## Supporting information for

# Efficient 4,5-Dihydro-1*H*-imidazol-5-one Formation from Amidines and Ketones under Transition-Metal Free Conditions

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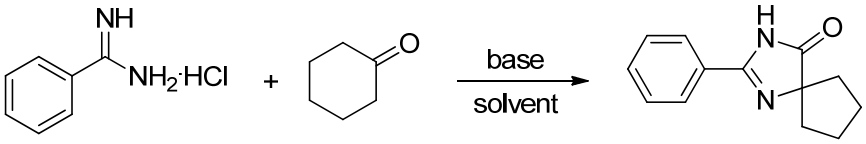
## General information:

All experiments were carried out under an atmosphere of oxygon. Flash column chromatography was performed over silica gel 48-75 μm. $^{1}H$ NMR and $^{13}C$ NMR spectra were recorded on Bruker-AV (400 or 500 MHz and 100 or 125 MHz, respectively) instrument internally referenced to $SiMe_4$, chloroform or dimethyl sulfoxide signals. Chemical shifts are referenced to solvent residual peak (2.50 ppm $^{1}H$, 39.50 ppm $^{13}C$ for DMSO – $d_6$, and 0 ppm $^{1}H$ for tetramethylsilane, 77.00 ppm $^{13}C$ for $CDCl_3$). MS analyses were performed on Agilent 5975 GC-MS instrument (EI). The new compounds were characterized by $^{1}H$ NMR, $^{13}C$ NMR, MS and HRMS. The structure of known compounds were further corroborated by comparing their $^{1}H$ NMR, $^{13}C$ NMR and MS data with those of literature. All reagents were used as received from commercial sources without further purification.

**Table S1. Optimization of the Reaction Conditions**[a]




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

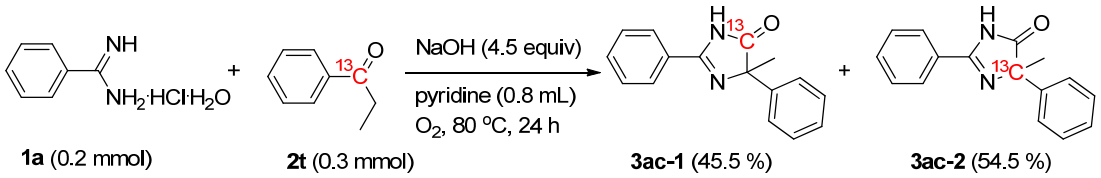
| entry | base (equiv) | T (°C) | solvent | yield (%)[b] |
|---|---|---|---|---|
| 1 | | 80 | NMP | trace |
| 2 | | 80 | pyridine | 5 |
| 3 | $K_2CO_3$ (2) | 80 | pyridine | 6 |
| 4 | NaOH (2) | 80 | pyridine | 33 |
| 5 | KOH (2) | 80 | pyridine | 23 |
| 6 | $^t$BuOK (2) | 80 | pyridine | 28 |
| 7 | NaOH (4) | 80 | pyridine | 88 |
| 8 | NaOH (4.5) | 80 | pyridine | 93 |
| 9 | NaOH (5) | 80 | pyridine | 91 |
| 10 | NaOH (4.5) | 80 | NMP | 20 |
| 11 | NaOH (4.5) | 80 | quinoline | 89 |
| 12 | NaOH (4.5) | 80 | toluene | 50 |
| 13[c] | NaOH (4.5) | 80 | pyridine | 75 |
| 14 | NaOH (4.5) | 60 | pyridine | 87 |
| 15 | NaOH (4.5) | 100 | pyridine | 88 |
| 16[d] | NaOH (4.5) | 80 | pyridine | 21 |

[a]Conditions: **1a** (0.2 mmol), **2a** (0.3 mmol), solvent (0.8 mL), 24 h, under oxygen unless otherwise noted. [b]GC yield based on **1a** using 1,3,5-trimethylbenzene as internal standard. [c]0.2 mmol of **2a** was used. [d]24 h under air.

## Control experiments:

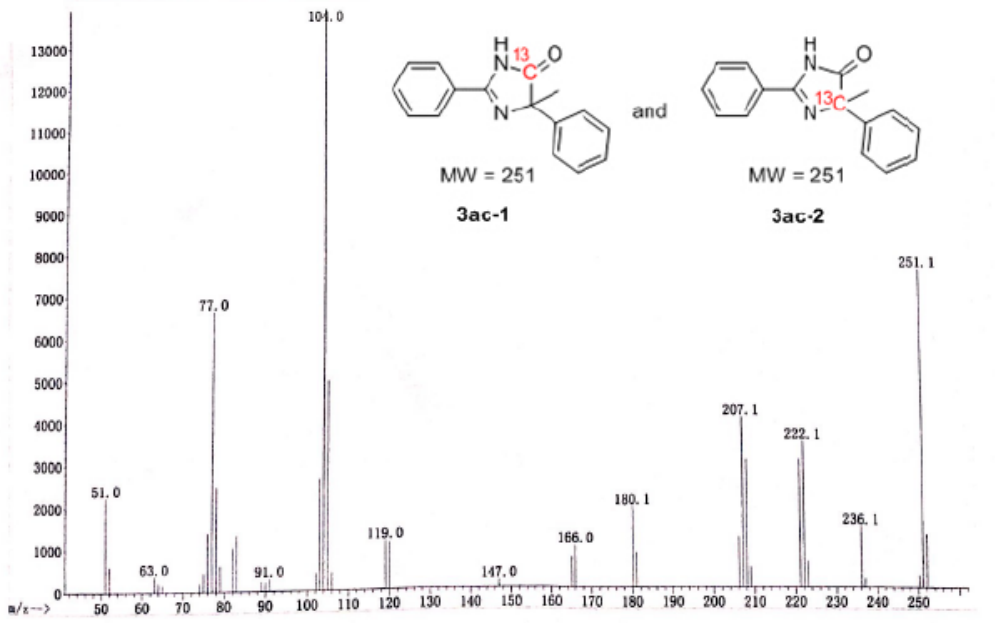
### 1) $^{13}$C labeling reaction

The reaction of **1a** with $^{13}$C labeled propiophenone (**2t**) was carried out under the standard reaction conditions (Scheme S1). The corresponding product **3ac** was isolated and analysed by MS (EI) and NMR. The $^{13}$C labeling reaction showed that the $^{13}$C-labeled carbon atom of **2t** appeared in two different sites in the product **3ac** (**3ac-1** and **3ac-2)**. The ratio of **3ac-1**/**3ac-2** was calculated based on the $^{13}$C NMR spectra.

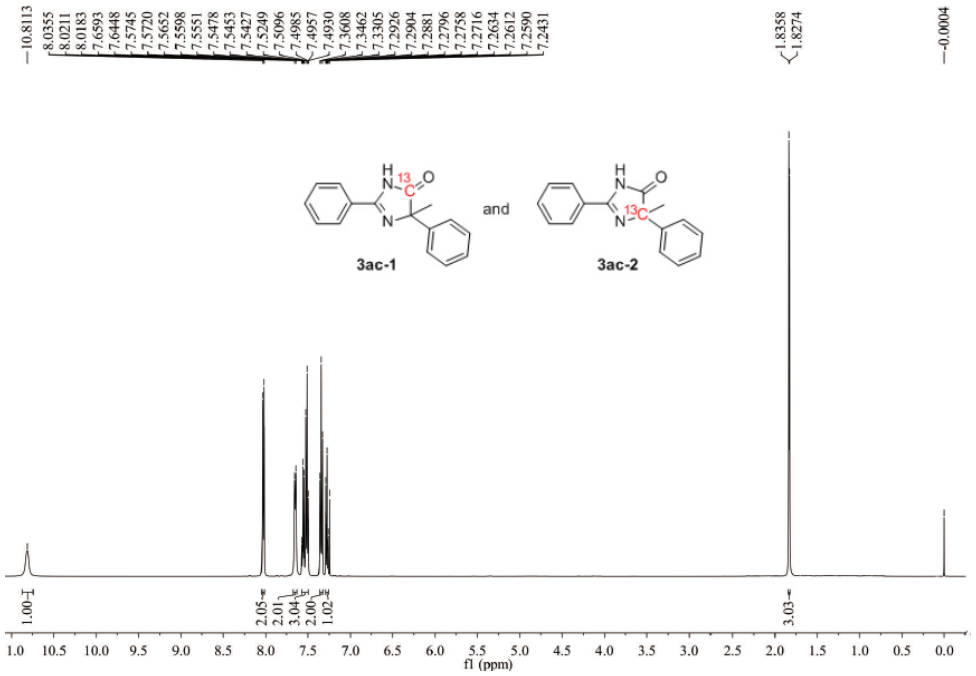



Scheme S1 $^{13}$C labeling reaction of **1a** with **2t**.

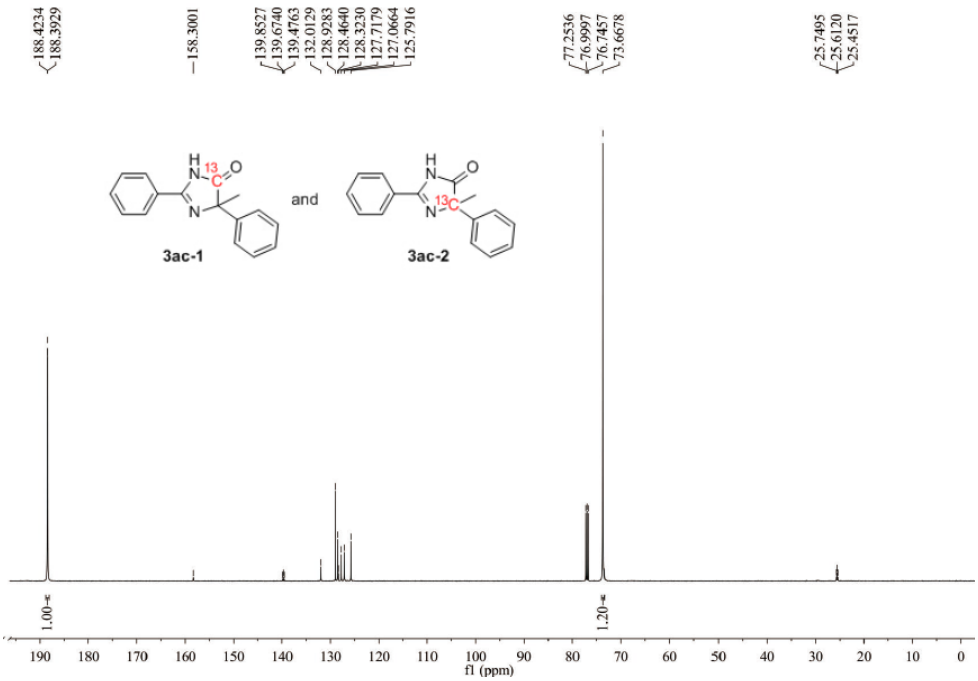
## MS analysis of $^{13}$C labeling products of 3ac-1 and 3ac-2



## $^{1}$H NMR spectra of 3ac-1 and 3ac-2

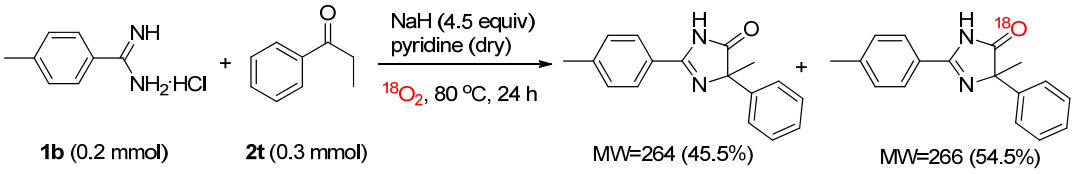



## $^{13}$C NMR spectra of 3ac-1 and 3ac-2



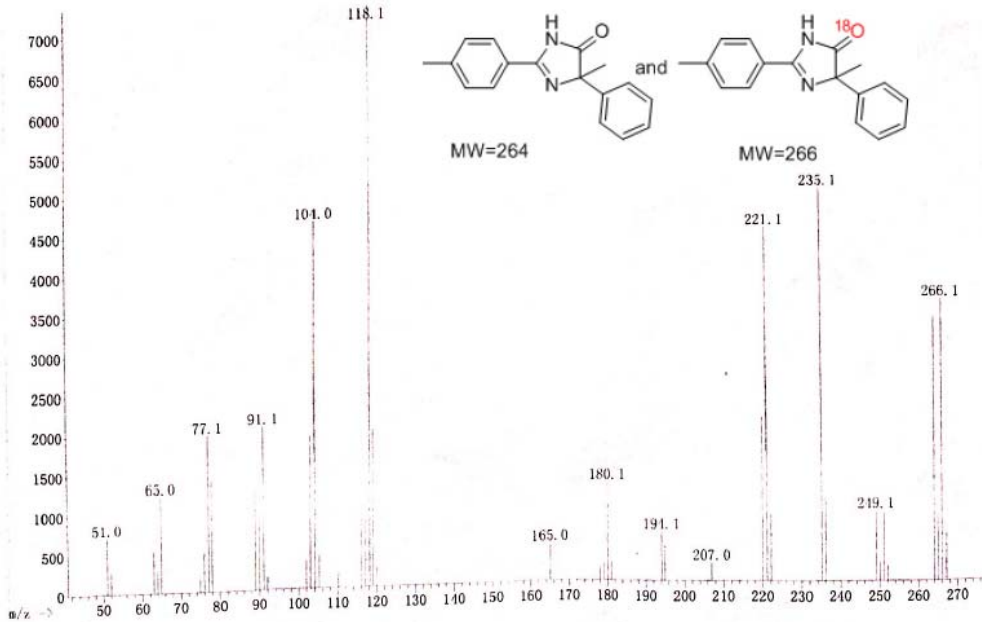
**2) $^{18}O$ labeling reaction**

The reaction of 4-methylbenzimidamide hydrochloride (**1b**) with propiophenone (**2t**) was carried out under $^{18}O_2$ atmosphere with modified reaction conditions (Scheme S2). To avoid any other oxygen source, NaH was used as the base to replace NaOH. The reaction result showed that more than half of the product is $^{18}O$-labeled as calculated by MS (by comparing to the product spectra under $O_2$). This means part of the oxygen atoms in the product came from the molecular oxygen.




Scheme S2 Reaction of **1b** with **2t** under $^{18}O_2$ atmosphere.

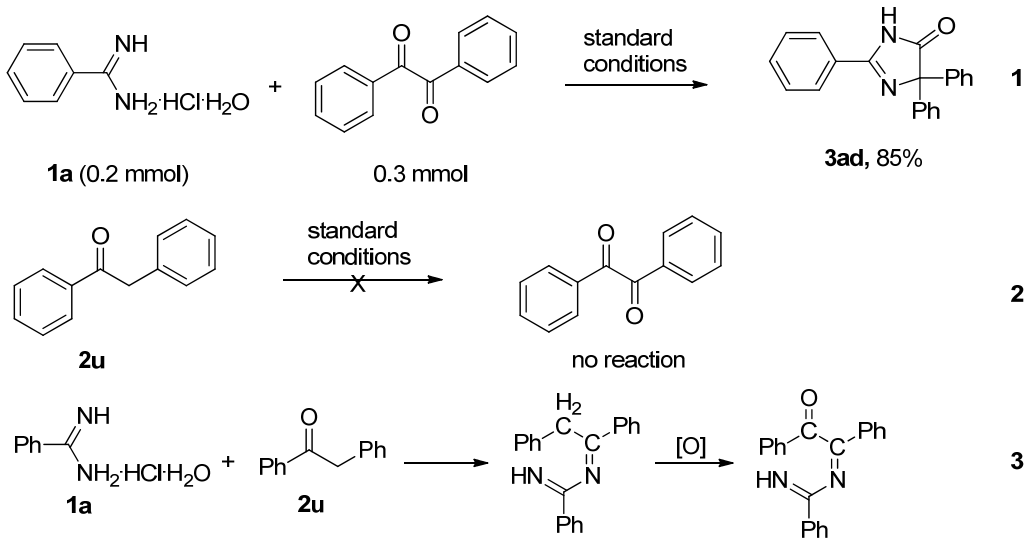
**The MS spectra of reaction under $^{18}O_2$**




**3) Reaction of 1a with benzil**

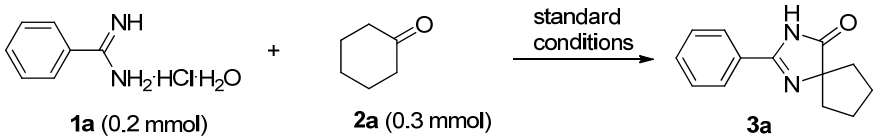
Under the standard reaction conditions, benzamidine hydrochloride hydrate (**1a**) smoothly reacted with benzil to afford the corresponding produce **3ad** in 85% yield (Scheme S3, eq.1). However,

1,2-diphenylethanone (**2u**) could not be converted into benzil under the standard condition (Scheme S3, eq.2). This means the $CH_2$ group *ortho* to the carbonyl group in ketone might be oxidized into the carbonyl group and serves as a key intermediate under the standard reaction conditions with the aid of amidine substrate (Scheme S3, eq.3).



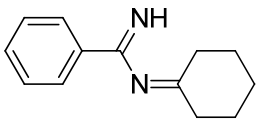

Scheme S3 Reaction of **1a** with benzil.

**4) HRMS monitoring result of reaction 1a with 2a**

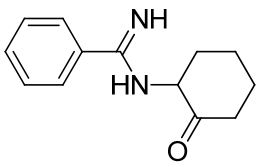



Scheme S4 Reaction of **1a** with **2a**.

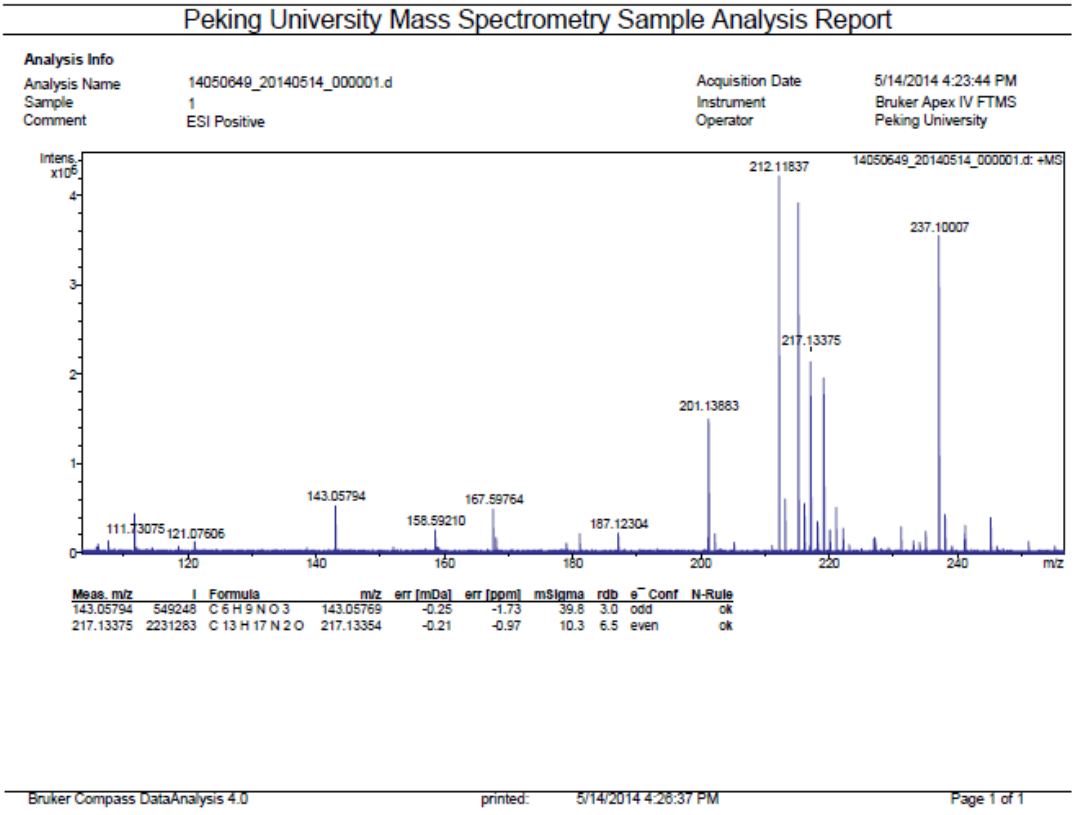
The reaction of **1a** with **2a** was carried out under the standard conditions for 4 h and the mixture was directly analysed by HRMS (Scheme S4). The HRMS analysis result showed that two possible intermediates were detected with $[M+H]^+$ = 201.13883, $[M+H]^+$ =217.13375 which could be:



Chemical Formula: $C_{13}H_{16}N_2$
Exact Mass: 200.13



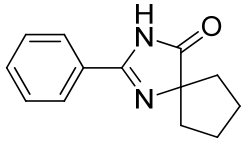
Chemical Formula: $C_{13}H_{16}N_2O$
Exact Mass: 216.13




**General procedure: 2-phenyl-1,3-diazaspiro[4.4]non-1-en-4-one (3a)**:

A 10 mL oven-dried reaction vessel was charged with NaOH (36 mg, 0.9 mmol), benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol). The reaction vessel was purged with oxygen for three times and was added cyclohexanone (**2a**, 31.2 μL, 0.3 mmol), pyridine (0.8 mL) by syringe. The sealed vessel was stirred at 80 $^{o}$C for 24 h. After cooling to room temperature, the volatiles were removed under vacuum and the residue was purified by column chromatography (silica gel, petroleum ether/ethyl acetate = 9:1) to give **3a** as white solid; yield: 36.8 mg (86%). m.p.: 201-203 °C.

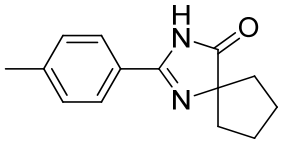
**2-Phenyl-1,3-diazaspiro[4.4]non-1-en-4-one (3a, CAS: 779309-80-7)[1]**




$^{1}$H NMR (400 MHz, $CDCl_3$, ppm) $\delta$ 10.02 (br, 1H), 7.92 (d, $J$ = 6.8 Hz, 2H), 7.54-7.49 (m, 3H), 2.07-1.97 (m, 8H); $^{13}$C NMR (100 MHz, DMSO-$d_6$, ppm) $\delta$ 188.0, 157.4, 131.5, 128.7, 128.4,

126.7, 77.4, 37.0, 25.5; MS (EI) *m/z* (%) 214, 171, 104 (100), 83, 54.

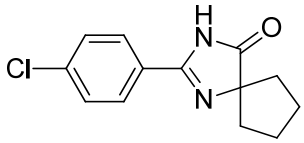
**2-(*p*-Tolyl)-1,3-diazaspiro[4.4]non-1-en-4-one (3b)**



The reaction was conducted with 4-methylbenzimidamide hydrochloride (**1b**, 34.1 mg. 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 40.1 mg, 88% yield of **3b** as white solid. m.p.: 194-196 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.14 (br, 1H), 7.80 (d, $J$ = 8.0 Hz, 2H), 7.31 (d, $J$ = 8.1 Hz, 2H), 2.42 (s, 3H), 2.07-1.94 (m, 8H); $^{13}$C NMR (125 MHz, DMSO-$d_6$, ppm) $\delta$ 187.8, 156.6, 136.2, 128.8, 128.5, 126.7, 77.5, 37.0, 25.5, 21.0; MS (EI) *m/z* (%) 228, 185, 118 (100), 83, 65; HRMS calcd. for: $C_{14}H_{17}ON_2$ $[M+H]^+$ 229.1335, found 229.1333.

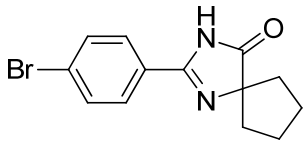
**2-(4-Chlorophenyl)-1,3-diazaspiro[4.4]non-1-en-4-one (3c)**



The reaction was conducted with 4-chlorobenzamidine hydrochloride (**1c**, 38.2 mg. 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 43.2 mg, 87% yield of **3c** as white solid. m.p.: 258-259 °C.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) $\delta$ 10.02 (br, 1H), 7.86 (d, $J$ = 7.7 Hz , 2H), 7.49 (d, $J$ = 8.2 Hz , 2H), 2.06-1.94 (m, 8H); $^{13}$C NMR (125 MHz, DMSO-$d_6$, ppm) $\delta$ 187.9, 156.7, 136.2, 128.9, 128.5, 127.7, 77.6, 37.0, 25.5; MS (EI) *m/z* (%) 248, 138 (100), 83, 75, 54; HRMS calcd. for: $C_{13}H_{14}ON_2Cl[M+H]^+$ 249.0789, found 249.0787.

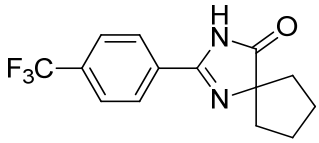
**2-(4-Bromophenyl)-1,3-diazaspiro[4.4]non-1-en-4-one (3d)**



The reaction was conducted with 4-bromobenzamidine hydrochloride (**1d**, 47.1 mg. 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 49.1 mg, 84% yield of **3d** as white solid. m.p.: 279-281 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 9.98 (br, 1H), 7.78 (d, $J$ = 8.2 Hz, 2H), 7.65 (d, $J$ = 8.5 Hz, 2H), 2.11-1.94 (m, 8H); $^{13}$C NMR (125 MHz, DMSO-$d_6$, ppm) $\delta$ 187.9, 156.8, 131.8, 128.7, 128.0, 125.2, 77.6, 37.0, 25.5; MS (EI) *m/z* (%) 292, 184, 83 (100), 76, 54; HRMS calcd. for: $C_{13}H_{14}ON_2Br[M+H]^+$ 293.0284, found 293.0282.

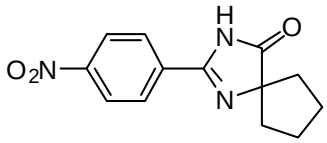
**2-(4-(Trifluoromethyl)phenyl)-1,3-diazaspiro[4.4]non-1-en-4-one (3e)**



The reaction was conducted with 4-(trifluoromethyl)benzamidine hydrochloride (**1e**, 44.9 mg. 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 50.8 mg, 90% yield of **3e** as white solid. m.p.: 217-219 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.81 (br, 1H), 8.09 (d, $J$ = 8.2 Hz, 2H), 7.79 (d, $J$ = 8.3 Hz, 2H), 2.14-1.94 (m, 8H); $^{13}$C NMR (125 MHz, DMSO-$d_6$, ppm) $\delta$ 187.7, 156.7, 132.6, 131.3 (q, $J$ = 33.6 Hz), 127.6, 126.0 (q, $J$ = 274.5 Hz), 125.6, 77.8, 37.0, 25.5; MS (EI) *m/z* (%) 282, 239, 172 (100), 145, 54; HRMS calcd. for: $C_{14}H_{14}ON_2F_3$ $[M+H]^+$ 283.1053, found 283.1049.

**2-(4-nitrophenyl)-1,3-diazaspiro[4.4]non-1-en-4-one (3f)**

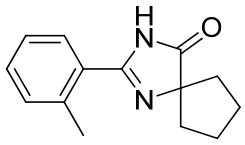


The reaction was conducted with 4-nitrobenzimidamide hydrochloride (**1f**, 40.2 mg, 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column

chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 38.3 mg. 74% yield of **3f** as white solid. m.p.: 262-264 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.86 (br, 1H), 8.38 (d, $J$ = 8.8 Hz, 2H), 8.15 (d, $J$ = 8.7 Hz, 2H), 2.14-1.95 (m, 8H); $^{13}$C NMR (125 MHz, DMSO-$d_6$, ppm) $\delta$ 187.7, 156.4, 149.1, 134.4, 128.1, 123.9, 78.0, 37.0, 25.5; MS (EI) *m/z* (%) 259, 149, 103, 83 (100), 54; HRMS calcd. for: $C_{13}H_{14}O_3N_3$ $[M+H]^+$ 260.1030, found 260.1033.

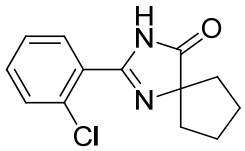
**2-(*o*-Tolyl)-1,3-diazaspiro[4.4]non-1-en-4-one (3g)**



The reaction was conducted with 2-methylbenzimidamide hydrochloride (**1g**, 34.1 mg, 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 40.6 mg. 89% yield of **3g** as white solid. m.p.: 143-145 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 8.86 (br, 1H), 7.50-7.48 (m, 1H), 7.39-7.36 (m, 1H), 7.30-7.28 (m, 2H), 2.52 (s, 3H ), 2.08-1.92 (m, 8H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 189.6, 158.3, 137.2, 131.2, 130.4, 128.8, 128.4, 125.9, 78.6, 37.5, 26.0, 20.4; MS (EI) *m/z* (%) 228, 158, 118 (100), 83, 54; HRMS calcd. for: $C_{14}H_{17}ON_2$ $[M+H]^+$ 229.1335, found 229.1333.

**2-(2-Chlorophenyl)-1,3-diazaspiro[4.4]non-1-en-4-one (3h, CAS: 1316305-72-2)[2]**

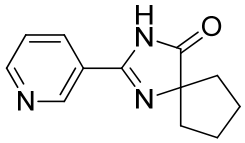


The reaction was conducted with 2-chlorobenzimidamide hydrochloride (**1h**, 38.2 mg, 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 42.2 mg. 85% yield of **3h** as white solid. m.p.: 183-185 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 8.68 (br, 1H), 7.92 (d, $J$ = 7.4 Hz, 1H), 7.46-7.45 (m, 2H), 7.41-7.37 (m, 1H), 2.07-1.94 (m, 8H); $^{13}$C NMR (125 MHz, DMSO-$d_6$, ppm) $\delta$ 187.9, 157.3,

132.0, 130.7, 129.9, 129.2, 127.3, 126.7, 77.6, 36.7, 25.5; MS (EI) *m/z* (%) 248, 185, 138 (100), 83, 54.

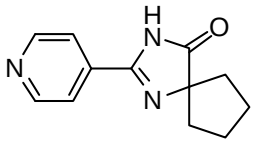
**2-(Pyridin-3-yl)-1,3-diazaspiro[4.4]non-1-en-4-one (3i)**



The reaction was conducted with 3-pyridinecarboximidamide hydrochloride (**1i**, 31.5 mg, 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 30.5 mg. 71% yield of **3i** as white solid. m.p.: 193-195 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.98 (br, 1H), 9.20 (d, $J$ = 1.6 Hz, 1H), 8.79-8.78 (m, 1H), 8.32-8.29 (m, 1H), 7.48-7.46 (m, 1H), 2.13-1.93 (m, 8H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.6, 155.7, 152.0, 150.0, 134.5, 125.0, 123.7, 78.8, 37.5, 26.1; MS (EI) *m/z* (%) 215, 172, 105 (100), 78, 54; HRMS calcd. for: $C_{12}H_{14}ON_3$ $[M+H]^+$ 216.1131, found 216.1130.

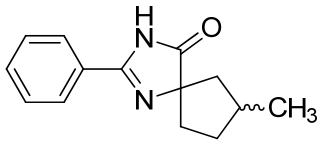
**2-(Pyridin-4-yl)-1,3-diazaspiro[4.4]non-1-en-4-one (3j)**



The reaction was conducted with 4-pyridinecarboximidamide hydrochloride (**1j**, 31.5 mg, 0.2 mmol) and cyclohexanone (**2a**, 31.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 32.3 mg. 75% yield of **3j** as white solid. m.p.: 215-217 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.89 (br, 1H), 8.82 (d, $J$ = 5.1 Hz, 2H), 7.83 (d, $J$ = 6.0 Hz, 2H), 2,14-1.93 (m, 8H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.8, 155.9, 150.6, 136.1, 120.7, 79.5, 37.6, 26.2; MS (EI) *m/z* (%) 215, 172, 105 (100), 83, 54; HRMS calcd. for: $C_{12}H_{14}ON_3$ $[M+H]^+$ 216.1131, found 216.1135.

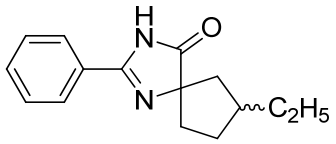
**7-Methyl-2-phenyl-1,3-diazaspiro[4.4]non-1-en-4-one (3k)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 4-methylcyclohexanone (**2b**, 36.8 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 36.5 mg. 80% yield of **3k** as white solid.

$^1$H NMR (500 MHz, $CDCl_3$, mixture, ppm) δ 10.27 (br, 0.5H), 10.19 (br, 0.5H), 7.92 (d, *J* = 7.6 Hz, 2H), 7.55-7.49 (m, 3H), 2.61-2.53 (m, 0.5H), 2.48-2.40 (m, 0.5H), 2.22-1.93 (m, 4H), 1.75-1.54 (m, 2H), 1.15 (t, *J* = 7.0 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, mixture, ppm) δ 191.2, 190.6, 157.3, 131.6, 128.8, 128.6, 126.9, 79.2, 78.6, 46.1, 44.8, 37.6, 36.7, 35.4, 35.1, 35.0, 34.5, 19.9, 19.6; MS (EI) *m/z* (%) 228, 185, 104 (100), 82, 51; HRMS calcd. for: $C_{14}H_{17}ON_2$ $[M+H]^+$ 229.1335, found 229.1334.

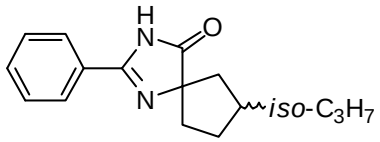
**7-Ethyl-2-phenyl-1,3-diazaspiro[4.4]non-1-en-4-one (3l)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 4-ethylcyclohexanone (**2c**, 42.3 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 40.7 mg. 84% yield of **3l** as white solid.

$^1$H NMR (500 MHz, $CDCl_3$, mixture, ppm) δ 10.15 (br, 0.5H), 10.05 (br, 0.5H), 7.92-7.90 (m, 2H), 7.55-7.48 (m, 3H), 2.43-1.92 (m, 5H), 1.76-1.47 (m, 4H), 0.97-0.94 (m, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, mixture, ppm) δ 191.1, 190.5, 157.3, 131.6, 128.8, 128.7, 126.9, 78.9, 78.3, 44.1, 42.8, 42.6, 42.3, 37.4, 36.4, 32.7, 32.1, 28.4, 28.2, 13.0, 12.9; MS (EI) *m/z* (%) 242, 173, 104 (100), 82, 54; HRMS calcd. for: $C_{15}H_{19}ON_2$ $[M+H]^+$ 243.1492, found 243.1489.

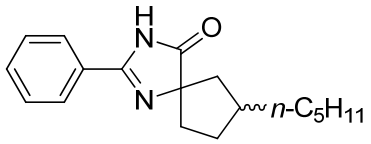
**2-phenyl-7-(*iso*-propyl)-1,3-diazaspiro[4.4]non-1-en-4-one (3m)**




The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 4-(*iso*-propyl)cyclohexanone (**2d**, 46.5 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 42.5 mg. 83% yield of **3m** as white solid.

$^1$H NMR (500 MHz, $CDCl_3$, mixture, ppm) δ 10.12 (br, 0.5H), 9.99 (br, 0.5H), 7.92-7.90 (t, $J$ = 6.1 Hz, 2H), 7.55-7.48 (m, 3H), 2.18-1.58 (m, 8H), 0.98-0.97 (m, 3H), 0.94-0.92 (m, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, mixture, ppm) δ 191.1, 190.6, 157.3, 131.6, 128.8, 128.7, 128.66, 126.9, 126.88, 78.9, 78.3, 48.3, 47.9, 42.8, 41.4, 37.4, 36.3, 33.4, 33.3, 31.3, 30.6, 21.6, 21.56, 21.55, 21.5; MS (EI) *m/z* (%) 256, 228, 173, 104 (100), 82; HRMS calcd. for: $C_{16}H_{21}ON_2$ $[M+H]^+$ 257.1648, found 257.1649.

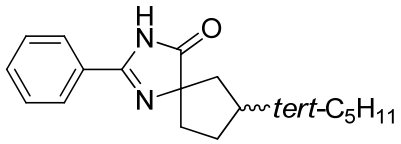
**7-(*n*-Pentyl)-2-phenyl-1,3-diazaspiro[4.4]non-1-en-4-one (3n)**




The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 4-(*n*-pentyl)cyclohexanone (**2e**, 57 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 48.3 mg. 85% yield of **3n** as white solid.

$^1$H NMR (500 MHz, $CDCl_3$, mixture, ppm) δ 10.14 (br, 0.5H), 10.04 (br, 0.5H), 7.92-7.90 (m, 2H), 7.55-7.49 (m, 3H), 2.49-2.28 (m, 1H), 2.22-1.92 (m, 4H), 1.76-1.25 (m, 10H), 0.89 (t, $J$ = 6.8 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, mixture, ppm) δ 191.1, 190.6, 157.2, 131.6, 128.8, 128.7, 126.9, 78.9, 78.3, 44.5, 43.1, 40.8, 40.5, 37.4, 36.4, 35.5, 35.3, 33.1, 32.5, 32.0, 31.9, 29.7, 28.34, 28.31, 22.63, 22.62, 14.1; MS (EI) *m/z* (%) 284, 173, 104 (100), 82, 55; HRMS calcd. for: $C_{18}H_{25}ON_2$ $[M+H]^+$ 285.1961, found 285.1958.

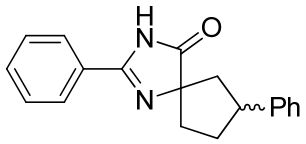
**7- (*tert*-Pentyl)-2-phenyl-1,3-diazaspiro[4.4]non-1-en-4-one (3o)**




The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 4-(*tert*-pentyl)cyclohexanone (**2f**, 55 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 40.3 mg. 71% yield of **3o** as white solid.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) δ 9.97 (br, 1H), 7.92 (d, $J$ = 5.2 Hz, 2H), 7.56-7.48 (m, 3H), 2.38-2.31 (m, 1H), 2.02-1.82 (m, 6H), 1.32-1.27 (q, $J$ = 7.5 Hz, 2H), 0.90-0.85 (m, 9H); $^{13}$C NMR (125 MHz, $CDCl_3$, mixture, ppm) δ 191.3, 190.7, 157.4, 157.35, 131.5, 128.7, 128.66, 128.62, 126.9, 126.87, 78.6, 77.9, 49.0, 48.8, 38.9, 37.7, 36.8, 36.3, 34.4, 34.2, 33.8, 33.7, 27.2, 26.5, 23.9, 23.8, 23.75, 23.70, 8.3, 8.2; MS (EI) *m/z* (%) 284, 255, 173 (100), 82, 55; HRMS calcd. for: $C_{18}H_{25}ON_2$ $[M+H]^+$ 285.1961, found 285.1957.

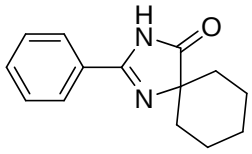
**2,7-Diphenyl-1,3-diazaspiro[4.4]non-1-en-4-one (3p)**




The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 4-phenylcyclohexanone (**2g**, 50.2 mg, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 44.1 mg. 76% yield of **3p** as white solid.

$^1$H NMR (400 MHz, $CDCl_3$, mixture, ppm) δ 10.18 (br, 0.5H), 10.03 (br, 0.5H), 7.96 (d, $J$ = 6.4 Hz, 2H), 7.57-7.52 (m, 3H), 7.41-7.32 (m, 4H), 7.23-7.22 (m, 1H), 3.75-3.71 (m, 0.5H), 3.60-3.56 (m, 0.5H), 2.52-2.07 (m, 6H); $^{13}$C NMR (125 MHz, $CDCl_3$, mixture, ppm) δ 190.7, 190.3, 157.7, 157.6, 144.2, 144.0, 131.8, 131.7, 128.92, 128.91, 128.7, 128.4, 127.3, 127.2, 126.92, 126.90, 126.3, 126.2, 78.7, 78.2, 46.0, 45.6, 45.4, 44.3, 37.6, 36.6, 35.2, 34.3; MS (EI) *m/z* (%) 290, 173 (100), 104, 77, 51; HRMS calcd. for: $C_{19}H_{19}ON_2$ $[M+H]^+$ 291.1492, found 291.1491.

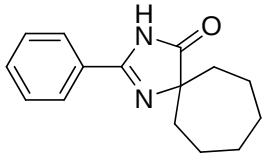
**2-Phenyl-1,3-diazaspiro[4.5]dec-1-en-4-one (3q, CAS: 34935-85-8)[3]**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and cycloheptanone (**2h**, 35.4 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 36.9 mg. 81% yield of **3q** as white solid. m.p.: 175-177 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) δ 10.24 (br, 1H), 7.95-7.93 (m, 2H), 7.55-7.49 (m, 3H), 1.91-1.50 (m, 10H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) δ 190.7, 157.2, 131.5, 128.8, 128.7, 127.0, 72.1, 33.2, 25.3, 21.6; MS (EI) *m/z* (%) 228, 173, 104 (100), 97, 54.

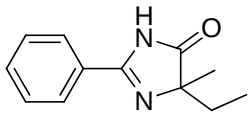
**2-Phenyl-1,3-diazaspiro[4.6]undec-1-en-4-one (3r)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and cyclooctanone (**2i**, 39.5 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 43.1 mg. 89% yield of **3r** as white solid. m.p.: 164-165 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) δ 10.11 (br, 1H), 7.93 (d, $J$ = 6.9 Hz, 2H), 7.56-7.49 (m, 3H), 1.92-1.68 (m, 12H); $^{13}$C NMR (100 MHz, $C_2D_6SO$, ppm) δ 188.4, 156.7, 131.0, 128.2, 128.0, 126.3, 72.5, 35.4, 29.1, 22.0; MS (EI) *m/z* (%) 242,173, 104 (100), 83, 54; HRMS calcd. for: $C_{15}H_{19}ON_2$ $[M+H]^+$ 243.1492, found 243.1490.

**4-Ethyl-4-methyl-2-phenyl-1*H*-imidazol-5(4*H*)-one (3s)**

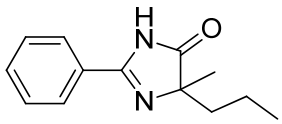


The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 2-pentanone (**2j**, 31.9 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 27.5 mg. 68% yield of

**3s** as white solid. m.p.: 150-151 °C.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) $\delta$ 10.23 (br, 1H), 7.94 (d, $J$ = 7.0 Hz, 2H), 7.57-7.51 (m, 3H), 1.92 (q, $J$ = 7.0 Hz, 2H), 1.47 (s, 3H), 0.84 (t, $J$ = 7.4 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.1, 158.1, 131.7, 128.8, 128.4, 126.9, 73.0, 30.9, 22.9, 8.2; MS (EI) *m/z* (%) 202, 173, 104 (100), 77, 51; HRMS calcd. for: $C_{12}H_{15}ON_2$ $[M+H]^+$ 203.1179, found 203.1176.

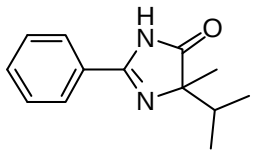
**4-Methyl-2-phenyl-4-propyl-1*H*-imidazol-5(4*H*)-one (3t)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 2-hexanone (**2k**, 37.1 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 30.7 mg. 71% yield of **3t** as white solid. m.p.: 130-132 °C.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) $\delta$ 10.16 (br, 1H), 7.94 (d, $J$ = 6.6 Hz, 2H), 7.57-7.50 (m, 3H), 1.86 (t, $J$ = 8.4 Hz, 2H), 1.46 (s, 3H), 1.34-1.14 (m, 2H), 0.88 (t, $J$ = 7.2 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.2, 157.9, 131.7, 128.8, 128.4, 126.9, 72.7, 39.9, 23.3, 17.1, 14.0; MS (EI) *m/z* (%) 216, 173, 104 (100), 77, 51; HRMS calcd. for: $C_{13}H_{17}ON_2$ $[M+H]^+$ 217.1335, found 217.1334.

**4-Methyl-2-phenyl-4-(*iso*-propyl)-1*H*-imidazol-5(4*H*)-one (3u)**

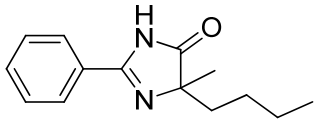


The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 4-methylpentan-2-one (**2l**, 37.6 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 26.4 mg. 61% yield of **3u** as white solid. m.p.: 120-123 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.23 (br, 1H), 7.94 (d, $J$ = 7.1 Hz, 2H), 7.56-7.50 (m, 3H), 2.16-2.10 (m, 1H), 1.45 (s, 3H), 1.08 (d, $J$ = 6.8 Hz, 3H), 0.92 (d, $J$ = 6.8 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.3, 158.0, 131.7, 128.9, 128.5, 126.9, 75.2, 34.9, 21.2, 16.9, 16.8; MS (EI)

*m/z* (%) 216, 174, 104 (100), 77, 51; HRMS calcd. for: $C_{13}H_{17}ON_2$ $[M+H]^+$ 217.1335, found 217.1337.

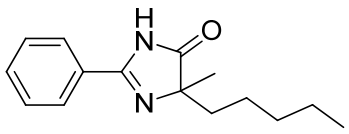
**4-Butyl-4-methyl-2-phenyl-1*H*-imidazol-5(4*H*)-one (3v)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 2-heptanone (**2m**, 41.8 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 36.8 mg. 80% yield of **3v** as white solid. m.p.: 94-95 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.29 (br, 1H), 7.94 (d, $J$ = 7.1 Hz, 2H), 7.58-7.51 (m, 3H), 1.87 (t, $J$ = 8.1 Hz, 2H), 1.46 (s, 3H), 1.31-1.10 (m, 4H), 0.84 (t, $J$ = 7.2 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.3, 158.0, 131.7, 128.8, 128.4, 126.9, 72.5, 37.6, 25.8, 23.3, 22.6, 13.8; MS (EI) *m/z* (%) 230, 144, 104 (100), 77, 57; HRMS calcd. for: $C_{14}H_{19}ON_2$ $[M+H]^+$ 231.1492, found 231.1491.

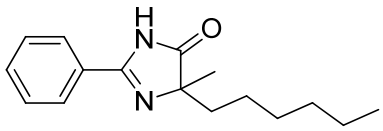
**4-Methyl-4-pentyl-2-phenyl-1*H*-imidazol-5(4*H*)-one (3w)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 2-octanone (**2n**, 47 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 37.6 mg. 77% yield of **3w** as white solid. m.p.: 99-100 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.16 (br, 1H), 7.94 (d, $J$ = 7.3 Hz, 2H), 7.58-7.50 (m, 3H), 1.87 (t, $J$ = 8.0 Hz, 2H), 1.46 (s, 3H), 1.28-1.11 (m, 6H), 0.82 (t, $J$ = 6.9 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.3, 157.9, 131.7, 128.8, 128.4, 126.9, 72.7, 37.8, 31.7, 23.4, 23.3, 22.3, 13.9; MS (EI) *m/z* (%) 244, 174, 104 (100), 77, 51; HRMS calcd. for: $C_{15}H_{21}ON_2$ $[M+H]^+$ 245.1648, found 245.1646.

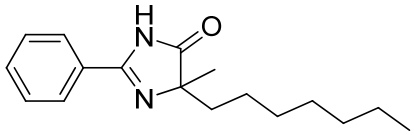
**4-Hexyl-4-methyl-2-phenyl-1*H*-imidazol-5(4*H*)-one (3x)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 2-nonanone (**2o**, 51.5 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 38.7 mg. 75% yield of **3x** as white solid. m.p.: 73-75 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.30 (br, 1H), 7.95 (d, $J$ = 7.2 Hz, 2H), 7.58-7.51 (m, 3H), 1.87 (t, $J$ = 7.7 Hz, 2H), 1.47 (s, 3H), 1.25-1.12 (m, 8H), 0.82 (t, $J$ = 6.9 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.2, 157.8, 131.7, 128.8, 128.4, 126.9, 72.7, 37.9, 31.6, 29.3, 23.7, 23.4, 22.5, 14.0; MS (EI) *m/z* (%) 258, 187, 104 (100), 77, 57; HRMS calcd. for: $C_{16}H_{23}ON_2$ $[M+H]^+$ 259.1805, found 259.1803.

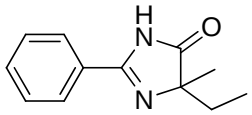
**4-Heptyl-4-methyl-2-phenyl-1*H*-imidazol-5(4*H*)-one (3y)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 2-decanone (**2p**, 57.2 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 39.7 mg. 73% yield of **3y** as white solid. m.p.: 90-92 °C.

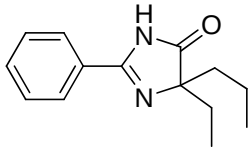
$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.00 (br, 1H), 7.95 (d, $J$ = 5.2 Hz, 2H), 7.59-7.51 (m, 3H), 1.88 (t, $J$ = 7.3 Hz, 2H), 1.47 (s, 3H), 1.25-1.12 (m, 10H), 0.83 (t, $J$ = 7.0 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 190.3, 157.9, 131.7, 128.8, 128.4, 126.9, 72.7, 37.9, 31.7, 29.5, 29.0, 23.8, 23.3, 22.5, 14.0; MS (EI) *m/z* (%) 272, 174, 104 (100), 77, 57; HRMS calcd. for: $C_{17}H_{25}ON_2$ $[M+H]^+$ 273.1961, found 273.1959.

**4-Ethyl-4-methyl-2-phenyl-1*H*-imidazol-5(4*H*)-one (3z)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 3-pentanone (**2q**, 31.9 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 29.9 mg. 74% yield of **3z** as white solid. This product is same as **3s**.

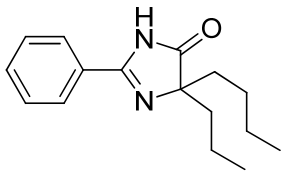
**4-Ethyl-2-phenyl-4-propyl-1*H*-imidazol-5(4*H*)-one (3aa)**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 4-heptanone (**2r**, 41.8 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 32.2 mg. 70% yield of **3aa** as white solid. m.p.: 144-146 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 10.11 (br, 1H), 7.93 (d, $J$ = 7.1 Hz, 2H), 7.58-7.50 (m, 3H), 1.96-1.81 (m, 4H), 1.31-1.13 (m, 2H), 0.87 (t, $J$ = 7.3 Hz, 3H), 0.81 (t, $J$ = 7.4 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 189.7, 158.4, 131.7, 128.8, 128.3, 126.9, 77.2, 39.2, 30.3, 16.9, 14.1, 7.9; MS (EI) *m/z* (%) 230, 187, 104 (100), 77, 51; HRMS calcd. for: $C_{14}H_{19}ON_2$ $[M+H]^+$ 231.1492, found 231.1491.

**4-Butyl-2-phenyl-4-propyl-1*H*-imidazol-5(4*H*)-one (3ab)**

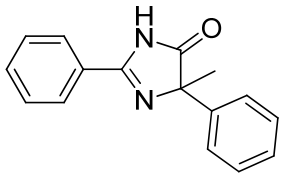


The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 5-nonanone (**2s**, 52 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 41.8 mg. 81% yield of **3ab** as white solid. m.p.: 114-116 °C.

$^1$H NMR (500 MHz, $CDCl_3$, ppm) $\delta$ 9.98 (br, 1H), 7.94 (d, $J$ = 4.7 Hz, 2H), 7.59-7.51 (m, 3H),

1.93-1.8 (m, 4H), 1.32-1.09 (m, 6H), 0.86 (t, $J$ = 7.3 Hz, 3H), 0.82 (t, $J$ = 7.2 Hz, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 189.7, 158.3, 131.7, 128.7, 128.3, 126.9, 76.2, 39.5, 37.1, 25.5, 22.6, 16.8, 14.0, 13.8; MS (EI) *m/z* (%) 258, 187, 104 (100), 77, 57; HRMS calcd. for: $C_{16}H_{23}ON_2$ $[M+H]^+$ 259.1805, found 259.1804.

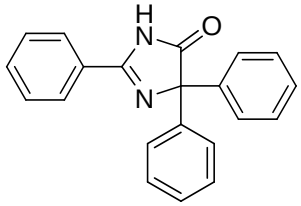
**4-Methyl-2,4-diphenyl-1*H*-imidazol-5(4*H*)-one (3ac)[4]**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and propiophenone (**2t**, 39.8 μL, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 42.5 mg. 85% yield of **3ac** as white solid. m.p.: 161-163 °C.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) $\delta$ 10.14 (br, 1H), 8.01 (d, $J$ = 7.0 Hz, 2H), 7.65 (d, $J$ = 7.6 Hz, 2H), 7.61-7.52 (m, 3H), 7.38-7.27 (m, 3H), 1.84 (s, 3H); $^{13}$C NMR (125 MHz, $CDCl_3$, ppm) $\delta$ 188.7, 158.9, 139.5, 132.0, 128.8, 128.4, 128.2, 127.7, 127.1, 125.7, 73.5, 25.4; MS (EI) *m/z* (%) 250, 180, 104 (100), 77, 51; HRMS calcd. for: $C_{16}H_{15}ON_2$ $[M+H]^+$ 251.1179, found 251.1178.

**2,4,4-triphenyl-1*H*-imidazol-5(4*H*)-one (3ad, CAS: 37068-60-3)[5]**



The reaction was conducted with benzamidine hydrochloride hydrate (**1a**, 35.6 mg, 0.2 mmol) and 1,2-diphenylethanone (**2u**, 58.9 mg, 0.3 mmol). The crude mixture was purified by flash column chromatography on silica (petroleum ether/ethyl acetate = 9:1) to provide 53.7 mg. 86% yield of **3ad** as white solid. m.p.: 224-225 °C.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) $\delta$ 10.05 (br, 1H), 8.01 (d, $J$ = 7.2 Hz, 2H), 7.62-7.49 (m, 7H), 7.35-7.27 (m, 6H); $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) $\delta$ 185.9, 158.2, 140.3, 132.1, 129.2, 128.9, 128.4, 127.8, 127.4, 127.2, 84.1; MS (EI) *m/z* (%) 312, 269, 180 (100), 104, 51.

## References


[1] I. Panov, P. Drabina, Z. Padělková, J. Hanusek and M. Sedlák, *J. Heterocyclic Chem.*, 2010, **47**, 1356.

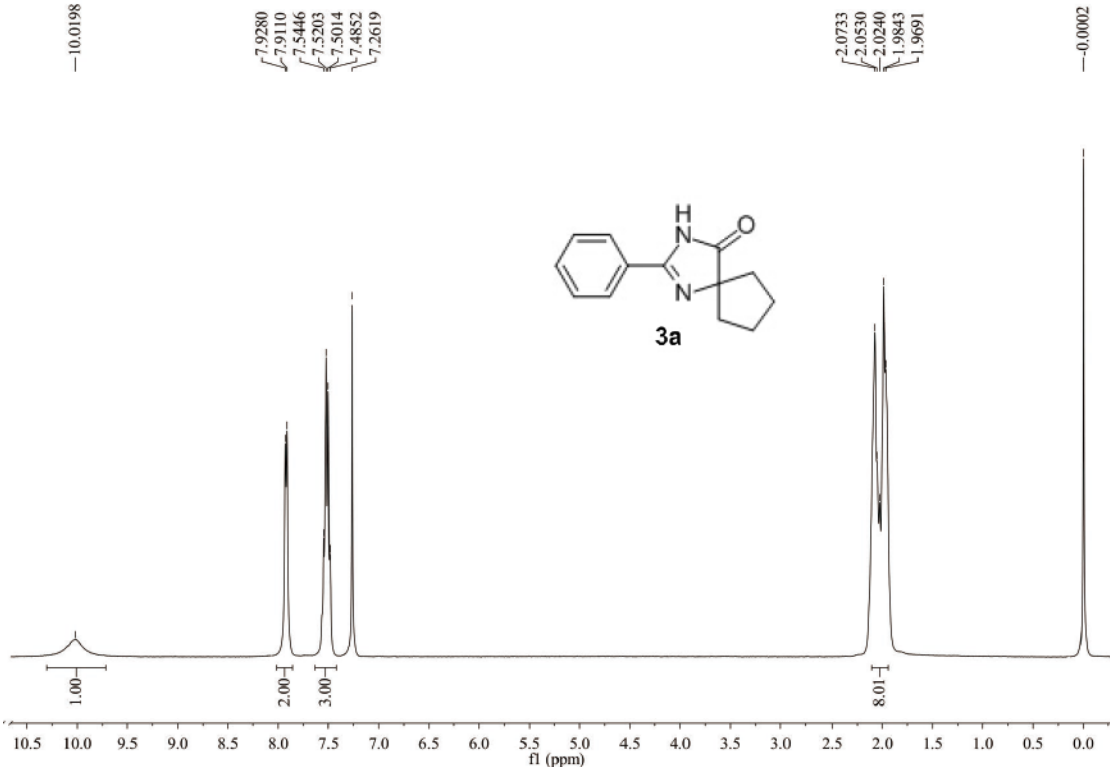
[2] M. R. Wood, I. M. Bell, J. J. Kim, H. G. Selnick and C. A. Stump, WO2010/21919 A1, 2010.

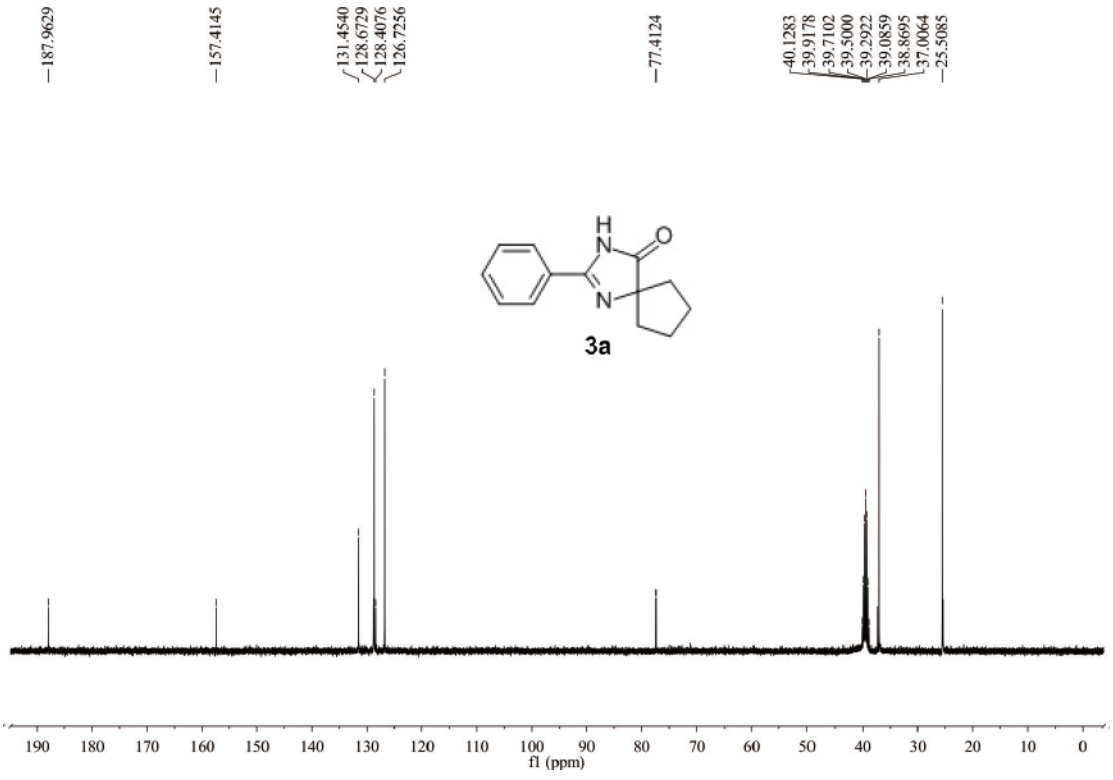
[3] T. Toda, S. Morimura, E. Mori, H. Horiuchi and K. Murayama, *Bull. Chem. Soc. Jpn.*, 1971, **44**, 3445.

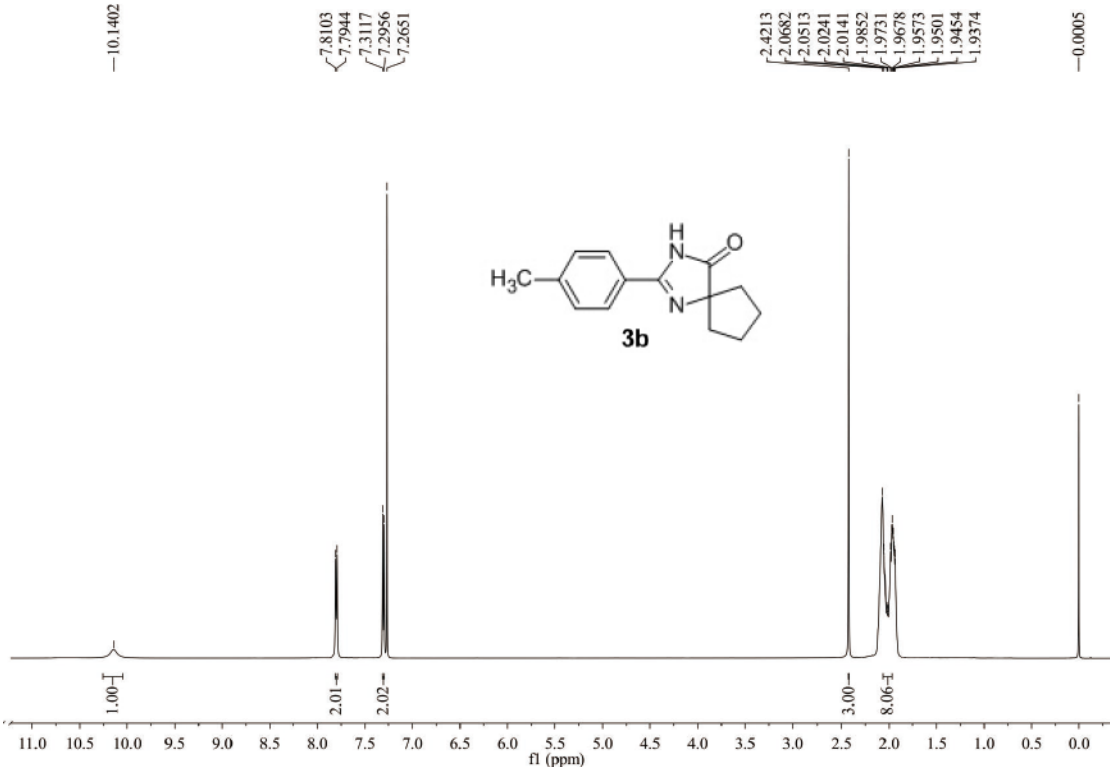
[4] Sakaguchi and Tanabe; *Yakugaku Zasshi*, 1977, **97**, 223.

[5] K. Gillman, M. Higgins, G. Poindexter, M. Browning, W. Clarke, S. Flowers, J. Grace, J. Hogan, R. McGovern, L. Iben, G. Mattson, A. Ortiz, S. Rassnick, J. Russell and I. Antal-Zimanyi, *Bioorgan. Med. Chem*., 2006, **14**, 5517.

# $^{1}$H and $^{13}$C NMR of products

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7.2956
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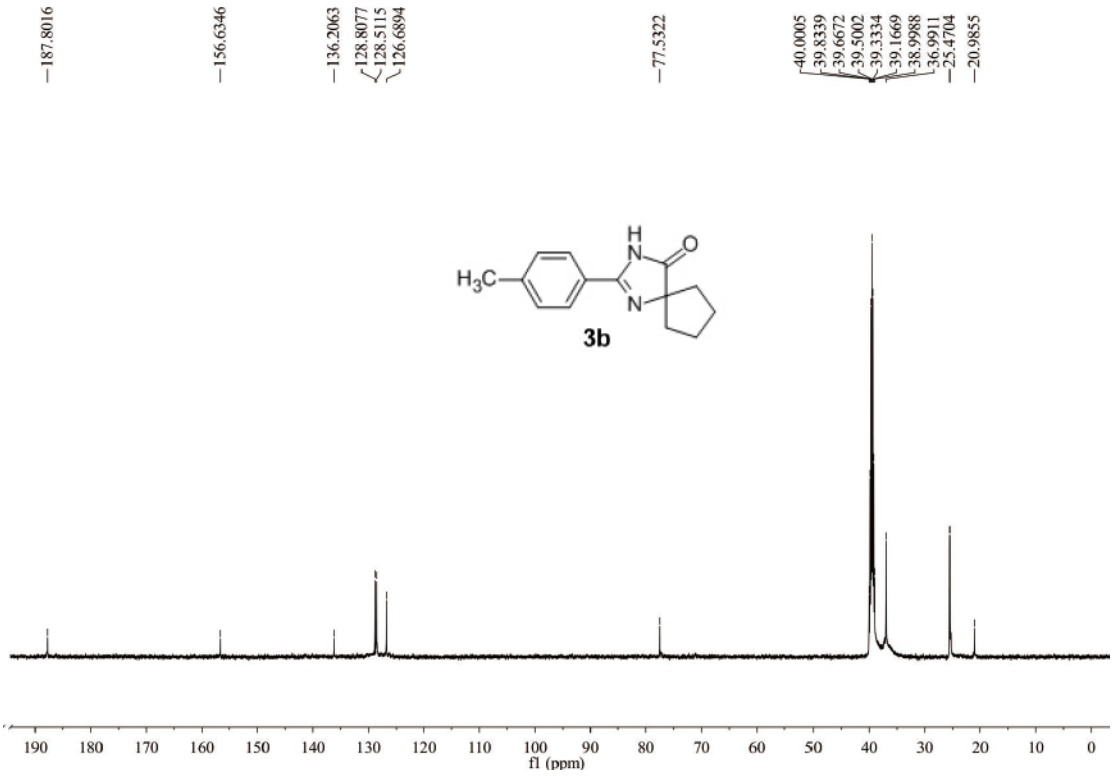
2.0513
2.0241
2.0141
1.9852
1.9731
1.9678
1.9573
1.9501
1.9454
1.9374
0.0005
H
N
O
H3C
N
3b
1.00
2.01
2.02
3.00
8.06
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)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36.9911
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H
N
O
H3C
N
3b
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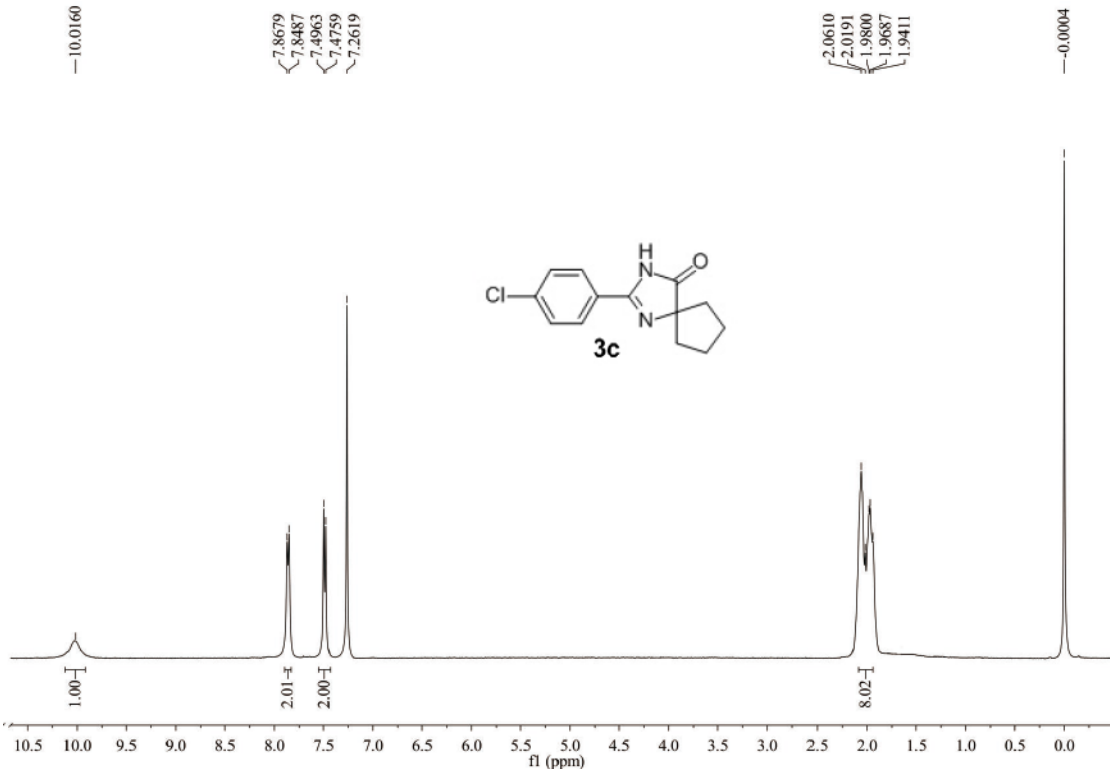
f1 (ppm)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1.9800
1.9687
1.9411
0.0004
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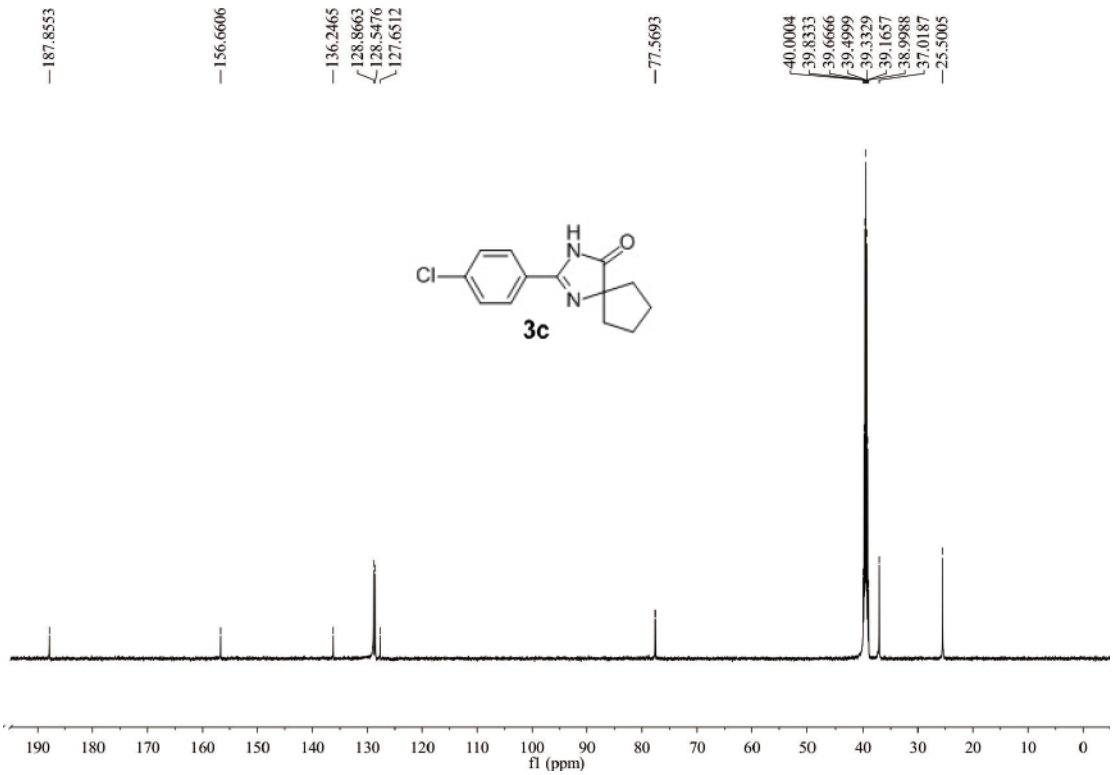
H
N
O
N
3c
1.00
2.01
2.00
8.02
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)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39.4999
39.3329
39.1657
38.9988
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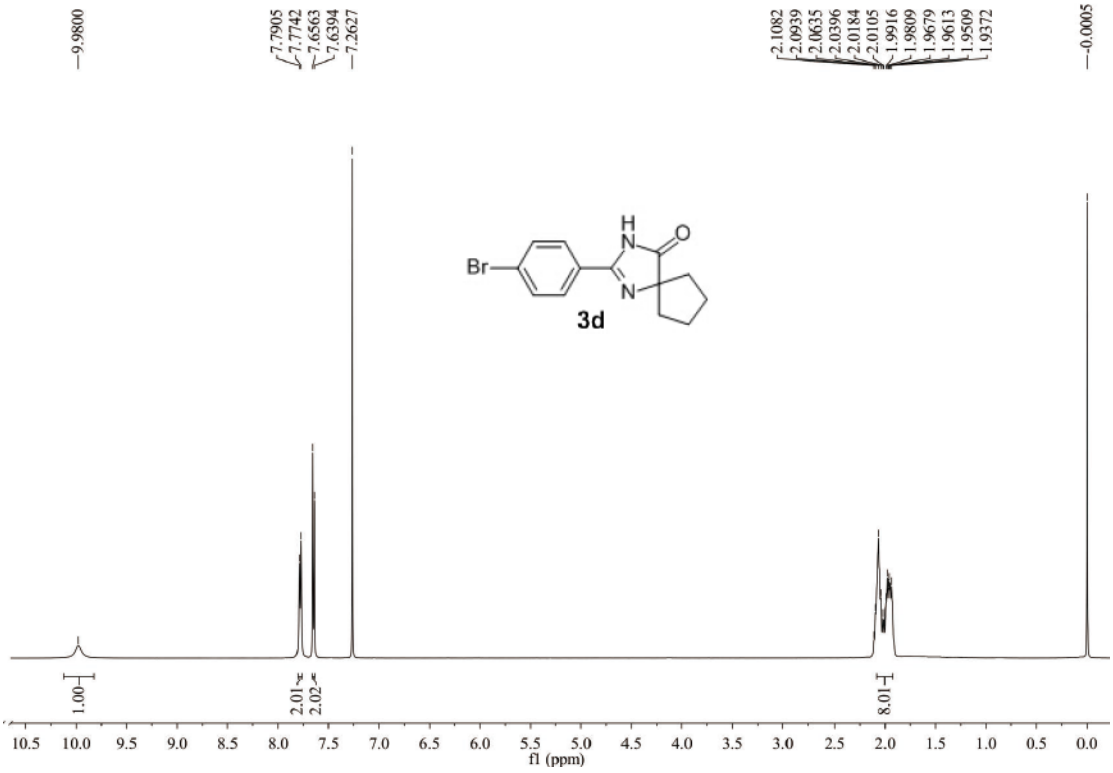
H
N
O
N
3c
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)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2.0184
2.0105
1.9916
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1.9679
1.9613
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Br
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N
O
N
3d
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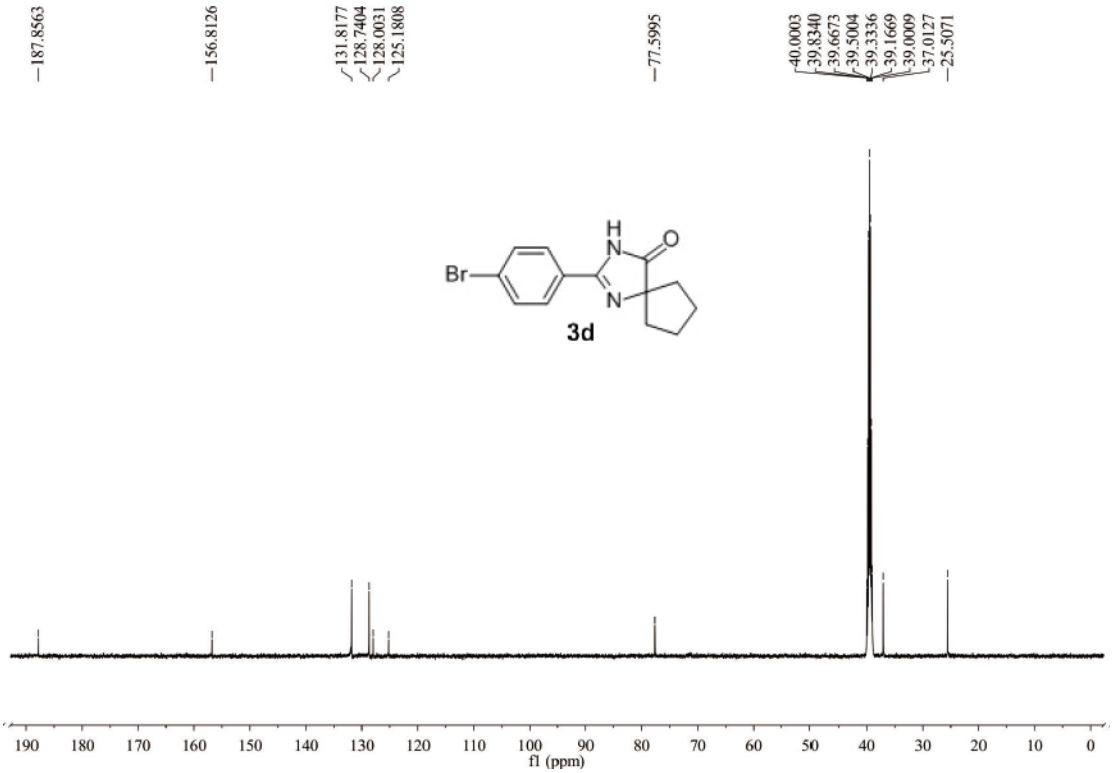
2.01
2.02
8.01
fl (ppm)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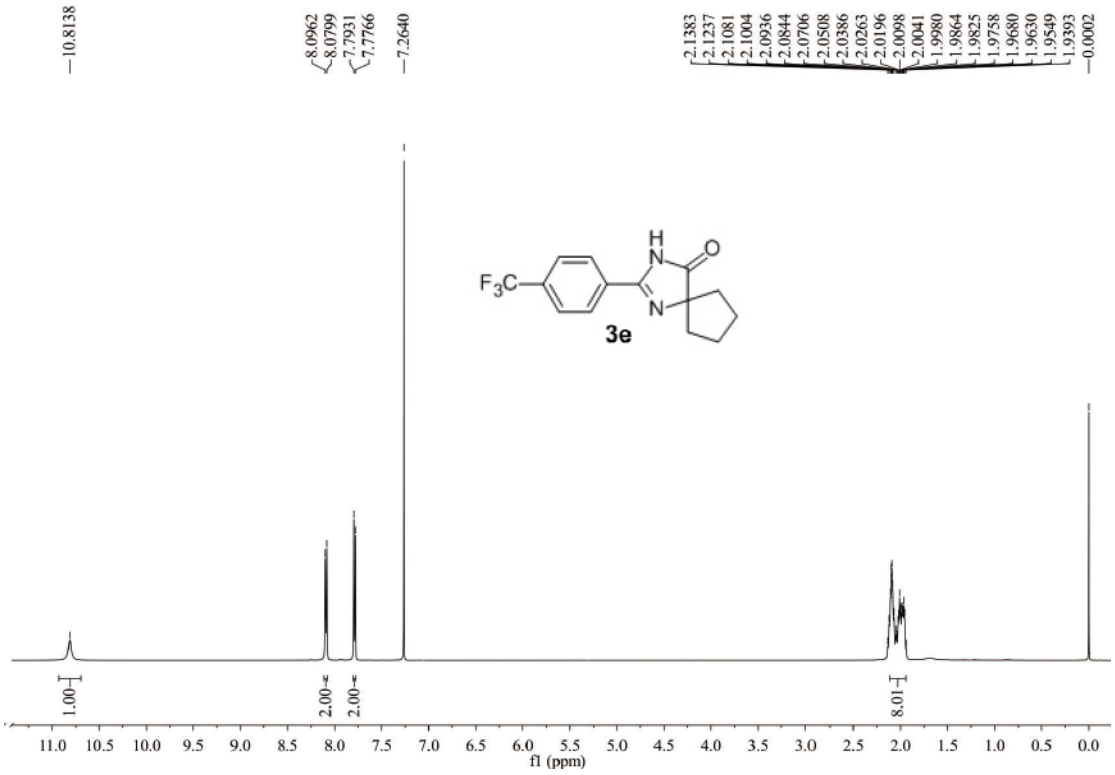
Br
H
N
O
N
3d
fl (ppm)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2.0098
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O
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2.00
2.00
8.01
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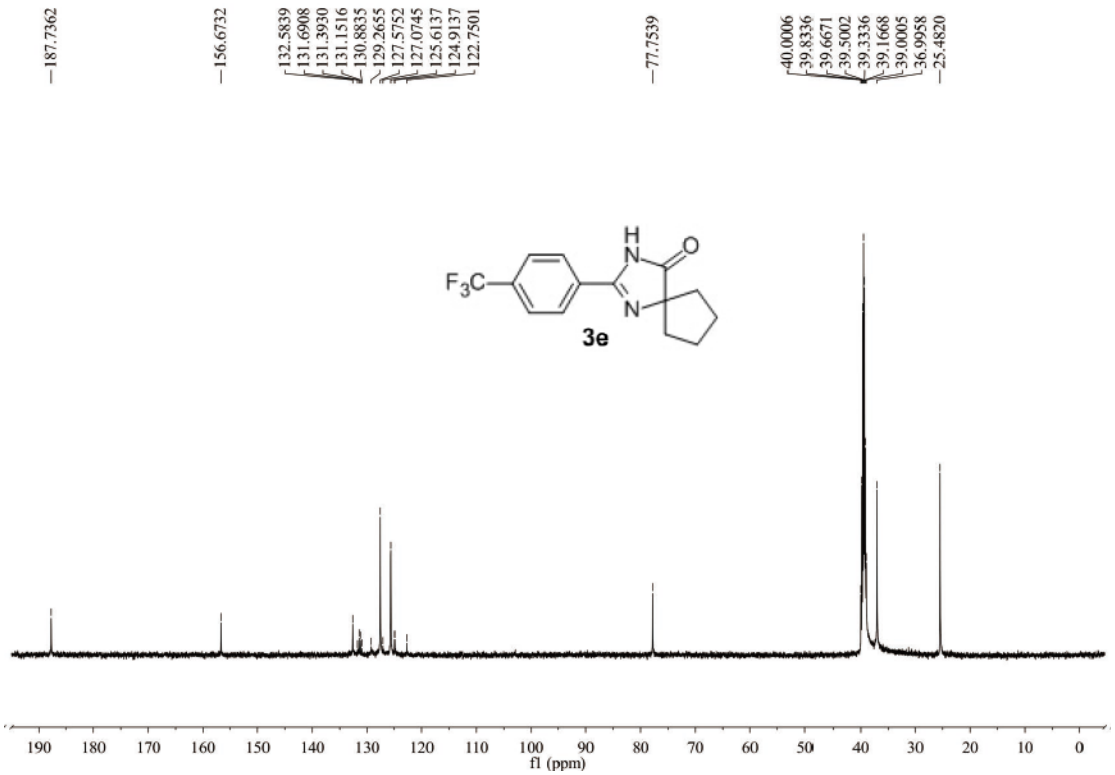
f1 (ppm)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N
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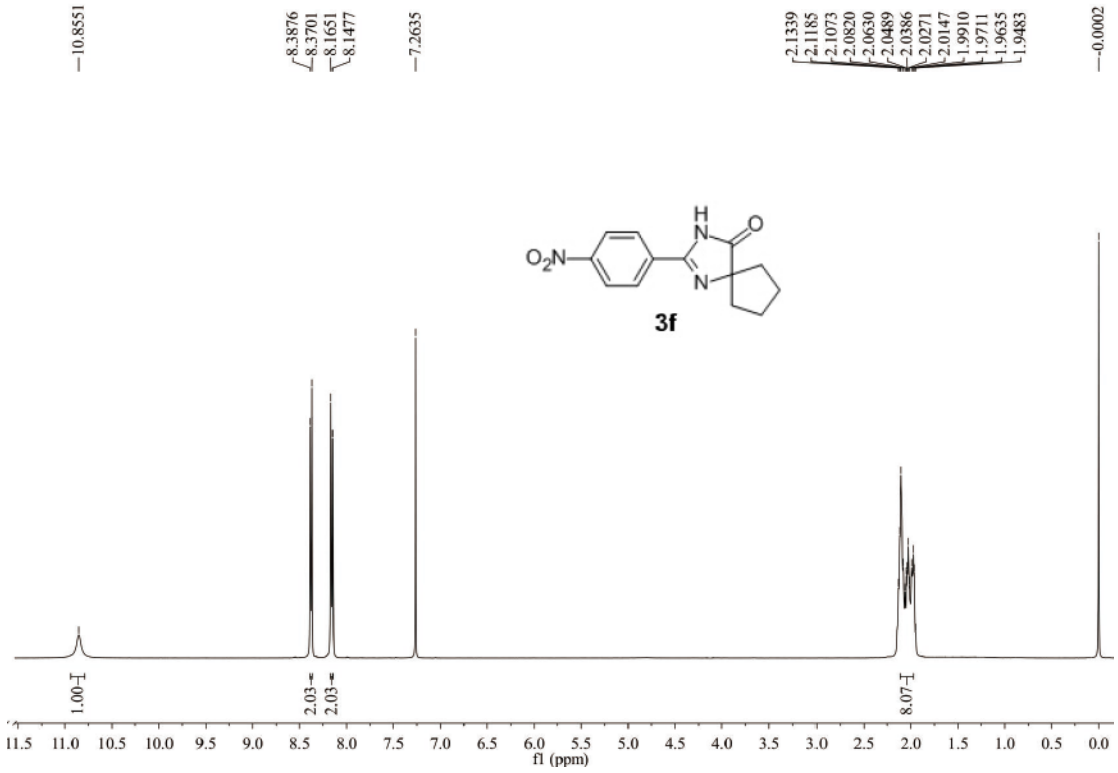
f1 (ppm)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2.0386
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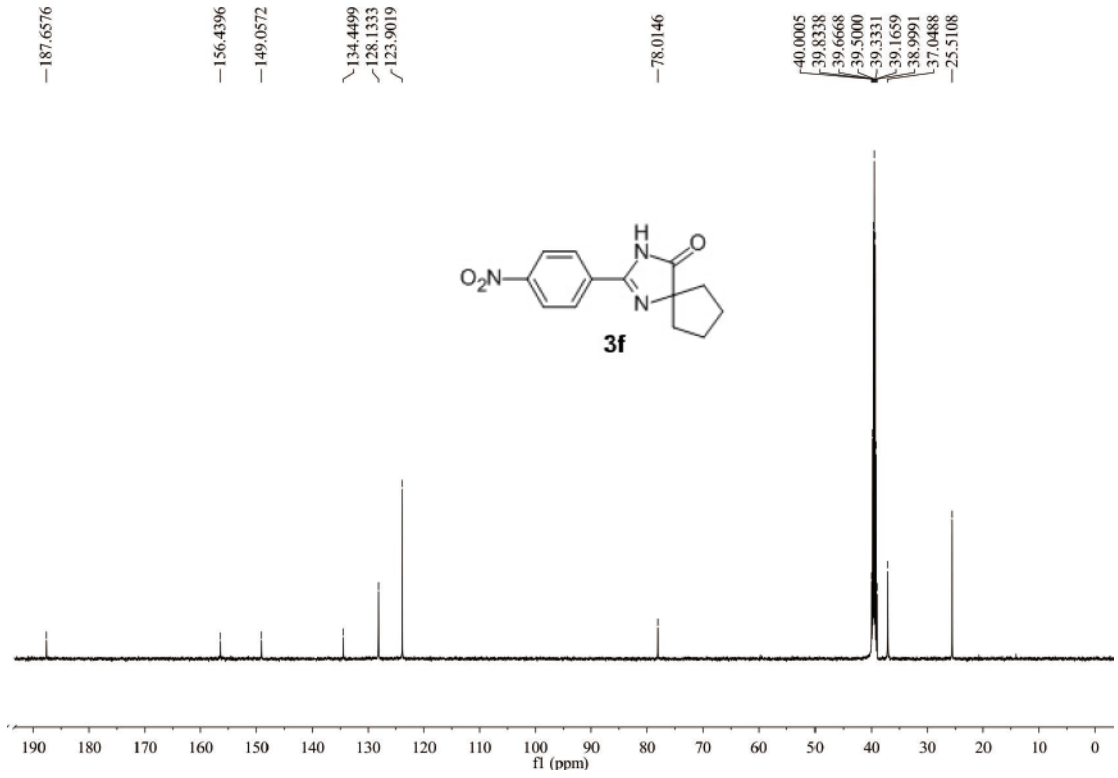
f1 (ppm)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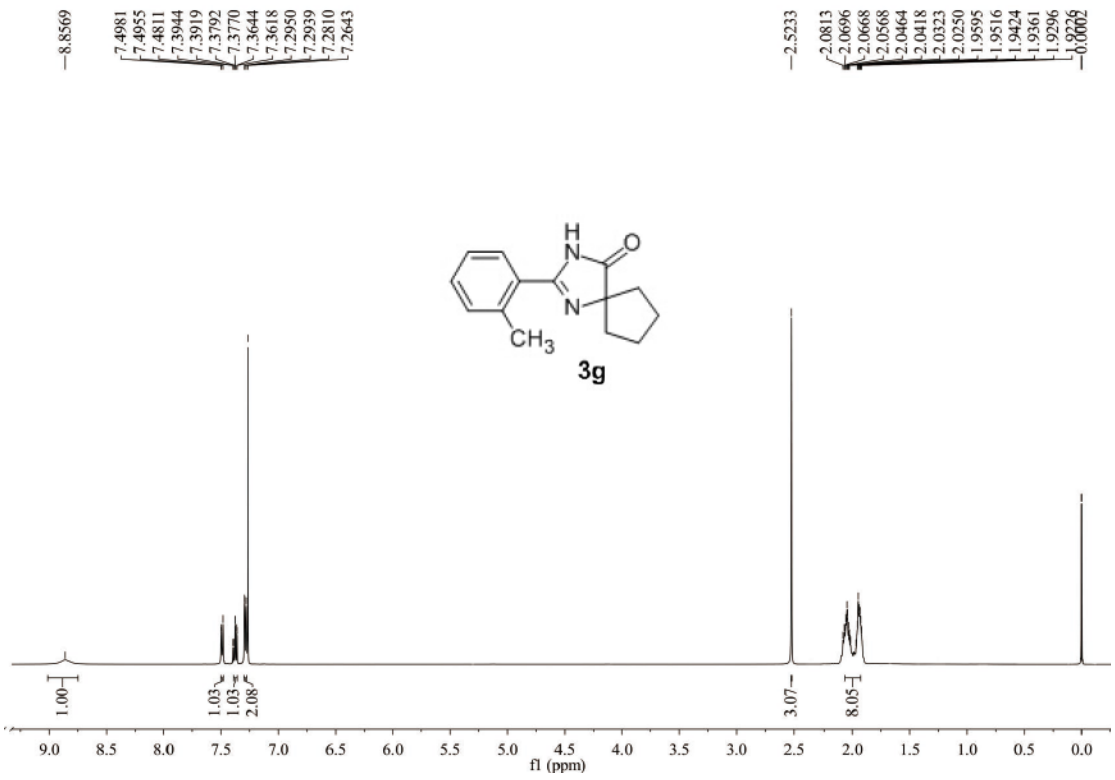
f1 (ppm)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7.2643
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2.0464
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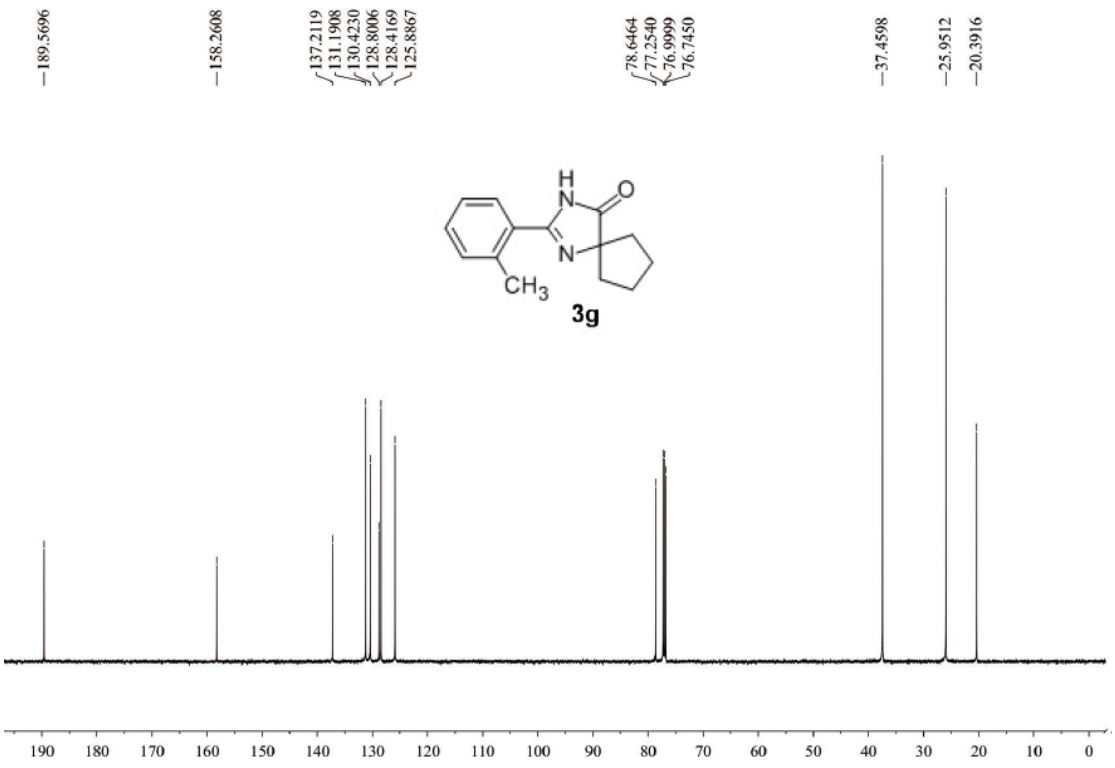
f1 (ppm)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76.9999
76.7450
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20.3916
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O
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CH3
3g
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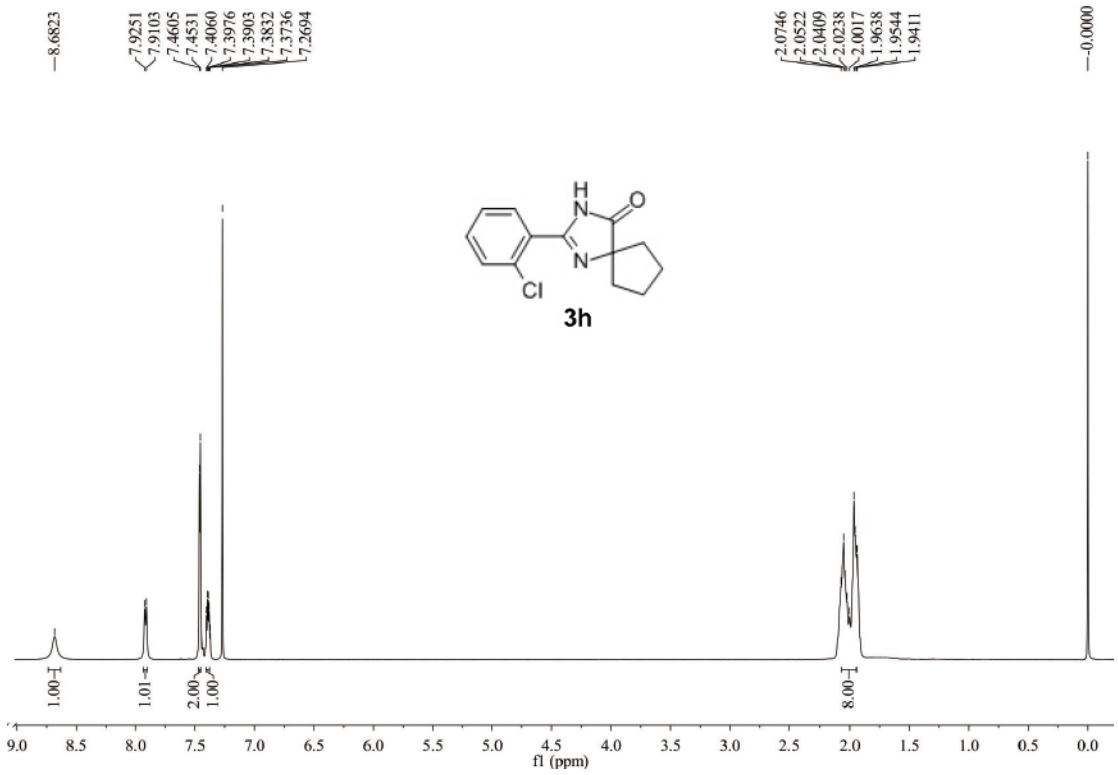
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2.0522
2.0409
2.0238
2.0017
1.9638
1.9544
1.9411
0.0000
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N
O
N
Cl
3h
1.00
1.01
2.00
1.00
8.00
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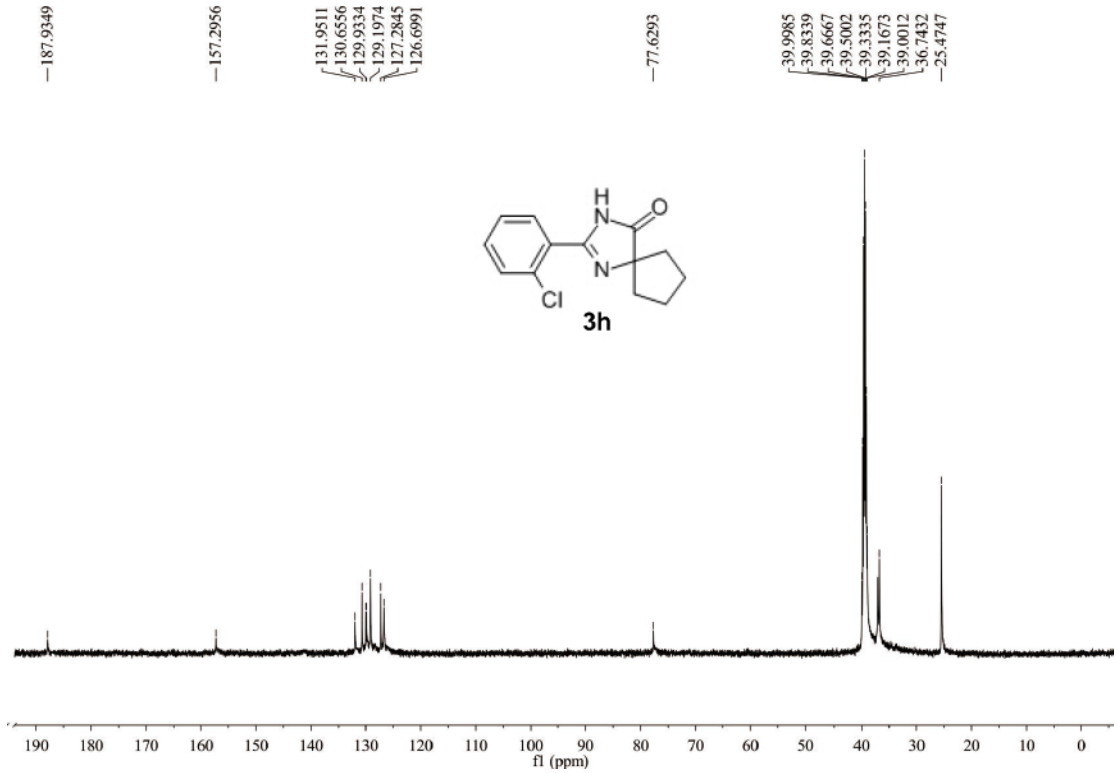
f1 (ppm)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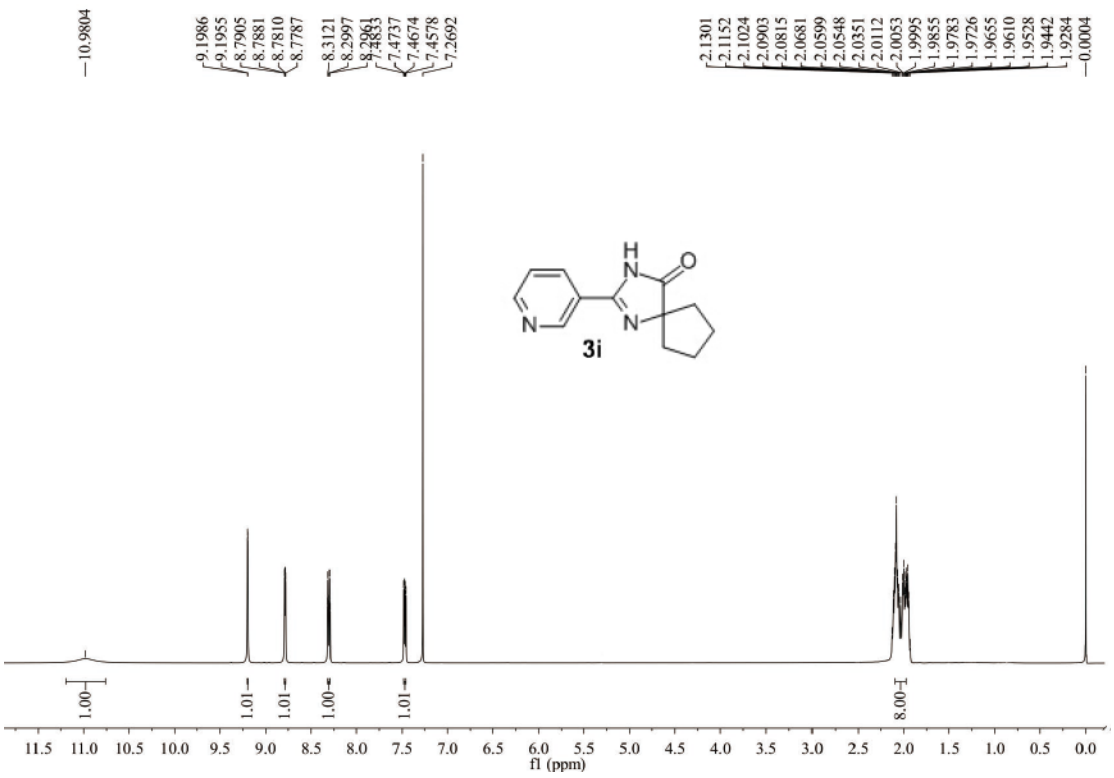
f1 (ppm)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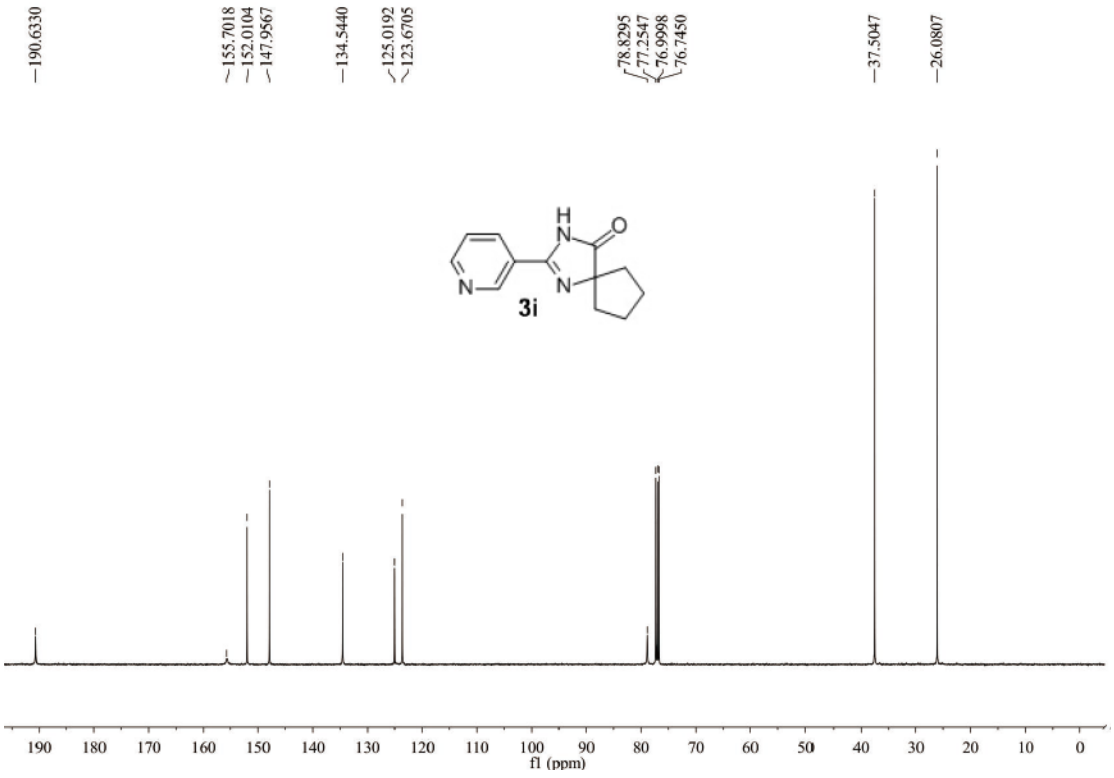
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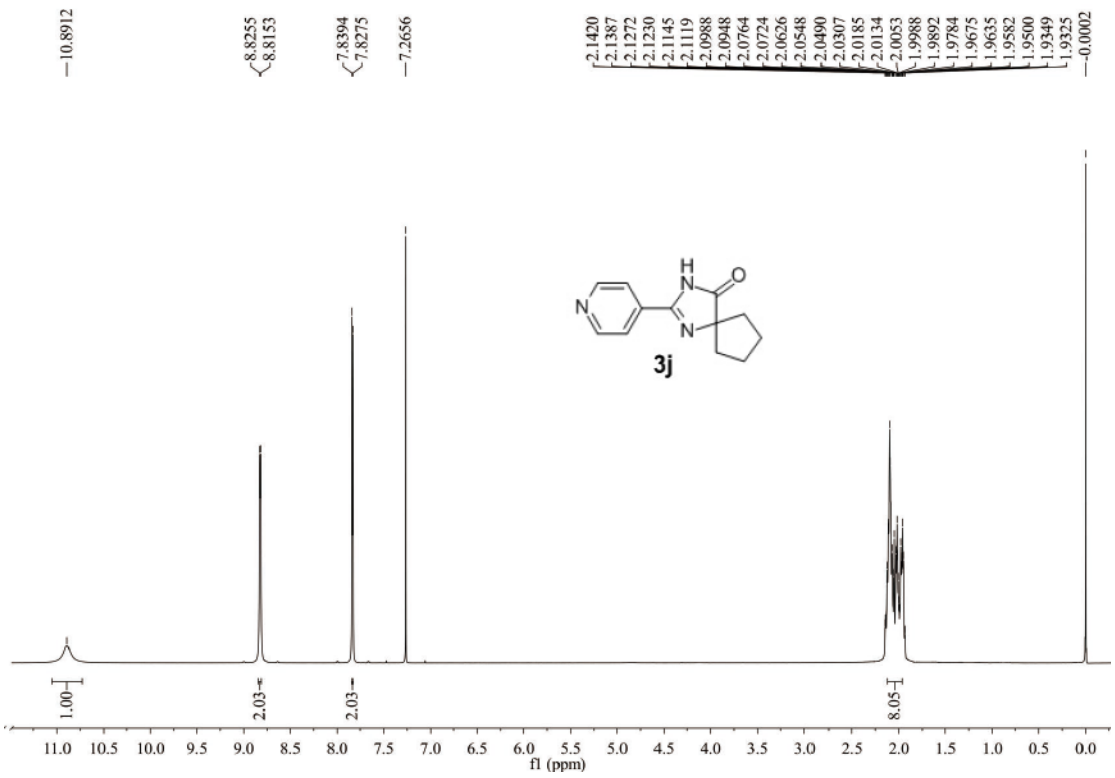
f1 (ppm)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2.0626
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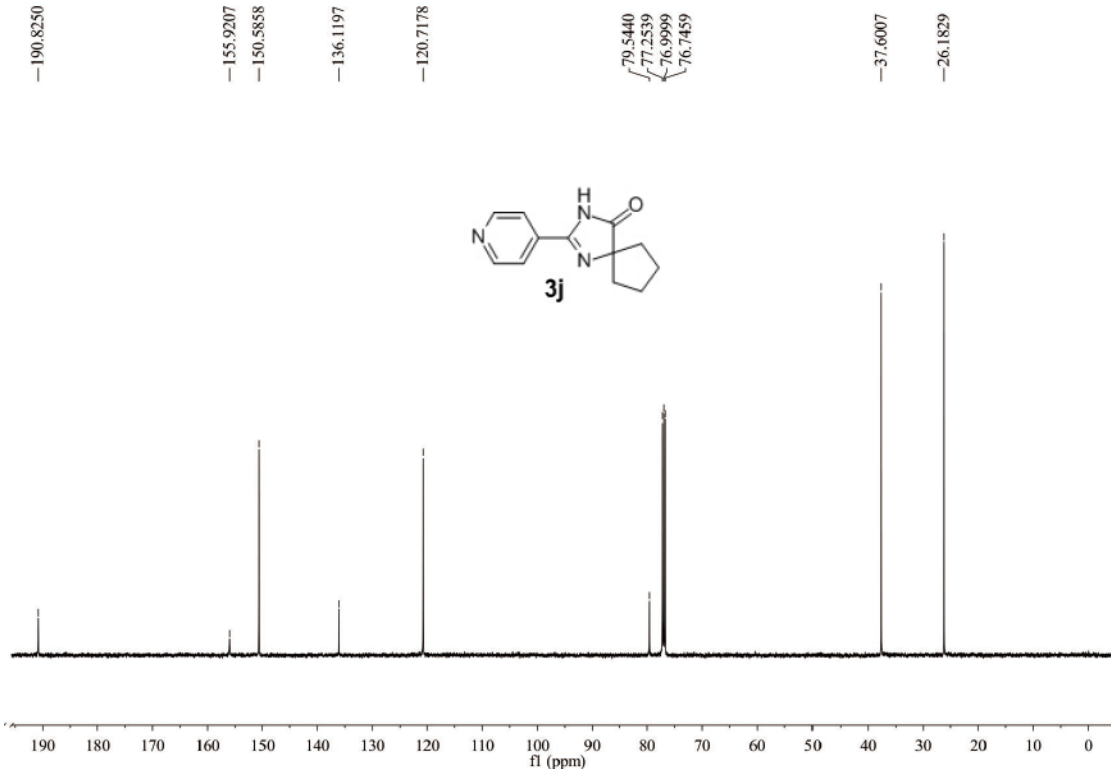
2.0490
2.0307
2.0185
2.0134
2.0053
1.9988
1.9892
1.9784
1.9675
1.9635
1.9582
1.9500
1.9349
1.9325
0.0002
3j
1.00
2.03
2.03
8.05
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)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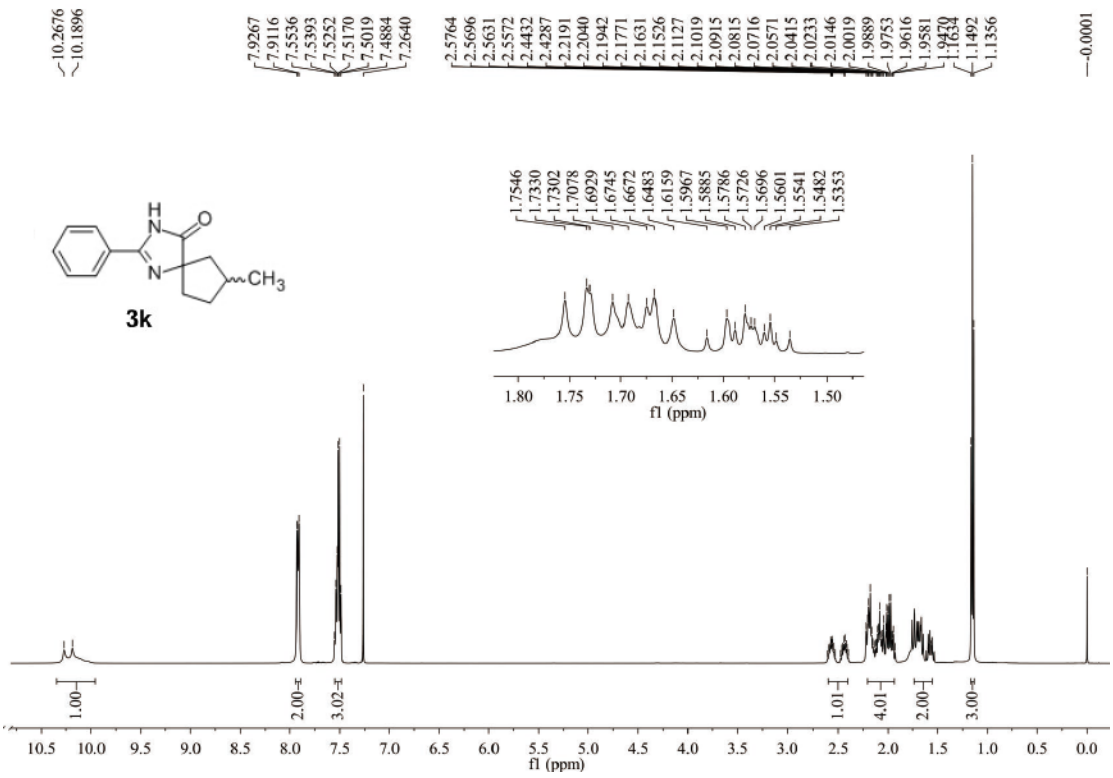
76.7459
37.6007
26.1829
3j
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)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7.5252
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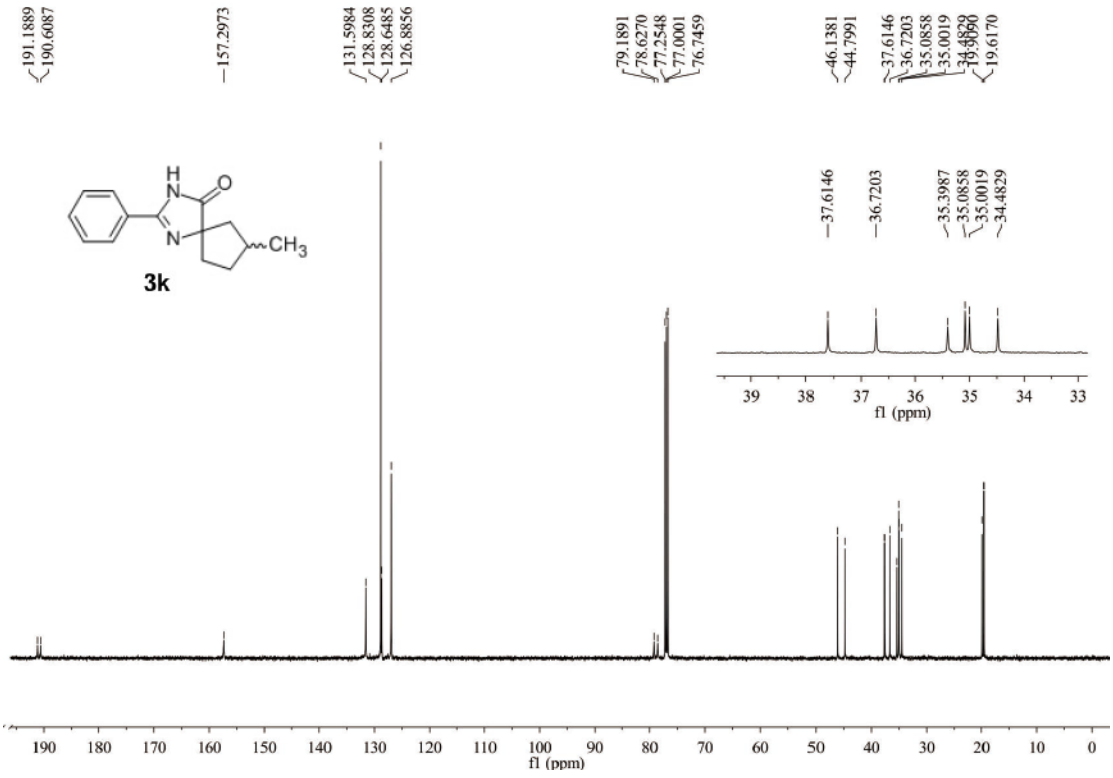
7.4884
7.2640
2.5764
2.5696
2.5631
2.5572
2.4432
2.4287
2.2191
2.2040
2.1942
2.1771
2.1631
2.1526
2.1127
2.1019
2.0915
2.0815
2.0716
2.0571
2.0415
2.0233
2.0146
2.0019
1.9889
1.9753
1.9616
1.9581
1.9420
1.1634
1.1492
1.1356
-0.0001
1.7546
1.7330
1.7302
1.7078
1.6929
1.6745
1.6672
1.6483
1.6159
1.5967
1.5885
1.5786
1.5726
1.5696
1.5601
1.5541
1.5482
1.5353
3k
1.80 1.75 1.70 1.65 1.60 1.55 1.50
f1 (ppm)
1.00
2.00
3.02
1.01
4.01
2.00
3.00
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)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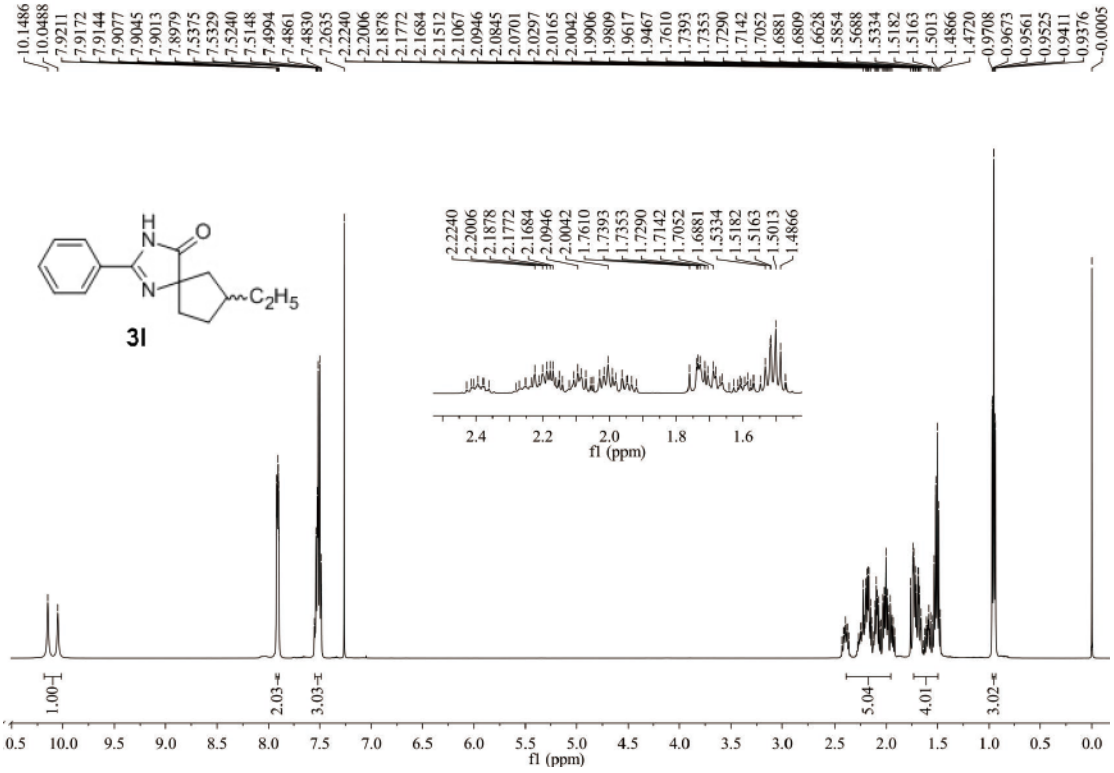
46.1381
44.7991
37.6146
36.7203
35.0858
35.0019
34.4829
19.6170
3k
39 38 37 36 35 34 33
f1 (ppm)
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)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7.5329
7.5240
7.5148
7.4994
7.4861
7.4830
7.2635
2.2240
2.2006
2.1878
2.1772
2.1684
2.1512
2.1067
2.0946
2.0845
2.0701
2.0297
2.0165
2.0042
1.9906
1.9809
1.9617
1.9467
1.7610
1.7393
1.7353
1.7290
1.7142
1.7052
1.6881
1.6809
1.6628
1.5854
1.5688
1.5334
1.5182
1.5163
1.5013
1.4866
1.4720
0.9708
0.9673
0.9561
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0.0005
3I
fl (ppm)
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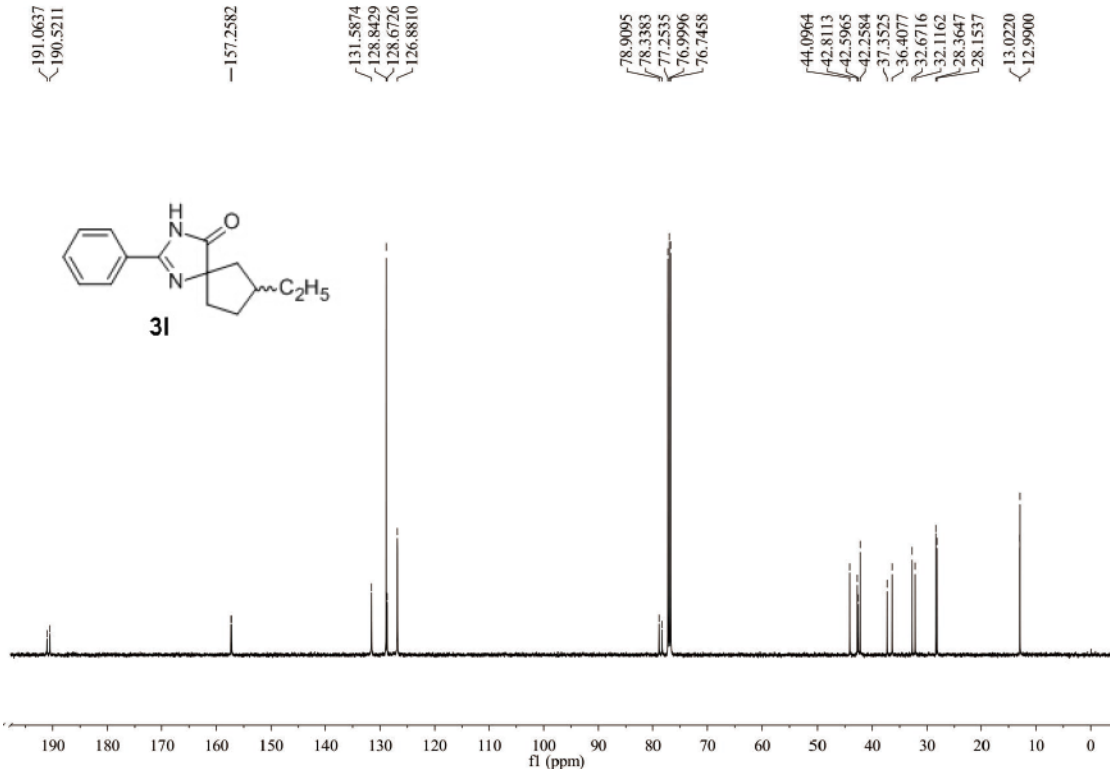
2.03
3.03
5.04
4.01
3.02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76.7458
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42.8113
42.5965
42.2584
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32.1162
28.3647
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13.0220
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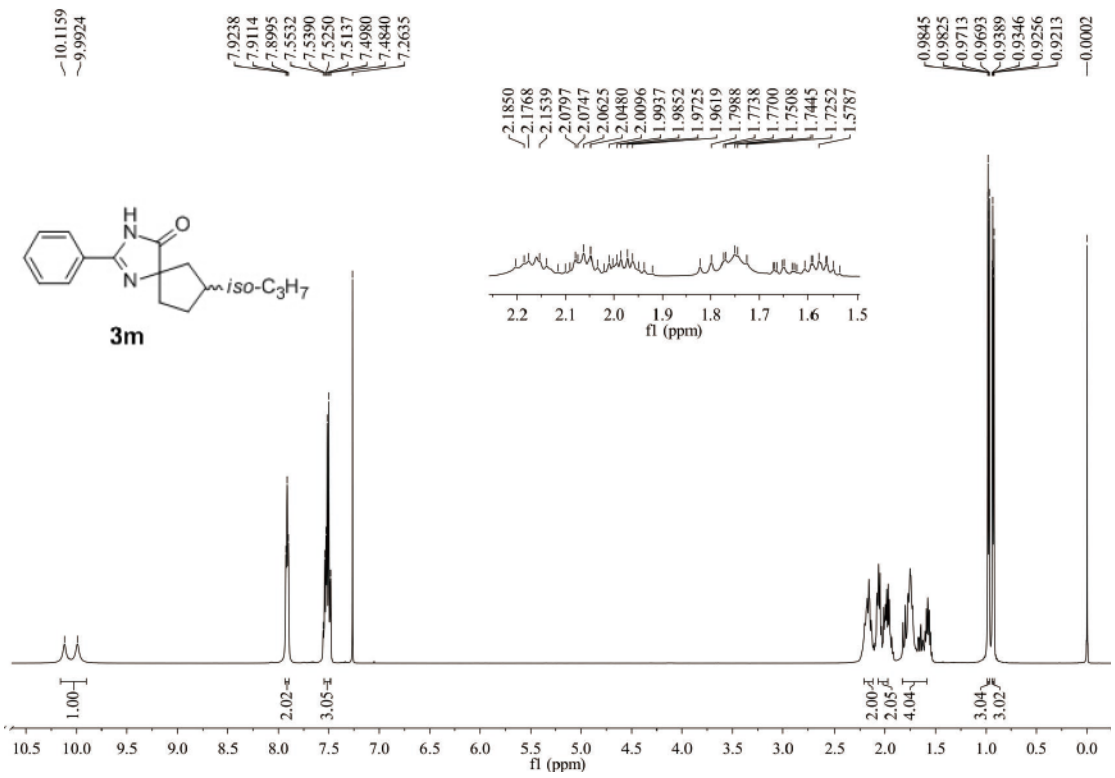
3I
fl (ppm)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7.5532
7.5390
7.5250
7.5137
7.4980
7.4840
7.2635
2.1850
2.1768
2.1539
2.0797
2.0747
2.0625
2.0480
2.0096
1.9937
1.9852
1.9725
1.9619
1.7988
1.7738
1.7700
1.7508
1.7445
1.7252
1.5787
0.9845
0.9825
0.9713
0.9693
0.9389
0.9346
0.9256
0.9213
0.0002
iso-C3H7
3m
f1 (ppm)
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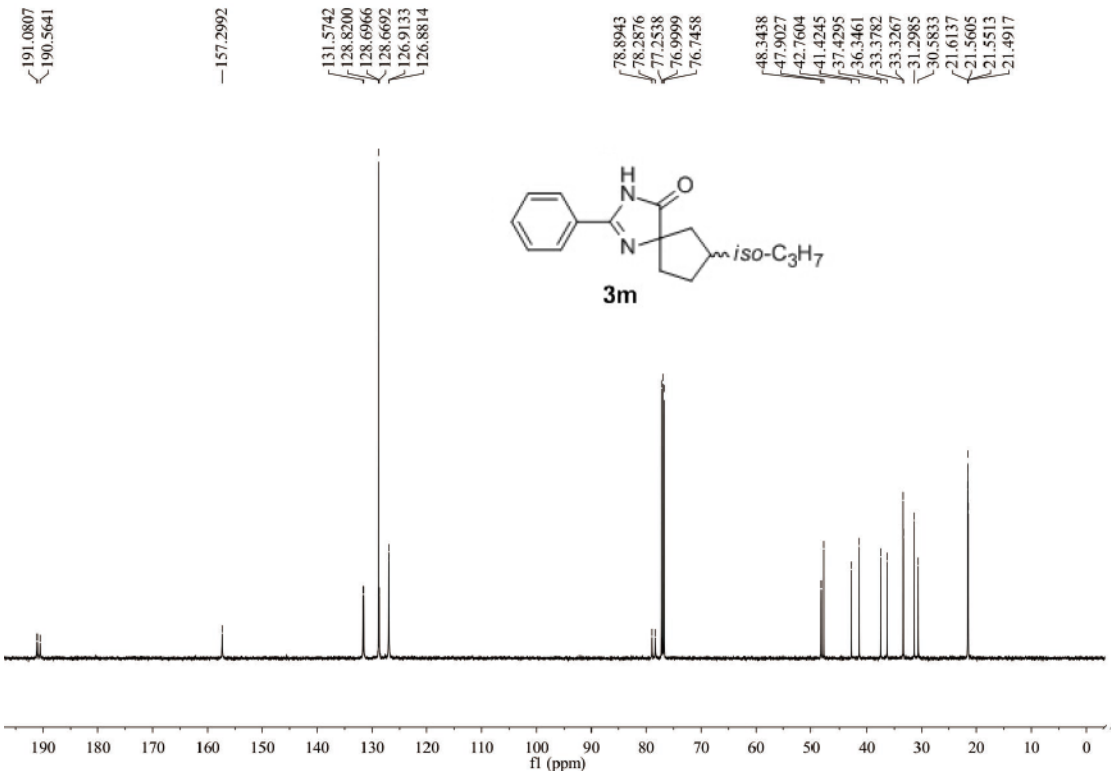
2.02
3.05
2.00
2.05
4.04
3.04
3.02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157.2992
131.5742
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78.8943
78.2876
77.2538
76.9999
76.7458
48.3438
47.9027
42.7604
41.4245
37.4295
36.3461
33.3782
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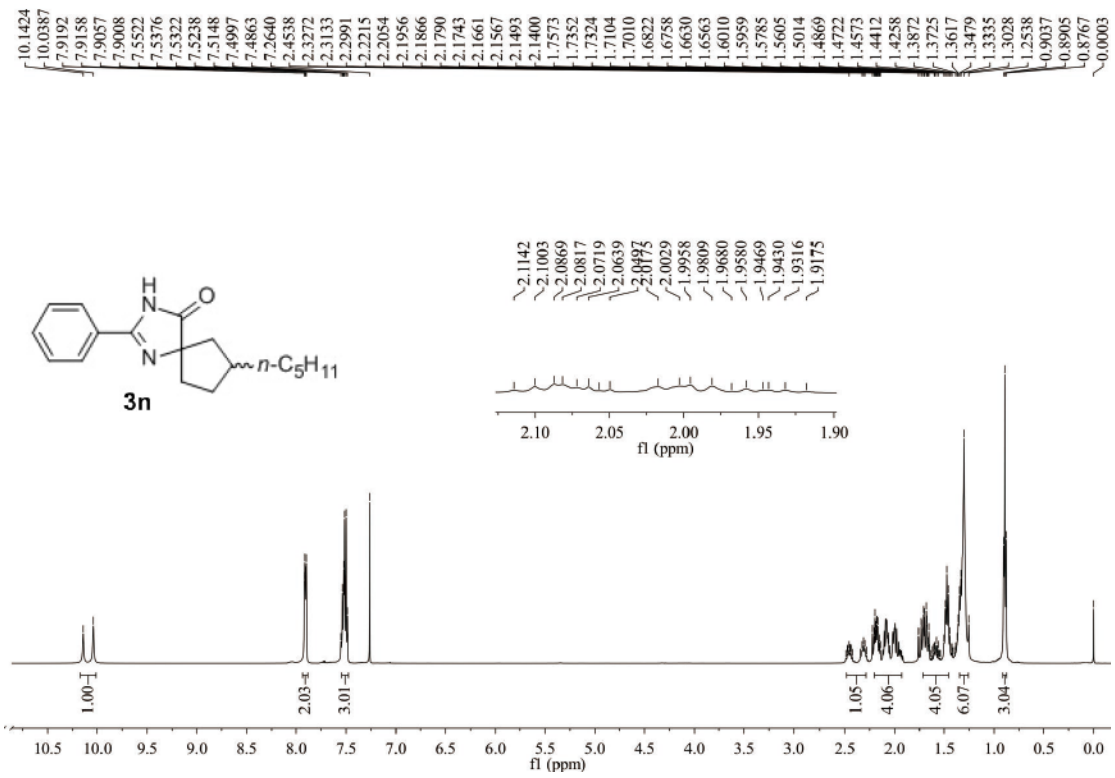
21.4917
iso-C3H7
3m
f1 (ppm)


3n
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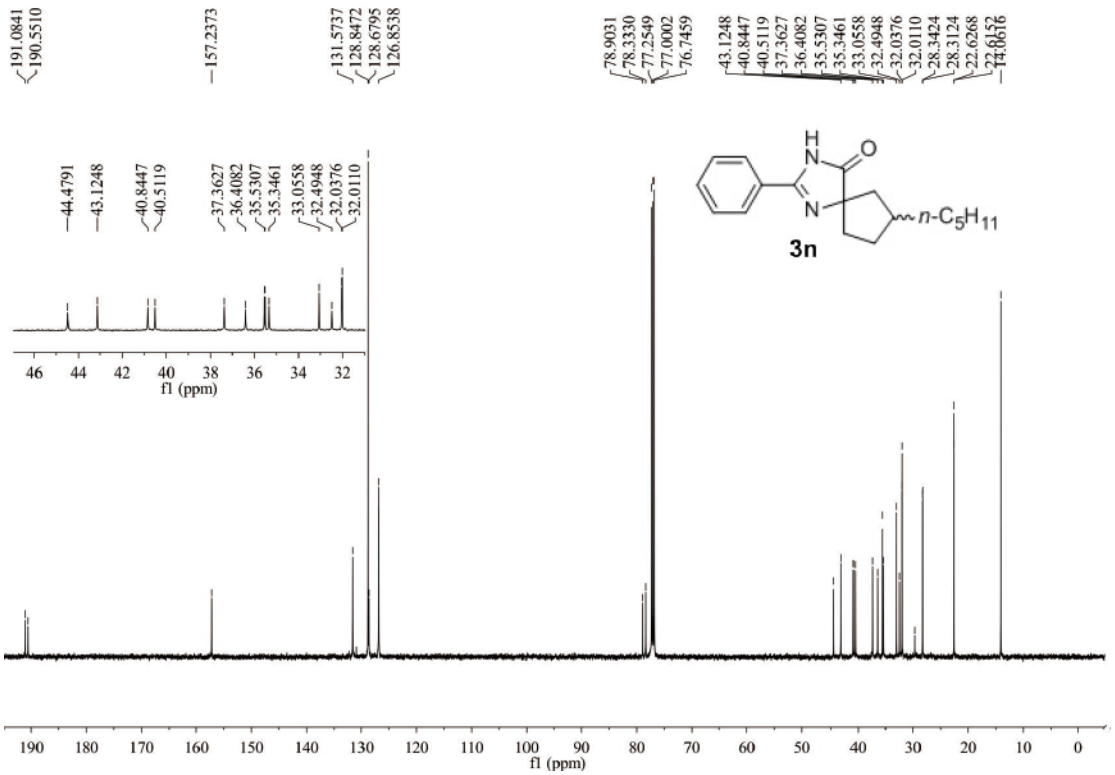
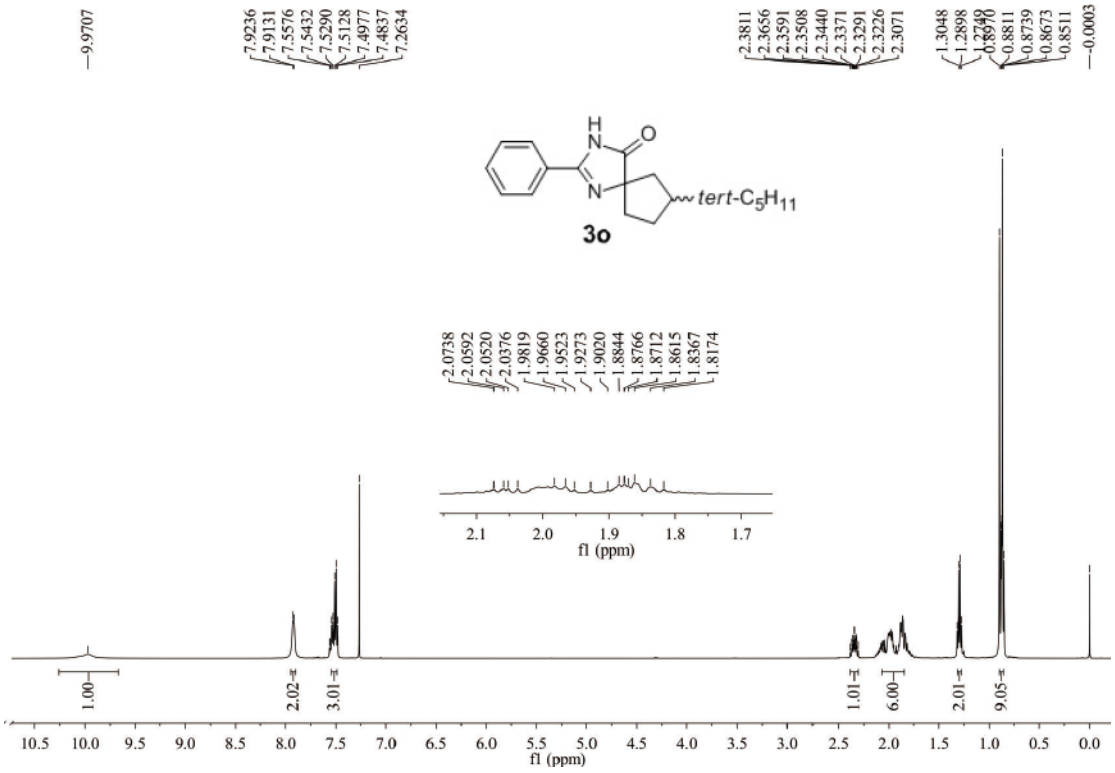
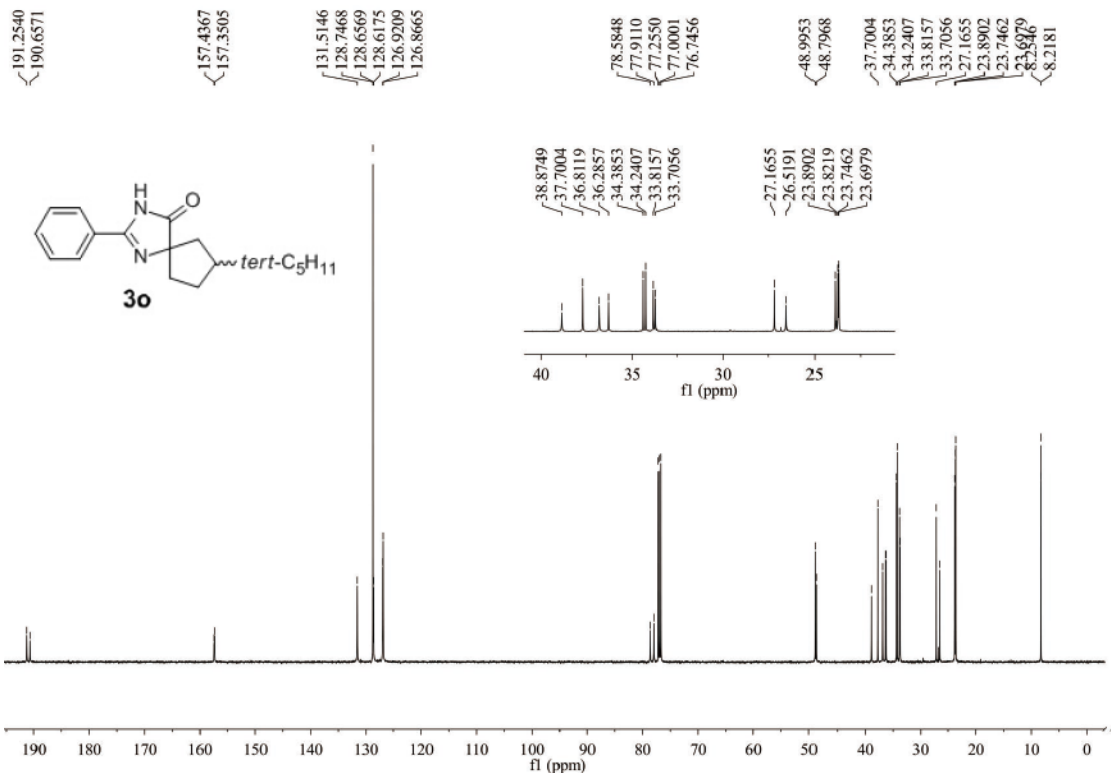
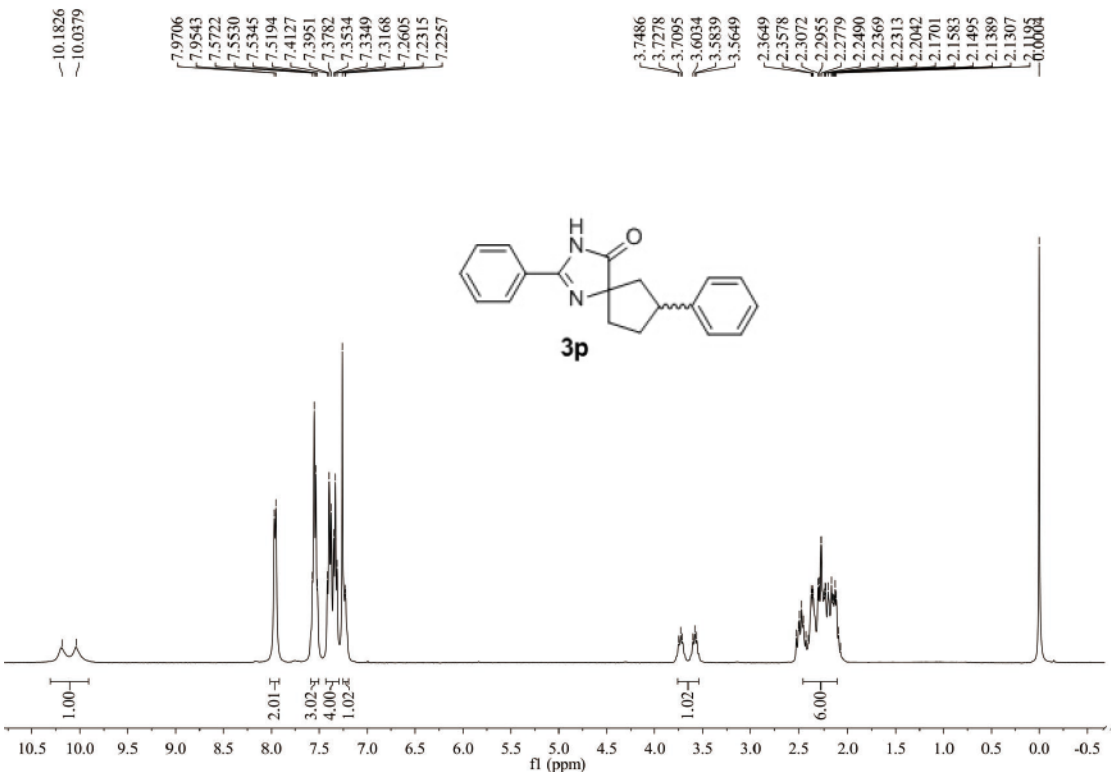
fl (ppm)


3n
n-C5H11
fl (ppm)


3o
tert-C5H11
fl (ppm)


3o
tert-C5H11
fl (ppm)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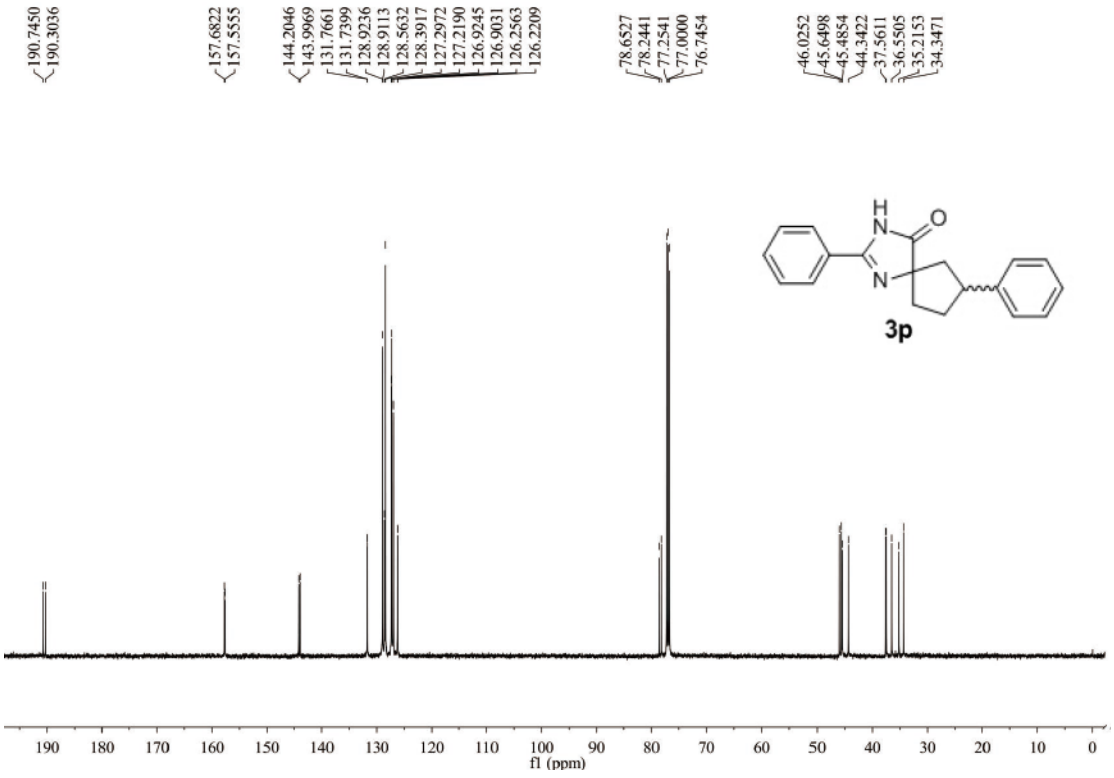
7.3782
7.3534
7.3349
7.3168
7.2605
7.2315
7.2257
3.7486
3.7278
3.7095
3.6034
3.5839
3.5649
2.3649
2.3578
2.3072
2.2955
2.2779
2.2490
2.2369
2.2313
2.2042
2.1701
2.1583
2.1495
2.1389
2.1307
0.0004
3p
1.00
2.01
3.02
4.00
1.02
1.02
6.00
fl (ppm)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157.6822
157.5555
144.2046
143.9969
131.7661
131.7399
128.9236
128.9113
128.5632
128.3917
127.2972
127.2190
126.9245
126.9031
126.2563
126.2209
78.6527
78.2441
77.2541
77.0000
76.7454
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37.5611
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35.2153
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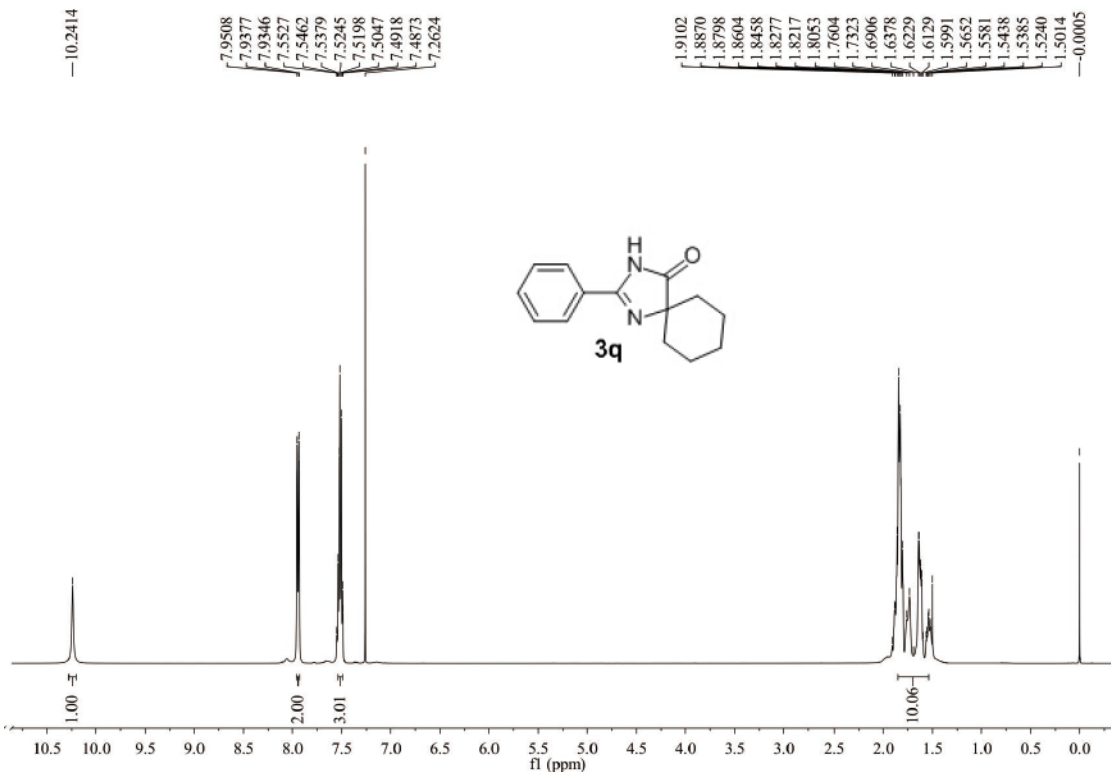
3p
fl (ppm)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7.5462
7.5379
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7.5198
7.5047
7.4918
7.4873
7.2624
1.9102
1.8870
1.8798
1.8604
1.8458
1.8277
1.8217
1.8053
1.7604
1.7323
1.6906
1.6378
1.6229
1.6129
1.5991
1.5652
1.5581
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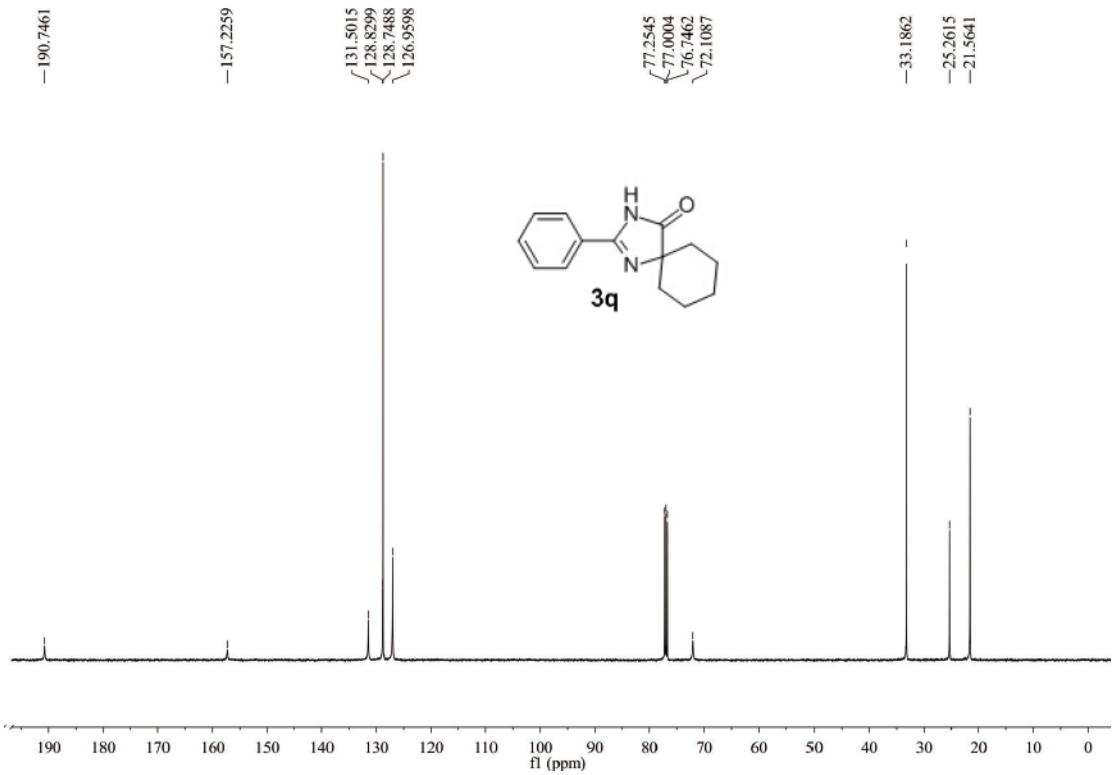
1.5240
1.5014
0.0005
3q
1.00
2.00
3.01
10.06
fl (ppm)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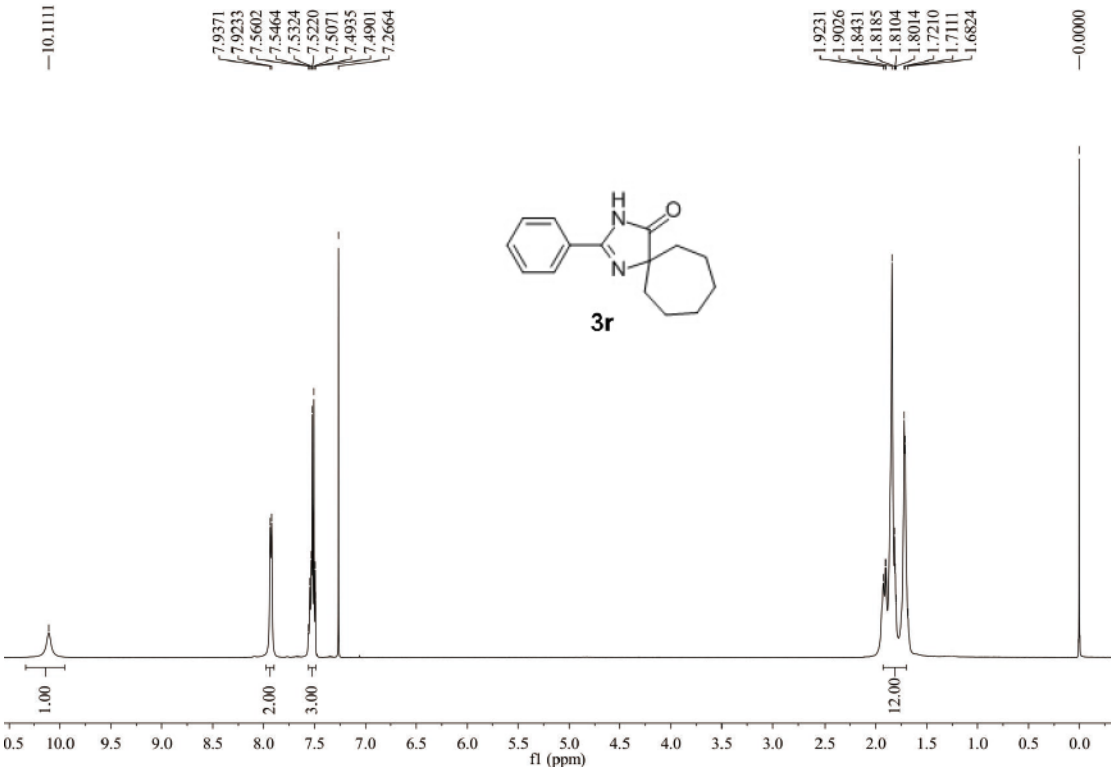
128.7488
126.9598
77.2545
77.0004
76.7462
72.1087
33.1862
25.2615
21.5641
3q
fl (ppm)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7.4901
7.2664
1.9231
1.9026
1.8431
1.8185
1.8104
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1.7111
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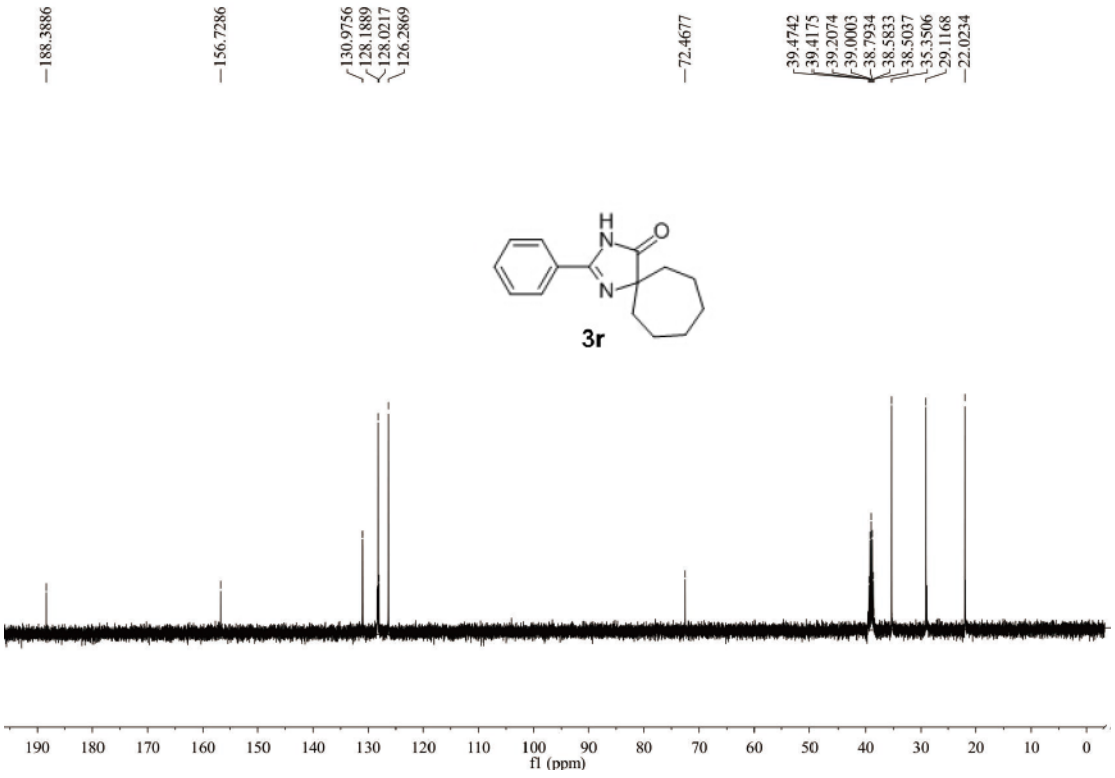
1.00
2.00
3.00
12.00
fl (ppm)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29.1168
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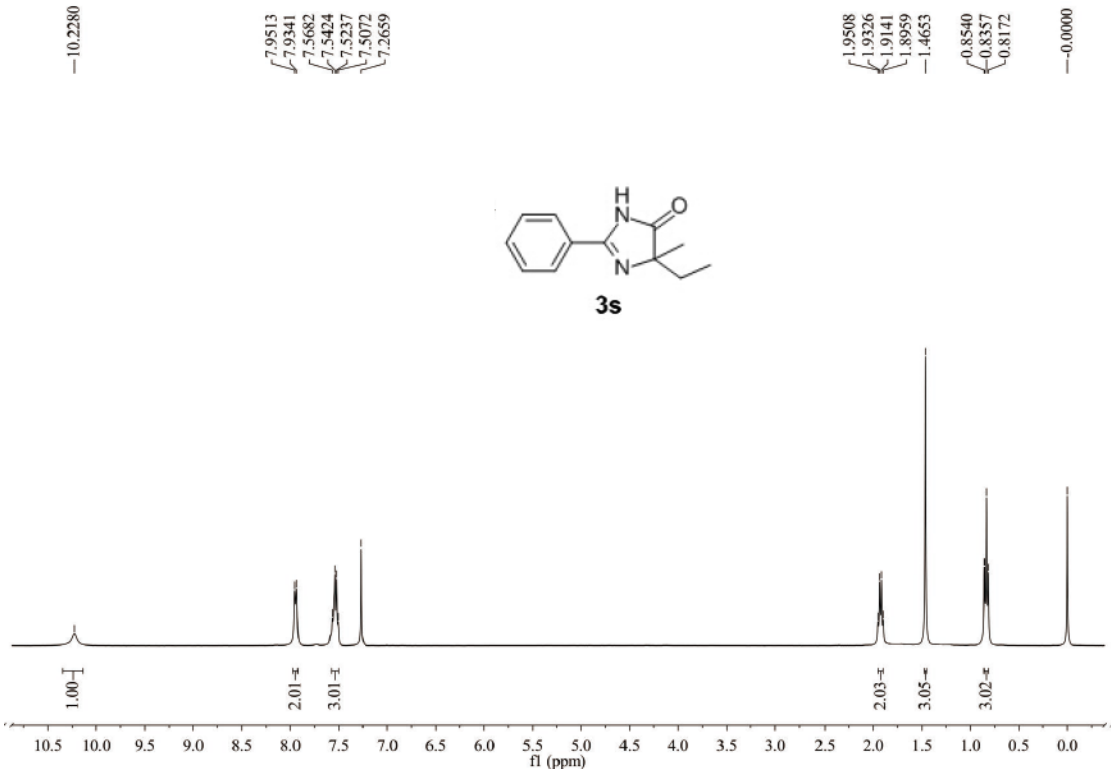
3r
fl (ppm)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1.9141
1.8959
1.4653
0.8540
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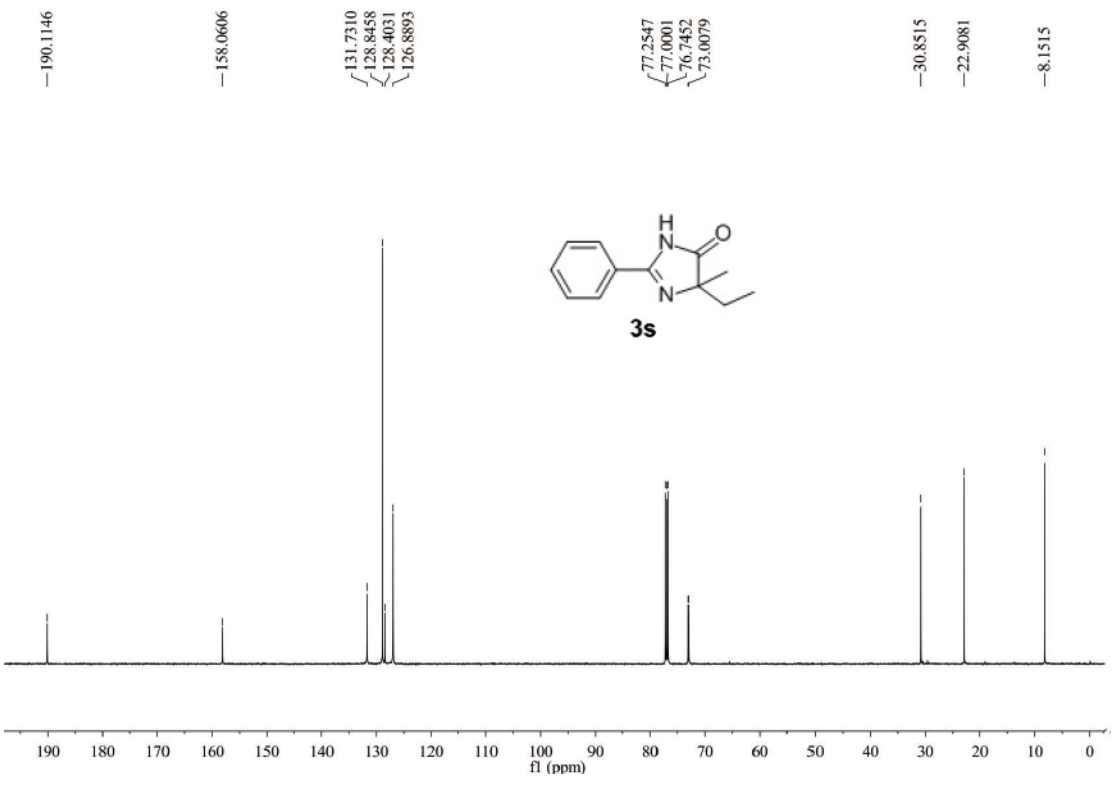
0.0000
3s
1.00
2.01
3.01
2.03
3.05
3.02
f1 (ppm)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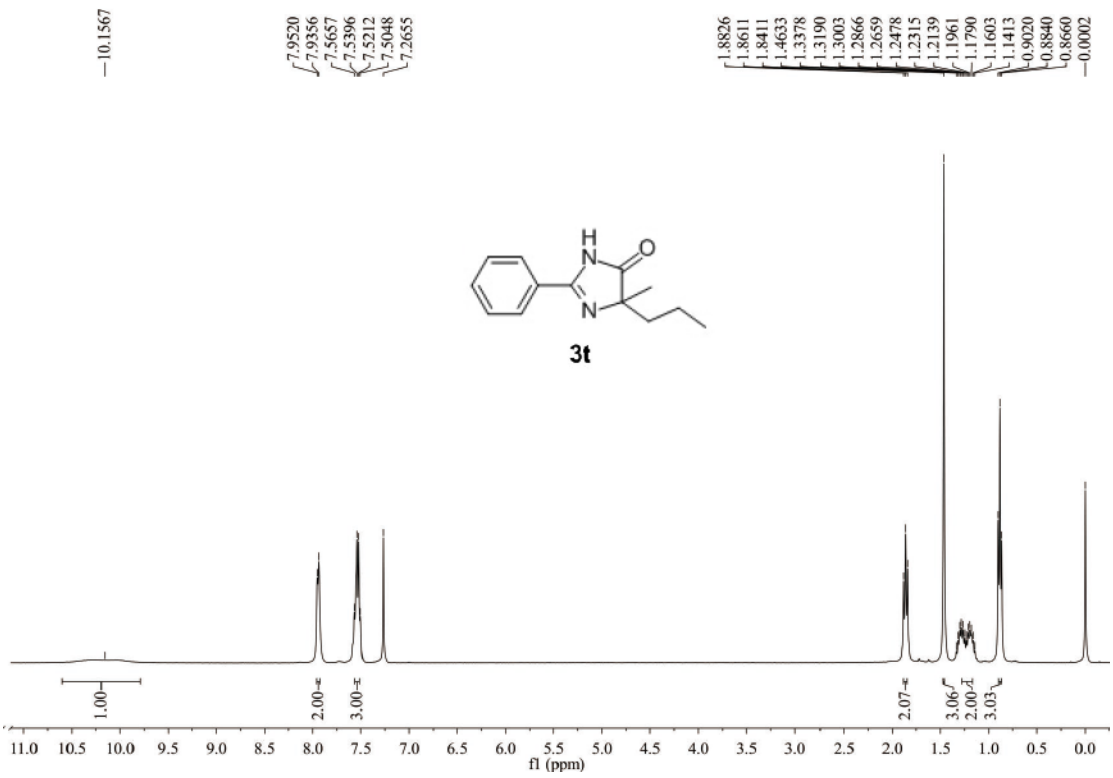
3s
f1 (ppm)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1.3003
1.2866
1.2659
1.2478
1.2315
1.2139
1.1961
1.1790
1.1603
1.1413
0.9020
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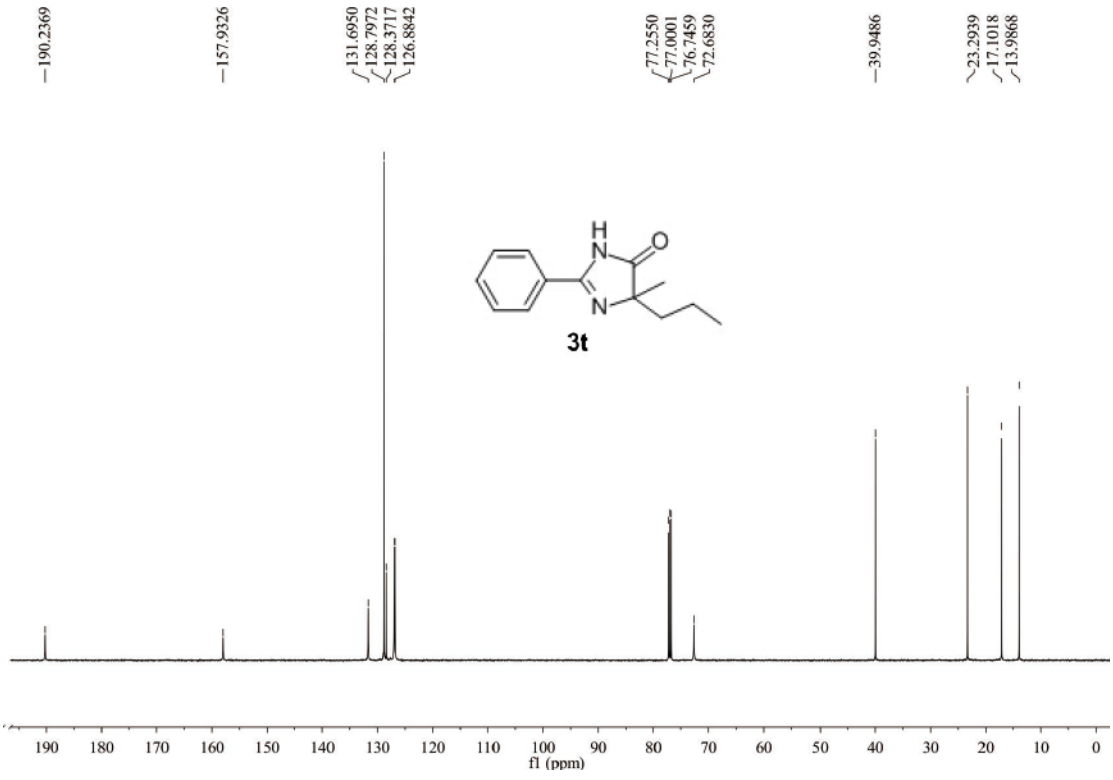
3t
1.00
2.00
3.00
2.07
3.06
2.00
3.03
f1 (ppm)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76.7459
72.6830
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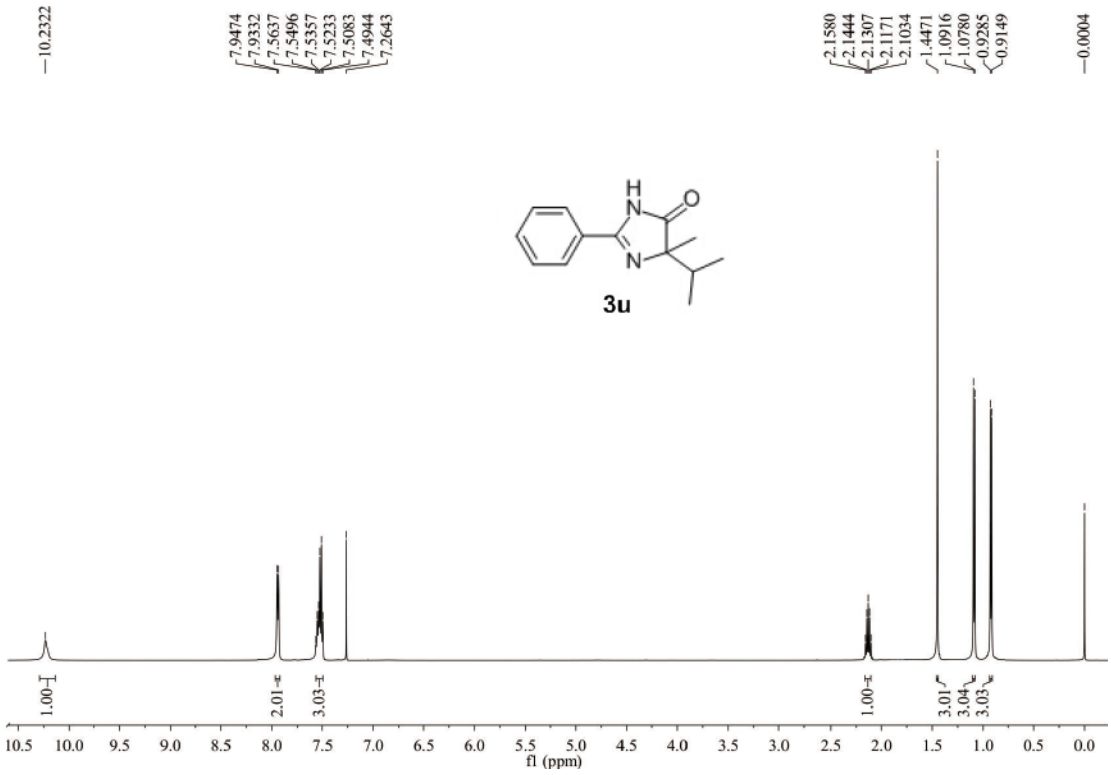
17.1018
13.9868
3t
f1 (ppm)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2.1307
2.1171
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1.0916
1.0780
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3u
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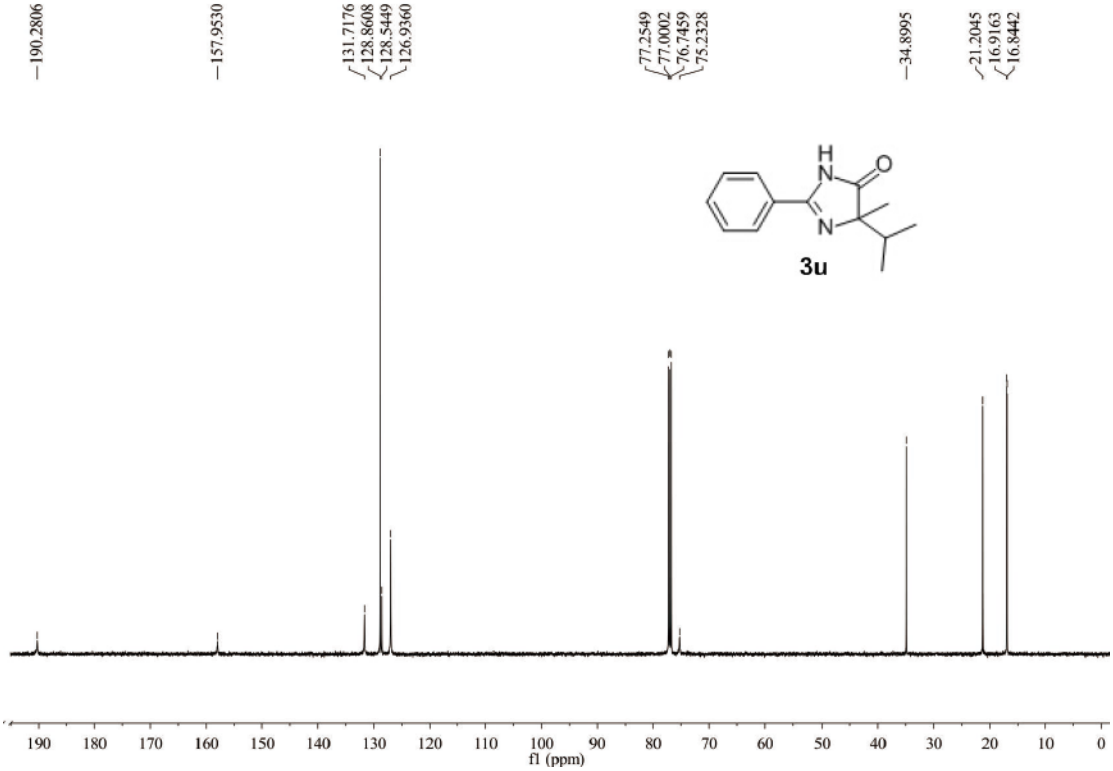
2.01
3.03
1.00
3.01
3.04
3.03
f1 (ppm)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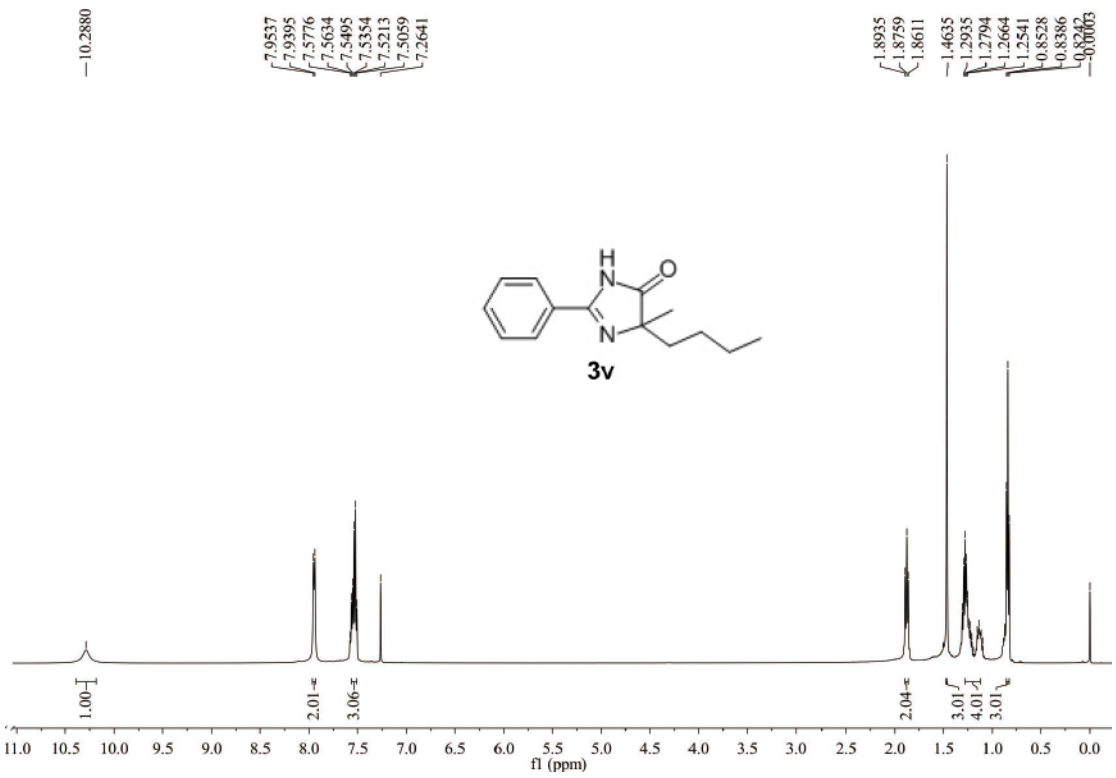
3u
f1 (ppm)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1.8759
1.8611
1.4635
1.2935
1.2794
1.2664
1.2541
0.8528
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3v
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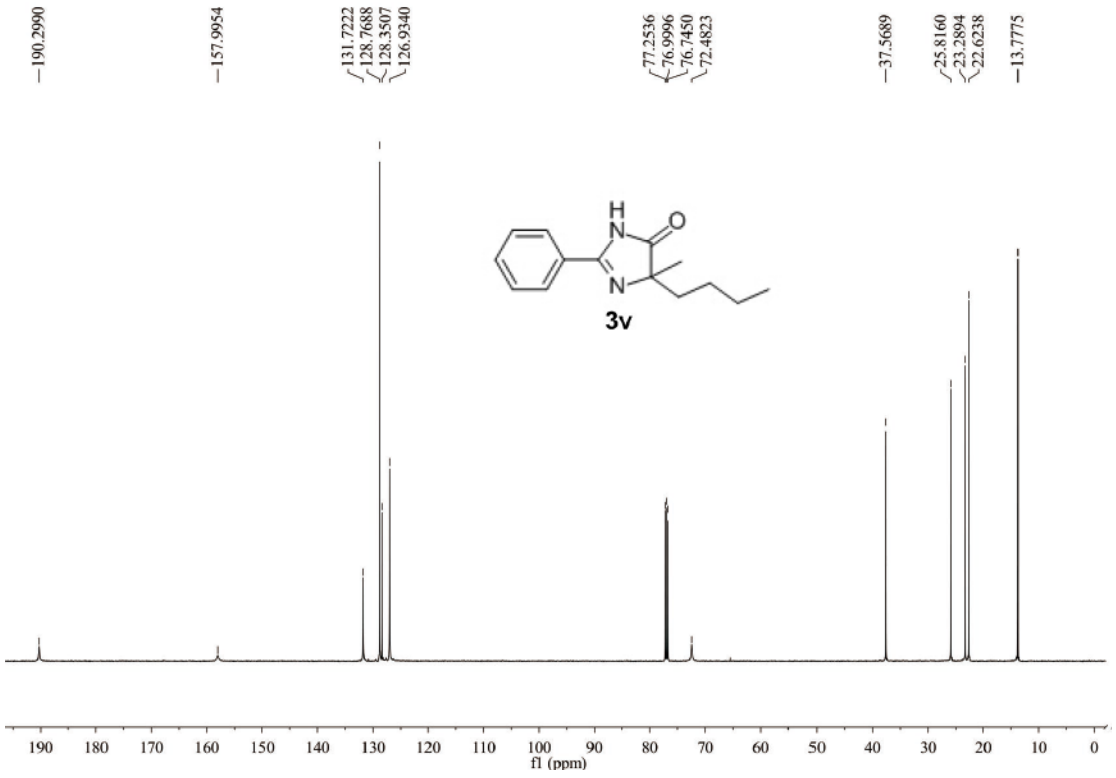
2.01
3.06
2.04
3.01
4.01
3.01
fl (ppm)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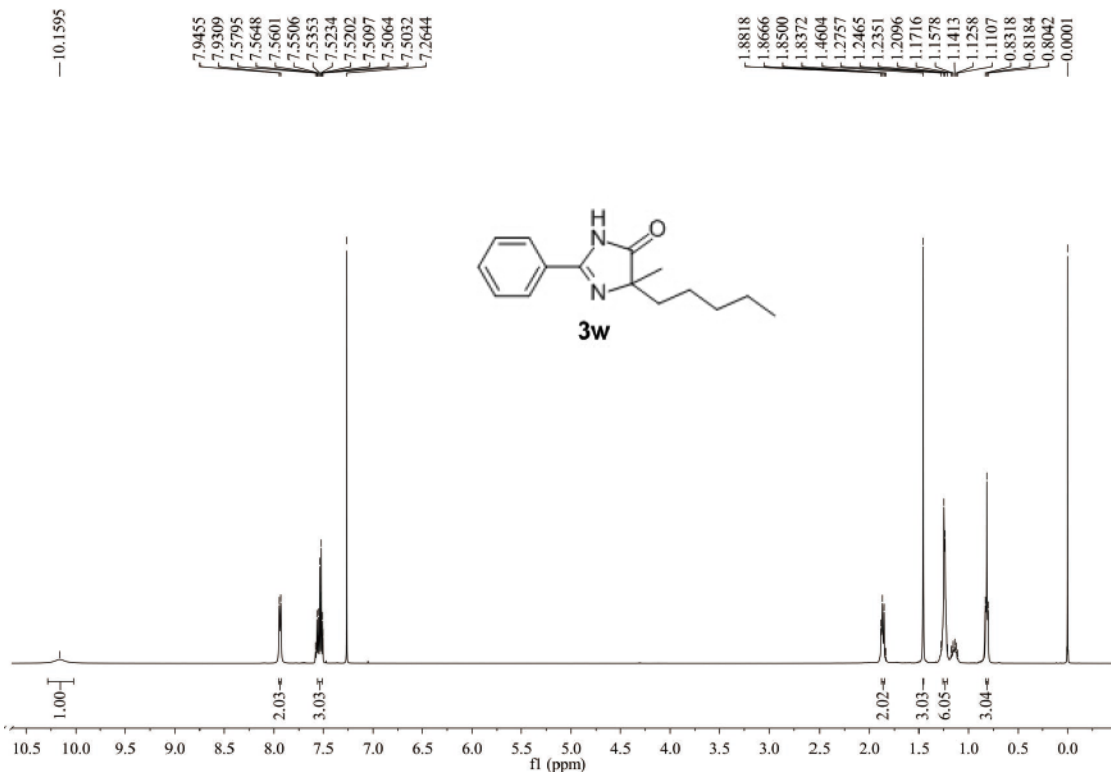
22.6238
13.7775
3v
fl (ppm)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1.1716
1.1578
1.1413
1.1258
1.1107
0.8318
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0.8042
0.0001
3w
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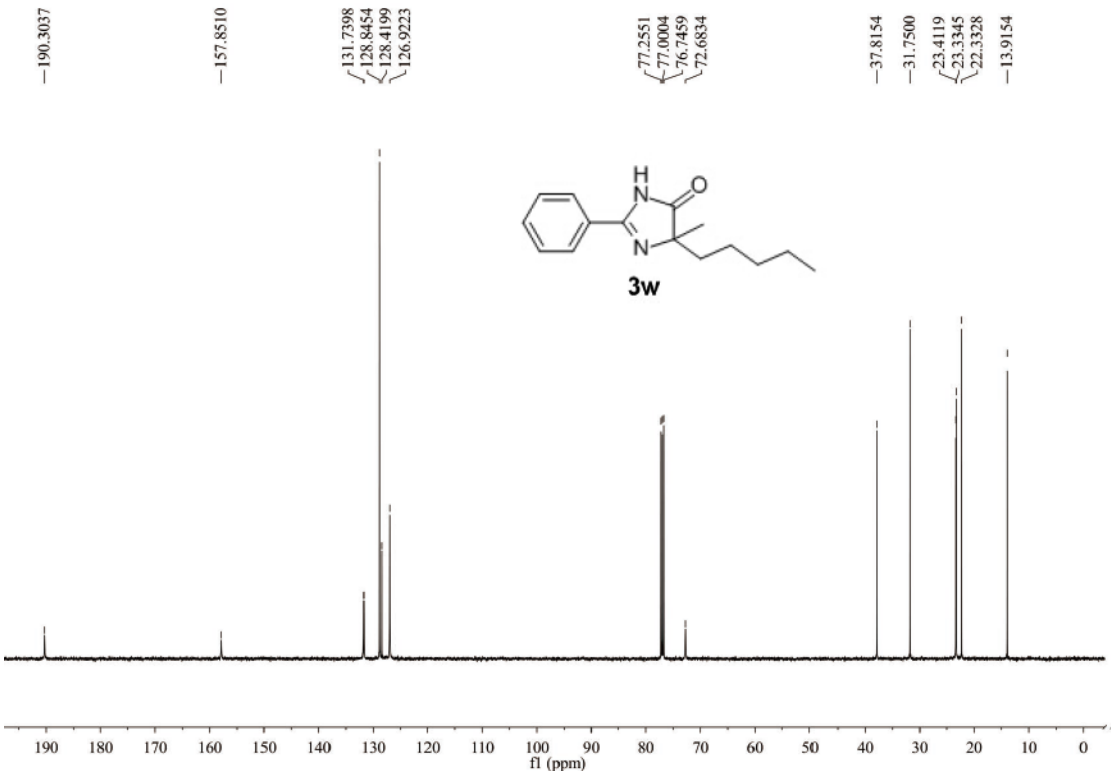
3.03
2.02
3.03
6.05
3.04
fl (ppm)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22.3328
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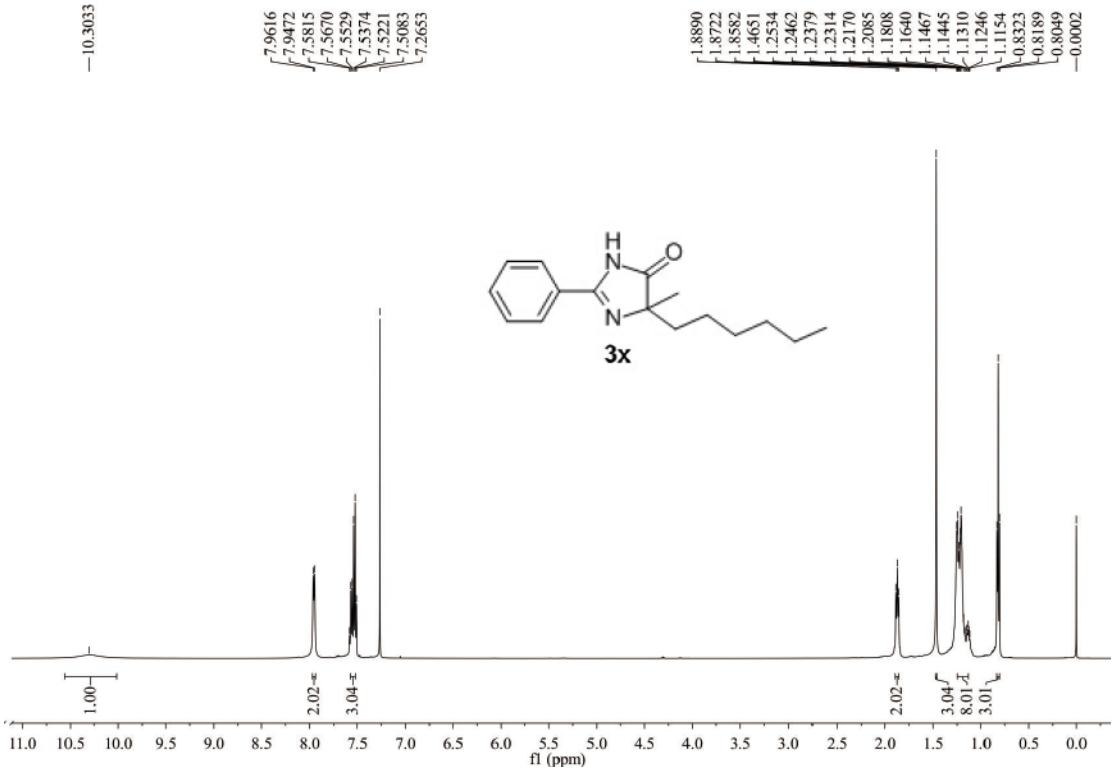
3w
fl (ppm)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7.5221
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7.2653
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1.8722
1.8582
1.4651
1.2534
1.2462
1.2379
1.2314
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1.2085
1.1808
1.1640
1.1467
1.1445
1.1310
1.1246
1.1154
0.8323
0.8189
0.8049
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3x
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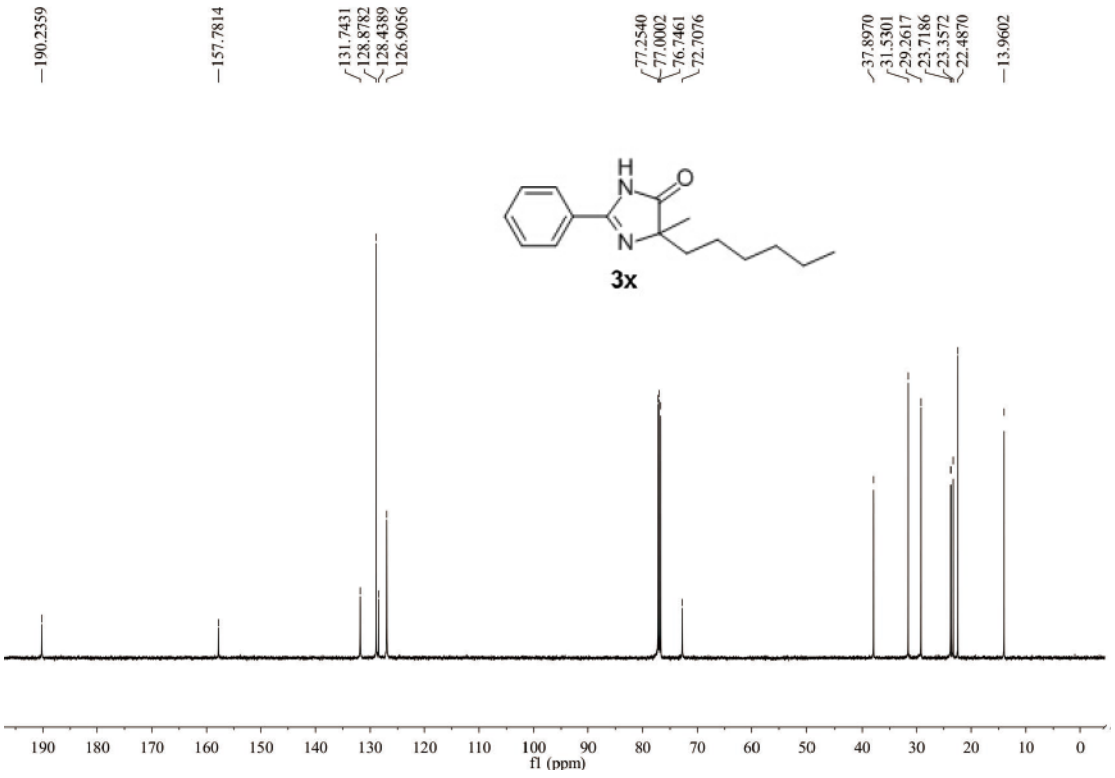
2.02
3.04
2.02
3.04
8.01
3.01
f1 (ppm)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22.4870
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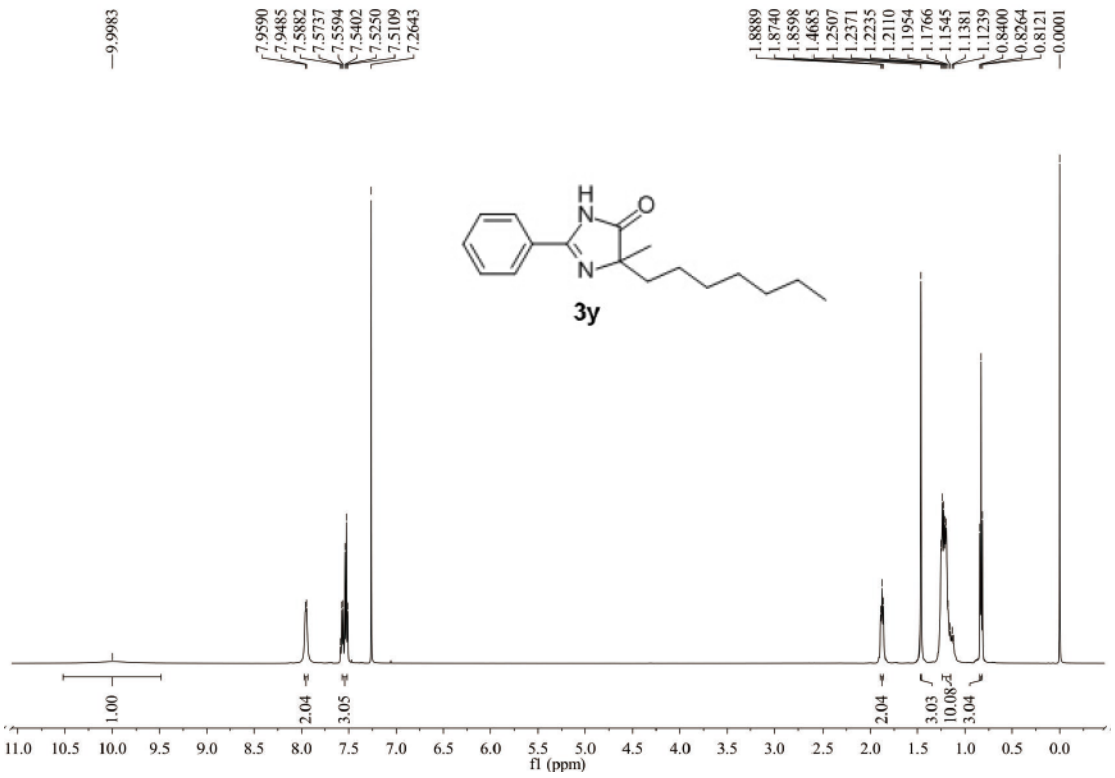
3x
f1 (ppm)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1.8889
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1.4685
1.2507
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1.2235
1.2110
1.1954
1.1766
1.1545
1.1381
1.1239
0.8400
0.8264
0.8121
0.0001
3y
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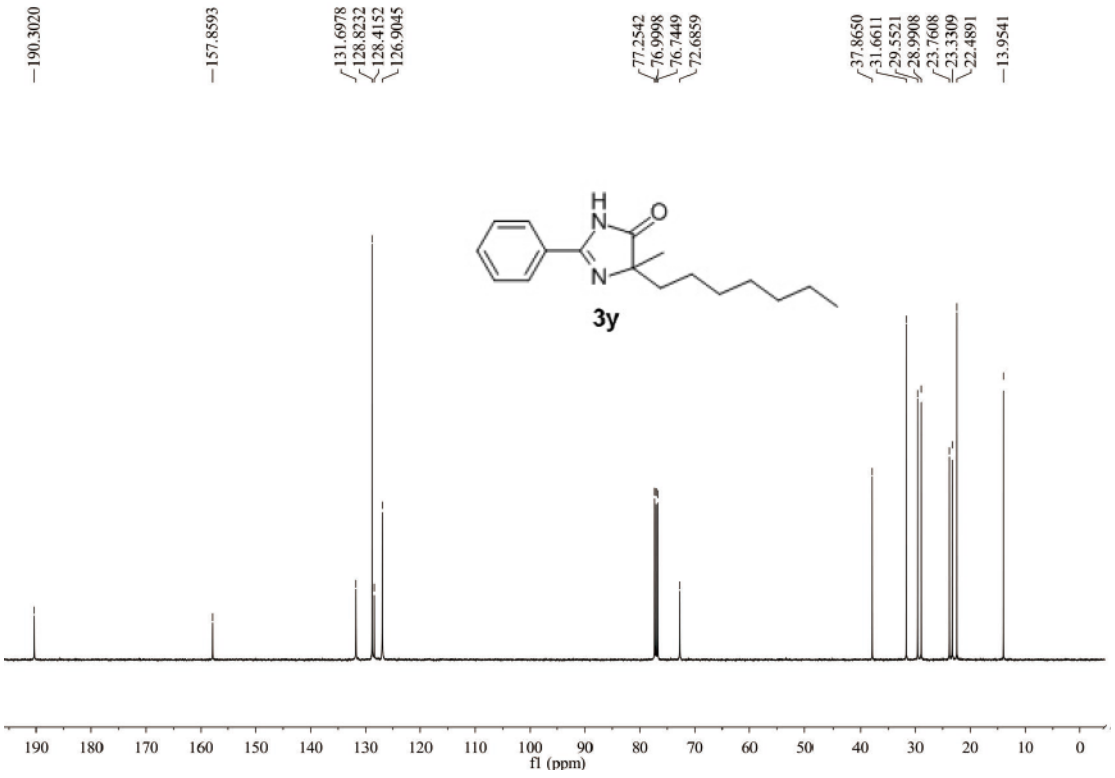
2.04
3.05
2.04
3.03
10.08
3.04
fl (ppm)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31.6611
29.5521
28.9908
23.7608
23.3309
22.4891
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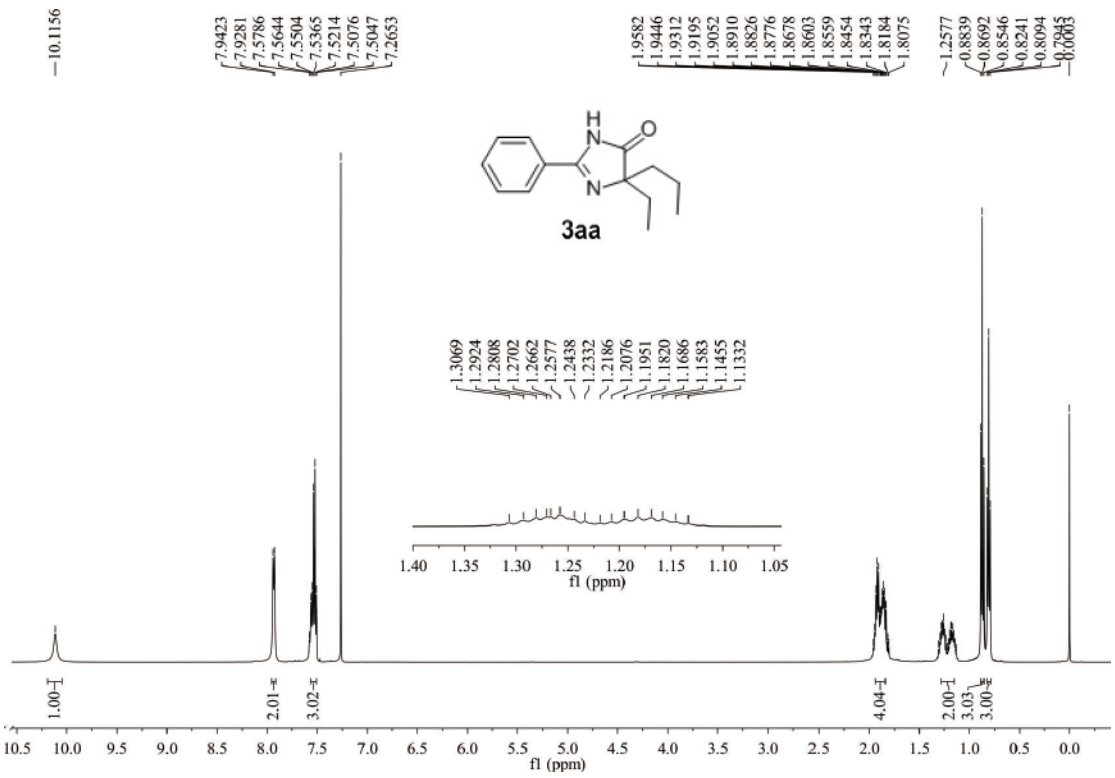
3y
fl (ppm)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1.9446
1.9312
1.9195
1.9052
1.8910
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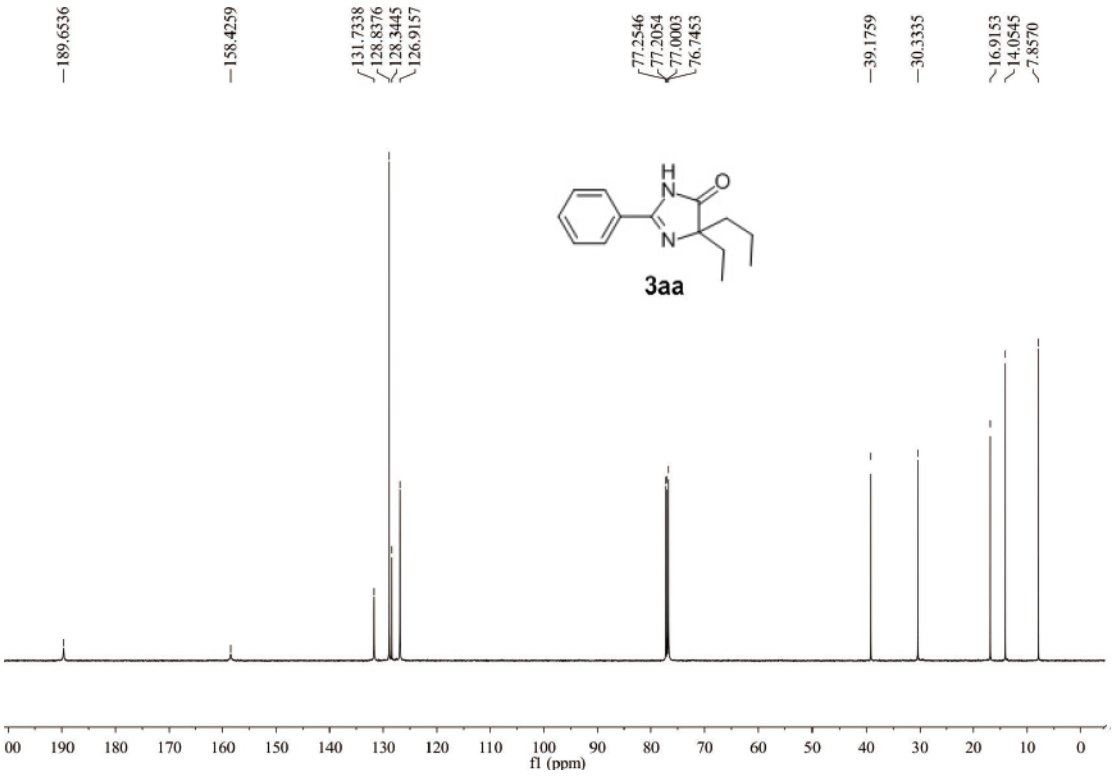
1.8776
1.8678
1.8603
1.8559
1.8454
1.8343
1.8184
1.8075
1.2577
0.8839
0.8692
0.8546
0.8241
0.8094
0.7945
0.0003
3aa
1.3069
1.2924
1.2808
1.2702
1.2662
1.2577
1.2438
1.2332
1.2186
1.2076
1.1951
1.1820
1.1686
1.1583
1.1455
1.1332
1.40 1.35 1.30 1.25 1.20 1.15 1.10 1.05
f1 (ppm)
1.00
2.01
3.02
4.04
2.00
3.03
3.00
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)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77.0003
76.7453
39.1759
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14.0545
7.8570
3aa
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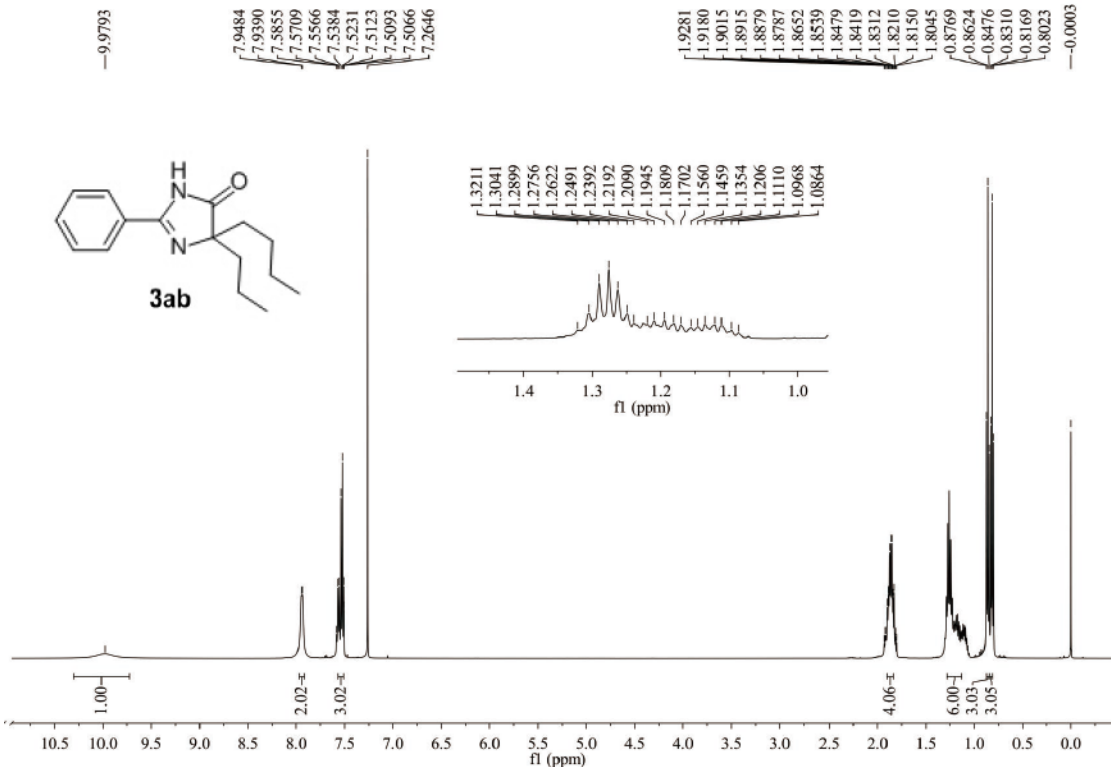
f1 (ppm)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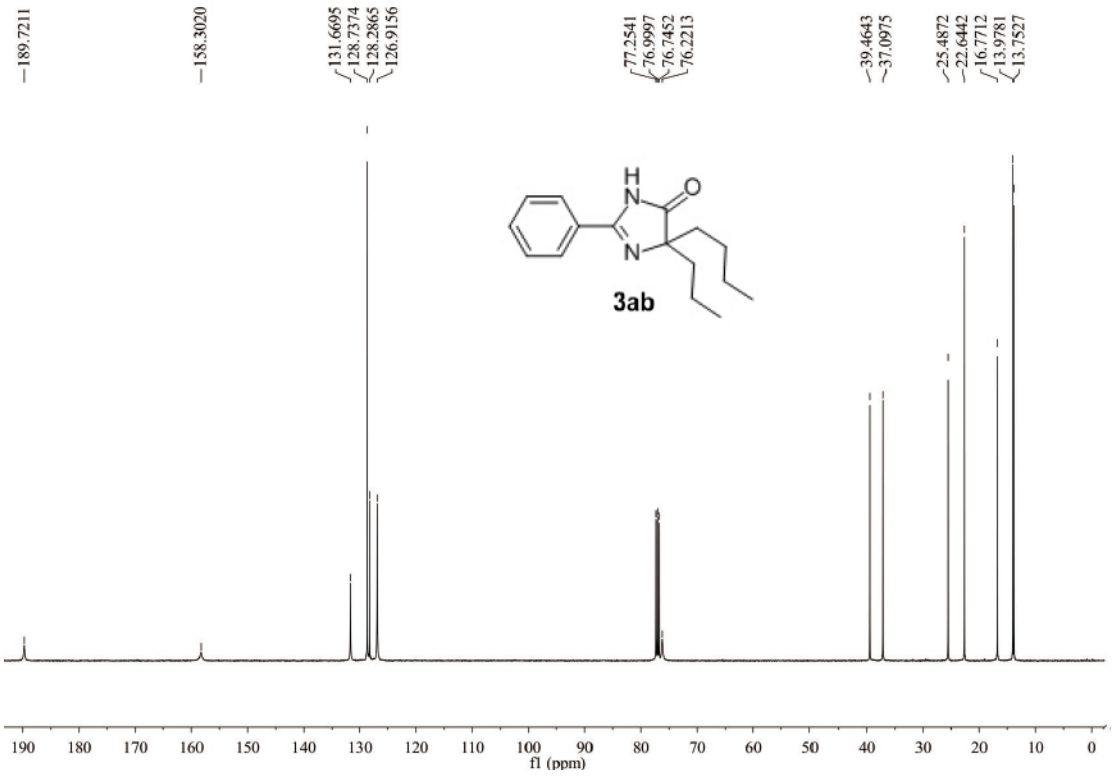
7.5123
7.5093
7.5066
7.2646
1.9281
1.9180
1.9015
1.8915
1.8879
1.8787
1.8652
1.8539
1.8479
1.8419
1.8312
1.8210
1.8150
1.8045
0.8769
0.8624
0.8476
0.8310
0.8169
0.8023
0.0003
3ab
1.3211
1.3041
1.2899
1.2756
1.2622
1.2491
1.2392
1.2192
1.2090
1.1945
1.1809
1.1702
1.1560
1.1459
1.1354
1.1206
1.1110
1.0968
1.0864
f1 (ppm)
1.00
2.02
3.02
4.06
6.00
3.03
3.05
f1 (ppm)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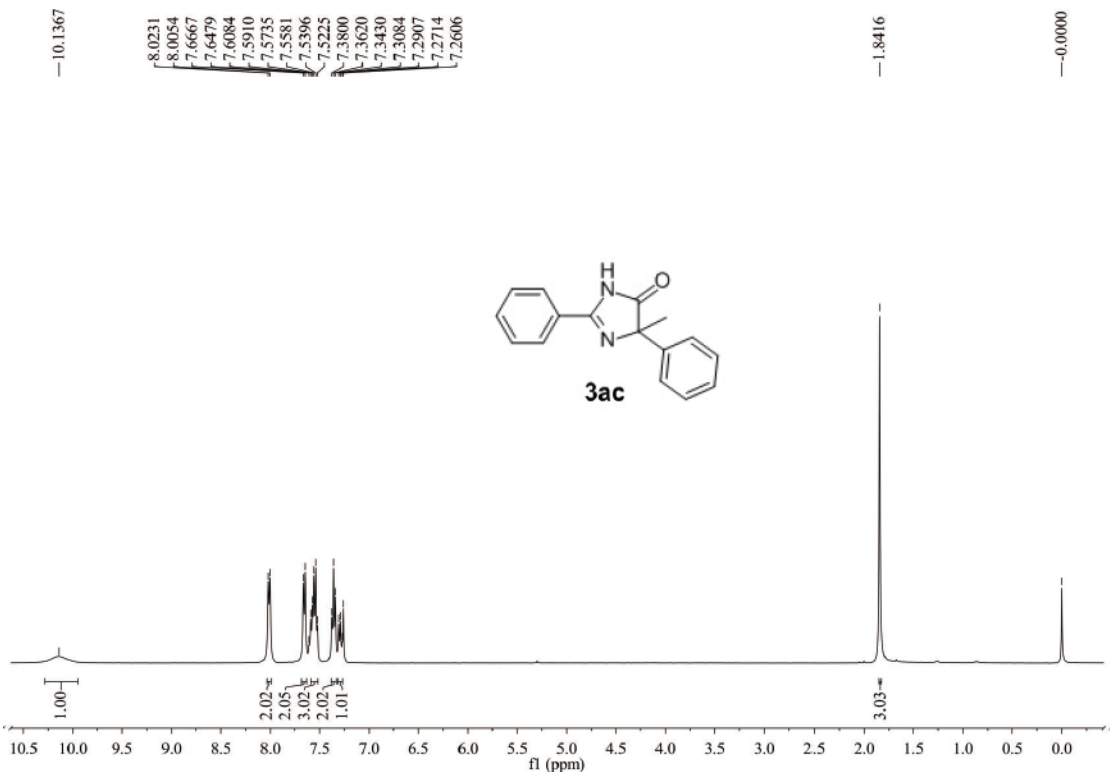
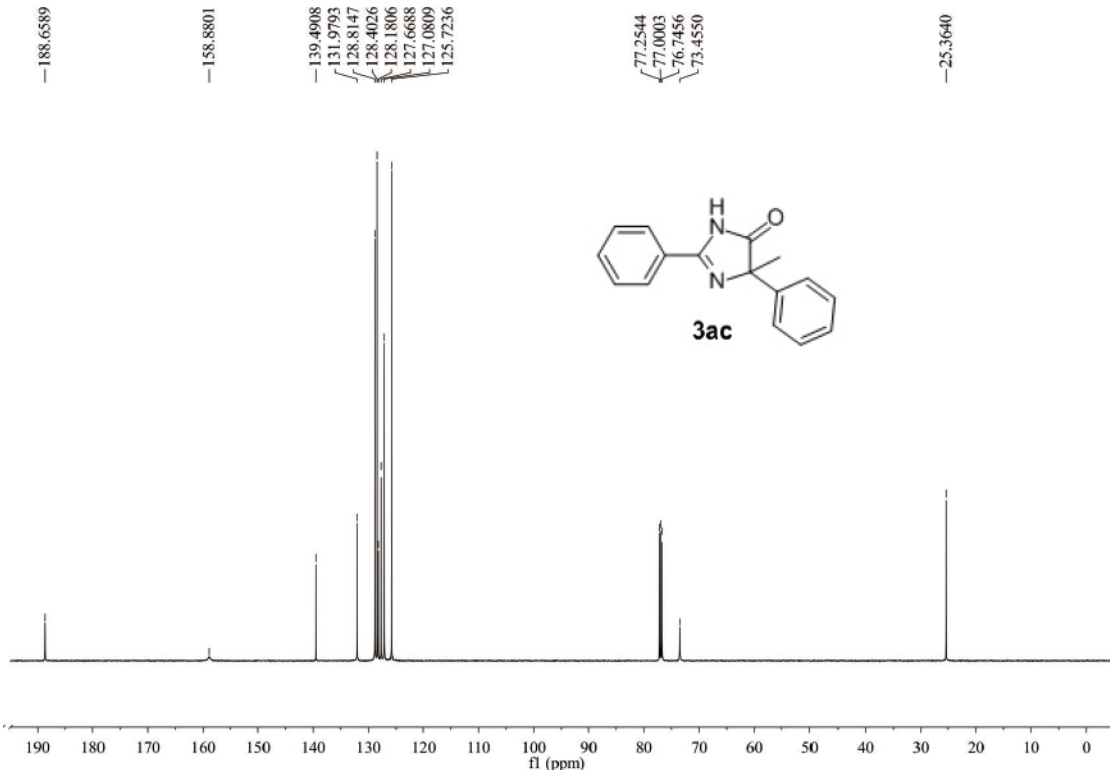
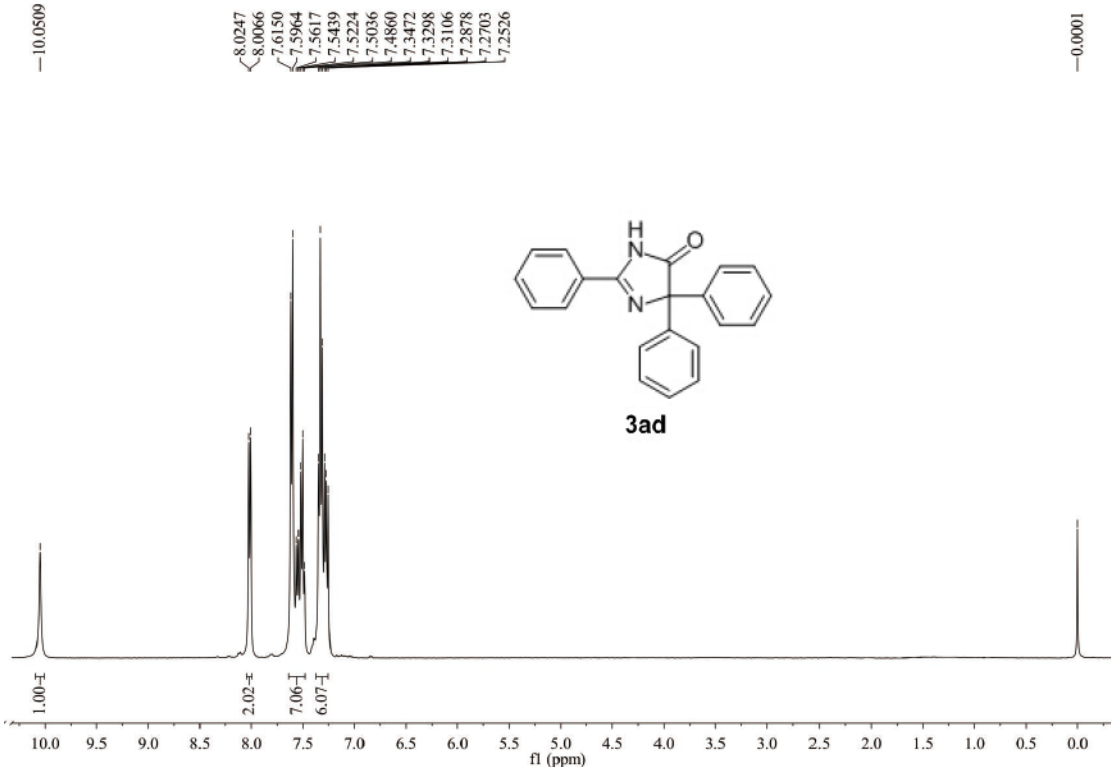
131.6695
128.7374
128.2865
126.9156
77.2541
76.9997
76.7452
76.2213
39.4643
37.0975
25.4872
22.6442
16.7712
13.9781
13.7527
3ab
f1 (ppm)

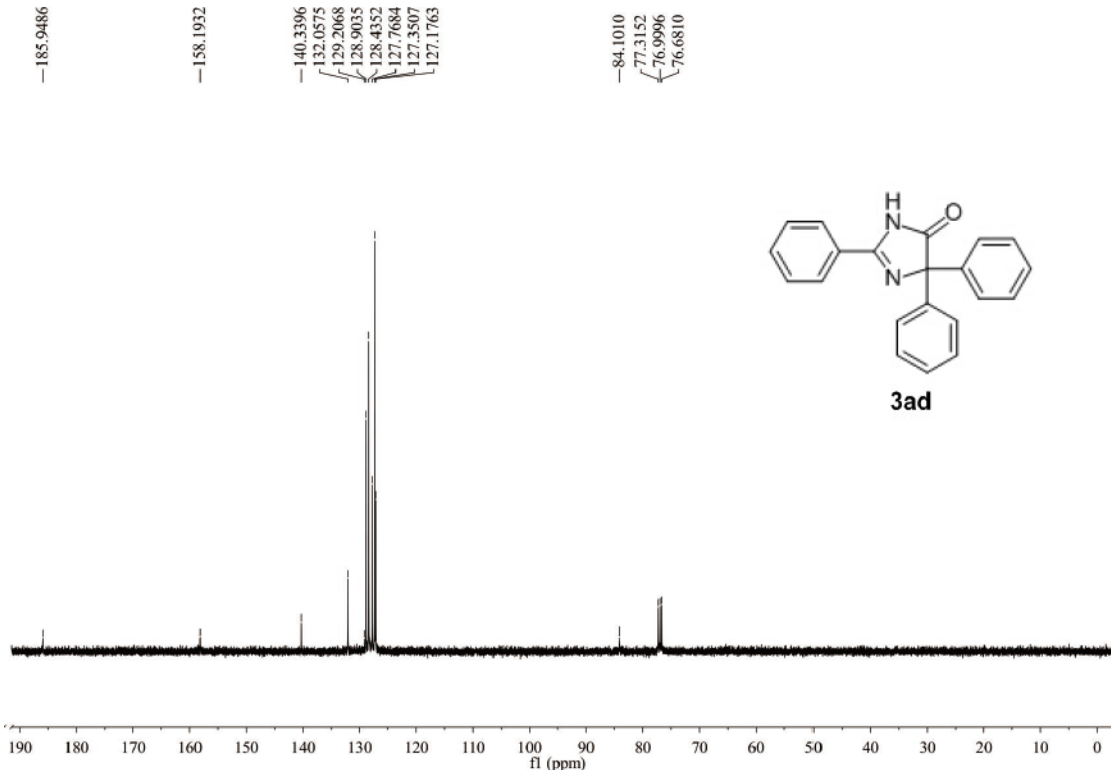

10.1367
8.0231
8.0054
7.6667
7.6479
7.6084
7.5910
7.5735
7.5581
7.5396
7.5225
7.3800
7.3620
7.3430
7.3084
7.2907
7.2714
7.2606
1.8416
0.0000
3ac
1.00
2.02
2.05
3.02
2.02
1.01
3.03
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


188.6589
158.8801
139.4908
131.9793
128.8147
128.4026
128.1806
127.6688
127.0809
125.7236
77.2544
77.0003
76.7456
73.4550
25.3640
3ac
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


10.0509
8.0247
8.0066
7.6150
7.5964
7.5617
7.5439
7.5224
7.5036
7.4860
7.3472
7.3298
7.3106
7.2878
7.2703
7.2526
0.0001
1.00
2.02
7.06
6.07
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)



3ad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140.3396
132.0575
129.2068
128.9035
128.4352
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127.3507
127.1763
84.1010
77.3152
76.9996
76.6810
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)



3ad

## X-ray structure of 3y:

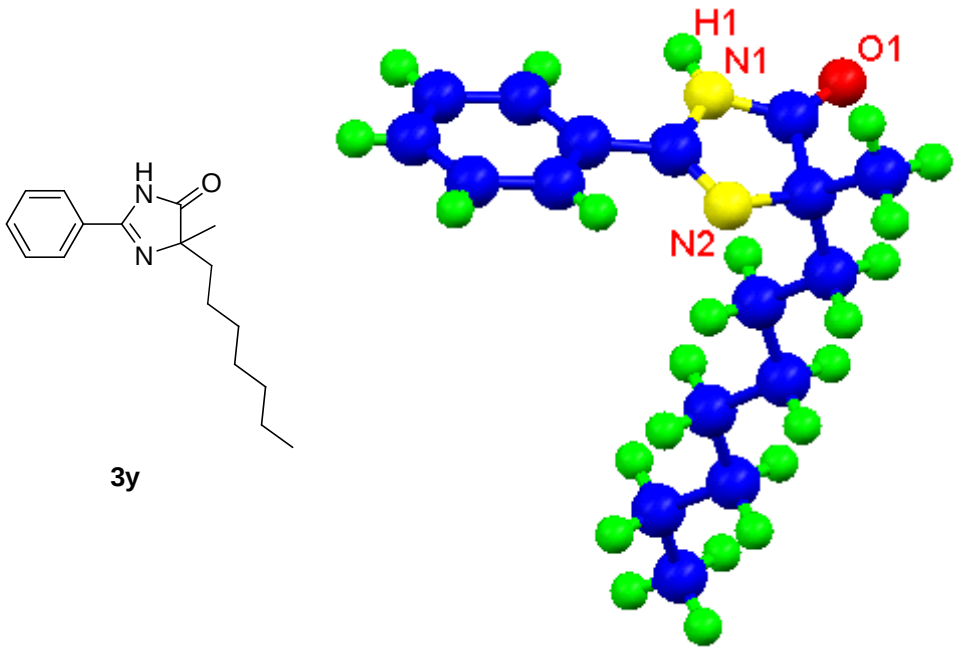


**Figure S1. X-ray structure of 3y**

**Table 1**. Crystal data and structure refinement for **3y**.

| | | |
|---|---|---|
| Identification code | **3y** | |
| Empirical formula | C17 H24 N2 O | |
| Formula weight | 272.38 | |
| Temperature | 173.1500 K | |
| Wavelength | 0.71073 Å | |
| Crystal system | Monoclinic | |
| Space group | C 1 2/c 1 | |
| Unit cell dimensions | a = 29.745(11) Å | α= 90°. |
| | b = 5.802(2) Å | β= 112.103(4)°. |
| | c = 19.967(8) Å | γ = 90°. |
| Volume | 3192(2) $Å^3$ | |
| Z | 8 | |
| Density (calculated) | 1.133 $Mg/m^3$ | |
| Absorption coefficient | 0.071 $mm^{-1}$ | |
| F(000) | 1184 | |
| Crystal size | 0.5 x 0.21 x 0.18 $mm^3$ | |
| Theta range for data collection | 2.957 to 27.458°. | |
| Index ranges | -38<=h<=38, -7<=k<=6, -20<=l<=25 | |
| Reflections collected | 12176 | |
| Independent reflections | 3635 [R(int) = 0.0277] | |

| | |
|---|---|
| Completeness to theta = 26.000° | 99.6 % |
| Absorption correction | Semi-empirical from equivalents |
| Max. and min. transmission | 1.0000 and 0.8989 |
| Refinement method | Full-matrix least-squares on $F^2$ |
| Data / restraints / parameters | 3635 / 0 / 187 |
| Goodness-of-fit on $F^2$ | 1.118 |
| Final R indices [I>2sigma(I)] | R1 = 0.0525, wR2 = 0.1152 |
| R indices (all data) | R1 = 0.0593, wR2 = 0.1190 |
| Extinction coefficient | n/a |
| Largest diff. peak and hole | 0.221 and -0.174 e.$Å^{-3}$ |

**Table 2.** Atomic coordinates (x $10^4$) and equivalent isotropic displacement parameters ($Å^2$x $10^3$) for **3y**. U(eq) is defined as one third of the trace of the orthogonalized $U^{ij}$ tensor.

| | x | y | z | U(eq) |
|---|---|---|---|---|
| O1 | 2859(1) | 2055(2) | 5992(1) | 31(1) |
| N1 | 2929(1) | 384(2) | 4983(1) | 26(1) |
| N2 | 3551(1) | -2109(2) | 5476(1) | 26(1) |
| C1 | 3193(1) | -2011(2) | 4154(1) | 25(1) |
| C2 | 2946(1) | -668(2) | 3549(1) | 28(1) |
| C3 | 2937(1) | -1301(3) | 2873(1) | 31(1) |
| C4 | 3173(1) | -3277(3) | 2798(1) | 32(1) |
| C5 | 3413(1) | -4644(3) | 3395(1) | 34(1) |
| C6 | 3424(1) | -4026(2) | 4071(1) | 31(1) |
| C7 | 3237(1) | -1316(2) | 4887(1) | 24(1) |
| C8 | 3485(1) | -867(2) | 6077(1) | 24(1) |
| C9 | 3055(1) | 729(2) | 5706(1) | 24(1) |
| C10 | 3366(1) | -2554(3) | 6574(1) | 32(1) |
| C11 | 3940(1) | 551(2) | 6500(1) | 27(1) |
| C12 | 4107(1) | 2172(3) | 6039(1) | 31(1) |
| C13 | 4545(1) | 3628(3) | 6481(1) | 31(1) |
| C14 | 4726(1) | 5125(3) | 6008(1) | 33(1) |
| C15 | 5134(1) | 6772(3) | 6421(1) | 34(1) |
| C16 | 5295(1) | 8247(3) | 5928(1) | 48(1) |
| C17 | 5670(1) | 10068(3) | 6314(1) | 51(1) |

**Table 3**. Bond lengths [Å] and angles [°] for **3y**.

| | |
|---|---|
| O1-C9 | 1.2294(16) |
| N1-C7 | 1.4068(17) |
| N1-C9 | 1.3622(17) |
| N2-C7 | 1.2804(17) |
| N2-C8 | 1.4748(17) |
| C1-C2 | 1.3926(19) |
| C1-C6 | 1.3965(19) |
| C1-C7 | 1.4759(18) |
| C2-C3 | 1.3913(19) |
| C3-C4 | 1.381(2) |
| C4-C5 | 1.386(2) |
| C5-C6 | 1.386(2) |
| C8-C9 | 1.5259(18) |
| C8-C10 | 1.5274(19) |
| C8-C11 | 1.5379(18) |
| C11-C12 | 1.5225(19) |
| C12-C13 | 1.5259(19) |
| C13-C14 | 1.522(2) |
| C14-C15 | 1.521(2) |
| C15-C16 | 1.513(2) |
| C16-C17 | 1.520(2) |
| | |
| C9-N1-C7 | 108.27(11) |
| C7-N2-C8 | 107.26(11) |
| C2-C1-C6 | 119.23(12) |
| C2-C1-C7 | 121.89(12) |
| C6-C1-C7 | 118.84(12) |
| C3-C2-C1 | 120.35(13) |
| C4-C3-C2 | 120.00(13) |
| C3-C4-C5 | 120.00(13) |
| C4-C5-C6 | 120.40(13) |
| C5-C6-C1 | 120.00(13) |
| N1-C7-C1 | 120.48(11) |
| N2-C7-N1 | 114.48(12) |
| N2-C7-C1 | 125.01(12) |
| N2-C8-C9 | 104.20(10) |

| | |
|---|---|
| N2-C8-C10 | 110.48(11) |
| N2-C8-C11 | 110.39(11) |
| C9-C8-C10 | 110.38(11) |
| C9-C8-C11 | 110.27(11) |
| C10-C8-C11 | 110.93(11) |
| O1-C9-N1 | 126.55(12) |
| O1-C9-C8 | 127.71(12) |
| N1-C9-C8 | 105.74(11) |
| C12-C11-C8 | 114.71(11) |
| C11-C12-C13 | 113.18(12) |
| C14-C13-C12 | 112.41(12) |
| C15-C14-C13 | 114.66(12) |
| C16-C15-C14 | 112.67(13) |
| C15-C16-C17 | 114.52(15) |

Symmetry transformations used to generate equivalent atoms:

**Table 4**. Anisotropic displacement parameters ($Å^2$ x $10^3$) for **3y**. The anisotropic displacement factor exponent takes the form: $-2\pi^2[ h^2 a^{*2}U^{11} + ... + 2 h k a^* b^* U^{12} ]$

| | $U^{11}$ | $U^{22}$ | $U^{33}$ | $U^{23}$ | $U^{13}$ | $U^{12}$ |
|---|---|---|---|---|---|---|
| O1 | 33(1) | 32(1) | 26(1) | -3(1) | 10(1) | 7(1) |
| N1 | 27(1) | 28(1) | 22(1) | -1(1) | 7(1) | 6(1) |
| N2 | 30(1) | 24(1) | 25(1) | 0(1) | 11(1) | 2(1) |
| C1 | 24(1) | 25(1) | 25(1) | -3(1) | 10(1) | -3(1) |
| C2 | 29(1) | 27(1) | 29(1) | -1(1) | 13(1) | 3(1) |
| C3 | 31(1) | 36(1) | 26(1) | 1(1) | 11(1) | 1(1) |
| C4 | 34(1) | 38(1) | 26(1) | -9(1) | 14(1) | -3(1) |
| C5 | 39(1) | 29(1) | 36(1) | -7(1) | 16(1) | 4(1) |
| C6 | 36(1) | 27(1) | 29(1) | -1(1) | 12(1) | 4(1) |
| C7 | 26(1) | 21(1) | 27(1) | -1(1) | 11(1) | 1(1) |
| C8 | 28(1) | 22(1) | 22(1) | 1(1) | 10(1) | 3(1) |
| C9 | 26(1) | 24(1) | 23(1) | -2(1) | 9(1) | -1(1) |
| C10 | 39(1) | 29(1) | 31(1) | 3(1) | 16(1) | -1(1) |
| C11 | 28(1) | 28(1) | 23(1) | 2(1) | 7(1) | 1(1) |
| C12 | 30(1) | 32(1) | 27(1) | 2(1) | 8(1) | -2(1) |
| C13 | 30(1) | 32(1) | 29(1) | -1(1) | 9(1) | -2(1) |

| | | | | | | |
|---|---|---|---|---|---|---|
| C14 | 30(1) | 35(1) | 31(1) | 1(1) | 8(1) | -3(1) |
| C15 | 31(1) | 35(1) | 34(1) | 0(1) | 10(1) | -3(1) |
| C16 | 43(1) | 54(1) | 45(1) | 5(1) | 14(1) | -15(1) |
| C17 | 39(1) | 43(1) | 70(1) | 0(1) | 20(1) | -10(1) |

**Table 5**. Hydrogen coordinates (x $10^4$) and isotropic displacement parameters (Å$^2$ x $10^3$) for **3y**.

| | x | y | z | U(eq) |
|---|---|---|---|---|
| H2 | 2785 | 659 | 3598 | 33 |
| H3 | 2773 | -394 | 2471 | 37 |
| H4 | 3171 | -3689 | 2347 | 38 |
| H5 | 3568 | -5984 | 3342 | 41 |
| H6 | 3584 | -4953 | 4470 | 37 |
| H10A | 3084 | -3436 | 6298 | 48 |
| H10B | 3304 | -1713 | 6944 | 48 |
| H10C | 3636 | -3574 | 6793 | 48 |
| H11A | 3876 | 1454 | 6863 | 33 |
| H11B | 4203 | -502 | 6750 | 33 |
| H12A | 3841 | 3189 | 5772 | 37 |
| H12B | 4188 | 1270 | 5691 | 37 |
| H13A | 4459 | 4607 | 6808 | 37 |
| H13B | 4806 | 2617 | 6771 | 37 |
| H14A | 4837 | 4129 | 5712 | 39 |
| H14B | 4455 | 6017 | 5685 | 39 |
| H15A | 5025 | 7766 | 6722 | 41 |
| H15B | 5408 | 5889 | 6737 | 41 |
| H16A | 5012 | 9007 | 5582 | 58 |
| H16B | 5429 | 7250 | 5659 | 58 |
| H17A | 5755 | 10918 | 5966 | 77 |
| H17B | 5954 | 9336 | 6652 | 77 |
| H17C | 5538 | 11102 | 6568 | 77 |
| H1 | 2687(6) | 1070(30) | 4647(9) | 34(4) |

**Table 6**. Torsion angles [°] for **3y**.

| | |
|---|---|
| N2-C8-C9-O1 | 177.74(13) |
| N2-C8-C9-N1 | -2.14(13) |
| N2-C8-C11-C12 | 53.31(15) |
| C1-C2-C3-C4 | 0.3(2) |
| C2-C1-C6-C5 | 1.3(2) |
| C2-C1-C7-N1 | 16.74(19) |
| C2-C1-C7-N2 | -161.40(13) |
| C2-C3-C4-C5 | 0.8(2) |
| C3-C4-C5-C6 | -0.9(2) |
| C4-C5-C6-C1 | -0.2(2) |
| C6-C1-C2-C3 | -1.4(2) |
| C6-C1-C7-N1 | -165.93(12) |
| C6-C1-C7-N2 | 15.9(2) |
| C7-N1-C9-O1 | -178.42(13) |
| C7-N1-C9-C8 | 1.46(14) |
| C7-N2-C8-C9 | 2.08(14) |
| C7-N2-C8-C10 | 120.63(12) |
| C7-N2-C8-C11 | -116.30(12) |
| C7-C1-C2-C3 | 175.94(13) |
| C7-C1-C6-C5 | -176.08(13) |
| C8-N2-C7-N1 | -1.31(15) |
| C8-N2-C7-C1 | 176.91(12) |
| C8-C11-C12-C13 | 177.37(11) |
| C9-N1-C7-N2 | -0.13(16) |
| C9-N1-C7-C1 | -178.45(12) |
| C9-C8-C11-C12 | -61.29(14) |
| C10-C8-C9-O1 | 59.12(18) |
| C10-C8-C9-N1 | -120.76(12) |
| C10-C8-C11-C12 | 176.12(12) |
| C11-C8-C9-O1 | -63.80(17) |
| C11-C8-C9-N1 | 116.32(12) |
| C11-C12-C13-C14 | 176.41(12) |
| C12-C13-C14-C15 | 174.75(12) |
| C13-C14-C15-C16 | -179.13(14) |
| C14-C15-C16-C17 | 174.74(14) |

Symmetry transformations used to generate equivalent atoms:

**Table 7**. Hydrogen bonds for **3y** [Å and °].

| D-H...A | d(D-H) | d(H...A) | d(D...A) | <(DHA) |
|---|---|---|---|---|
| N1-H1...O1#1 | 0.873(17) | 1.973(17) | 2.8386(16) | 171.1(15) |

Symmetry transformations used to generate equivalent atoms:

#1 -x+1/2,-y+1/2,-z+1