

Formation of Ylidenehydrazines Enabled by Manganese-Catalyzed Acceptorless Dehydrogenative Coupling

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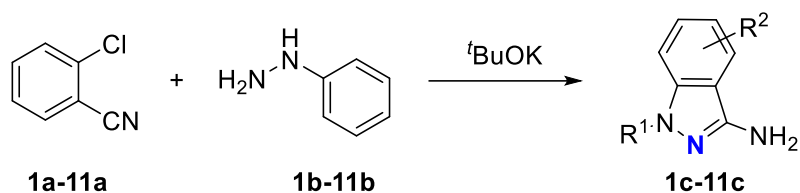
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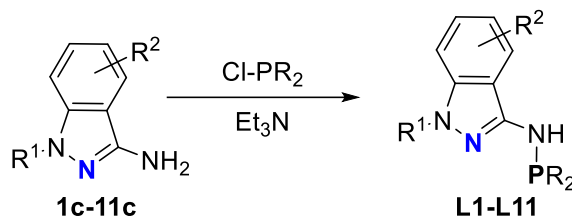
1. General

Experimental: All reactions and manipulations with air sensitive compounds being present were performed under dry argon (Ar 5.0) or nitrogen (N₂ 5.0), using Schlenk and glove box techniques. Deuterated solvents were bought from Cambridge Isotope Laboratories, distilled accordingly, and stored over molecular sieves (3 Å). Other chemicals were purchased from commercial vendors and used without further purification. NMR spectra were collected on a Varian INOVA 300 and 400 MHz spectrometer. Chemical shifts (δ) are reported in ppm relative to residual solvent signal. Coupling constants (J) are given in Hz (coupling patterns: s: singlet, s_br: broad singlet, d: doublet, t: triplet, q: quartet, m: multiplet). GC analyses were carried out using an Agilent Technologies 6890N system equipped with a Machinery-Nagel (MN) Optima 5 HT column (30 m, 320 μ m, 0.25 μ m) or an Agilent Technologies 6850 system equipped with a MN Optima 17 column (30 m, 320 μ m, 0.25 μ m). GC/MS analyses were carried out on an Agilent 7890A/MSD 5975C system equipped with a HP-5MS column (30 m, 320 μ m, 0.25 μ m). High resolution mass spectra (HRMS) were recorded on Bruker MicroTOF-QII mass (ESI). MN silica gel 60 (0.040 – 0.063 mm particle size) was used for flash column chromatography.

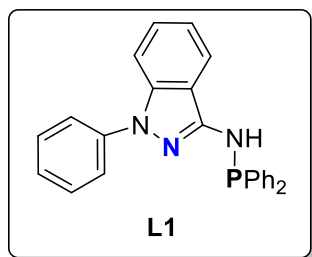
2. Synthesis of NNP-Ligands



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, base, hydrazines (**1b-11b**) and diglyme. After stirring of five minutes, nitriles (**1a-11a**) were added the mixture reaction. Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated oil bath (design temperature). After design time the reaction was cooled, quenched with half-saturated brine and extracted with dichloromethane (4 x 15 mL). The combined organic phase was dried over Na_2SO_4 and concentrated. A small aliquot of the organic phase was analyzed by GC or GC-MS to monitor product formation. Purification of the remainder by column chromatography on silica gel gave the corresponding products **1c-11c** (pentane/ethyl ether = 15/1 – 5/1).^[1]



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, catalyst, ligand, solvent, amines (**1c-11c**, 1.0 mmol), Cl-PR_2 (1.2 mmol), Et_3N (1.5 mmol) and THF (2.0 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (30 °C) for 12 hours. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/ethoxyethane = 20:1 – 1:1) on Al_2O_3 to give the product **L1-L11** in the reported yields.



***N*-(diphenylphosphanyl)-1-phenyl-1H-indazol-3-amine (L1)**

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 349.9 mg, 89% yield.

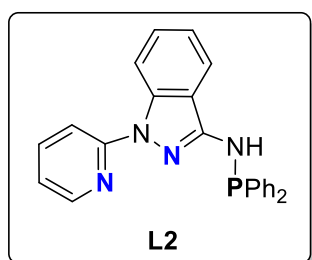
¹H NMR (400 MHz, CDCl₃) δ 8.05 (d, *J* = 8.1 Hz, 1H), 7.62 (dd, *J* = 8.2, 2.5 Hz, 3H), 7.52 (td, *J* = 7.6, 1.8 Hz, 4H), 7.40 (t, *J* = 7.9 Hz, 2H), 7.34 – 7.31 (m, 7H), 7.17 (t, *J* = 7.5 Hz, 1H), 7.06 (t, *J* = 7.5 Hz, 1H), 5.20 (d, *J* = 7.2 Hz, 1H) ppm.

¹³C NMR (101 MHz, CDCl₃) δ 148.98, 148.79, 140.35, 140.33, 140.23, 140.07, 131.27, 131.06, 129.14, 129.10, 128.49, 128.43, 127.61, 125.08, 121.72, 121.64, 121.60, 119.83, 116.69, 116.67, 110.13 ppm.

³¹P NMR (162 MHz, CDCl₃) δ 32.45 ppm.

HRMS (ESI) calcd. for C₂₅H₂₁N₃P [M+H]: 394.1473, found: 394.1474.

Elemental Analysis calcd for C₂₅H₂₀N₃P: C, 76.32; H, 5.12; N, 10.68; found: C, 76.34; H, 5.16; N, 10.63.



***N*-(diphenylphosphanyl)-1-(pyridin-2-yl)-1H-indazol-3-amine (L2)**

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 323.2 mg, 82% yield.

¹H NMR (400 MHz, CDCl₃) δ 8.82 (d, *J* = 8.6 Hz, 1H), 8.44 (dd, *J* = 4.9, 1.2 Hz, 1H), 7.98 (d, *J* = 8.1 Hz, 1H), 7.81 (d, *J* = 8.4 Hz, 1H), 7.72 – 7.67 (m, 1H), 7.59 – 7.55 (m, 4H), 7.52 – 7.48 (m, 1H), 7.43 – 7.40 (m, 6H), 7.20 (t, *J* = 7.5 Hz, 1H), 7.01

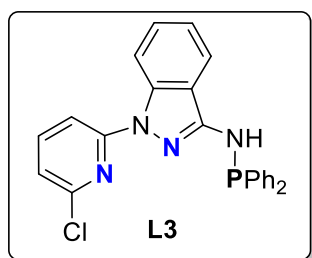
– 6.98 (m, 1H), 5.14 (d, $J = 6.4$ Hz, 1H) ppm.

^{13}C NMR (101 MHz, CDCl_3) δ 154.42, 149.82, 149.66, 147.58, 140.42, 140.22, 140.10, 138.00, 131.60, 131.39, 129.45, 128.77, 128.71, 128.64, 121.32, 120.65, 120.55, 118.36, 118.10, 118.09, 115.56, 112.71 ppm.

^{31}P NMR (162 MHz, CDCl_3) δ 32.08 ppm.

HRMS (ESI) calcd. for $\text{C}_{24}\text{H}_{20}\text{N}_4\text{P}$ [$\text{M}+\text{H}$]: 395.1425, found: 395.1422.

Elemental Analysis calcd for $\text{C}_{24}\text{H}_{19}\text{N}_4\text{P}$: C, 73.09; H, 4.86; N, 14.2; found: C, 73.04; H, 4.88; N, 14.19.



1-(6-chloropyridin-2-yl)-N-(diphenylphosphanyl)-1H-indazol-3-amine (L3)

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 325.4 mg, 76% yield.

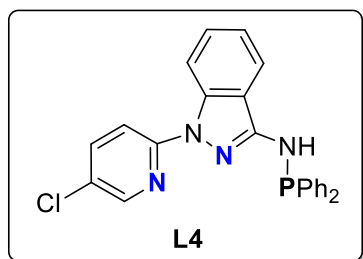
^1H NMR (400 MHz, CDCl_3) δ 8.77 (d, $J = 8.6$ Hz, 1H), 7.94 (d, $J = 8.0$ Hz, 1H), 7.70 (d, $J = 8.1$ Hz, 1H), 7.61 (t, $J = 7.9$ Hz, 1H), 7.57 – 7.51 (m, 5H), 7.43 – 7.40 (m, 6H), 7.22 (t, $J = 7.5$ Hz, 1H), 6.98 (d, $J = 7.6$ Hz, 1H), 5.13 (d, $J = 6.2$ Hz, 1H) ppm.

^{13}C NMR (101 MHz, CDCl_3) δ 153.58, 150.29, 150.13, 148.76, 140.21, 140.09, 139.82, 139.70, 131.45, 131.24, 129.38, 128.93, 128.65, 128.59, 121.74, 120.39, 120.30, 118.34, 117.56, 115.68, 110.31 ppm.

^{31}P NMR (162 MHz, CDCl_3) δ 32.1 ppm.

HRMS (ESI) calcd. for $\text{C}_{24}\text{H}_{19}\text{ClN}_4\text{P}$ [$\text{M}+\text{H}$]: 429.1036, found: 429.1031.

Elemental Analysis calcd for $\text{C}_{24}\text{H}_{18}\text{ClN}_4\text{P}$: C, 67.22; H, 4.23; N, 13.06; found: C, 67.28; H, 4.28; N, 13.07.



1-(5-chloropyridin-2-yl)-*N*-(diphenylphosphanyl)-1H-indazol-3-amine (L4)

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 346.8 mg, 81% yield.

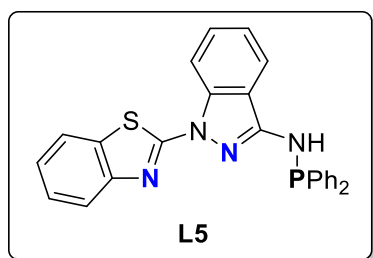
¹H NMR (400 MHz, CDCl₃) δ 8.69 (d, *J* = 8.6 Hz, 1H), 8.32 (d, *J* = 2.3 Hz, 1H), 7.92 (d, *J* = 8.1 Hz, 1H), 7.72 (d, *J* = 8.9 Hz, 1H), 7.57 (dd, *J* = 8.9, 2.5 Hz, 1H), 7.55 – 7.49 (m, 4H), 7.47 – 7.44 (m, 1H), 7.39 – 7.35 (m, 6H), 7.16 (t, *J* = 7.5 Hz, 1H), 5.15 (d, *J* = 6.2 Hz, 1H) ppm.

¹³C NMR (101 MHz, CDCl₃) δ 152.50, 150.09, 149.93, 145.89, 140.20, 139.95, 139.83, 137.65, 131.52, 131.31, 129.42, 128.76, 128.71, 128.65, 125.38, 121.55, 120.57, 120.48, 118.26, 118.25, 115.47, 113.46 ppm.

³¹P NMR (162 MHz, CDCl₃) δ 32.26 ppm.

HRMS (ESI) calcd. for C₂₄H₁₉ClN₄P [M+H]: 429.1036, found: 429.1032.

Elemental Analysis calcd for C₂₄H₁₈ClN₄P: C, 67.22; H, 4.23; N, 13.06; found: C, 67.26; H, 4.27; N, 13.08.



1-(benzo[d]thiazol-2-yl)-*N*-(diphenylphosphanyl)-1H-indazol-3-amine (L5)

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 387.1 mg, 86% yield.

¹H NMR (400 MHz, CDCl₃) δ 8.72 (d, *J* = 8.5 Hz, 1H), 8.09 (d, *J* = 8.1 Hz, 1H), 7.85 (d, *J* = 8.1 Hz, 1H), 7.73 (d, *J* = 7.9 Hz, 1H), 7.58 – 7.51 (m, 5H), 7.41 – 7.36 (m,

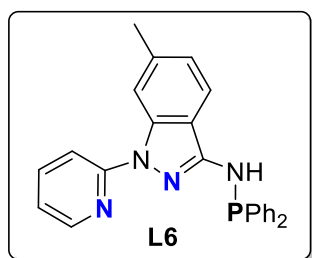
7H), 7.24 – 7.20 (m, 2H), 5.35 (d, $J = 5.3$ Hz, 1H) ppm.

^{13}C NMR (101 MHz, CDCl_3) δ 160.81, 152.26, 151.35, 151.17, 140.13, 139.39, 139.27, 132.02, 131.50, 131.29, 129.56, 129.48, 128.66, 128.59, 126.01, 123.28, 122.69, 121.69, 121.55, 121.34, 121.07, 118.74, 114.58 ppm.

^{31}P NMR (162 MHz, CDCl_3) δ 33.08 ppm.

HRMS (ESI) calcd. for $\text{C}_{26}\text{H}_{20}\text{N}_4\text{PS}$ [$\text{M}+\text{H}$]: 451.1146, found: 451.1143.

Elemental Analysis calcd for $\text{C}_{26}\text{H}_{19}\text{N}_4\text{PS}$: C, 69.32; H, 4.25; N, 12.44; found: C, 69.37; H, 4.27; N, 12.47.



***N*-(diphenylphosphanyl)-6-methyl-1-(pyridin-2-yl)-1H-indazol-3-amine (L6)**

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 355.1 mg, 87% yield.

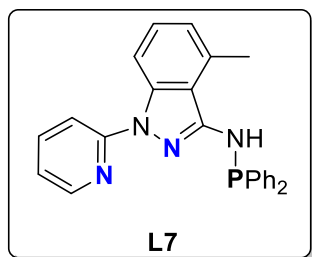
^1H NMR (400 MHz, CDCl_3) δ 8.61 (s, 1H), 8.37 (d, $J = 3.9$ Hz, 1H), 7.78 (dd, $J = 27.5, 8.3$ Hz, 2H), 7.51 – 7.48 (m, 5H), 7.30 – 7.28 (m, 6H), 6.94 (d, $J = 8.2$ Hz, 1H), 6.86 (dd, $J = 6.6, 5.3$ Hz, 1H), 5.24 (d, $J = 6.5$ Hz, 1H), 2.46 (s, 3H) ppm.

^{13}C NMR (101 MHz, CDCl_3) δ 153.24, 148.62, 148.45, 146.25, 139.76, 139.03, 138.91, 137.86, 136.68, 130.34, 130.13, 128.13, 127.48, 127.42, 122.06, 119.15, 119.04, 116.91, 115.06, 115.05, 113.92, 111.50, 21.09 ppm.

^{31}P NMR (162 MHz, CDCl_3) δ 32.13 ppm.

HRMS (ESI) calcd. for $\text{C}_{25}\text{H}_{22}\text{N}_4\text{P}$ [$\text{M}+\text{H}$]: 409.1582, found: 409.1586.

Elemental Analysis calcd for $\text{C}_{25}\text{H}_{21}\text{N}_4\text{P}$: C, 73.52; H, 5.18; N, 13.72; found: C, 73.53; H, 5.13; N, 13.78.



***N*-(diphenylphosphanyl)-4-methyl-1-(pyridin-2-yl)-1H-indazol-3-amine (L7)**

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 338.8 mg, 83% yield.

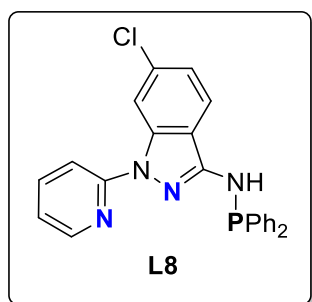
¹H NMR (400 MHz, CDCl₃) δ 8.70 (d, *J* = 8.6 Hz, 1H), 8.42 (dd, *J* = 4.9, 1.0 Hz, 1H), 7.82 (d, *J* = 8.4 Hz, 1H), 7.67 – 7.60 (m, 5H), 7.45 – 7.40 (m, 6H), 7.35 (dd, *J* = 8.3, 7.3 Hz, 1H), 6.99 – 6.90 (m, 1H), 6.91 (d, *J* = 7.1 Hz, 1H), 5.05 (d, *J* = 4.9 Hz, 1H), 2.70 (s, 3H) ppm.

¹³C NMR (101 MHz, CDCl₃) δ 154.32, 150.05, 149.95, 147.18, 140.34, 140.32, 140.18, 140.03, 137.70, 131.43, 131.22, 130.40, 129.13, 128.53, 128.47, 122.71, 118.09, 117.54, 117.53, 113.40, 112.91, 19.44 ppm.

³¹P NMR (162 MHz, CDCl₃) δ 32.67 ppm.

HRMS (ESI) calcd. for C₂₅H₂₂N₄P [M+H]: 409.1582, found: 409.1584.

Elemental Analysis calcd for C₂₅H₂₁N₄P: C, 73.52; H, 5.18; N, 13.72; found: C, 73.58; H, 5.14; N, 13.76.



6-chloro-*N*-(diphenylphosphanyl)-1-(pyridin-2-yl)-1H-indazol-3-amine (L8)

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 338.2 mg, 79% yield.

¹H NMR (400 MHz, CDCl₃) δ 8.86 (s, 1H), 8.44 – 8.38 (m, 1H), 7.90 (d, *J* = 8.5 Hz,

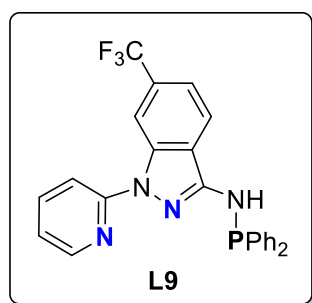
1H), 7.75 (d, $J = 8.4$ Hz, 1H), 7.67 (d, $J = 7.4$ Hz, 1H), 7.52 – 7.36 (m, 11H), 7.12 (d, $J = 8.5$ Hz, 1H), 6.99 – 6.96 (m, 1H), 5.19 (d, $J = 6.0$ Hz, 1H) ppm.

^{13}C NMR (101 MHz, CDCl_3) δ 153.85, 149.51, 149.34, 147.44, 140.43, 139.80, 139.68, 137.95, 134.72, 131.39, 131.19, 129.38, 128.65, 128.58, 121.94, 121.60, 121.49, 118.59, 115.26, 112.42 ppm.

^{31}P NMR (162 MHz, CDCl_3) δ 32.64 ppm.

HRMS (ESI) calcd. for $\text{C}_{24}\text{H}_{19}\text{ClN}_4\text{P}$ [M+H]: 429.1036, found: 429.1039.

Elemental Analysis calcd for $\text{C}_{24}\text{H}_{18}\text{ClN}_4\text{P}$: C, 67.22; H, 4.23; N, 13.06; found: C, 67.25; H, 4.27; N, 13.08.



***N*-(diphenylphosphanyl)-1-(pyridin-2-yl)-6-(trifluoromethyl)-1H-indazol-3-amine (L9)**

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 342 mg, 74% yield.

^1H NMR (400 MHz, CDCl_3) δ 9.20 (s, 1H), 8.48 – 8.46 (m, 1H), 8.13 (d, $J = 8.5$ Hz, 1H), 7.82 (d, $J = 8.4$ Hz, 1H), 7.73 – 7.69 (m, 1H), 7.59 – 7.55 (m, 4H), 7.42 – 7.40 (M, 7H), 7.04 (ddd, $J = 7.1, 4.9, 0.9$ Hz, 1H), 5.26 (d, $J = 6.3$ Hz, 1H) ppm.

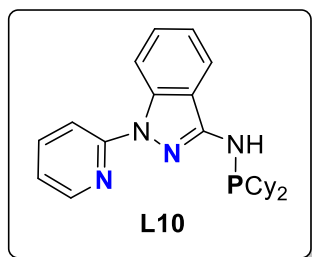
^{13}C NMR (101 MHz, CDCl_3) δ 153.80, 149.43, 149.26, 147.55, 139.66, 139.54, 139.23, 138.05, 131.41, 131.20, 130.40, 130.09, 129.45, 128.68, 128.62, 125.84, 123.13, 121.54, 121.43, 119.58, 118.89, 117.55, 113.33, 113.29, 112.52 ppm.

^{31}P NMR (162 MHz, CDCl_3) δ 32.97 ppm.

^{19}F NMR (376 MHz, CDCl_3) δ -61.48 ppm.

HRMS (ESI) calcd. for $\text{C}_{25}\text{H}_{19}\text{F}_3\text{N}_4\text{P}$ [M+H]: 463.1299, found: 463.1296.

Elemental Analysis calcd for $\text{C}_{25}\text{H}_{18}\text{F}_3\text{N}_4\text{P}$: C, 64.94; H, 3.92; N, 12.12; found: C, 64.97; H, 3.95; N, 12.17.



***N*-(dicyclohexylphosphanyl)-1-(pyridin-2-yl)-1H-indazol-3-amine (L10)**

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 325.1 mg, 80% yield.

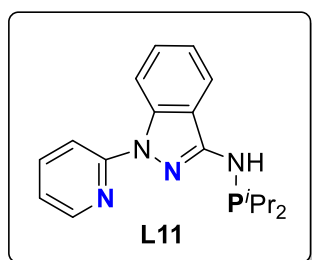
¹H NMR (400 MHz, CDCl₃) δ 8.78 (d, *J* = 8.5 Hz, 1H), 8.43 – 8.42 (m, 1H), 8.17 (d, *J* = 7.7 Hz, 1H), 7.82 (d, *J* = 8.3 Hz, 1H), 7.73 – 7.69 (m, 1H), 7.46 (t, *J* = 7.8 Hz, 1H), 7.17 (t, *J* = 7.5 Hz, 1H), 6.99 – 6.96 (m, 1H), 4.60 (d, *J* = 4.9 Hz, 1H), 1.87 – 1.67 (m, 12H), 1.32 – 1.19 (m, 10H) ppm.

¹³C NMR (101 MHz, CDCl₃) δ 154.33, 147.52, 140.45, 137.81, 128.28, 121.92, 121.78, 120.74, 117.98, 115.16, 112.41, 36.68, 36.56, 29.08, 28.90, 27.19, 27.12, 26.83, 26.75 ppm.

³¹P NMR (162 MHz, CDCl₃) δ 48.74 ppm.

HRMS (ESI) calcd. for C₂₄H₃₂N₄P [M+H]: 407.2364, found: 407.2369.

Elemental Analysis calcd for C₂₄H₃₁N₄P: C, 70.91; H, 7.69; N, 13.78; found: C, 70.95; H, 7.67; N, 13.74.



***N*-(diisopropylphosphanyl)-1-(pyridin-2-yl)-1H-indazol-3-amine (L11)**

The title compound was prepared according to the general procedure and purified by column chromatography to give a white solid 264.2 mg, 81% yield.

¹H NMR (400 MHz, CDCl₃) δ 8.83 (d, *J* = 8.6 Hz, 1H), 8.46 – 8.44 (m, 1H), 8.15 (d, *J* = 8.1 Hz, 1H), 7.86 (d, *J* = 8.4 Hz, 1H), 7.71 (ddd, *J* = 8.5, 7.3, 1.9 Hz, 1H), 7.49

(ddd, $J = 8.3, 7.1, 1.0$ Hz, 1H), 7.21 – 7.18 (m, 1H), 6.97 (ddd, $J = 7.2, 4.9, 0.9$ Hz, 1H), 4.62 (d, $J = 5.7$ Hz, 1H), 2.00 – 1.90 (m, 2H), 1.19 – 1.13 (m, 12H) ppm.

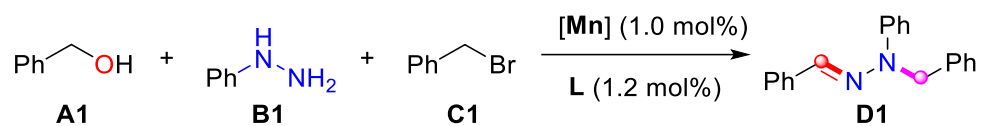
^{13}C NMR (101 MHz, CDCl_3) δ 154.21, 151.32, 151.18, 147.37, 140.27, 137.67, 128.17, 121.48, 121.35, 120.70, 117.91, 117.86, 115.13, 112.27, 26.79, 26.66, 18.83, 18.63, 17.25, 17.17 ppm.

^{31}P NMR (162 MHz, CDCl_3) δ 55.61 ppm.

HRMS (ESI) calcd. for $\text{C}_{18}\text{H}_{24}\text{N}_4\text{P}$ [$\text{M}+\text{H}$]: 327.1738, found: 327.1734.

Elemental Analysis calcd for $\text{C}_{18}\text{H}_{23}\text{N}_4\text{P}$: C, 66.24; H, 7.10; N, 17.17; found: C, 66.26; H, 7.13; N, 17.15.

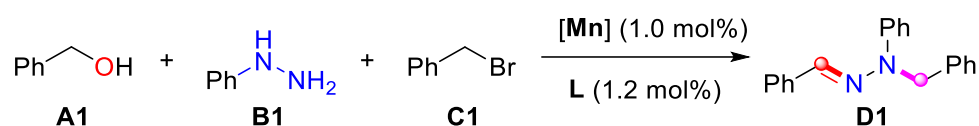
3. Screening of reaction parameters



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, catalyst, ligand, solvent, phenyl methanol (**A1**), phenyl hydrazine (**B1**) and (bromomethyl)benzene (**C1**). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (design temperature) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the product **D1** in the reported yields.

Entry	Parameter
Table S1	The difference of ligand screening
Table S2	The difference of catalyst screening
Table S3	The difference of base screening
Table S4	The difference of solvent screening
Table S5	The ratio of substrates screening
Table S6	Screening the loading of <i>t</i>-BuOK
Table S7	The loading of solvent screening
Table S8	The Pd-catalyst loading screening
Table S9	Reaction temperature screening
Table S10	Reaction time screening
Table S11	The reaction system screening

Table S1. The difference of ligand screening ^[a]



Entry	Ligand	D1 (%)	D1:E1
1	L1	<5	8:1
2	L2	62	>20:1
3	L3	41	>20:1
4	L4	54	18:1
5	L5	22	19:1
6	L6	60	18:1
7	L7	63	18:1
8	L8	51	17:1
9	L9	47	18:1
10	L10	82	19:1
11	L11	91	>20:1
12	-	0	-

[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (1.0 mol%), **L1-L11** (1.2 mol%), ^tBuOK (1.0 mmol), THF (3.0 mL), 100 °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

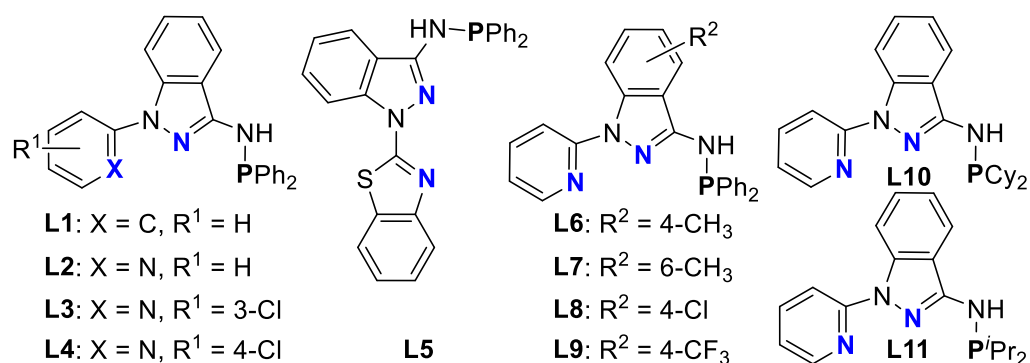
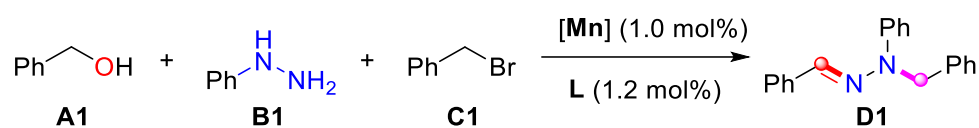


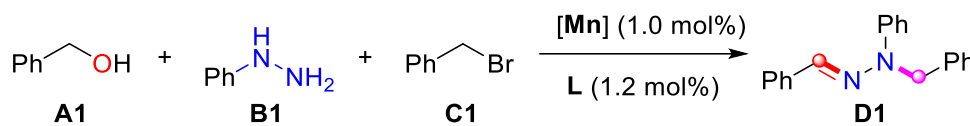
Table S2. The difference of catalyst screening^[a]



Entry	Catalyst	D1 (%)
1	MnBr(CO) ₅	91
2	Mn	0
3	MnO	0
4	MnO ₂	0
5	Mn ₃ O ₄	0
6	MnS	0
7	Mn(acac) ₃	0
8	Mn(OTf) ₂	0
9	Mn(NO ₃) ₂	0
10	Mn(NO ₃) ₂ •4H ₂ O	0
11	MnSO ₄ •H ₂ O	0
12 ^b	Mn(OAc) ₃ •4H ₂ O	0
13 ^b	MnTe ₂	0
14	MnCO ₃	0
15	MnF ₂	0
16	MnF ₃	0
17	MnCl ₂	0
18	MnCl ₂ •4H ₂ O	0
19	MnBr ₂	0
20	MnBr ₂ •4H ₂ O	0
21	-	0

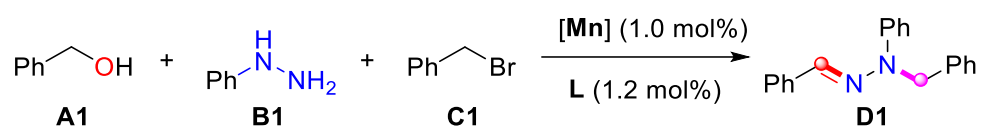
[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), Catalyst (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), THF (3.0 mL), 100 °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

Table S3. The difference of base screening^[a]



Entry	Base (5 mol%)	D1 (%)
1	LiOH	0
2	<i>t</i> -BuOLi	11
3	LiHMDS	9
4	Na ₂ CO ₃	0
5	NaOH	<5
6	<i>t</i> -BuONa	63
7	NaHMDS	64
8	NaH	41
9	K ₂ CO ₃	0
10	K ₃ PO ₄	0
11	KOH	<5
12	<i>t</i>-BuOK	91
13	KHMDS	82
14	KH	43
15	Cs ₂ CO ₃	0
16	CsOH	<5
17	<i>t</i> -BuOCs	26
18	Et ₃ N	0
19	Pyridine	0
20	DBU	0
21	TMEDA	0

[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), Base (1.0 mmol), THF (3.0 mL), 100 °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

Table S4. The difference of solvent screening^[a]

Entry	Solvent (3 mL)	D1 (%)
1	anisole	72
2	THF	91
3	1,4-dioxane	80
4	DME	12
5	toluene	67
6	xylene	61
7	mesitylene	52
8	DCM	<5
9	CH ₃ CN	<5
10	pyridine	13
11	methanol	<5
12	ethanol	<5
13	isopropanol	<5
14	<i>t</i> -AmOH	26
15	DMF	<5
16	DMAc	<5
17	NMP	<5
18	DMSO	<5

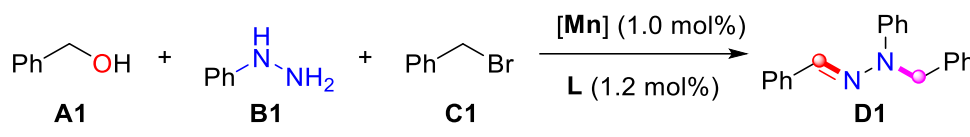
[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), solvent (3.0 mL), 100 °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

Table S5. The ratio of substrates screening^[a]

$\text{Ph-CH}_2\text{-OH}$ (A1) + Ph-NH-NH_2 (B1) + $\text{Ph-CH}_2\text{-Br}$ (C1) $\xrightarrow[\text{L (1.2 mol\%)}]{\text{[Mn] (1.0 mol\%)}}$ $\text{Ph-CH=N-N(Ph)-CH}_2\text{-Ph}$ (D1)

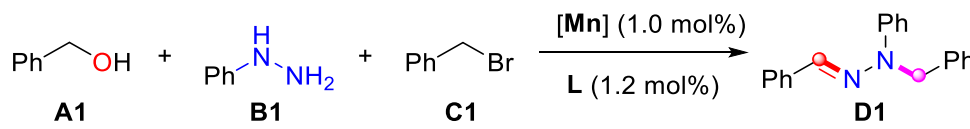
Entry	A1(mmol)	B1(mmol)	C1(mmol)	D1 (%)
1	0.5	0.5	0	0
2	0.5	0.5	0.5	51
3	0.5	0.5	0.6	60
4	0.5	0.5	0.7	63
5	0.5	0.5	0.8	63
6	0	0.5	0.7	0
7	0.6	0.5	0.7	77
8	0.7	0.5	0.7	86
9	0.8	0.5	0.7	91
10	0.9	0.5	0.7	91
11	0.8	0.4	0.7	87
12	0.8	0.6	0.7	90
13	0.8	0.7	0.7	90

[a] Reaction condition: **A1** (x mmol), **B1** (x mmol), **C1** (x mmol), MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), THF (3.0 mL), 100 °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

Table S6. Screening the loading of *t*-BuOK^[a]

Entry	<i>t</i> -BuOK (mmol)	D1 (%)
1	0	0
2	0.25	0
3	0.5	<5
4	0.75	86
5	1.0	91
6	1.25	91
7	2.0	90

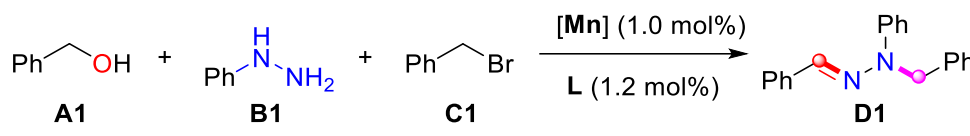
[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), *t*BuOK (X mmol), THF (3.0 mL), 100 °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

Table S7. The loading of solvent screening^[a]

Entry	THF (mL)	D1 (%)
1	0	23
2	0.5	61
3	1.0	76
4	1.5	80
5	2.0	88
6	3.0	91
7	4.0	90

[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), *t*BuOK (1.0 mmol), THF (x mL), 100 °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

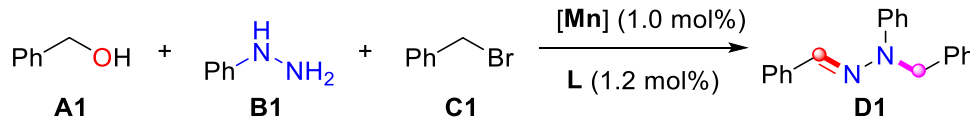
Table S8. The Pd-catalyst loading screening^[a]



Entry	MnBr(CO) ₅	L11	D1 (%)
1	0		0
2	0.0005	0.0006	13
3	0.001	0.0012	42
4	0.005	0.006	76
5	0.01	0.012	91
6	0.02	0.024	91

[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (x mol%), **L11** (x mol%), ^tBuOK (1.0 mmol), solvent (3.0 mL), 100 °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

Table S9. Reaction temperature screening^[a]



Entry	T (° C)	D1 (%)
1	RT	0
2	80	61
3	90	80
4	100	91
5	110	89
6	120	82

[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), THF (3.0 mL), x °C, 6 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

Table S10. Reaction time screening^[a]

Entry	t (h)	D1 (%)
1	3	65
2	6	91
3	10	93
4	12	96
5	14	96

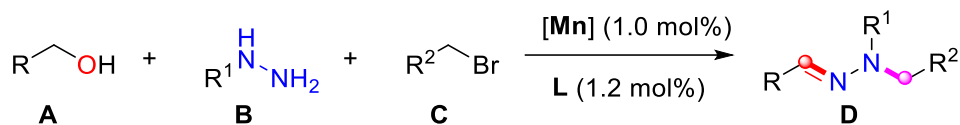
[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), THF (3.0 mL), 100 °C, x h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

Table S11. The reaction system screening^[a]

Entry	System	atmosphere	D1 (%)
1	open	N ₂	93
2	open	O ₂	<5
3	open	air	36
4	seal	N ₂	96
5	seal	O ₂	<5
6	seal	air	46

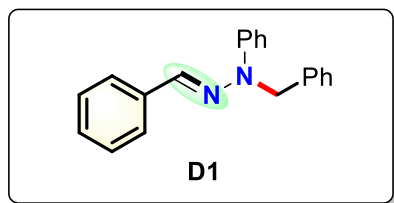
[a] Reaction condition: **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol), MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), THF (3.0 mL), 100 °C, 12 h. Yields of **D1** was determined by GC analysis using *n*-cetane as the internal standard.

4. General Procedure for the products



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), alcohols (**A**, 0.8 mmol), hydrazines (**B**, 0.5 mmol) and alkyl halides (**C**, 0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for 12 hours. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the products **D** in the reported yields.

5. Characterization Data ^[2]



1-benzyl-2-benzylidene-1-phenylhydrazine (D1)

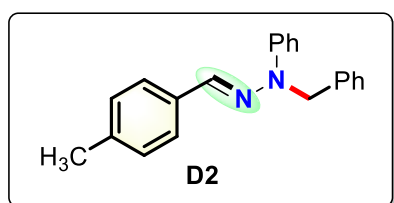
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 131.7 mg, 92% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.64 – 7.55 (m, 2H), 7.42 – 7.36 (m, 2H), 7.35 – 7.24 (m, 7H), 7.20 (q, *J* = 7.3 Hz, 4H), 6.96 – 6.88 (m, 1H), 5.10 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.99, 136.67, 135.78, 132.59, 129.33, 129.17, 128.68, 128.02, 127.45, 126.35, 126.17, 120.90, 114.92, 50.48 ppm.

HRMS (ESI) calcd. for C₂₀H₁₉N₂ [M+H]: 287.1548, found: 287.1549.

Elemental Analysis calcd for C₂₀H₁₈N₂: C, 83.88; H, 6.34; N, 9.78; found: C, 83.86; H, 6.36; N, 9.76.



1-benzyl-2-(4-methylbenzylidene)-1-phenylhydrazine (D2)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 144.2 mg, 96% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

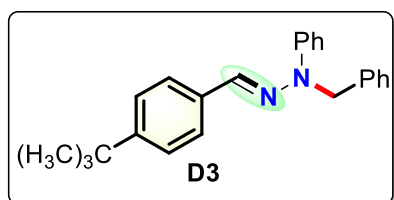
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.52 (d, *J* = 8.1 Hz, 2H), 7.44 – 7.31 (m, 6H), 7.28 – 7.20 (m, 4H), 7.14 (d, *J* = 8.0 Hz, 2H), 6.94 (t, *J* = 7.2 Hz, 1H), 5.17 (s, 2H), 2.34 (s, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.90, 137.82, 135.78, 133.77, 132.73,

129.20, 129.11, 128.96, 127.22, 126.12, 126.03, 120.52, 114.66, 50.41, 21.30 ppm.

HRMS (ESI) calcd. for $C_{21}H_{21}N_2$ [M+H]: 301.1705, found: 301.1705.

Elemental Analysis calcd for $C_{21}H_{20}N_2$: C, 83.96; H, 6.71; N, 9.33; found: C, 83.92; H, 6.72; N, 9.32.



1-benzyl-2-(4-(tert-butyl)benzylidene)-1-phenylhydrazine (D3)

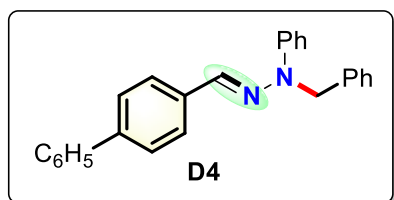
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 164.4 mg, 96% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

1H NMR (400 MHz, 23.0 °C, $CDCl_3$) δ 7.57 (d, J = 8.4 Hz, 2H), 7.46 – 7.26 (m, 10H), 7.23 (d, J = 7.0 Hz, 2H), 6.94 (t, J = 7.2 Hz, 1H), 5.18 (s, 2H), 1.31 (s, 9H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, $CDCl_3$) δ 151.11, 147.92, 135.72, 133.81, 132.63, 129.13, 128.97, 127.24, 126.07, 125.94, 125.46, 120.56, 114.72, 50.35, 34.66, 31.28 ppm.

HRMS (ESI) calcd. for $C_{24}H_{27}N_2$ [M+H]: 343.2174, found: 343.2176.

Elemental Analysis calcd for $C_{24}H_{26}N_2$: C, 84.17; H, 7.65; N, 8.18; found: C, 84.12; H, 7.64; N, 8.14.



2-([1,1'-biphenyl]-4-ylmethylene)-1-benzyl-1-phenylhydrazine (D4)

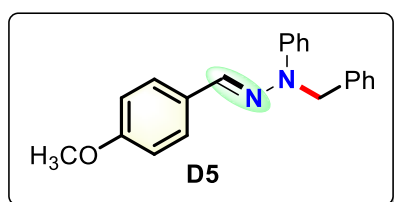
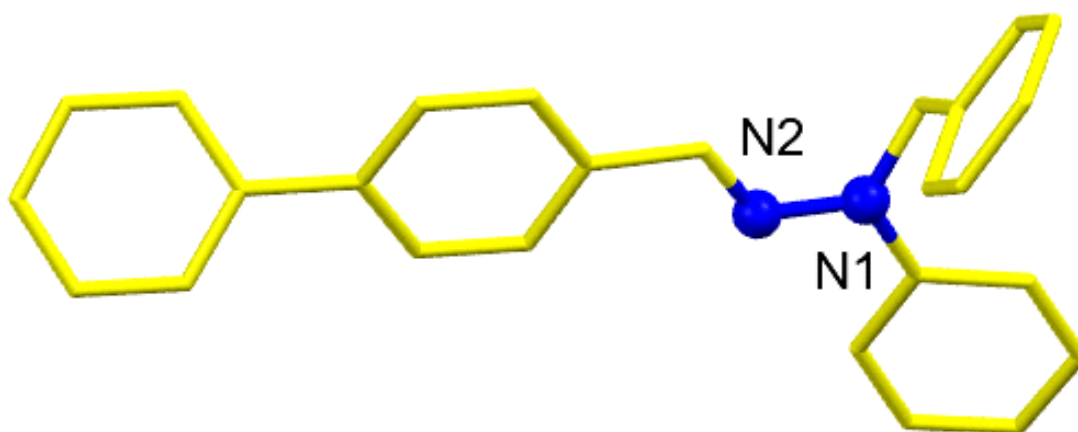
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 146.8 mg, 81% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.70 (d, $J = 8.4$ Hz, 2H), 7.65 – 7.50 (m, 4H), 7.46 – 7.42 (m, 4H), 7.37 – 7.31 (m, 5H), 7.31 – 7.26 (m, 3H), 7.25 (d, $J = 1.4$ Hz, 1H), 6.97 (t, $J = 7.2$ Hz, 1H), 5.21 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.85, 140.77, 135.61, 132.09, 129.18, 129.04, 128.80, 127.89, 127.33, 127.22, 126.94, 126.58, 126.05, 120.83, 114.86, 50.50 ppm.

HRMS (ESI) calcd. for $\text{C}_{26}\text{H}_{23}\text{N}_2$ [$\text{M}+\text{H}$]: 363.1861, found: 363.1860.

Elemental Analysis calcd for $\text{C}_{26}\text{H}_{22}\text{N}_2$: C, 86.15; H, 6.12; N, 7.73; found: C, 86.16; H, 6.16; N, 7.76.



1-benzyl-2-(4-methoxybenzylidene)-1-phenylhydrazine (D5)

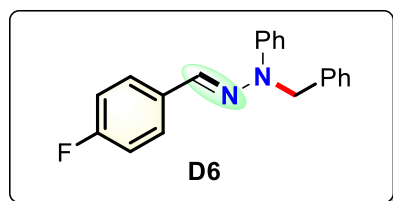
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 151.9mg, 96% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.56 (d, $J = 8.7$ Hz, 2H), 7.40 – 7.23 (m, 10H), 6.90 (dd, $J = 26.3, 7.9$ Hz, 3H), 5.16 (s, 2H), 3.81 (s, 3H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 159.59, 147.94, 135.90, 132.54, 129.39, 129.10, 128.96, 127.48, 127.21, 126.05, 120.37, 114.55, 113.97, 55.32, 50.47 ppm.

HRMS (ESI) calcd. for $C_{21}H_{21}N_2O$ [M+H]: 317.1654, found: 317.1653.

Elemental Analysis calcd for $C_{21}H_{20}N_2O$: C, 79.72; H, 6.37; N, 8.85; O, 5.06; found: C, 79.74; H, 6.36; N, 8.84.



1-benzyl-2-(4-fluorobenzylidene)-1-phenylhydrazine (D6)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 141.5 mg, 93% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

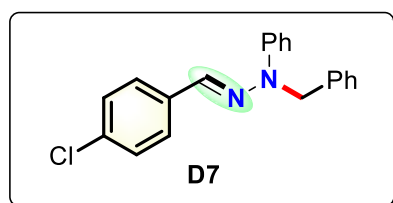
1H NMR (400 MHz, 23.0 °C, $CDCl_3$) δ 7.67 – 7.52 (m, 2H), 7.40 – 7.20 (m, 10H), 7.09 – 6.88 (m, 3H), 5.17 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, $CDCl_3$) δ 163.75, 161.29, 147.79, 135.57, 132.74, 131.30, 129.16, 129.02, 127.70, 127.62, 127.32, 125.99, 120.80, 115.58, 115.36, 114.77, 50.55 ppm.

^{19}F NMR (377 MHz, 23.0 °C, $CDCl_3$) δ -113.75 ppm.

HRMS (ESI) calcd. for $C_{20}H_{18}FN_2$ [M+H]: 305.1454, found: 305.1455.

Elemental Analysis calcd for $C_{20}H_{17}FN_2$: C, 78.92; H, 5.63; F, 6.24; N, 9.20; found: C, 78.93; H, 5.64; N, 9.23.



1-benzyl-2-(4-chlorobenzylidene)-1-phenylhydrazine (D7)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 150.8 mg, 94% yield. Purification by

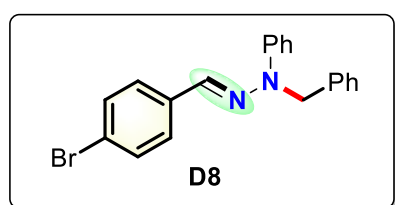
column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.59 – 7.50 (m, 2H), 7.44 – 7.24 (m, 12H), 6.97 (t, *J* = 7.2 Hz, 1H), 5.18 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.69, 135.39, 135.06, 133.32, 131.13, 129.18, 129.05, 128.66, 127.37, 127.25, 125.98, 121.03, 114.91, 50.61 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈ClN₂ [M+H]: 321.1159, found: 321.1155.

Elemental Analysis calcd for C₂₀H₁₇ClN₂: C, 74.88; H, 5.34; Cl, 11.05; N, 8.73; found: C, 74.85; H, 5.33; N, 8.74.



1-benzyl-2-(4-bromobenzylidene)-1-phenylhydrazine (D8)

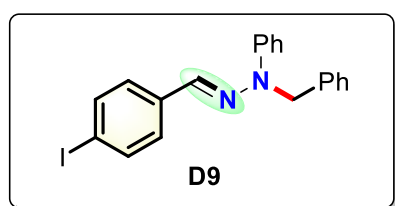
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 166.2 mg, 91% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.46 (q, *J* = 8.7 Hz, 4H), 7.41 – 7.29 (m, 7H), 7.23 (d, *J* = 7.1 Hz, 2H), 6.97 (t, *J* = 7.2 Hz, 1H), 5.18 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.70, 135.52, 135.38, 131.61, 131.13, 129.21, 129.07, 127.57, 127.40, 125.99, 121.52, 121.09, 114.95, 99.99, 50.63 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈BrN₂ [M+H]: 365.0653, found: 365.0655.

Elemental Analysis calcd for C₂₀H₁₇BrN₂: C, 65.76; H, 4.69; Br, 21.88; N, 7.67; found: C, 65.74; H, 4.66; N, 7.64.



1-benzyl-2-(4-iodobenzylidene)-1-phenylhydrazine (D9)

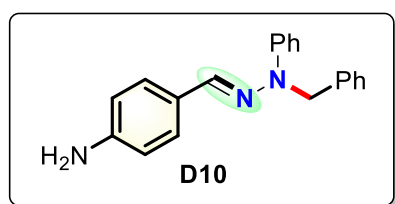
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 152.5 mg, 74% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.72 (d, $J = 8.4$ Hz, 2H), 7.57 (s, 1H), 7.49 – 7.38 (m, 4H), 7.33 (q, $J = 11.7, 8.0$ Hz, 4H), 7.25 (t, $J = 7.3$ Hz, 1H), 7.19 (d, $J = 7.2$ Hz, 2H), 6.92 (t, $J = 7.2$ Hz, 1H), 5.32 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.42, 137.86, 136.38, 136.22, 132.02, 129.68, 129.24, 128.30, 127.48, 126.70, 120.93, 114.71, 94.11, 48.67, 40.61, 40.40, 40.19, 39.98, 39.77, 39.56, 39.36 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{IN}_2$ [$\text{M}+\text{H}$]: 413.0515, found: 413.0517.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{IN}_2$: C, 58.27; H, 4.16; I, 30.78; N, 6.79; found: C, 58.21; H, 4.15; N, 6.73.



4-((2-benzyl-2-phenylhydrazineylidene)methyl)aniline (D10)

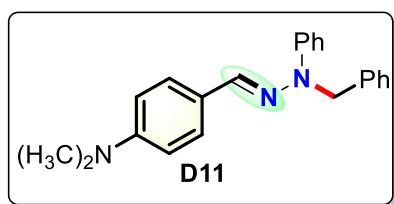
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 135.6 mg, 90% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.44 (s, 1H), 7.42 (s, 1H), 7.37 (d, $J = 7.9$ Hz, 2H), 7.34 – 7.28 (m, 5H), 7.26 (d, $J = 4.4$ Hz, 1H), 7.22 (dd, $J = 7.4, 3.6$ Hz, 3H), 6.90 (t, $J = 7.2$ Hz, 1H), 6.62 (d, $J = 8.5$ Hz, 2H), 5.12 (s, 2H), 3.69 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 148.12, 146.57, 136.21, 133.29, 129.14, 128.98, 127.60, 127.30, 127.20, 126.11, 120.10, 115.05, 114.42, 50.41 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{20}\text{N}_3$ [$\text{M}+\text{H}$]: 302.1657, found: 302.1656.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{19}\text{N}_3$: C, 79.70; H, 6.35; N, 13.94; found: C, 79.74; H, 6.35; N, 13.93.



4-((2-benzyl-2-phenylhydrazineylidene)methyl)-N,N-dimethylaniline (D11)

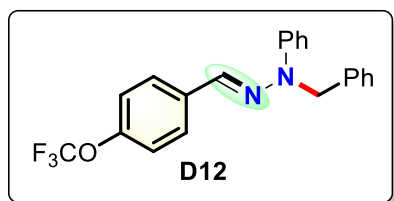
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 135.1 mg, 82% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.51 (d, J = 8.8 Hz, 2H), 7.37 (d, J = 8.9 Hz, 3H), 7.34 – 7.26 (m, 4H), 7.26 – 7.17 (m, 3H), 6.96 – 6.82 (m, 1H), 6.68 (d, J = 8.7 Hz, 2H), 5.14 (s, 2H), 2.95 (s, 6H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 150.48, 148.18, 136.35, 133.62, 129.12, 128.96, 127.41, 127.16, 126.14, 125.09, 119.92, 114.34, 112.34, 50.37, 40.53 ppm.

HRMS (ESI) calcd. for $\text{C}_{22}\text{H}_{24}\text{N}_3$ [$\text{M}+\text{H}$]: 330.1970, found: 330.1972.

Elemental Analysis calcd for $\text{C}_{22}\text{H}_{23}\text{N}_3$: C, 80.21; H, 7.04; N, 12.76; found: C, 80.23; H, 7.05; N, 12.75.



1-benzyl-1-phenyl-2-(4-(trifluoromethoxy)benzylidene)hydrazine (D12)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 179.6 mg, 97% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

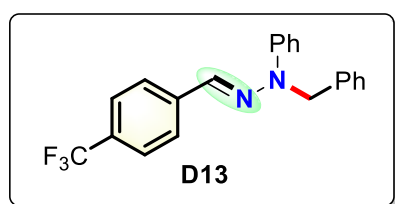
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.59 (d, J = 8.8 Hz, 2H), 7.38 (d, J = 7.8 Hz, 2H), 7.31 (t, J = 8.3, 6.1, 3.0 Hz, 5H), 7.26 (d, J = 7.2 Hz, 1H), 7.22 – 7.17 (m, 2H), 7.14 (d, J = 8.2 Hz, 2H), 6.96 (t, J = 7.2 Hz, 1H), 5.14 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.67, 148.66, 147.77, 135.42, 130.79, 129.29, 129.15, 127.48, 127.35, 126.05, 121.18, 121.15, 115.03, 50.61 ppm.

^{19}F NMR (377 MHz, 23.0 °C, CDCl_3) δ -57.73 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{18}\text{F}_3\text{N}_2\text{O}$ [M+H]: 371.1371, found: 371.1373.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{17}\text{F}_3\text{N}_2\text{O}$: C, 68.10; H, 4.63; F, 15.39; N, 7.56; O, 4.32; found: C, 68.17; H, 4.65; N, 7.51.



1-benzyl-1-phenyl-2-(4-(trifluoromethyl)benzylidene)hydrazine (D13)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 164.8 mg, 93% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

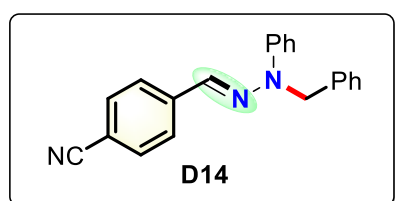
^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.70 (d, J = 8.2 Hz, 2H), 7.56 (d, J = 8.2 Hz, 2H), 7.42 (dd, J = 8.9, 1.1 Hz, 2H), 7.38 – 7.32 (m, 5H), 7.29 (d, J = 7.3 Hz, 1H), 7.24 (d, J = 7.0 Hz, 2H), 7.00 (t, J = 7.2 Hz, 1H), 5.21 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 147.58, 137.05, 135.11, 133.92, 130.58, 129.81, 129.25, 129.19, 129.12, 128.12, 127.48, 127.00, 126.11, 125.98, 125.74, 125.42, 122.25, 121.48, 118.91, 115.21, 101.79, 99.99, 50.72 ppm.

^{19}F NMR (377 MHz, 23.0 °C, CDCl_3) δ -62.42 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{18}\text{F}_3\text{N}_2$ [M+H]: 355.1422, found: 355.1425.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{17}\text{F}_3\text{N}_2$: C, 71.18; H, 4.84; F, 16.08; N, 7.91; found: C, 71.14; H, 4.86; N, 7.93.



4-((2-benzyl-2-phenylhydrazineylidene)methyl)benzonitrile (D14)

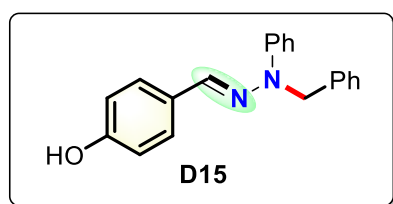
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 133.9 mg, 86% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.67 (d, $J = 8.3$ Hz, 2H), 7.58 (d, $J = 8.3$ Hz, 2H), 7.41 (d, $J = 8.1$ Hz, 2H), 7.37 – 7.30 (m, 5H), 7.22 (d, $J = 7.4$ Hz, 2H), 7.02 (t, $J = 7.2$ Hz, 1H), 5.21 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.40, 140.96, 134.84, 132.32, 129.96, 129.30, 129.18, 127.58, 126.31, 125.96, 121.90, 119.23, 115.43, 110.35, 50.89 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{18}\text{N}_3$ [$\text{M}+\text{H}$]: 312.1501, found: 312.1500.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{17}\text{N}_3$: C, 81.00; H, 5.50; N, 13.49; found: C, 81.02; H, 5.55; N, 13.42.



4-((2-benzyl-2-phenylhydrazineylidene)methyl)phenol (D15)

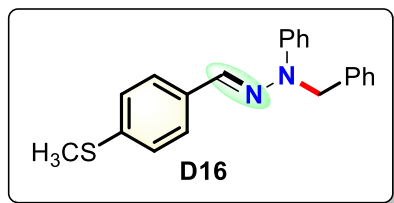
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 137.6 mg, 91% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.50 (d, $J = 8.6$ Hz, 2H), 7.41 – 7.19 (m, 11H), 6.92 (t, $J = 7.2$ Hz, 1H), 6.77 (d, $J = 8.6$ Hz, 2H), 5.13 (s, 2H), 4.92 (s, 1H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 155.53, 148.00, 135.95, 132.46, 129.66, 129.19, 129.03, 127.74, 127.29, 126.08, 120.47, 115.54, 114.64, 50.50 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{19}\text{N}_2\text{O}$ [$\text{M}+\text{H}$]: 303.1497, found: 303.1498.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{18}\text{N}_2\text{O}$: C, 79.44; H, 6.00; N, 9.26; O, 5.29; found: C, 79.43; H, 6.02; N, 9.24.



1-benzyl-2-(4-(methylthio)benzylidene)-1-phenylhydrazine (D16)

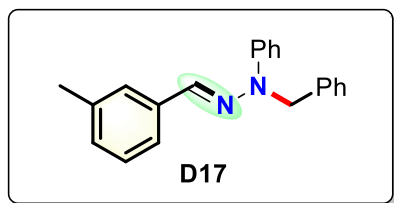
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 151.3 mg, 91% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.57 – 7.46 (m, 2H), 7.43 – 7.36 (m, 2H), 7.36 – 7.29 (m, 5H), 7.26 – 7.15 (m, 5H), 6.99 – 6.86 (m, 1H), 5.15 (s, 2H), 2.46 (s, 3H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.86, 138.14, 135.70, 133.63, 132.04, 129.20, 129.06, 128.47, 127.84, 127.34, 126.75, 126.60, 126.59, 126.07, 120.77, 114.80, 50.49, 15.89 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2\text{S}$ [$\text{M}+\text{H}$]: 333.1425, found: 333.1423.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2\text{S}$: C, 75.87; H, 6.06; N, 8.43; S, 9.64; found: C, 75.84; H, 6.03; N, 8.45.



1-benzyl-2-(3-methylbenzylidene)-1-phenylhydrazine (D17)

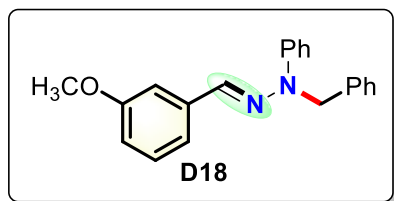
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 139.7 mg, 93% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.80 (d, $J = 7.6$ Hz, 1H), 7.51 (s, 1H), 7.36 (d, $J = 7.9$ Hz, 2H), 7.27 (t, $J = 8.8, 3.8, 3.2$ Hz, 4H), 7.21 – 7.18 (m, 3H), 7.16 – 6.97 (m, 3H), 6.89 (t, $J = 7.2$ Hz, 1H), 5.13 (s, 2H), 2.15 (s, 3H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.06, 135.66, 135.34, 134.35, 131.82, 130.64, 129.19, 129.01, 127.66, 127.32, 126.15, 126.09, 125.72, 120.73, 114.88, 50.68, 19.74 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2$ [M+H]: 301.1705, found: 301.1707.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2$: C, 83.96; H, 6.71; N, 9.33; found: C, 83.93; H, 6.73; N, 9.32.



1-benzyl-2-(3-methoxybenzylidene)-1-phenylhydrazine (D18)

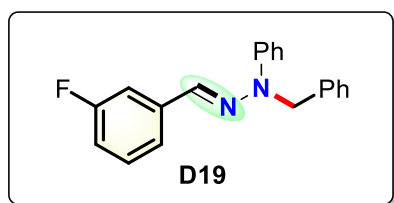
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 153.5 mg, 97% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.40 (d, $J = 7.9$ Hz, 2H), 7.38 – 7.28 (m, 5H), 7.25 (q, $J = 5.0, 4.6$ Hz, 5H), 7.15 (d, $J = 7.7$ Hz, 1H), 6.96 (t, $J = 7.2$ Hz, 1H), 6.80 (dd, $J = 8.0, 2.0$ Hz, 1H), 5.19 (s, 2H), 3.84 (s, 3H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 159.84, 147.82, 137.98, 135.59, 132.36, 129.48, 129.17, 129.03, 127.32, 126.04, 120.84, 119.28, 114.86, 113.96, 110.63, 55.29, 50.54 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2\text{O}$ [M+H]: 317.1654, found: 317.1658.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2\text{O}$: C, 79.72; H, 6.37; N, 8.85; O, 5.06; found: C, 79.76; H, 6.34; N, 8.86.



1-benzyl-2-(3-fluorobenzylidene)-1-phenylhydrazine (D19)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 135.4 mg, 89% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

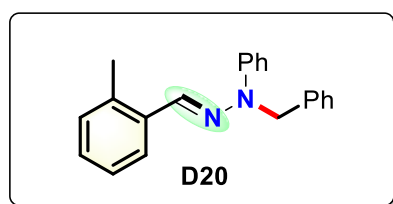
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.42 – 7.22 (m, 13H), 7.05 – 6.85 (m, 2H), 5.18 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 164.40, 161.97, 147.67, 139.02, 138.94, 135.33, 131.04, 131.01, 129.96, 129.88, 129.22, 129.08, 127.41, 126.00, 122.22, 122.19, 121.18, 115.02, 114.69, 114.48, 112.27, 112.04, 50.64 ppm.

$^{19}\text{F NMR}$ (377 MHz, 23.0 °C, CDCl_3) δ -113.45 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{FN}_2$ [M+H]: 305.1454, found: 305.1454.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{FN}_2$: C, 78.92; H, 5.63; F, 6.24; N, 9.20; found: C, 78.94; H, 5.65; N, 9.23.



1-benzyl-2-(2-methylbenzylidene)-1-phenylhydrazine (D20)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 138.2 mg, 92% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

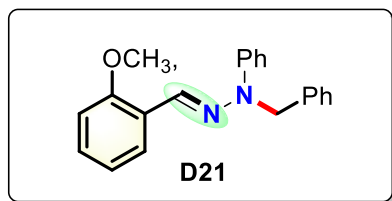
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.87 (d, $J = 7.4$ Hz, 1H), 7.58 (s, 1H), 7.43 (d, $J = 8.0$ Hz, 2H), 7.36 – 7.30 (m, 4H), 7.26 (t, $J = 3.1$ Hz, 2H), 7.23 – 7.11 (m, 3H), 7.09 (d, $J = 7.2$ Hz, 1H), 6.95 (t, $J = 7.2$ Hz, 1H), 5.19 (s, 2H), 2.22 (s, 3H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 148.07, 135.67, 135.35, 134.36, 131.82, 130.65, 129.21, 129.02, 128.77, 128.25, 127.67, 127.33, 126.16, 126.10, 125.73, 120.74, 114.88, 77.37, 77.05, 76.73, 50.68, 19.75, 0.04 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2$ [M+H]: 301.1705, found: 301.1704.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2$: C, 83.96; H, 6.71; N, 9.33; found: C, 83.92;

H, 6.73; N, 9.32.



1-benzyl-2-(2-methoxybenzylidene)-1-phenylhydrazine (D21)

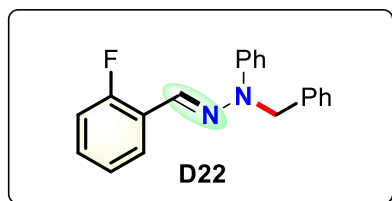
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 147.1 mg, 93% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 8.04 (dd, $J = 7.7, 1.6$ Hz, 1H), 7.82 (s, 1H), 7.41 (d, $J = 8.0$ Hz, 2H), 7.34 – 7.20 (m, 8H), 6.98 (t, $J = 7.5$ Hz, 1H), 6.93 (t, $J = 7.2$ Hz, 1H), 6.83 (d, $J = 8.2$ Hz, 1H), 5.19 (s, 2H), 3.72 (s, 3H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 156.99, 148.01, 135.96, 129.11, 128.87, 128.48, 127.14, 126.28, 125.50, 125.24, 120.90, 120.45, 114.69, 111.17, 55.63, 50.27 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2\text{O}$ [$\text{M}+\text{H}$]: 317.1654, found: 317.1655.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2\text{O}$: C, 79.72; H, 6.37; N, 8.85; O, 5.06; found: C, 79.73; H, 6.35; N, 8.83.



1-benzyl-2-(2-fluorobenzylidene)-1-phenylhydrazine (D22)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 121.7 mg, 80% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

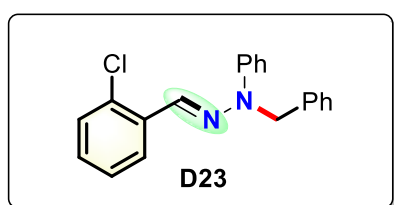
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 8.03 (t, $J = 7.6, 2.0$ Hz, 1H), 7.60 (s, 1H), 7.42 (d, $J = 7.8$ Hz, 2H), 7.34 (dd, $J = 5.7, 5.2, 3.3$ Hz, 4H), 7.27 – 7.10 (m, 6H), 7.04 – 6.91 (m, 2H), 5.20 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 161.71, 147.76, 135.26, 129.17, 129.00, 128.92, 128.84, 127.35, 126.10, 125.93, 125.38, 125.32, 124.16, 121.05, 115.59, 115.38, 114.96, 50.46 ppm.

^{19}F NMR (377 MHz, 23.0 °C, CDCl_3) δ -122.56 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{FN}_2$ [M+H]: 305.1454, found: 305.1456.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{FN}_2$: C, 78.92; H, 5.63; F, 6.24; N, 9.20; found: C, 78.94; H, 5.63; N, 9.23.



1-benzyl-2-(2-chlorobenzylidene)-1-phenylhydrazine (D23)

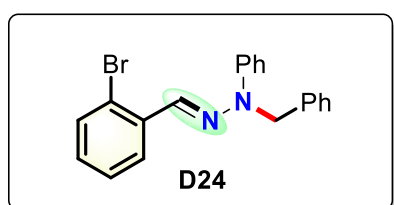
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 128.3 mg, 80% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 8.08 (dd, $J = 7.8, 1.5$ Hz, 1H), 7.76 (s, 1H), 7.44 (d, $J = 7.9$ Hz, 2H), 7.40 – 7.30 (m, 4H), 7.27 (d, $J = 6.8$ Hz, 6H), 7.15 (t, $J = 7.6, 1.7$ Hz, 1H), 6.99 (t, $J = 7.2$ Hz, 1H), 5.21 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 147.82, 135.28, 133.79, 132.82, 129.60, 129.38, 129.22, 129.01, 128.54, 127.41, 126.79, 126.31, 126.25, 121.20, 115.14, 50.91 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{ClN}_2$ [M+H]: 321.1159, found: 321.1158.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{ClN}_2$: C, 74.88; H, 5.34; Cl, 11.05; N, 8.73; found: C, 74.86; H, 5.34; N, 8.74.



1-benzyl-2-(2-bromobenzylidene)-1-phenylhydrazine (D24)

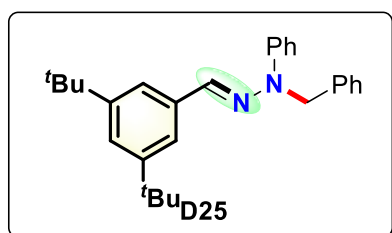
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 138.8 mg, 76% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 8.06 (dd, $J = 7.9, 1.6$ Hz, 1H), 7.71 (s, 1H), 7.50 – 7.42 (m, 3H), 7.37 – 7.31 (m, 4H), 7.30 – 7.24 (m, 4H), 7.12 – 7.03 (m, 1H), 6.99 (t, $J = 7.2$ Hz, 1H), 5.20 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.86, 135.28, 135.21, 132.85, 132.04, 129.24, 129.02, 128.87, 127.44, 127.39, 126.68, 126.32, 123.19, 121.24, 115.20, 51.09 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{BrN}_2$ [$\text{M}+\text{H}$]: 365.0653, found: 365.0653.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{BrN}_2$: C, 65.76; H, 4.69; Br, 21.88; N, 7.67; found: C, 65.74; H, 4.63; N, 7.64.



1-benzyl-2-(3,5-di-tert-butylbenzylidene)-1-phenylhydrazine (D25)

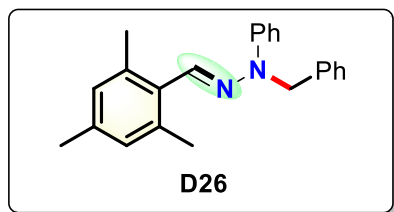
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 187.3 mg, 94% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.48 (d, $J = 1.8$ Hz, 2H), 7.43 (s, 1H), 7.39 (d, $J = 7.8$ Hz, 2H), 7.36 – 7.31 (m, 4H), 7.31 – 7.24 (m, 4H), 6.94 (t, $J = 7.2$ Hz, 1H), 5.19 (s, 2H), 1.34 (s, 18H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 150.98, 147.95, 135.89, 135.73, 133.55, 129.15, 129.00, 127.22, 126.09, 122.42, 120.58, 120.50, 114.66, 50.40, 34.86, 31.45 ppm.

HRMS (ESI) calcd. for $\text{C}_{28}\text{H}_{35}\text{N}_2$ [$\text{M}+\text{H}$]: 399.2800, found: 399.2803.

Elemental Analysis calcd for C₂₈H₃₄N₂: C, 84.37; H, 8.60; N, 7.03; found: C, 84.34; H, 8.62; N, 7.04.



1-benzyl-1-phenyl-2-(2,4,6-trimethylbenzylidene)hydrazine (D26)

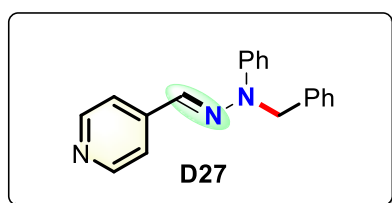
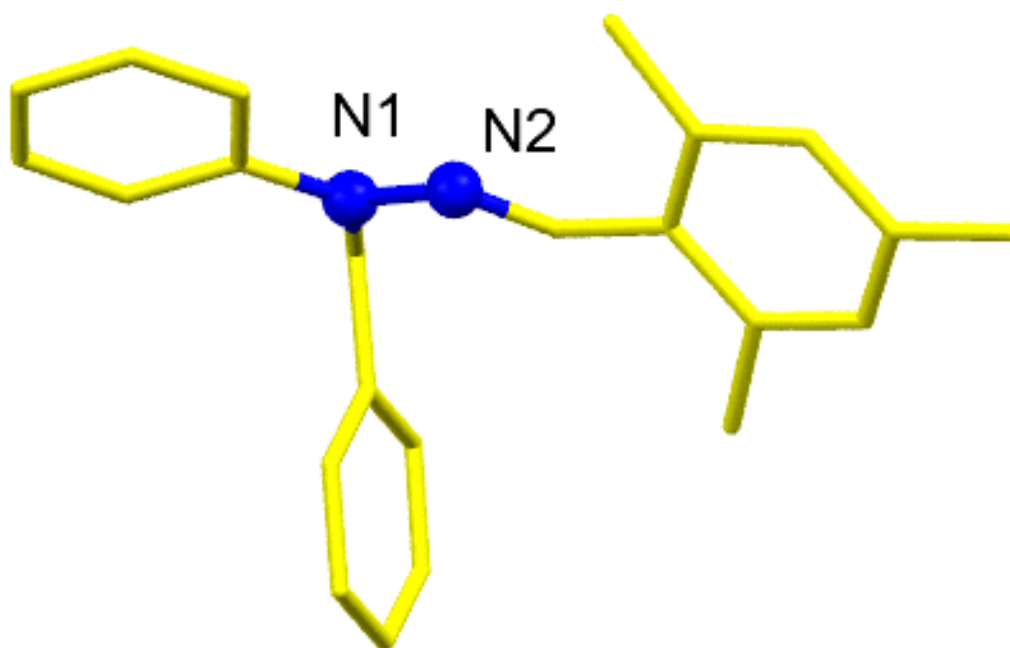
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 136.3 mg, 83% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.58 (s, 1H), 7.38 – 7.24 (m, 10H), 6.93 (t, *J* = 7.1 Hz, 1H), 6.83 (s, 2H), 5.21 (s, 2H), 2.27 (s, 6H), 2.25 (s, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 148.25, 136.95, 136.50, 135.75, 133.64, 130.06, 129.40, 129.18, 128.99, 127.27, 126.14, 120.37, 114.51, 50.19, 21.45, 21.01 ppm.

HRMS (ESI) calcd. for C₂₃H₂₅N₂ [M+H]: 329.2018, found: 329.2017.

Elemental Analysis calcd for C₂₃H₂₄N₂: C, 84.11; H, 7.37; N, 8.53; found: C, 84.14; H, 7.34; N, 8.52.



4-((2-benzyl-2-phenylhydrazineylidene)methyl)pyridine (D27)

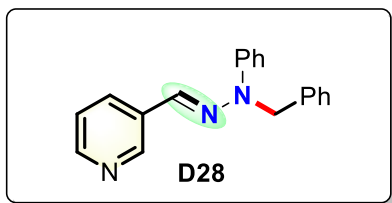
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 102.1 mg, 71% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 8.53 (d, $J = 4.9$ Hz, 2H), 7.48 – 7.21 (m, 12H), 7.03 (t, $J = 7.2$ Hz, 1H), 5.21 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 149.71, 147.34, 134.70, 129.30, 129.18, 127.60, 125.95, 122.05, 120.26, 115.53, 50.92 ppm.

HRMS (ESI) calcd. for $\text{C}_{19}\text{H}_{18}\text{N}_3$ [$\text{M}+\text{H}$]: 288.1501, found: 288.1500.

Elemental Analysis calcd for $\text{C}_{19}\text{H}_{17}\text{N}_3$: C, 79.41; H, 5.96; N, 14.62; found: C, 79.42; H, 5.94; N, 14.67.



3-((2-benzyl-2-phenylhydrazineylidene)methyl)pyridine (D28)

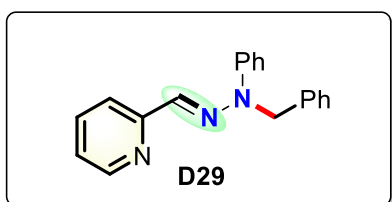
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 120.7 mg, 84% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 8.71 (s, 1H), 8.46 (d, $J = 3.9$ Hz, 1H), 8.02 (d, $J = 7.9, 1.8$ Hz, 1H), 7.44 – 7.33 (m, 6H), 7.26 (d, $J = 4.3$ Hz, 5H), 7.00 (t, $J = 7.2$ Hz, 1H), 5.21 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 148.49, 148.12, 147.57, 135.10, 132.44, 129.26, 129.14, 128.76, 127.51, 125.98, 123.55, 121.42, 115.10, 50.66 ppm.

HRMS (ESI) calcd. for $\text{C}_{19}\text{H}_{18}\text{N}_3$ [$\text{M}+\text{H}$]: 288.1501, found: 288.1501.

Elemental Analysis calcd for $\text{C}_{19}\text{H}_{17}\text{N}_3$: C, 79.41; H, 5.96; N, 14.62; found: C, 79.48; H, 5.94; N, 14.63.



2-((2-benzyl-2-phenylhydrazineylidene)methyl)pyridine (D29)

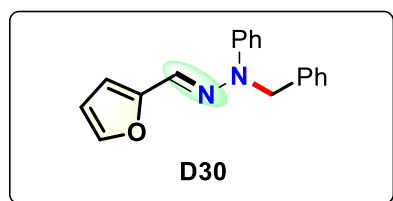
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 137.9 mg, 96% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 8.45 (dd, $J = 4.9, 1.7, 0.9$ Hz, 1H), 8.04 (dd, $J = 8.1, 0.9$ Hz, 1H), 7.65 (t, $J = 7.5, 1.3$ Hz, 1H), 7.43 (d, $J = 9.0, 1.6$ Hz, 2H), 7.39 – 7.29 (m, 4H), 7.26 – 7.20 (m, 3H), 7.10 (dd, $J = 7.4, 4.9, 1.2$ Hz, 1H), 7.06 – 6.92 (m, 1H), 5.22 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 155.72, 149.00, 147.59, 136.17, 134.92, 133.25, 129.24, 129.05, 127.42, 126.05, 122.10, 121.51, 119.31, 115.27, 50.80 ppm.

HRMS (ESI) calcd. for $\text{C}_{19}\text{H}_{18}\text{N}_3$ [M+H]: 288.1501, found: 288.1505.

Elemental Analysis calcd for $\text{C}_{19}\text{H}_{17}\text{N}_3$: C, 79.41; H, 5.96; N, 14.62; found: C, 79.45; H, 5.94; N, 14.63.



1-benzyl-2-(furan-2-ylmethylene)-1-phenylhydrazine (D30)

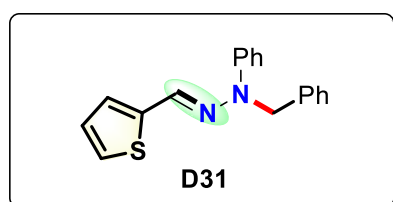
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 99.5 mg, 72% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.46 – 7.26 (m, 9H), 7.23 (d, J = 7.0 Hz, 2H), 6.96 (t, J = 7.2 Hz, 1H), 6.46 (d, J = 3.3 Hz, 1H), 6.41 (d, J = 1.8 Hz, 1H), 5.14 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 151.99, 147.61, 142.33, 135.31, 129.18, 129.06, 127.40, 126.02, 123.42, 121.04, 115.06, 111.49, 108.03, 50.72 ppm.

HRMS (ESI) calcd. for $\text{C}_{18}\text{H}_{17}\text{N}_2\text{O}$ [M+H]: 277.1341, found: 277.1340.

Elemental Analysis calcd for $\text{C}_{18}\text{H}_{16}\text{N}_2\text{O}$: C, 78.24; H, 5.84; N, 10.14; O, 5.79; found: C, 78.22; H, 5.88; N, 10.19.



1-benzyl-1-phenyl-2-(thiophen-2-ylmethylene)hydrazine (D31)

The title compound was prepared according to the general procedure and purified by

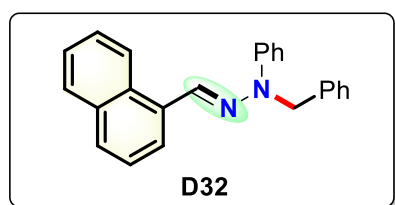
column chromatography to give a yellow solid 133.1 mg, 91% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.55 (s, 1H), 7.38 – 7.24 (m, 9H), 7.21 – 7.15 (m, 1H), 6.95 (t, *J* = 3.9 Hz, 3H), 5.15 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.47, 142.42, 135.45, 129.18, 129.07, 127.58, 127.39, 127.14, 126.02, 125.83, 125.09, 120.84, 114.74, 50.76 ppm.

HRMS (ESI) calcd. for C₁₈H₁₇N₂S [M+H]: 293.1112, found: 293.1110.

Elemental Analysis calcd for C₁₈H₁₆N₂S: C, 73.94; H, 5.52; N, 9.58; S, 10.96; found: C, 73.99; H, 5.51; N, 9.57.



(1-benzyl-2-(naphthalen-1-ylmethylene)-1-phenylhydrazine (D32)

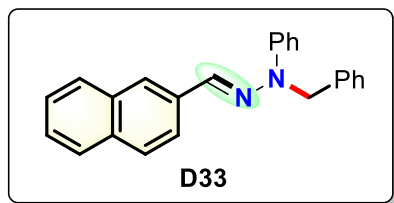
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 1146.4 mg, 87% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 8.28 (dd, *J* = 6.3, 3.5 Hz, 1H), 8.07 (s, 1H), 7.97 – 7.88 (m, 1H), 7.83 (dd, *J* = 6.2, 3.4 Hz, 1H), 7.76 (d, *J* = 8.2 Hz, 1H), 7.52 – 7.41 (m, 5H), 7.42 – 7.26 (m, 7H), 7.06 – 6.91 (m, 1H), 5.30 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 164.65, 162.19, 147.69, 138.58, 138.51, 136.33, 132.68, 130.70, 130.62, 129.23, 128.56, 128.04, 126.24, 121.65, 121.62, 121.01, 114.86, 114.48, 114.27, 113.24, 113.02, 50.14 ppm.

HRMS (ESI) calcd. for C₂₄H₂₁N₂ [M+H]: 337.1705, found: 337.1704.

Elemental Analysis calcd for C₂₄H₂₀N₂: C, 85.68; H, 5.99; N, 8.33; found: C, 85.65; H, 5.98; N, 8.36.



1-benzyl-2-(naphthalen-2-ylmethylene)-1-phenylhydrazine (D33)

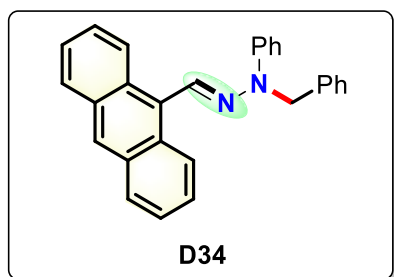
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 156.4 mg, 93% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 8.06 (dd, *J* = 8.6, 1.5 Hz, 1H), 7.87 – 7.71 (m, 4H), 7.56 (s, 1H), 7.52 – 7.39 (m, 4H), 7.36 (q, *J* = 8.8, 6.9, 4.7 Hz, 4H), 7.29 (t, *J* = 6.0 Hz, 3H), 6.98 (t, *J* = 7.2 Hz, 1H), 5.24 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.93, 135.73, 134.35, 133.57, 133.34, 132.74, 129.27, 129.12, 128.30, 127.95, 127.86, 127.40, 126.61, 126.32, 126.11, 125.96, 123.15, 120.92, 114.95, 50.60 ppm.

HRMS (ESI) calcd. for C₂₄H₂₁N₂ [M+H]: 337.1705, found: 337.1705.

Elemental Analysis calcd for C₂₄H₂₀N₂: C, 85.68; H, 5.99; N, 8.33; found: C, 85.67; H, 5.96; N, 8.34.



2-(anthracen-9-ylmethylene)-1-benzyl-1-phenylhydrazine (D34)

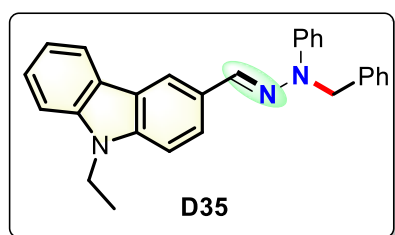
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 160.4 mg, 83% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 8.57 (s, 1H), 8.48 (s, 1H), 8.24 (d, *J* = 8.8 Hz, 2H), 8.09 (d, *J* = 8.1 Hz, 2H), 7.64 – 7.20 (m, 12H), 6.95 (t, *J* = 7.2 Hz, 1H), 5.64 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.01, 136.52, 131.98, 131.52, 129.83, 129.52, 129.45, 129.20, 128.40, 127.78, 127.64, 126.97, 126.68, 126.33, 125.81, 125.51, 123.96, 120.99, 114.69, 48.88, 40.61, 40.40, 40.19, 39.98, 39.77, 39.57, 39.36 ppm.

HRMS (ESI) calcd. for $\text{C}_{28}\text{H}_{23}\text{N}_2$ [M+H]: 387.1861, found: 387.1863.

Elemental Analysis calcd for $\text{C}_{28}\text{H}_{22}\text{N}_2$: C, 87.01; H, 5.74; N, 7.25; found: C, 87.04; H, 5.75; N, 7.27.



3-((2-benzyl-2-phenylhydrazineylidene)methyl)-9-ethyl-9H-carbazole (D35)

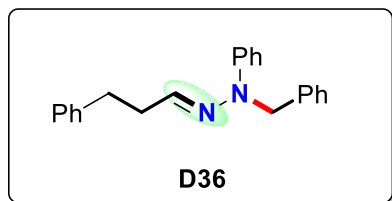
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 185.6 mg, 92% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 8.35 – 8.17 (m, 1H), 8.09 (d, J = 7.7 Hz, 1H), 7.88 (dd, J = 8.6, 1.5 Hz, 1H), 7.63 (s, 1H), 7.51 – 7.42 (m, 3H), 7.41 – 7.28 (m, 8H), 7.22 (t, J = 7.4 Hz, 1H), 6.94 (t, J = 7.2 Hz, 1H), 5.23 (s, 2H), 4.35 (q, J = 7.2 Hz, 2H), 1.42 (t, J = 7.2 Hz, 3H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.16, 140.33, 140.01, 136.20, 134.21, 129.17, 129.01, 127.83, 127.23, 126.15, 125.77, 124.07, 123.11, 123.04, 120.59, 120.24, 119.04, 118.92, 114.60, 108.62, 50.62, 37.66, 13.84 ppm.

HRMS (ESI) calcd. for $\text{C}_{28}\text{H}_{26}\text{N}_3$ [M+H]: 404.2127, found: 404.2126.

Elemental Analysis calcd for $\text{C}_{28}\text{H}_{25}\text{N}_3$: C, 83.34; H, 6.24; N, 10.41; found: C, 83.38; H, 6.29; N, 10.47.



1-benzyl-1-phenyl-2-(3-phenylpropylidene)hydrazine (D36)

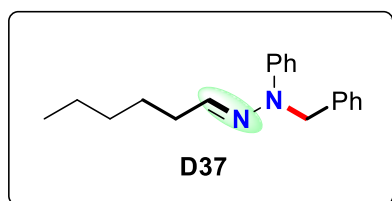
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 111.6 mg, 71% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.46 – 7.26 (m, 6H), 7.25 – 7.21 (m, 3H), 7.19 – 7.12 (m, 3H), 7.12 – 7.06 (m, 2H), 6.92 – 6.81 (m, 1H), 6.72 (t, $J = 5.0$ Hz, 1H), 4.98 (s, 2H), 2.81 (t, $J = 7.7$ Hz, 2H), 2.67 – 2.51 (m, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 148.22, 141.49, 136.19, 135.35, 129.12, 128.99, 128.90, 128.55, 128.51, 128.45, 128.36, 128.32, 127.12, 126.09, 125.87, 119.97, 114.25, 50.47, 34.74, 33.78 ppm.

HRMS (ESI) calcd. for $\text{C}_{22}\text{H}_{23}\text{N}_2$ [$\text{M}+\text{H}$]: 315.1861, found: 315.1863.

Elemental Analysis calcd for $\text{C}_{22}\text{H}_{22}\text{N}_2$: C, 84.04; H, 7.05; N, 8.91; found: C, 84.08; H, 7.02; N, 8.95.



1-benzyl-2-pentylidene-1-phenylhydrazine (D37)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 83.9 mg, 63% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

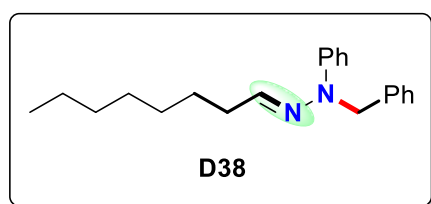
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.31 (t, $J = 7.3$ Hz, 2H), 7.27 – 7.21 (m, 5H), 7.16 (d, $J = 7.1$ Hz, 2H), 6.86 (dd, $J = 8.4, 5.3, 3.2$ Hz, 1H), 6.70 (t, $J = 5.3$ Hz, 1H), 4.98 (s, 2H), 2.32 – 2.23 (m, 2H), 1.44 (p, $J = 7.2$ Hz, 2H), 1.30 (p, $J = 14.7, 7.2$ Hz,

2H), 0.88 (t, $J = 7.3$ Hz, 3H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.57, 137.14, 136.67, 129.39, 129.13, 127.35, 126.37, 120.06, 114.43, 50.73, 32.95, 29.83, 22.51, 14.24 ppm.

HRMS (ESI) calcd. for $\text{C}_{18}\text{H}_{23}\text{N}_2$ [M+H]: 267.1861, found: 267.1860.

Elemental Analysis calcd for $\text{C}_{18}\text{H}_{22}\text{N}_2$: C, 81.16; H, 8.32; N, 10.52; found: C, 81.17; H, 8.34; N, 10.51.



1-benzyl-2-octylidene-1-phenylhydrazine (D38)

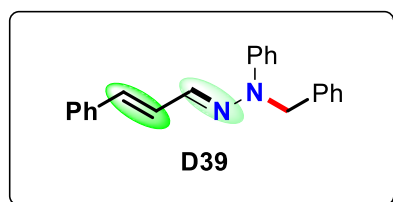
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 100.1 mg, 68% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.53 – 7.07 (m, 13H), 6.85 (dd, $J = 6.4, 2.9$ Hz, 1H), 6.69 (t, $J = 5.2$ Hz, 1H), 4.98 (s, 2H), 2.37 – 2.12 (m, 2H), 1.49 – 1.41 (m, 2H), 1.26 (d, $J = 13.8$ Hz, 10H), 0.87 (t, $J = 6.8$ Hz, 3H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.30, 136.92, 136.39, 129.10, 128.97, 128.85, 128.51, 128.26, 128.03, 127.44, 127.07, 126.09, 119.77, 114.16, 113.53, 50.44, 32.97, 31.83, 29.16, 29.08, 27.45, 22.68, 14.15 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{27}\text{N}_2$ [M+H]: 295.2174, found: 295.2172.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{26}\text{N}_2$: C, 81.58; H, 8.90; N, 9.51; found: C, 81.59; H, 8.93; N, 9.56.



1-benzyl-1-phenyl-2-(3-phenylallylidene)hydrazine (D39)

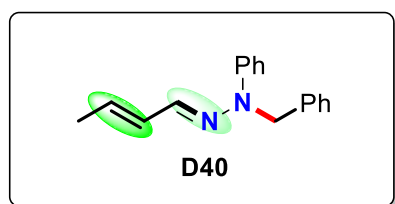
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 126.5 mg, 81% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.42 – 7.28 (m, 12H), 7.24 – 7.18 (m, 3H), 7.10 – 7.01 (m, 1H), 6.95 (t, $J = 6.8$ Hz, 1H), 6.56 (d, $J = 16.0$ Hz, 1H), 5.13 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.56, 137.04, 135.63, 135.41, 133.35, 129.17, 129.05, 128.67, 127.66, 127.39, 127.34, 126.34, 126.01, 120.93, 114.90, 50.70 ppm.

HRMS (ESI) calcd. for $\text{C}_{22}\text{H}_{21}\text{N}_2$ [M+H]: 313.1705, found: 313.1704.

Elemental Analysis calcd for $\text{C}_{22}\text{H}_{20}\text{N}_2$: C, 84.58; H, 6.45; N, 8.97; found: C, 84.57; H, 6.44; N, 8.91.



1-benzyl-2-but-2-en-1-ylidene-1-phenylhydrazine (D40)

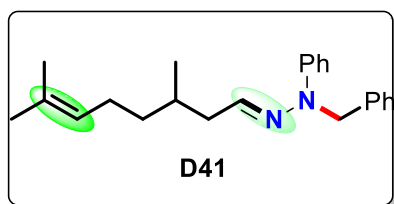
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 100.1 mg, 80% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.31 – 7.19 (m, 7H), 7.16 (t, $J = 6.8$ Hz, 2H), 7.09 (d, $J = 8.8$ Hz, 1H), 6.88 (t, $J = 8.4, 5.9, 2.4$ Hz, 1H), 6.31 (dd, $J = 15.5, 8.8, 1.6$ Hz, 1H), 5.71 (dd, $J = 15.5, 6.8$ Hz, 1H), 5.01 (s, 2H), 1.77 (d, $J = 6.8$ Hz, 3H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.86, 136.09, 135.78, 131.68, 130.33, 129.16, 129.01, 127.25, 126.07, 120.40, 114.59, 50.45, 18.39 ppm.

HRMS (ESI) calcd. for $\text{C}_{17}\text{H}_{19}\text{N}_2$ [M+H]: 251.1548, found: 251.1546.

Elemental Analysis calcd for $\text{C}_{17}\text{H}_{18}\text{N}_2$: C, 81.56; H, 7.25; N, 11.19; found: C, 81.58; H, 7.22; N, 11.15.



1-benzyl-2-(3,7-dimethyloct-6-en-1-ylidene)-1-phenylhydrazine (D41)

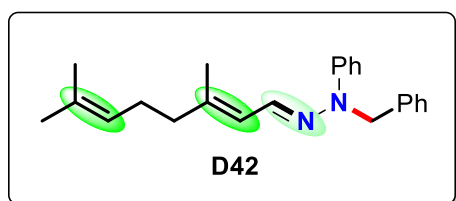
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 118.8 mg, 71% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.48 – 7.26 (m, 8H), 7.17 (d, *J* = 7.1 Hz, 2H), 6.93 – 6.80 (m, 1H), 6.67 (t, *J* = 5.6 Hz, 1H), 5.00 (s, 2H), 2.27 (d, *J* = 14.1, 5.7 Hz, 1H), 2.13 (dd, *J* = 14.0, 7.4, 5.9 Hz, 1H), 1.94 (d, *J* = 16.4, 8.7 Hz, 2H), 1.67 – 1.64 (m, 3H), 1.57 (s, 3H), 1.34 – 1.21 (m, 2H), 1.16 – 1.08 (m, 1H), 0.82 (d, *J* = 6.7 Hz, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 163.3, 160.0, 141.5, 141.5, 127.1, 127.0, 115.4, 115.2, 69.8, 25.3 ppm.

HRMS (ESI) calcd. for C₂₃H₃₁N₂ [M+H]: 335.2487, found: 335.2485.

Elemental Analysis calcd for C₂₃H₃₀N₂: C, 82.59; H, 9.04; N, 8.37; found: C, 82.57; H, 9.05; N, 8.36.



1-benzyl-2-(3,7-dimethylocta-2,6-dien-1-ylidene)-1-phenylhydrazine (D42)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 136.3 mg, 82% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

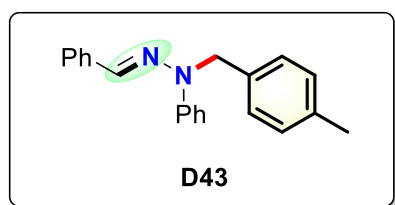
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.39 – 7.13 (m, 11H), 6.94 – 6.80 (m, 1H), 5.13

– 5.07 (m, 1H), 5.03 (d, $J = 5.0$ Hz, 2H), 2.09 (q, $J = 11.7, 8.4$ Hz, 2H), 2.05 – 1.97 (m, 1H), 1.92 (q, $J = 7.2$ Hz, 1H), 1.85 – 1.76 (m, 2H), 1.67 (s, 2H), 1.61 (s, 3H), 1.59 (s, 1H), 1.46 (s, 1H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.03, 141.79, 141.49, 136.26, 136.24, 133.53, 132.09, 131.94, 129.17, 129.04, 129.02, 128.49, 127.86, 127.28, 127.26, 126.18, 126.11, 123.93, 123.89, 123.66, 123.05, 120.36, 120.32, 114.68, 114.66, 50.88, 50.83, 40.14, 32.81, 26.78, 26.54, 25.81, 25.80, 24.23, 17.80, 17.67, 17.06 ppm.

HRMS (ESI) calcd. for $\text{C}_{23}\text{H}_{29}\text{N}_2$ [M+H]: 333.2331, found: 333.2330.

Elemental Analysis calcd for $\text{C}_{23}\text{H}_{28}\text{N}_2$: C, 83.09; H, 8.49; N, 8.43; found: C, 83.08; H, 8.45; N, 8.41.



2-benzylidene-1-(4-methylbenzyl)-1-phenylhydrazine (D43)

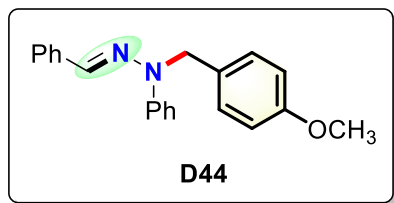
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 136.7 mg, 91% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.60 (d, $J = 7.9$ Hz, 2H), 7.42 – 7.34 (m, 3H), 7.30 (t, $J = 7.1$ Hz, 4H), 7.25 – 7.19 (m, 1H), 7.11 (s, 4H), 7.03 – 6.87 (m, 1H), 5.11 (s, 2H), 2.30 (s, 3H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.00, 137.01, 136.70, 132.63, 132.51, 129.79, 129.25, 128.60, 126.29, 126.08, 120.77, 114.88, 50.27, 21.16 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2$ [M+H]: 301.1705, found: 301.1705.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2$: C, 83.96; H, 6.71; N, 9.33; found: C, 83.97; H, 6.74; N, 9.31.



2-benzylidene-1-(4-methoxybenzyl)-1-phenylhydrazine (D44)

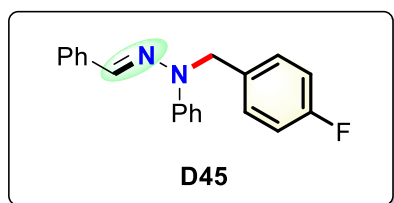
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 142.4 mg, 90% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.62 (d, *J* = 7.7 Hz, 2H), 7.43 – 7.37 (m, 3H), 7.32 (td, *J* = 8.8, 8.1, 2.6 Hz, 4H), 7.26 – 7.20 (m, 1H), 7.14 (d, *J* = 8.5 Hz, 2H), 6.94 (t, *J* = 7.2 Hz, 1H), 6.86 (d, *J* = 8.6 Hz, 2H), 5.11 (s, 2H), 3.77 (s, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 166.49, 158.84, 147.88, 136.59, 132.49, 129.15, 128.51, 127.82, 127.40, 127.20, 126.17, 122.34, 120.70, 115.01, 114.82, 114.43, 99.99, 55.31, 49.92 ppm.

HRMS (ESI) calcd. for C₂₁H₂₁N₂O [M+H]: 317.1654, found: 317.1653.

Elemental Analysis calcd for C₂₁H₂₀N₂O: C, 79.72; H, 6.37; N, 8.85; O, 5.06; found: C, 79.74; H, 6.35; N, 8.86.



2-benzylidene-1-(4-fluorobenzyl)-1-phenylhydrazine (D45)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 146.1 mg, 96% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.64 – 7.53 (m, 2H), 7.37 (dd, *J* = 8.9, 1.1 Hz, 2H), 7.33 – 7.26 (m, 5H), 7.26 – 7.15 (m, 2H), 7.03 – 6.83 (m, 4H), 5.07 (s, 2H) ppm.

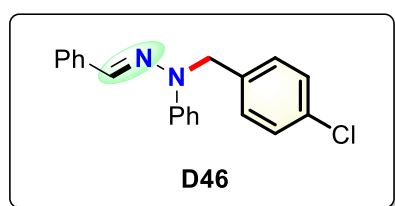
¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 164.75, 162.29, 147.80, 138.72, 138.65,

136.47, 132.78, 130.83, 130.74, 129.37, 128.70, 128.17, 126.38, 121.78, 121.75, 121.14, 114.97, 114.58, 114.37, 113.35, 113.13, 50.15, 50.13 ppm.

^{19}F NMR (377 MHz, 23.0 °C, CDCl_3) δ -111.83 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{FN}_2$ [M+H]: 305.1454, found: 305.1455.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{FN}_2$: C, 78.92; H, 5.63; F, 6.24; N, 9.20; found: C, 78.94; H, 5.65; N, 9.24.



2-benzylidene-1-(4-chlorobenzyl)-1-phenylhydrazine (D46)

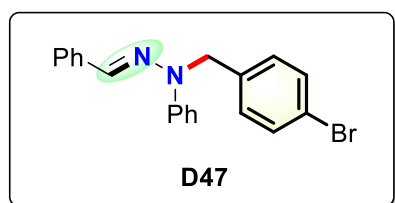
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 150.8 mg, 94% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.65 – 7.54 (m, 2H), 7.37 – 7.23 (m, 10H), 7.12 (d, J = 8.4 Hz, 2H), 6.94 (t, J = 7.1 Hz, 1H), 5.07 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 147.73, 136.40, 134.25, 133.16, 132.72, 129.34, 129.28, 129.15, 128.70, 128.66, 128.13, 127.58, 126.31, 121.09, 114.93, 49.92 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{ClN}_2$ [M+H]: 321.1159, found: 321.1155.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{ClN}_2$: C, 74.88; H, 5.34; Cl, 11.05; N, 8.73; found: C, 74.87; H, 5.35; N, 8.74.



2-benzylidene-1-(4-bromobenzyl)-1-phenylhydrazine (D47)

The title compound was prepared according to the general procedure and purified by

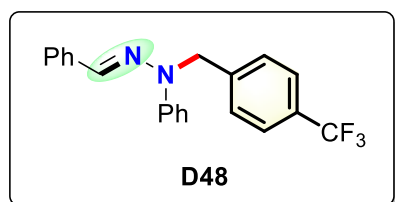
column chromatography to give a yellow solid 164.4 mg, 90% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.59 (dd, *J* = 8.2, 1.1 Hz, 2H), 7.44 – 7.39 (m, 2H), 7.35 – 7.28 (m, 6H), 7.24 – 7.20 (m, 1H), 7.06 (d, *J* = 8.5 Hz, 2H), 6.97 – 6.91 (m, 1H), 5.04 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.72, 136.39, 134.80, 132.74, 132.23, 129.35, 128.67, 128.15, 127.96, 126.33, 121.22, 121.11, 114.93, 49.98 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈BrN₂ [M+H]: 365.0653, found: 365.0653.

Elemental Analysis calcd for C₂₀H₁₇BrN₂: C, 65.76; H, 4.69; Br, 21.88; N, 7.67; found: C, 65.73; H, 4.64; N, 7.63.



2-benzylidene-1-phenyl-1-(4-(trifluoromethyl)benzyl)hydrazine (D48)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 166.6 mg, 94% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

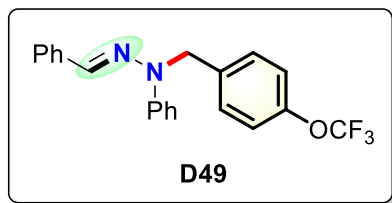
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.64 – 7.55 (m, 4H), 7.38 – 7.27 (m, 9H), 7.27 – 7.21 (m, 1H), 6.96 (t, *J* = 7.1 Hz, 1H), 5.16 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.66, 140.04, 140.03, 136.27, 132.78, 129.99, 129.67, 129.37, 128.67, 128.59, 128.22, 126.53, 126.33, 126.26, 126.16, 126.13, 126.09, 126.05, 125.52, 122.82, 121.23, 114.94, 50.15 ppm.

¹⁹F NMR (377 MHz, 23.0 °C, CDCl₃) δ -62.35 ppm.

HRMS (ESI) calcd. for C₂₁H₁₈F₃N₂ [M+H]: 355.1422, found: 355.1425.

Elemental Analysis calcd for C₂₁H₁₇F₃N₂: C, 71.18; H, 4.84; F, 16.08; N, 7.91; found: C, 71.16; H, 4.86; N, 7.93.



2-benzylidene-1-phenyl-1-(4-(trifluoromethoxy)benzyl)hydrazine (D49)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 159.3 mg, 86% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

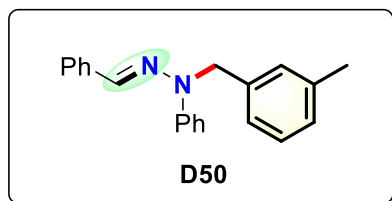
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.63 (d, *J* = 7.3 Hz, 2H), 7.40 – 7.32 (m, 6H), 7.27 (d, *J* = 9.3 Hz, 4H), 7.19 (d, *J* = 8.3 Hz, 2H), 6.97 (t, *J* = 7.1 Hz, 1H), 5.18 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 148.44, 147.63, 136.30, 134.37, 132.65, 129.26, 128.58, 128.08, 127.49, 126.24, 121.63, 121.06, 114.86, 53.44, 49.83 ppm.

¹⁹F NMR (377 MHz, 23.0 °C, CDCl₃) δ -57.86 ppm.

HRMS (ESI) calcd. for C₂₁H₁₈F₃N₂O [M+H]: 371.1371, found: 371.1373.

Elemental Analysis calcd for C₂₁H₁₇F₃N₂O: C, 68.10; H, 4.63; F, 15.39; N, 7.56; O, 4.32; found: C, 68.15; H, 4.64; N, 7.53.



2-benzylidene-1-(3-methylbenzyl)-1-phenylhydrazine (D50)

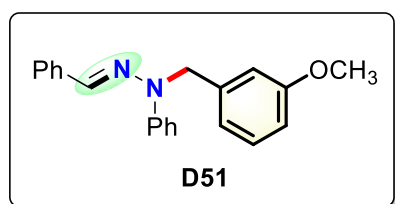
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 132.2 mg, 88% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.61 (d, *J* = 7.5 Hz, 2H), 7.40 (d, *J* = 8.0 Hz, 2H), 7.36 (s, 1H), 7.30 (t, *J* = 7.7 Hz, 4H), 7.24 – 7.16 (m, 2H), 7.03 (q, *J* = 8.1 Hz, 3H), 6.93 (t, *J* = 7.2 Hz, 1H), 5.10 (s, 2H), 2.29 (s, 3H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.17, 139.00, 136.83, 135.92, 132.65, 129.39, 129.14, 128.74, 128.32, 128.06, 126.77, 126.44, 123.32, 120.93, 115.02, 50.75, 21.73 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2$ [M+H]: 301.1705, found: 301.1705.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2$: C, 83.96; H, 6.71; N, 9.33; found: C, 83.98; H, 6.75; N, 9.32.



2-benzylidene-1-(3-methoxybenzyl)-1-phenylhydrazine (D51)

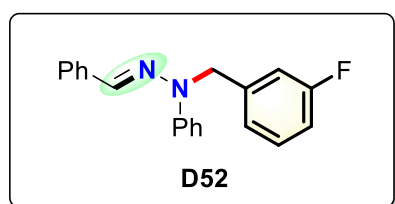
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 144.1 mg, 91% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.65 – 7.59 (m, 2H), 7.43 – 7.36 (m, 3H), 7.34 – 7.27 (m, 4H), 7.26 – 7.20 (m, 2H), 6.98 – 6.91 (m, 1H), 6.85 – 6.75 (m, 3H), 5.13 (s, 2H), 3.74 (s, 3H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 160.29, 147.95, 137.54, 136.57, 132.59, 130.16, 129.20, 128.56, 127.92, 126.26, 120.82, 118.30, 114.89, 112.58, 111.72, 55.27, 50.63 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2\text{O}$ [M+H]: 317.1654, found: 317.1653.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2\text{O}$: C, 79.72; H, 6.37; N, 8.85; O, 5.06; found: C, 79.73; H, 6.34; N, 8.82.



2-benzylidene-1-(3-fluorobenzyl)-1-phenylhydrazine (D52)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 140.2 mg, 92% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

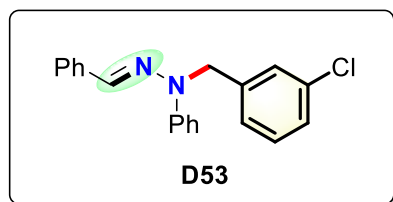
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 8.28 (q, *J* = 6.8, 3.3 Hz, 1H), 8.07 (s, 1H), 7.96 – 7.89 (m, 1H), 7.83 (dd, *J* = 6.2, 3.4 Hz, 1H), 7.76 (d, *J* = 8.2 Hz, 1H), 7.49 – 7.44 (m, 4H), 7.38 – 7.32 (m, 5H), 6.98 (t, *J* = 7.3 Hz, 1H), 5.30 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 164.65, 162.19, 147.69, 138.58, 138.52, 136.33, 132.68, 130.70, 130.62, 129.23, 128.56, 128.04, 126.24, 121.65, 121.62, 121.01, 114.87, 114.48, 114.27, 113.24, 113.02, 50.15 ppm.

¹⁹F NMR (377 MHz, 23.0 °C, CDCl₃) δ -112.10 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈FN₂ [M+H]: 305.1454, found: 305.1455.

Elemental Analysis calcd for C₂₀H₁₇FN₂: C, 78.92; H, 5.63; F, 6.24; N, 9.20; found: C, 78.93; H, 5.65; N, 9.22.



2-benzylidene-1-(3-chlorobenzyl)-1-phenylhydrazine (D53)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 146.1 mg, 91% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

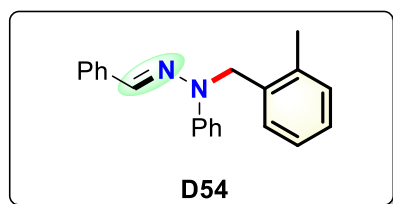
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.61 (d, *J* = 7.7 Hz, 2H), 7.38 – 7.28 (m, 7H), 7.22 (t, *J* = 7.2 Hz, 4H), 7.09 – 7.03 (m, 1H), 6.95 (t, *J* = 7.1 Hz, 1H), 5.08 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.74, 138.07, 136.39, 135.11, 132.75, 130.45, 129.34, 128.66, 128.15, 127.74, 126.35, 126.25, 124.30, 121.13, 114.94, 50.14 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈ClN₂ [M+H]: 321.1159, found: 321.1155.

Elemental Analysis calcd for C₂₀H₁₇ClN₂: C, 74.88; H, 5.34; Cl, 11.05; N, 8.73; found:

C, 74.85; H, 5.31; N, 8.73.



2-benzylidene-1-(2-methylbenzyl)-1-phenylhydrazine (D54)

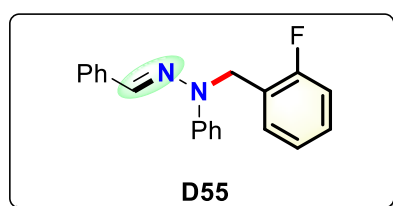
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 124.7 mg, 83% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.73 – 7.51 (m, 2H), 7.40 – 7.28 (m, 6H), 7.26 – 7.17 (m, 4H), 7.08 (t, *J* = 7.4 Hz, 1H), 7.01 – 6.84 (m, 2H), 5.06 (s, 2H), 2.44 (s, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.80, 136.56, 134.68, 132.62, 132.24, 130.54, 129.14, 128.53, 127.86, 127.26, 126.57, 126.22, 125.80, 125.60, 120.73, 114.77, 77.36, 77.05, 76.73, 49.02, 19.09, 0.03 ppm.

HRMS (ESI) calcd. for C₂₁H₂₁N₂ [M+H]: 301.1705, found: 301.1705.

Elemental Analysis calcd for C₂₁H₂₀N₂: C, 83.96; H, 6.71; N, 9.33; found: C, 83.99; H, 6.76; N, 9.32.



2-benzylidene-1-(2-fluorobenzyl)-1-phenylhydrazine (D55)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 124.7 mg, 83% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.68 – 7.55 (m, 2H), 7.37 (d, *J* = 7.9 Hz, 2H), 7.34 – 7.26 (m, 5H), 7.22 – 7.16 (m, 2H), 7.09 (dd, *J* = 10.6, 7.0 Hz, 1H), 7.01 – 6.90

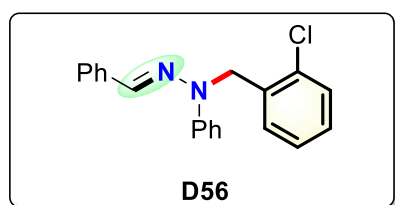
(m, 3H), 5.15 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 161.79, 159.36, 147.74, 136.54, 132.61, 130.23, 129.39, 129.21, 129.13, 128.71, 128.59, 128.16, 128.09, 128.05, 127.93, 126.43, 124.76, 124.72, 122.53, 122.39, 121.08, 115.76, 115.56, 114.92, 44.45, 44.40 ppm.

^{19}F NMR (377 MHz, 23.0 °C, CDCl_3) δ -117.56 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{FN}_2$ [M+H]: 305.1454, found: 305.1455.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{FN}_2$: C, 78.92; H, 5.63; F, 6.24; N, 9.20; found: C, 78.95; H, 5.67; N, 9.23.



2-benzylidene-1-(2-chlorobenzyl)-1-phenylhydrazine (D56)

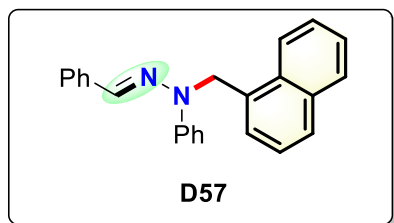
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 138.1 mg, 86% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.61 (d, J = 7.4 Hz, 2H), 7.43 (d, J = 7.4 Hz, 1H), 7.38 – 7.28 (m, 6H), 7.26 – 7.17 (m, 3H), 7.09 (t, J = 7.4 Hz, 1H), 7.04 – 6.97 (m, 1H), 6.94 (t, J = 7.1 Hz, 1H), 5.16 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 147.58, 136.43, 132.65, 132.58, 132.41, 129.89, 129.31, 128.81, 128.64, 128.11, 127.68, 127.43, 126.38, 121.01, 114.77, 48.59 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{ClN}_2$ [M+H]: 321.1159, found: 321.1155.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{ClN}_2$: C, 74.88; H, 5.34; Cl, 11.05; N, 8.73; found: C, 74.89; H, 5.36; N, 8.71.



2-benzylidene-1-(naphthalen-1-ylmethyl)-1-phenylhydrazine (D57)

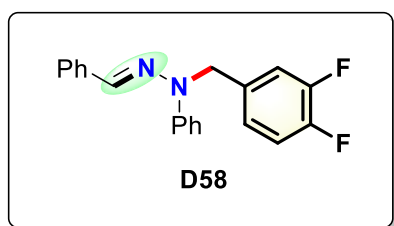
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 136.3 mg, 81% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 8.27 (d, *J* = 8.3 Hz, 1H), 8.02 (d, *J* = 7.6 Hz, 1H), 7.84 (d, *J* = 8.2 Hz, 1H), 7.79 – 7.58 (m, 4H), 7.52 (s, 1H), 7.50 – 7.22 (m, 8H), 7.03 – 6.81 (m, 2H), 5.76 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.52, 136.73, 134.15, 133.08, 130.81, 130.07, 129.67, 129.16, 129.09, 128.35, 127.88, 126.73, 126.54, 126.44, 126.01, 123.92, 122.74, 120.71, 114.60, 47.37, 40.62, 40.41, 40.21, 40.00, 39.79, 39.58, 39.37 ppm.

HRMS (ESI) calcd. for C₂₄H₂₁N₂ [M+H]: 337.1705, found: 337.1705.

Elemental Analysis calcd for C₂₄H₂₀N₂: C, 85.68; H, 5.99; N, 8.33; found: C, 85.67; H, 5.95; N, 8.37.



2-benzylidene-1-(3,4-difluorobenzyl)-1-phenylhydrazine (D58)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 148.3 mg, 92% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

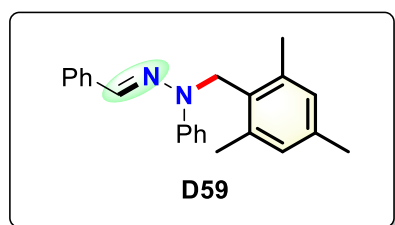
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.71 – 7.53 (m, 2H), 7.41 – 7.27 (m, 7H), 7.27 – 7.19 (m, 1H), 7.13 – 6.90 (m, 4H), 5.06 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 152.19, 152.06, 150.94, 150.81, 149.71, 149.59, 148.48, 148.35, 147.60, 136.28, 132.85, 132.82, 129.38, 128.68, 128.24, 126.34, 122.11, 122.07, 122.05, 122.01, 121.26, 118.07, 117.90, 115.32, 115.15, 114.97, 49.62 ppm.

^{19}F NMR (377 MHz, 23.0 °C, CDCl_3) δ -136.32, -136.37, -139.38, -139.43 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{17}\text{F}_2\text{N}_2$ [M+H]: 323.1360, found: 323.1359.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{16}\text{F}_2\text{N}_2$: C, 74.52; H, 5.00; F, 11.79; N, 8.69; found: C, 74.54; H, 5.05; N, 8.66.



2-benzylidene-1-phenyl-1-(2,4,6-trimethylbenzyl)hydrazine (D59)

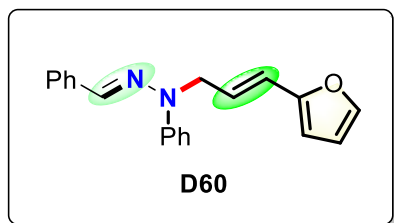
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 124.8 mg, 76% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.54 (d, J = 8.0 Hz, 2H), 7.25 (dd, J = 22.7, 15.4, 8.6 Hz, 8H), 7.06 – 6.95 (m, 1H), 6.80 (s, 2H), 4.90 (s, 2H), 2.23 (s, 9H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 147.41, 136.81, 136.78, 136.76, 133.69, 130.05, 129.91, 129.15, 128.57, 127.80, 126.17, 122.80, 119.77, 50.72, 20.95, 20.42 ppm.

HRMS (ESI) calcd. for $\text{C}_{23}\text{H}_{25}\text{N}_2$ [M+H]: 329.2018, found: 329.2017.

Elemental Analysis calcd for $\text{C}_{23}\text{H}_{24}\text{N}_2$: C, 84.11; H, 7.37; N, 8.53; found: C, 84.12; H, 7.35; N, 8.56.



2-((*E*)-benzylidene)-1-((*E*)-3-(furan-2-yl)allyl)-1-phenylhydrazine (D60)

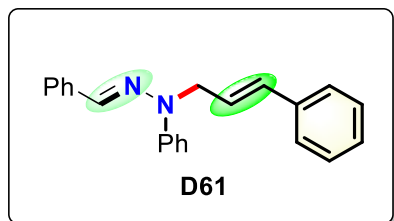
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 110.4 mg, 73% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, CDCl₃) δ 7.74 – 7.64 (m, 2H), 7.44 (s, 1H), 7.42 – 7.28 (m, 7H), 7.27 – 7.18 (m, 1H), 6.98 – 6.90 (m, 1H), 6.33 – 6.26 (m, 1H), 6.24 – 6.17 (m, 2H), 6.12 – 6.06 (m, 1H), 4.68 – 4.58 (m, 2H).

¹³C NMR (101 MHz, CDCl₃) δ 152.2, 147.6, 142.0, 136.6, 132.5, 129.2, 128.6, 128.0, 126.3, 120.8, 120.3, 119.6, 114.8, 111.4, 108.3, 48.1.

HRMS (ESI) calcd. for C₂₀H₁₉N₂O [M+H]: 303.1491, found: 303.1486.

Elemental Analysis calcd for C₂₀H₁₈N₂O: C, 79.44; H, 6.00; N, 9.26; found: C, 79.45; H, 6.04; N, 9.24.



2-(benzylidene)-1-cinnamyl-1-phenylhydrazine (D61)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 126.5 mg, 81% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

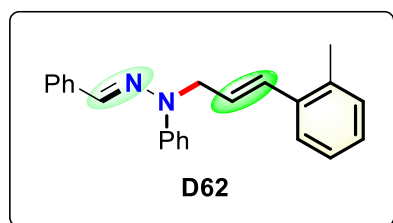
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.72 – 7.61 (m, 2H), 7.48 (s, 1H), 7.45 – 7.39 (m, 2H), 7.37 – 7.29 (m, 6H), 7.28 – 7.18 (m, 4H), 6.94 (t, *J* = 7.2 Hz, 1H), 6.44 (d, *J* = 16.1 Hz, 1H), 6.32 – 6.17 (m, 1H), 4.66 (dd, *J* = 3.8, 1.7 Hz, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.67, 136.69, 136.43, 132.49, 131.38,

129.21, 128.65, 127.95, 127.78, 126.48, 126.28, 121.80, 120.77, 114.96, 48.54 ppm.

HRMS (ESI) calcd. for $C_{22}H_{21}N_2$ [M+H]: 313.1705, found: 313.1704.

Elemental Analysis calcd for $C_{22}H_{20}N_2$: C, 84.58; H, 6.45; N, 8.97; found: C, 84.57; H, 6.44; N, 8.94.



2-(benzylidene)-1-phenyl-1-3-(p-tolyl)allylhydrazine (D62)

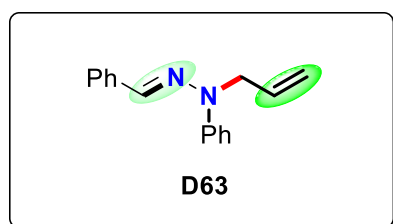
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 133.8 mg, 82% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

1H NMR (400 MHz, 23.0 °C, $CDCl_3$) δ 7.80 – 7.58 (m, 1H), 7.44 – 7.37 (m, 1H), 7.37 – 7.27 (m, 3H), 7.26 – 7.16 (m, 3H), 7.05 (d, J = 7.9 Hz, 2H), 7.01 – 6.84 (m, 3H), 6.40 (d, J = 16.1 Hz, 1H), 6.27 – 6.06 (m, 1H), 4.63 (dd, J = 4.2, 2.0 Hz, 2H), 2.27 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, $CDCl_3$) δ 147.70, 137.63, 136.75, 133.67, 132.47, 131.27, 129.35, 129.22, 128.65, 127.93, 126.39, 126.29, 120.73, 120.71, 114.96, 48.56, 29.84, 21.28 ppm.

HRMS (ESI) calcd. for $C_{23}H_{23}N_2$ [M+H]: 327.1861, found: 327.1863.

Elemental Analysis calcd for $C_{23}H_{22}N_2$: C, 84.63; H, 6.79; N, 8.58; found: C, 84.64; H, 6.78; N, 8.56.



1-allyl-2-benzylidene-1-phenylhydrazine (D63)

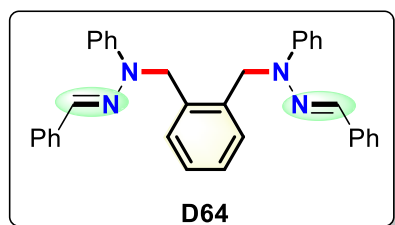
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 106.3 mg, 90% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.68 (d, $J = 7.4$ Hz, 2H), 7.42 (s, 1H), 7.39 – 7.23 (m, 8H), 6.93 (t, $J = 7.1$ Hz, 1H), 5.24 – 5.11 (m, 2H), 4.55 – 4.52 (m, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.61, 136.71, 132.28, 129.98, 129.07, 128.58, 127.83, 126.16, 120.60, 116.93, 114.80, 48.77 ppm.

HRMS (ESI) calcd. for $\text{C}_{16}\text{H}_{17}\text{N}_2$ [M+H]: 237.1392, found: 237.1391.

Elemental Analysis calcd for $\text{C}_{16}\text{H}_{16}\text{N}_2$: C, 81.32; H, 6.82; N, 11.85; found: C, 81.34; H, 6.83; N, 11.84.



1,2-bis((2-(benzylidene)-1-phenylhydrazine)methyl)benzene (D64)

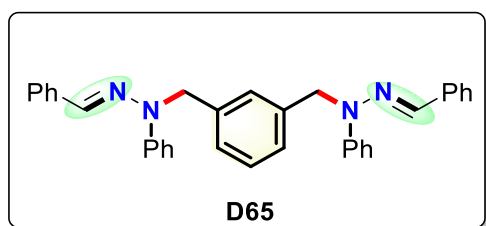
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 183.1 mg, 74% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.63 (d, $J = 7.8$ Hz, 4H), 7.41 (d, $J = 8.6$ Hz, 4H), 7.38 – 7.30 (m, 10H), 7.25 (dd, $J = 14.8, 7.7$ Hz, 3H), 7.16 (q, $J = 12.7, 9.1, 4.0$ Hz, 4H), 6.99 (t, $J = 7.1$ Hz, 2H), 5.16 (s, 4H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 147.65, 136.29, 132.67, 131.66, 129.34, 128.66, 128.16, 128.13, 126.79, 126.32, 121.34, 115.31, 48.60 ppm.

HRMS (ESI) calcd. for $\text{C}_{34}\text{H}_{31}\text{N}_4$ [M+H]: 495.2549, found: 495.2548.

Elemental Analysis calcd for $\text{C}_{34}\text{H}_{30}\text{N}_4$: C, 82.56; H, 6.11; N, 11.33; found: C, 82.55; H, 6.12; N, 11.34.



1,3-bis((2-(benzylidene)-1-phenylhydrazine)methyl)benzene (D65)

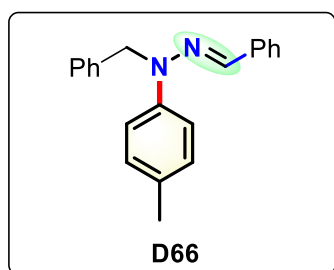
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 188.1 mg, 76% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.58 – 7.52 (m, 4H), 7.40 (d, *J* = 7.8 Hz, 4H), 7.33 – 7.22 (m, 14H), 7.15 – 7.11 (m, 3H), 6.95 (t, *J* = 7.2 Hz, 2H), 5.12 (s, 4H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.88, 136.80, 136.44, 132.67, 129.90, 129.25, 128.58, 127.94, 126.23, 125.22, 123.79, 120.89, 114.90, 50.45 ppm.

HRMS (ESI) calcd. for C₃₄H₃₁N₄ [M+H]: 495.2549, found: 495.2547.

Elemental Analysis calcd for C₃₄H₃₀N₄: C, 82.56; H, 6.11; N, 11.33; found: C, 82.56; H, 6.11; N, 11.33.



1-benzyl-2-benzylidene-1-(p-tolyl)hydrazine (D66)

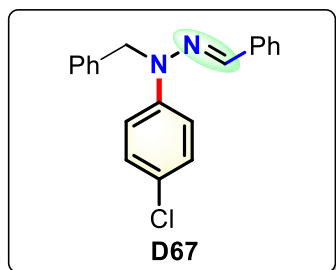
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 129.2 mg, 86% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.60 (d, *J* = 7.3 Hz, 2H), 7.34 – 7.27 (m, 7H), 7.23 (dd, *J* = 12.7, 7.1 Hz, 4H), 7.11 (d, *J* = 8.4 Hz, 2H), 5.13 (s, 2H), 2.30 (s, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 163.3, 160.0, 141.5, 141.5, 127.1, 127.0, 115.4, 115.2, 69.8, 25.3 ppm.

HRMS (ESI) calcd. for C₂₁H₂₁N₂ [M+H]: 301.1705, found: 301.1708.

Elemental Analysis calcd for C₂₁H₂₀N₂: C, 83.96; H, 6.71; N, 9.33; found: C, 83.95; H, 6.73; N, 9.32.



1-benzyl-2-benzylidene-1-(4-chlorophenyl)hydrazine (D67)

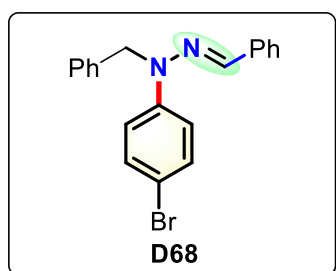
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 147.6 mg, 92% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.59 (s, 1H), 7.57 (s, 1H), 7.36 (s, 1H), 7.31 (q, *J* = 9.2, 8.1, 3.7 Hz, 6H), 7.26 – 7.20 (m, 4H), 7.17 (d, *J* = 7.2 Hz, 2H), 5.11 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 146.67, 136.45, 135.44, 133.45, 129.36, 129.28, 128.83, 128.41, 127.72, 126.54, 126.20, 125.85, 116.06, 50.52 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈ClN₂ [M+H]: 321.1159, found: 321.1155.

Elemental Analysis calcd for C₂₀H₁₇ClN₂: C, 74.88; H, 5.34; Cl, 11.05; N, 8.73; found: C, 74.87; H, 5.36; N, 8.71.



1-benzyl-2-benzylidene-1-(4-bromophenyl)hydrazine (D68)

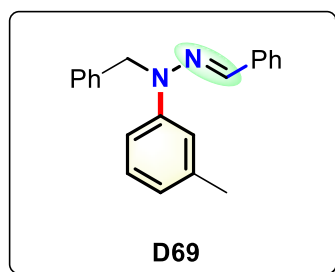
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 153.4 mg, 84% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.60 (d, *J* = 7.5 Hz, 2H), 7.38 (d, *J* = 6.1 Hz, 3H), 7.33 (t, *J* = 7.1 Hz, 4H), 7.29 – 7.22 (m, 4H), 7.19 (d, *J* = 7.4 Hz, 2H), 5.13 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 146.87, 136.18, 135.15, 133.35, 131.96, 129.13, 128.60, 128.22, 127.50, 126.33, 125.97, 116.26, 113.01, 50.18 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈BrN₂ [M+H]: 365.0653, found: 365.0653.

Elemental Analysis calcd for C₂₀H₁₇BrN₂: C, 65.76; H, 4.69; Br, 21.88; N, 7.67; found: C, 65.74; H, 4.65; N, 7.63.



1-benzyl-2-benzylidene-1-(m-tolyl)hydrazine (D69)

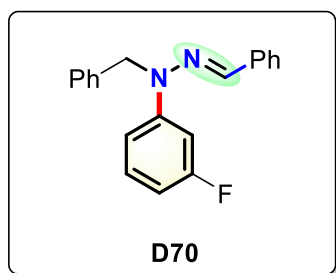
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 120.2 mg, 80% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.61 (d, *J* = 1.2 Hz, 1H), 7.59 (s, 1H), 7.34 (s, 1H), 7.31 (q, *J* = 7.7, 3.7 Hz, 4H), 7.22 (dd, *J* = 11.0, 3.7 Hz, 5H), 7.20 – 7.17 (m, 2H), 6.78 – 6.75 (m, 1H), 5.14 (s, 2H), 2.35 (s, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 147.93, 138.97, 136.59, 135.77, 132.36, 128.99, 128.49, 127.78, 127.24, 126.16, 126.04, 121.69, 115.51, 112.05, 50.57, 21.87 ppm.

HRMS (ESI) calcd. for C₂₁H₂₁N₂ [M+H]: 301.1705, found: 301.1705.

Elemental Analysis calcd for C₂₁H₂₀N₂: C, 83.96; H, 6.71; N, 9.33; found: C, 83.94; H, 6.70; N, 9.31.



1-benzyl-2-benzylidene-1-(3-fluorophenyl)hydrazine (D70)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 132.4 mg, 87% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

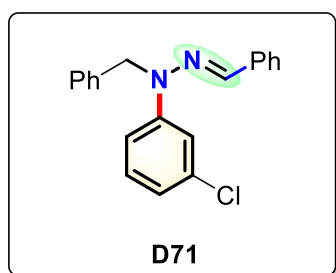
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.62 (d, $J = 7.5$ Hz, 2H), 7.41 (s, 1H), 7.33 (t, $J = 7.1$ Hz, 4H), 7.22 (t, $J = 11.5$ Hz, 6H), 7.04 (d, $J = 8.3$ Hz, 1H), 6.62 (t, $J = 8.0$ Hz, 1H), 5.15 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 165.13, 162.71, 149.58, 149.48, 136.12, 135.19, 133.60, 130.29, 130.19, 129.13, 128.61, 128.30, 127.49, 126.41, 125.97, 109.75, 109.73, 107.27, 107.05, 102.30, 102.03, 50.24 ppm.

$^{19}\text{F NMR}$ (377 MHz, 23.0 °C, CDCl_3) δ -112.11 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{FN}_2$ [$\text{M}+\text{H}$]: 305.1454, found: 305.1455.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{FN}_2$: C, 78.92; H, 5.63; F, 6.24; N, 9.20; found: C, 78.90; H, 5.61; N, 9.24.



1-benzyl-2-benzylidene-1-(3-chlorophenyl)hydrazine (D71)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 131.5 mg, 82% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

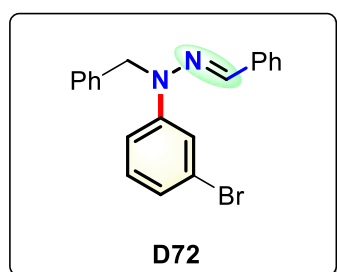
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.66 (dd, $J = 8.0, 1.3$ Hz, 1H), 7.55 – 7.51 (m,

2H), 7.41 (d, $J = 7.3$ Hz, 2H), 7.35 – 7.27 (m, 6H), 7.25 – 7.21 (m, 2H), 7.18 (s, 1H), 7.10 (t, $J = 7.9, 1.6$ Hz, 1H), 4.95 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 148.91, 136.08, 135.13, 135.10, 133.81, 130.13, 129.16, 128.62, 128.33, 127.52, 126.44, 125.96, 120.52, 114.85, 112.53, 50.23 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{ClN}_2$ [M+H]: 321.1159, found: 321.1155.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{ClN}_2$: C, 74.88; H, 5.34; Cl, 11.05; N, 8.73; found: C, 74.84; H, 5.35; N, 8.74.



1-benzyl-2-benzylidene-1-(3-bromophenyl)hydrazine (D72)

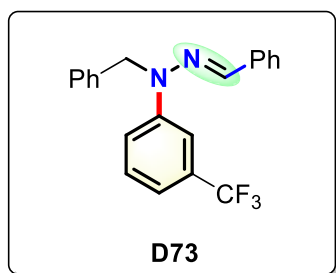
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 135.1 mg, 74% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.63 (t, $J = 2.0$ Hz, 1H), 7.61 (s, 1H), 7.59 (s, 1H), 7.39 (s, 1H), 7.33 (t, $J = 7.3$ Hz, 4H), 7.28 – 7.18 (m, 5H), 7.13 (t, $J = 8.0$ Hz, 1H), 7.04 (d, $J = 8.2$ Hz, 1H), 5.11 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 149.02, 136.07, 135.08, 133.90, 130.43, 129.18, 128.63, 128.36, 127.54, 126.45, 125.97, 123.46, 123.36, 117.73, 113.01, 50.23 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{BrN}_2$ [M+H]: 365.0653, found: 365.0653.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{BrN}_2$: C, 65.76; H, 4.69; Br, 21.88; N, 7.67; found: C, 65.74; H, 4.64; N, 7.64.



1-benzyl-2-benzylidene-1-(3-(trifluoromethyl)phenyl)hydrazine (D73)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 148.8 mg, 84% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

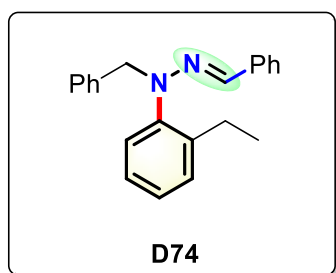
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.73 (s, 1H), 7.63 (d, *J* = 1.4 Hz, 1H), 7.61 (s, 1H), 7.45 (d, *J* = 9.5 Hz, 2H), 7.36 (t, *J* = 14.7, 7.7 Hz, 5H), 7.30 – 7.24 (m, 2H), 7.23 – 7.15 (m, 3H), 5.18 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 148.10, 135.97, 134.97, 134.17, 132.09, 131.77, 131.45, 131.14, 129.66, 129.21, 128.65, 128.45, 127.60, 126.47, 125.94, 125.71, 123.00, 117.32, 117.31, 117.07, 117.03, 116.99, 116.95, 111.44, 111.40, 111.36, 111.32, 50.22 ppm.

¹⁹F NMR (377 MHz, 23.0 °C, CDCl₃) δ -62.54 ppm.

HRMS (ESI) calcd. for C₂₁H₁₈F₃N₂ [M+H]: 355.1422, found: 355.1425.

Elemental Analysis calcd for C₂₁H₁₇F₃N₂: C, 71.18; H, 4.84; F, 16.08; N, 7.91; found: C, 71.14; H, 4.86; N, 7.91.



1-benzyl-2-benzylidene-1-(2-ethylphenyl)hydrazine (D74)

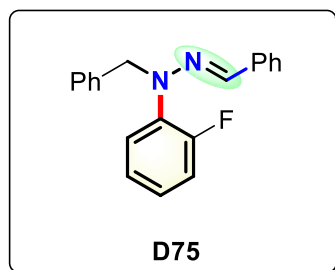
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 113.2 mg, 72% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.50 – 7.45 (m, 2H), 7.35 (d, *J* = 7.2 Hz, 2H), 7.32 – 7.23 (m, 6H), 7.22 (s, 1H), 7.18 (dd, *J* = 10.2, 4.9, 2.1 Hz, 2H), 7.01 (dd, *J* = 7.5, 1.6 Hz, 1H), 6.93 (s, 1H), 4.86 (s, 2H), 2.58 (q, *J* = 7.6 Hz, 2H), 1.13 (t, *J* = 7.6 Hz, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 144.14, 141.97, 138.07, 136.77, 133.41, 129.37, 128.43, 128.39, 128.32, 127.35, 127.22, 127.16, 127.11, 126.72, 125.73, 60.53, 24.22, 14.83 ppm.

HRMS (ESI) calcd. for C₂₂H₂₃N₂ [M+H]: 315.1861, found: 315.1864.

Elemental Analysis calcd for C₂₂H₂₂N₂: C, 84.04; H, 7.05; N, 8.91; found: C, 84.02; H, 7.03; N, 8.90.



1-benzyl-2-benzylidene-1-(2-fluorophenyl)hydrazine (D75)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 121.7 mg, 80% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

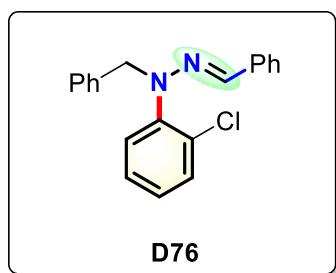
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.56 – 7.49 (m, 3H), 7.31 (dd, *J* = 15.6, 7.5, 5.7 Hz, 7H), 7.26 – 7.19 (m, 2H), 7.16 – 7.10 (m, 1H), 7.10 – 7.04 (m, 2H), 5.02 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 155.81, 153.36, 136.83, 136.46, 136.38, 136.24, 134.80, 128.71, 128.51, 127.99, 127.17, 126.66, 126.16, 124.82, 124.75, 124.70, 124.68, 124.63, 124.60, 116.73, 116.52, 56.48 ppm.

¹⁹F NMR (377 MHz, 23.0 °C, CDCl₃) δ -122.78 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈FN₂ [M+H]: 305.1454, found: 305.1455.

Elemental Analysis calcd for C₂₀H₁₇FN₂: C, 78.92; H, 5.63; F, 6.24; N, 9.20; found: C, 78.91; H, 5.61; N, 9.22.



1-benzyl-2-benzylidene-1-(2-chlorophenyl)hydrazine (D76)

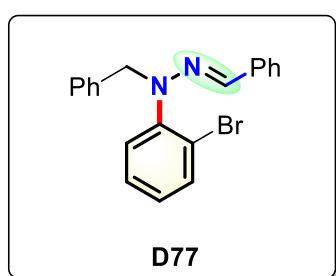
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 117.1 mg, 73% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.51 (d, $J = 7.2$ Hz, 2H), 7.42 (dd, $J = 8.0, 1.3$ Hz, 1H), 7.38 (d, $J = 7.4$ Hz, 2H), 7.33 – 7.17 (m, 9H), 7.12 (t, $J = 7.9, 1.8$ Hz, 1H), 4.94 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 144.74, 137.18, 136.27, 135.50, 130.71, 130.09, 128.52, 128.50, 127.95, 127.84, 127.69, 127.55, 127.18, 126.86, 126.12, 58.27 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{18}\text{ClN}_2$ [$\text{M}+\text{H}$]: 321.1159, found: 321.1155.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{17}\text{ClN}_2$: C, 74.88; H, 5.34; Cl, 11.05; N, 8.73; found: C, 74.87; H, 5.37; N, 8.72.



1-benzyl-2-benzylidene-1-(2-bromophenyl)hydrazine (D77)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 122.4 mg, 67% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

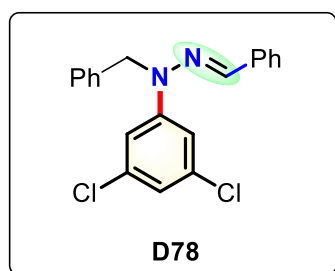
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.65 (dd, $J = 8.0, 1.3$ Hz, 1H), 7.55 – 7.49 (m, 2H), 7.40 (d, $J = 7.3$ Hz, 2H), 7.32 – 7.19 (m, 8H), 7.16 (s, 1H), 7.09 (t, $J = 7.9, 1.6$ Hz,

¹H), 4.94 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 145.69, 137.19, 136.24, 135.47, 133.87, 128.45, 128.44, 128.41, 128.35, 127.87, 127.55, 127.15, 126.05, 121.11, 58.77 ppm.

HRMS (ESI) calcd. for C₂₀H₁₈BrN₂ [M+H]: 365.0653, found: 365.0653.

Elemental Analysis calcd for C₂₀H₁₇BrN₂: C, 65.76; H, 4.69; Br, 21.88; N, 7.67; found: C, 65.72; H, 4.64; N, 7.63.



1-benzyl-2-benzylidene-1-(3,5-dichlorophenyl)hydrazine (D78)

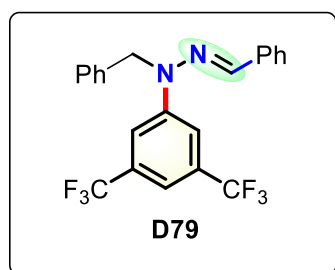
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 152.8 mg, 86% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.58 (d, *J* = 7.2 Hz, 2H), 7.39 (s, 1H), 7.31 (t, *J* = 7.4 Hz, 4H), 7.28 – 7.22 (m, 4H), 7.14 (t, *J* = 6.8 Hz, 2H), 6.87 (t, *J* = 1.6 Hz, 1H), 5.04 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 149.36, 135.66, 135.60, 135.08, 134.54, 129.33, 128.80, 128.73, 127.77, 126.69, 125.91, 120.21, 112.84, 50.04 ppm.

HRMS (ESI) calcd. for C₂₀H₁₇Cl₂N₂ [M+H]: 355.0769, found: 355.0767.

Elemental Analysis calcd for C₂₀H₁₆Cl₂N₂: C, 67.62; H, 4.54; Cl, 19.96; N, 7.89; found: C, 67.63; H, 4.55; N, 7.84.



1-benzyl-2-benzylidene-1-(3,5-bis(trifluoromethyl)phenyl)hydrazine (D79)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 192.2 mg, 91% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

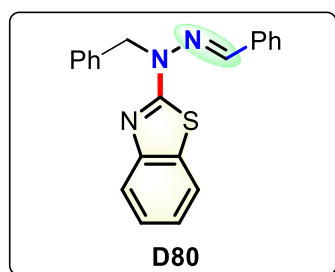
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.78 (s, 2H), 7.65 – 7.60 (m, 2H), 7.52 (s, 1H), 7.40 (s, 1H), 7.38 (s, 1H), 7.35 (d, $J = 7.7$ Hz, 3H), 7.33 – 7.27 (m, 2H), 7.20 (d, $J = 7.2$ Hz, 2H), 5.20 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 148.60, 136.00, 135.35, 134.14, 133.00, 132.67, 132.34, 132.02, 129.39, 129.03, 128.76, 127.90, 127.65, 126.72, 125.81, 124.94, 122.23, 119.52, 113.87, 113.84, 113.50, 113.46, 113.42, 113.38, 113.35, 50.03 ppm.

$^{19}\text{F NMR}$ (377 MHz, 23.0 °C, CDCl_3) δ -62.88 ppm.

HRMS (ESI) calcd. for $\text{C}_{22}\text{H}_{17}\text{F}_6\text{N}_2$ [$\text{M}+\text{H}$]: 423.1296, found: 423.1299.

Elemental Analysis calcd for $\text{C}_{22}\text{H}_{16}\text{F}_6\text{N}_2$: C, 62.56; H, 3.82; F, 26.99; N, 6.63; found: C, 62.53; H, 3.83; N, 6.65.



2-(1-benzyl-2-benzylidenehydrazineyl)benzo[d]thiazole (D80)

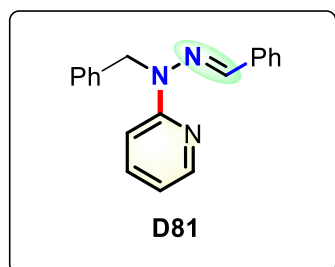
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 138.9 mg, 81% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.70 (d, $J = 7.7$ Hz, 1H), 7.67 – 7.56 (m, 4H), 7.38 – 7.21 (m, 10H), 7.16 (t, $J = 7.9$ Hz, 1H), 5.59 (s, 2H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 169.69, 152.22, 138.19, 134.49, 132.79, 129.50, 129.07, 128.71, 127.63, 126.94, 126.49, 125.82, 122.38, 120.97, 120.42, 49.25 ppm.

HRMS (ESI) calcd. for C₂₁H₁₈N₃S [M+H]: 344.1221, found: 344.1223.

Elemental Analysis calcd for C₂₁H₁₇N₃S: C, 73.44; H, 4.99; N, 12.24; S, 9.33; found: C, 73.45; H, 4.96; N, 12.22.



2-(1-benzyl-2-benzylidenehydrazineyl)pyridine (D81)

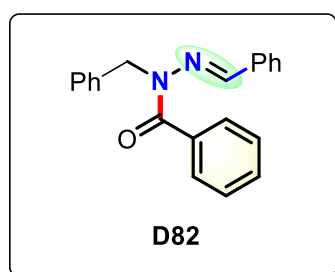
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 133.6 mg, 93% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 8.21 (dd, *J* = 5.4, 1.2 Hz, 1H), 7.84 (d, *J* = 8.6 Hz, 1H), 7.68 – 7.58 (m, 3H), 7.52 (s, 1H), 7.36 – 7.17 (m, 8H), 6.80 (t, *J* = 7.0, 5.0, 0.8 Hz, 1H), 5.62 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 157.87, 147.32, 137.69, 136.38, 136.10, 135.09, 128.84, 128.58, 128.38, 126.99, 126.43, 126.35, 115.89, 109.68, 46.14 ppm.

HRMS (ESI) calcd. for C₁₉H₁₈N₃ [M+H]: 288.1501, found: 288.1501.

Elemental Analysis calcd for C₁₉H₁₇N₃: C, 79.41; H, 5.96; N, 14.62; found: C, 79.44; H, 5.94; N, 14.61.



N-benzyl-N'-benzylidenebenzohydrazide (D82)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 100.5 mg, 64% yield. Purification by

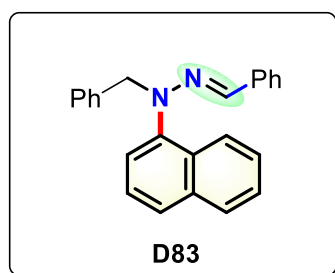
column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.89 – 7.77 (m, 2H), 7.68 (s, 1H), 7.55 – 7.40 (m, 3H), 7.41 – 7.19 (m, 10H), 5.43 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 171.51, 140.20, 135.19, 135.08, 134.62, 130.51, 130.05, 129.69, 129.10, 128.68, 127.54, 127.52, 127.23, 126.50, 45.54 ppm.

HRMS (ESI) calcd. for C₂₁H₁₉N₂O [M+H]: 315.1497, found: 315.1498.

Elemental Analysis calcd for C₂₁H₁₈N₂O: C, 80.23; H, 5.77; N, 8.91; O, 5.09; found: C, 80.20; H, 5.76; N, 8.94.



1-benzyl-2-benzylidene-1-(naphthalen-1-yl)hydrazine (D83)

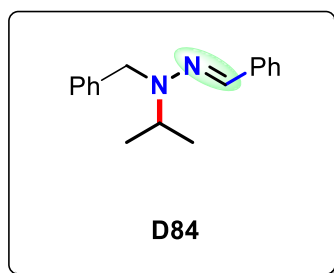
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 122.8 mg, 73% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 8.04 (d, *J* = 8.2 Hz, 1H), 7.88 (d, *J* = 7.6 Hz, 1H), 7.75 (d, *J* = 8.2 Hz, 1H), 7.51 – 7.39 (m, 7H), 7.34 – 7.23 (m, 6H), 7.21 – 7.16 (m, 1H), 7.06 (s, 1H), 5.06 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 143.49, 138.27, 136.83, 135.02, 134.95, 130.10, 128.76, 128.70, 128.67, 128.22, 127.80, 127.42, 127.21, 126.69, 126.46, 126.13, 126.10, 124.20, 123.72, 60.51 ppm.

HRMS (ESI) calcd. for C₂₄H₂₁N₂ [M+H]: 337.1705, found: 337.1704.

Elemental Analysis calcd for C₂₄H₂₀N₂: C, 85.68; H, 5.99; N, 8.33; found: C, 85.64; H, 5.94; N, 8.35.



1-benzyl-2-benzylidene-1-isopropylhydrazine (D84)

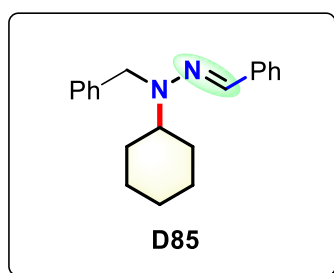
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 64.3 mg, 51% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.49 – 7.43 (m, 2H), 7.33 – 7.23 (m, 7H), 7.14 (t, $J = 7.3$ Hz, 1H), 7.07 (s, 1H), 4.36 (s, 2H), 3.80 (t, $J = 6.6$ Hz, 1H), 1.29 (d, $J = 6.6$ Hz, 6H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 138.19, 137.51, 130.69, 128.70, 128.33, 126.86, 126.80, 126.43, 125.32, 57.76, 52.61, 20.41 ppm.

HRMS (ESI) calcd. for $\text{C}_{17}\text{H}_{21}\text{N}_2$ [M+H]: 253.1705, found: 253.1706.

Elemental Analysis calcd for $\text{C}_{17}\text{H}_{20}\text{N}_2$: C, 80.91; H, 7.99; N, 11.10; found: C, 80.96; H, 7.95; N, 11.12.



1-benzyl-2-benzylidene-1-cyclohexylhydrazine (D85)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 81.8 mg, 56% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

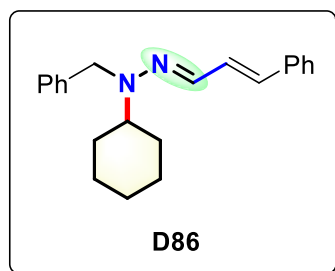
$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.47 – 7.42 (m, 2H), 7.35 – 7.22 (m, 7H), 7.16 – 7.09 (m, 1H), 7.03 (s, 1H), 4.44 (s, 2H), 3.36 (t, $J = 11.6, 3.7$ Hz, 1H), 2.05 – 1.97 (m, 2H), 1.84 (d, $J = 13.2$ Hz, 2H), 1.66 (d, $J = 14.3$ Hz, 1H), 1.58 (t, $J = 12.3, 3.4$ Hz, 2H),

1.38 – 1.28 (m, 2H), 1.18 (t, $J = 12.8, 3.4$ Hz, 1H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 138.25, 137.66, 129.66, 128.65, 128.31, 126.80, 126.59, 126.36, 125.23, 66.30, 52.97, 31.27, 25.91 ppm.

HRMS (ESI) calcd. for $\text{C}_{20}\text{H}_{25}\text{N}_2$ [$\text{M}+\text{H}$]: 293.2018, found: 293.2017.

Elemental Analysis calcd for $\text{C}_{20}\text{H}_{24}\text{N}_2$: C, 82.15; H, 8.27; N, 9.58; found: C, 82.14; H, 8.23; N, 9.56.



2-((*E*)-benzylidene)-1-cinnamyl-1-cyclohexylhydrazine (D86)

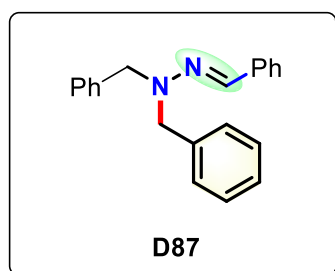
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 79.6 mg, 50% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, CDCl_3) δ 7.57 – 7.47 (m, 2H), 7.39 – 7.10 (m, 9H), 6.58 – 6.49 (m, 1H), 6.29 – 6.17 (m, 1H), 4.07 – 3.96 (m, 2H), 3.38 – 3.22 (m, 1H), 2.02 – 1.90 (m, 2H), 1.88 – 1.78 (m, 2H), 1.72 – 1.48 (m, 3H), 1.42 – 1.06 (m, 3H).

^{13}C NMR (101 MHz, CDCl_3) δ 137.8, 137.0, 130.8, 129.4, 128.6, 128.4, 127.4, 126.6, 126.3, 125.3, 125.2, 65.7, 50.6, 31.3, 26.0.

HRMS (ESI) calcd. for $\text{C}_{22}\text{H}_{27}\text{N}_2$ [$\text{M}+\text{H}$]: 319.2169, found: 319.2170.

Elemental Analysis calcd for $\text{C}_{22}\text{H}_{26}\text{N}_2$: C, 97; H, 8.23; N, 8.80; found: C, 82.94; H, 8.22; N, 8.82.



1,1-dibenzyl-2-benzylidenehydrazine (D87)

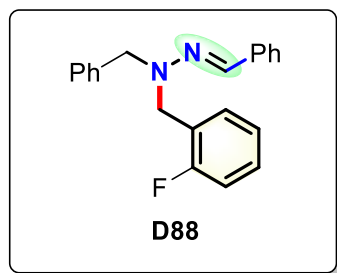
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 109.6 mg, 73% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

$^1\text{H NMR}$ (400 MHz, 23.0 °C, CDCl_3) δ 7.49 (d, $J = 7.3$ Hz, 2H), 7.34 – 7.25 (m, 10H), 7.24 – 7.16 (m, 3H), 4.51 (s, 4H) ppm.

$^{13}\text{C NMR}$ (101 MHz, 23.0 °C, CDCl_3) δ 137.67, 137.01, 131.88, 128.63, 128.49, 127.87, 127.71, 127.26, 125.62, 57.96 ppm.

HRMS (ESI) calcd. for $\text{C}_{21}\text{H}_{21}\text{N}_2$ [$\text{M}+\text{H}$]: 301.1705, found: 301.1704.

Elemental Analysis calcd for $\text{C}_{21}\text{H}_{20}\text{N}_2$: C, 83.96; H, 6.71; N, 9.33; found: C, 83.94; H, 6.73; N, 9.32.



2-((*E*)-benzylidene)-1-cinnamyl-1-(2-fluorobenzyl)hydrazine (D88)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 119.3 mg, 75% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

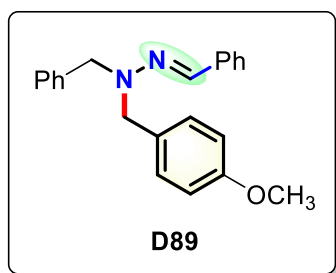
$^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.52 (d, $J = 7.2$ Hz, 2H), 7.39 – 7.21 (m, 8H), 7.20 – 7.10 (m, 3H), 7.08 – 6.95 (m, 2H), 6.49 (d, $J = 16.0$ Hz, 1H), 6.31 – 6.15 (m, 1H), 4.56 (s, 2H), 4.16 – 4.03 (m, 2H).

$^{13}\text{C NMR}$ (101 MHz, CDCl_3) δ 162.0, 159.6, 137.1, 137.0, 132.8, 132.3, 130.2, 129.1, 129.0, 128.8, 128.7, 127.8, 127.5, 126.6, 125.8, 125.0, 124.8, 124.4, 124.4, 115.6, 115.4, 56.5, 51.0.

$^{19}\text{F NMR}$ (376 MHz, CDCl_3) $\delta = -117.8$.

HRMS (ESI) calcd. for $\text{C}_{23}\text{H}_{21}\text{FN}_2$ [$\text{M}+\text{H}$]: 345.1761, found: 345.1762.

Elemental Analysis calcd for $\text{C}_{23}\text{H}_{20}\text{FN}_2$: C, 79.22; H, 6.02; F, 5.97; N, 8.80; found: C, 79.25; H, 6.04; N, 8.84.



2-((*E*)-benzylidene)-1-cinnamyl-1-(4-methoxybenzyl)hydrazine (D89)

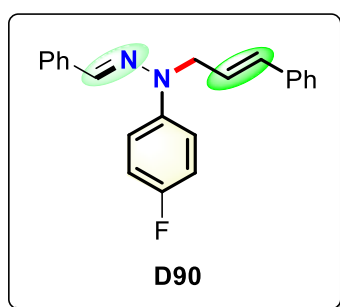
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 117.2 mg, 71% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, CDCl₃) δ 7.60 – 7.47 (m, 2H), 7.38 – 7.11 (m, 11H), 6.87 – 6.78 (m, 2H), 6.52 – 6.39 (m, 1H), 6.28 – 6.15 (m, 1H), 4.45 (s, 2H), 4.08 – 3.98 (m, 2H), 3.72 (s, 3H).

¹³C NMR (101 MHz, CDCl₃) δ 156.0, 137.2, 137.0, 132.6, 132.1, 129.8, 129.1, 128.7, 128.6, 127.7, 127.4, 126.5, 125.7, 125.1, 114.1, 57.3, 55.5, 55.4.

HRMS (ESI) calcd. for C₂₄H₂₅N₂O [M+H]: 357.1961, found: 357.1961.

Elemental Analysis calcd for C₂₄H₂₄N₂O: C, 79.97; H, 6.71; N, 8.48; O, 4.84; found: C, 79.96; H, 6.74; N, 8.42.



2-(benzylidene)-1-cinnamyl-1-(4-fluorophenyl)hydrazine (D90)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 189.2 mg, 74% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

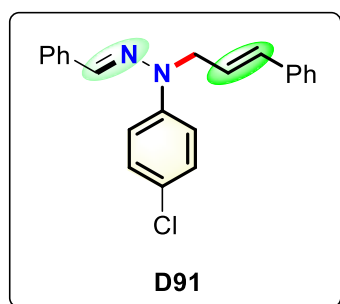
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.43 (s, 1H), 7.37 – 7.12 (m, 8H), 7.08 – 6.93 (m, 3H), 6.40 (d, *J* = 16.1 Hz, 1H), 6.29 – 6.09 (m, 2H), 4.72 – 4.45 (m, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 159.07, 156.70, 144.20, 144.18, 136.58, 136.35, 132.60, 131.47, 128.71, 128.64, 128.04, 127.90, 126.51, 126.26, 121.68, 116.31, 116.24, 115.82, 115.60, 49.08 ppm.

^{19}F NMR (377 MHz, 23.0 °C, CDCl_3) δ -123.94 ppm.

HRMS (ESI) calcd. for $\text{C}_{22}\text{H}_{20}\text{FN}_2$ [M+H]: 331.1610, found: 331.1613.

Elemental Analysis calcd for $\text{C}_{22}\text{H}_{19}\text{FN}_2$: C, 79.97; H, 5.80; F, 5.75; N, 8.48; found: C, 79.94; H, 5.82; N, 8.45.



2-(benzylidene)-1-(4-chlorophenyl)-1-cinnamylhydrazine (D91)

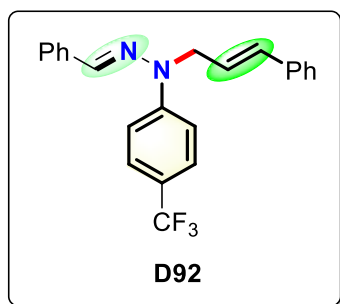
The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 140.5 mg, 81% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.77 – 7.57 (m, 2H), 7.47 (s, 1H), 7.42 – 7.04 (m, 12H), 6.38 (d, J = 16.1, 2.0 Hz, 1H), 6.18 (d, J = 16.0, 4.0 Hz, 1H), 4.59 (dd, J = 4.0, 2.0 Hz, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 146.21, 136.36, 136.25, 133.20, 131.53, 129.09, 128.72, 128.70, 128.25, 127.93, 126.50, 126.46, 126.39, 125.58, 121.31, 115.95, 48.36 ppm.

HRMS (ESI) calcd. for $\text{C}_{22}\text{H}_{20}\text{ClN}_2$ [M+H]: 347.1315, found: 347.1316.

Elemental Analysis calcd for $\text{C}_{22}\text{H}_{19}\text{ClN}_2$: C, 76.18; H, 5.52; Cl, 10.22; N, 8.08; found: C, 76.14; H, 5.55; N, 8.03.



2-(benzylidene)-1-cinnamyl-1-(4-(trifluoromethoxy)phenyl)hydrazine (D92)

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 171.1 mg, 90% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 20 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.66 (d, *J* = 7.7 Hz, 1H), 7.48 (s, 1H), 7.42 – 7.05 (m, 9H), 6.39 (d, *J* = 16.0 Hz, 1H), 6.19 (d, *J* = 16.0, 4.0 Hz, 2H), 4.78 – 4.43 (m, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 146.34, 142.99, 142.98, 136.33, 136.24, 133.44, 131.59, 128.73, 128.72, 128.32, 127.97, 126.51, 126.41, 122.16, 122.10, 121.26, 115.44, 48.44 ppm.

¹⁹F NMR (377 MHz, 23.0 °C, CDCl₃) δ -58.05 ppm.

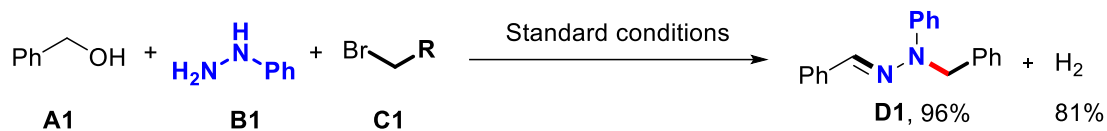
HRMS (ESI) calcd. for C₂₃H₂₀F₃N₂O [M+H]: 397.1528, found: 397.1527.

Elemental Analysis calcd for C₂₃H₁₉F₃N₂O: C, 69.69; H, 4.83; F, 14.38; N, 7.07; O, 4.04; found: C, 69.66; H, 4.86; N, 7.05.

6. Mechanism investigations

6.1 Control experiments

6.1.1



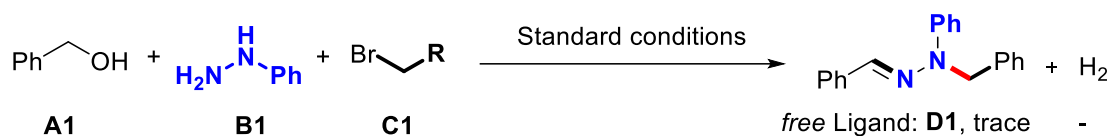
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.1.2



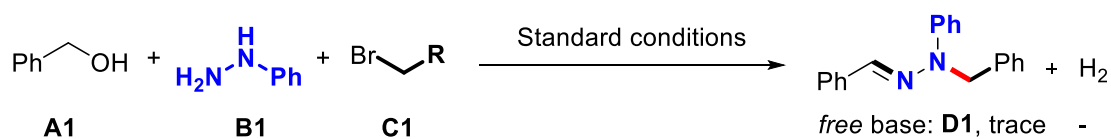
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.1.3



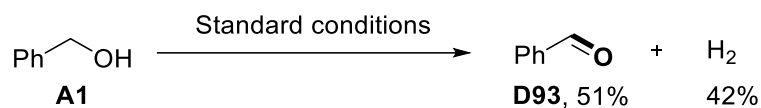
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), ^tBuOK (1.0 mmol), **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.1.4



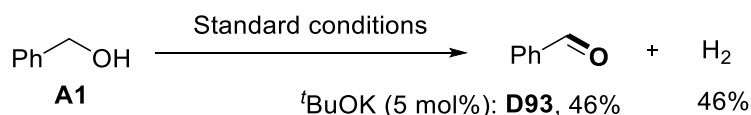
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.1.5



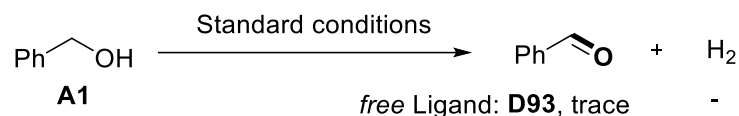
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), **A1** (0.8 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.1.6



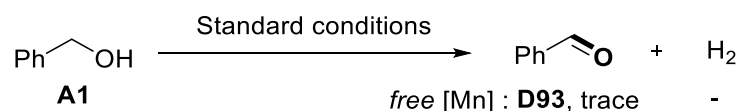
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (0.025 mmol), **A1** (0.8 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.1.7



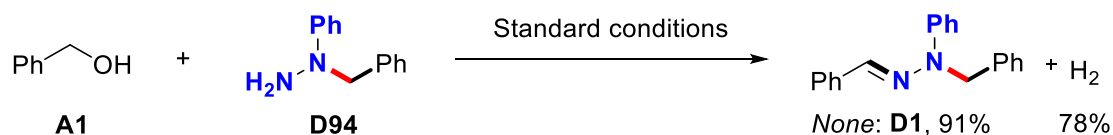
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), ^tBuOK (1.0 mmol), **A1** (0.8 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.1.8



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, **L11** (1.2 mol%), ^tBuOK (1.0 mmol), **A1** (0.8 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

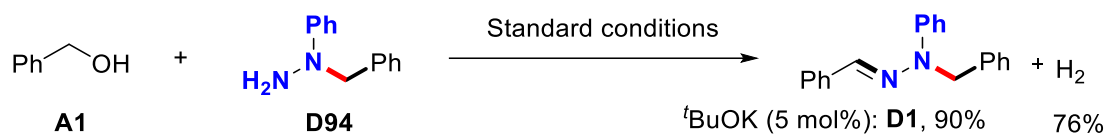
6.2.1



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), **A1** (0.8 mmol), **D94** (0.5 mmol) and THF (3 mL). Then the seal tube was

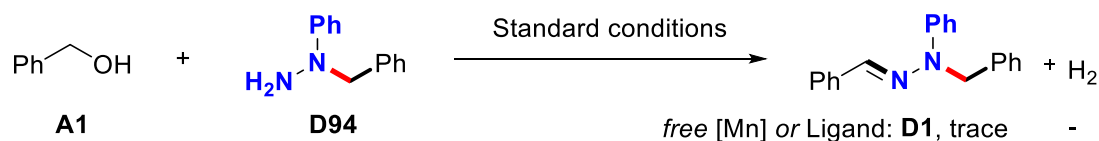
closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.2



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), *t*BuOK (1.0 mmol), **A1** (0.8 mmol), **D94** (0.5 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

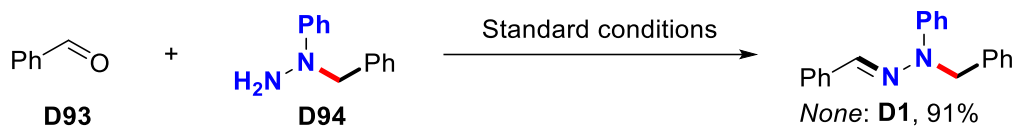
6.2.3



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), *t*BuOK (1.0 mmol), **A1** (0.8 mmol), **D94** (0.5 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic

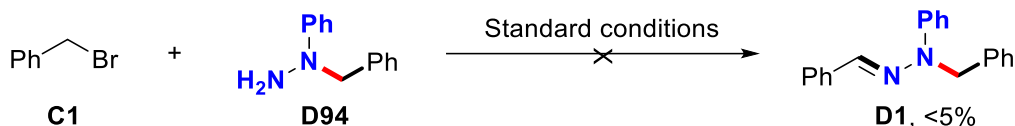
phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.4



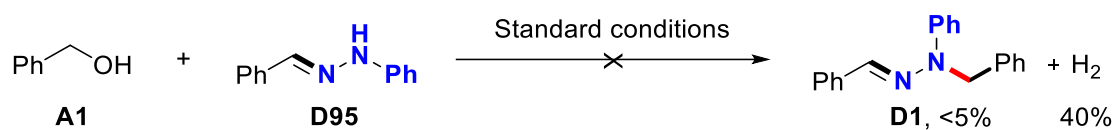
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), **D93** (0.8 mmol), **D94** (0.5 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.5



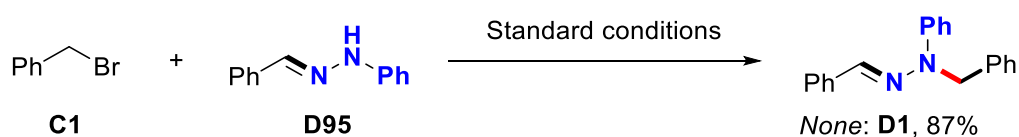
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), **A1** (0.8 mmol), **D94** (0.5 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.6



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), L11 (1.2 mol%), ^tBuOK (1.0 mmol), A1 (0.8 mmol), D95 (0.5 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.7



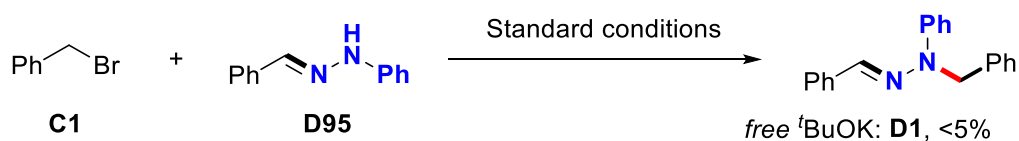
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), L11 (1.2 mol%), ^tBuOK (1.0 mmol), C1 (0.8 mmol), D95 (0.5 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.8



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), **C1** (0.8 mmol), **D95** (0.5 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.9



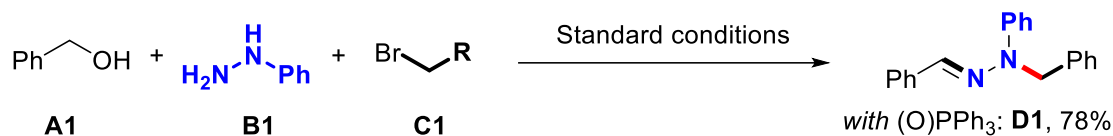
Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (1.0 mmol), **C1** (0.8 mmol), **D95** (0.5 mmol) and **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.10



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), mercury (2 drops), **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.11



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), $(\text{O})\text{PPh}_3$ (1.0 mmol) **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

6.2.12



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was

charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), TEMPO (1.0 mmol), **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

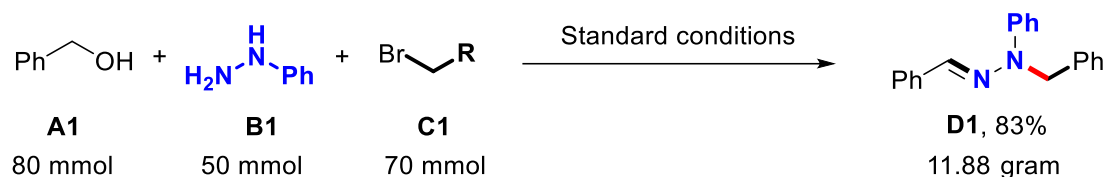
6.2.13



Using a nitrogen-filled glove box, an oven-dried pressure tube (38 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), Ph_2CHCH_2 (1.0 mmol), **A1** (0.8 mmol), **B1** (0.5 mmol) and **C1** (0.7 mmol) and THF (3 mL). Then the seal tube was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) for design time. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation in the reported yield.

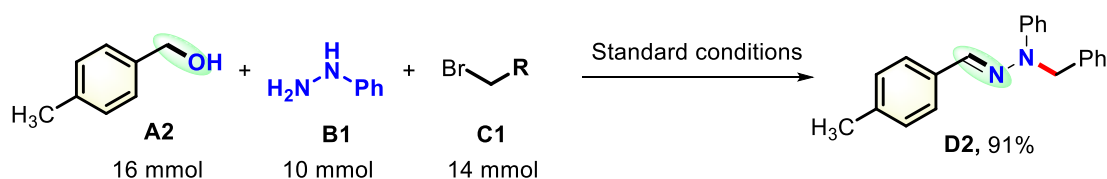
7. Gram-Scale Synthesis

7.1 Gram-Scale Synthesis of D1



Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (1000 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (100.0 mmol), **A1** (80 mmol), **B1** (50 mmol), **C1** (70 mmol) and THF (300 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N_2 atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the desired product **D1** 11.88 g, the yield is 83%.

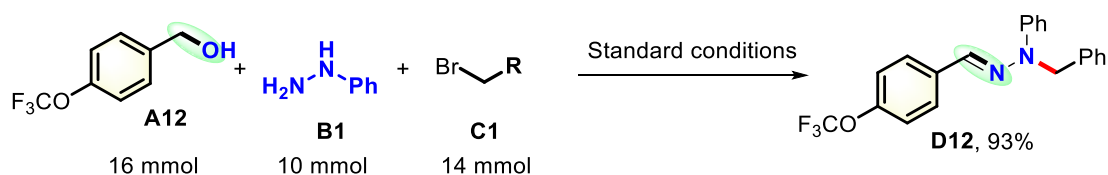
7.2 Gram-Scale Synthesis of D2



Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (200 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (100.0 mmol), **A2** (16 mmol), **B1** (10 mmol), **C1** (14 mmol) and THF (60 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N_2 atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas

were analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the desired product **D2** in 91% (2.73 g).

7.3 Gram-Scale Synthesis of **D12**



Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (200 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (100.0 mmol), **A12** (16 mmol), **B1** (10 mmol), **C1** (14 mmol) and THF (60 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N_2 atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the desired product **D12** in 93% (3.44 g).

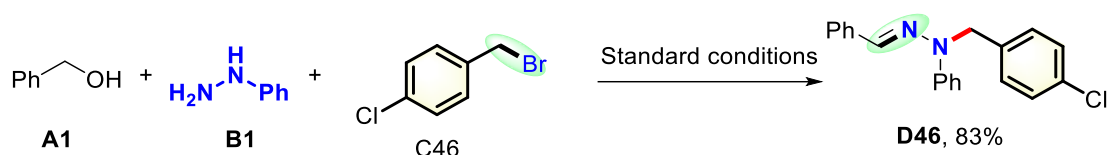
7.4 Gram-Scale Synthesis of **D43**



Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (200 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (100.0 mmol), **A1** (16 mmol), **B1** (10 mmol), **C43** (14 mmol) and THF (60 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove

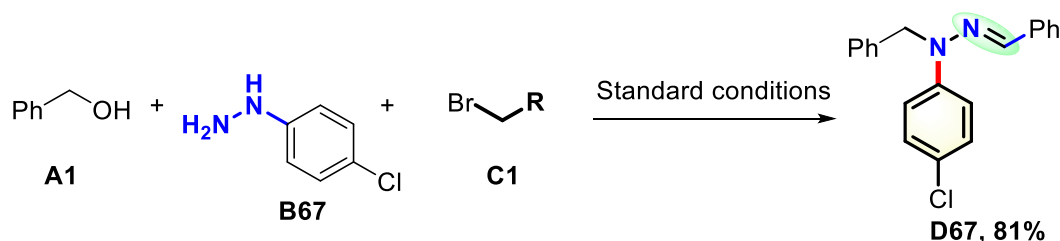
box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N₂ atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the desired product **D43** in 84% (2.52 g).

7.5 Gram-Scale Synthesis of D46



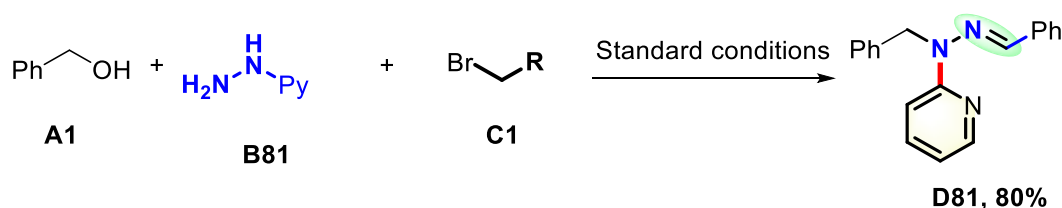
Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (200 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (100.0 mmol), **A1** (16 mmol), **B1** (10 mmol), **C46** (14 mmol) and THF (60 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N₂ atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the desired product **D46** in 84% (2.66 g).

7.6 Gram-Scale Synthesis of D67



Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (200 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (100.0 mmol), **A1** (16 mmol), **B67**(10 mmol), **C1** (14 mmol) and THF (60 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N₂ atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the desired product **D67** in 81% (2.59 g).

7.7 Gram-Scale Synthesis of **D81**



Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (200 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (10.0 mmol), **A1** (16 mmol), **B81** (10 mmol), **C1** (14 mmol) and THF (60 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N₂ atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase and gas were analyzed by GC and GC-MS to monitor product formation. The solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography (petroleum ether/dichloromethane = 20:1 – 5:1) on silica gel to give the desired product **D81** in 80% (2.29 g).

8. Time-conversion-plot reaction



Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (38 mL volume) was charged with a magnetic stirring bar, MnBr(CO)₅ (1.0 mol%), **L11** (1.2 mol%), ^tBuOK (10.0 mmol), **A1** (0.8 mmol), **B1** (0.5 mmol), **C1** (0.7 mmol) and THF (3 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N₂ atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase was analyzed by GC to monitor product formation. Composition of **D1**, **d93** and **D94** were determined by GC-analysis using *n*-Cetane as internal standard.

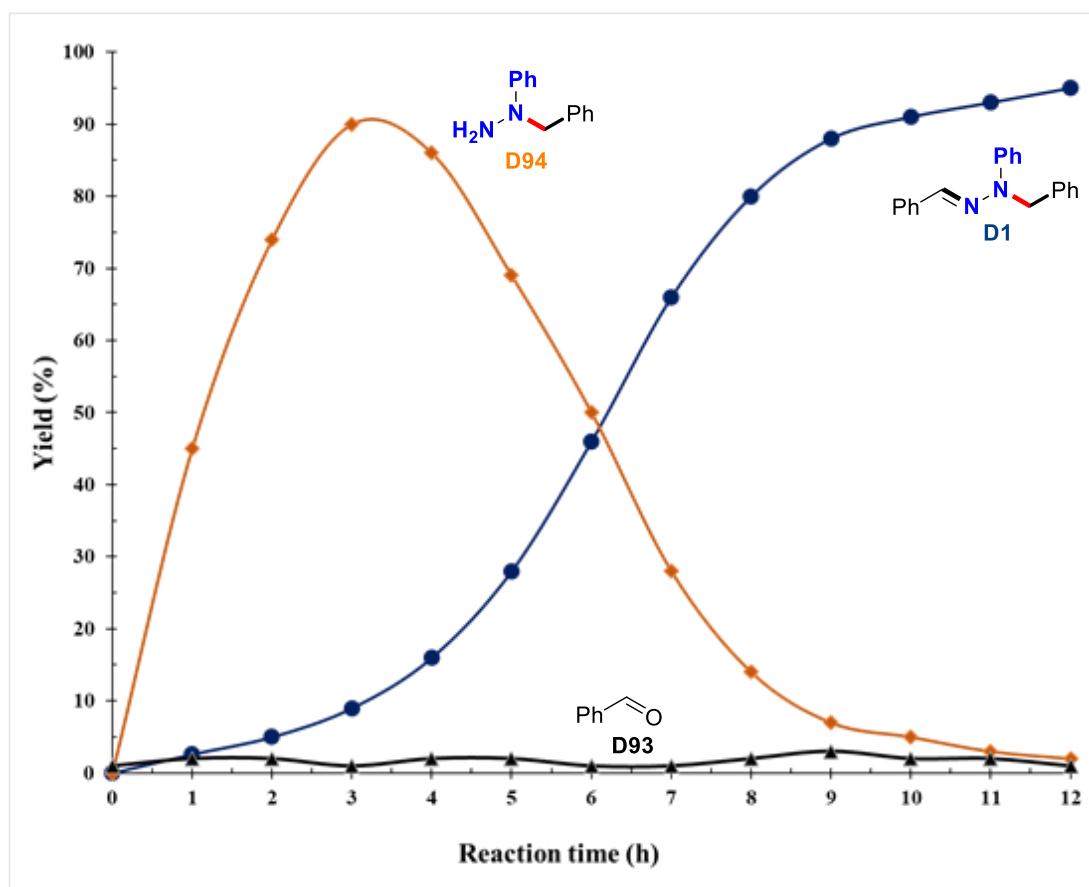
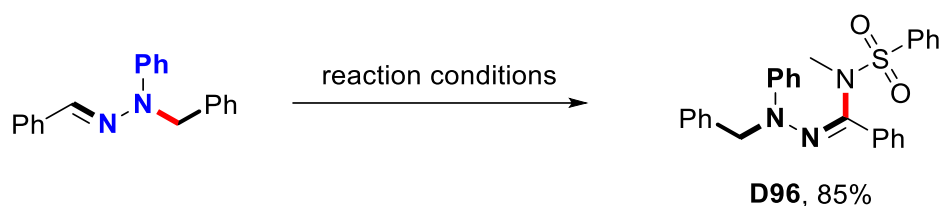


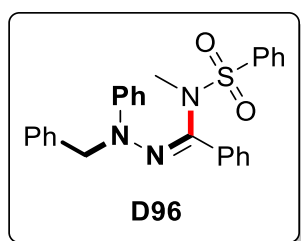
Figure S4 Time-conversion-plot for the reaction

9. Possible reactions in combination with hydrazone

9.1 Synthesis of D96 from hydrazone D1



Using a nitrogen-filled glove box, an oven-dried yang-type tube (50 mL volume) was charged with a magnetic stirring bar, **D1** (0.1 mmol), *N*-methyl-*N*-((phenylsulfonyl)oxy)benzenesulfonamide (0.2 mmol), *fac*-Ir(ppy)₃ (0.002 mmol), Na₂HPO₄ (0.2 mmol), and DCE (1.0 mL) were added in sequence under magnetic stirring. Then the seal tube was closed tightly with a teflon cap, immersed into a pre-cooled metal sand bath (25 °C) under visible-light irradiation. After 90 hours, a small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation, respectively. Purification of the remainder by column chromatography on silica gel gave the corresponding product **D96** (pentane/ethyl acetate = 50/1-5/1) in the reported yield.



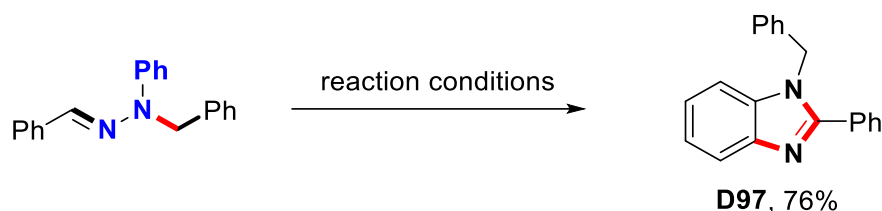
***N'*-benzyl-*N*-methyl-*N'*-phenyl-*N*-(phenylsulfonyl)benzohydrazonamide (Z:E = 1.9:1.0)^[3]**

The title compound was prepared according to the general procedure and purified by column chromatography to give the yellow oil 38.7 mg, 85% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 50 : 1 – 5:1).

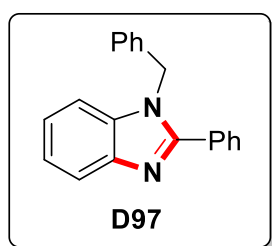
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.89 (d, *J* = 8.4 Hz, 1.32H), 7.67 – 7.55 (m, 1.82H), 7.51 (d, *J* = 8.0 Hz, 1.45H), 7.45 – 7.36 (m, 1.55H), 7.30 – 7.17 (m, 6.98H), 7.17 – 7.04 (m, 3.85H), 6.99 – 6.92 (m, 1.63H), 6.83 (t, *J* = 4.4 Hz, 0.66H), 6.73 (d, *J* = 8.4 Hz, 1.26H), 4.99 (s, 0.68H), 4.43 (s, 1.31H), 3.07 (s, 1.95H), 2.79 (s, 1.04H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 157.34, 150.24, 149.28, 141.69, 139.09, 138.76, 137.51, 136.82, 135.04, 132.95, 132.81, 132.68, 129.90, 129.38, 128.81, 128.76, 128.64, 128.35, 128.29, 128.22, 128.03, 127.92, 127.83, 127.62, 126.92, 126.81, 60.06, 56.53, 37.98, 35.12 ppm.

9.2 Synthesis of D97 from hydrazone D1



Under the nitrogen, an oven-dried yang-type tube (50 mL volume) was charged with a magnetic stirring bar, **D1** (0.5 mmol), $\text{Cu}(\text{OTf})_2$ (0.5 mmol) and toluene (2 mL) were added in sequence under magnetic stirring. Then the seal tube was closed tightly with a teflon cap, immersed into a pre-heated metal sand bath (110 °C). After 12 hours the reaction was cooled, a small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation, respectively. Purification of the remainder by column chromatography on silica gel gave the corresponding product **D97** (pentane/ethyl acetate = 50/1-5/1) in the reported yield.



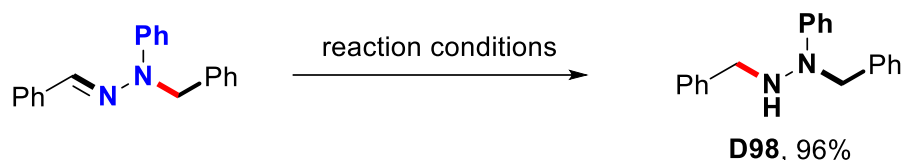
1-benzyl-2-phenyl-1H-benzo[d]imidazole^[4]

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 108.1 mg, 76% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 50 : 1 – 5:1).

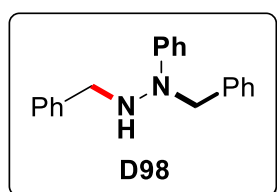
^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.91 (d, J = 8.0 Hz, 1H), 7.70 (d, J = 7.6 Hz, 2H), 7.47 (d, J = 7.2 Hz, 3H), 7.36-7.30 (m, 4H), 7.26-7.21 (m, 2H), 7.12 (d, J = 6.8 Hz, 2H), 5.47 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 154.32, 143.01, 136.53, 136.11, 132.72, 130.23, 129.51, 129.22, 128.93, 128.02, 126.11, 123.32, 123.01, 120.12, 110.71, 48.52 ppm.

9.3 Synthesis of D98 from hydrazone D1



Under the nitrogen, an oven-dried yang-type tube (50 mL volume) was charged with a magnetic stirring bar, **D1** (0.5 mmol), and EtOH (2 mL) were added in sequence under magnetic stirring. Then sodium amalgam (1.0 mmol) was added and the seal tube was closed tightly with a teflon cap, immersed into a pre-cooled metal sand bath (0 °C). After 12 hours the reaction was cooled, a small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation, respectively. Purification of the remainder by column chromatography on silica gel gave the corresponding product **D98** (pentane/ethyl acetate = 50/1-5/1) in the reported yield.



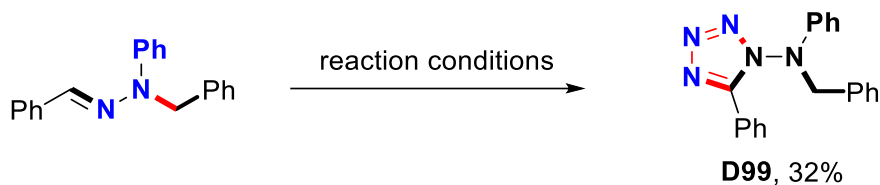
1,2-dibenzyl-1-phenylhydrazine^[5]

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 138.4 mg, 96% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 50 : 1 – 5:1).

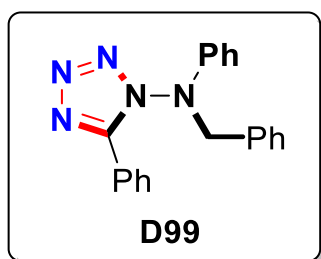
^1H NMR (400 MHz, 23.0 °C, CDCl_3) δ 7.20–7.35 (m, 9H), 7.02 (d, J = 8.4 Hz, 2H), 6.77–6.88 (m, 4H), 5.68 (s, 1H), 4.77 (s, 2H) ppm.

^{13}C NMR (101 MHz, 23.0 °C, CDCl_3) δ 151.72, 151.21, 140.52, 129.62, 128.91, 126.53, 126.11, 123.33, 113.23, 65.52 ppm.

9.4 Synthesis of D99 from hydrazone D1



An oven-dried three-necked round-bottomed flask (25 mL) was charged with hydrazone **D1** (0.5 mmol), LiClO₄ (0.5 mmol) and TMSN₃ (2.5 mmol). Then the flask was equipped with a condenser, a reticulated vitreous carbon (RVC) anode (100 PPI, 1 cm x 1 cm x 1 cm), and a platinum plate (1 cm x 1 cm) cathode. After MeOH (2 mL) and MeCN (4 mL) were added, the reaction mixture was stirred and electrolyzed at a constant current of 10 mA under 0 °C for 2 hours (2.5 F). When the reaction was finished, a small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation, respectively. Purification of the remainder by column chromatography on silica gel gave the corresponding product **D99** (pentane/ethyl acetate = 50/1-5/1) in the reported yield.



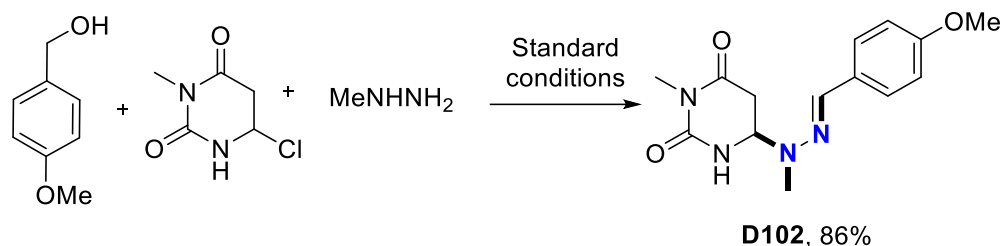
N-benzyl-*N*,5-diphenyl-1*H*-tetrazol-1-amine^[6]

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 52.4 mg, 32% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 50 : 1 – 5:1).

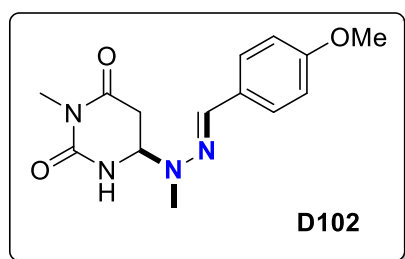
¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 7.72 (dd, *J* = 8.4, 1.2 Hz, 2H), 7.50 – 7.47 (m, 1H), 7.42 – 7.37 (m, 4H), 7.20 – 7.14 (m, 2H), 7.06 (dd, *J* = 8.4, 6.8 Hz, 2H), 6.99 (ddt, *J* = 8.8, 7.6, 1.2 Hz, 4H), 4.97 (s, 2H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 153.21, 147.62, 133.11, 131.42, 129.82, 129.21, 128.61, 128.41, 128.40, 124.12, 122.81, 117.00, 60.82 ppm.

9.5. Synthesis of **D102**



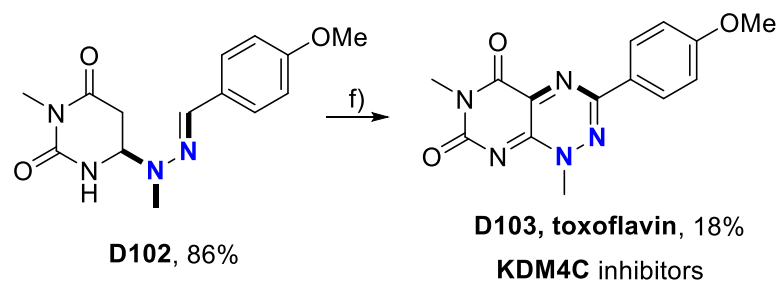
Using a nitrogen-filled glove box, an oven-dried Schlenk Flask (38 mL volume) was charged with a magnetic stirring bar, $\text{MnBr}(\text{CO})_5$ (1.0 mol%), **L11** (1.2 mol%), $t\text{BuOK}$ (1.0 mmol), PMPCH_2OH (0.8 mmol), MeNHNH_2 (0.5 mmol), 6-chloro-3-methyl-2,4-dihydropyrimidine-2,4(1H,3H)-dione (0.5 mmol) (0.7 mmol) and THF (3 mL). Then the Schlenk Flask was closed tightly with a teflon cap, removed from the glove box and immersed into a pre-heated metal bath (100 °C) condensation reflux 24 hours under N_2 atmosphere. After the reaction was finished, the reaction was analyzed by TLC to monitor product formation. Then a small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation. Purification of the remainder by column chromatography on silica gel gave the corresponding product **D102** (pentane/ethyl acetate) in the reported yield.



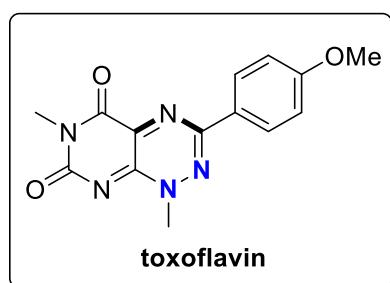
6-(2-(4-methoxybenzylidene)-1-methylhydrazinyl)-3-methyl-2,4-dihydropyrimidine-2,4(1H,3H)-dione^[7]

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 124.8 mg, 86% yield. Purification by column chromatography on silica gel (pentane/ethyl ether = 50 : 1 – 1:1, CH_3OH 5%). $^1\text{H NMR}$ (400 MHz, 23.0 °C, $\text{DMSO}-d_6$) δ 10.62 (s, 1H), 7.90–7.97 (m, 3H), 6.99 (d, $J = 8.4$ Hz, 2H), 5.18 (s, 1H), 3.81 (s, 3H), 3.12 (s, 3H), ppm.

9.6 Synthesis of Toxoflavin **D103**



Using a nitrogen-filled glove box, an oven-dried pressure tube (50 mL volume) was charged with a magnetic stirring bar, **D102** (0.5 mmol), CH₃COOH (4 mL) and H₂O (0.4 mL) were added in sequence under magnetic stirring. Then NaNO₂ (0.75 mmol) was added the seal tube was closed tightly with a teflon cap, removed from the glove box, immersed into a pre-cooled metal sand bath (0 °C). After 6 hours, the reaction mixture was diluted with water and extracted with DCM. Organic layers were combined, dried over sodium sulphate, filtered and evaporated under reduced pressure to obtain crude product. A small aliquot of the organic phase was analyzed by GC and GC-MS to monitor product formation. Purification of the remainder by column chromatography on silica gel gave the corresponding product **toxoflavin** (MeOH/EtOAc) in the reported yield.



3-(4-methoxyphenyl)-1,6-dimethylpyrimido[5,4-e][1,2,4]triazine-5,7(1H,6H)-dione^[7]

The title compound was prepared according to the general procedure and purified by column chromatography to give a yellow solid 26.9 mg, 18% yield. Purification by column chromatography on silica gel (MeOH/EtOAc = 50 : 1 – 5:1).

¹H NMR (400 MHz, 23.0 °C, CDCl₃) δ 8.13 (d, 2H, *J* = 8.4 Hz), 7.14 (d, 2H, *J* = 8.8 Hz), 4.03 (s, 3H), 3.85 (s, 3H), 3.27 (s, 3H) ppm.

¹³C NMR (101 MHz, 23.0 °C, CDCl₃) δ 162.32, 159.51, 154.52, 151.71, 149.72,

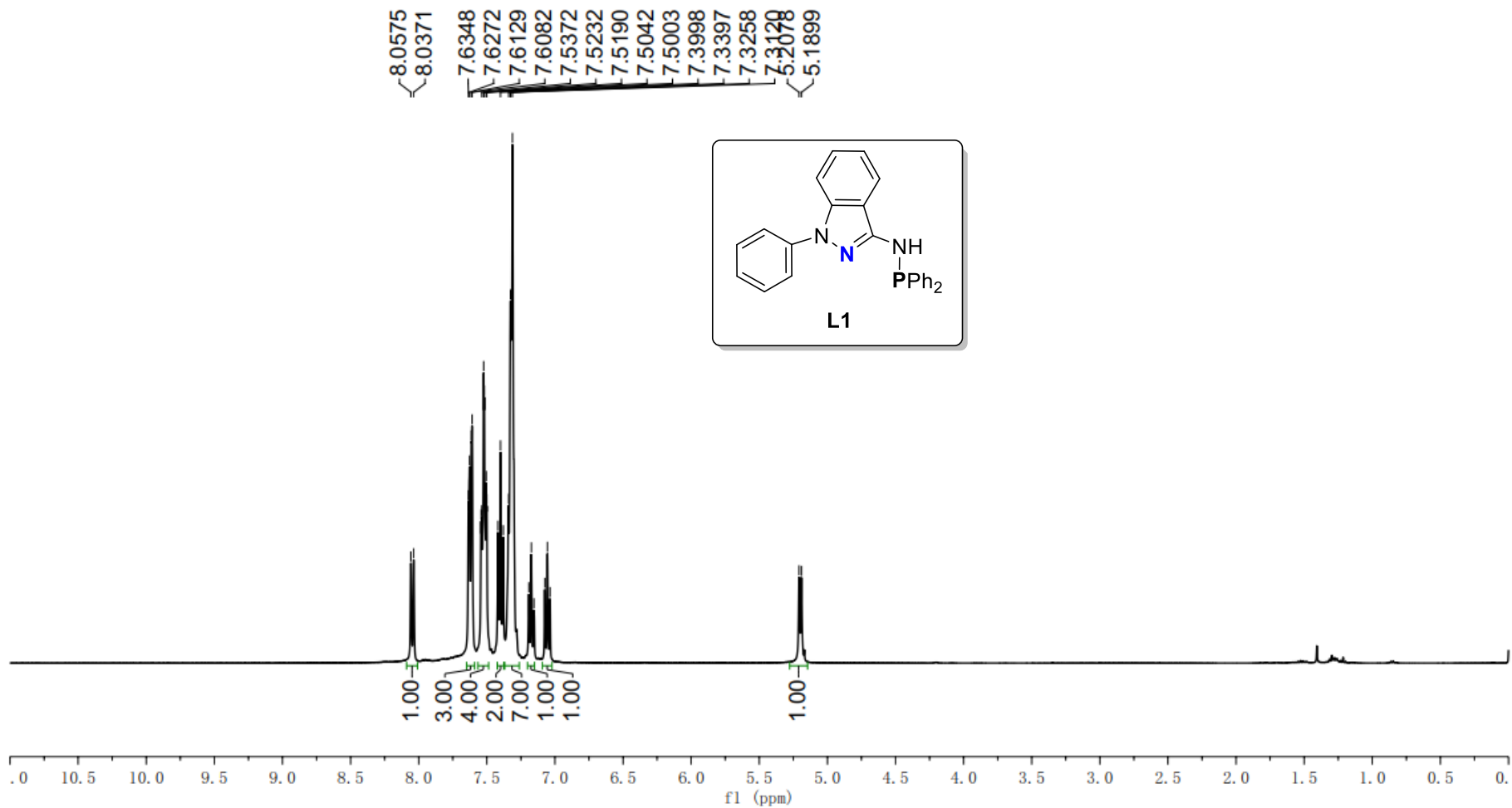
146.81, 128.82, 125.52, 115.21, 55.92, 43.22, 28.73 ppm.

9. References

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10. NMR Spectra

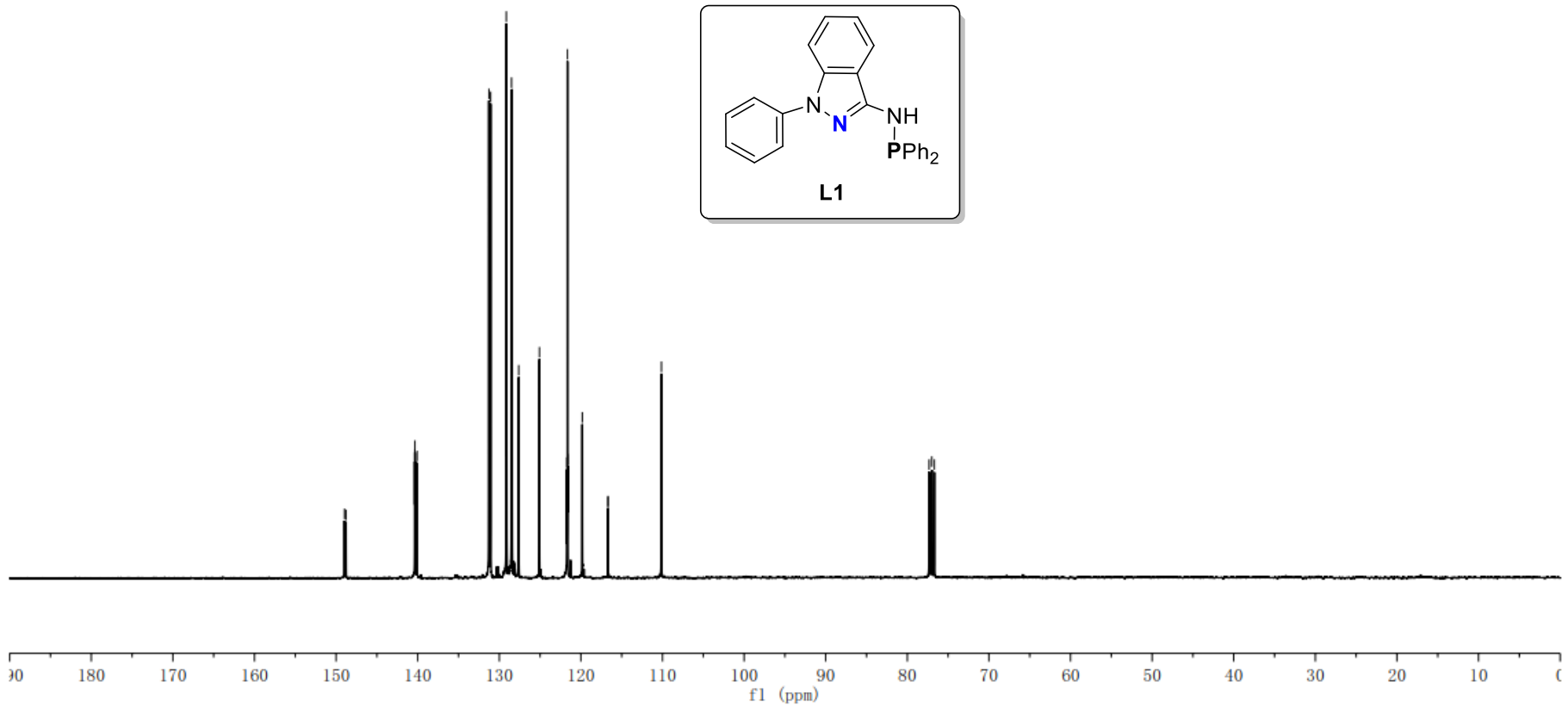
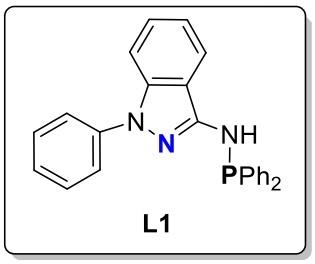
2023-06-01 yy0035 616-11-4



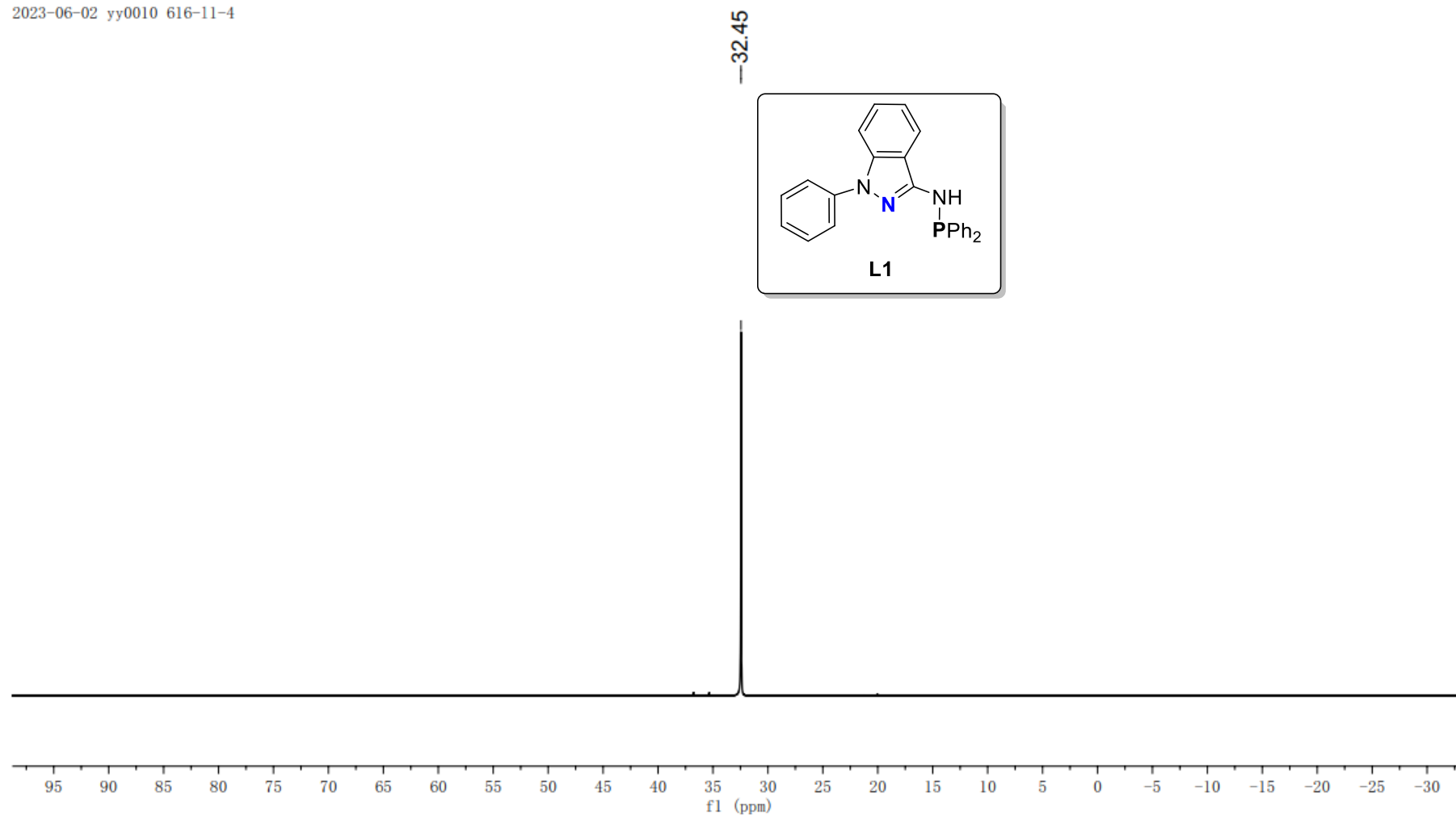
2023-06-02 yy0010 616-11-4

148.98
148.79
140.35
140.33
140.07
131.27
131.06
129.14
129.10
128.49
128.43
121.64
110.94

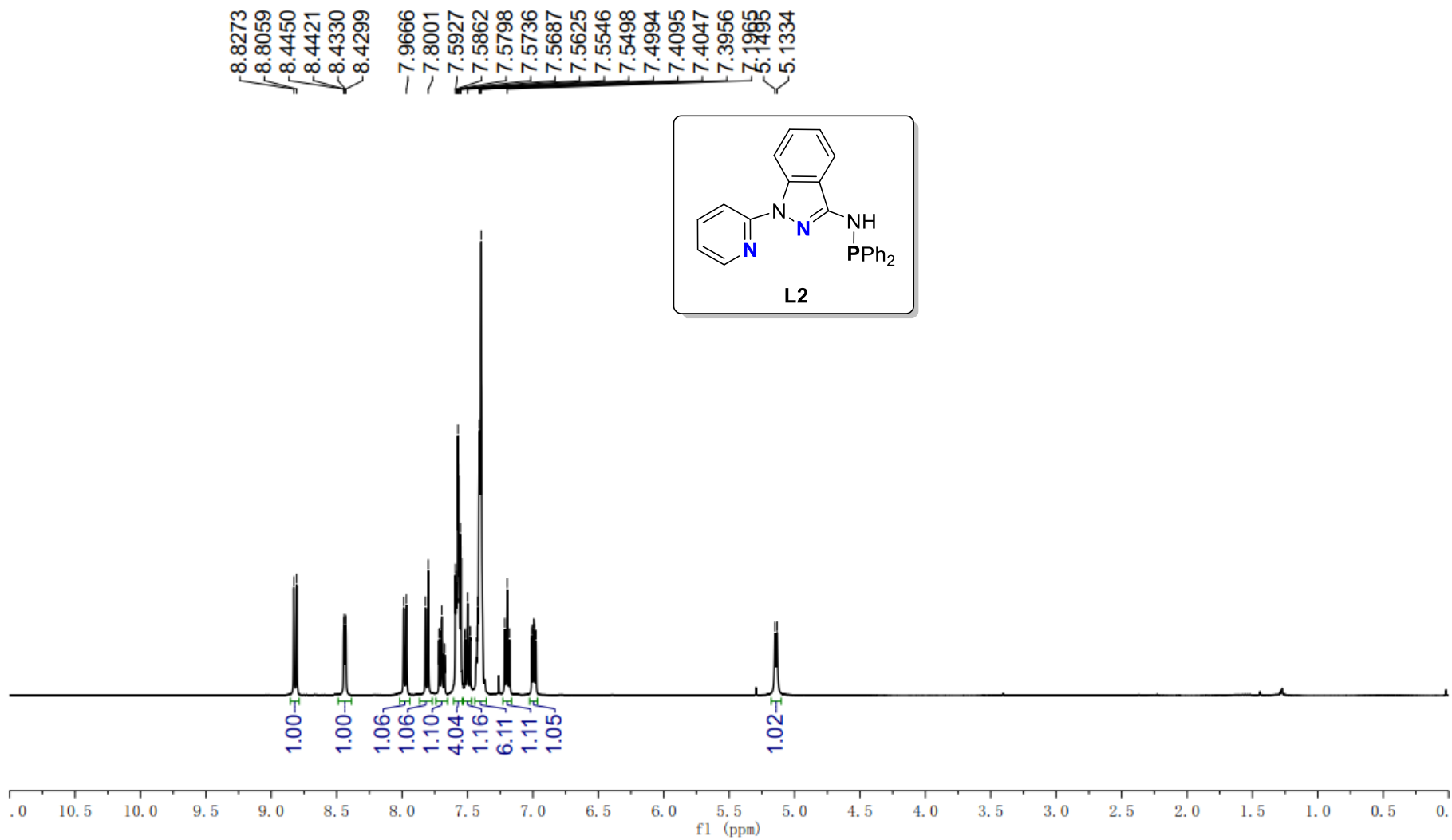
77.32
77.00
76.68



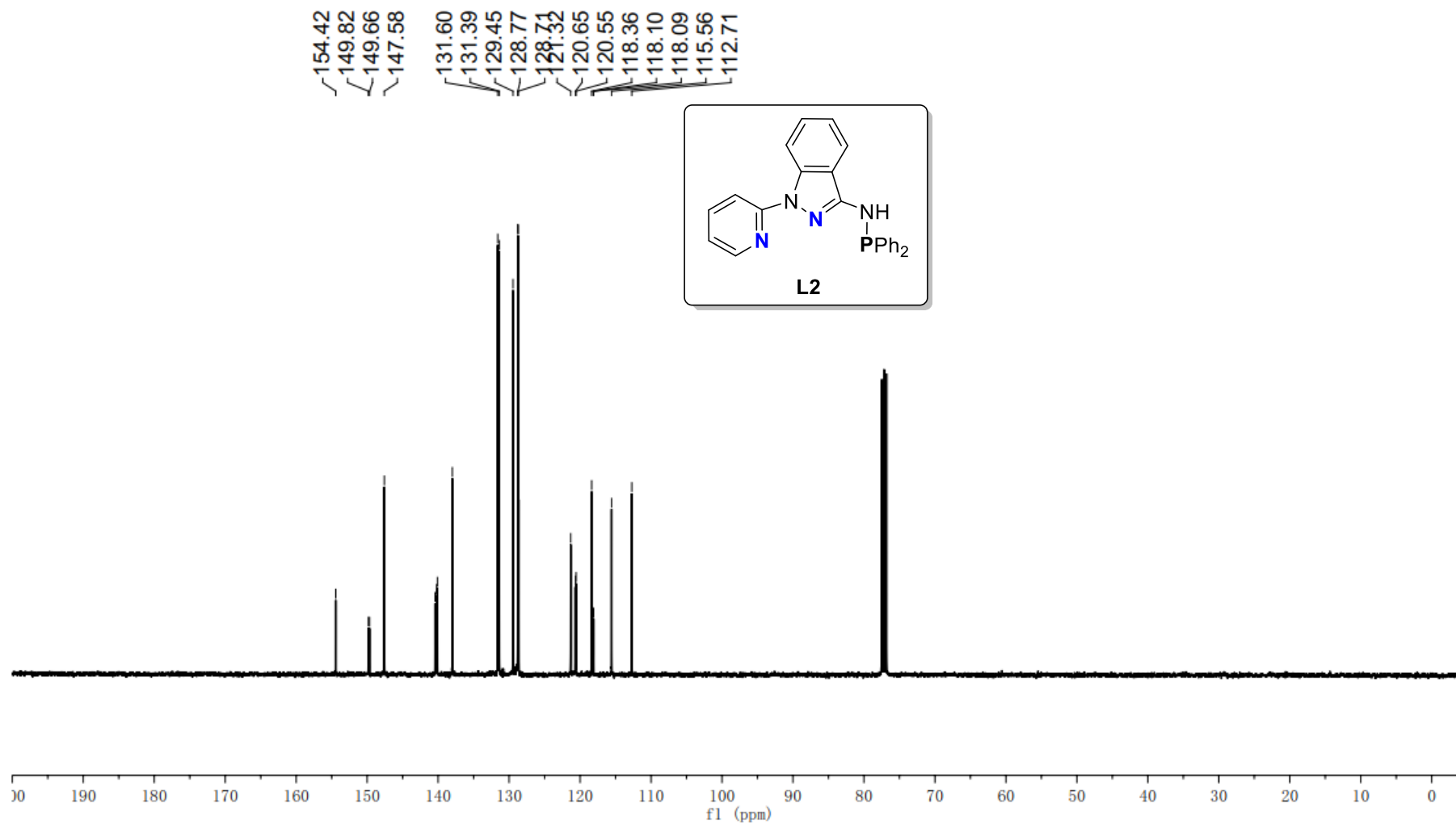
2023-06-02 yy0010 616-11-4



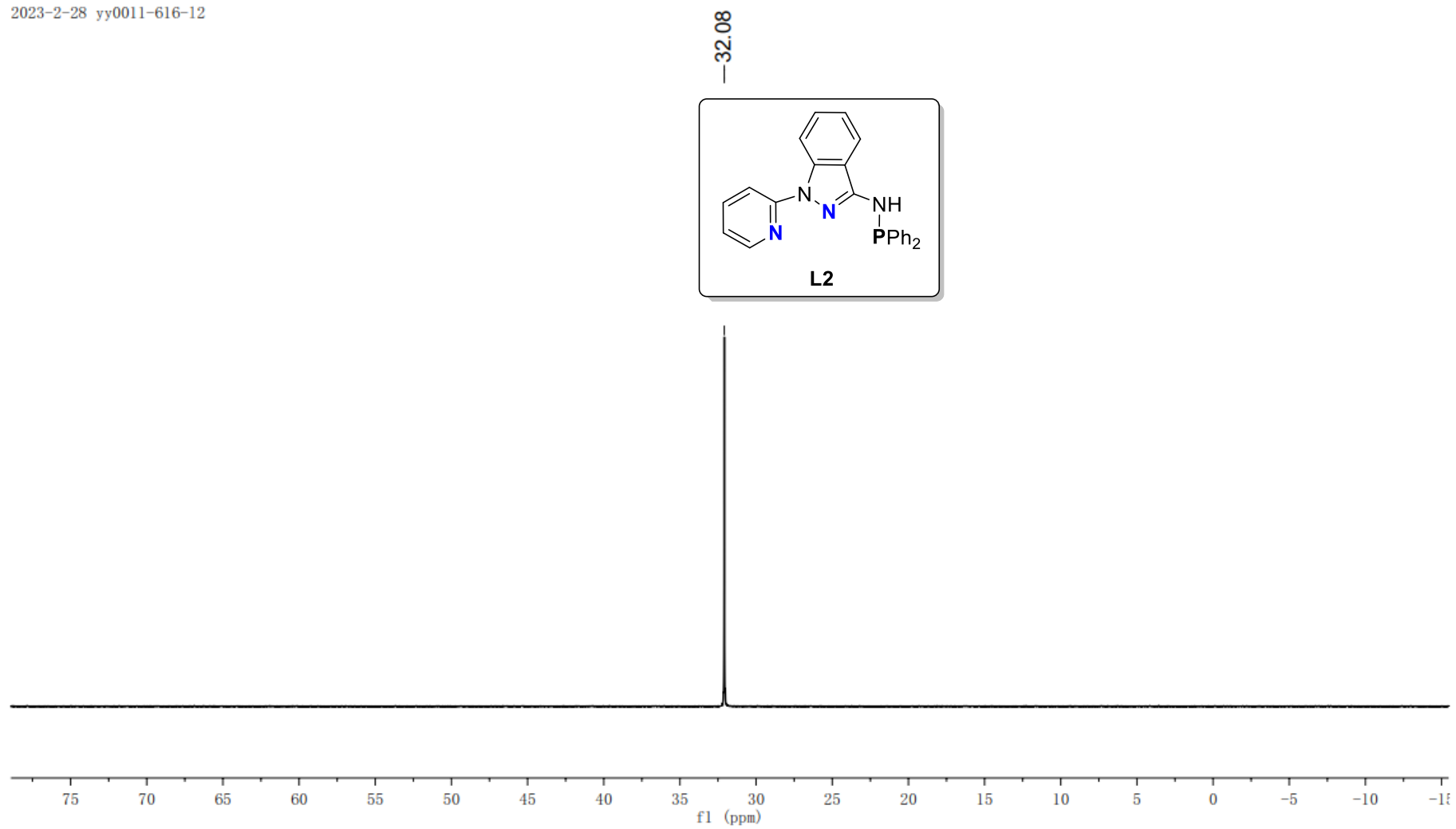
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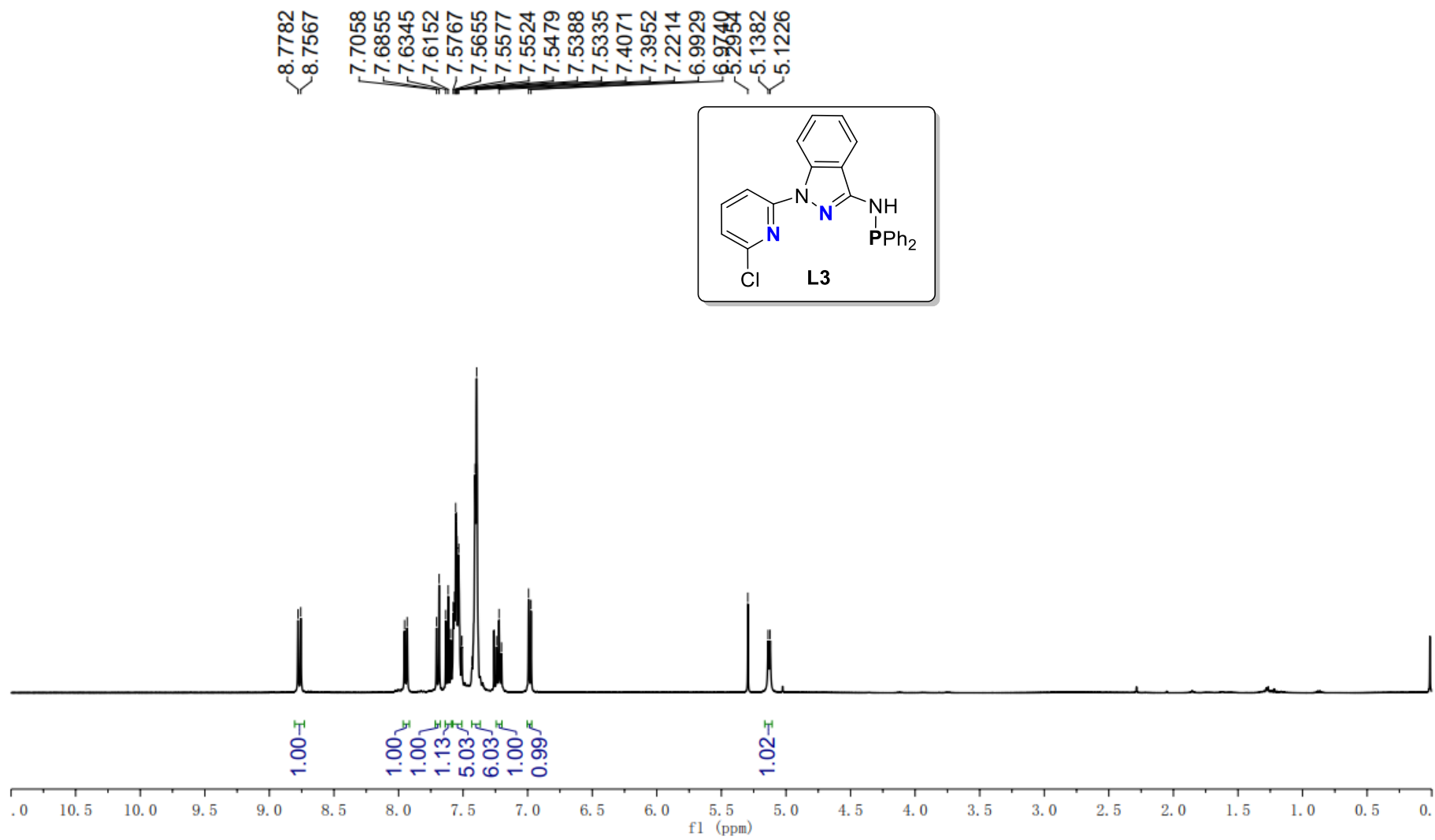
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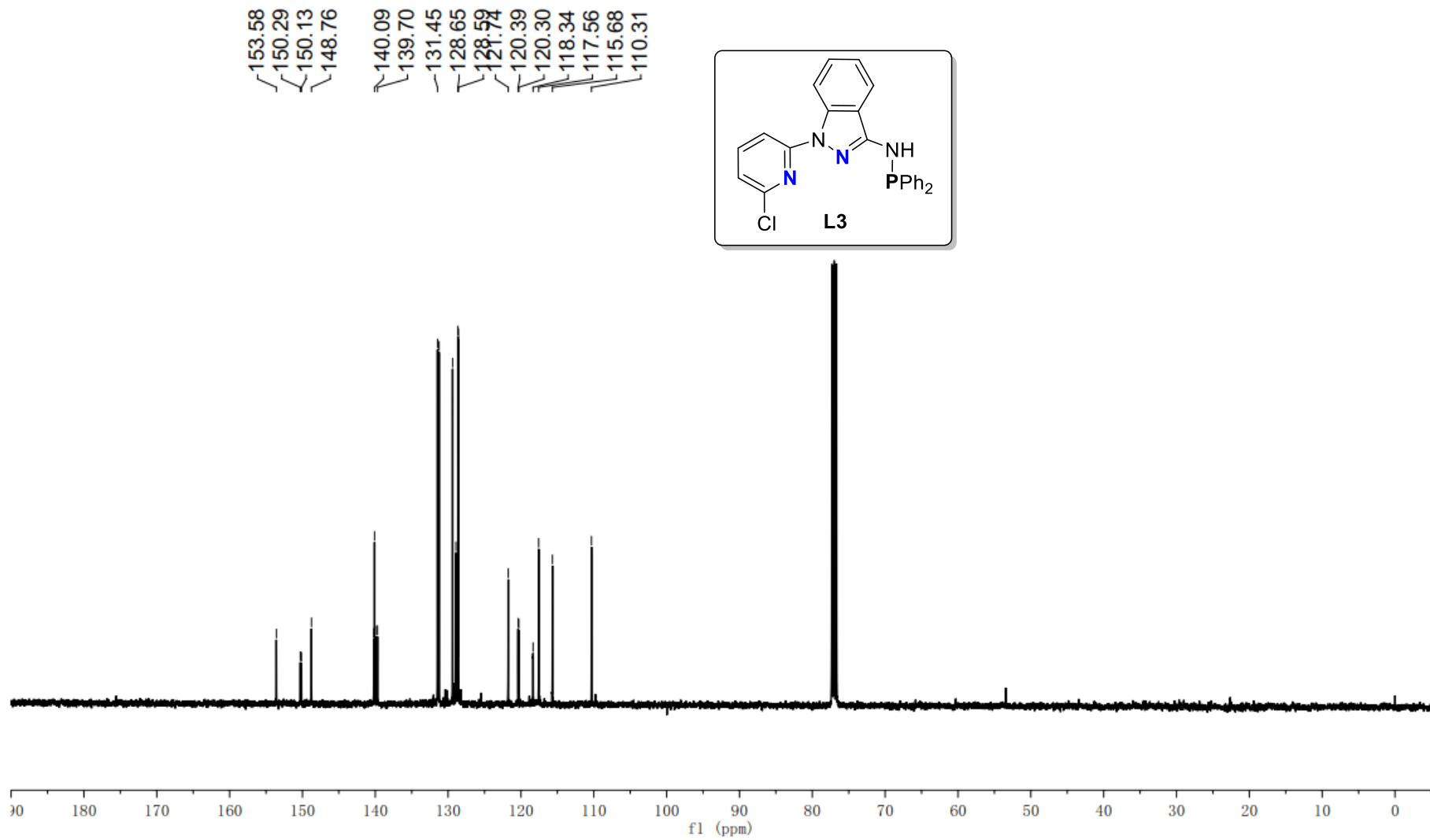


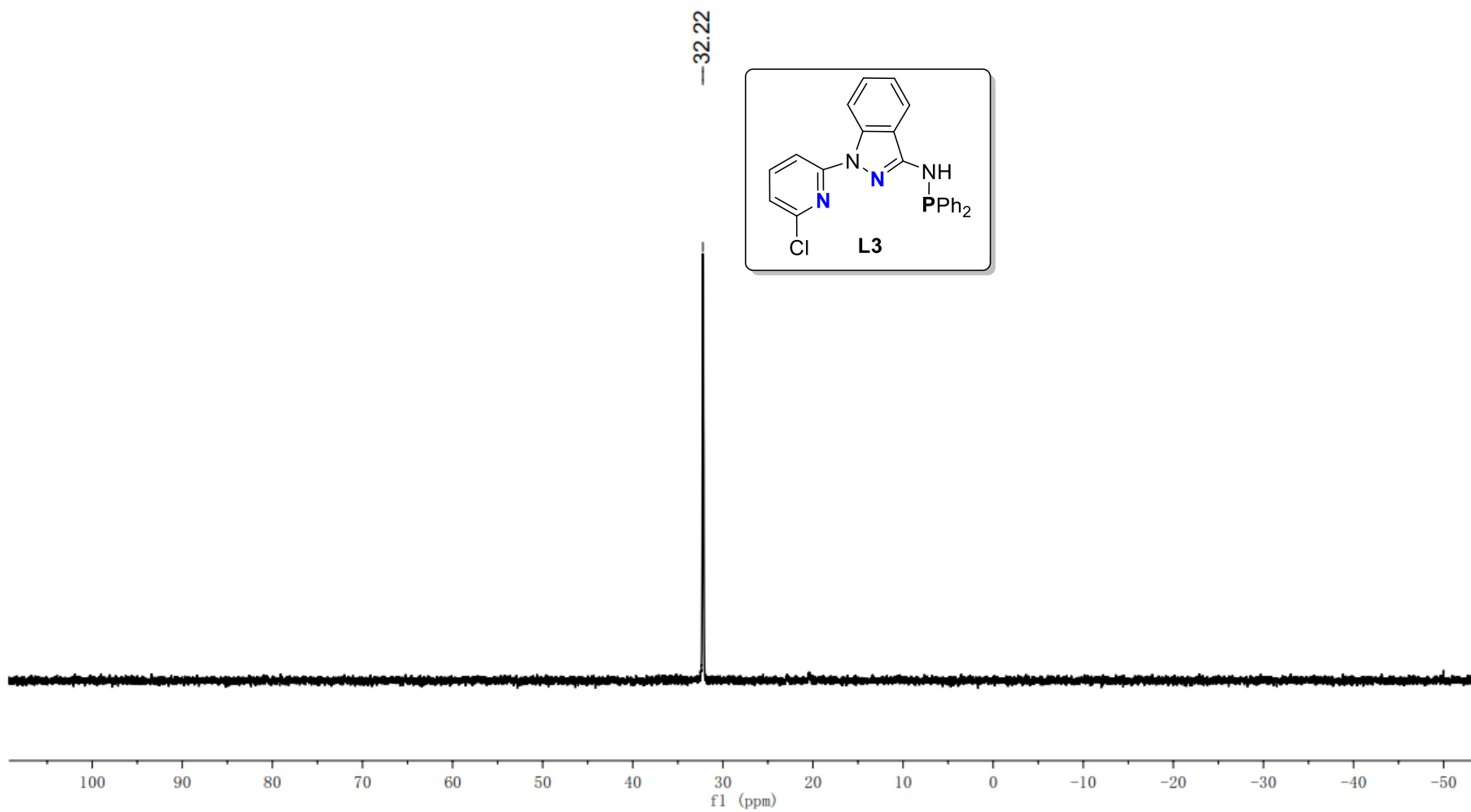
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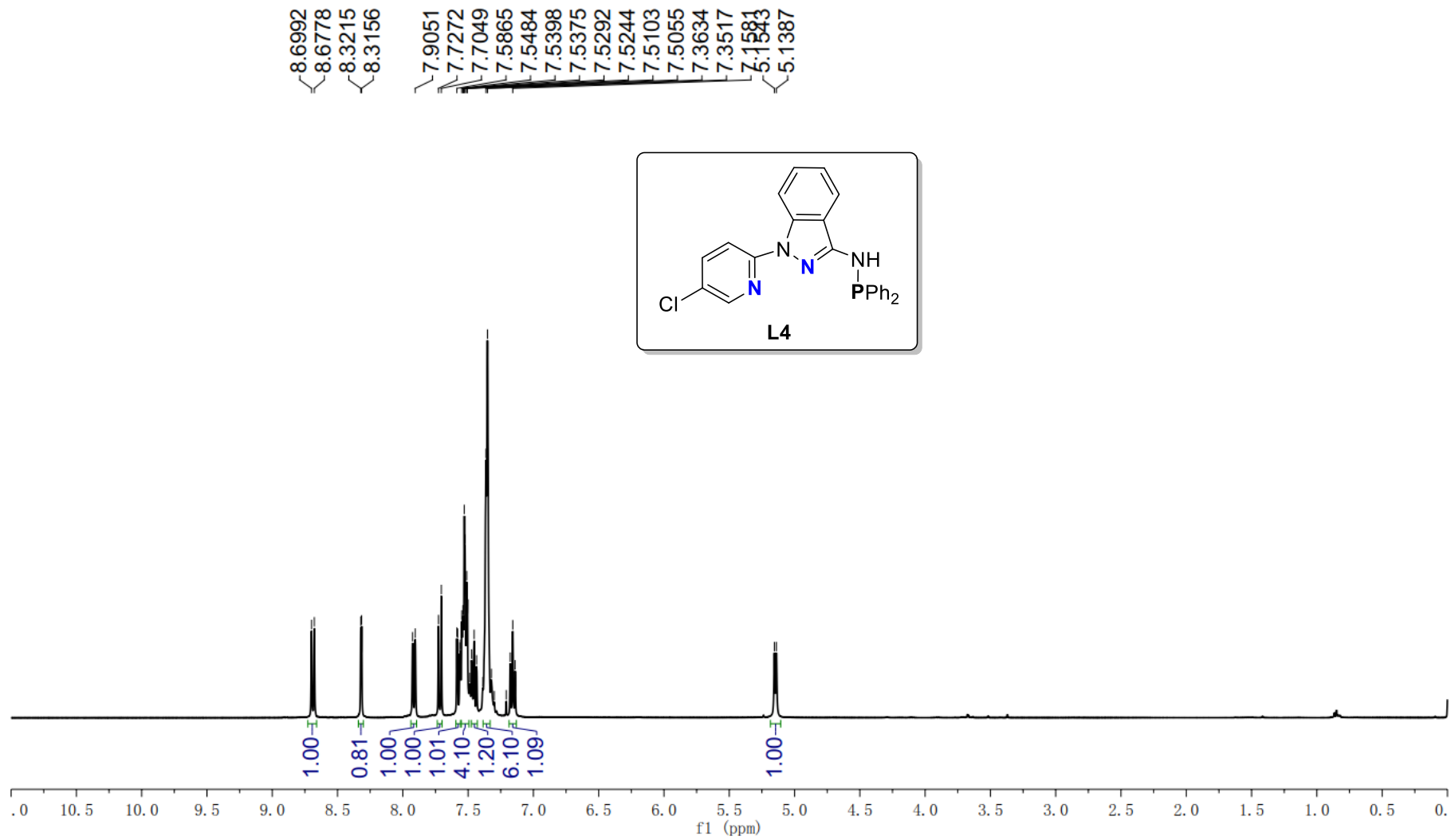


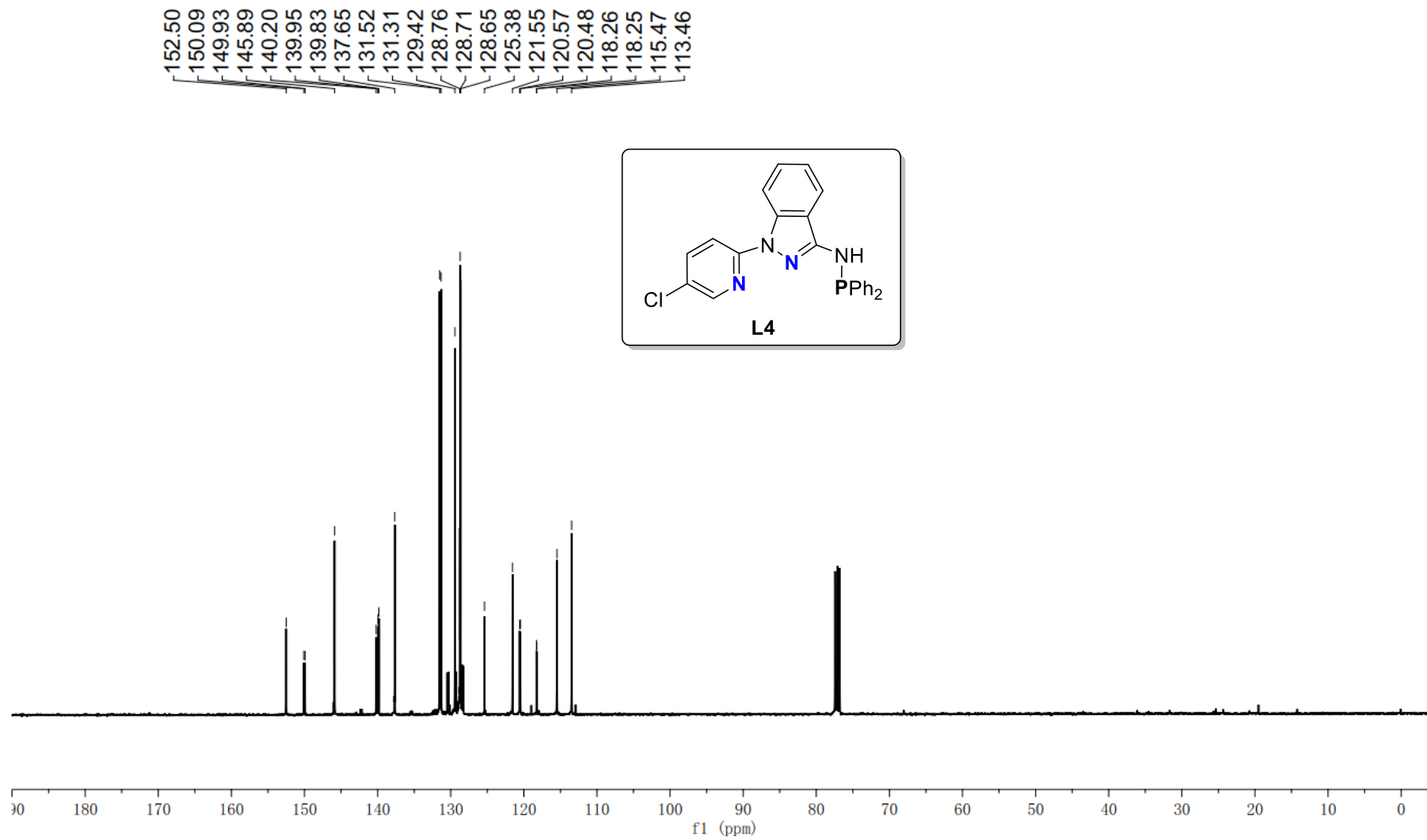
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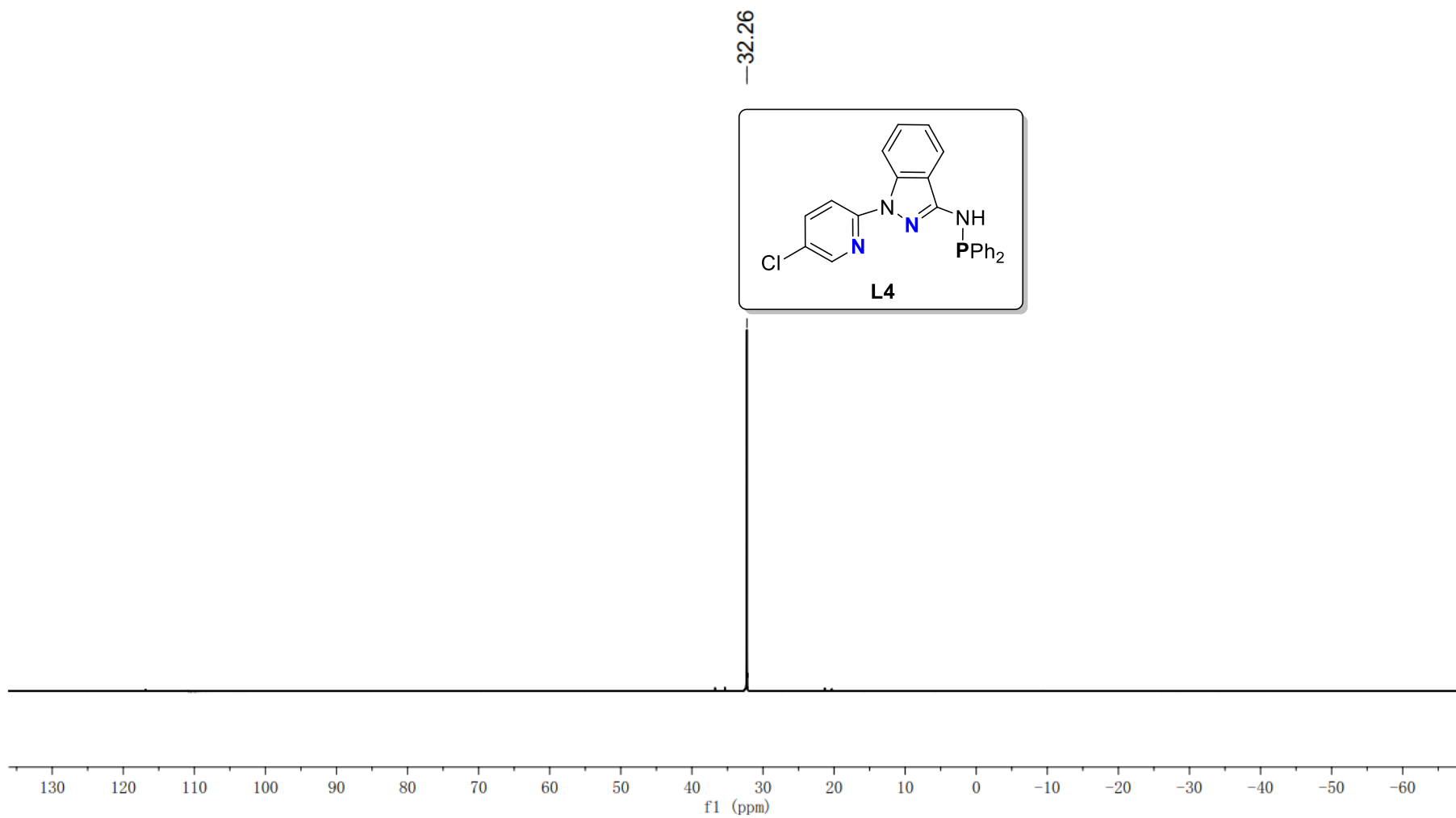




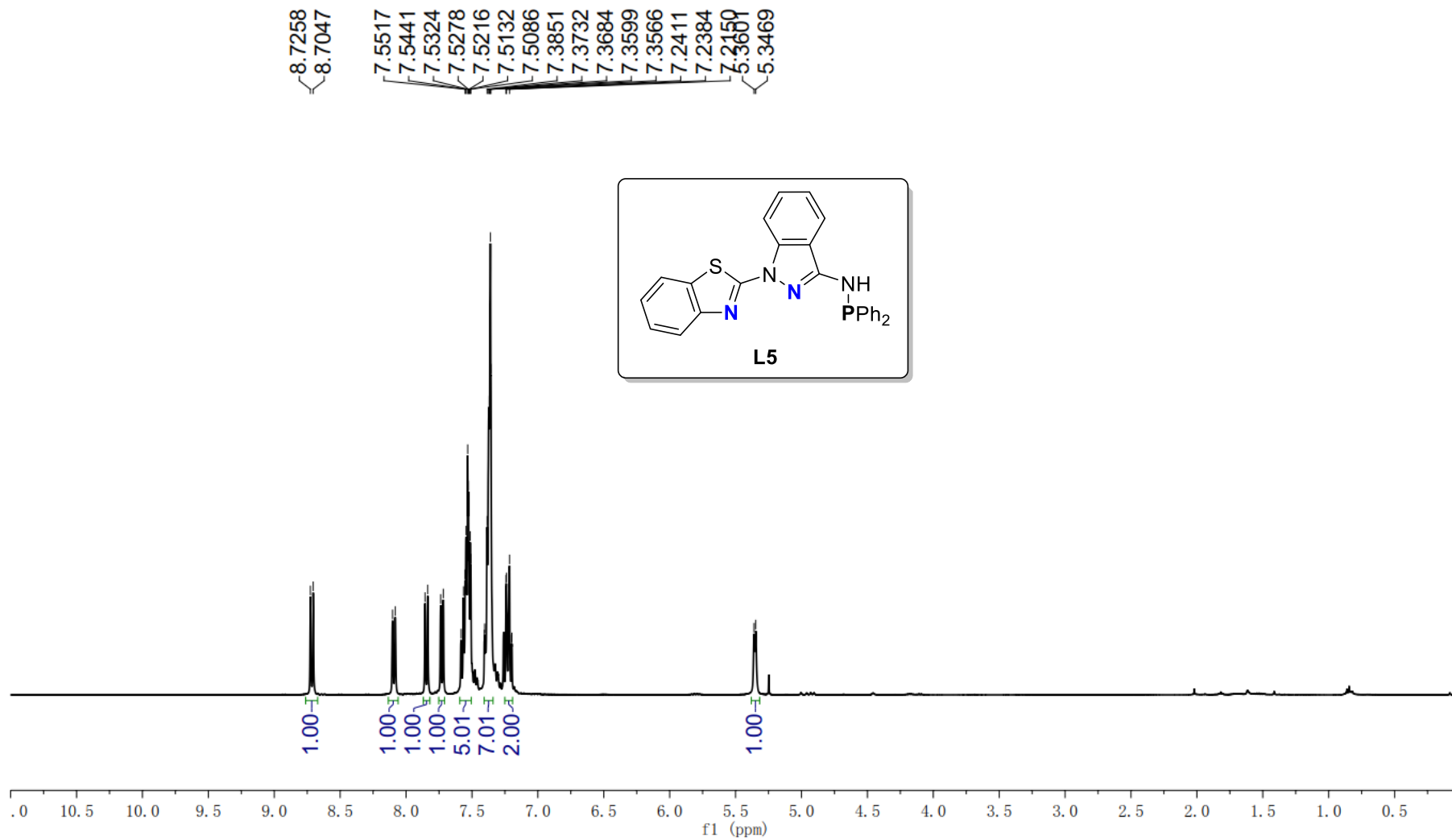




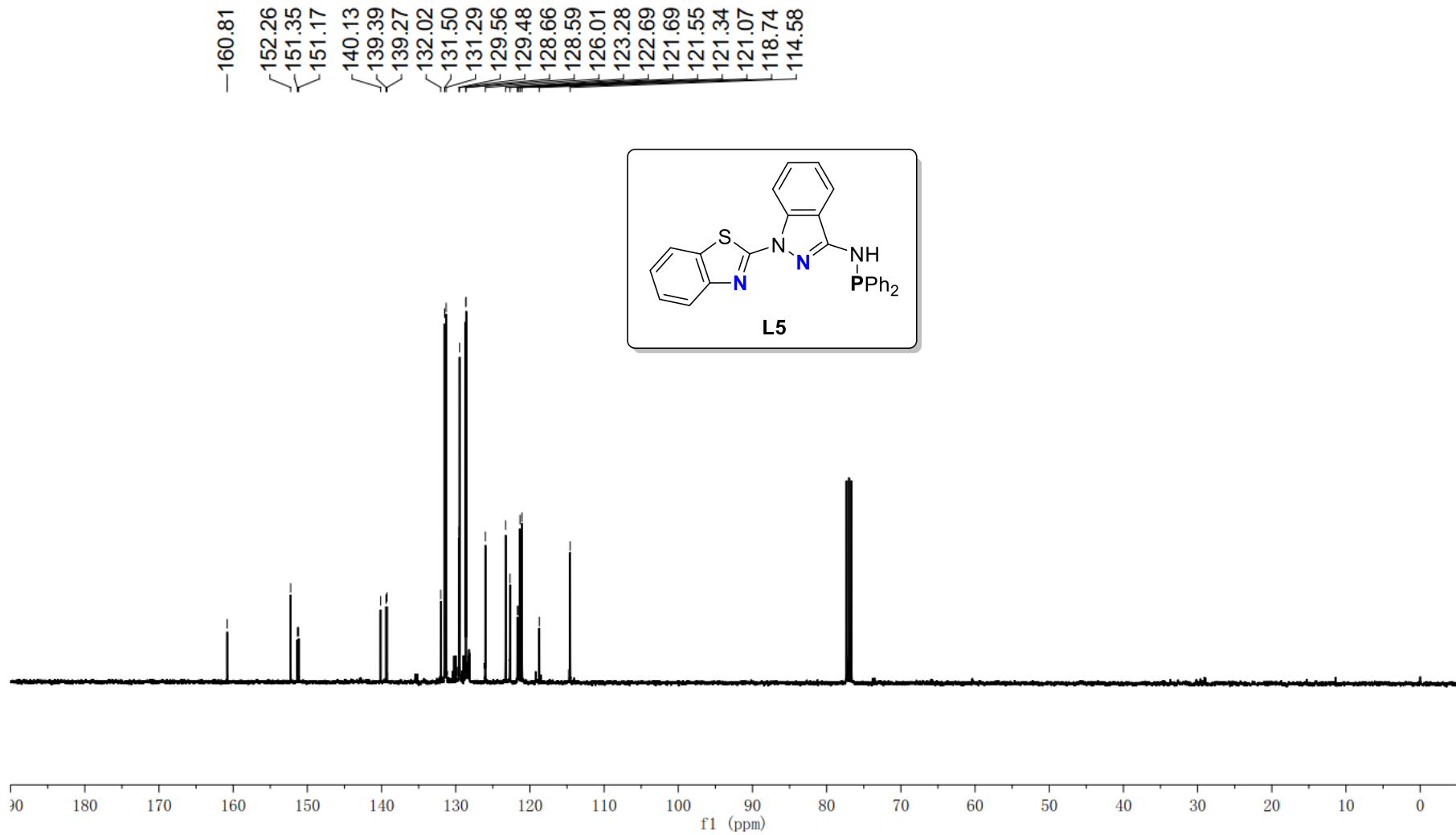




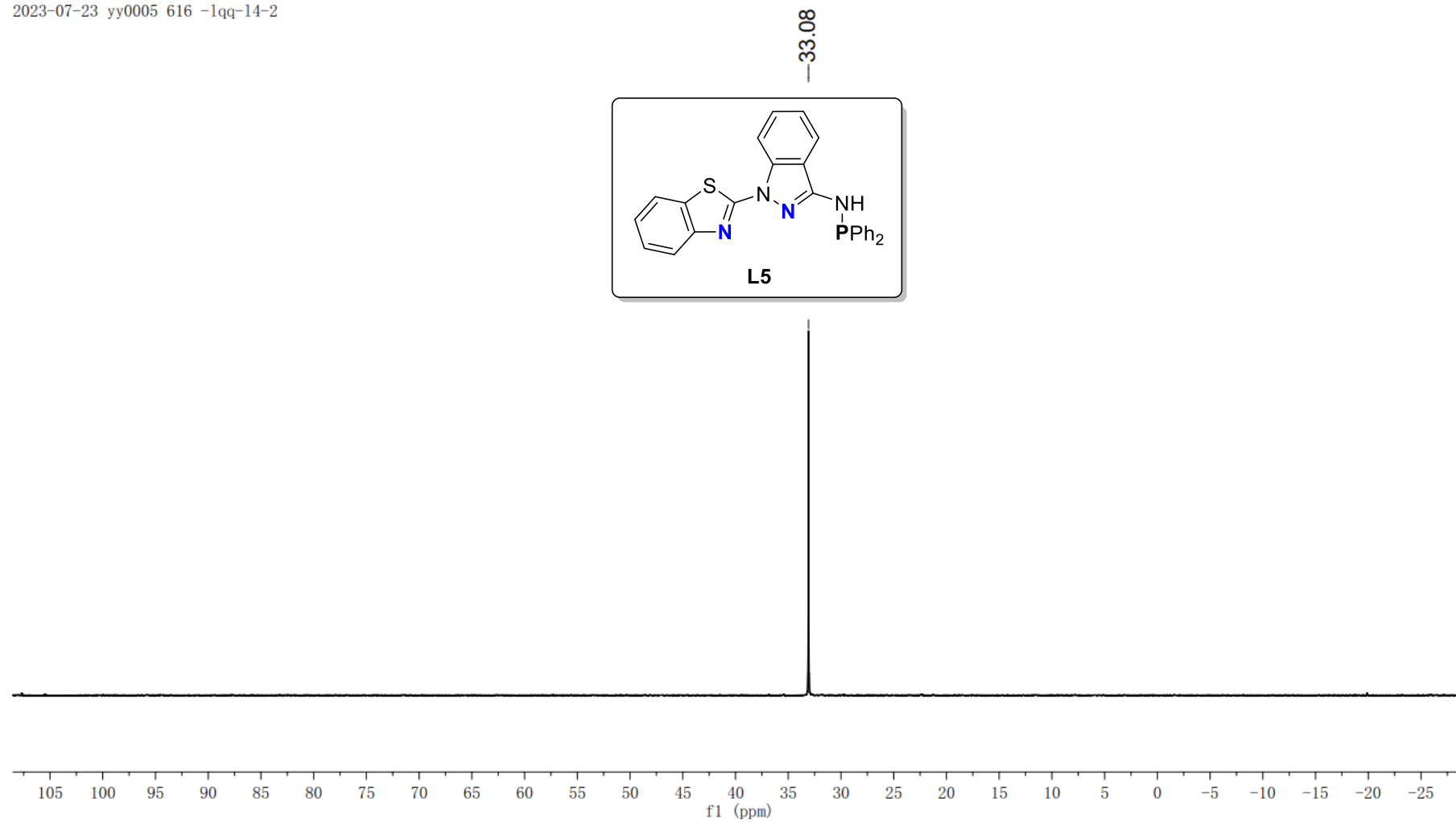
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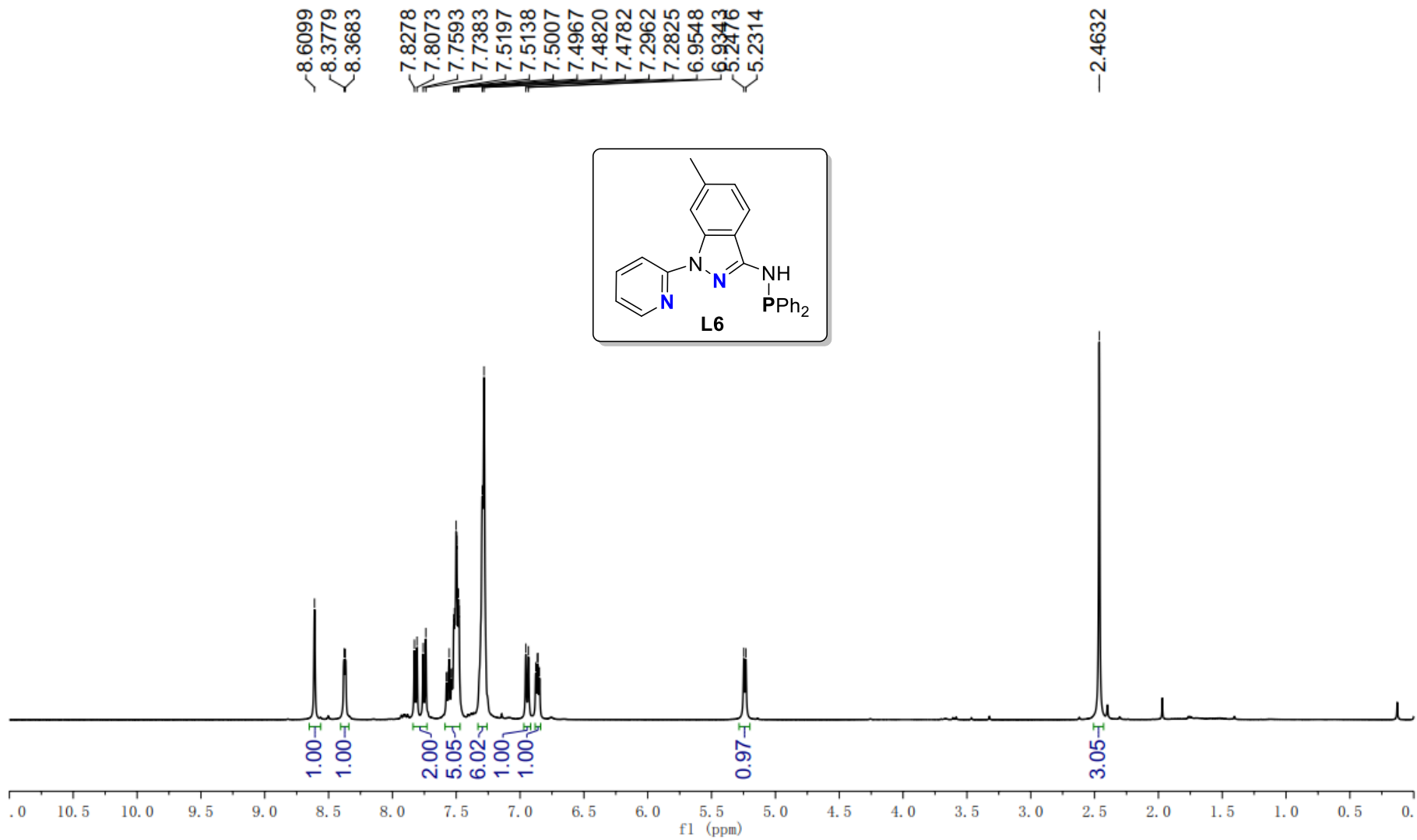


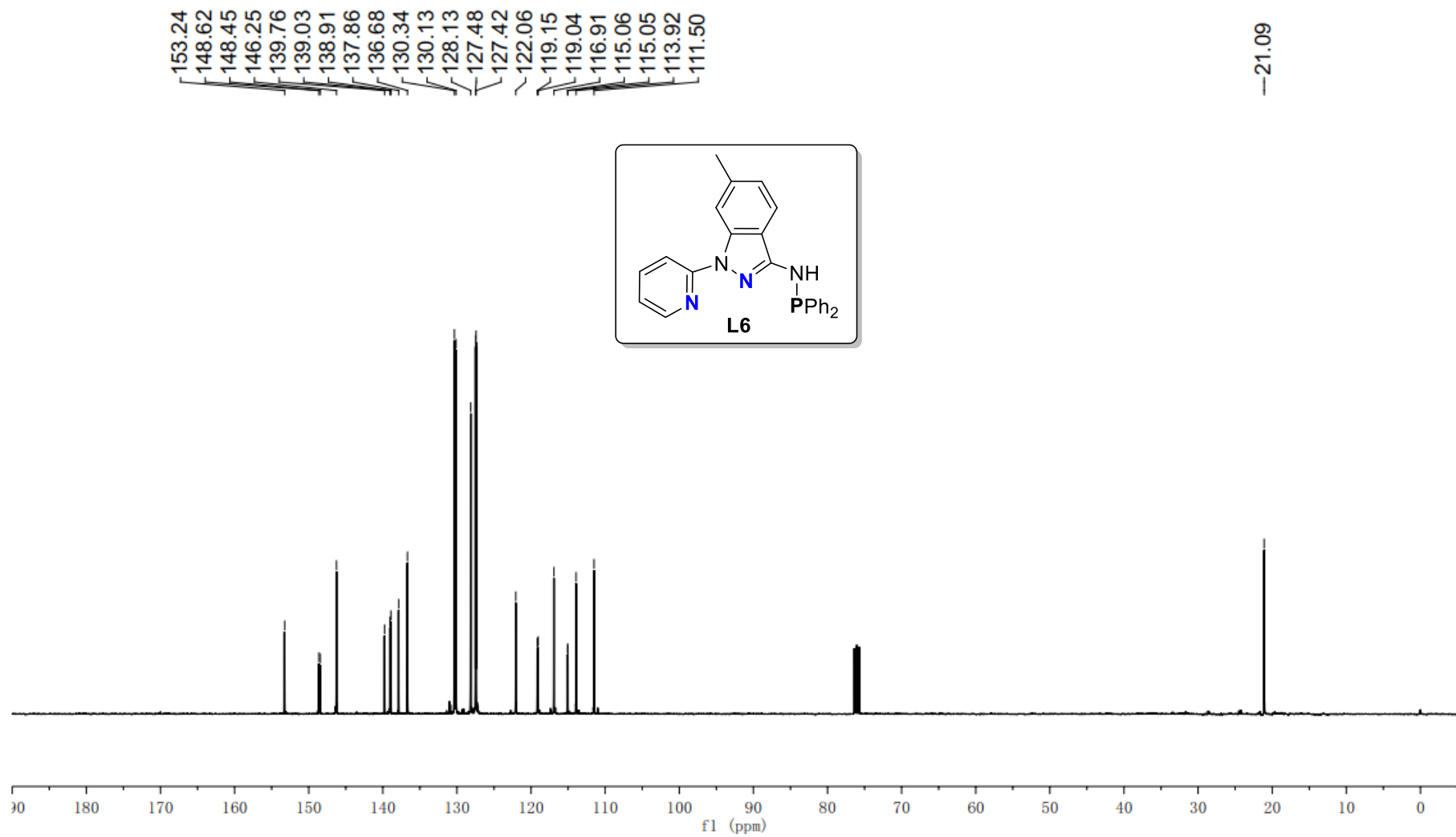
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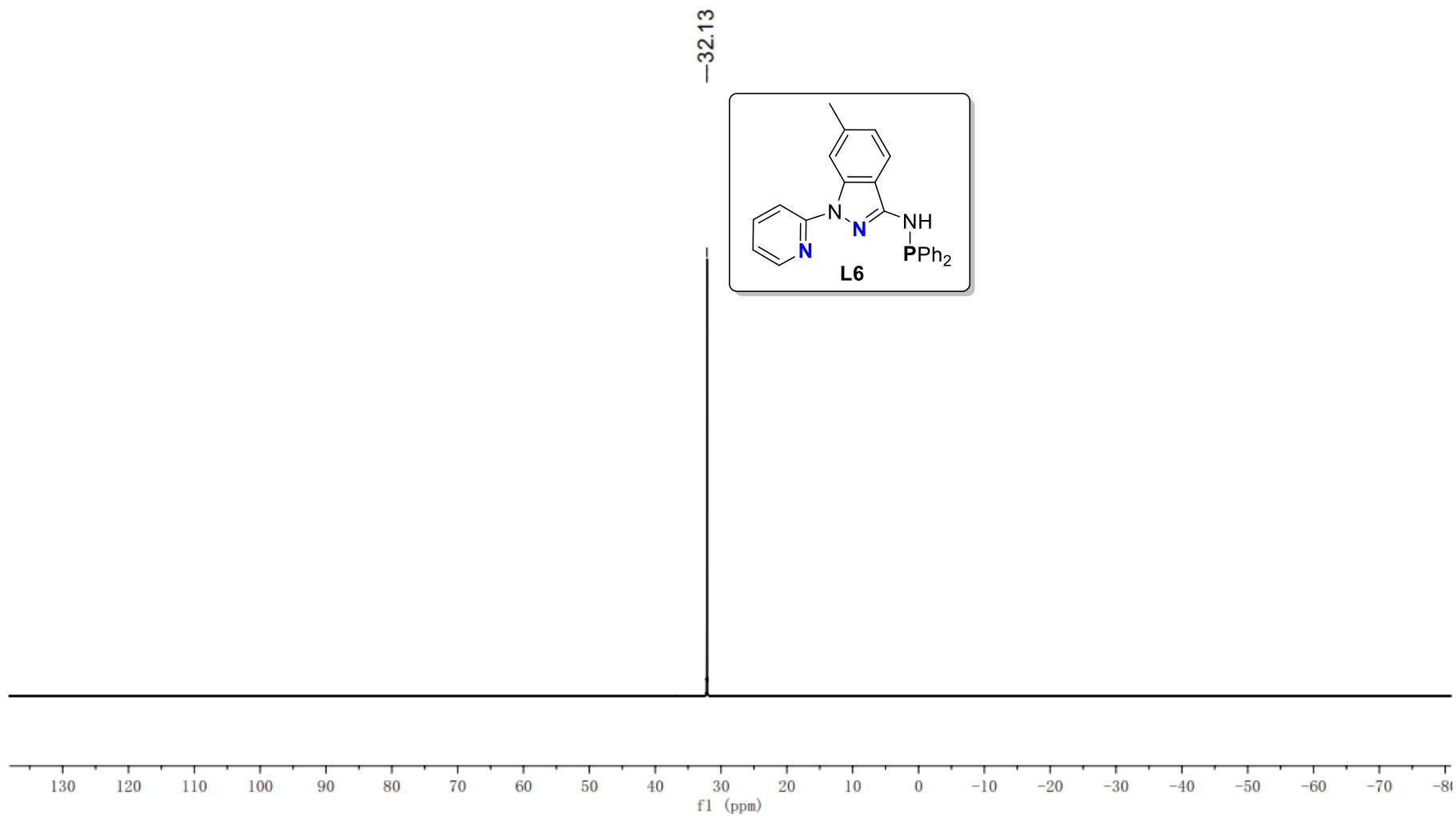


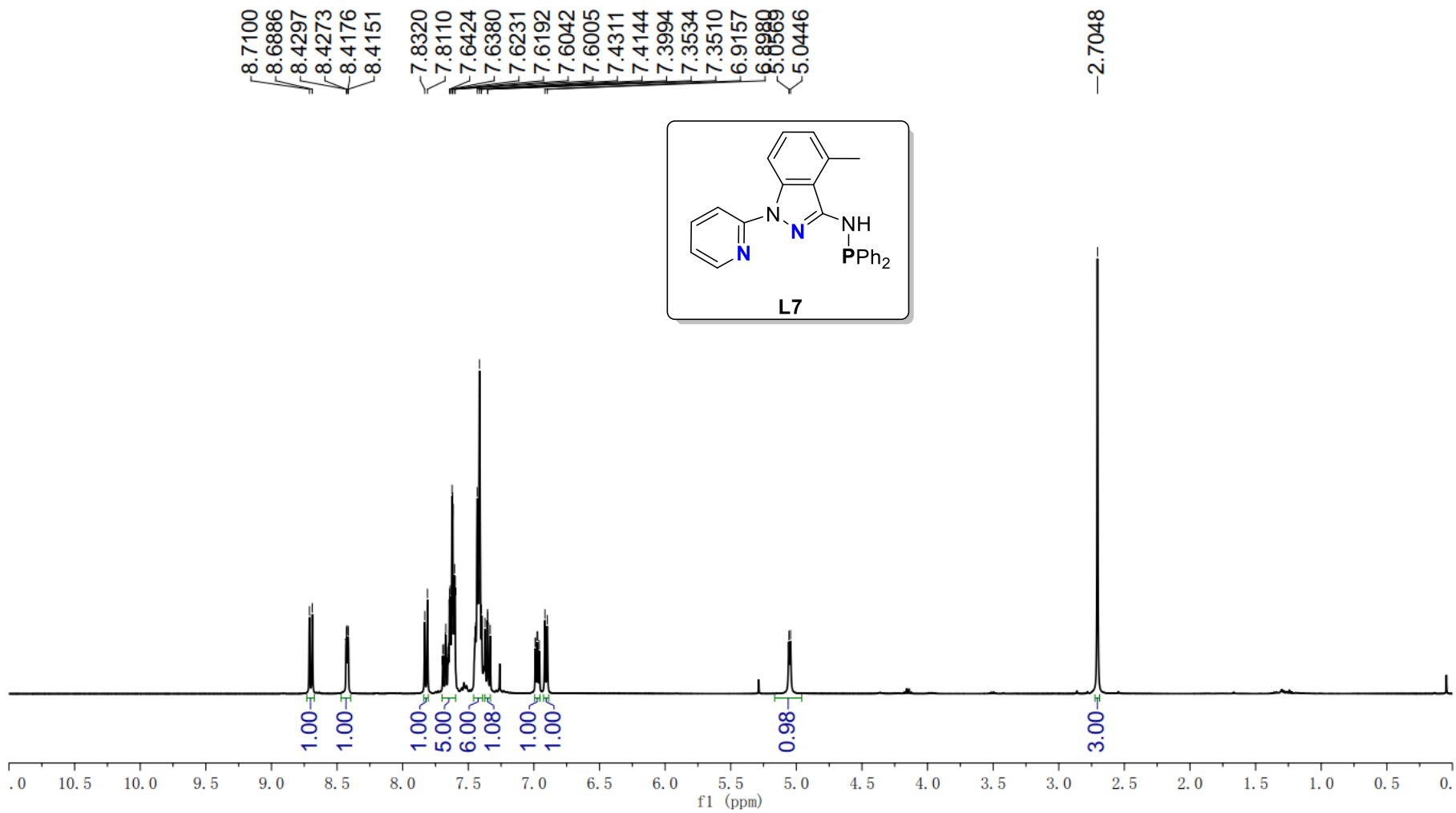
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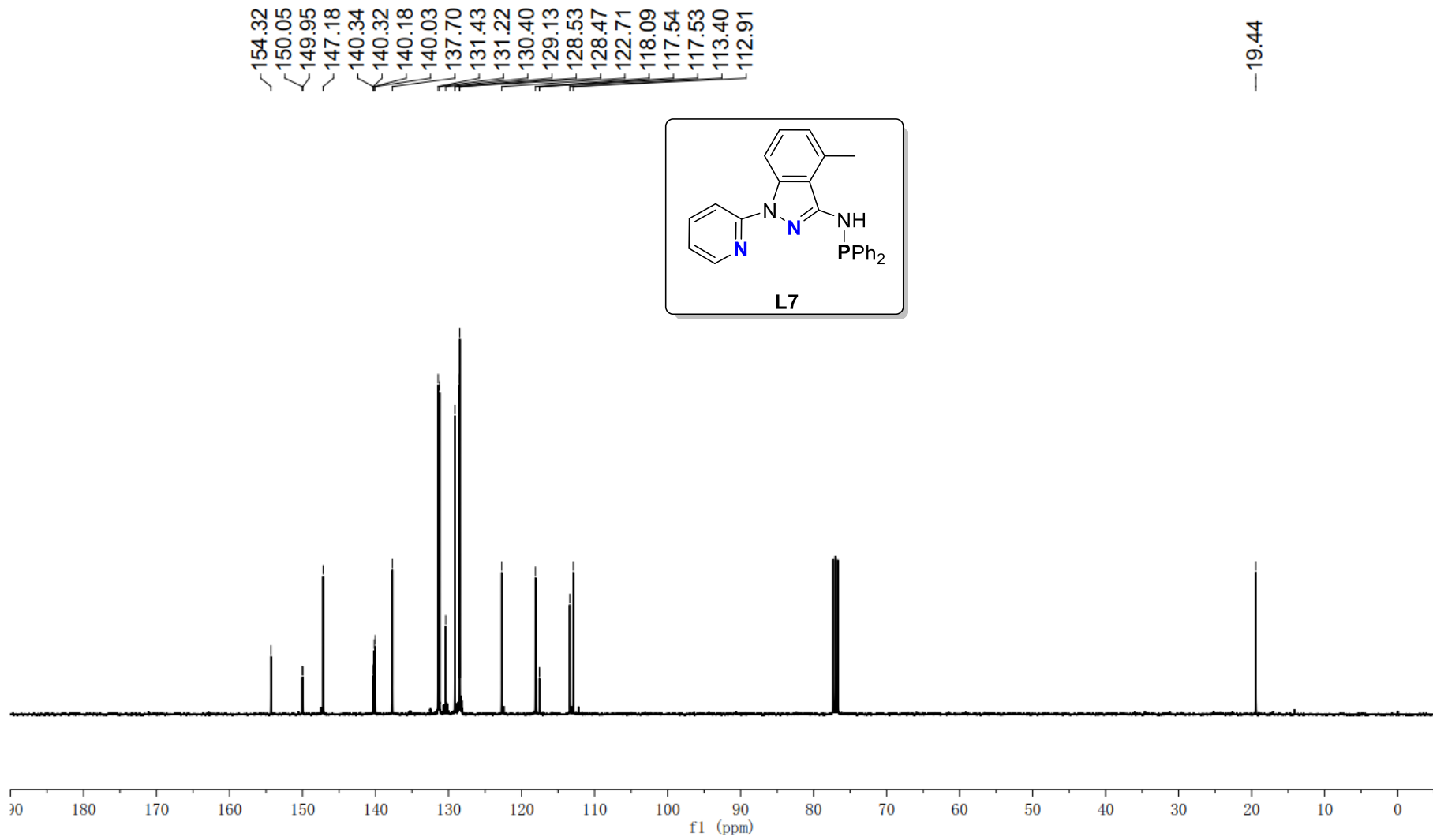


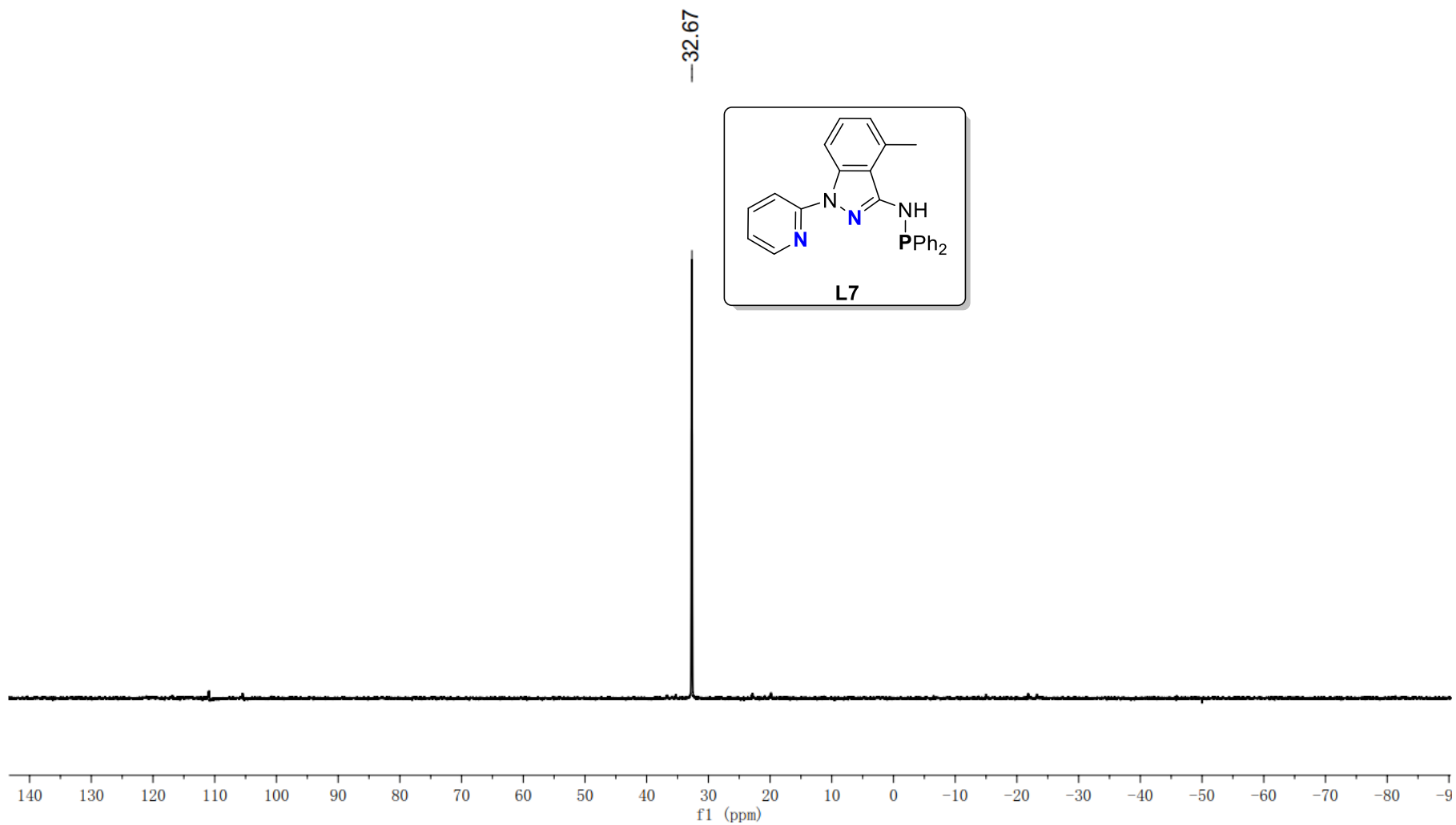


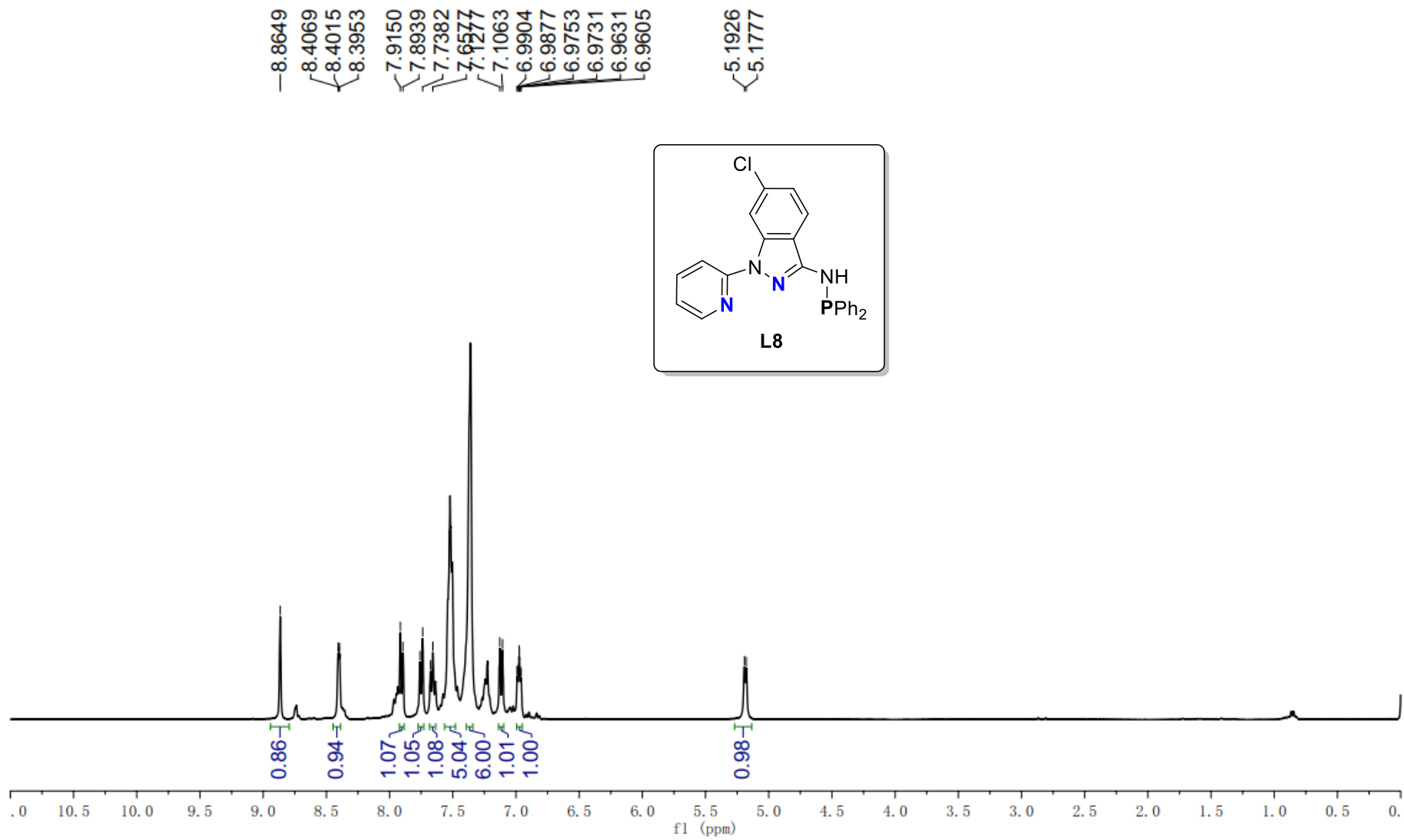




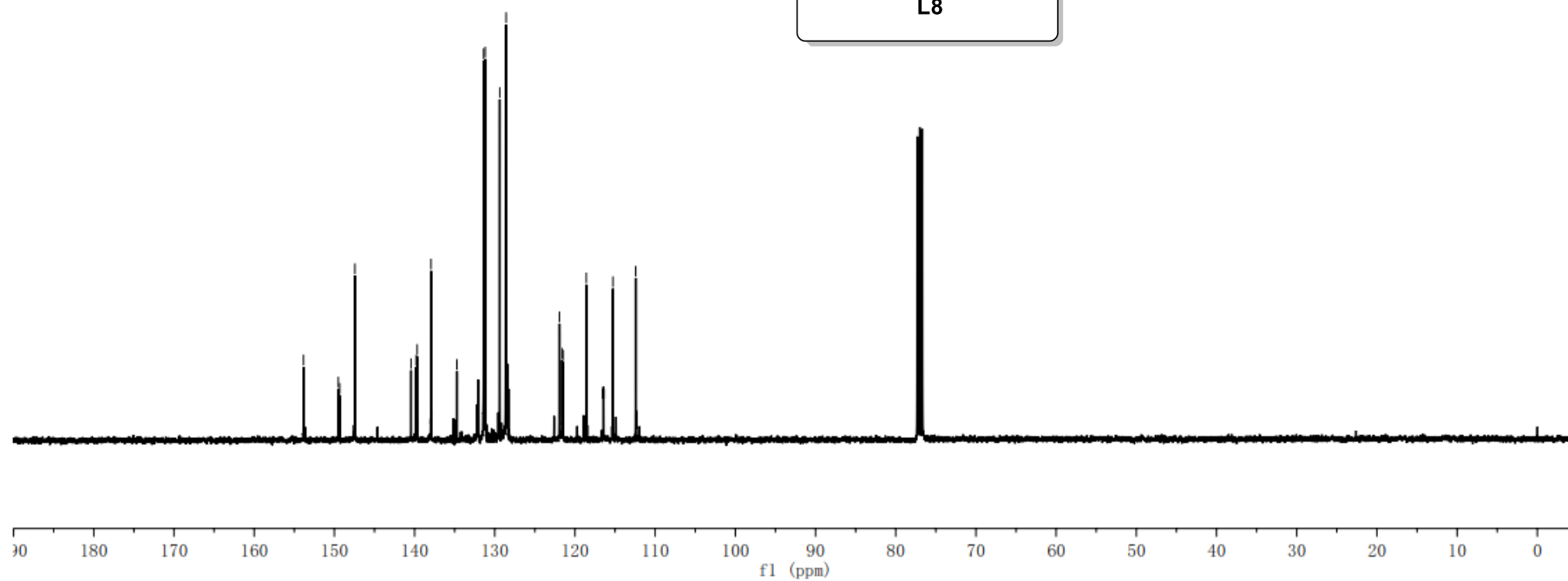
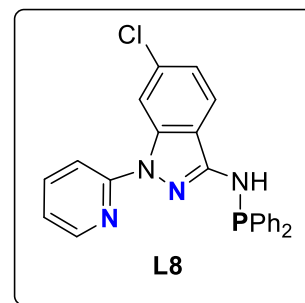


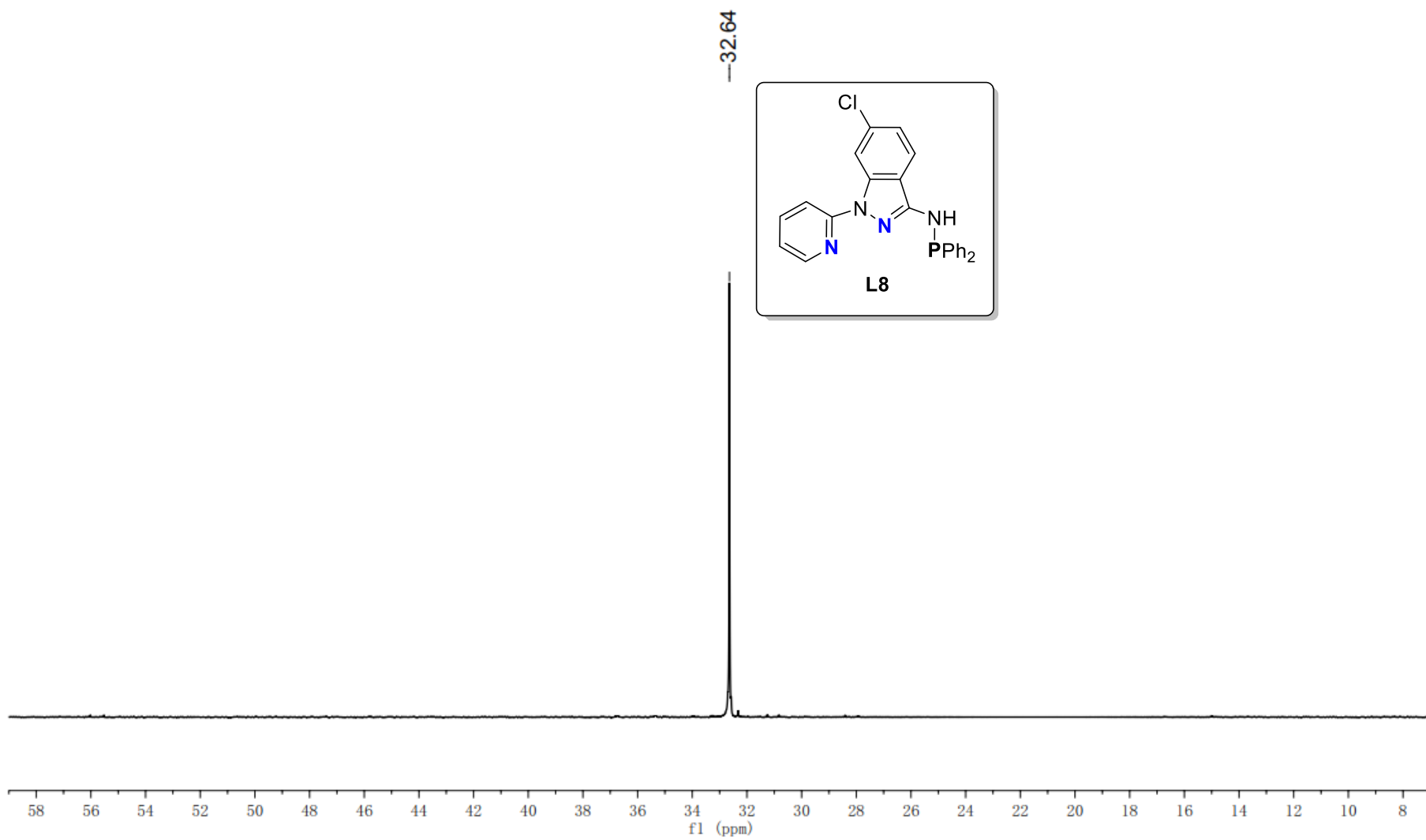


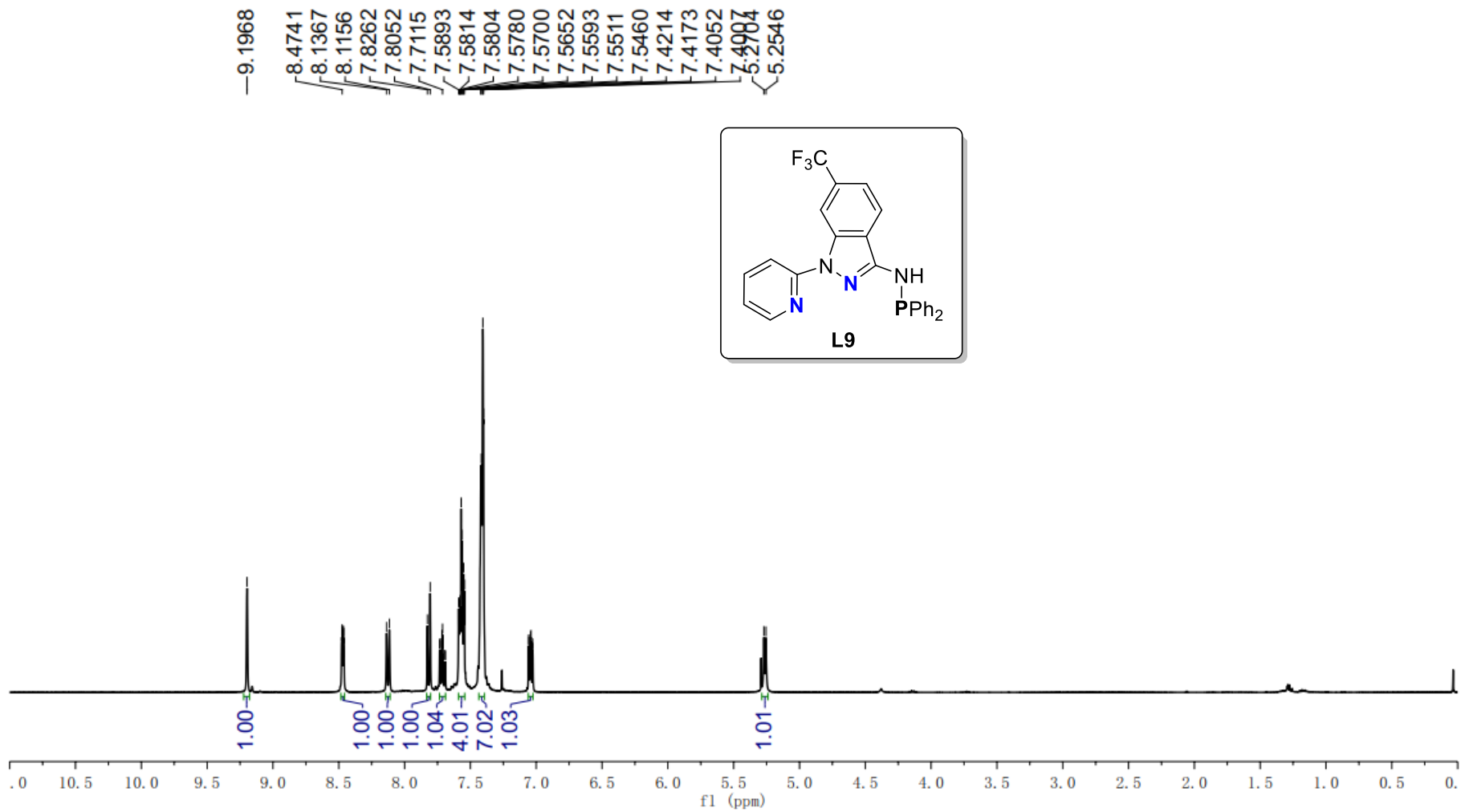


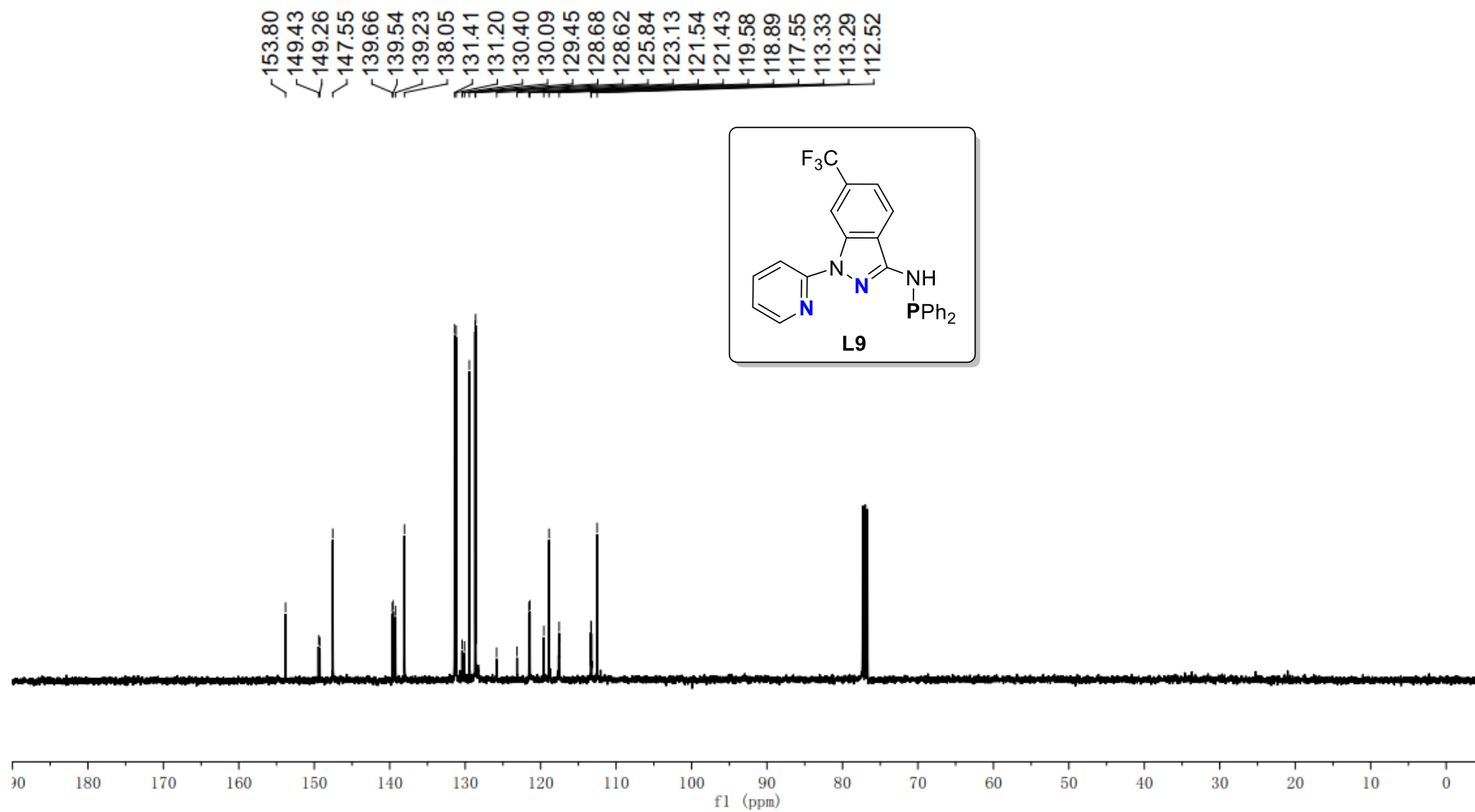


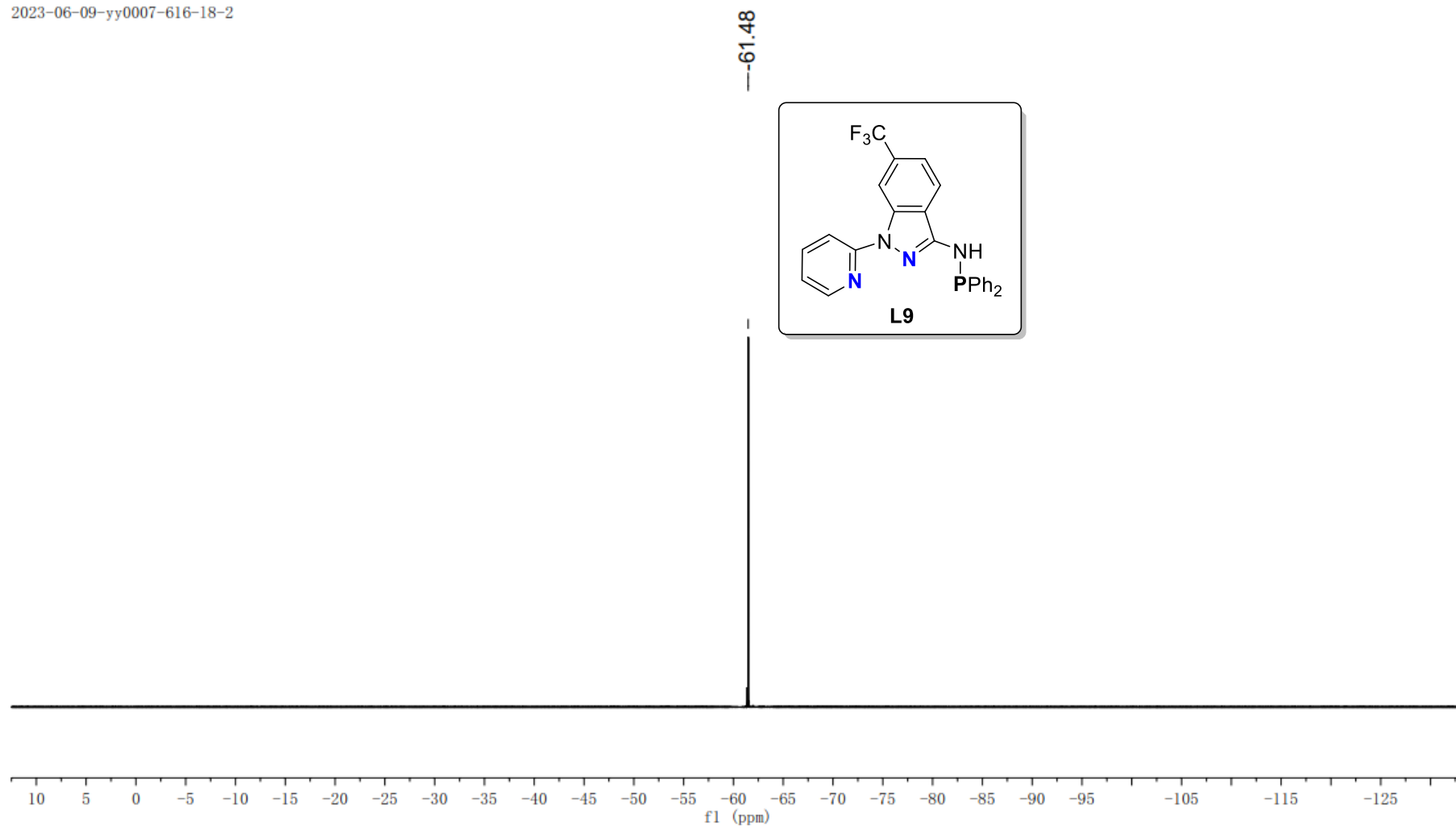
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139.80
139.68
137.95
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131.39
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128.58
121.94
121.60
121.49
118.59
115.26
112.42

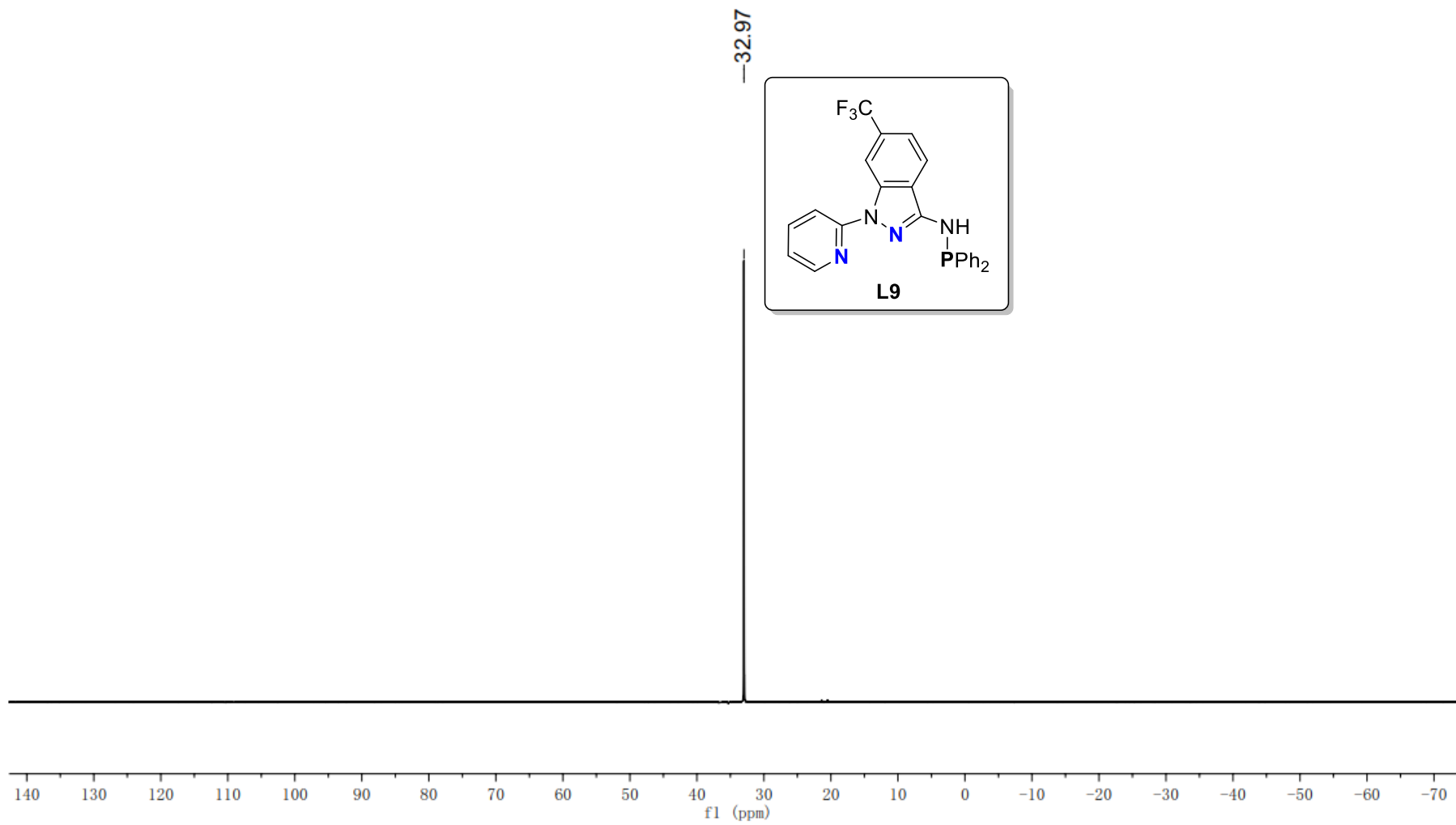


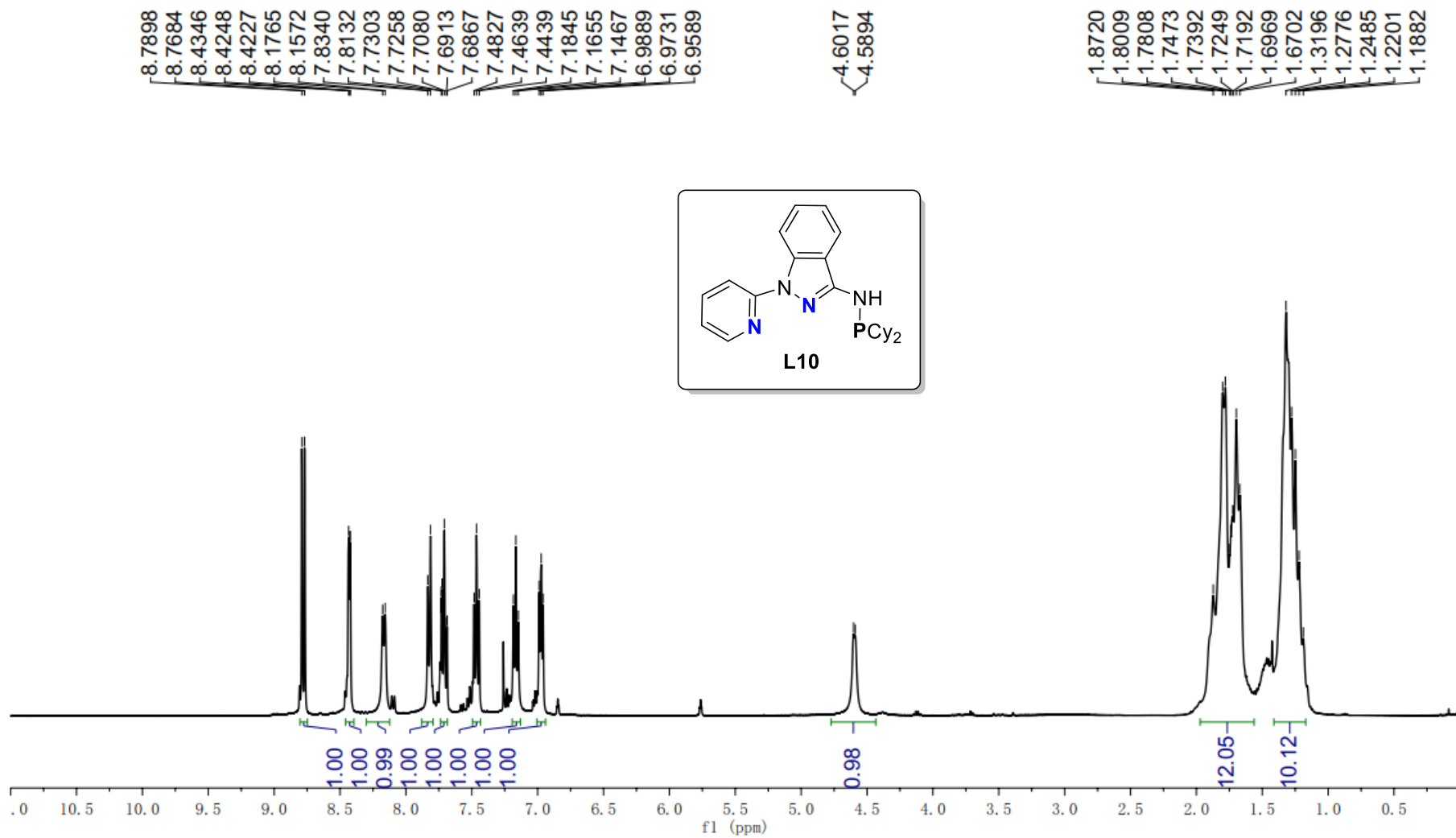


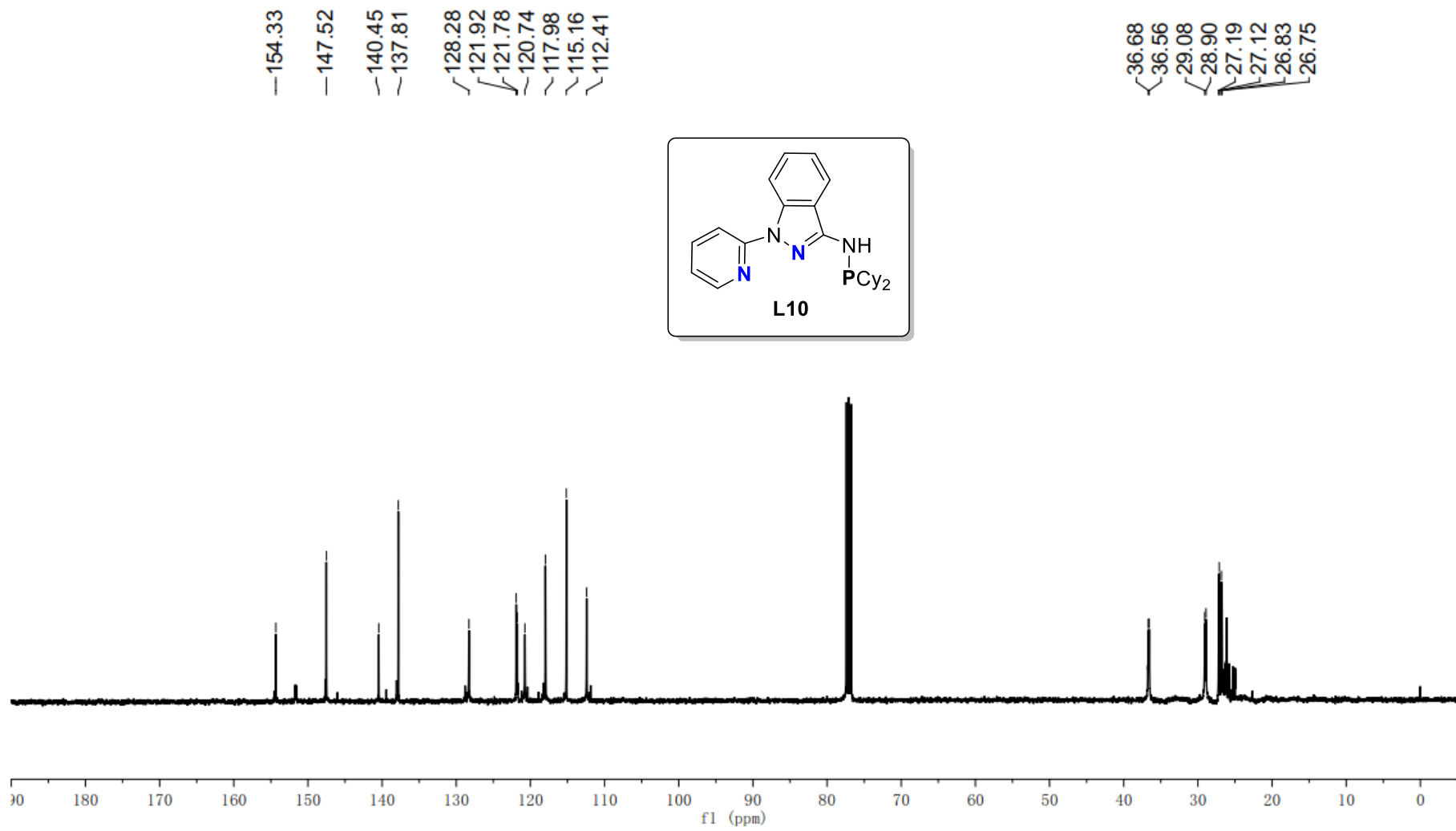


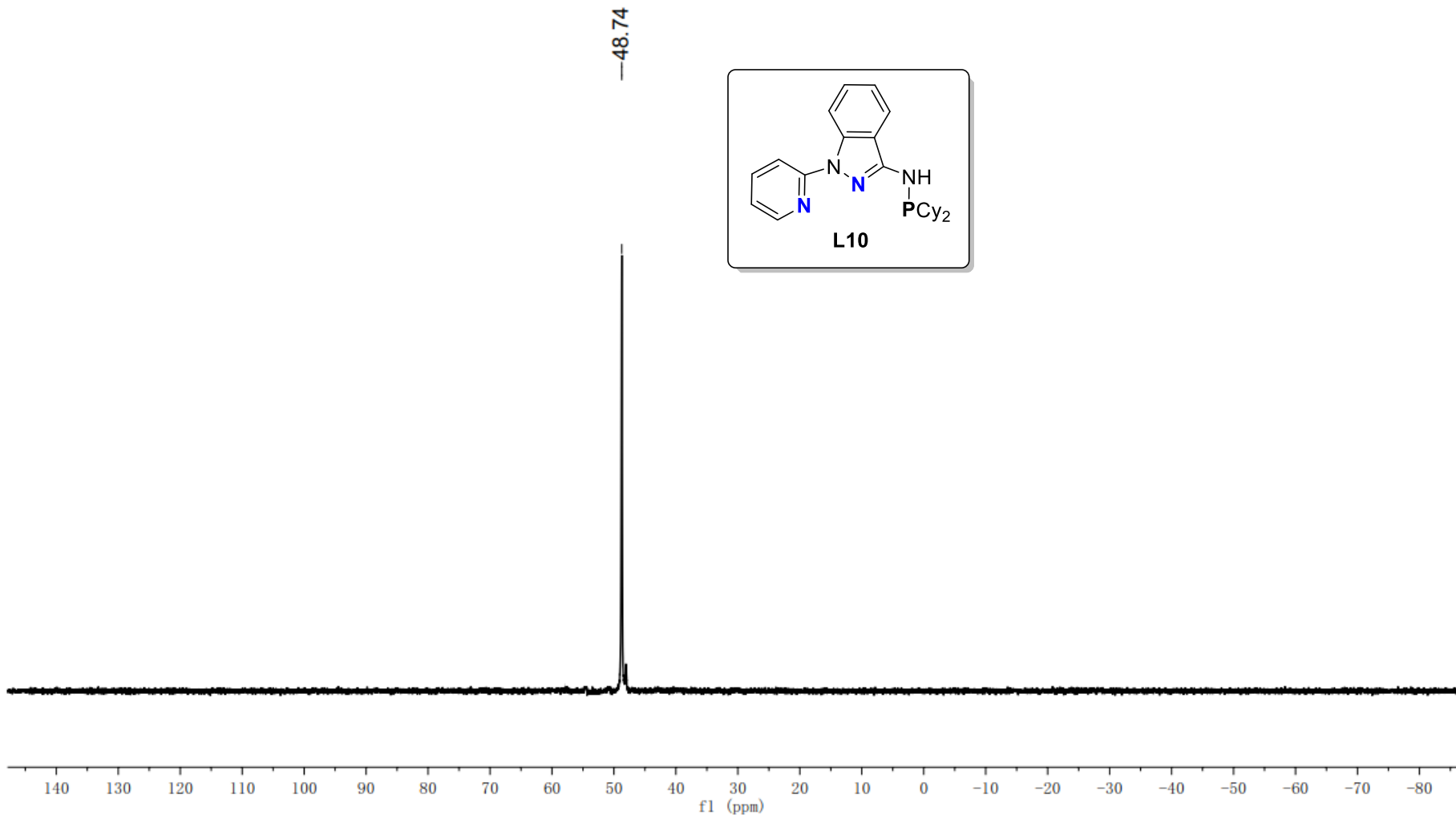


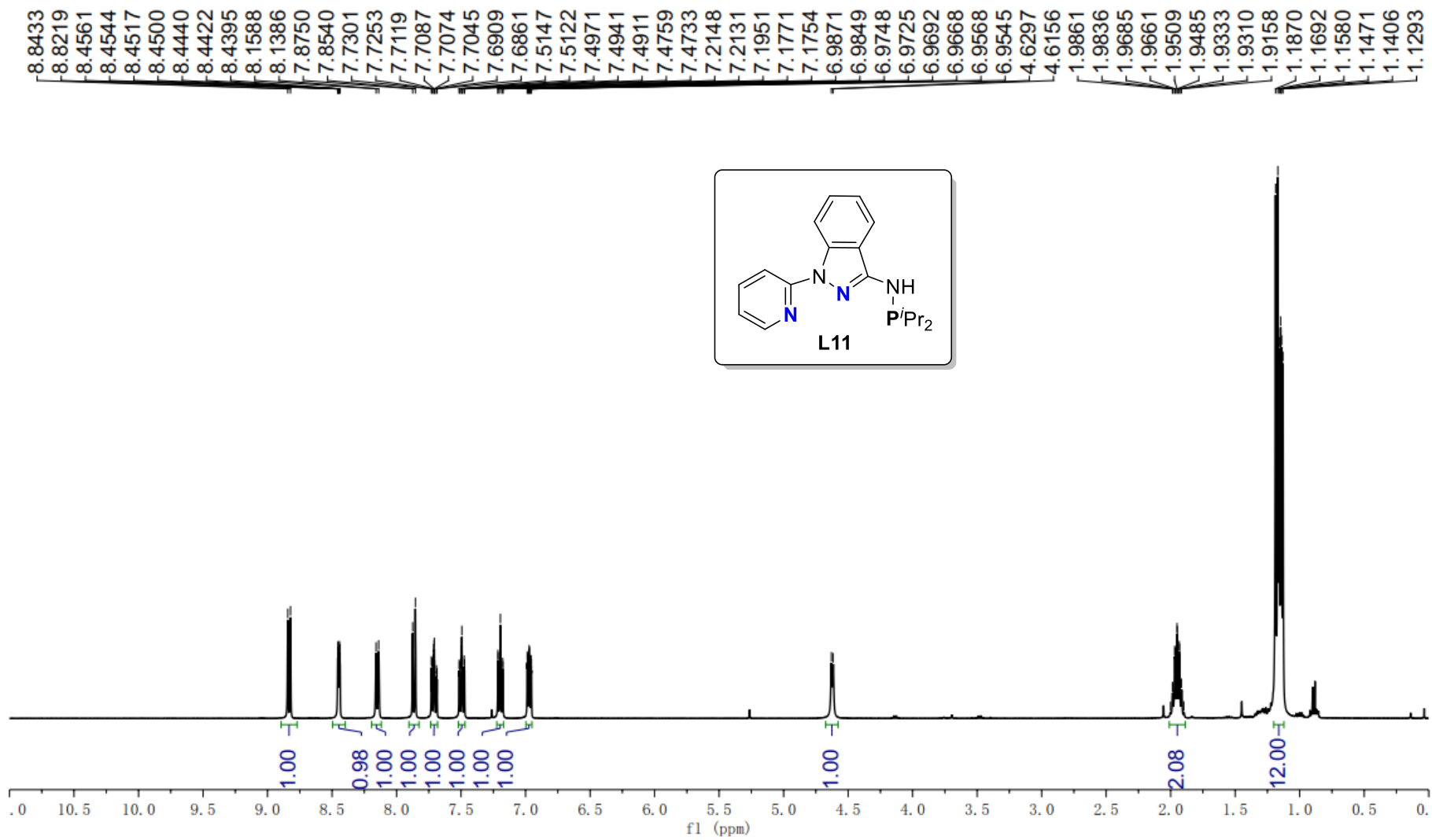






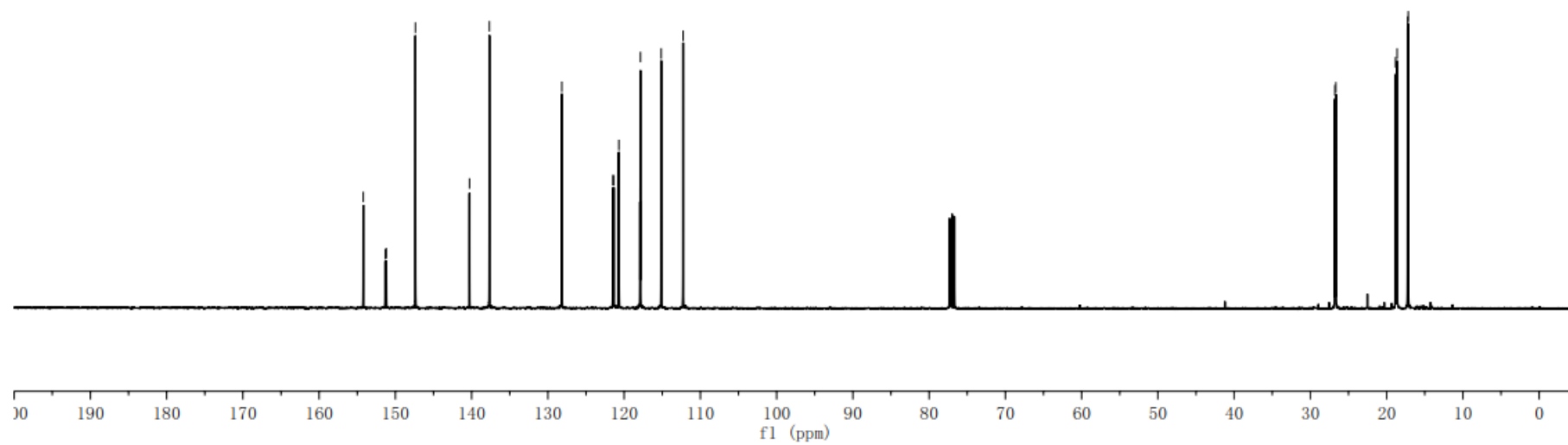
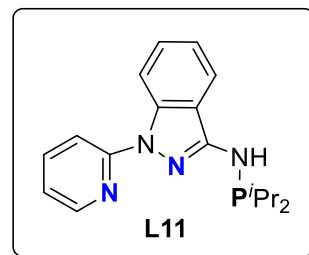


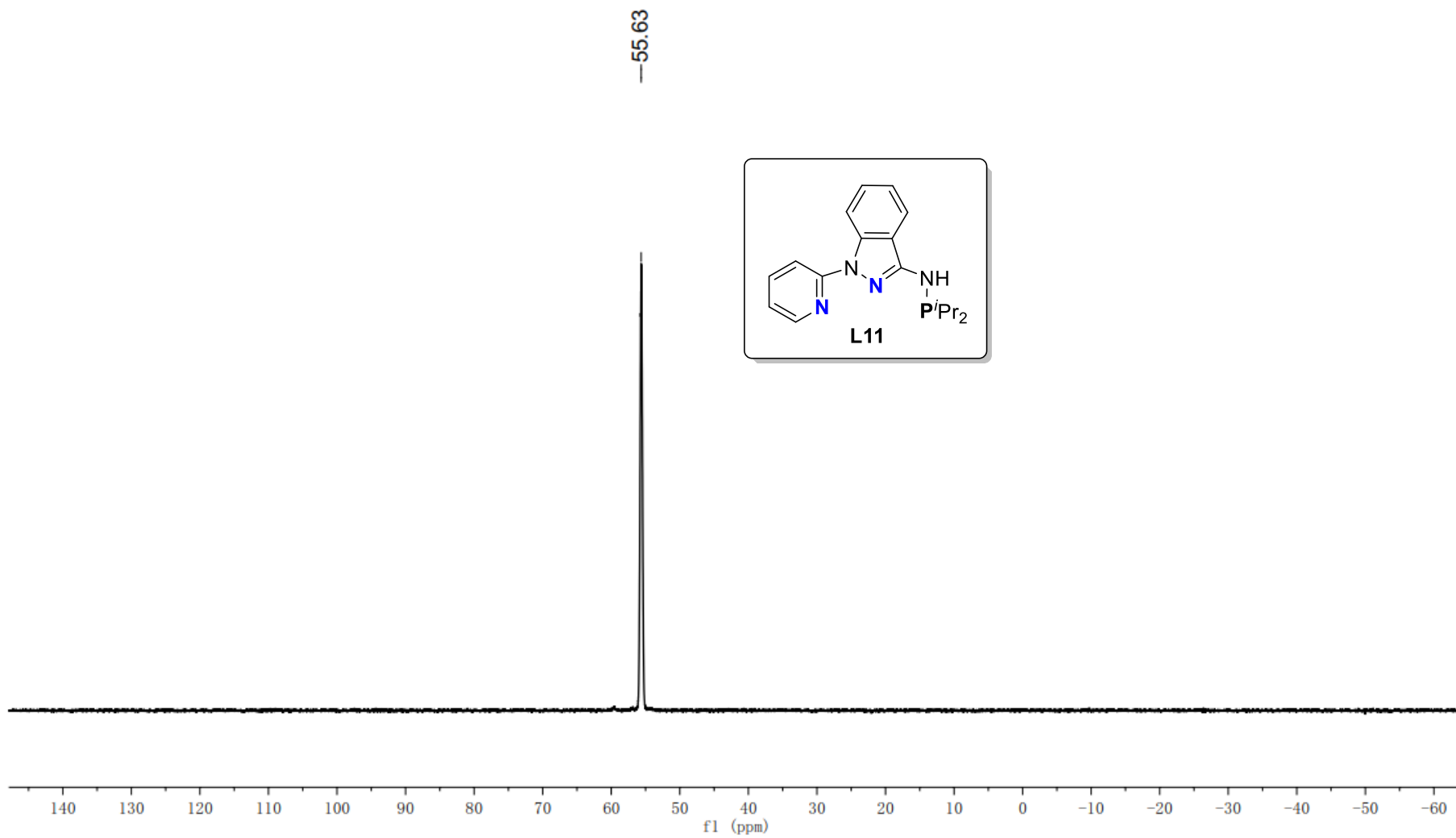




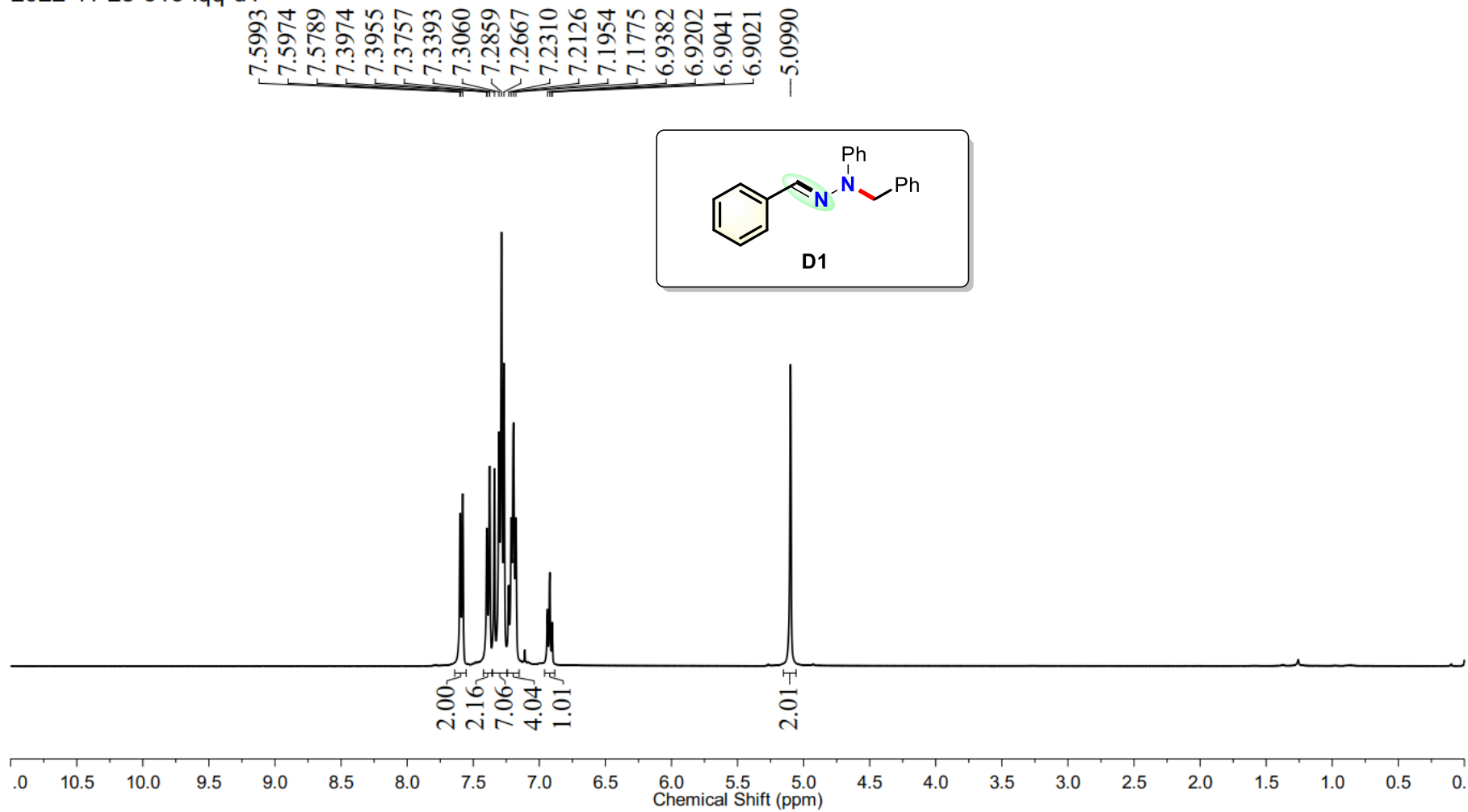
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121.48
121.35
120.70
117.91
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115.13
112.27

26.79
26.66
18.83
18.63
17.25
17.17





2022-11-28-616-lqq-d1

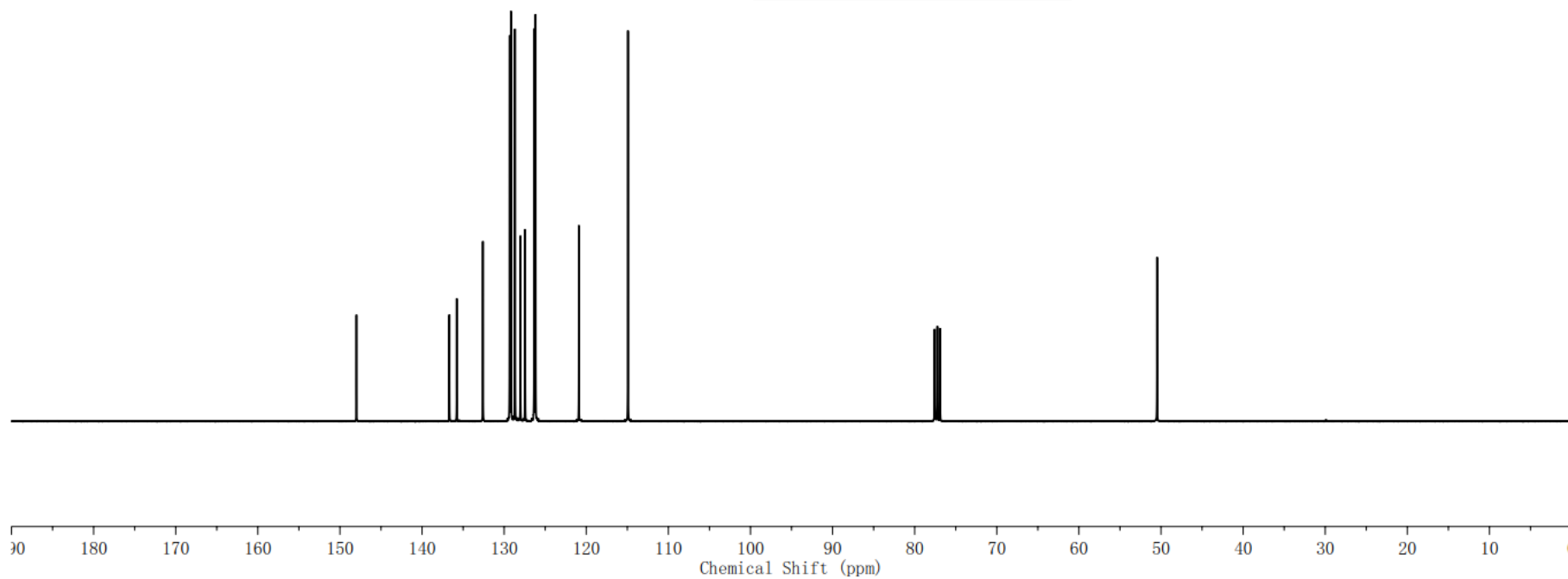
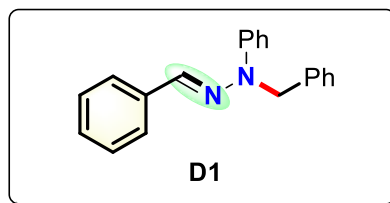


2022-12-02-yy0013-616-lqq-d1

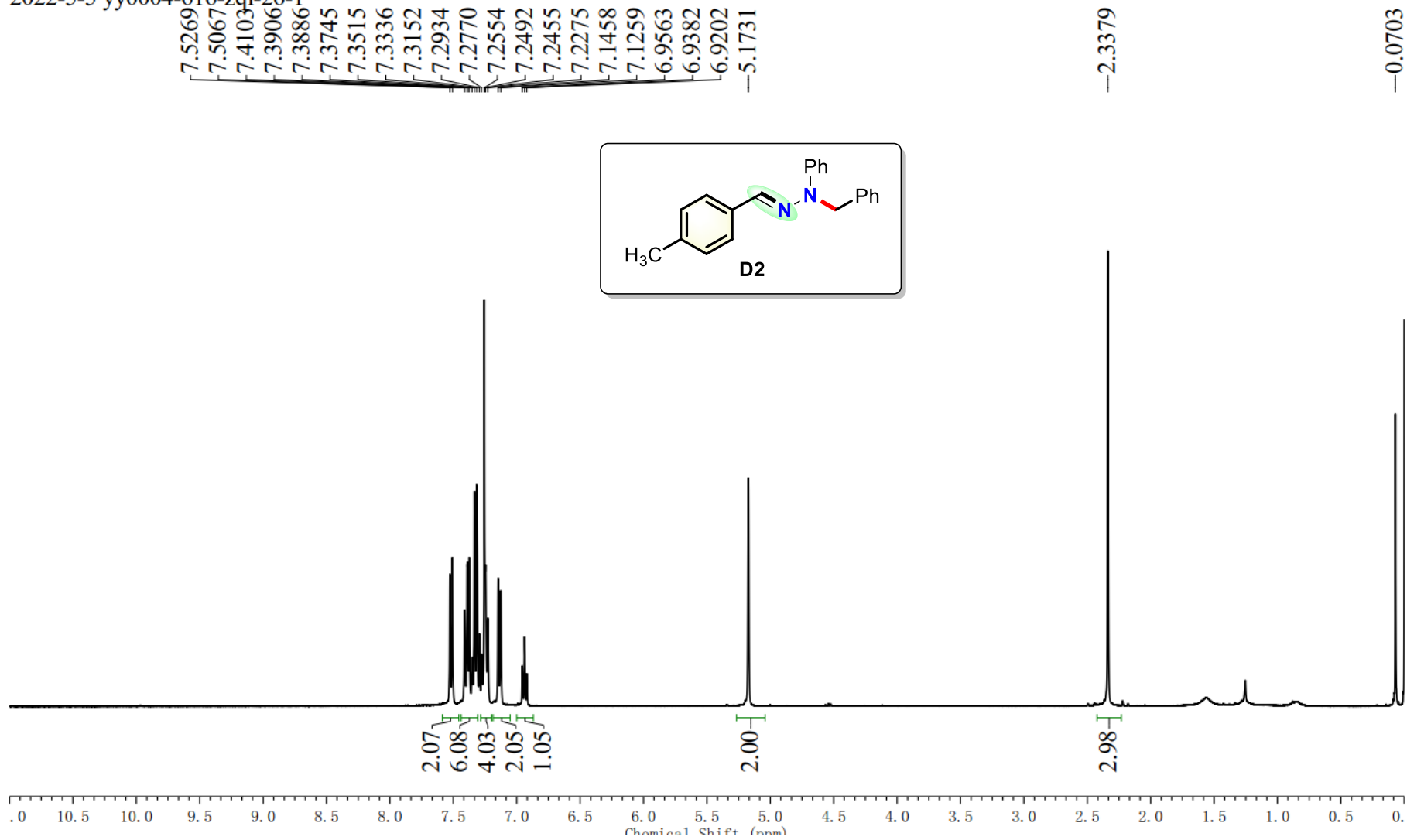
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126.35
126.17
120.90
114.92

77.56
77.24
76.92

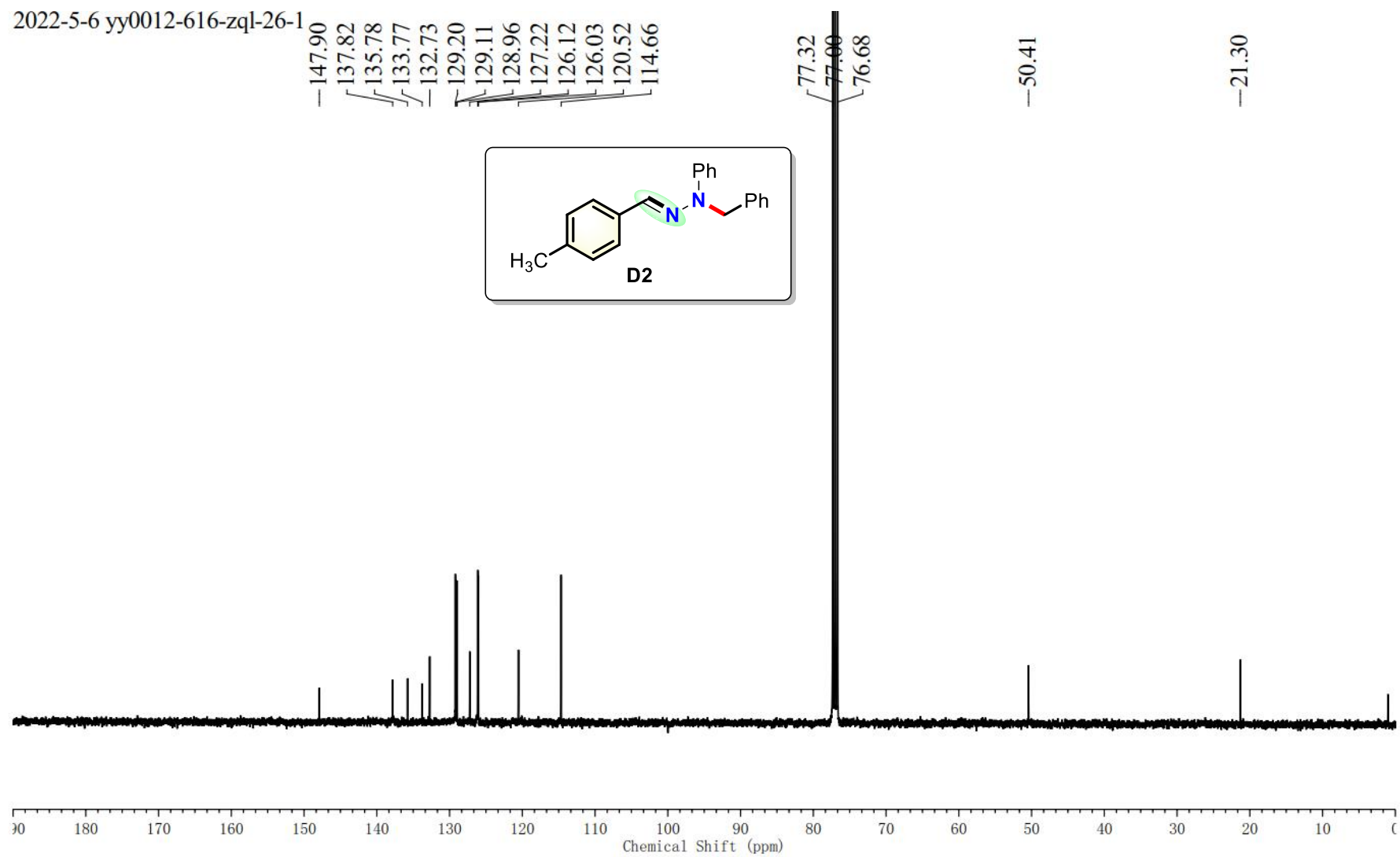
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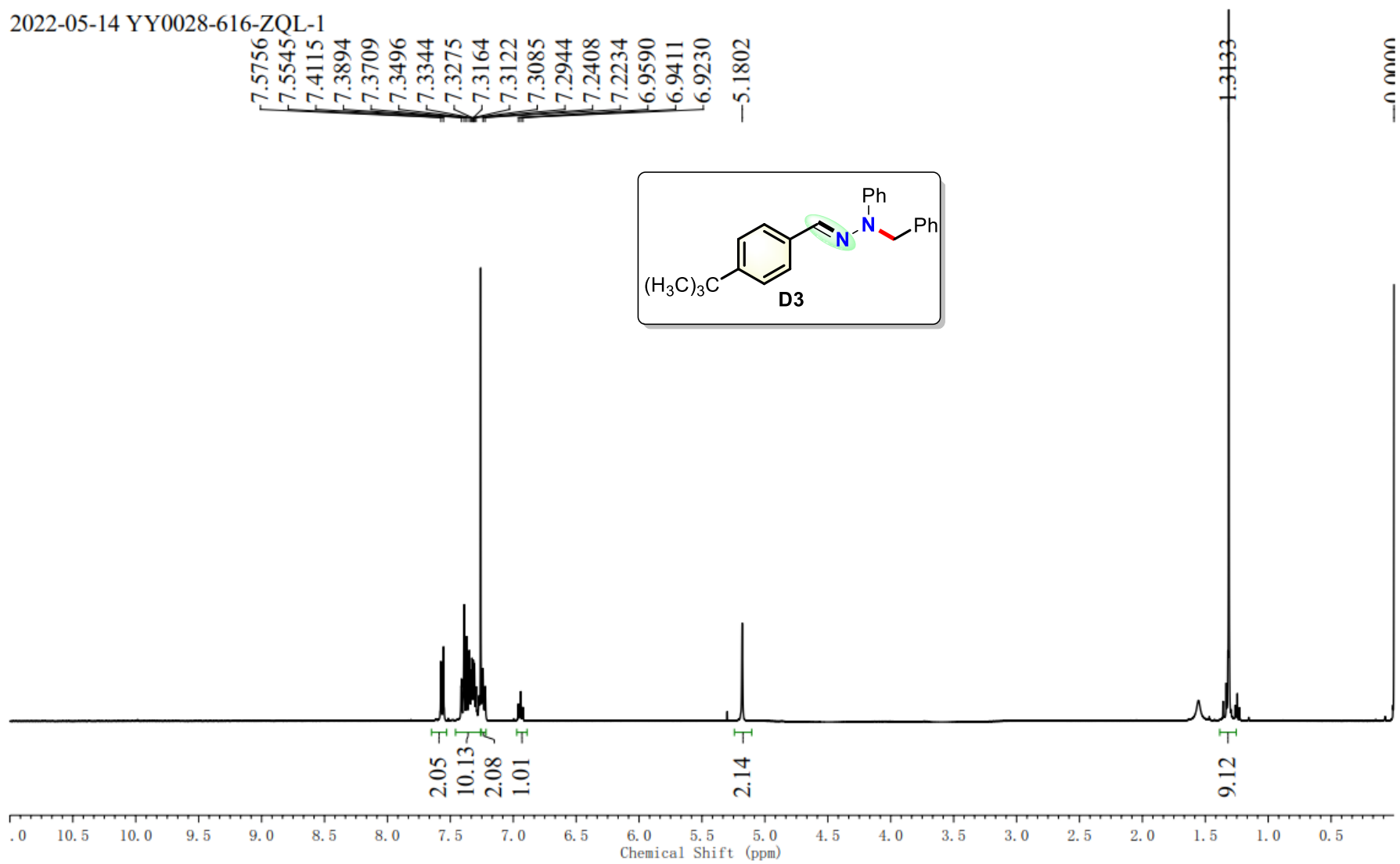
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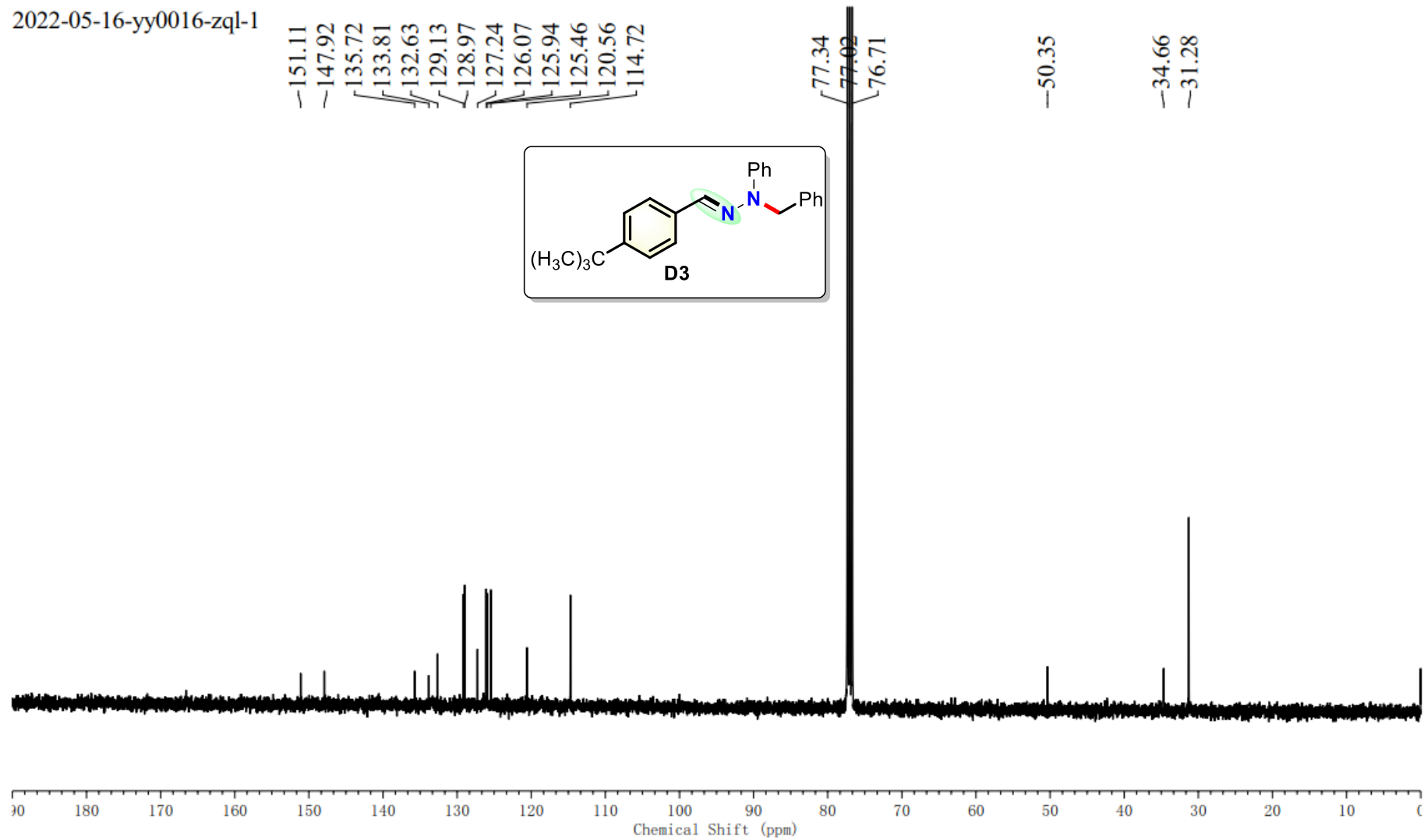
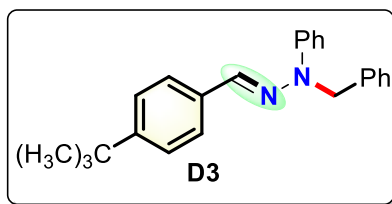


2022-05-14 YY0028-616-ZQL-1



2022-05-16-yy0016-zql-1

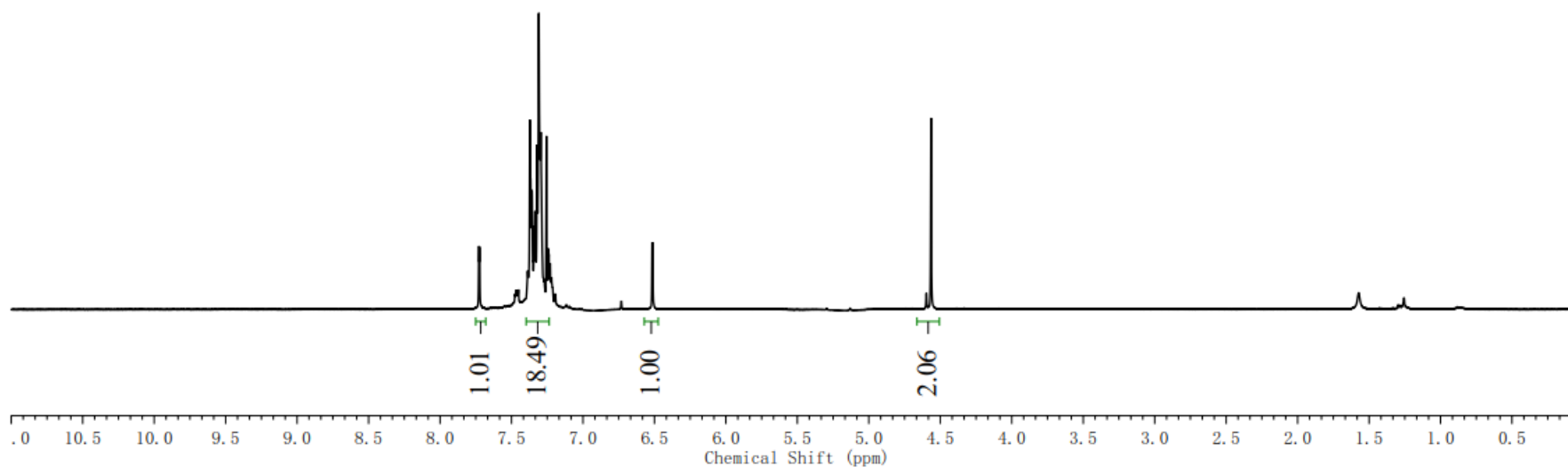
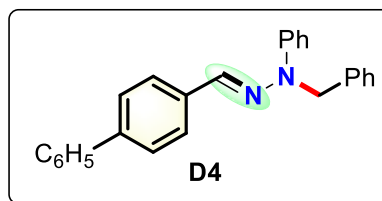
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128.97
127.24
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114.72
77.34
77.02
76.71
50.35
34.66
31.28



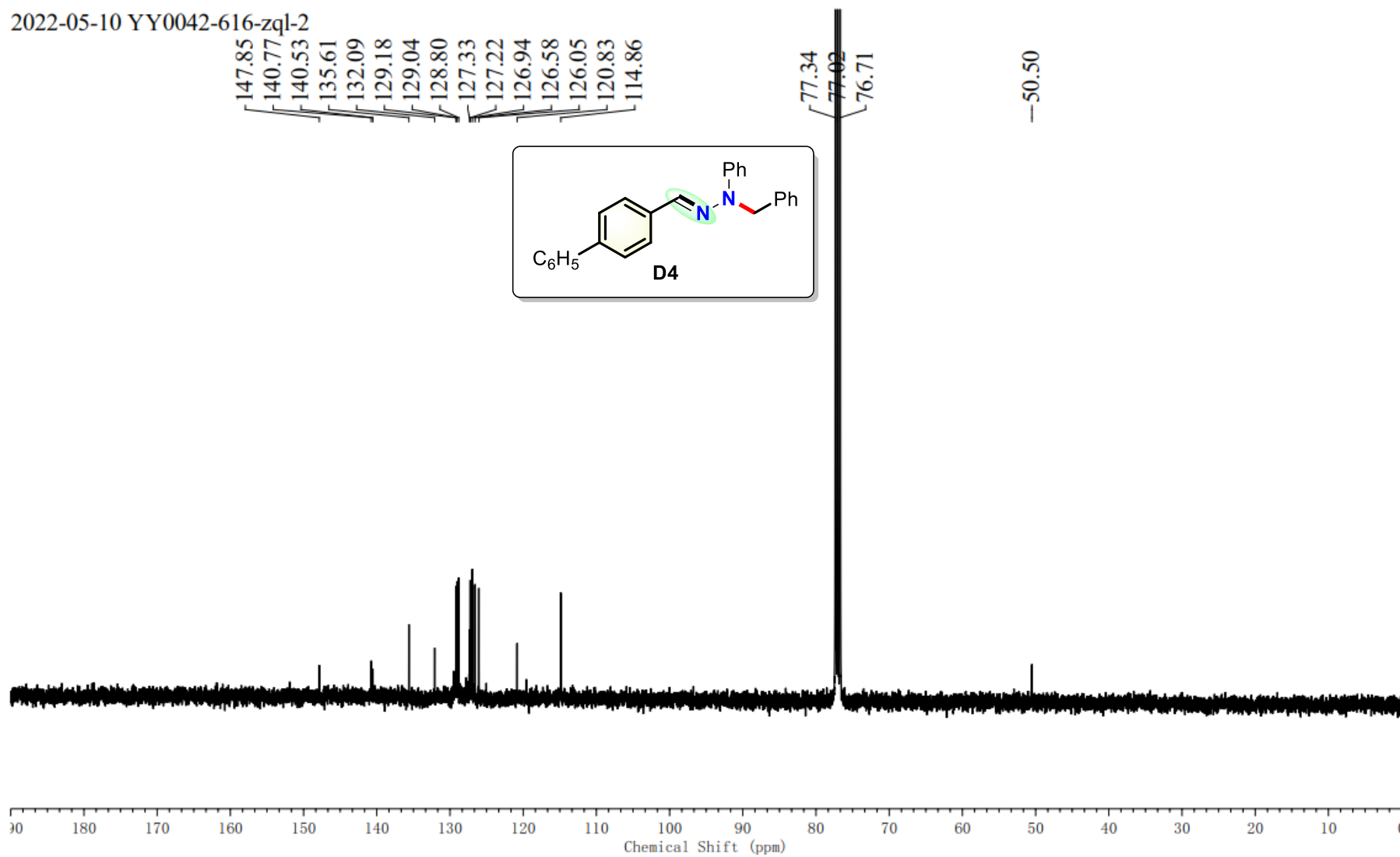
2022-5-7 yy0007-616-zql-1

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7.3206
7.3092
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7.2558
7.2443
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7.2293
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6.5115

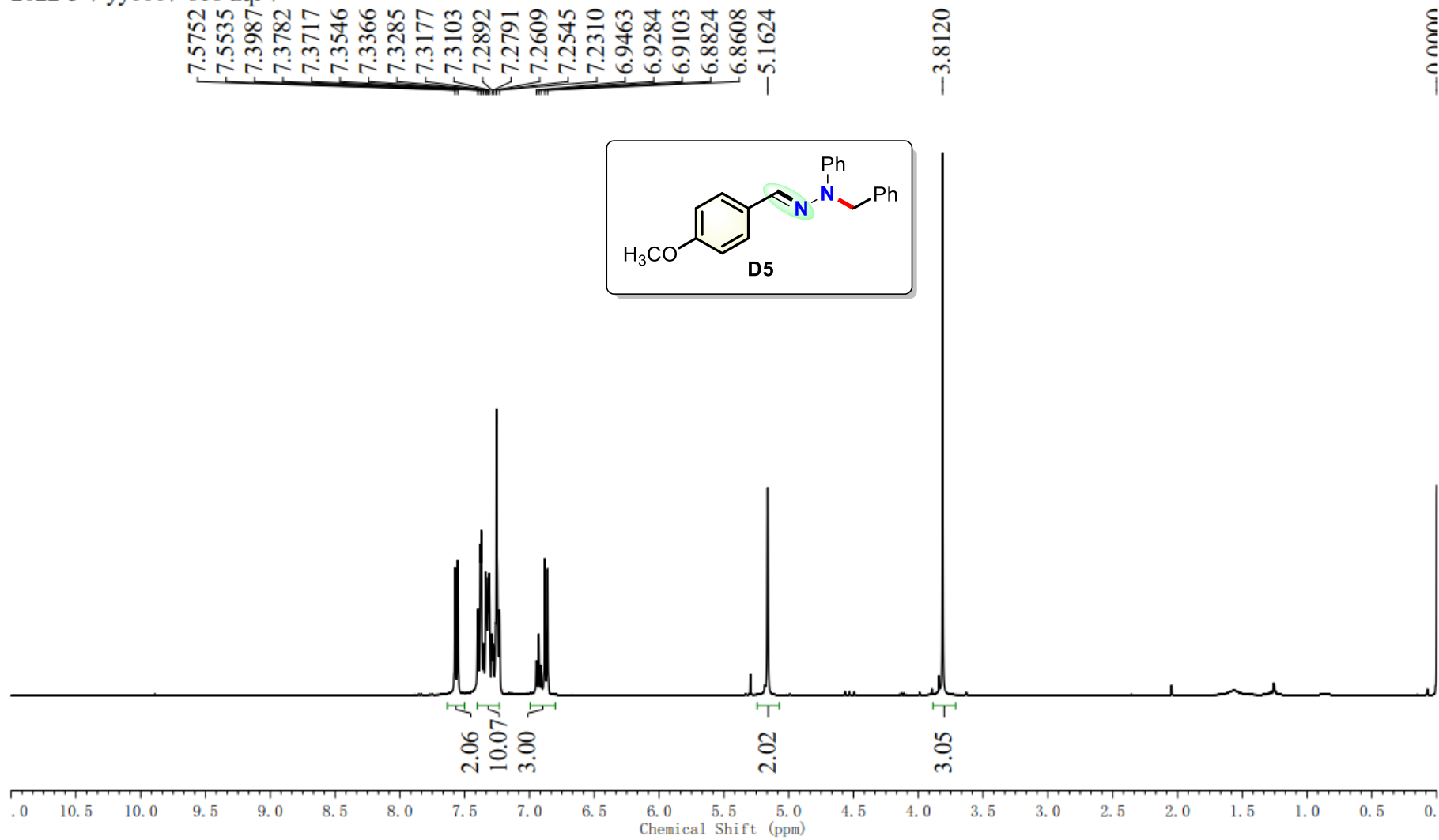
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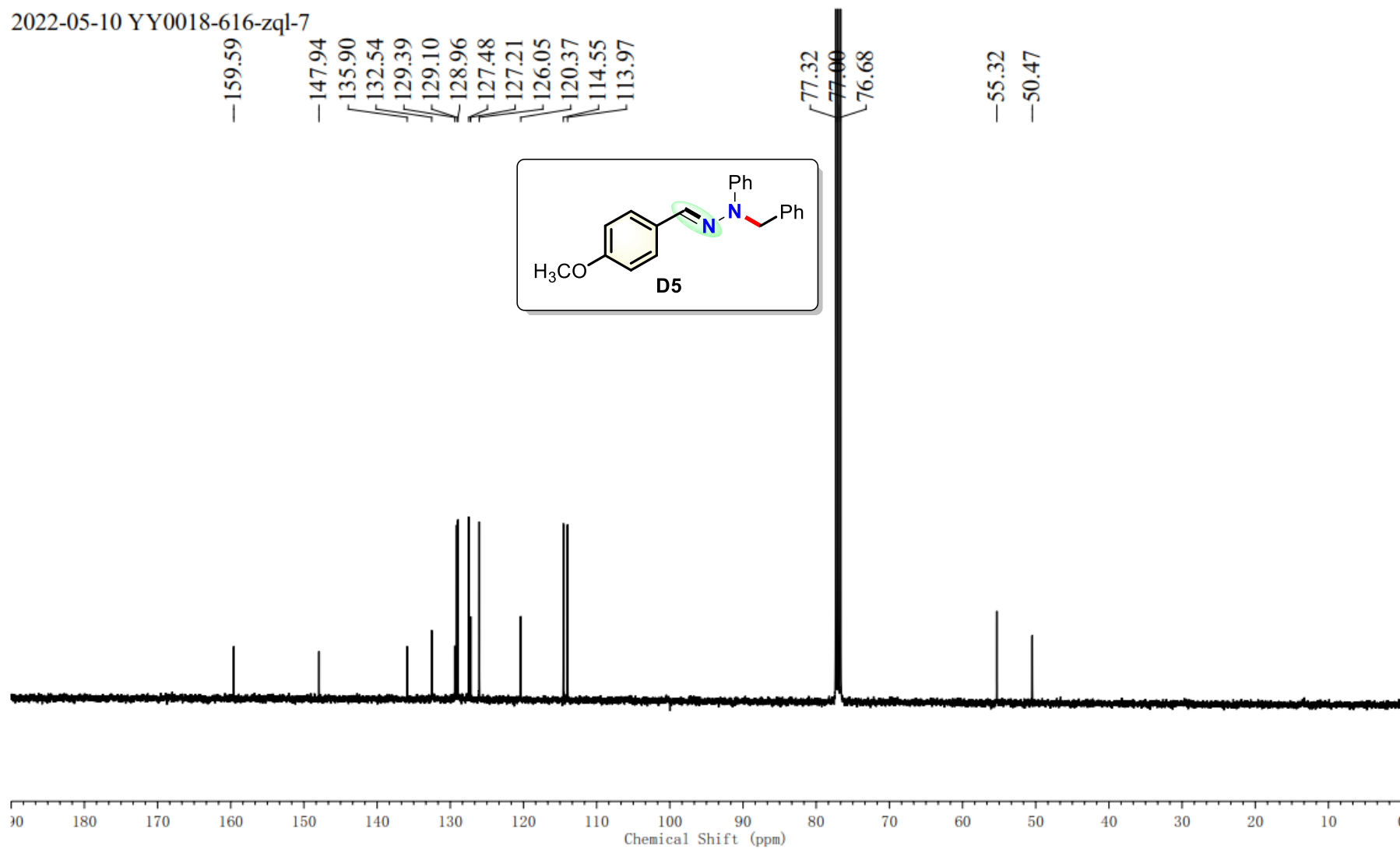
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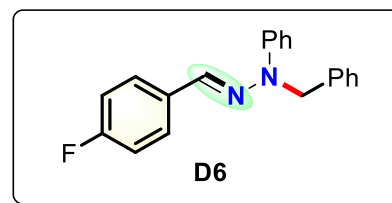
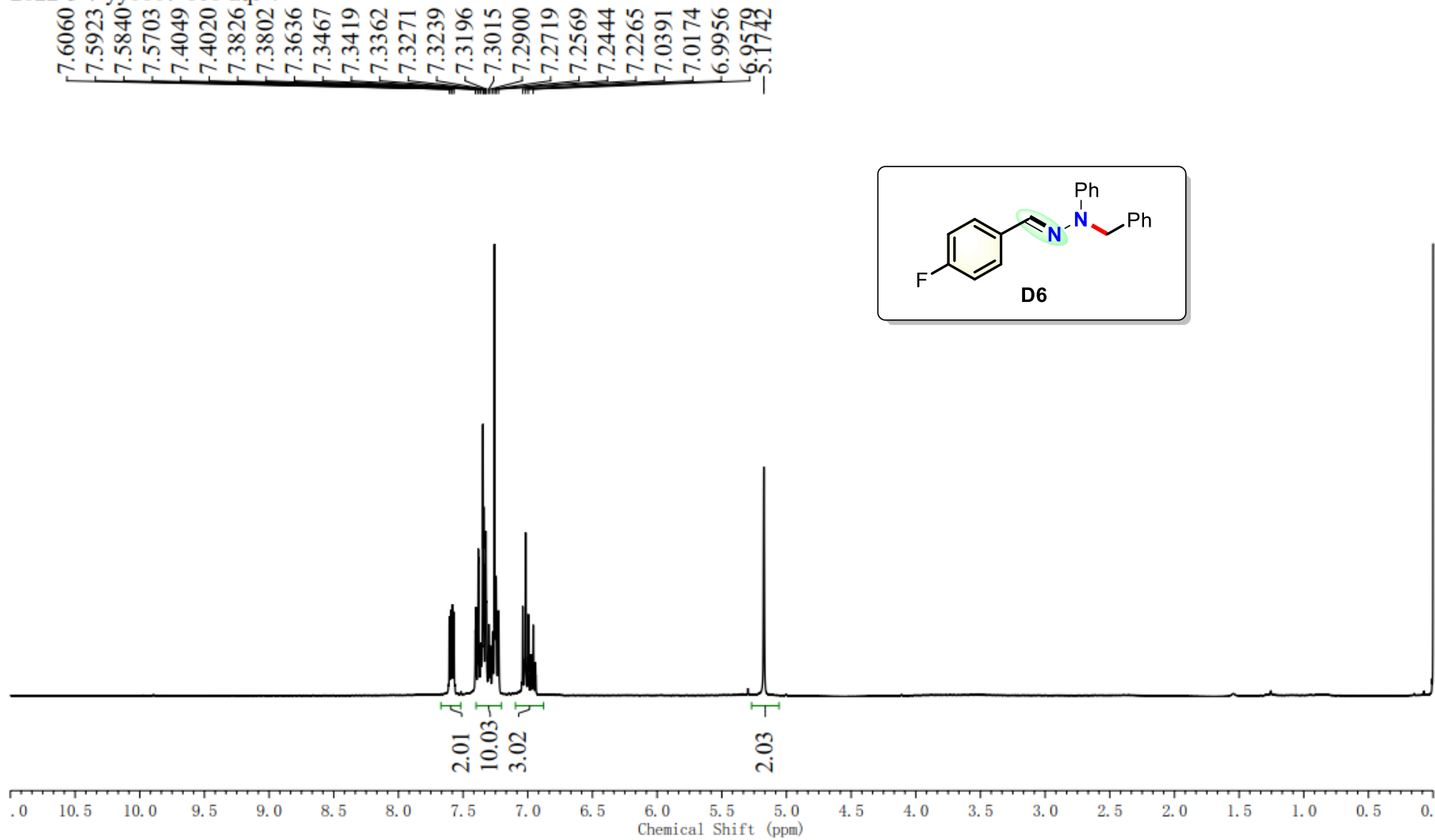
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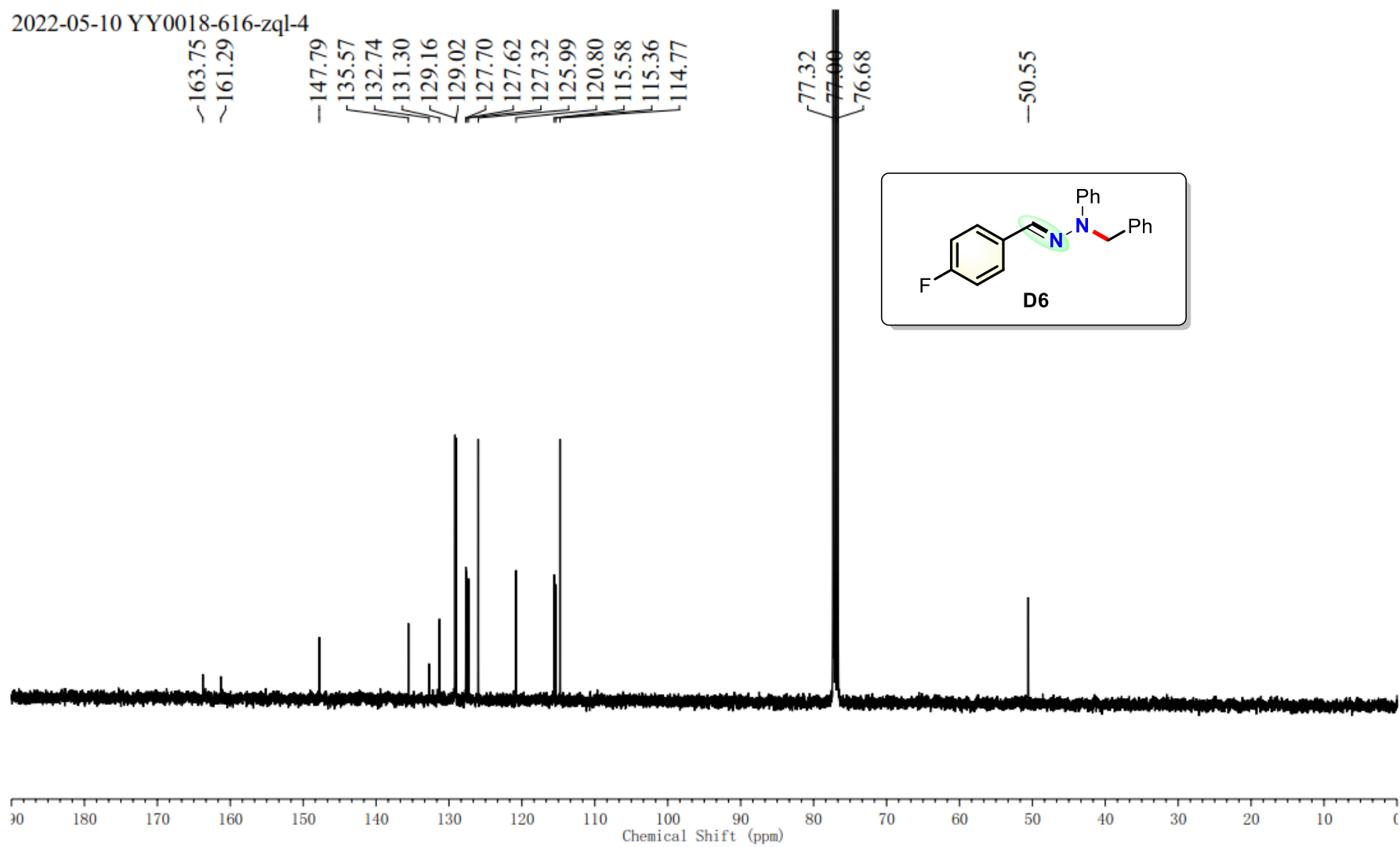
2022-05-10 YY0018-616-zql-7



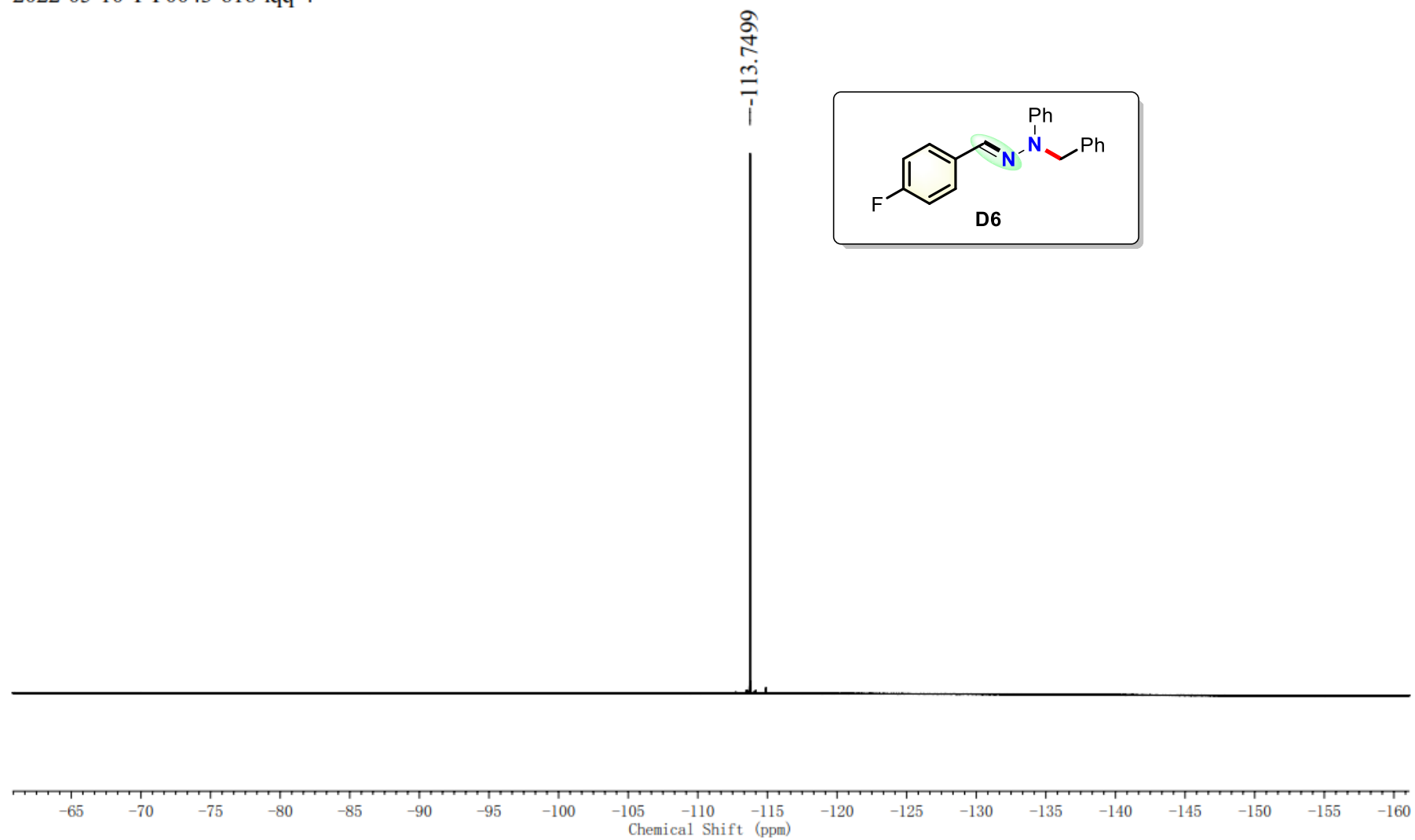
2022-5-7 yy0007-616-zql-4



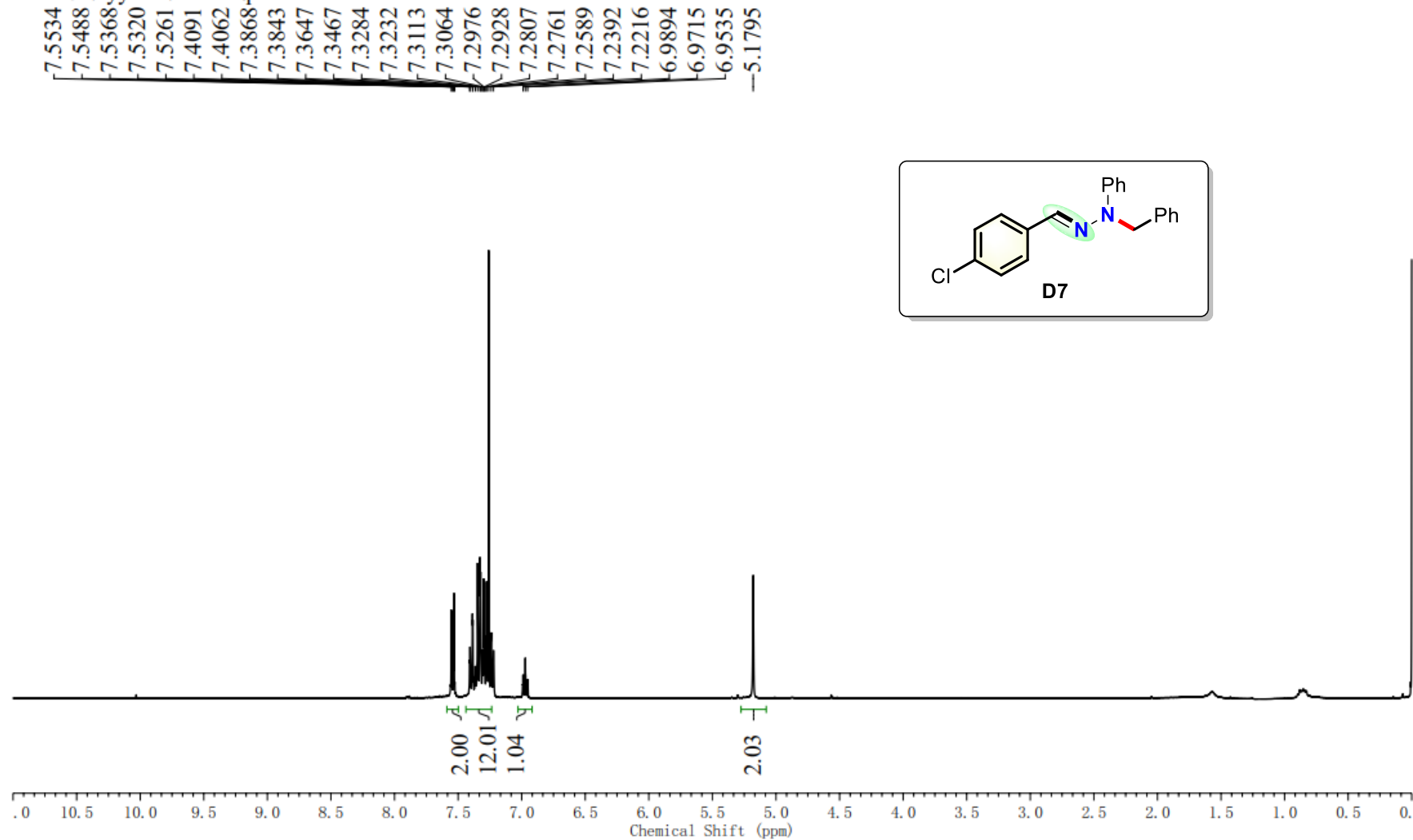
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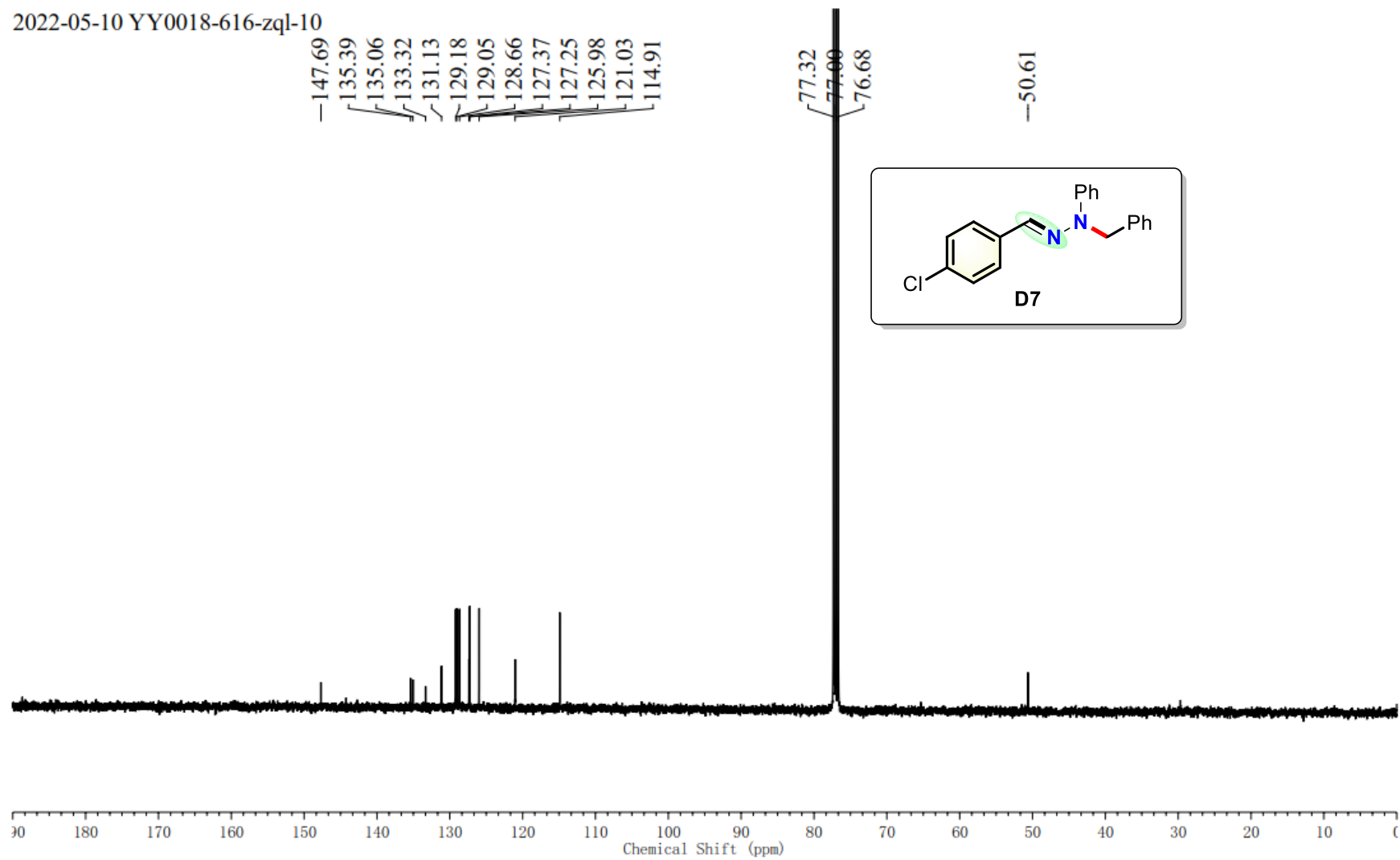
2022-05-10 YY0043-616-lqq-4



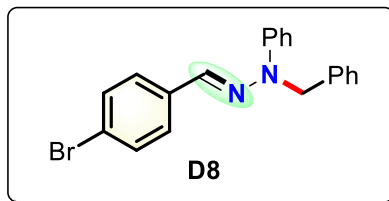
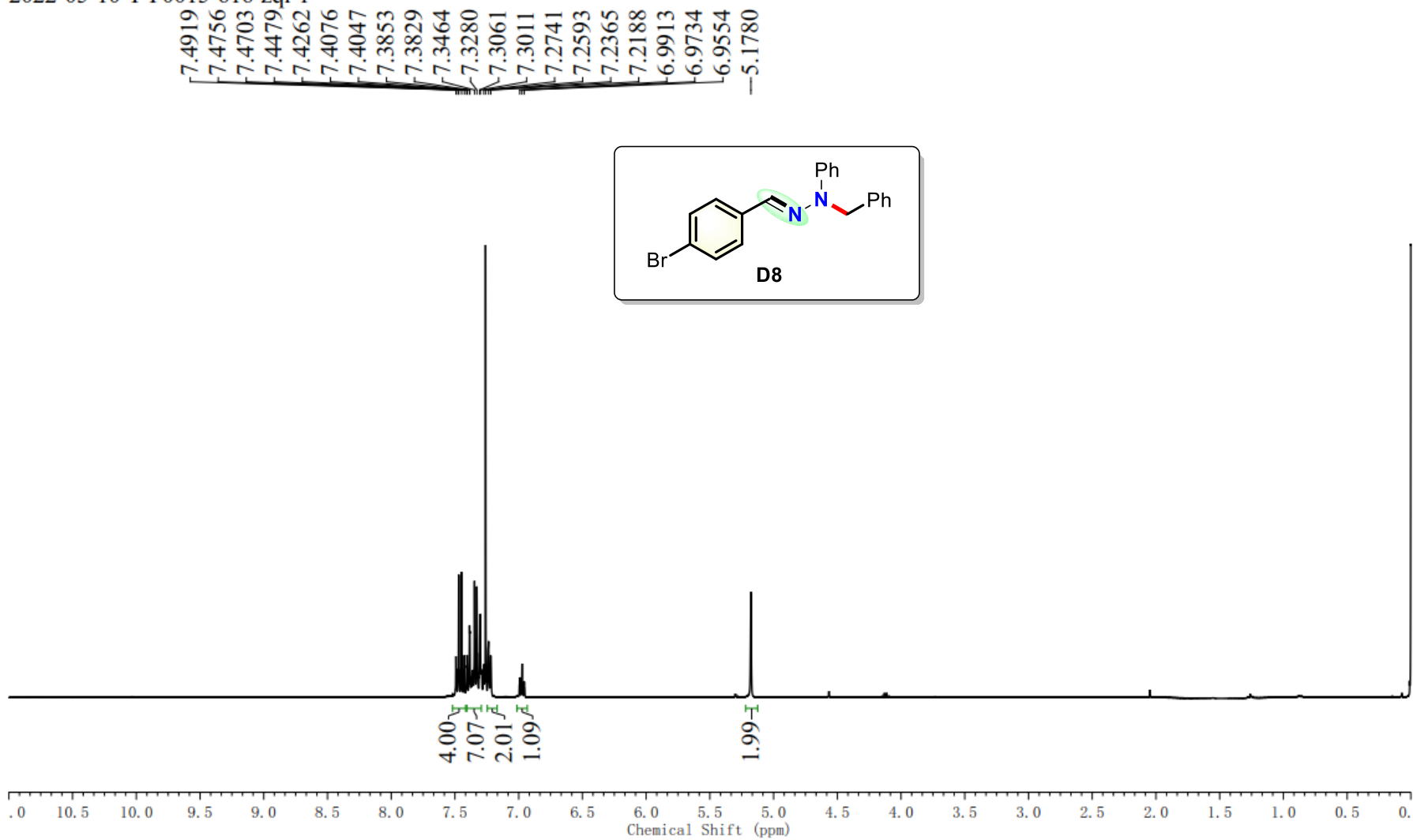
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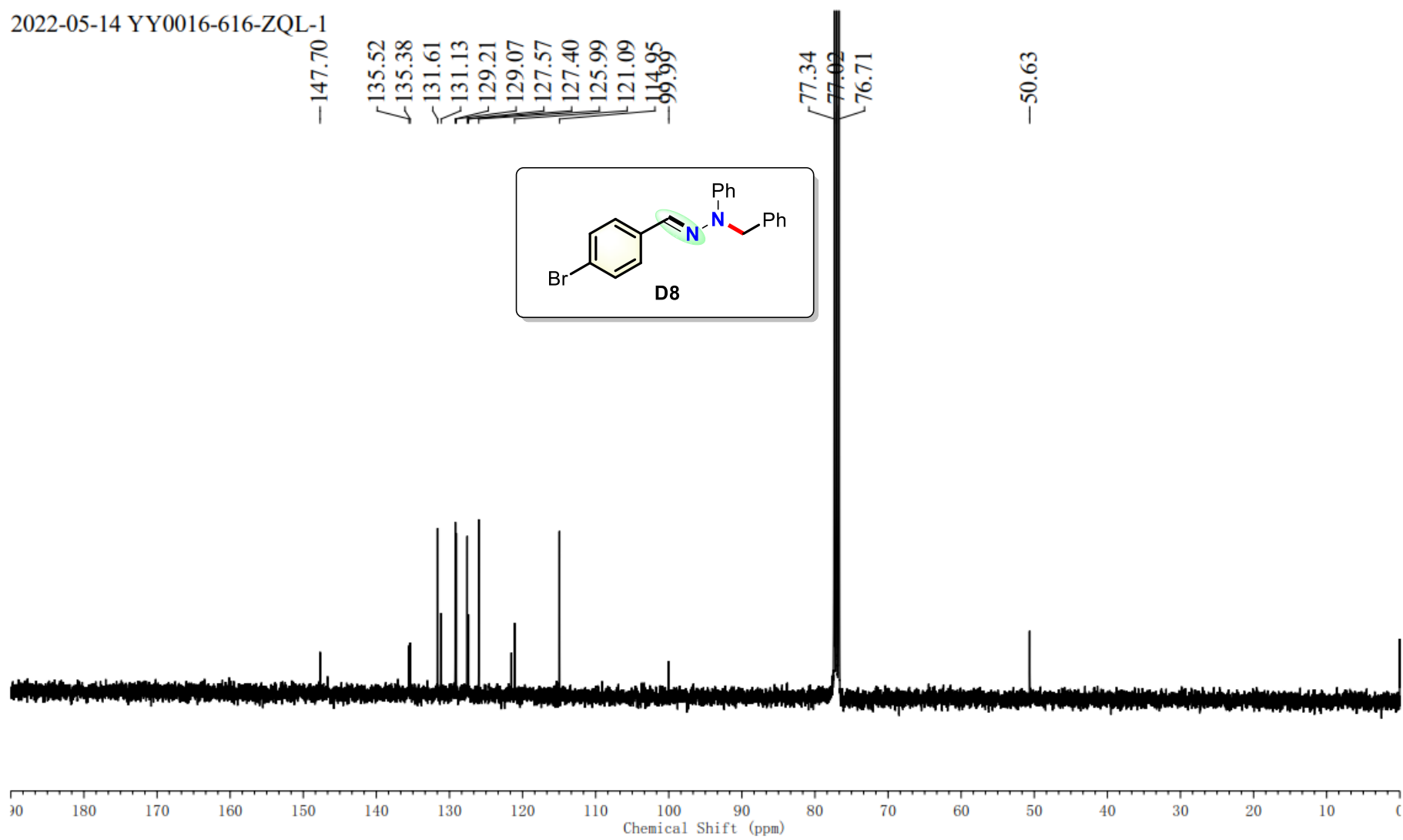
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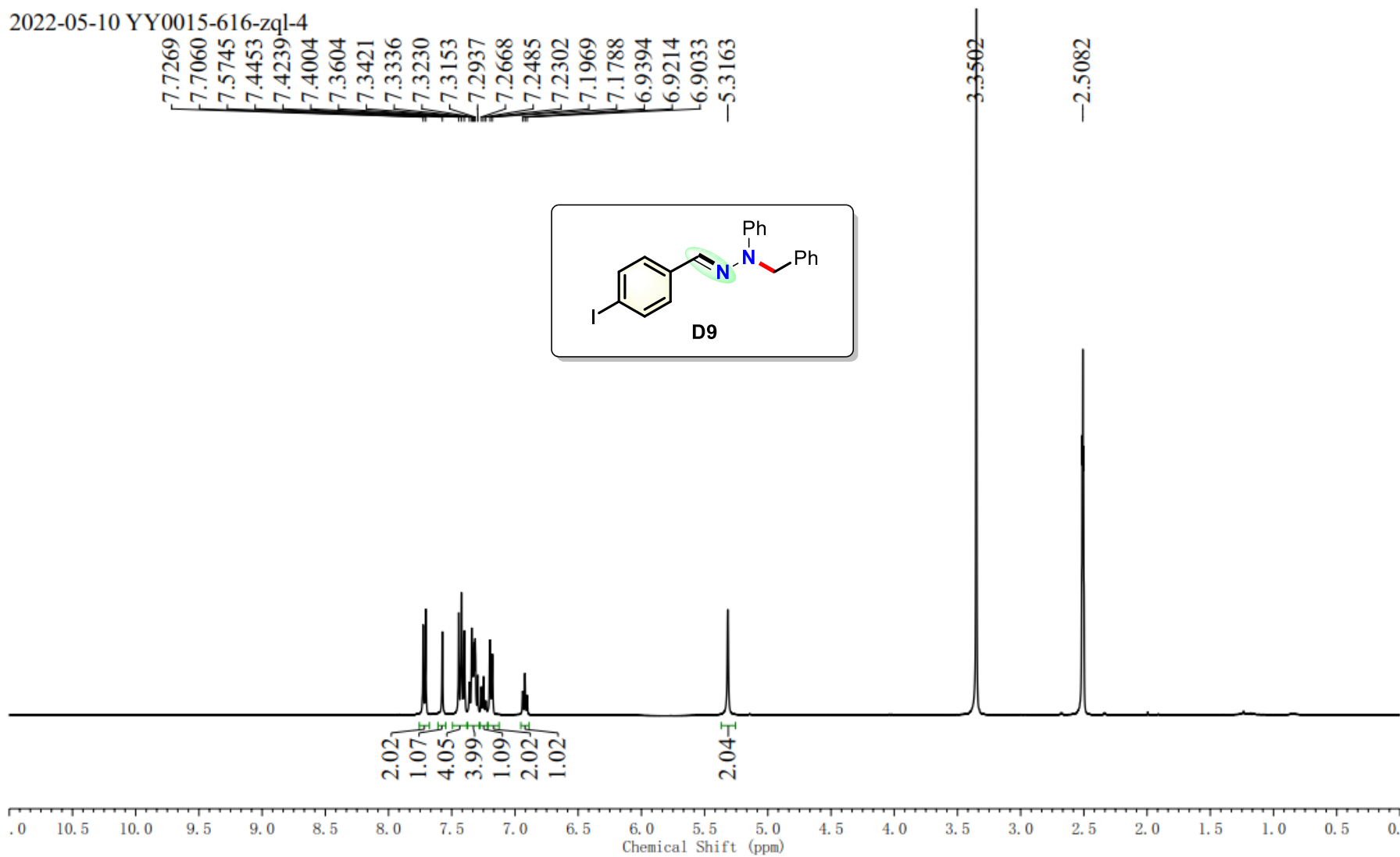
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2022-05-14 YY0016-616-ZQL-1



2022-05-10 YY0015-616-zql-4

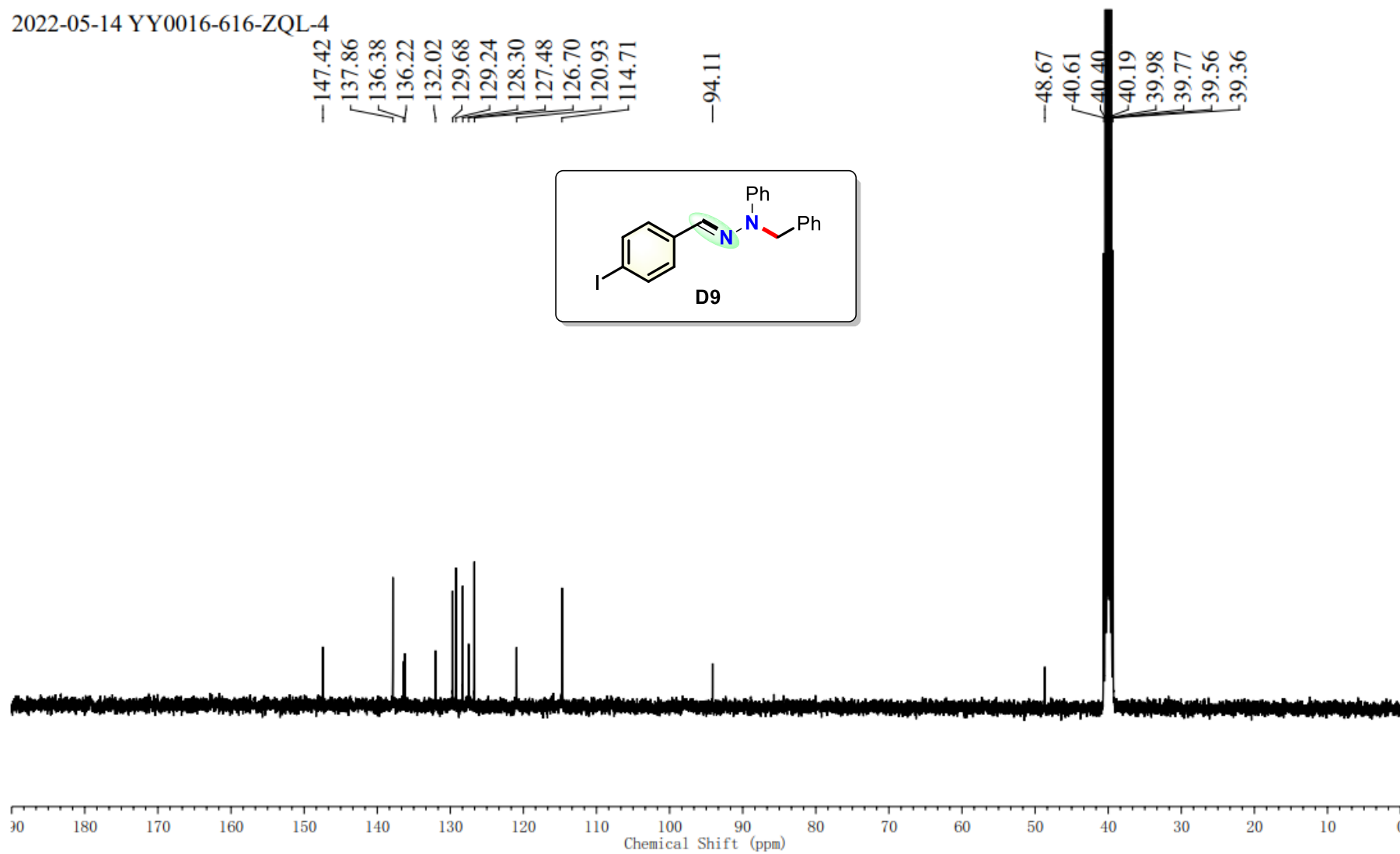
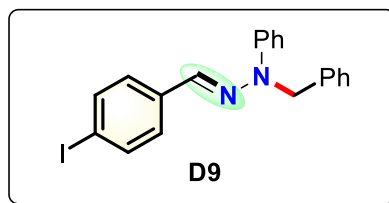


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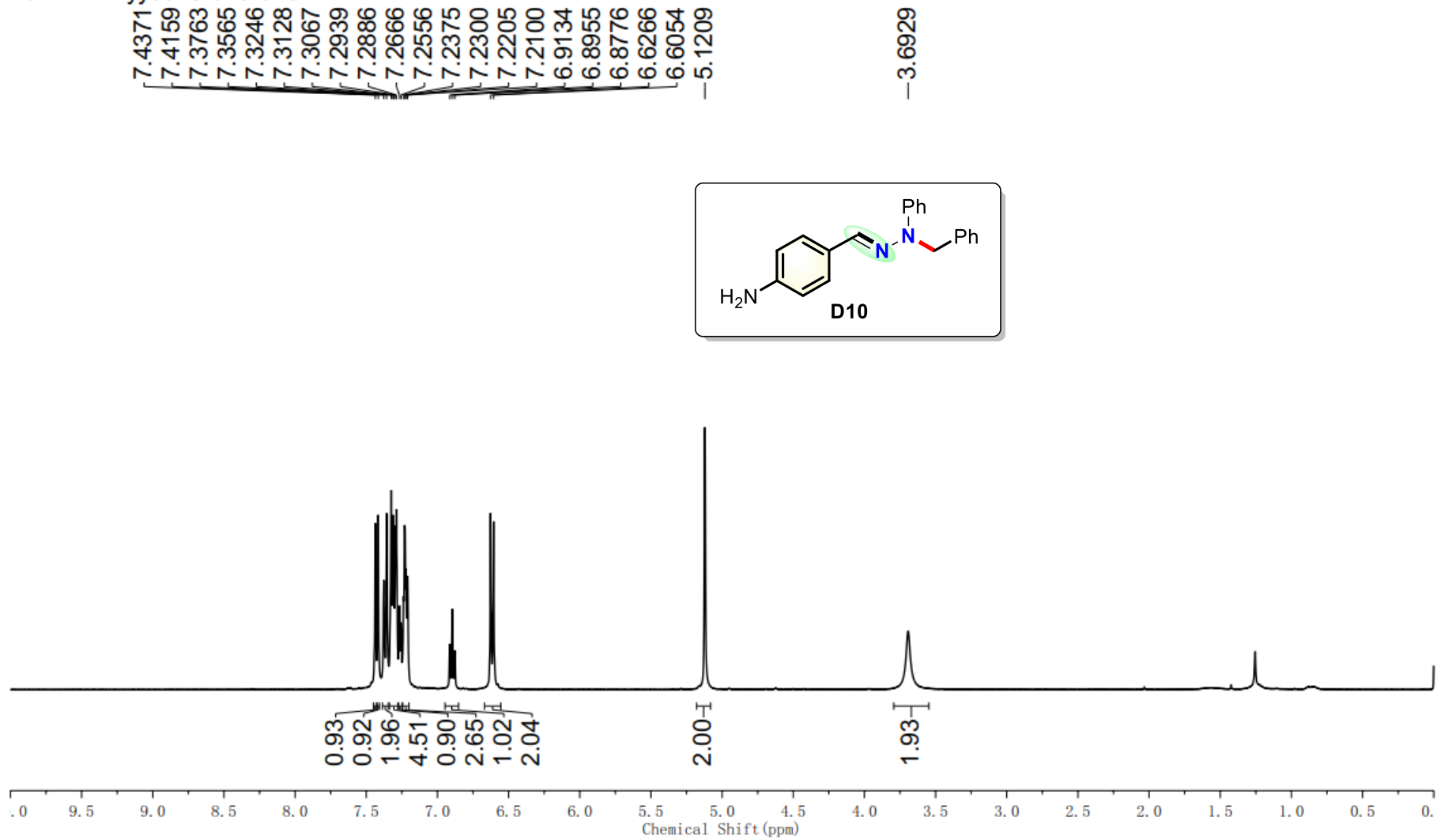
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127.48
126.70
120.93
114.71

94.11

48.67
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40.40
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39.98
39.77
39.56
39.36



2022-11-7-yy0025-616-e20

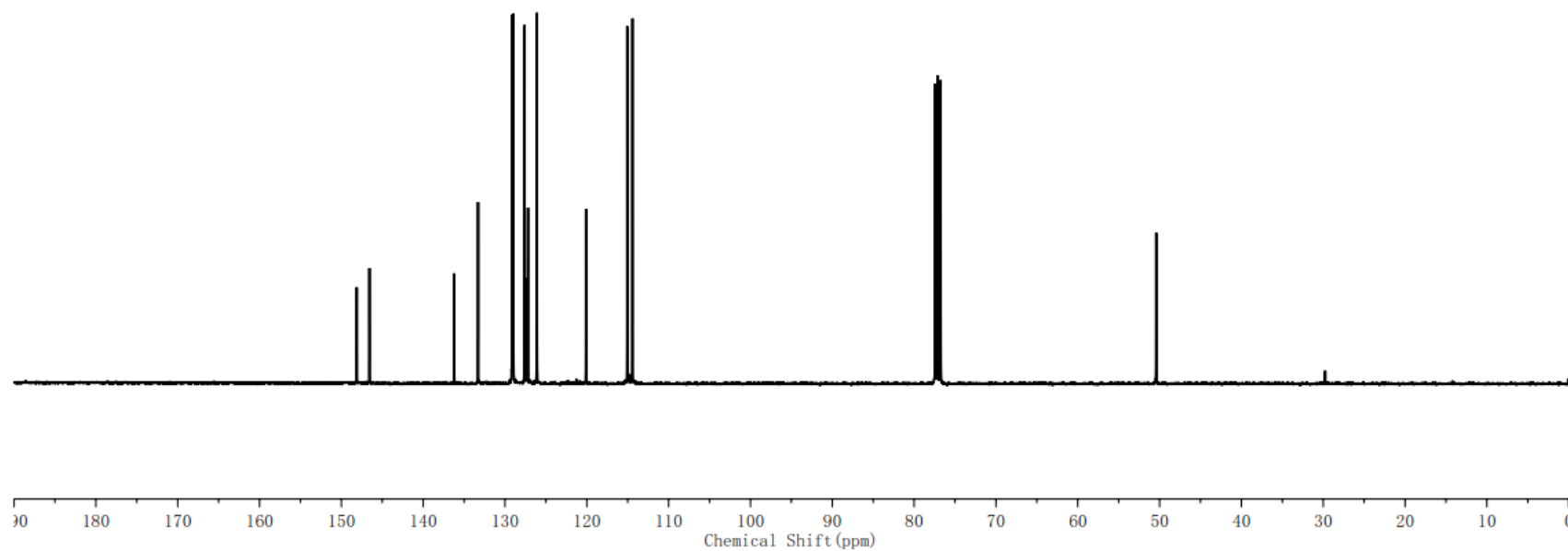
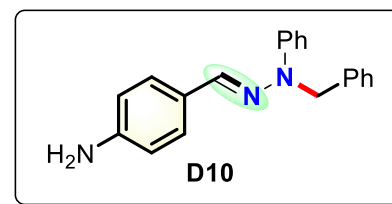


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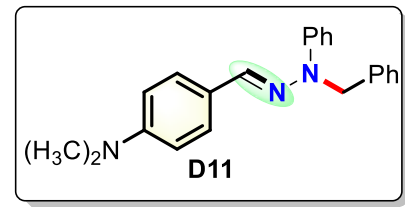
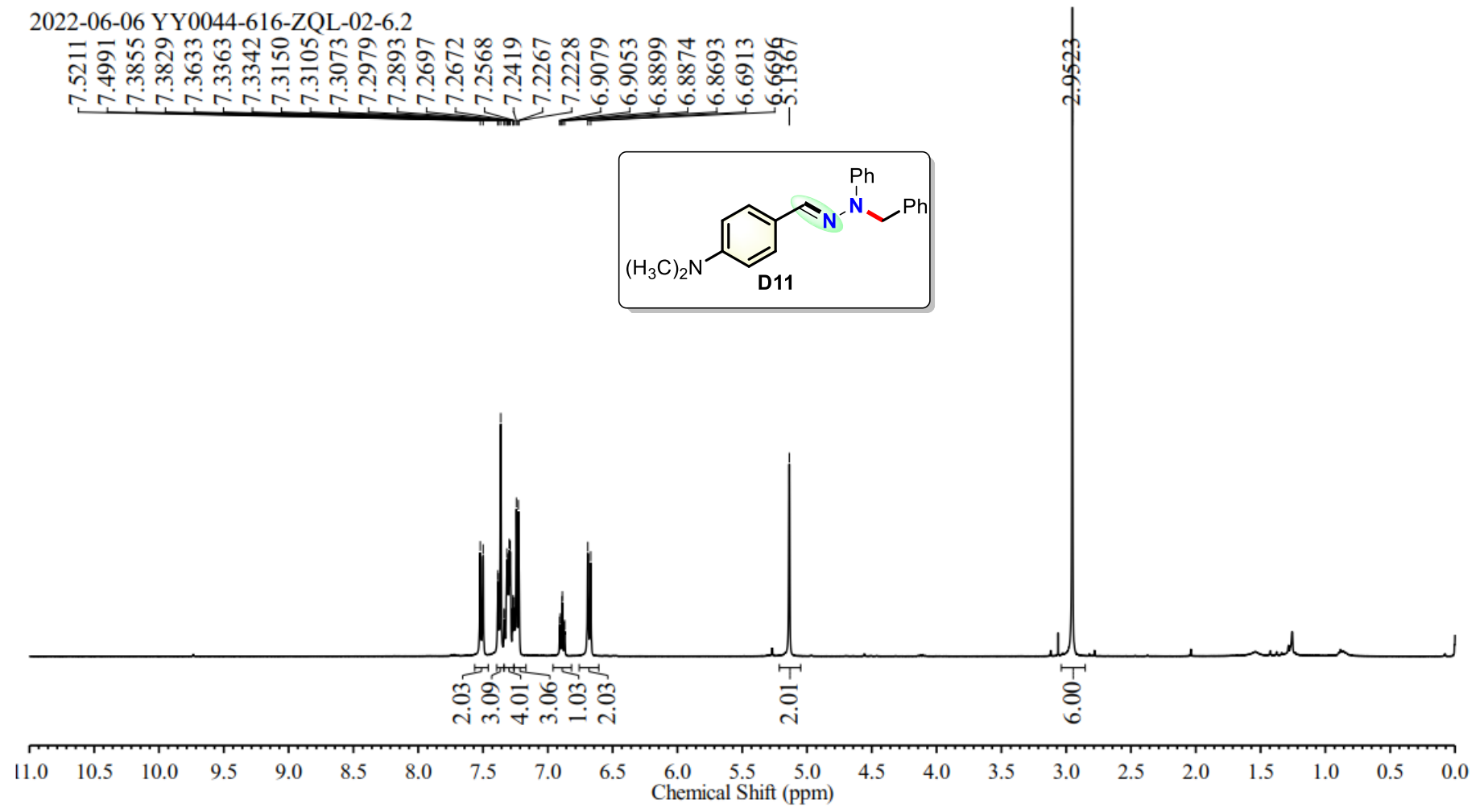
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146.57
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133.29
129.14
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127.60
127.30
127.20
126.11
120.10
115.05
114.42

77.41
77.09
76.78

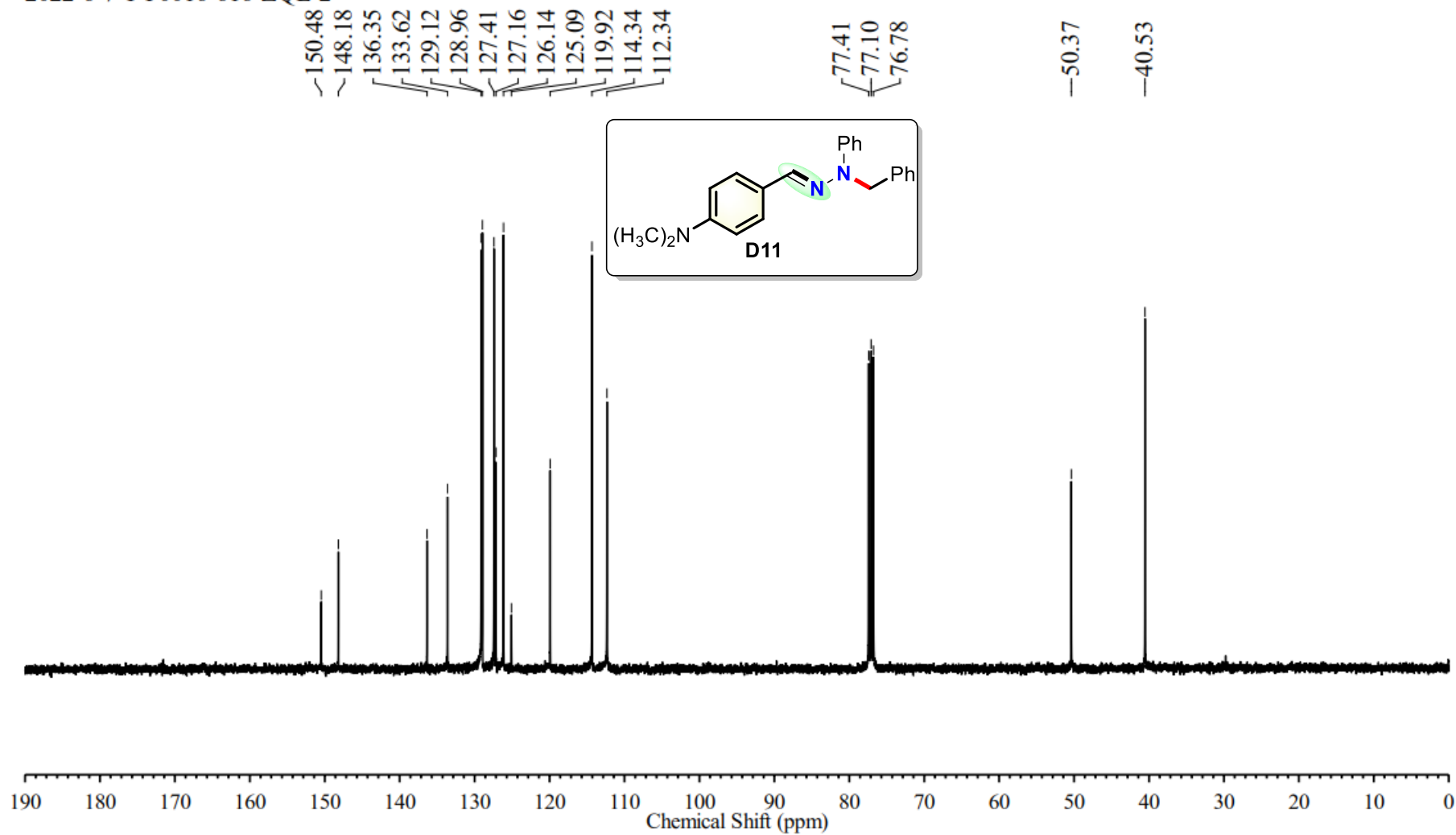
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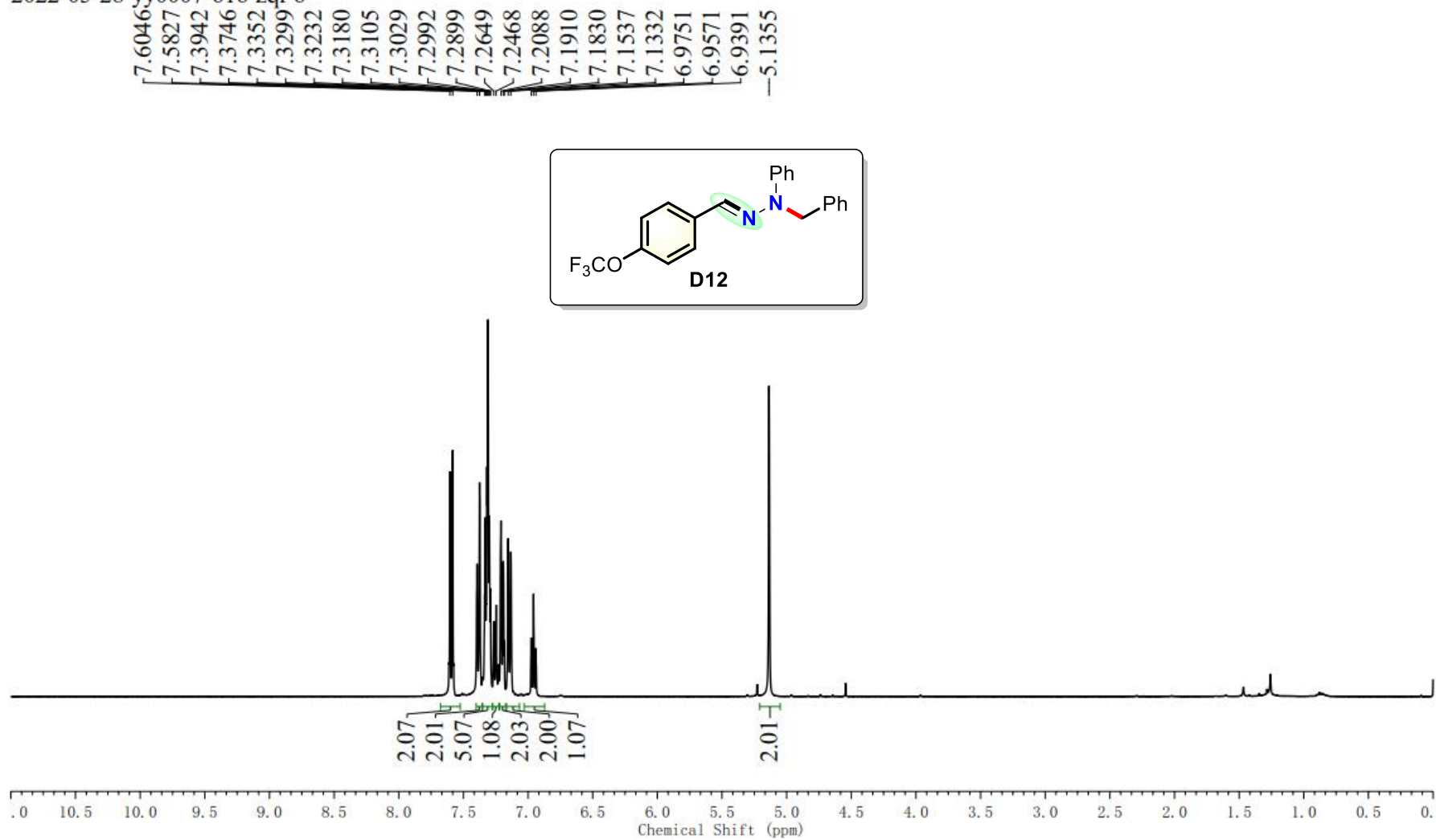
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2022-6-7 YY0016-616-ZQL-2

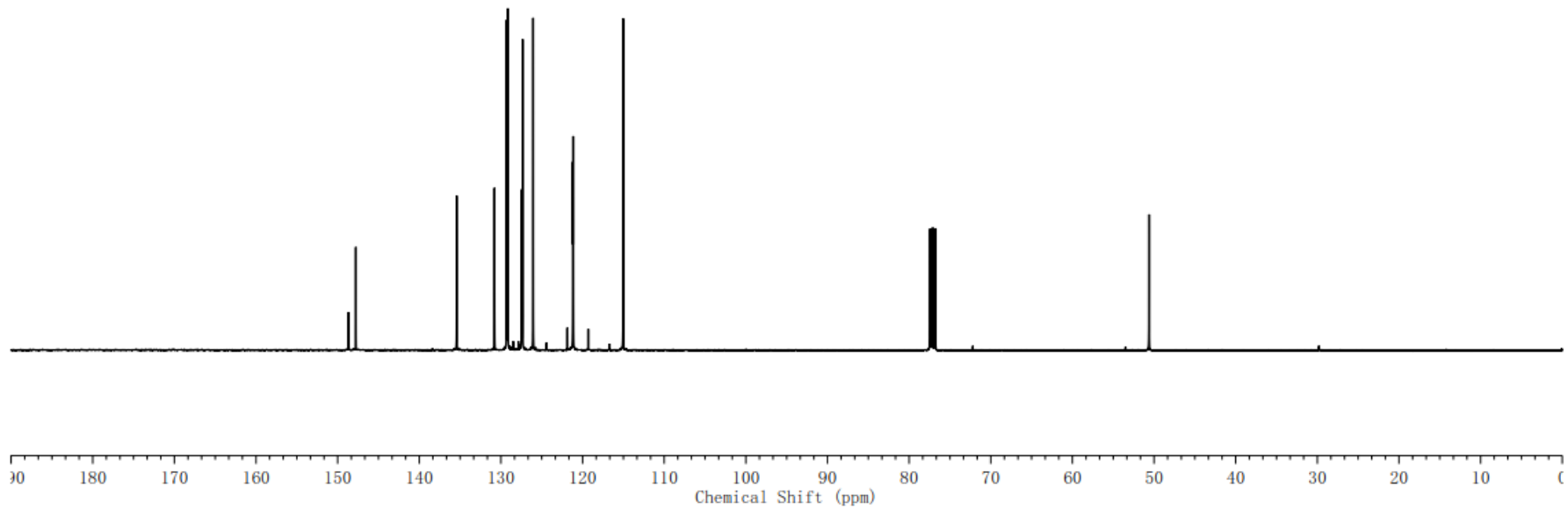
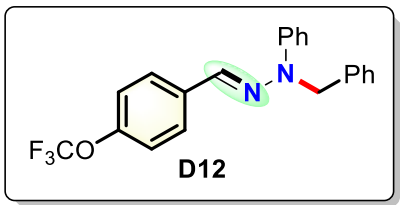


2022-05-28-yy0007-616-zq1-6

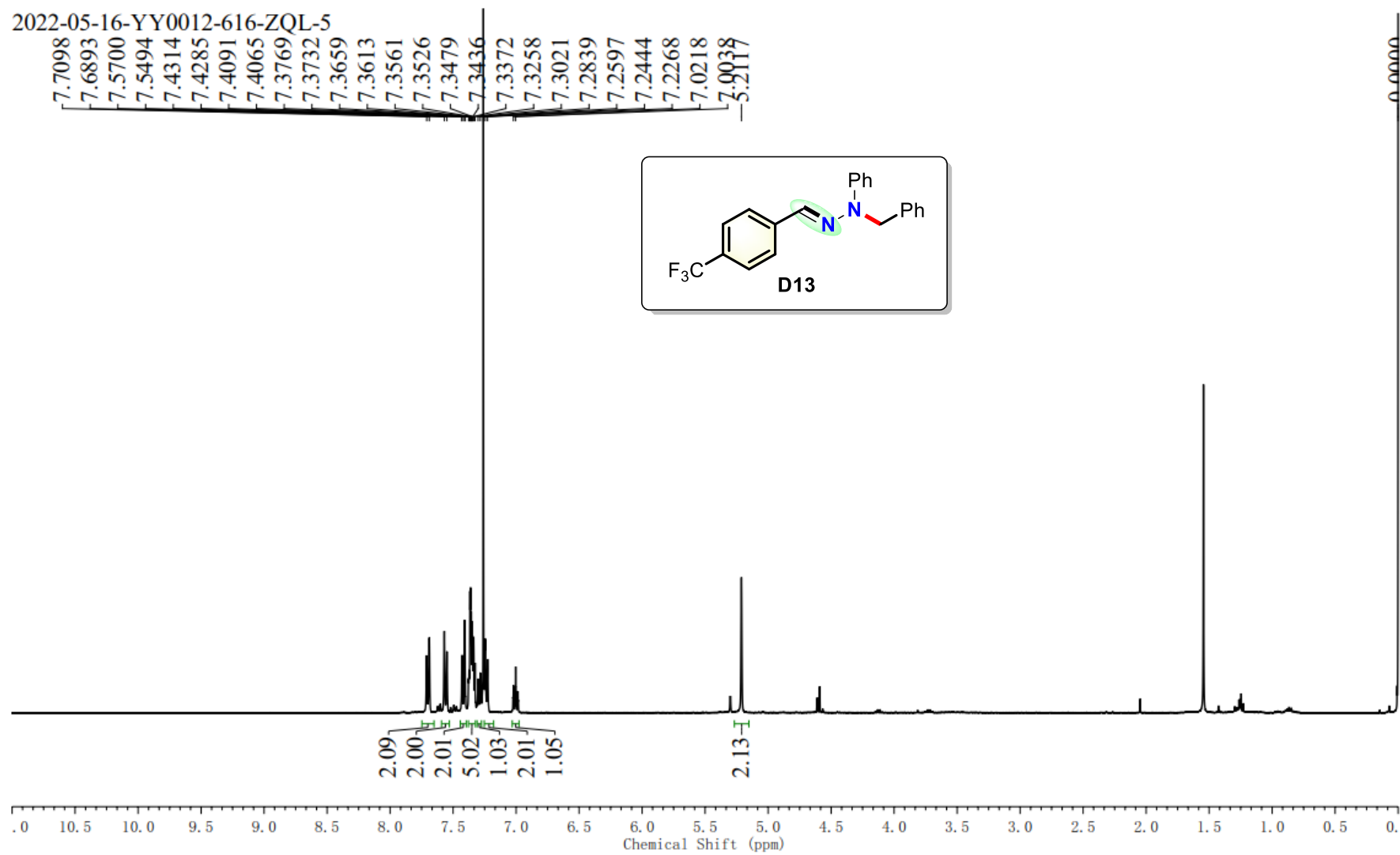


2022-05-30 YY0021 616-ZQL-0527-6

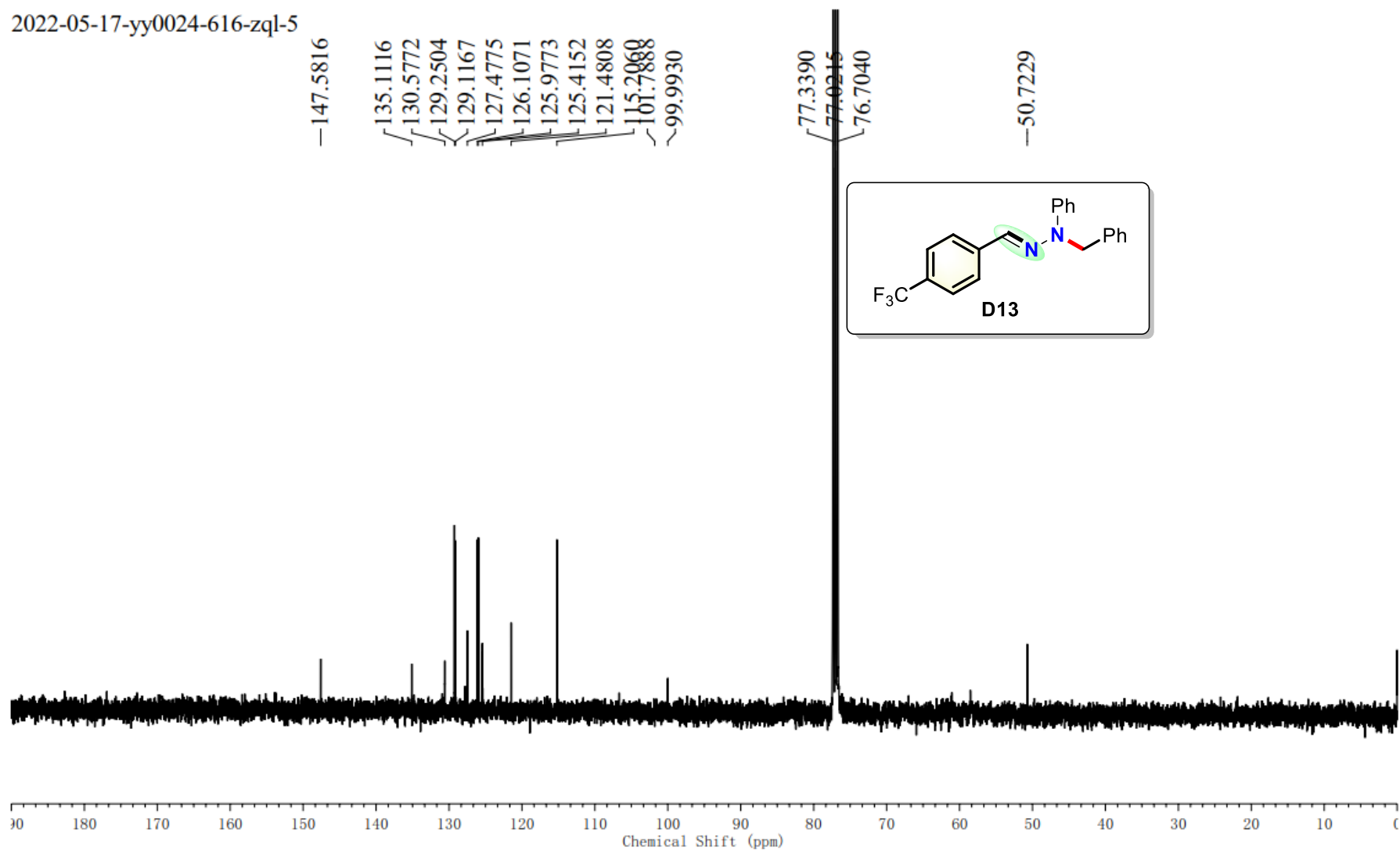
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129.15
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121.15
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77.43
77.11
76.79
50.61



2022-05-16-YY0012-616-ZQL-5

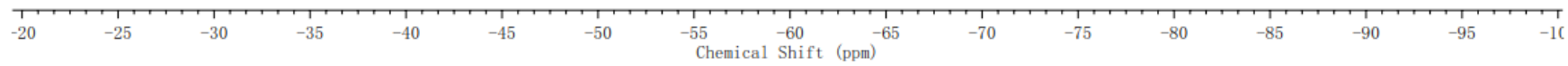
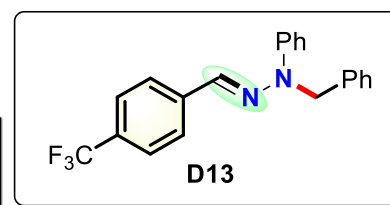


2022-05-17-yy0024-616-zql-5



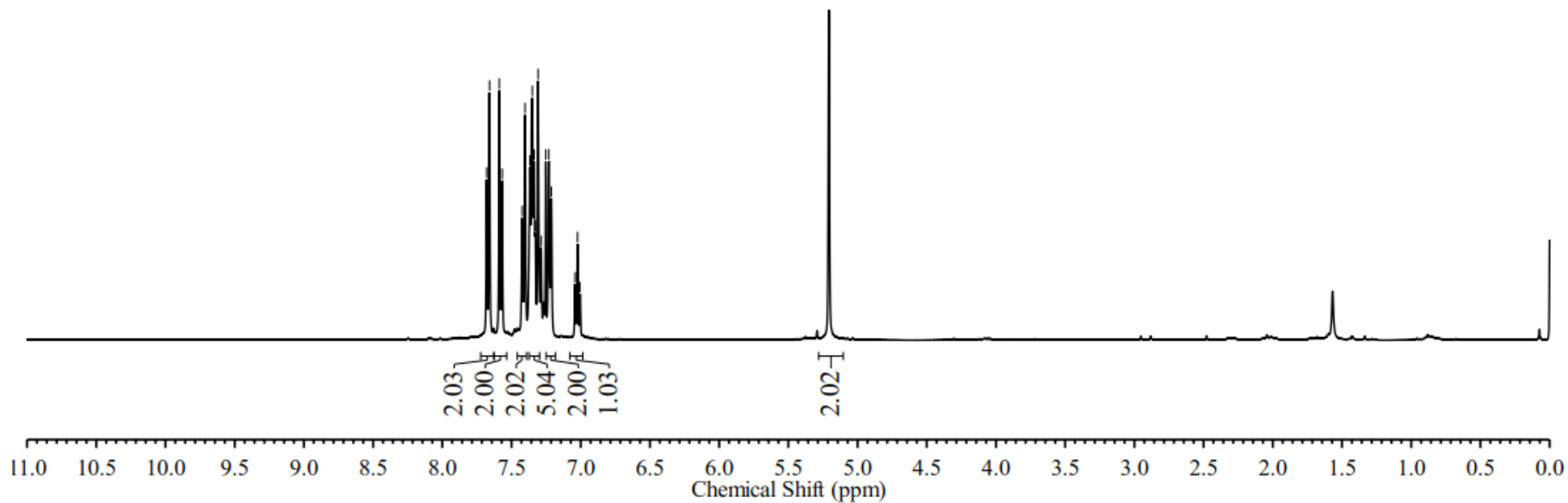
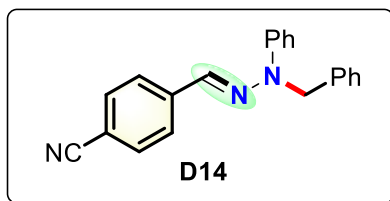
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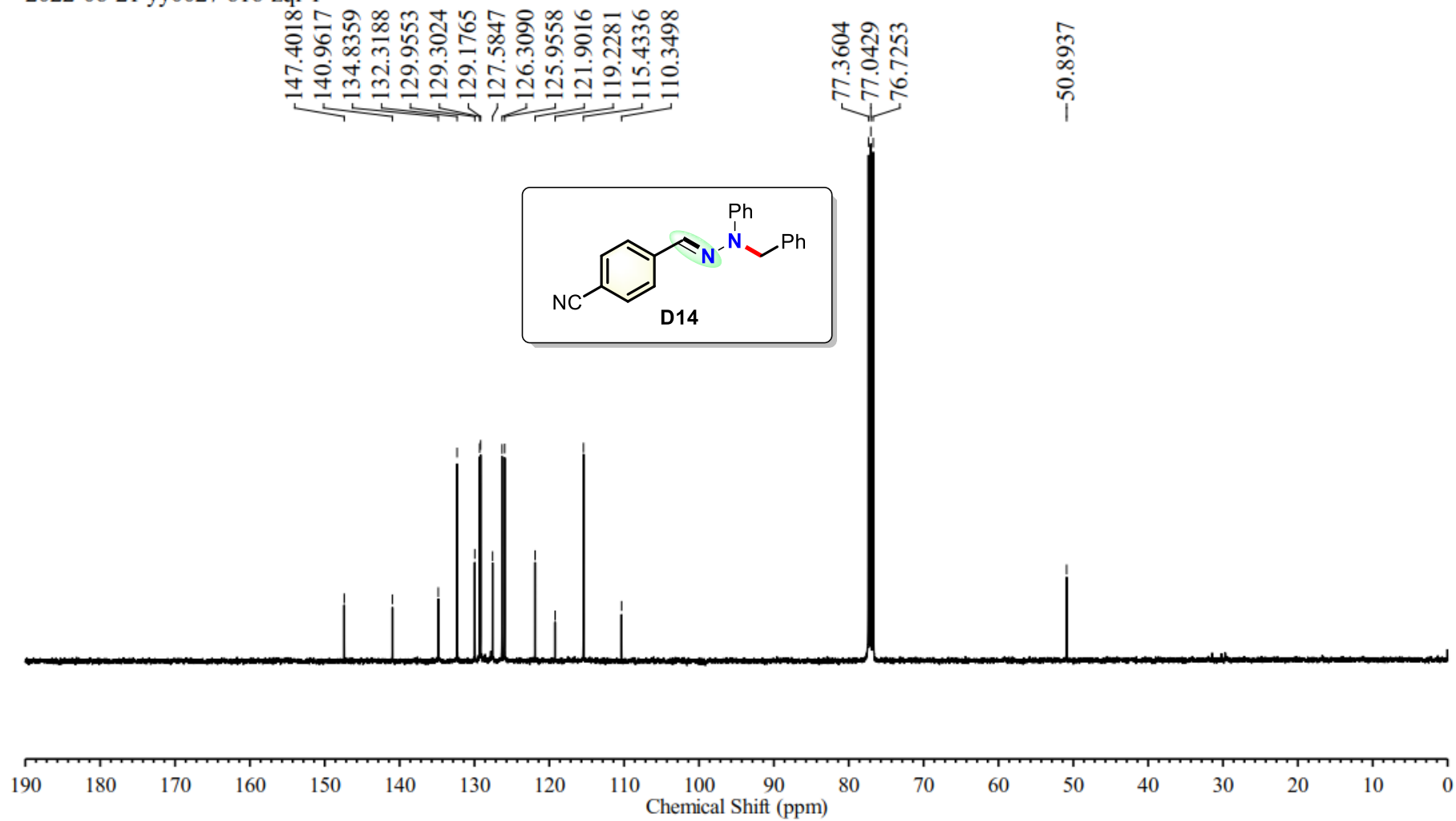


2022-06-20 yy0003-616-zql-1

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7.0229
7.0050

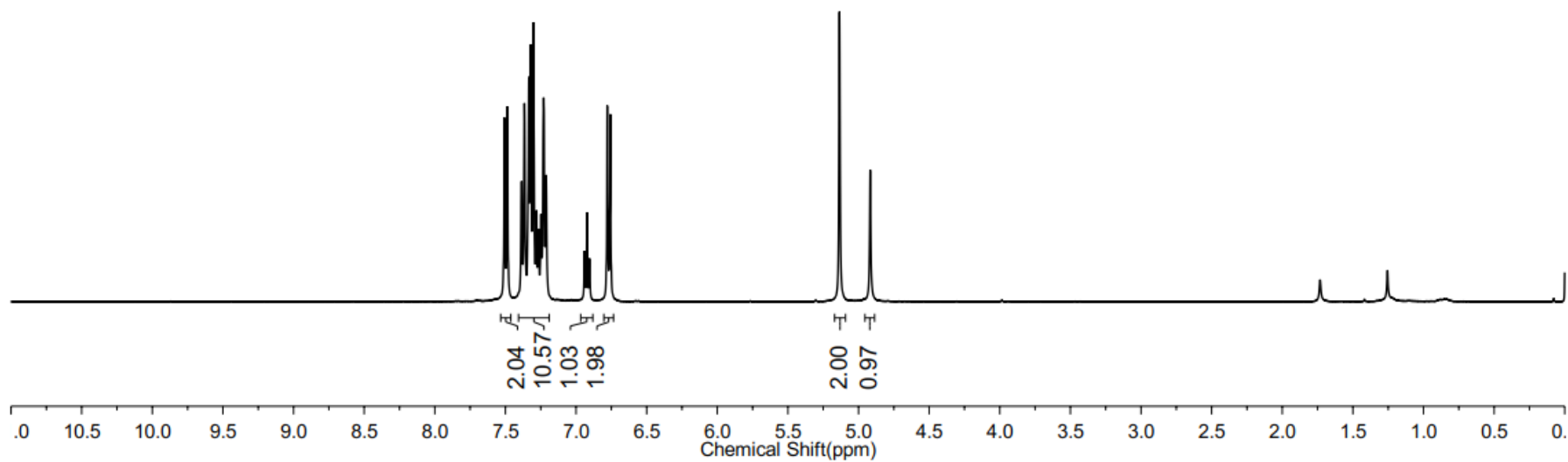
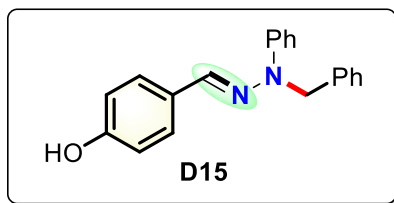


2022-06-21 yy0027 616-zql-1



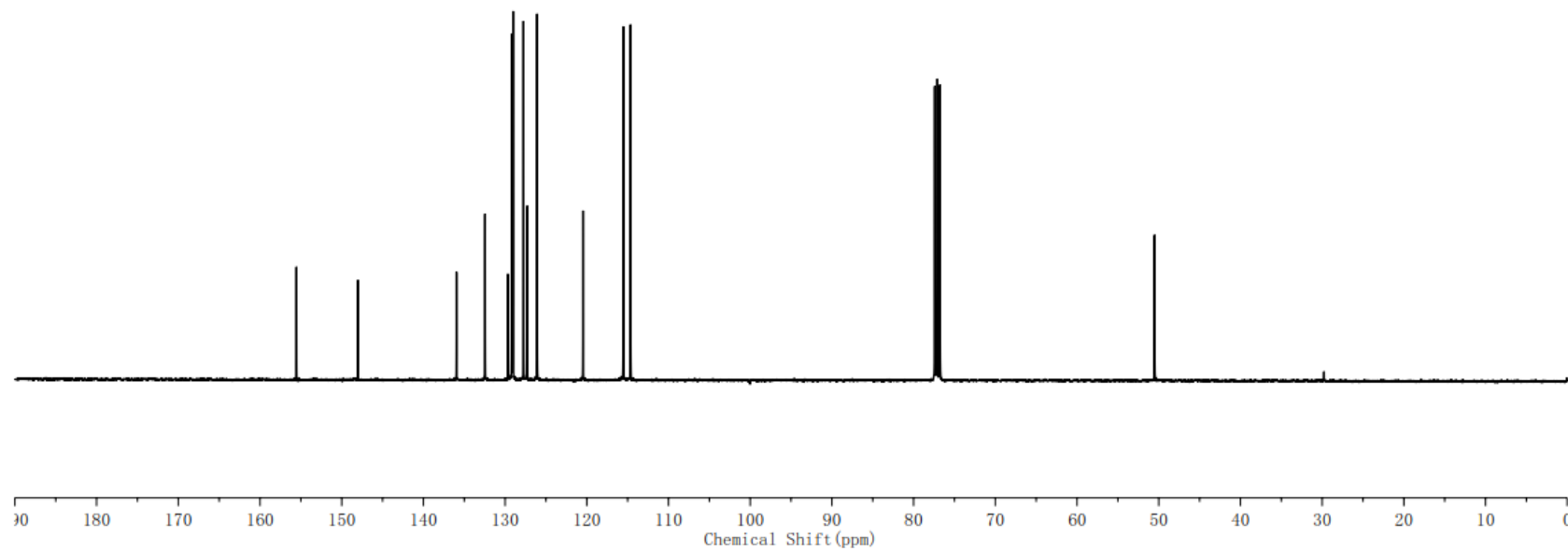
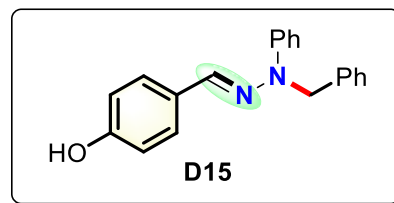
2022-11-7-yy0025-616-e26

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7.2483
7.2293
7.2118
6.9400
6.9221
6.9041
6.7790
6.7575
— 5.1350
— 4.9171

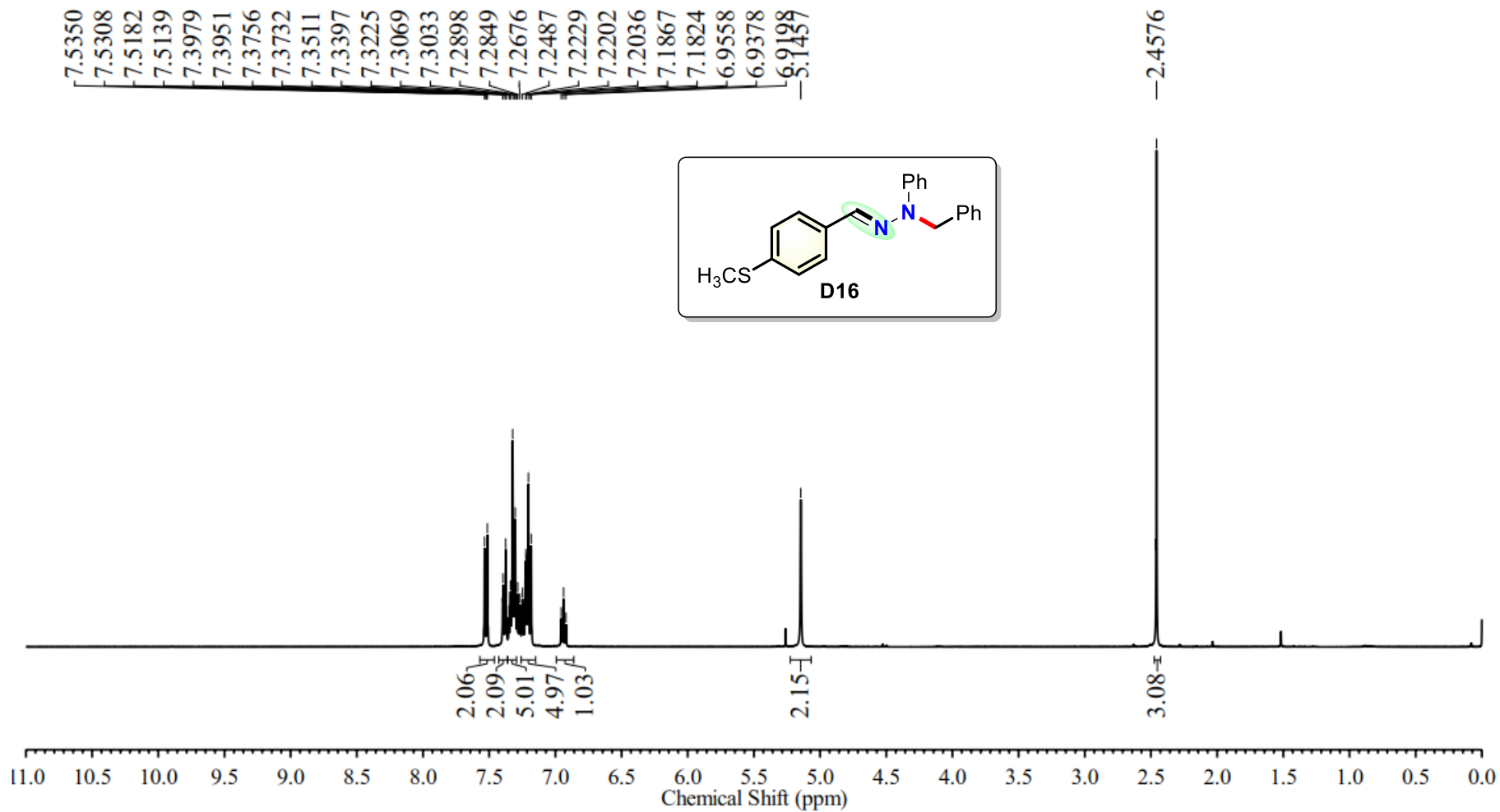


2022-11-10-yy0031-616-E26

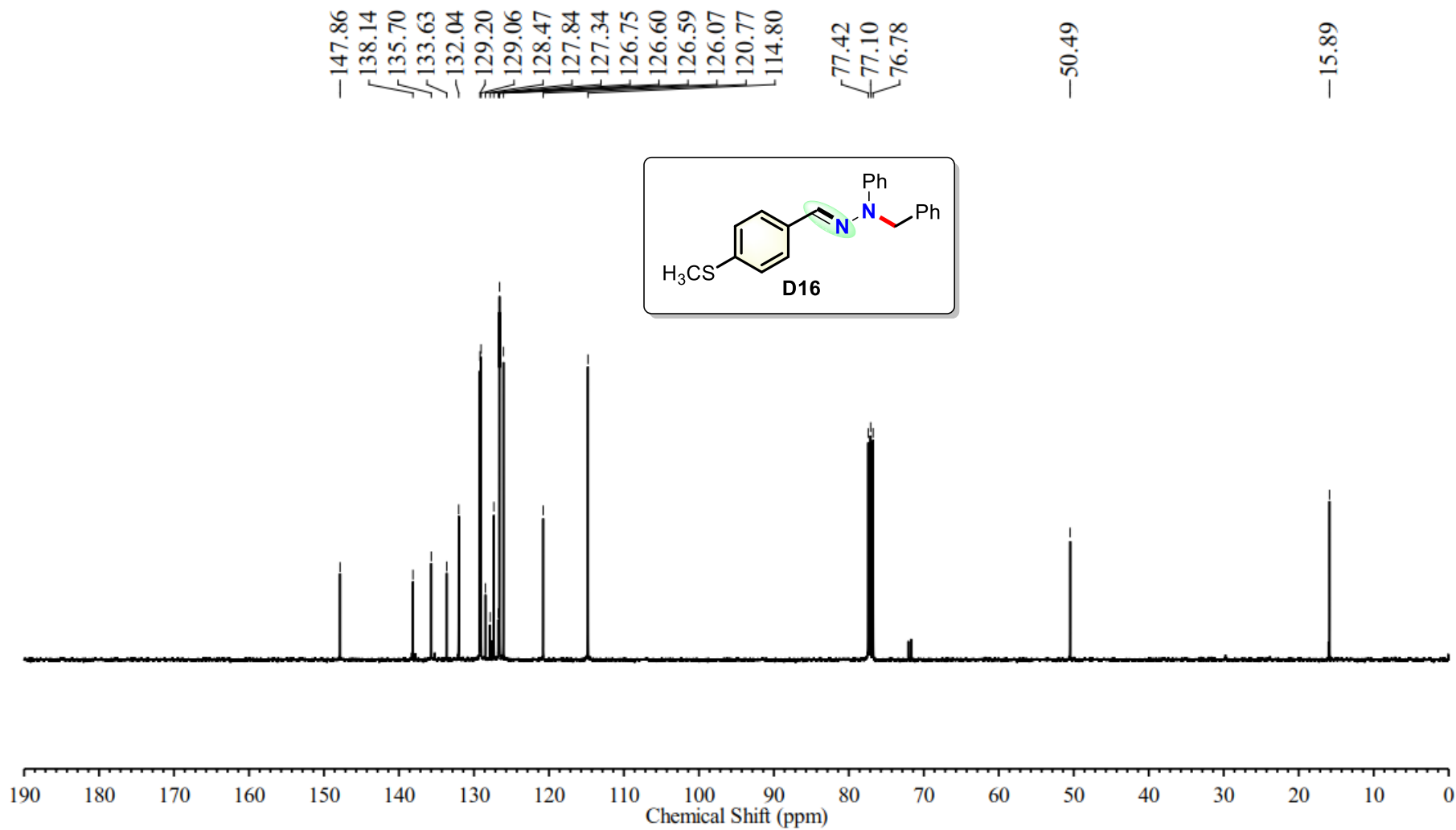
155.53
148.00
135.95
132.46
129.66
129.19
129.03
127.74
127.29
126.08
120.47
115.54
114.64
77.40
77.09
76.77
50.50



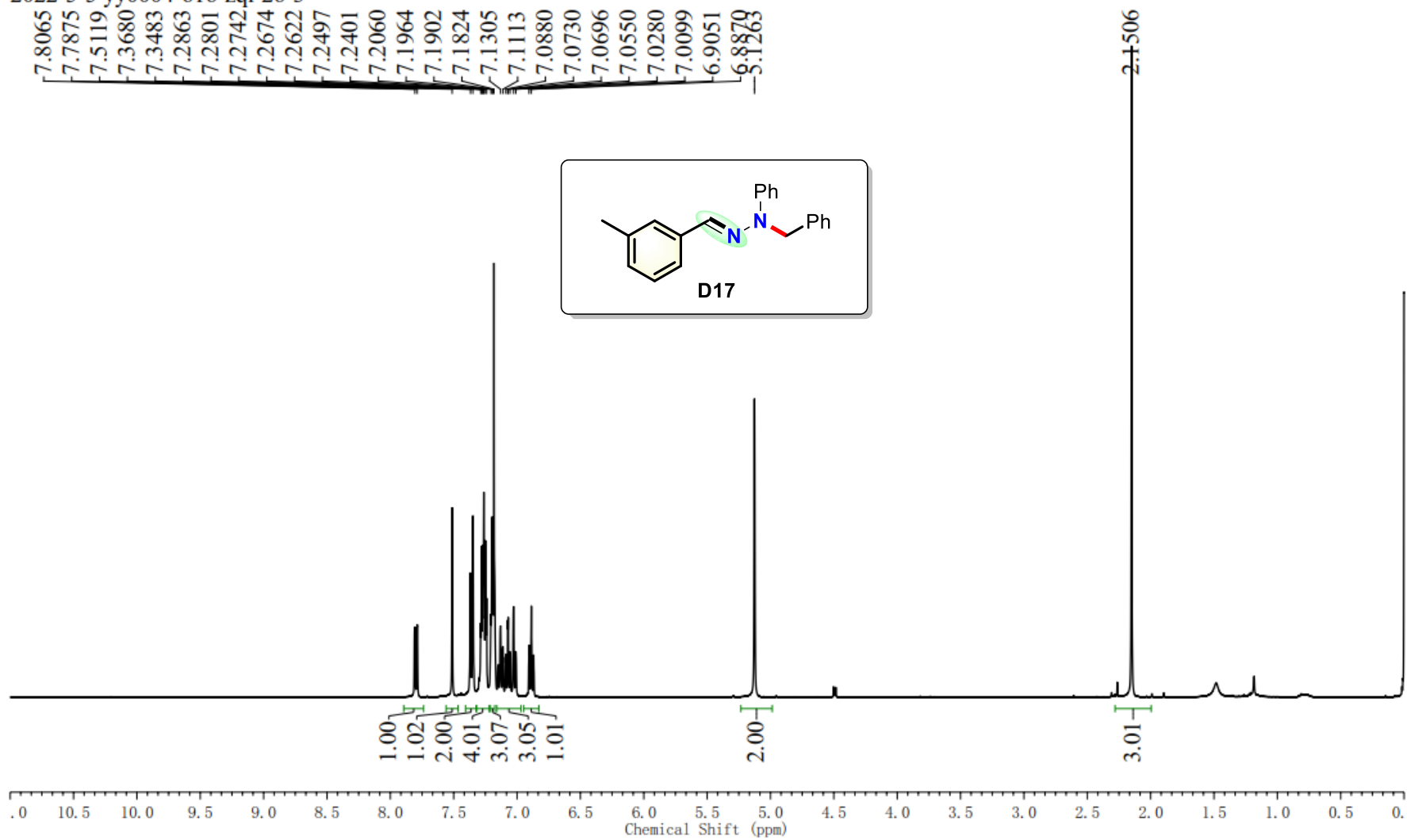
2022-06-06 YY0044-616-ZQL-04-6.2



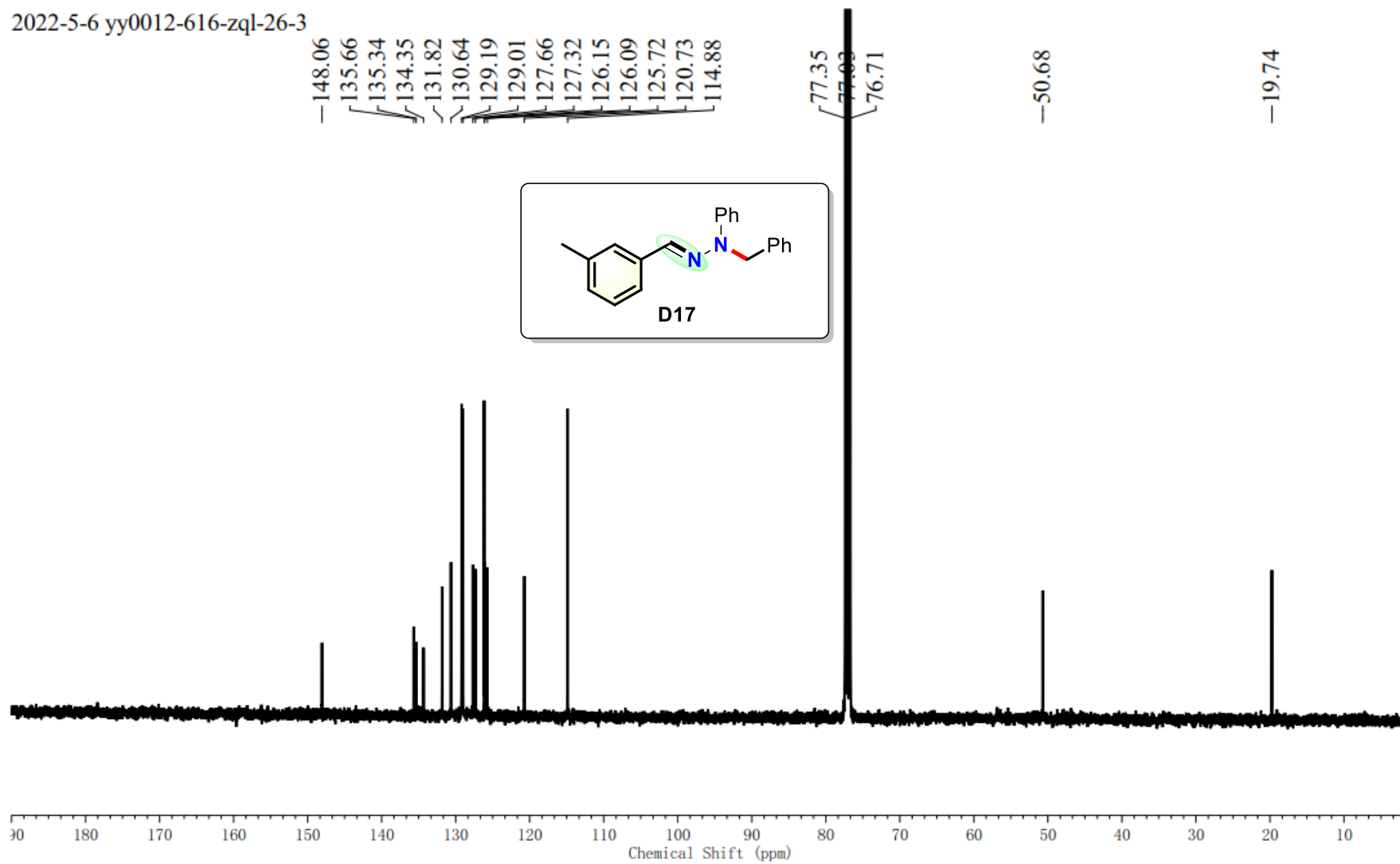
2022-6-7 YY0016-616-ZQL-4



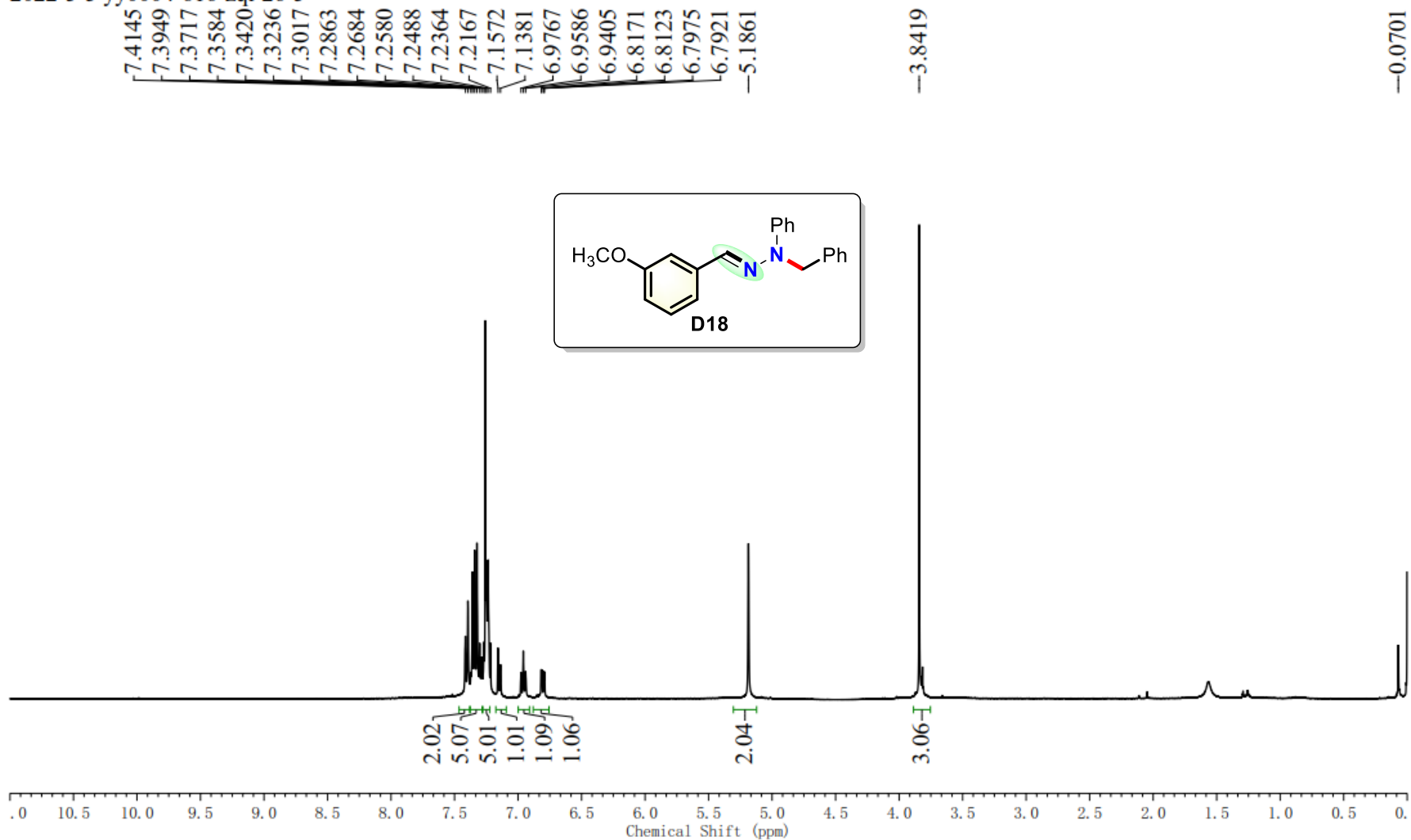
2022-5-5 yy0004-616-zql-26-3



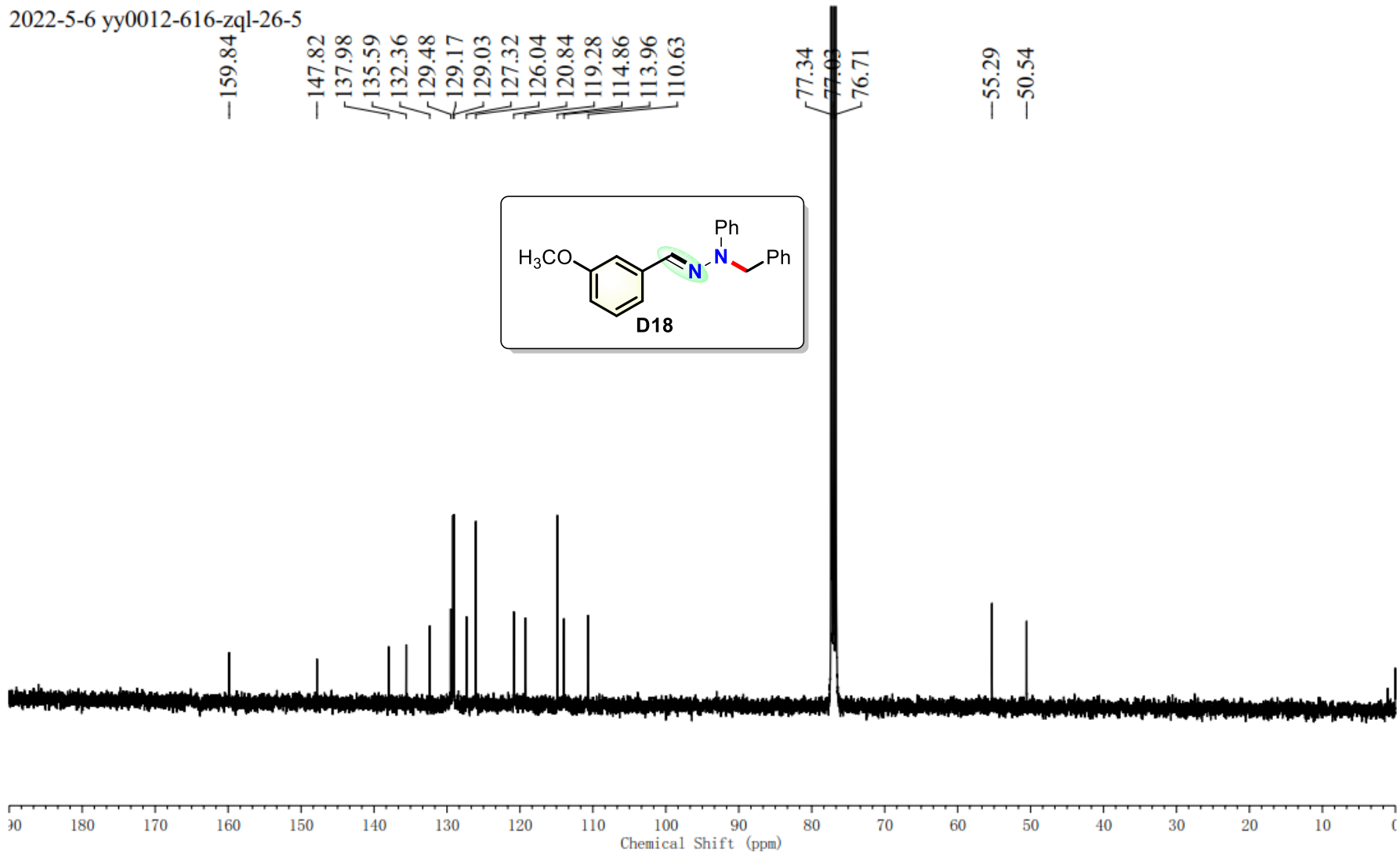
2022-5-6 yy0012-616-zql-26-3



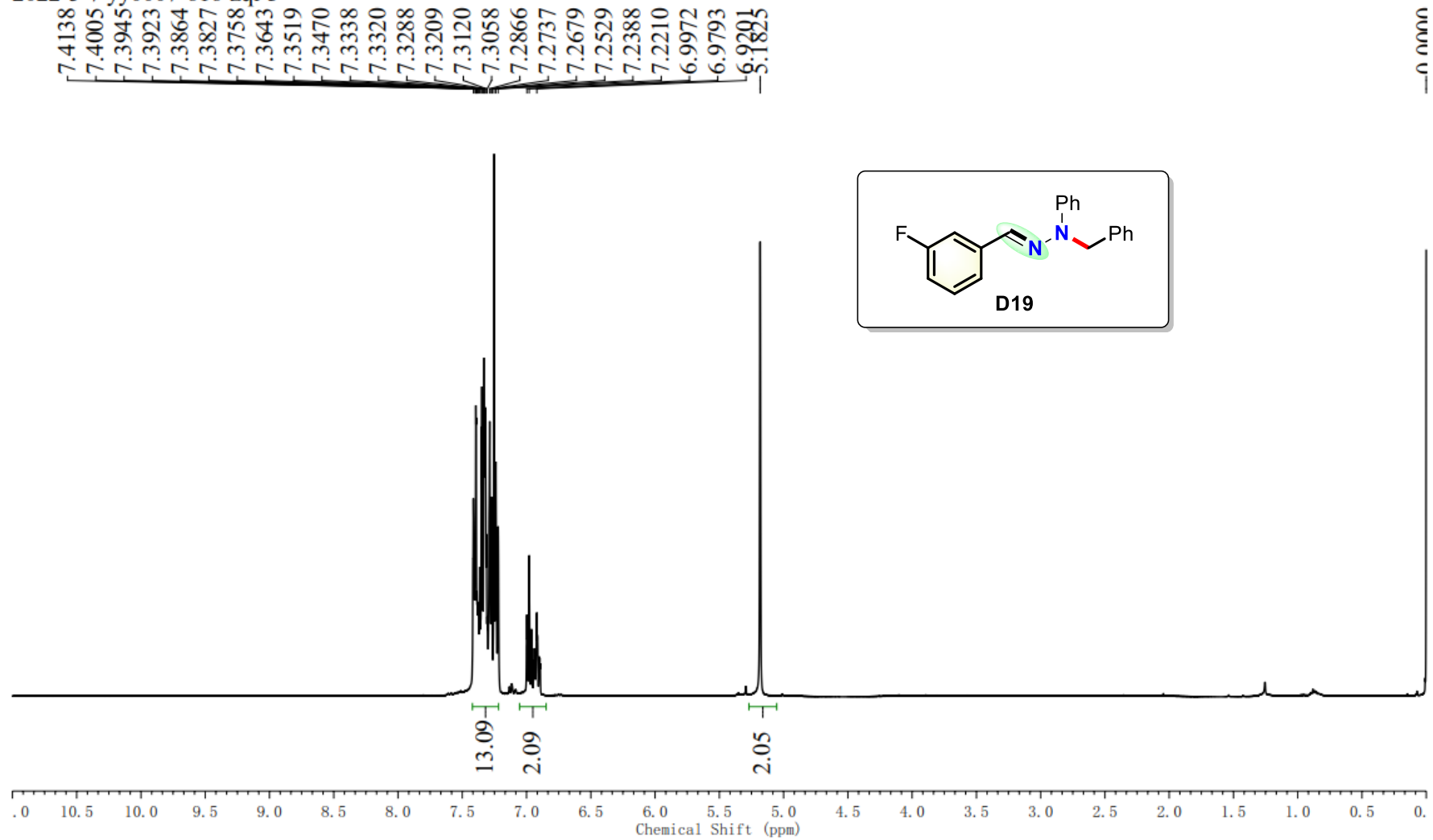
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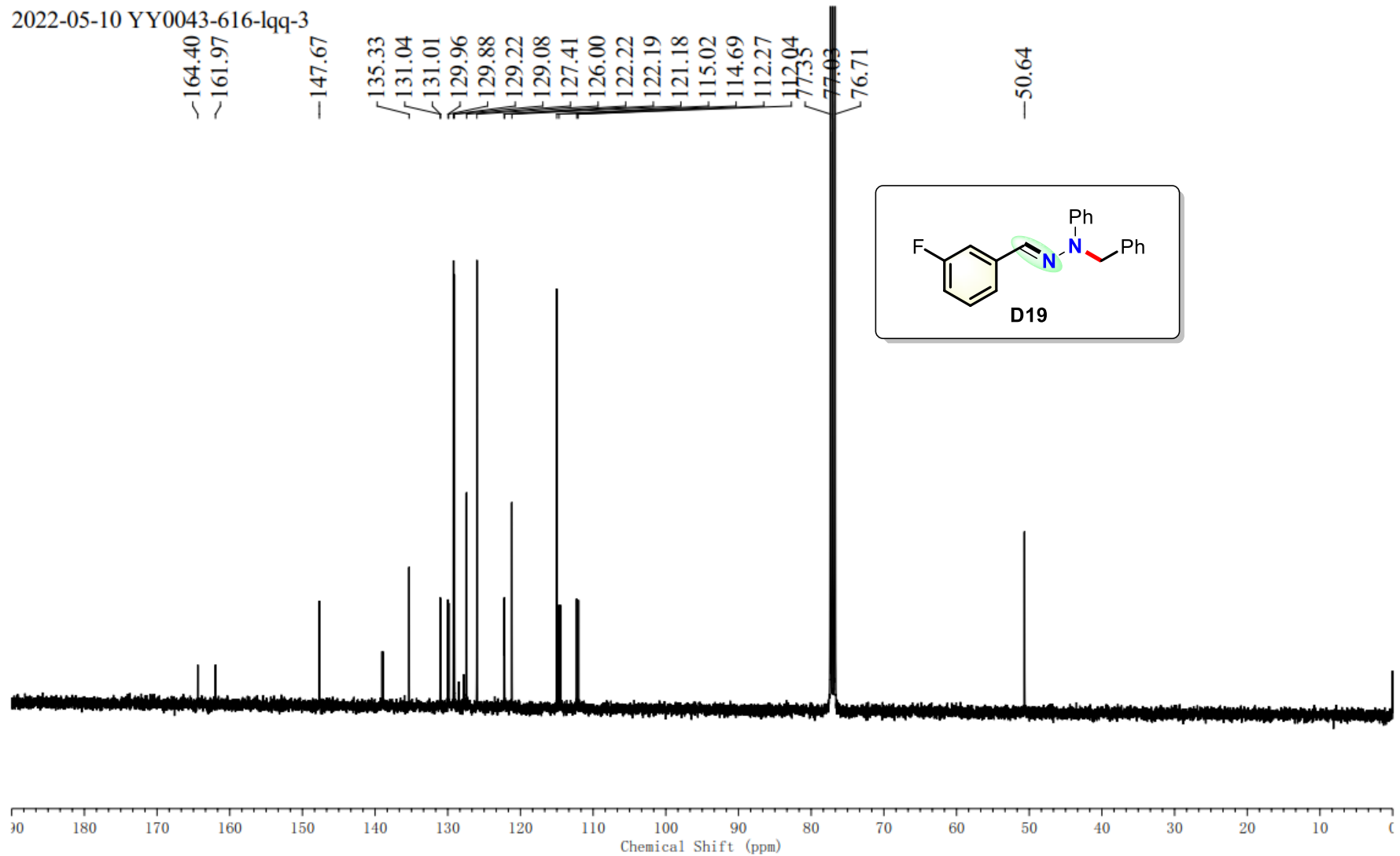
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2022-5-7 yy0007-616-zql-3

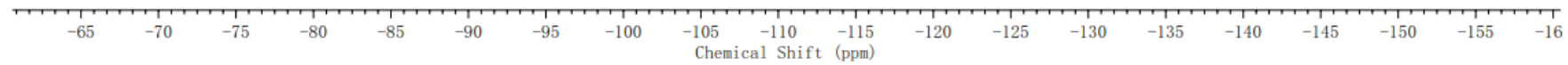
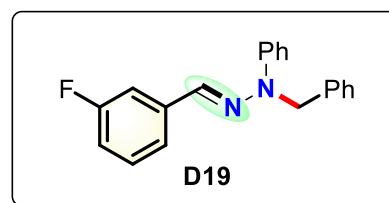


2022-05-10 YY0043-616-lqq-3

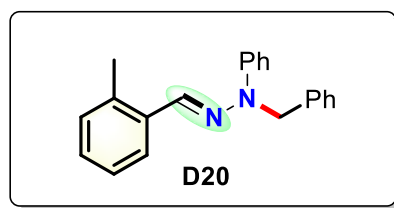
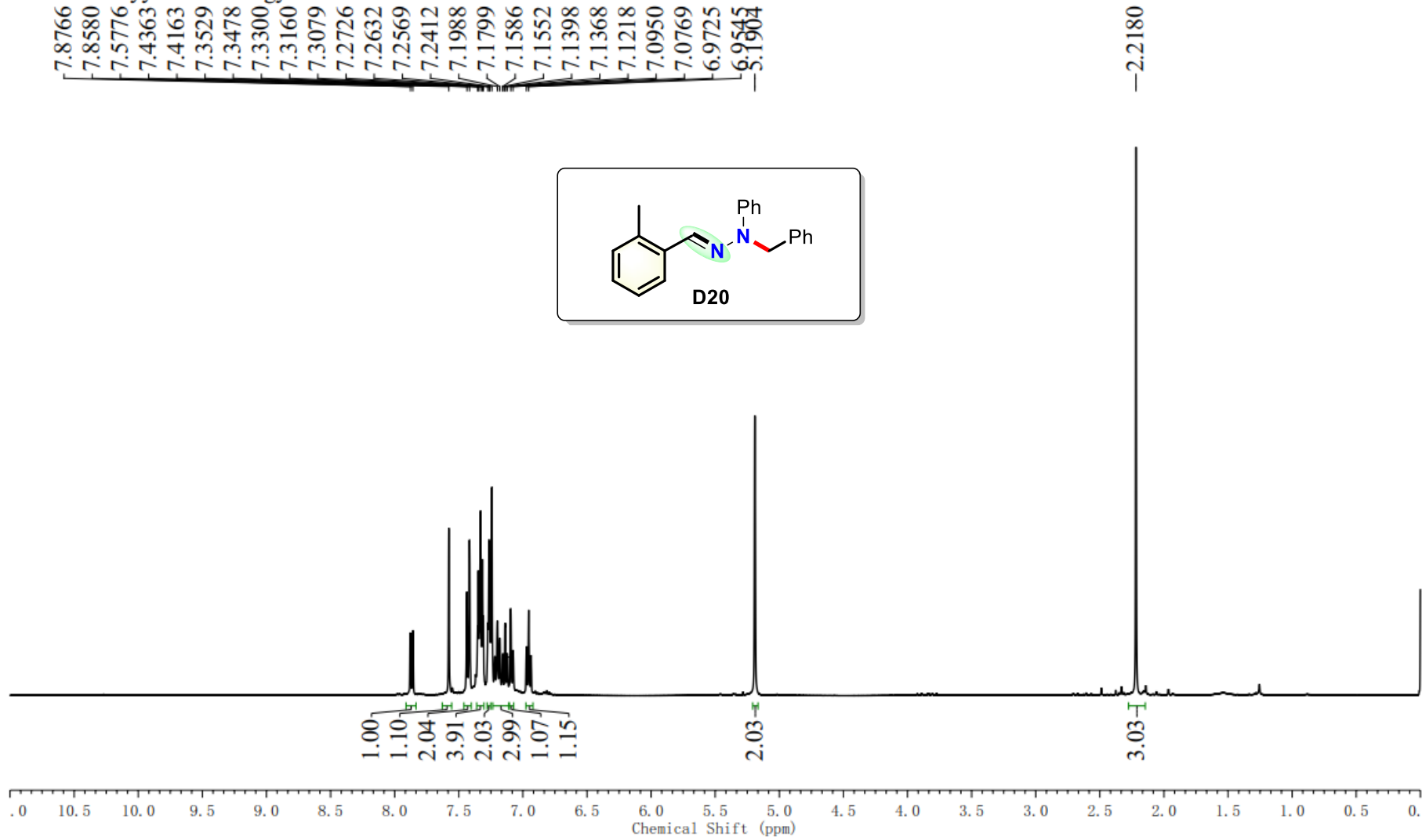


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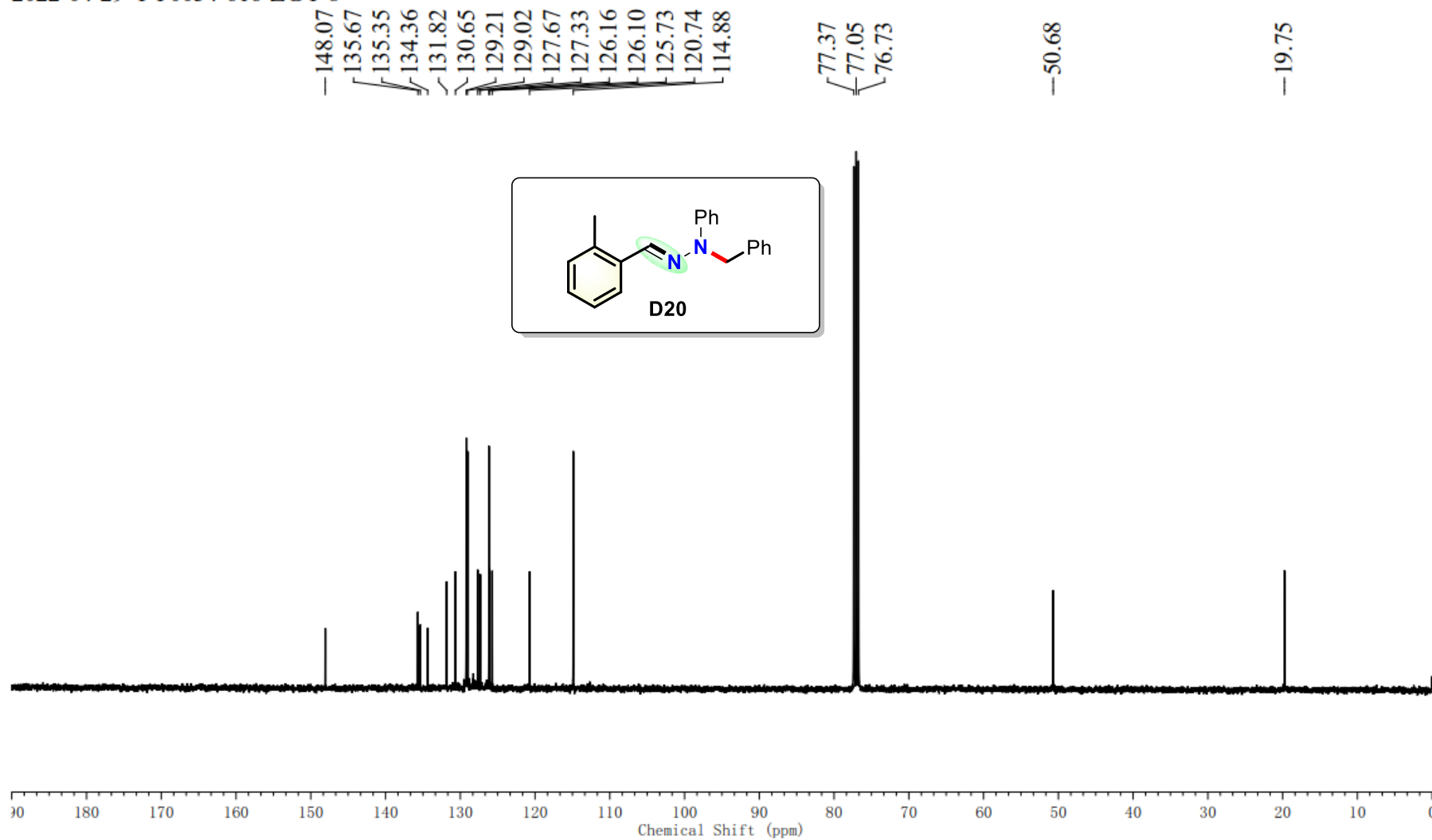
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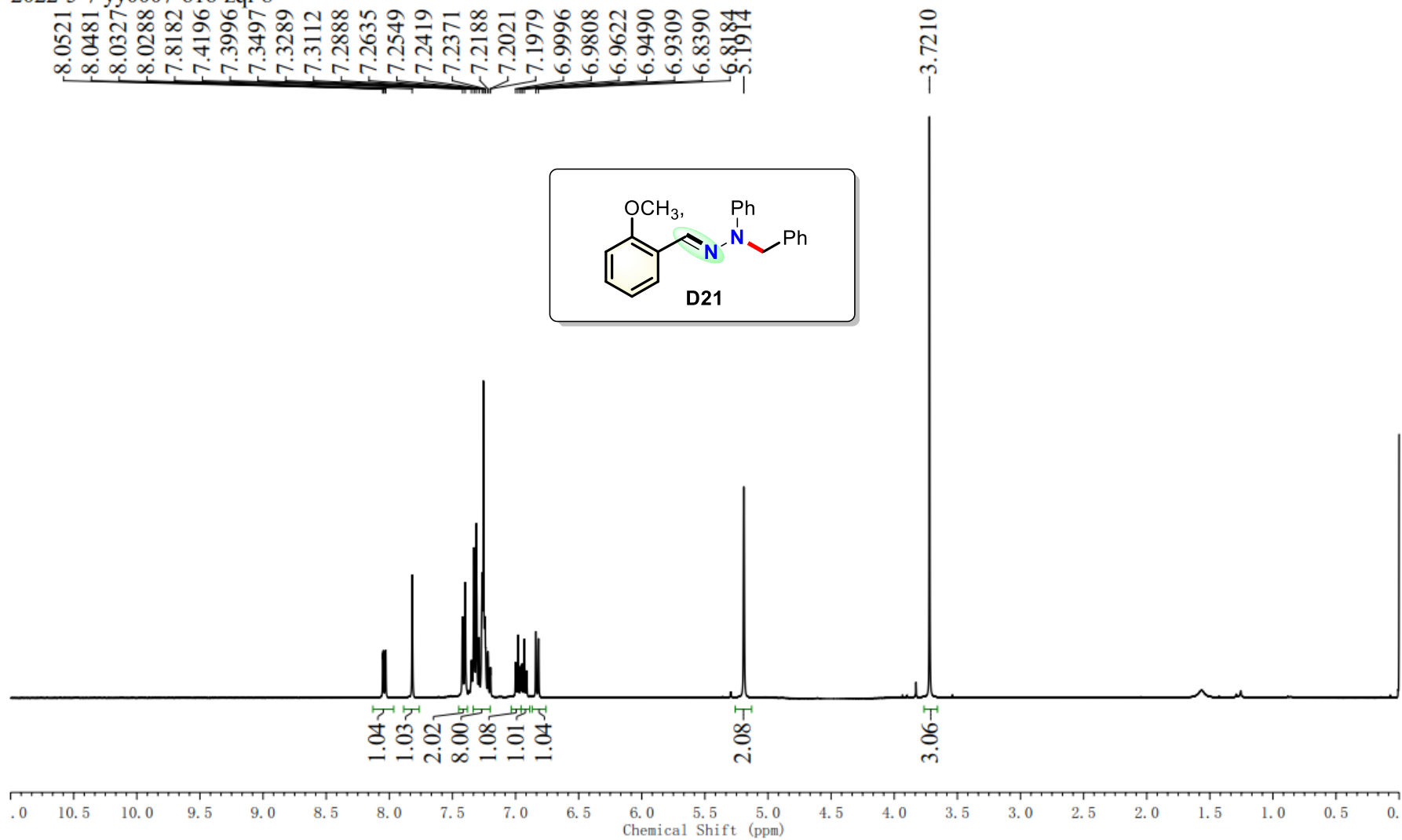
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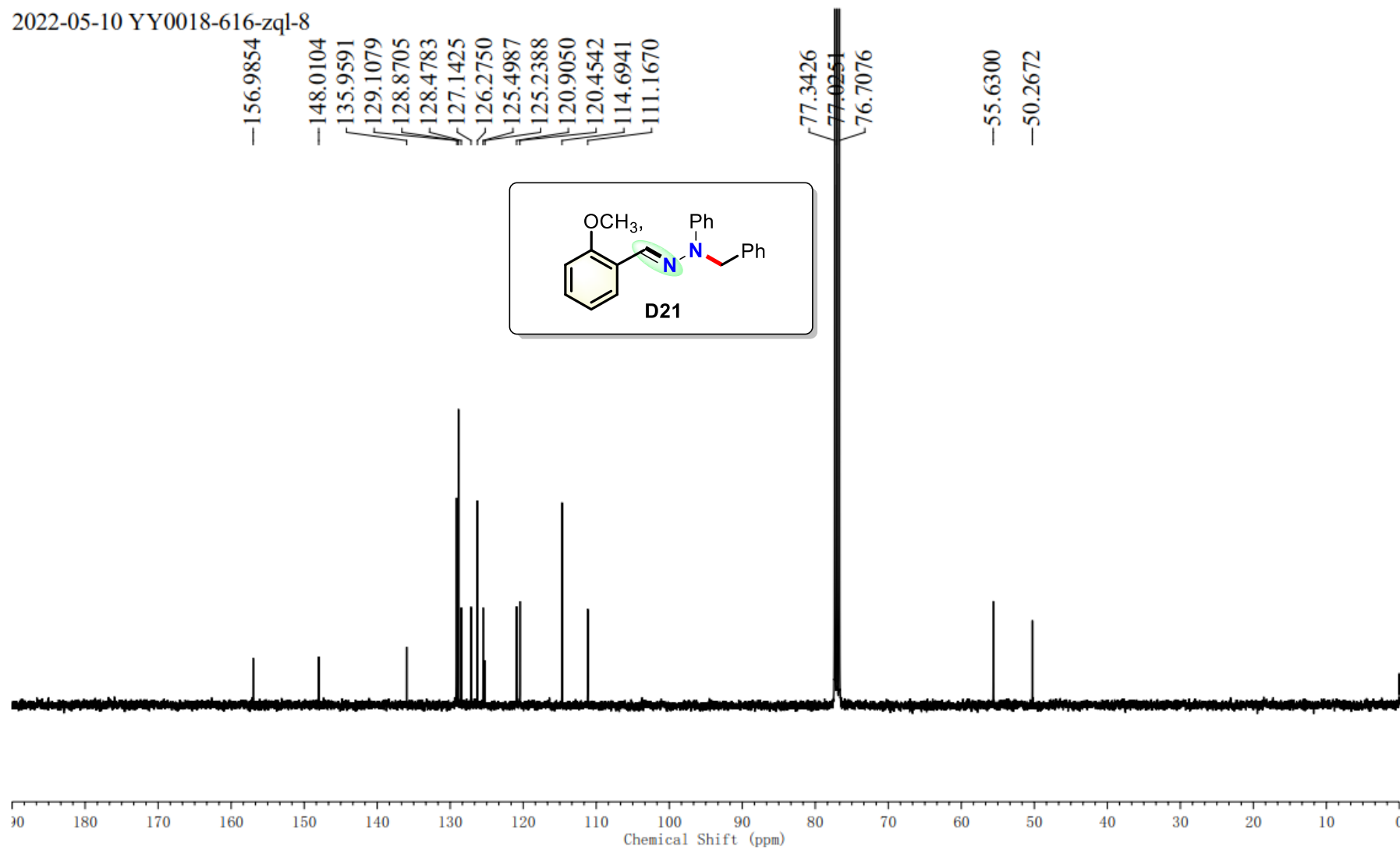
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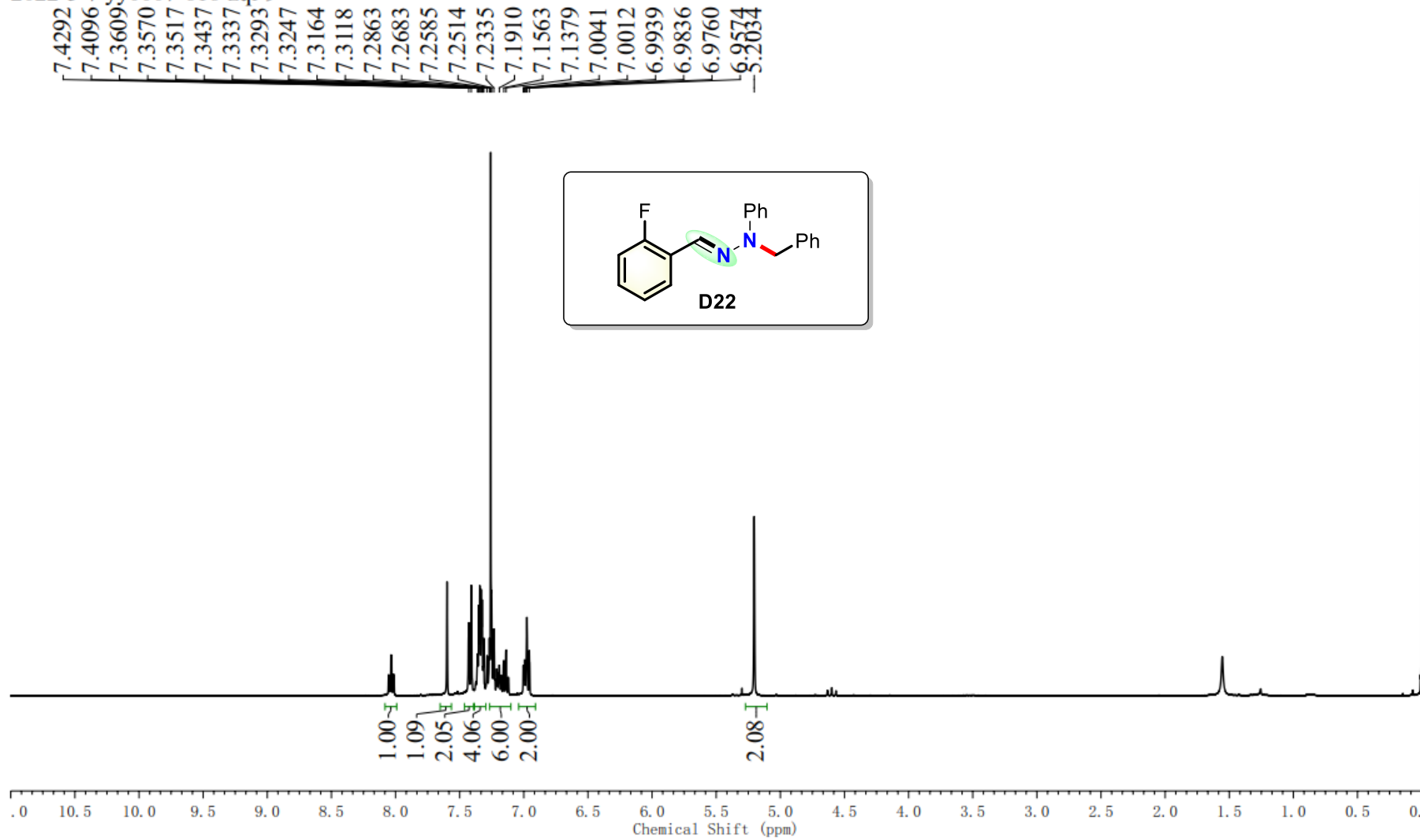
2022-5-7 yy0007-616-zq1-8



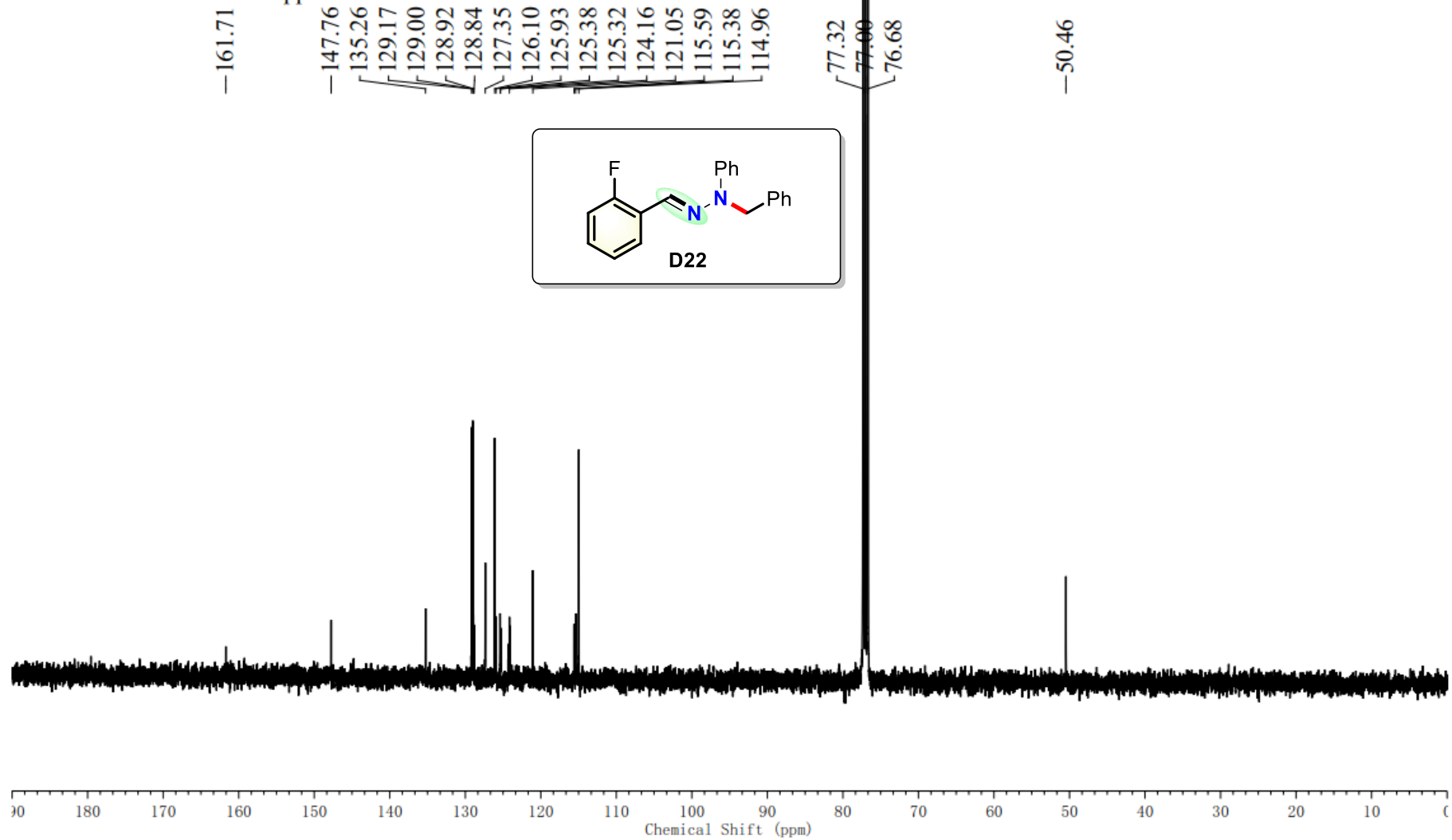
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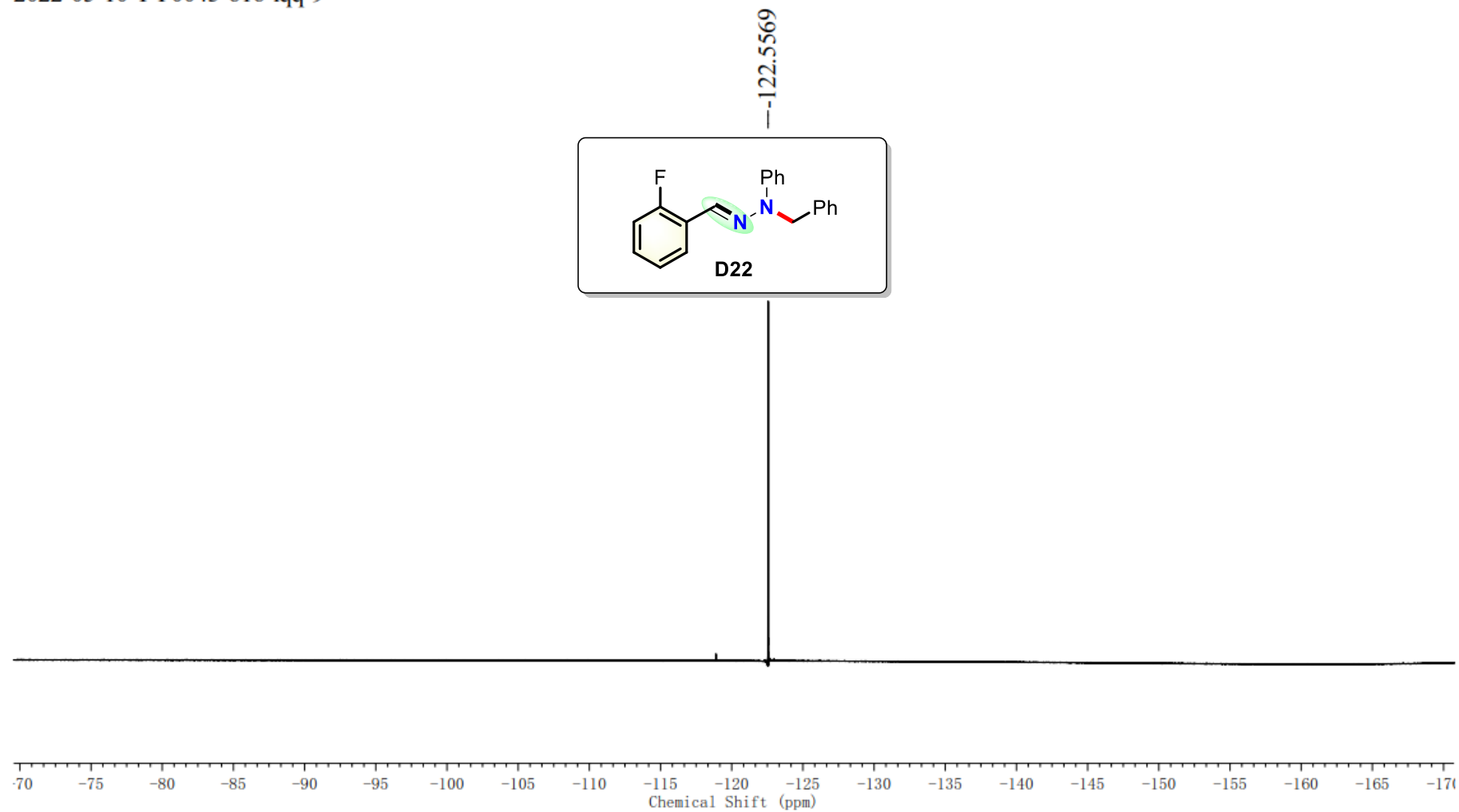
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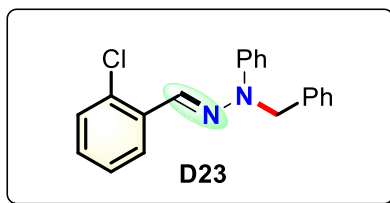
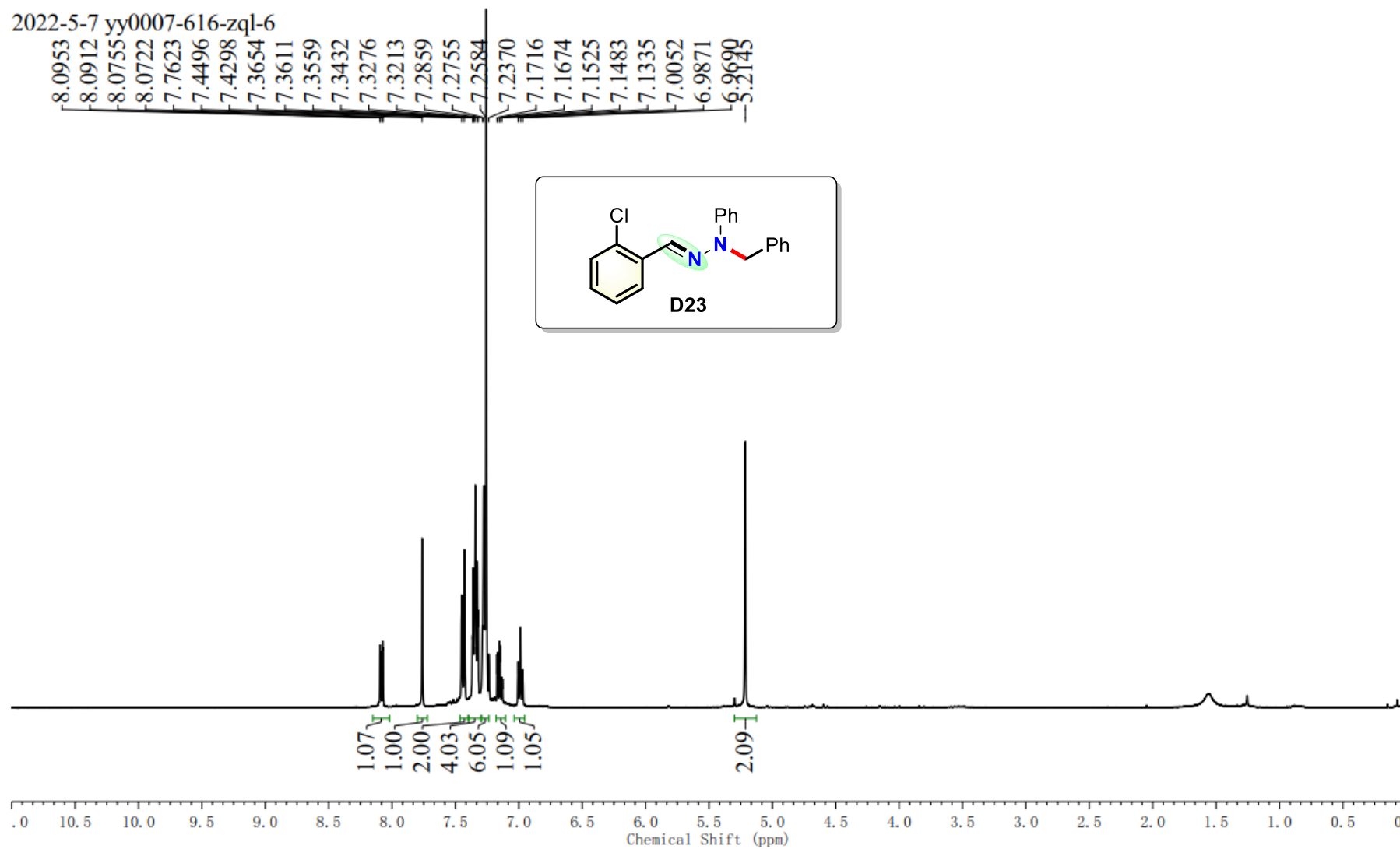
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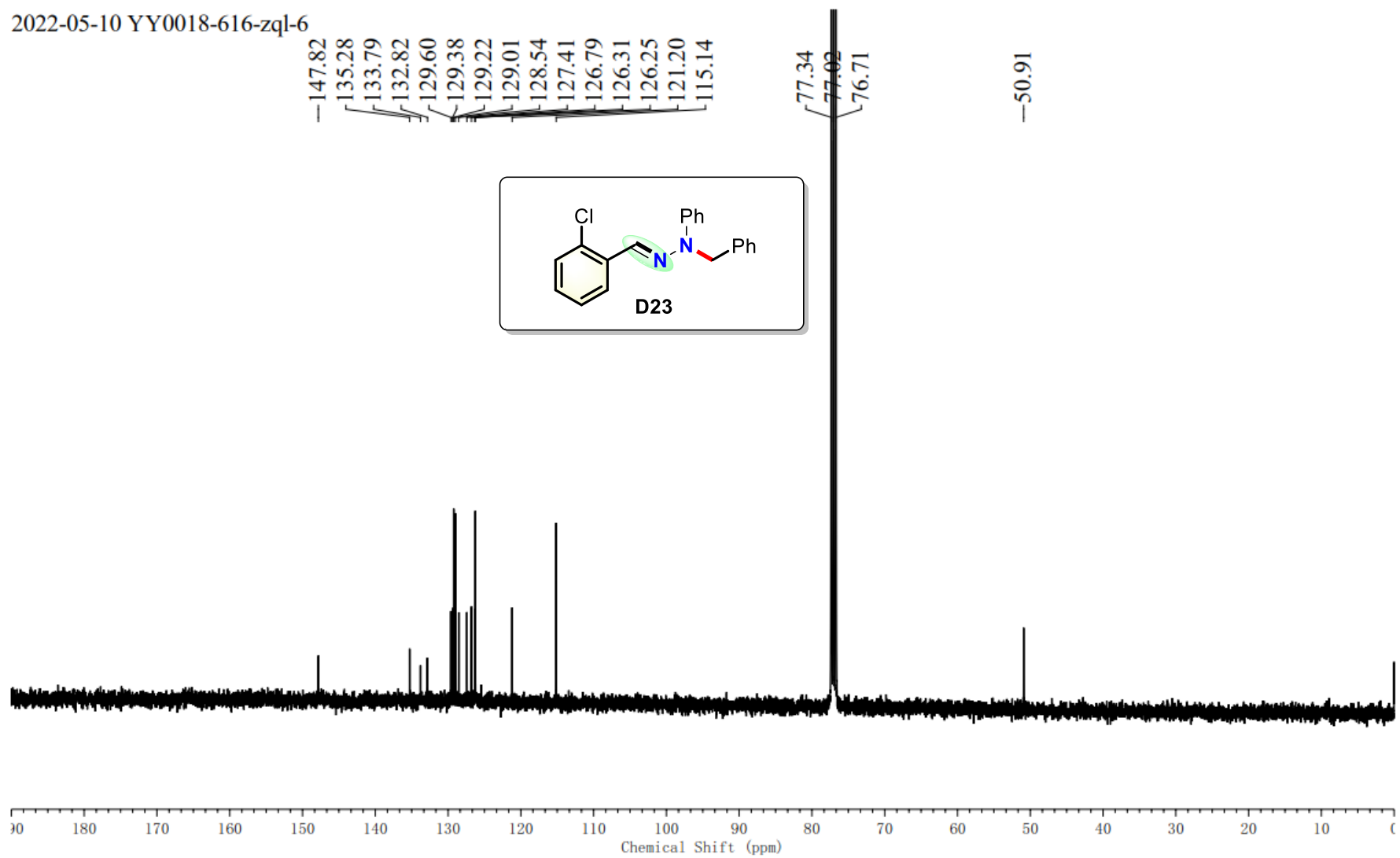
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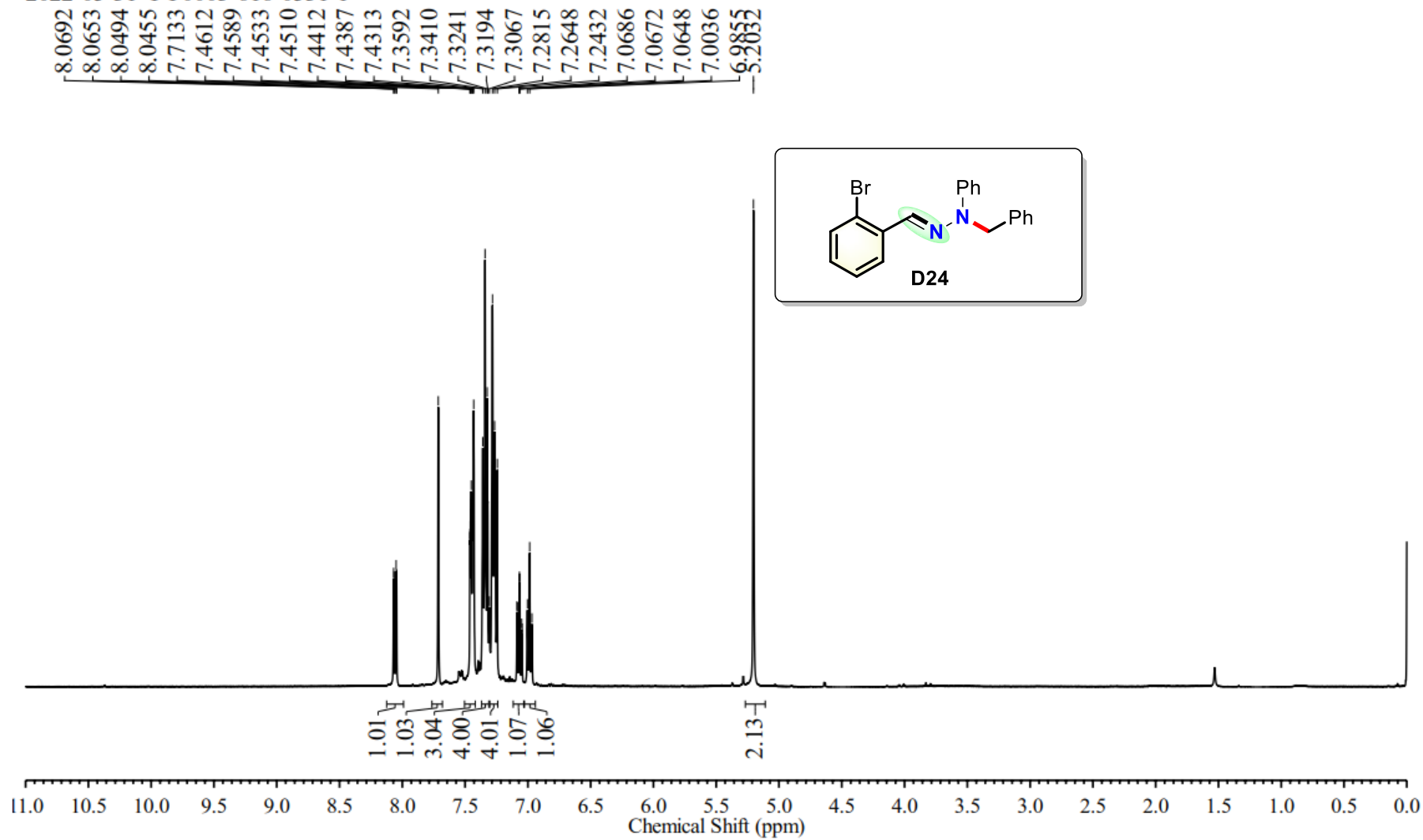
2022-5-7 yy0007-616-zql-6



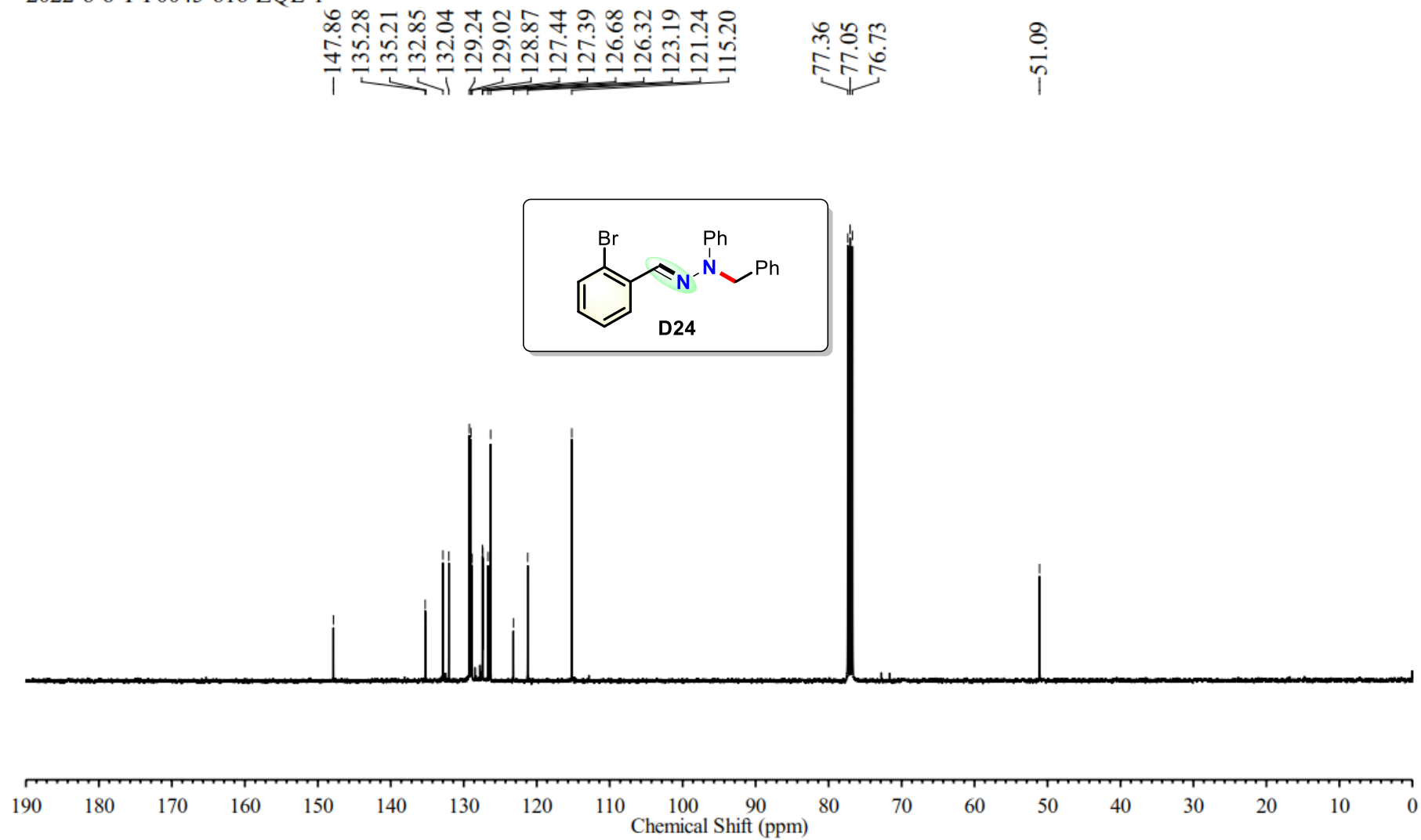
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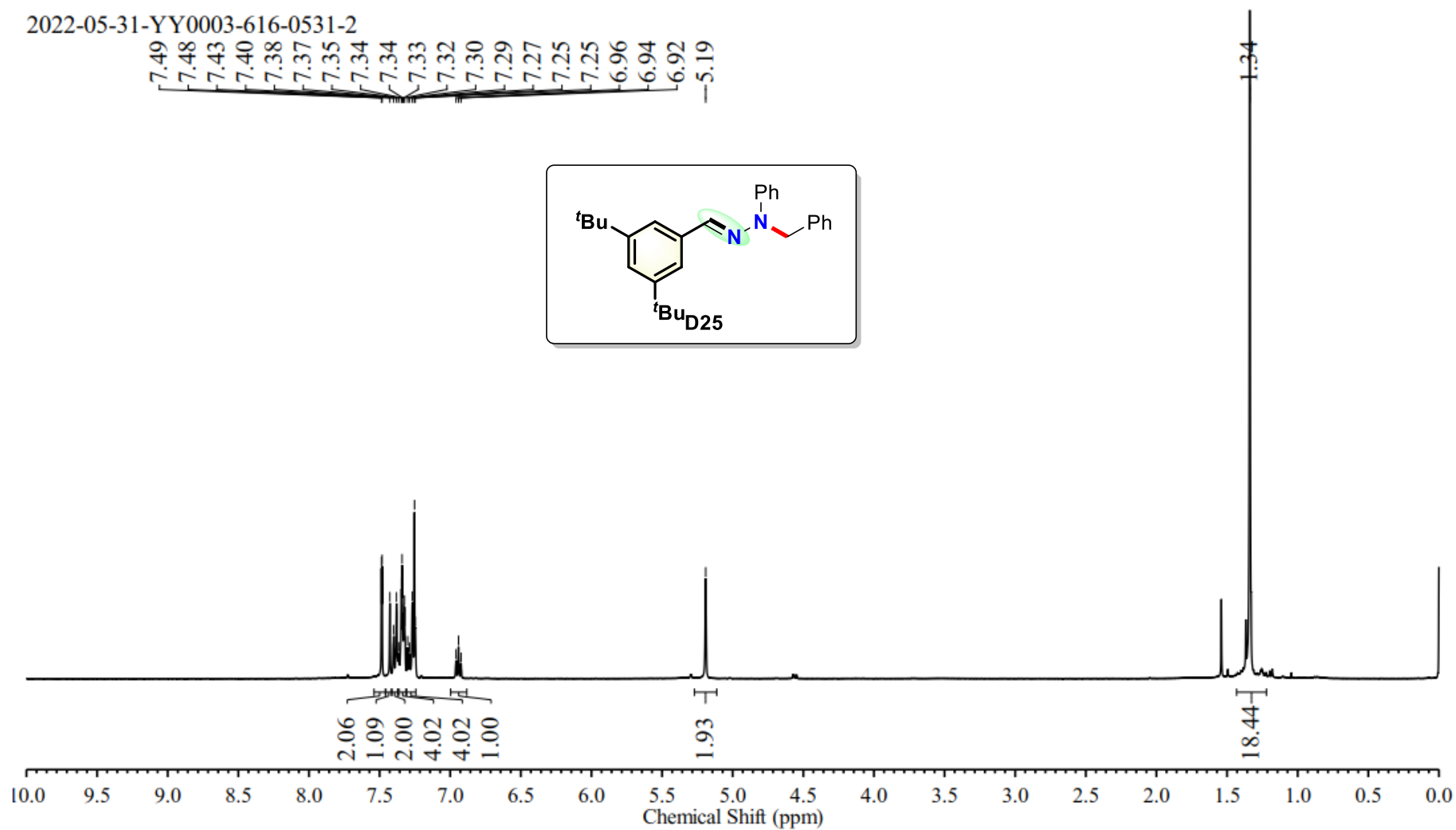
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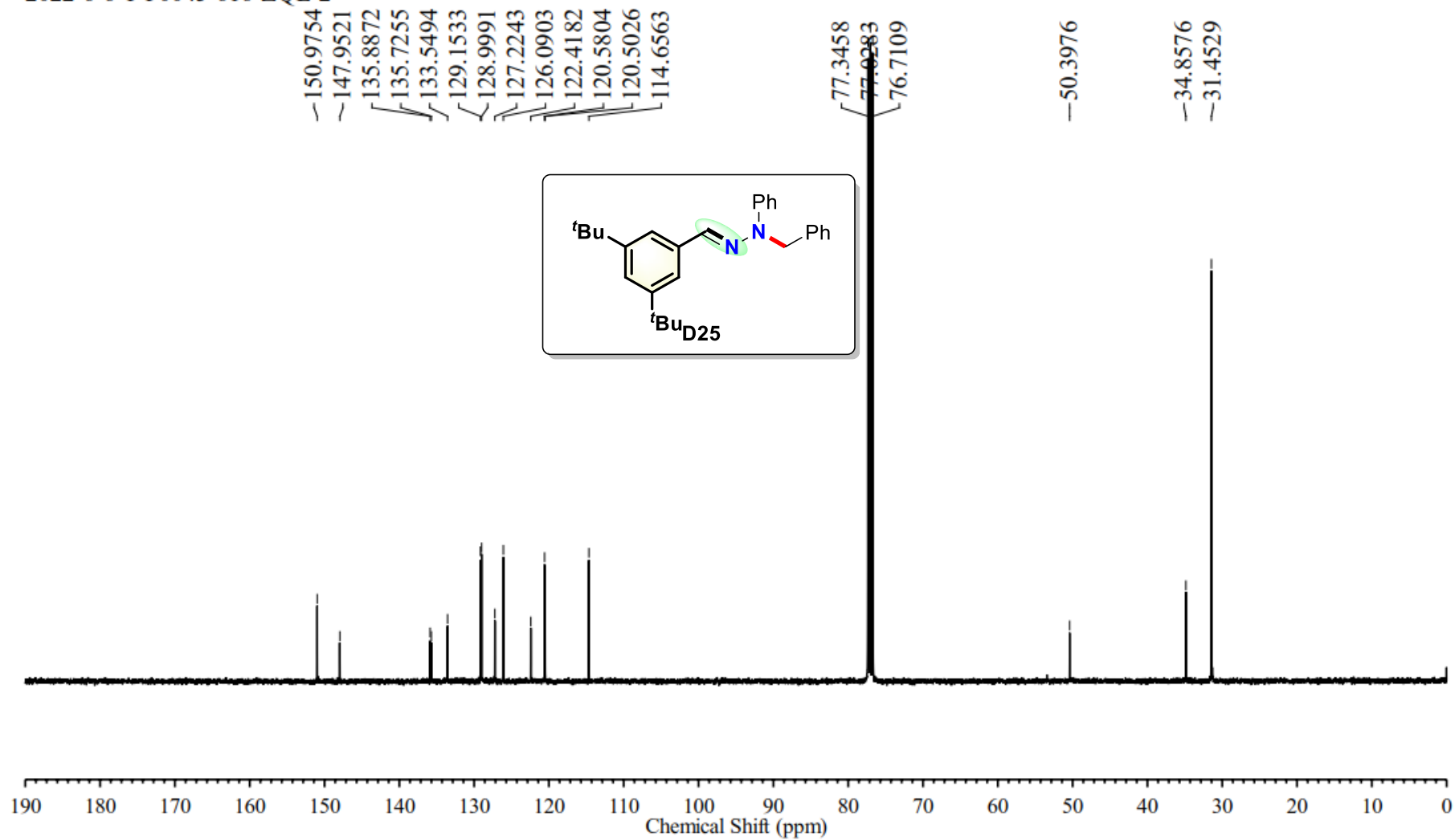
2022-6-6 YY0045-616-ZQL-1



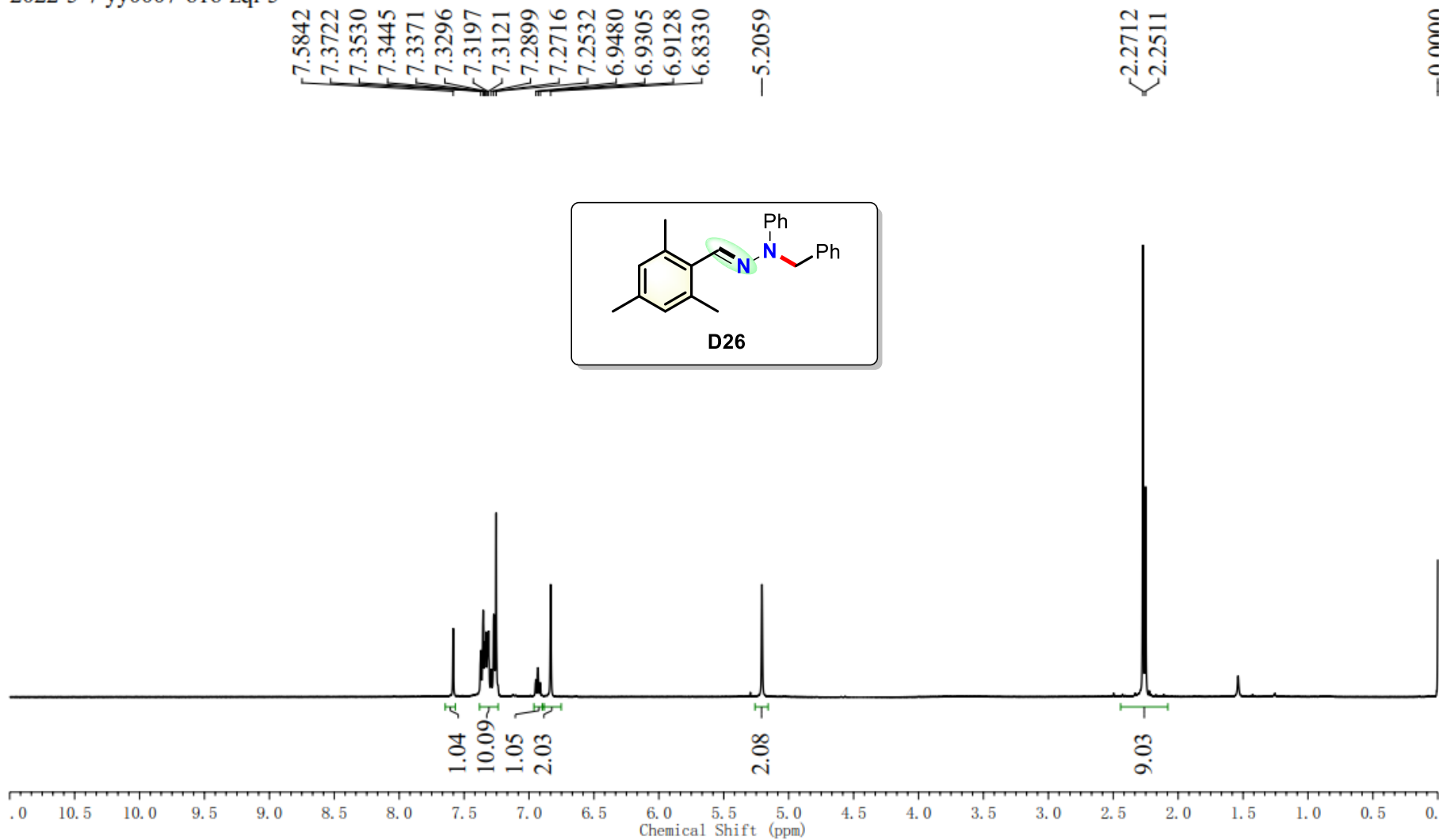
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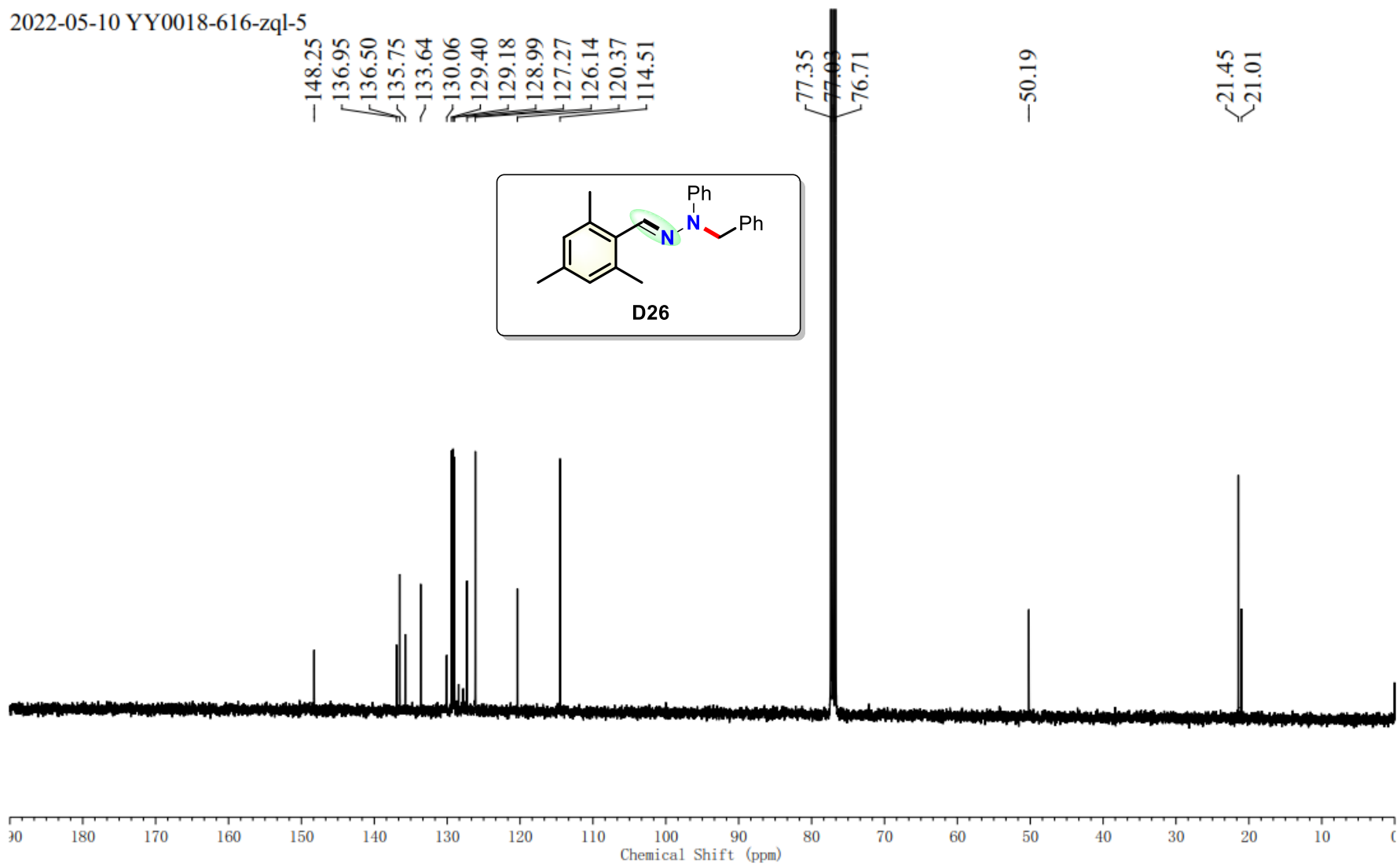
2022-6-6 YY0045-616-ZQL-2



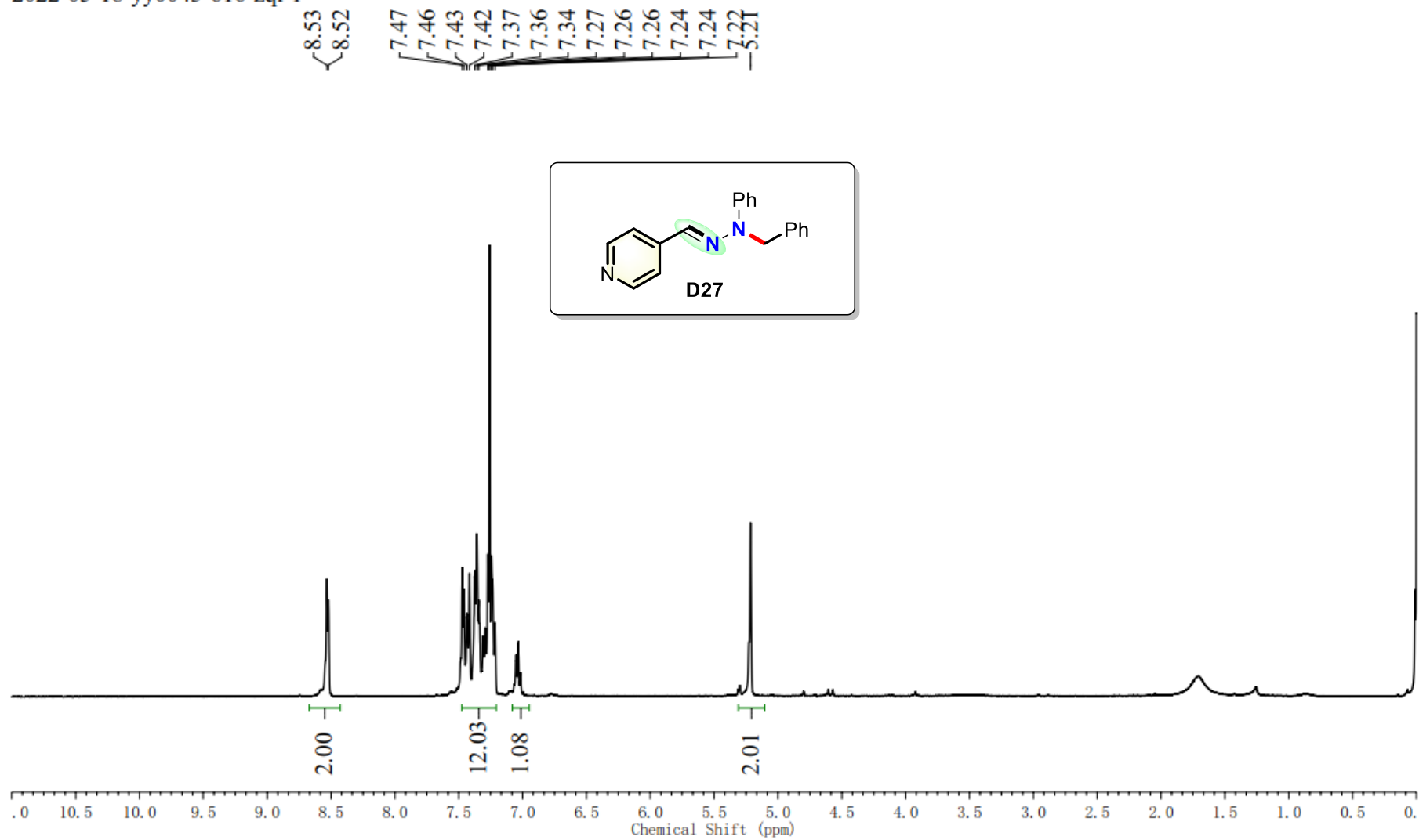
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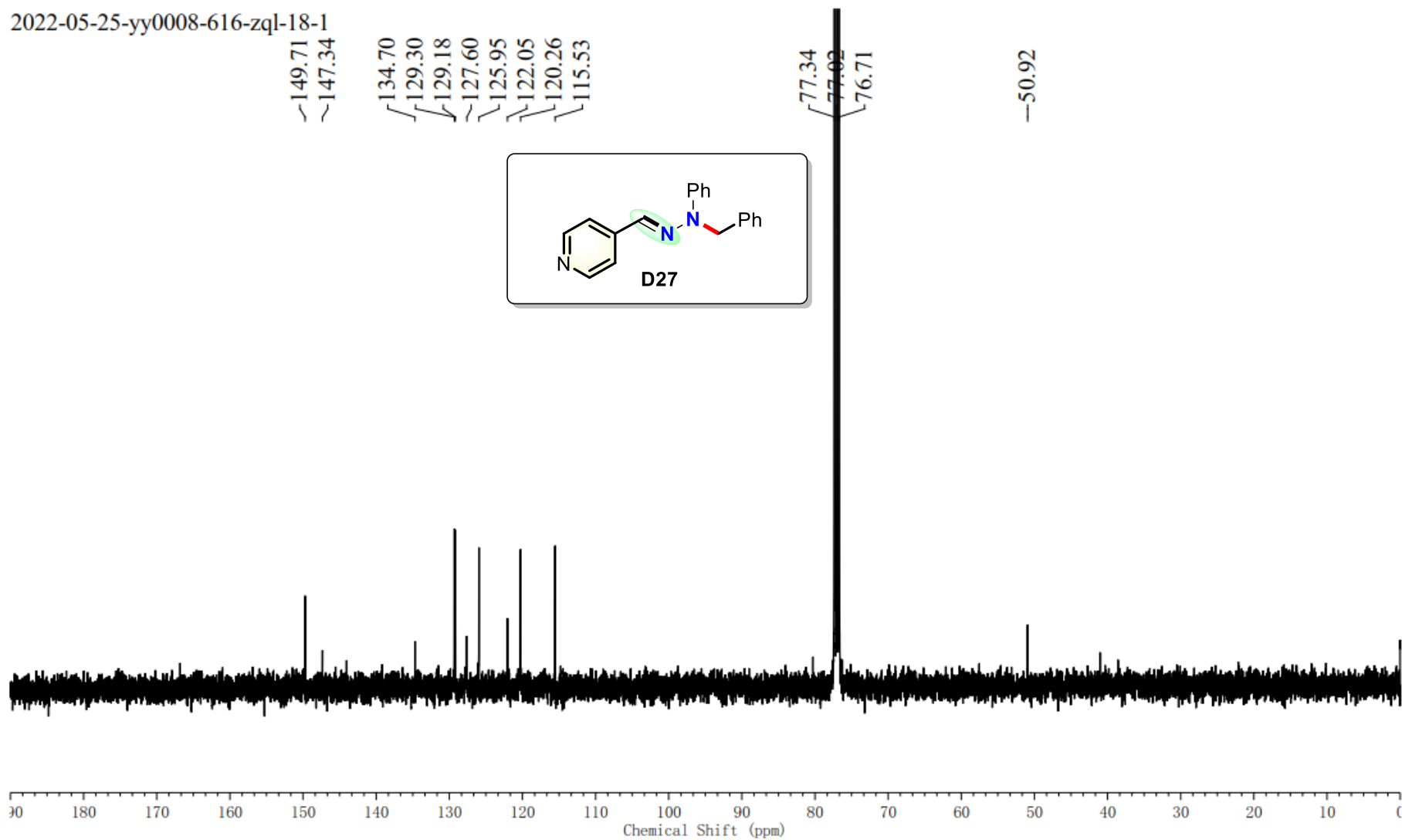
2022-05-10 YY0018-616-zql-5



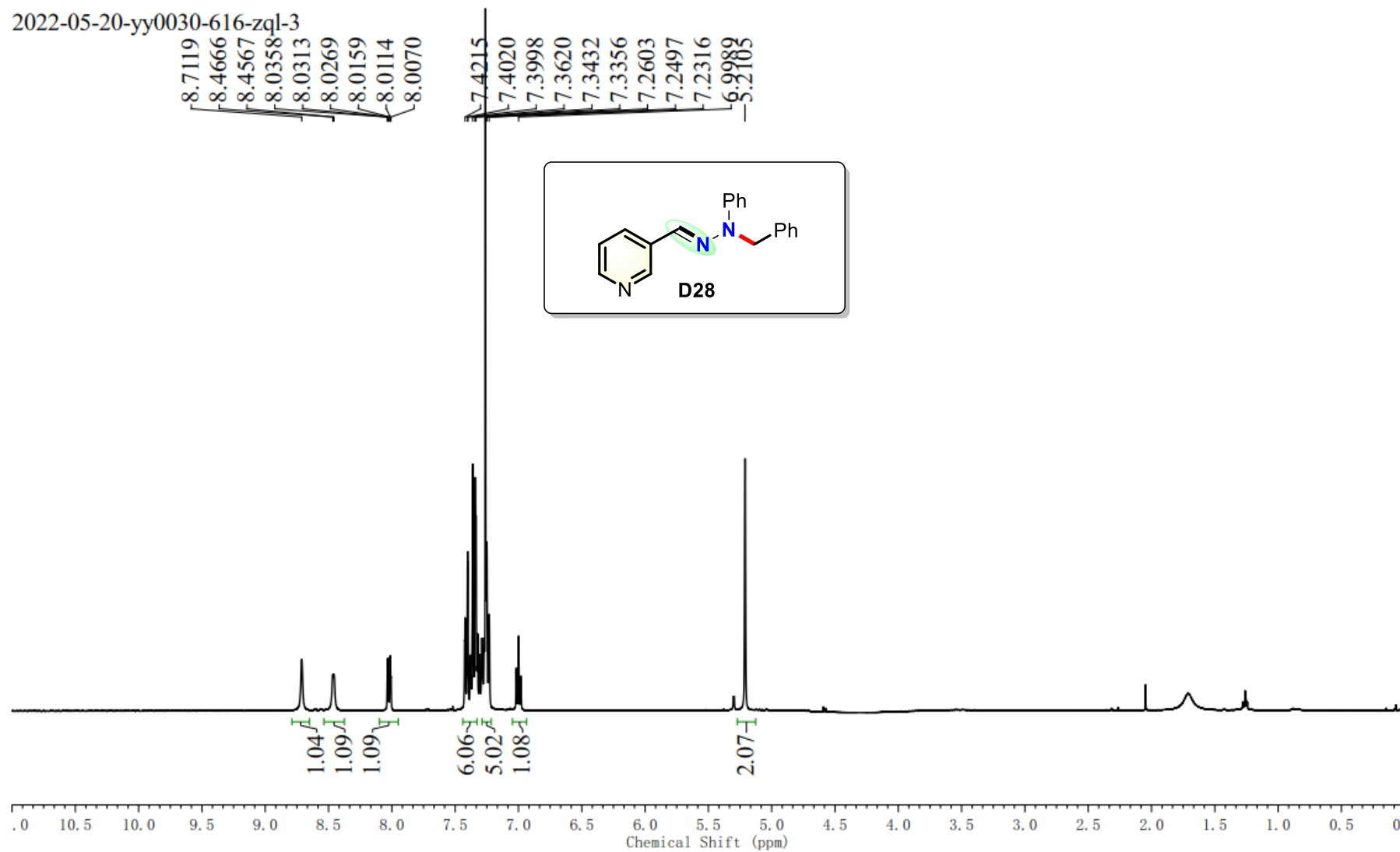
2022-05-18-yy0043-616-zql-1



2022-05-25-yy0008-616-zql-18-1



2022-05-20-yy0030-616-zql-3

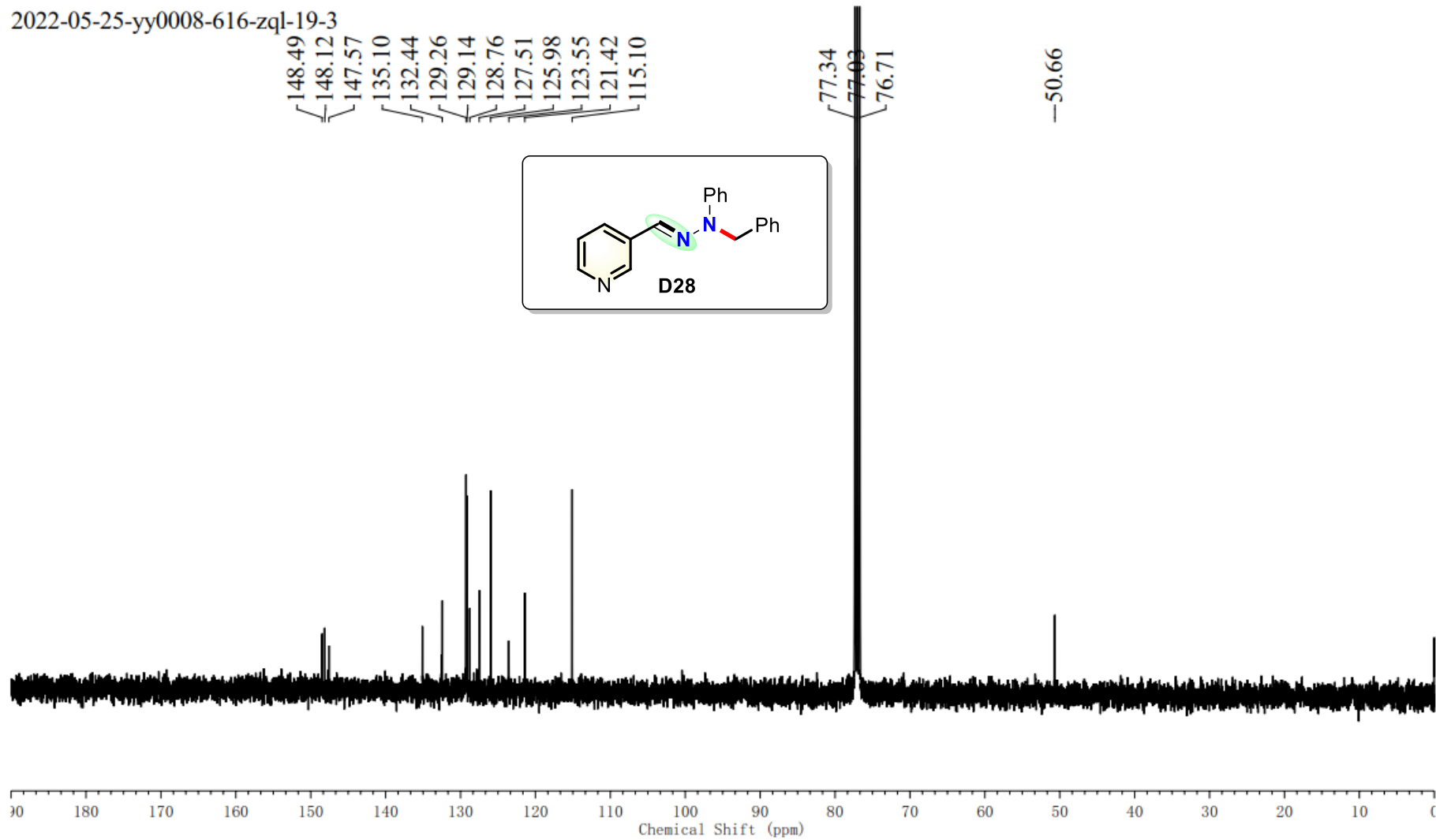
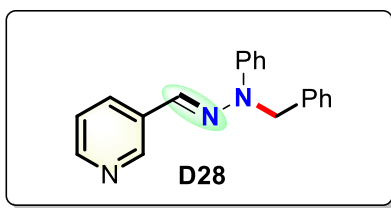


2022-05-25-yy0008-616-zql-19-3

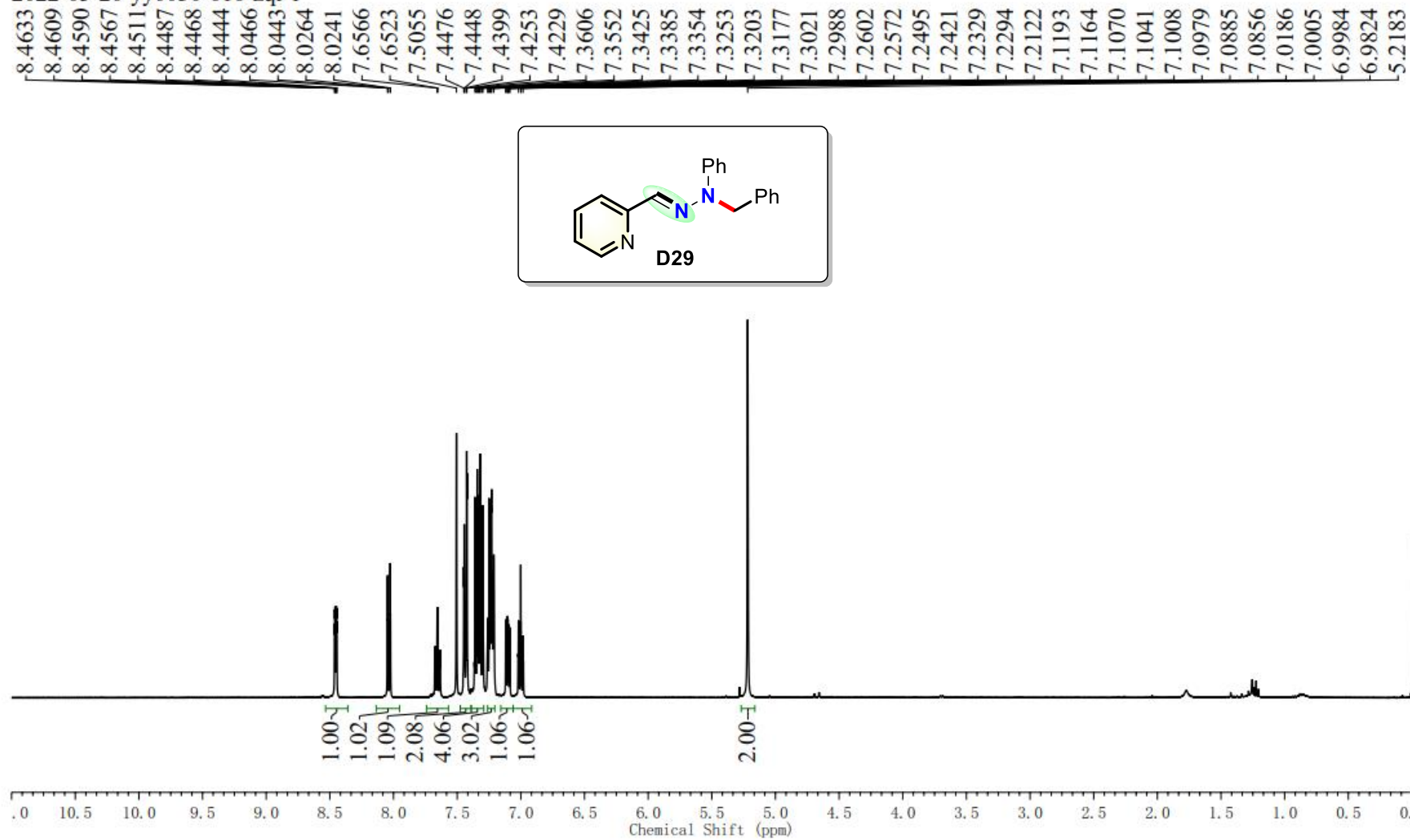
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148.12
147.57
135.10
132.44
129.26
129.14
128.76
127.51
125.98
123.55
121.42
115.10

77.34
77.03
76.71

—50.66



2022-05-20-yy0030-616-zql-1

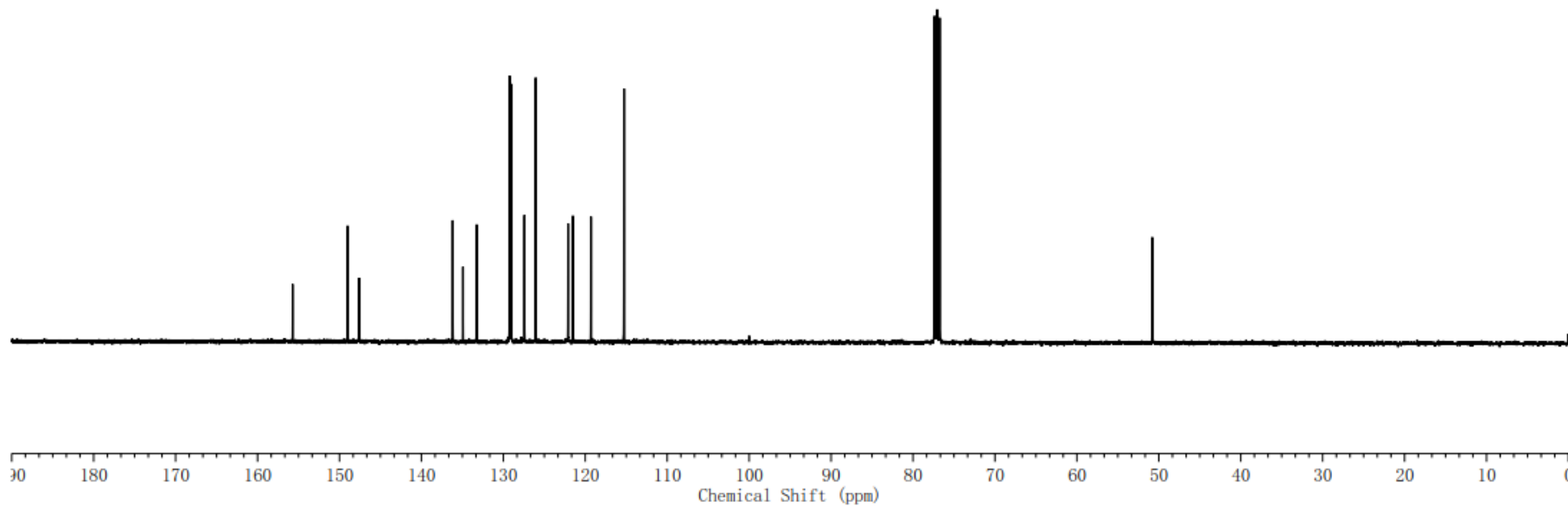
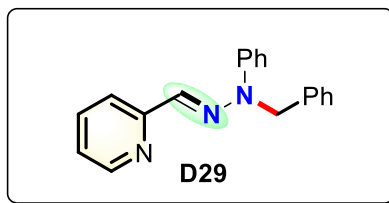


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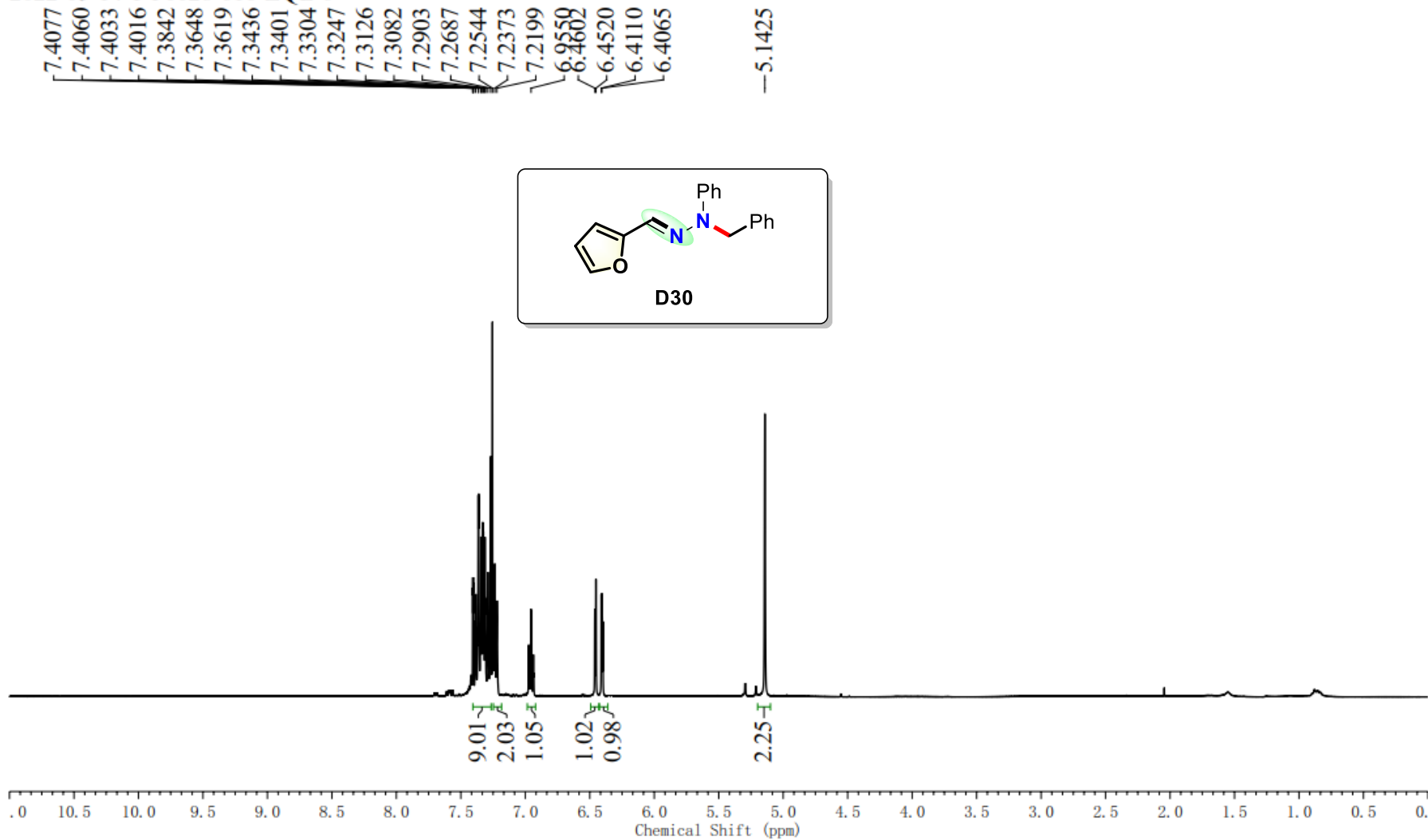
155.72
149.00
147.59
136.17
134.92
133.25
129.24
129.05
127.42
126.05
122.10
121.51
119.31
115.27

77.38
77.06
76.74

50.80



2022-05-14 YY0028-616-ZQL-6

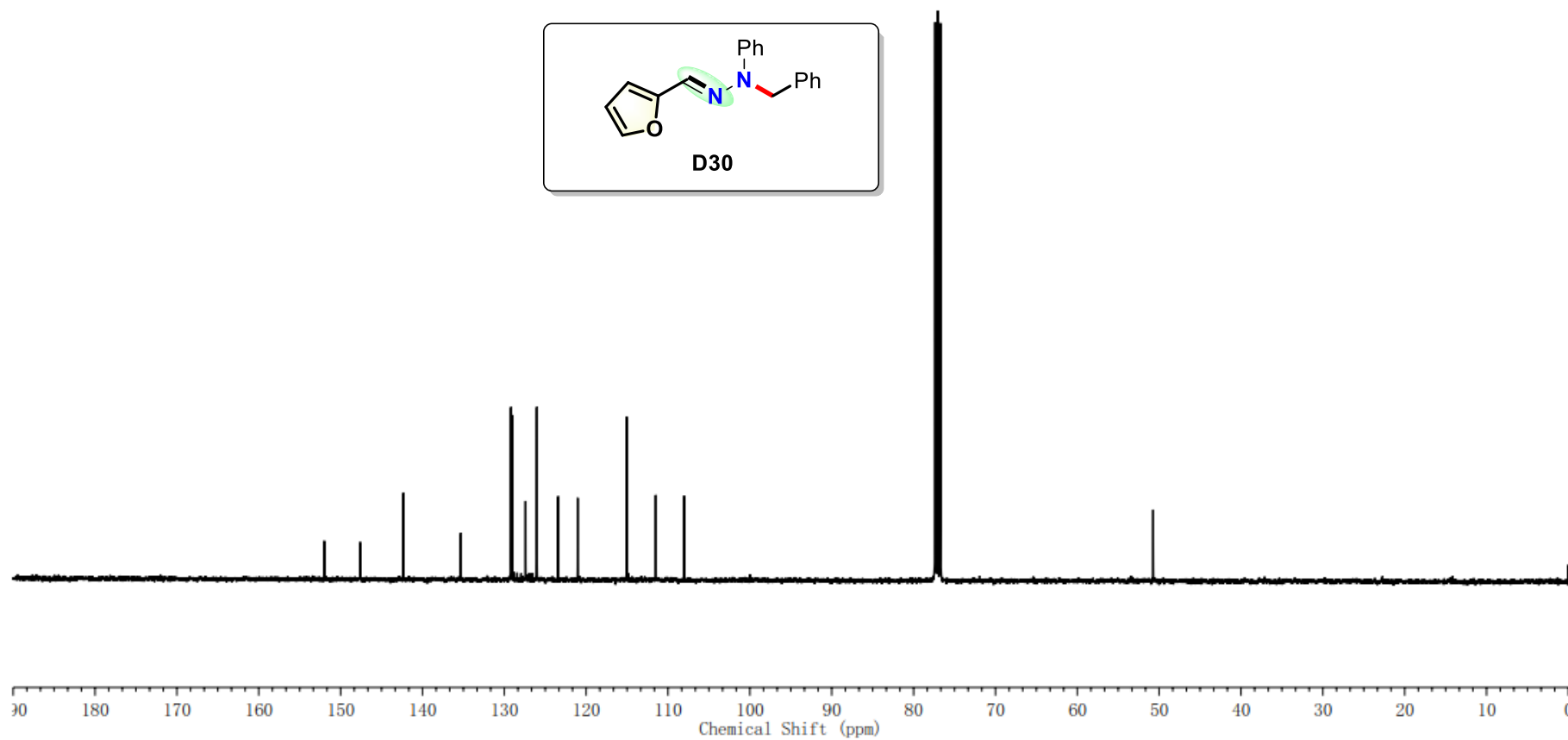
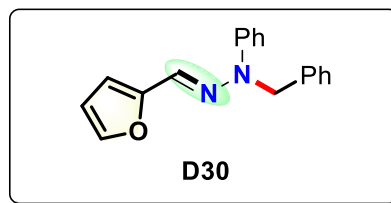


2022-05-16-yy0016-zql-6

151.99
147.61
142.33
135.31
129.18
129.06
127.40
126.02
123.42
121.04
115.06
111.49
108.03

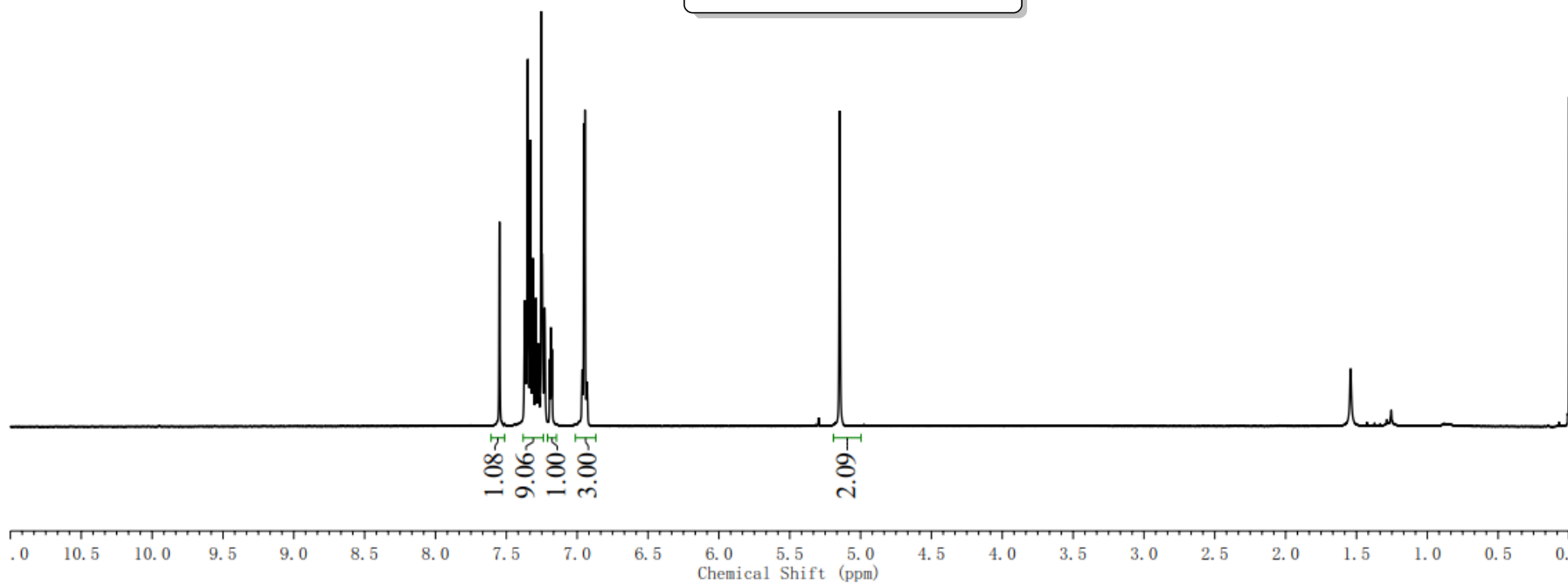
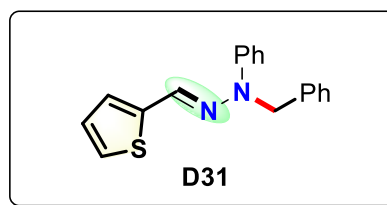
77.35
77.03
76.72

50.72

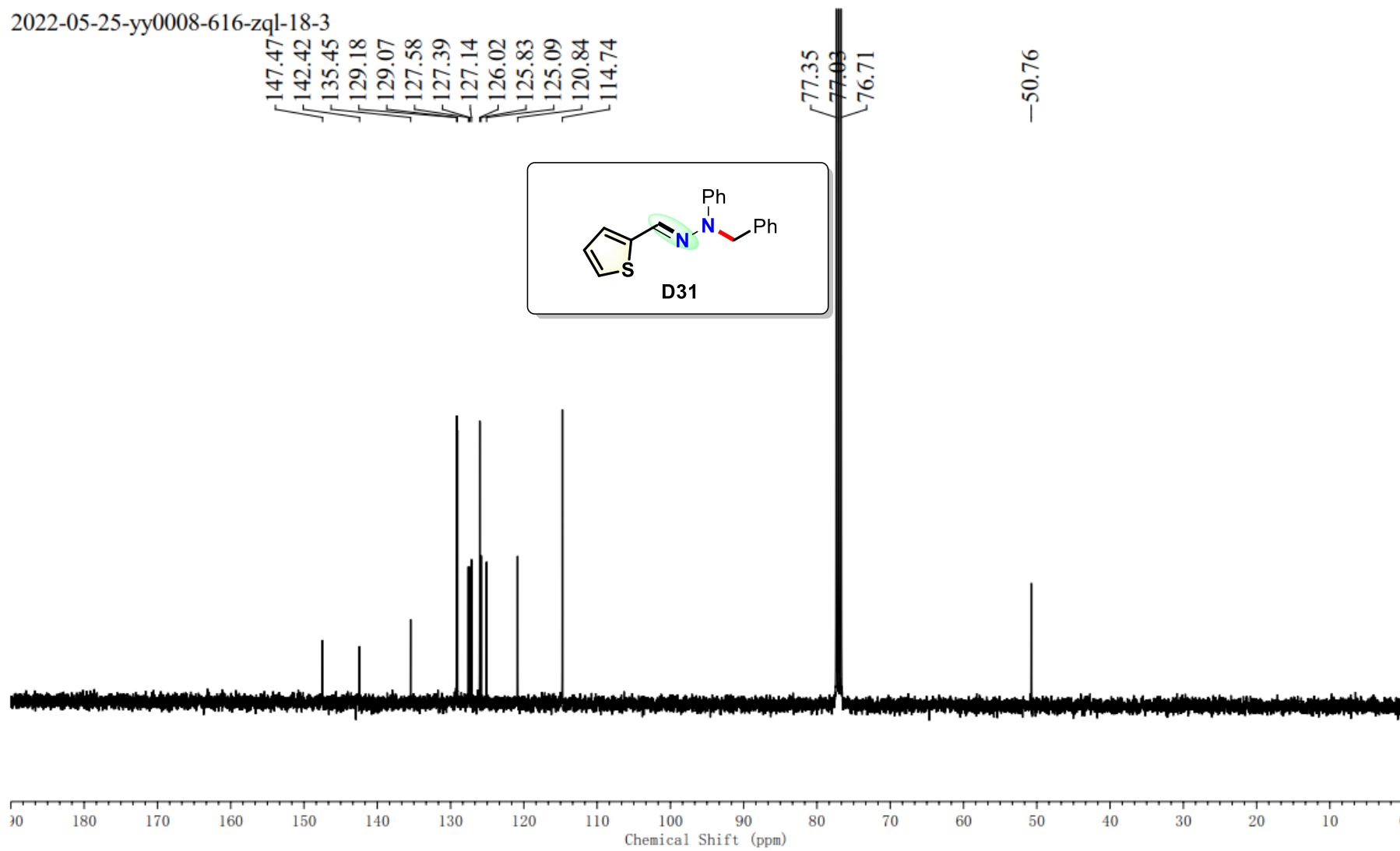


2022-05-18-yy0043-616-zql-3

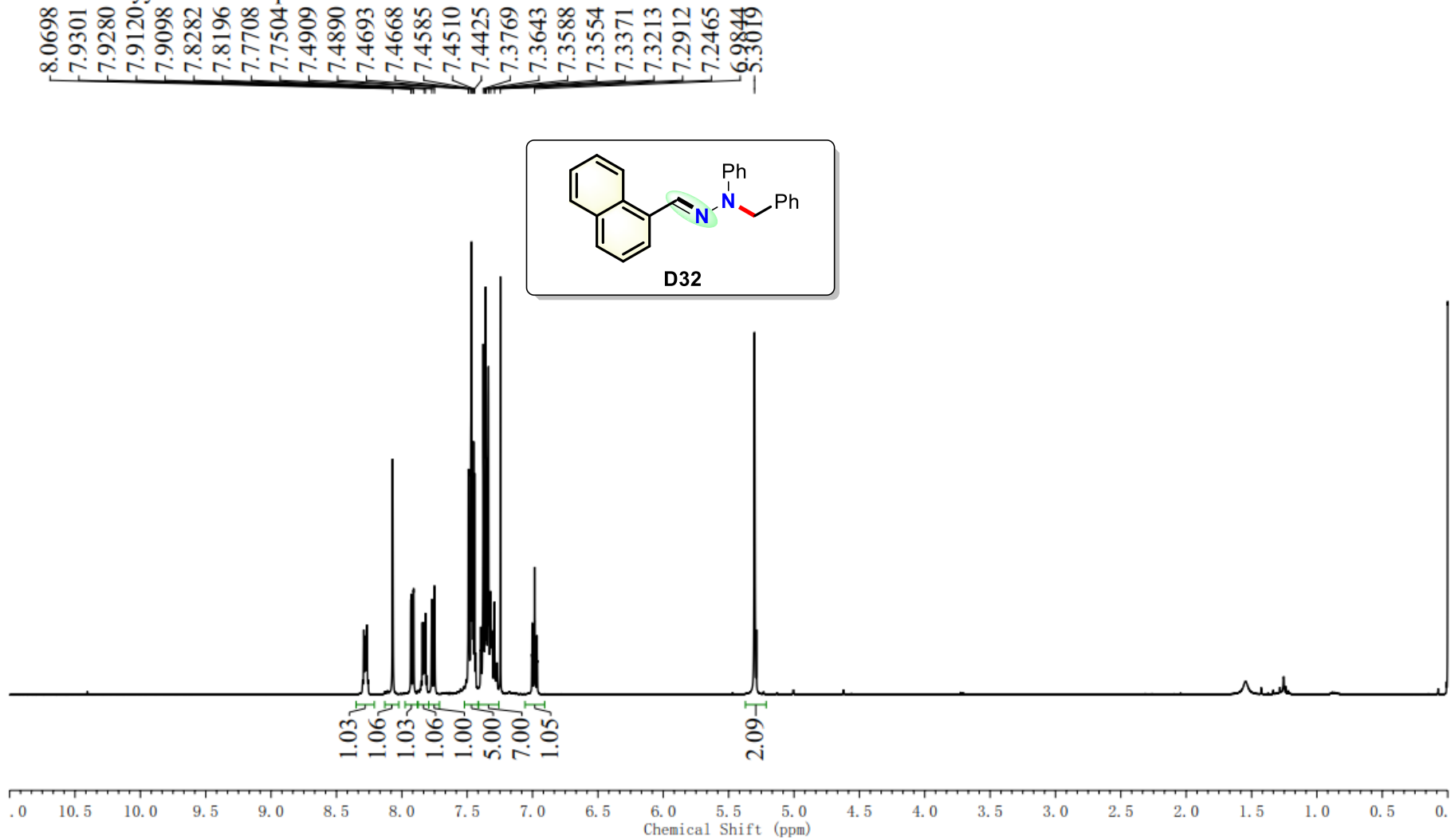
7.5485
7.3695
7.3486
7.3302
7.3129
7.2913
7.2746
7.2550
7.2480
7.2294
7.1934
7.1855
7.1775
6.9645
6.9528
6.9451
6.9300
— 5.1474



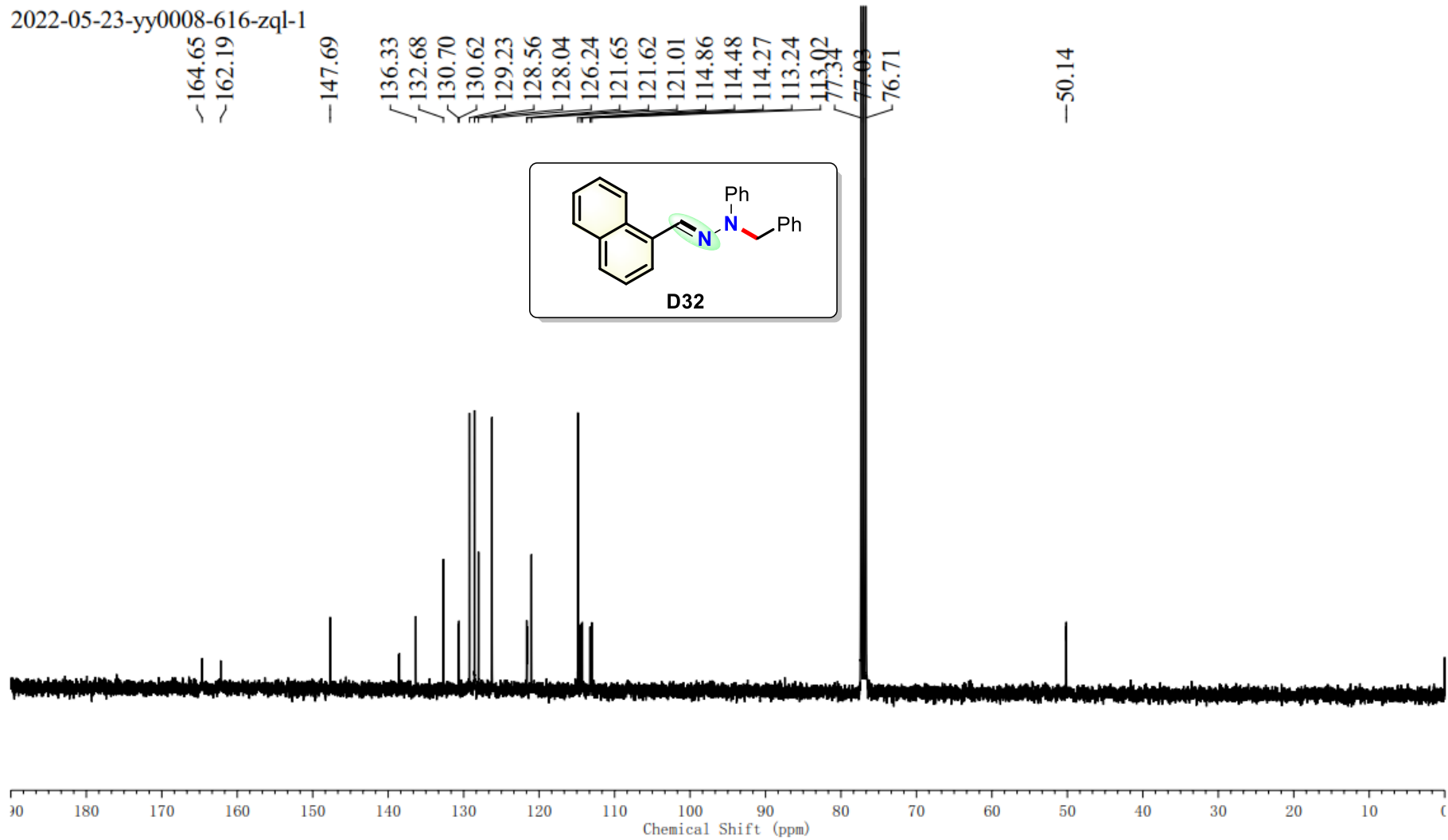
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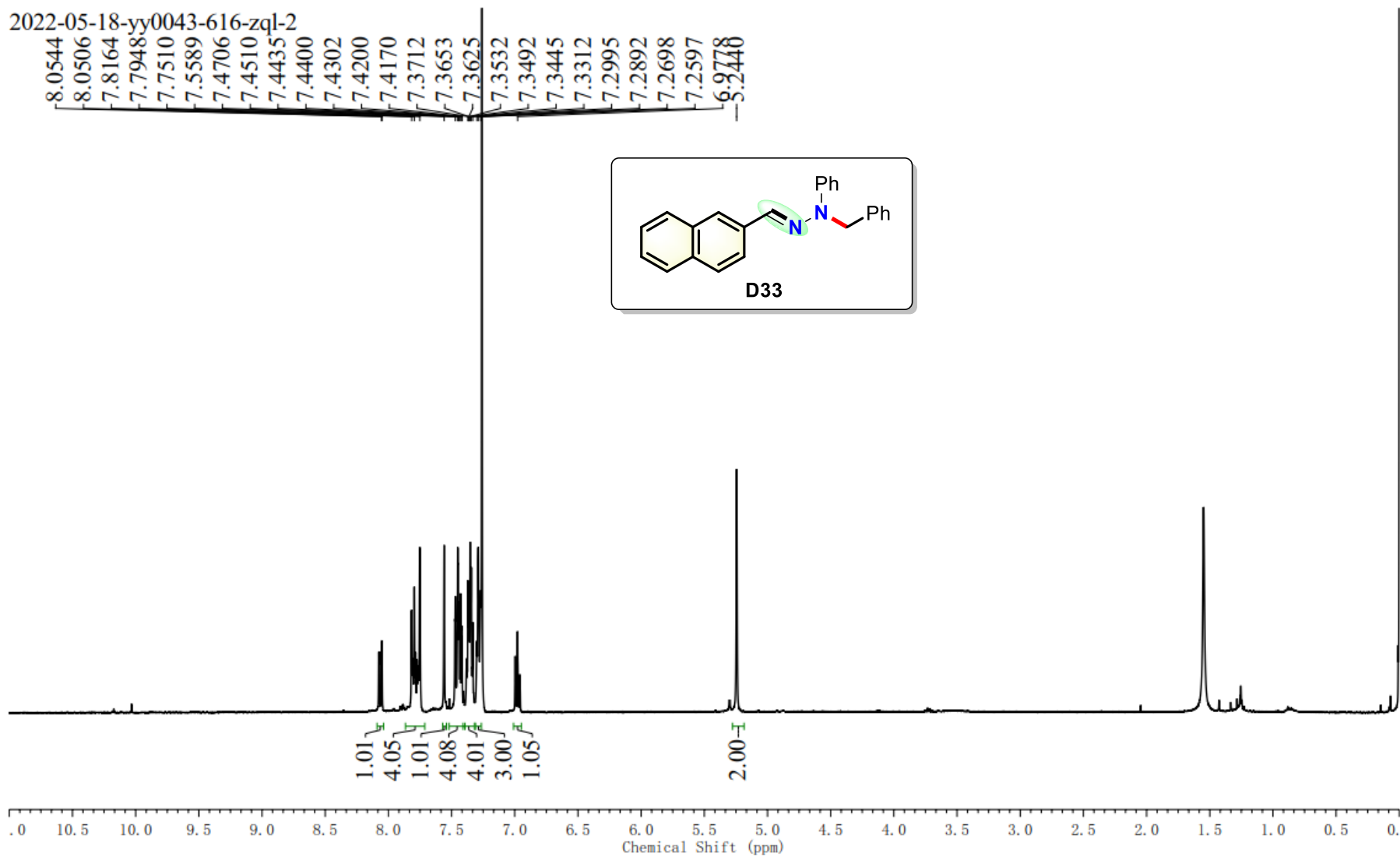
2022-05-20-yy0025-616-zql-1



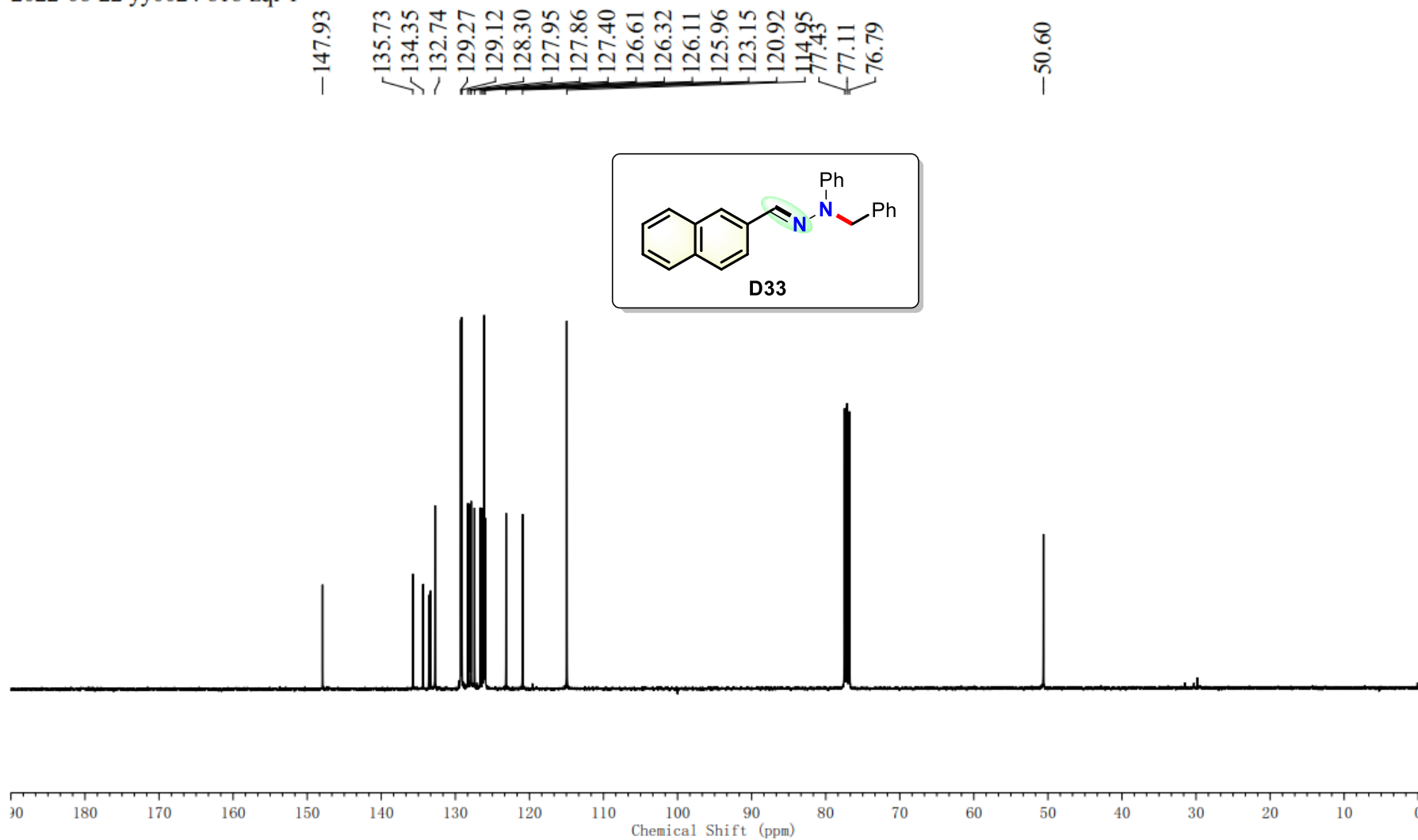
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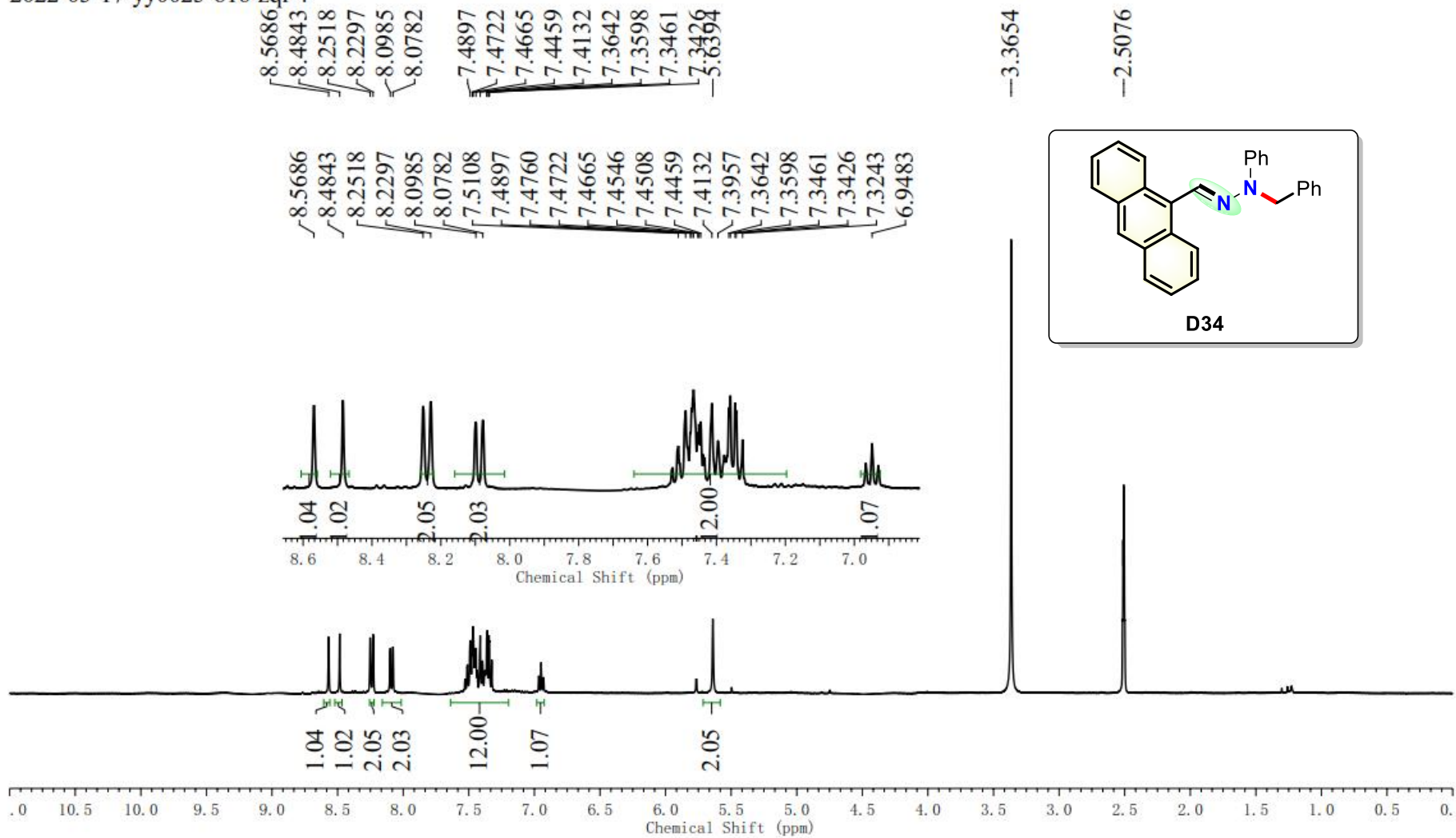
2022-05-18-yy0043-616-zq1-2



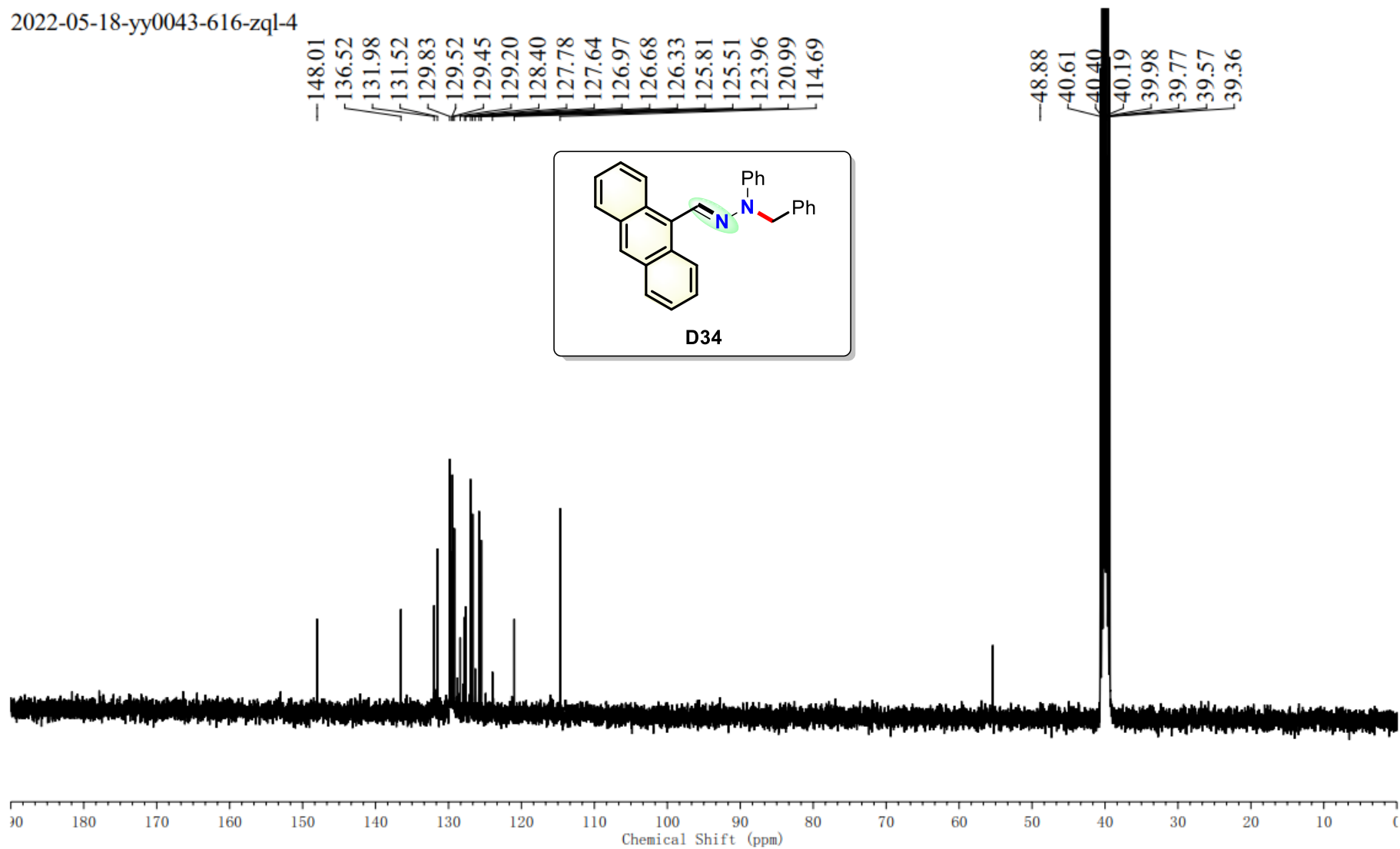
2022-06-22 yy0024 616-zql-1



2022-05-17-yy0025-616-zql-4

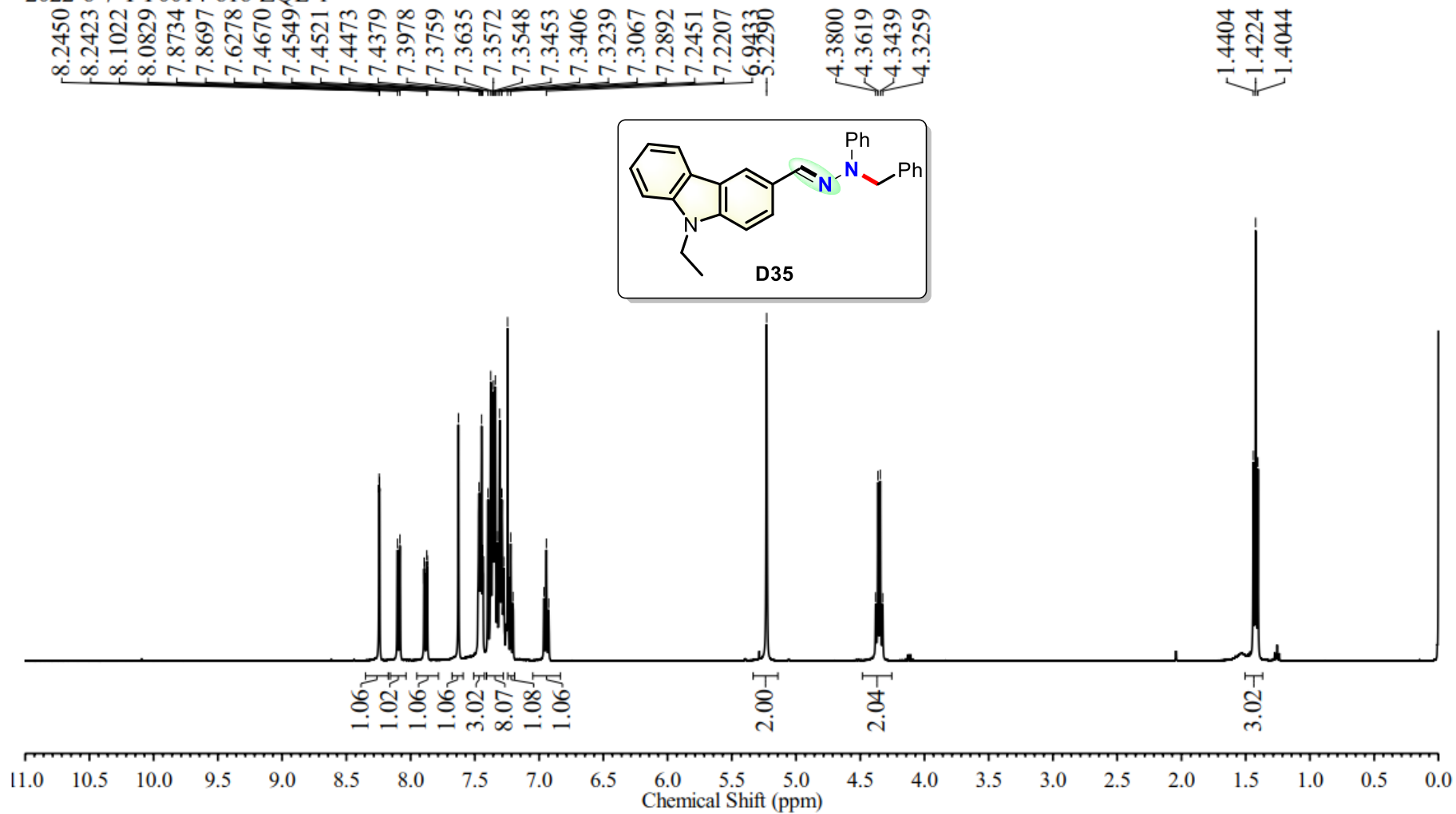


2022-05-18-yy0043-616-zql-4

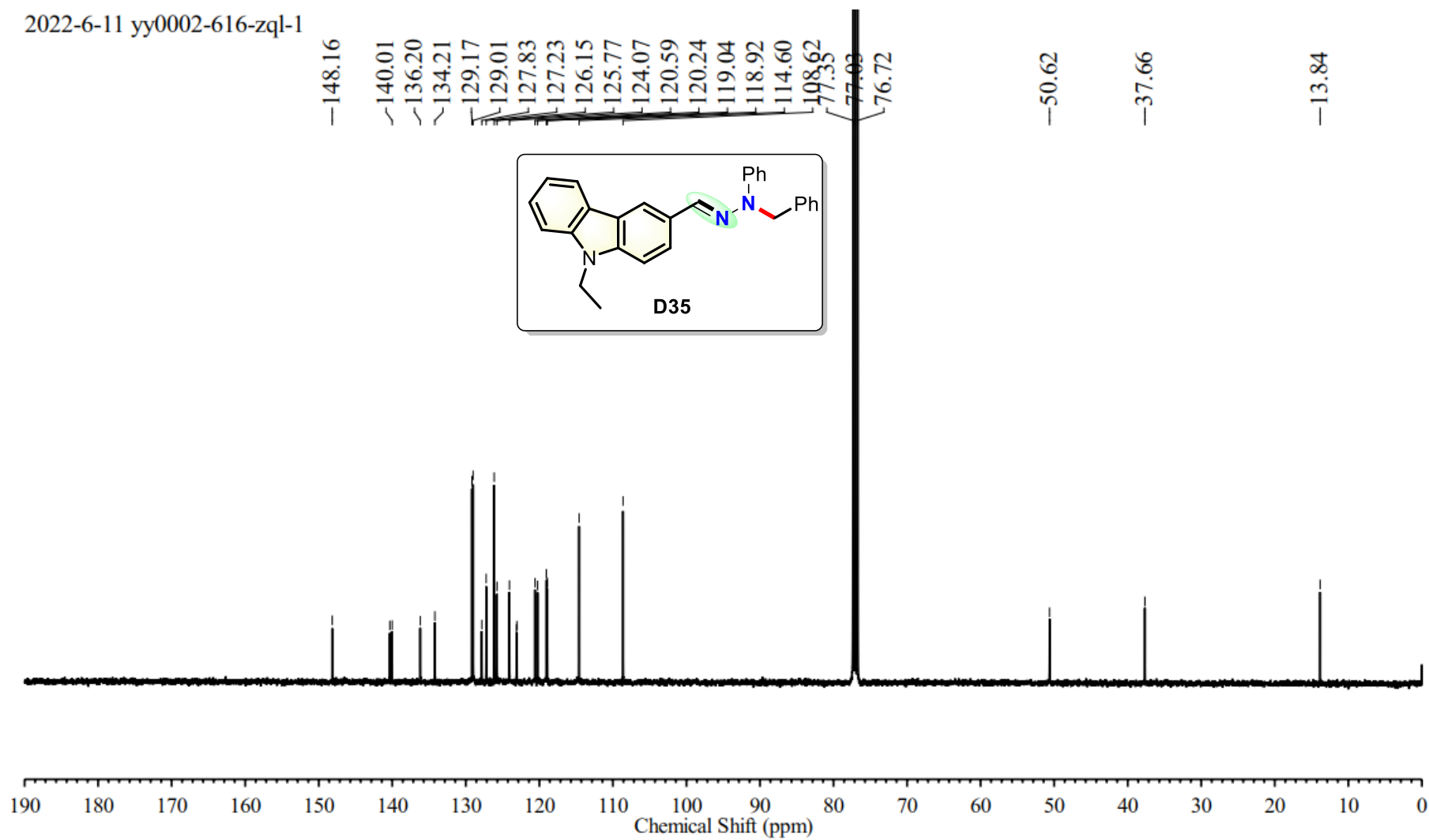


S210

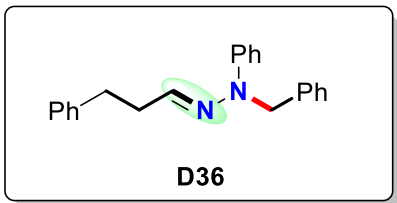
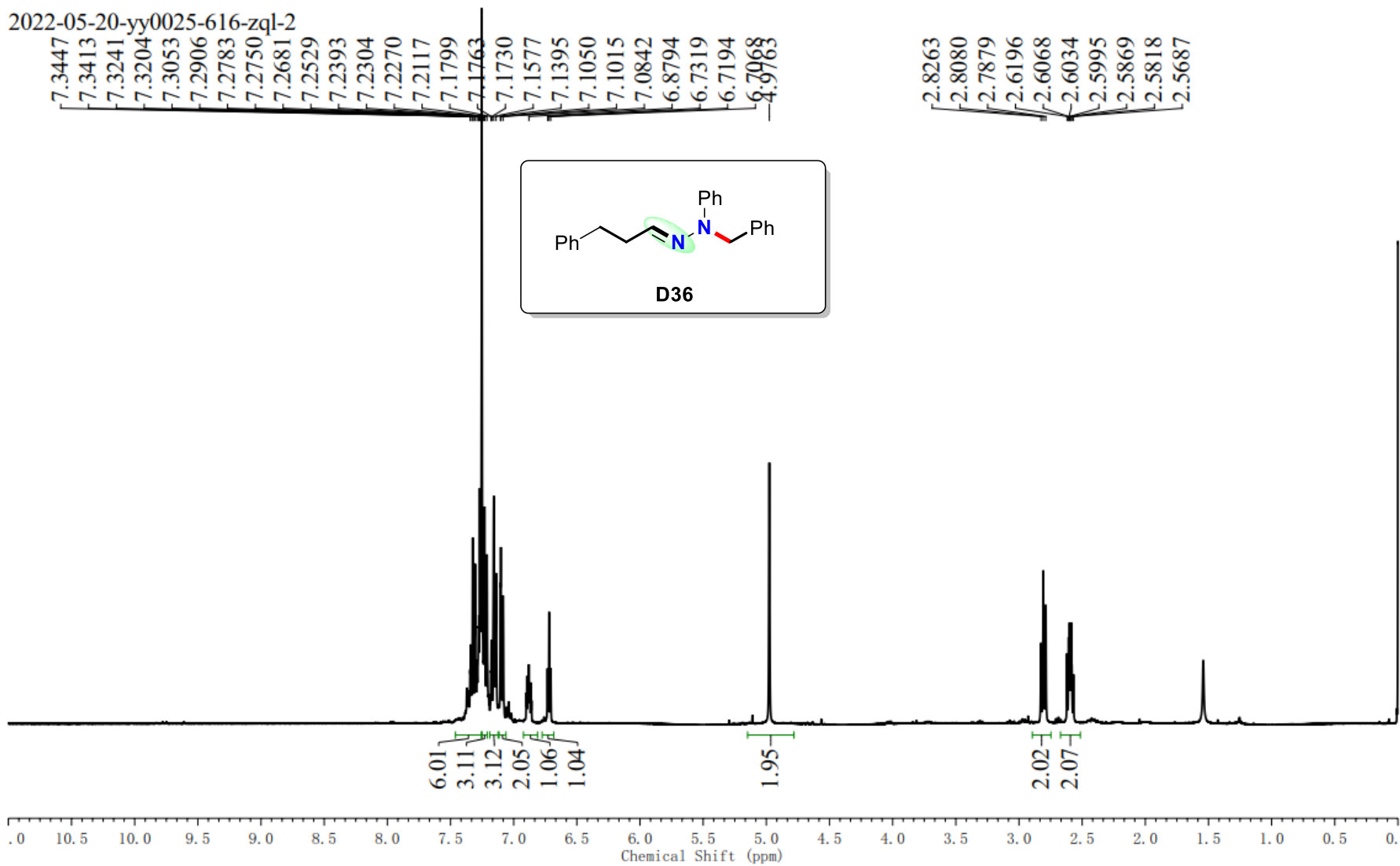
2022-6-7 YY0014-616-ZQL-1



2022-6-11 yy0002-616-zql-1



2022-05-20-yy0025-616-zql-2



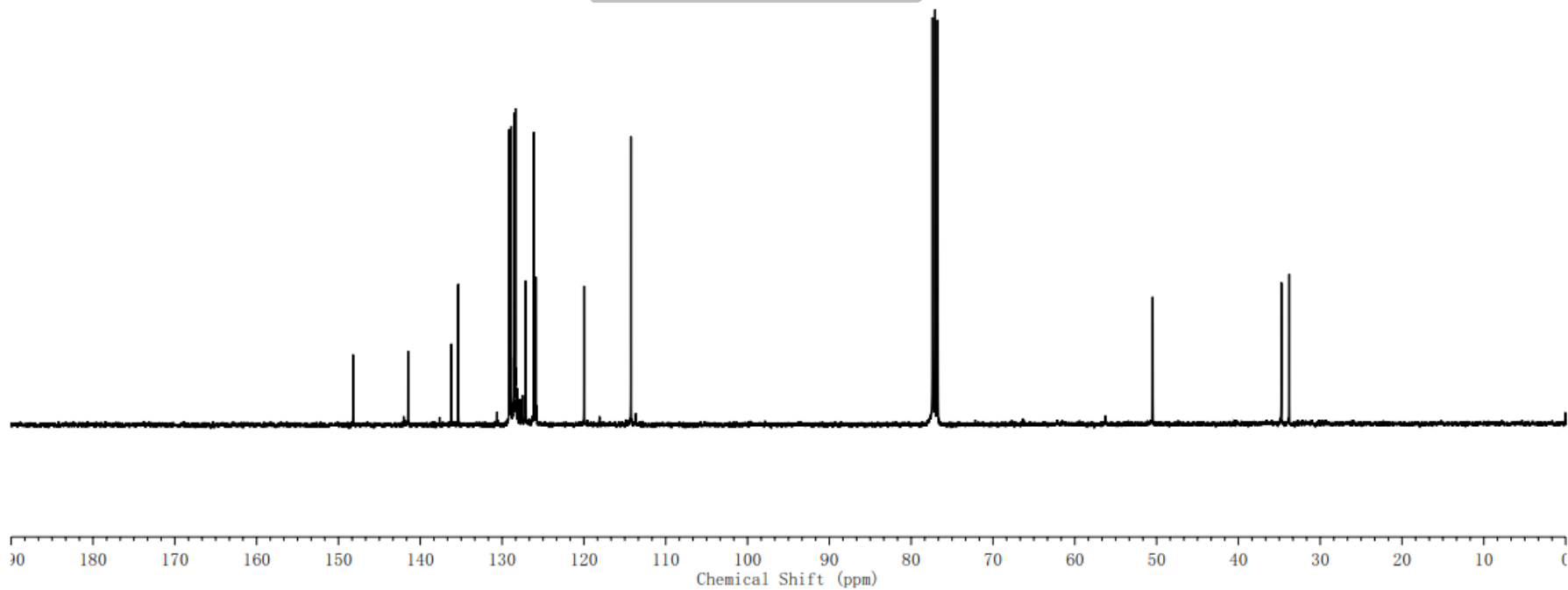
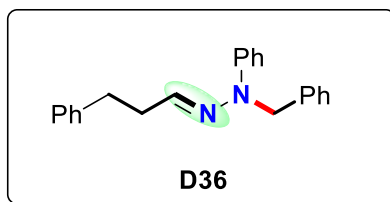
2022-06-22 yy0024 616-zql-2

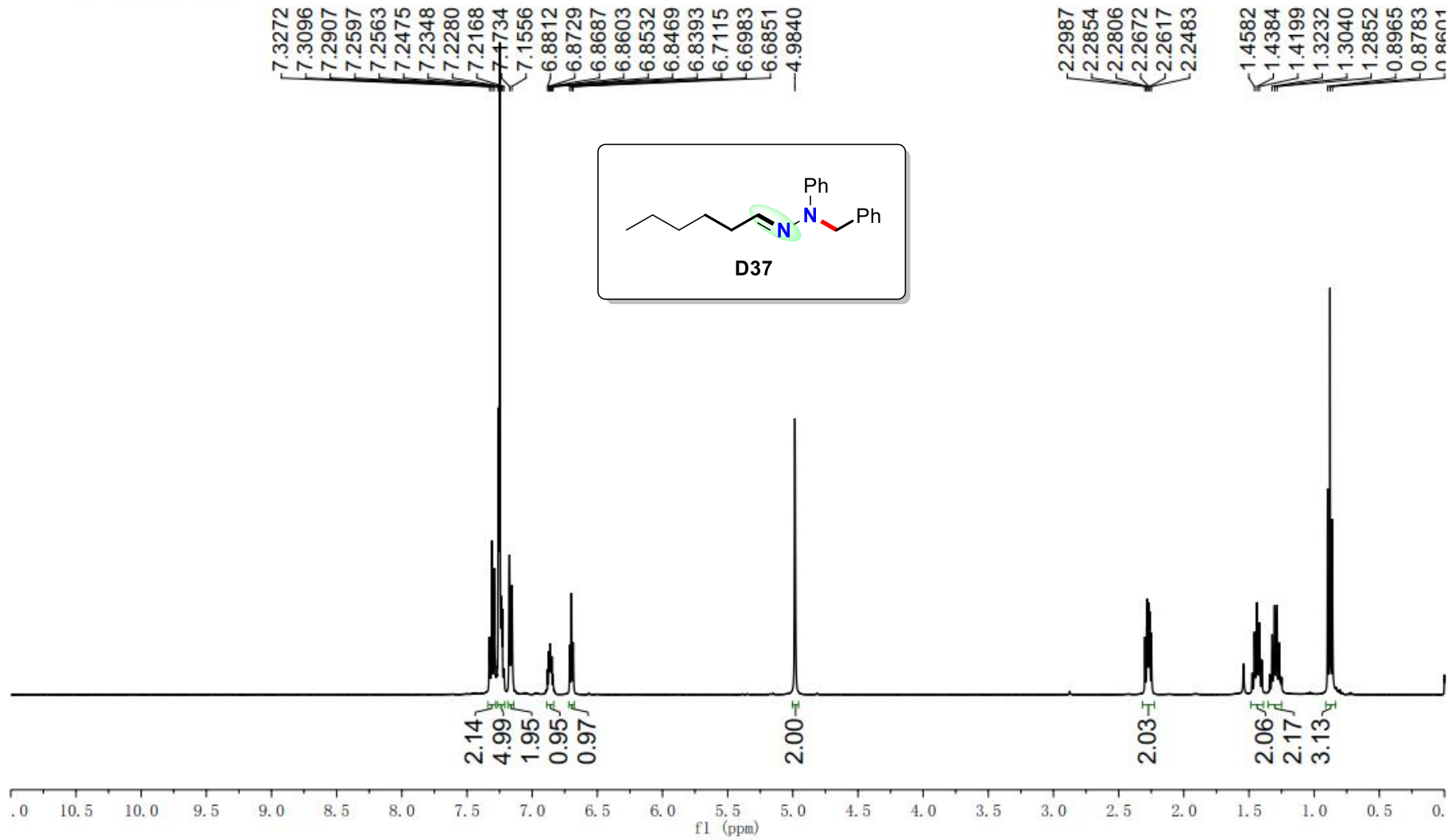
148.22
141.49
136.19
135.35
129.12
128.99
128.90
128.55
128.51
128.45
128.36
128.32
127.12
126.09
125.87
119.97
114.25

77.38
77.07
76.75

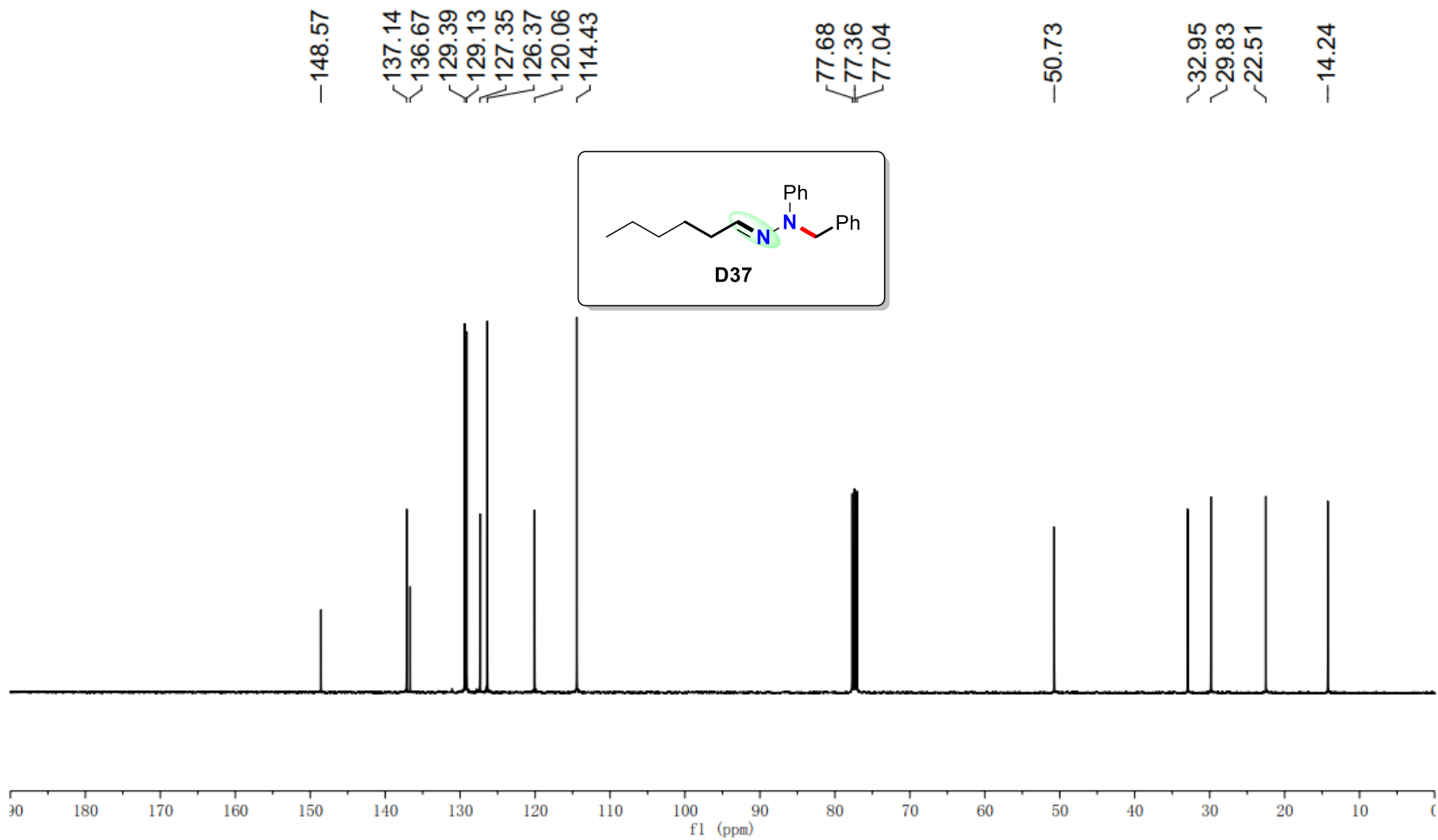
50.47

34.74
33.78

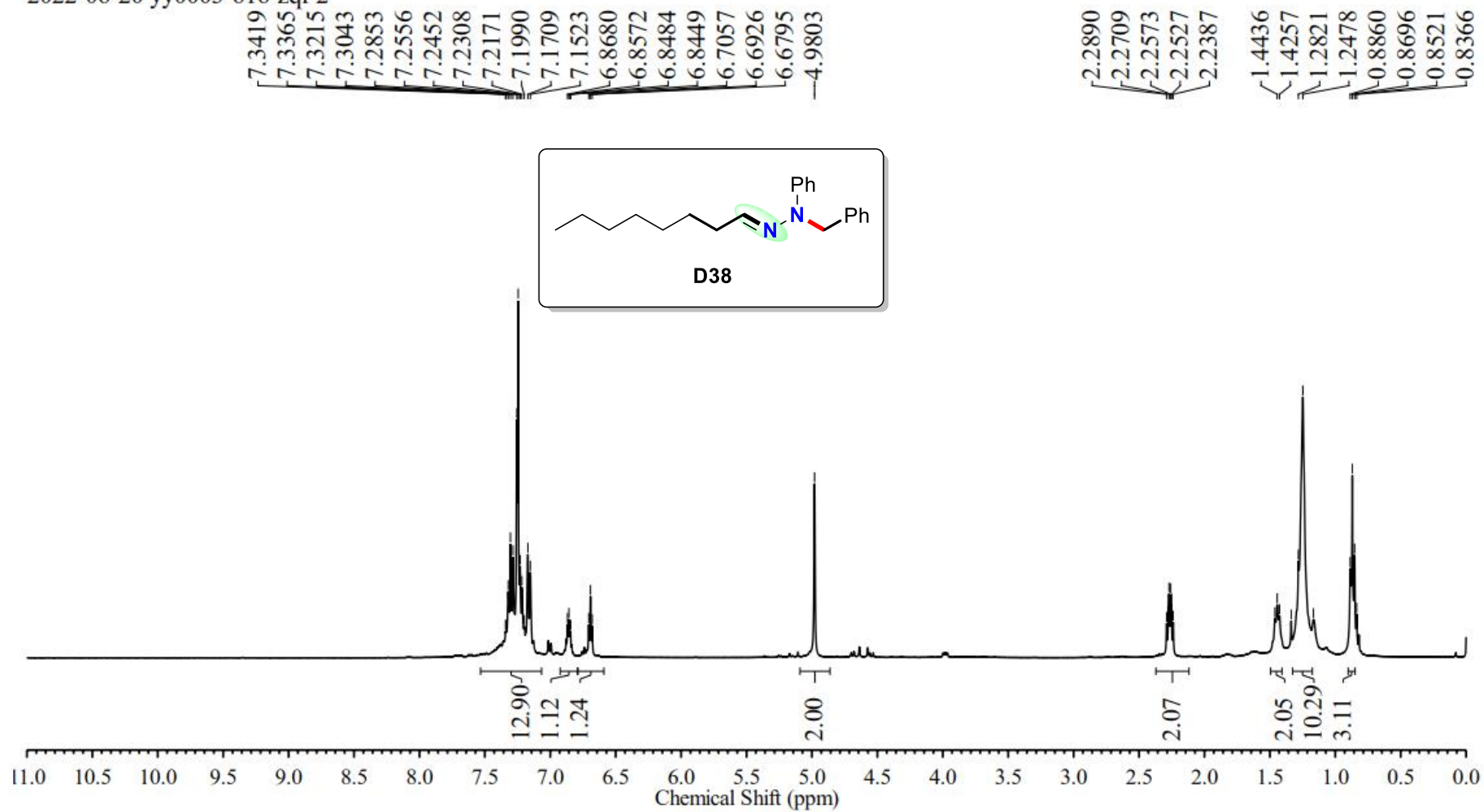




2022-9-19 yy0036-616-1qq-e37

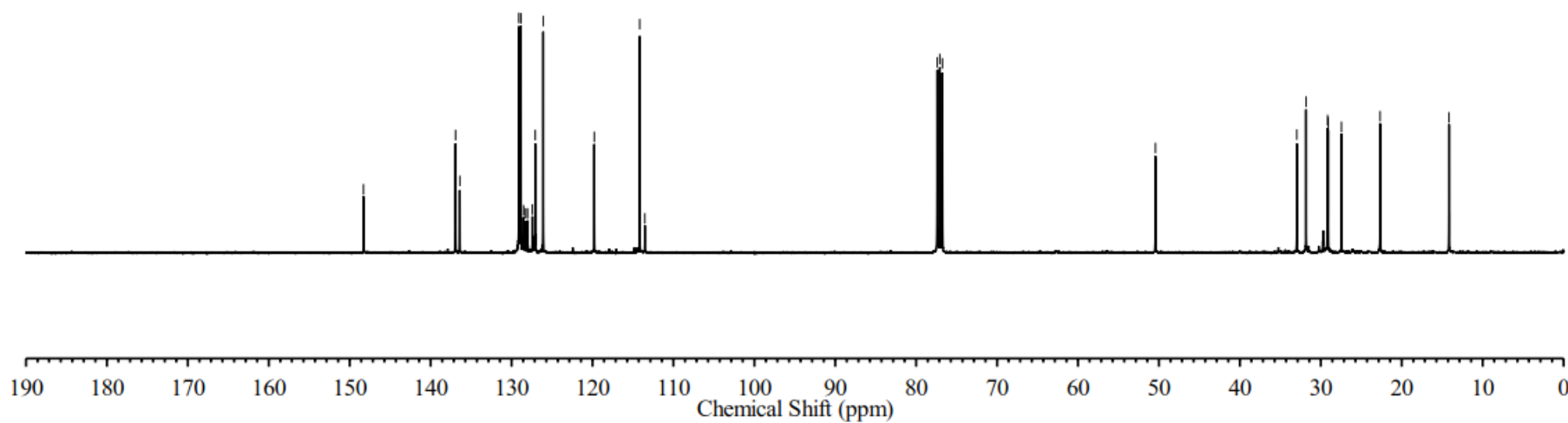
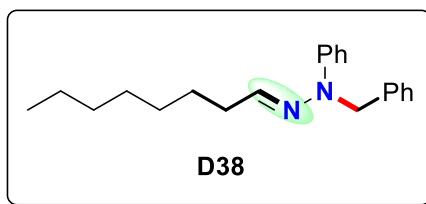


2022-06-20 yy0003-616-zql-2

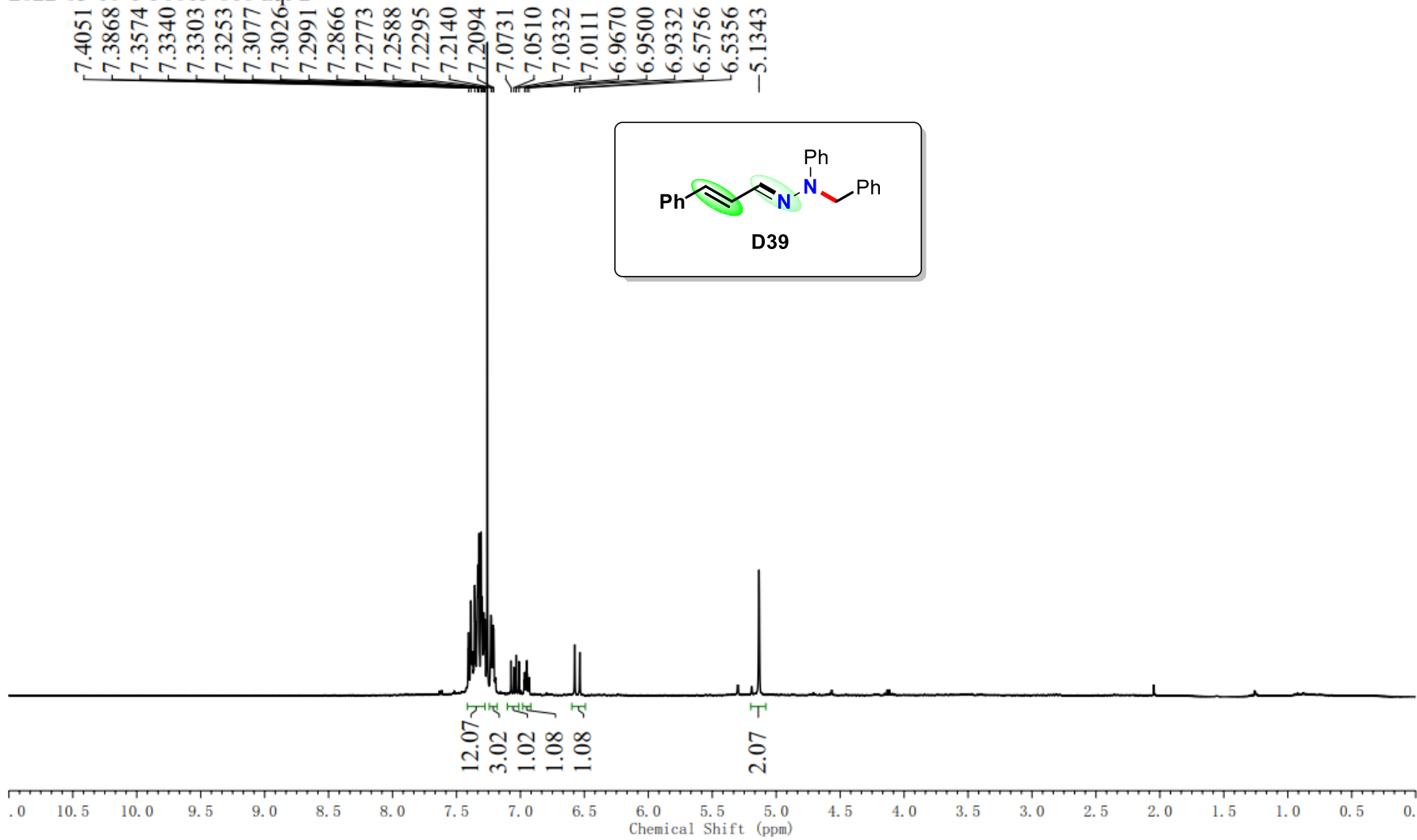


2022-06-21 yy0027 616-zql-2

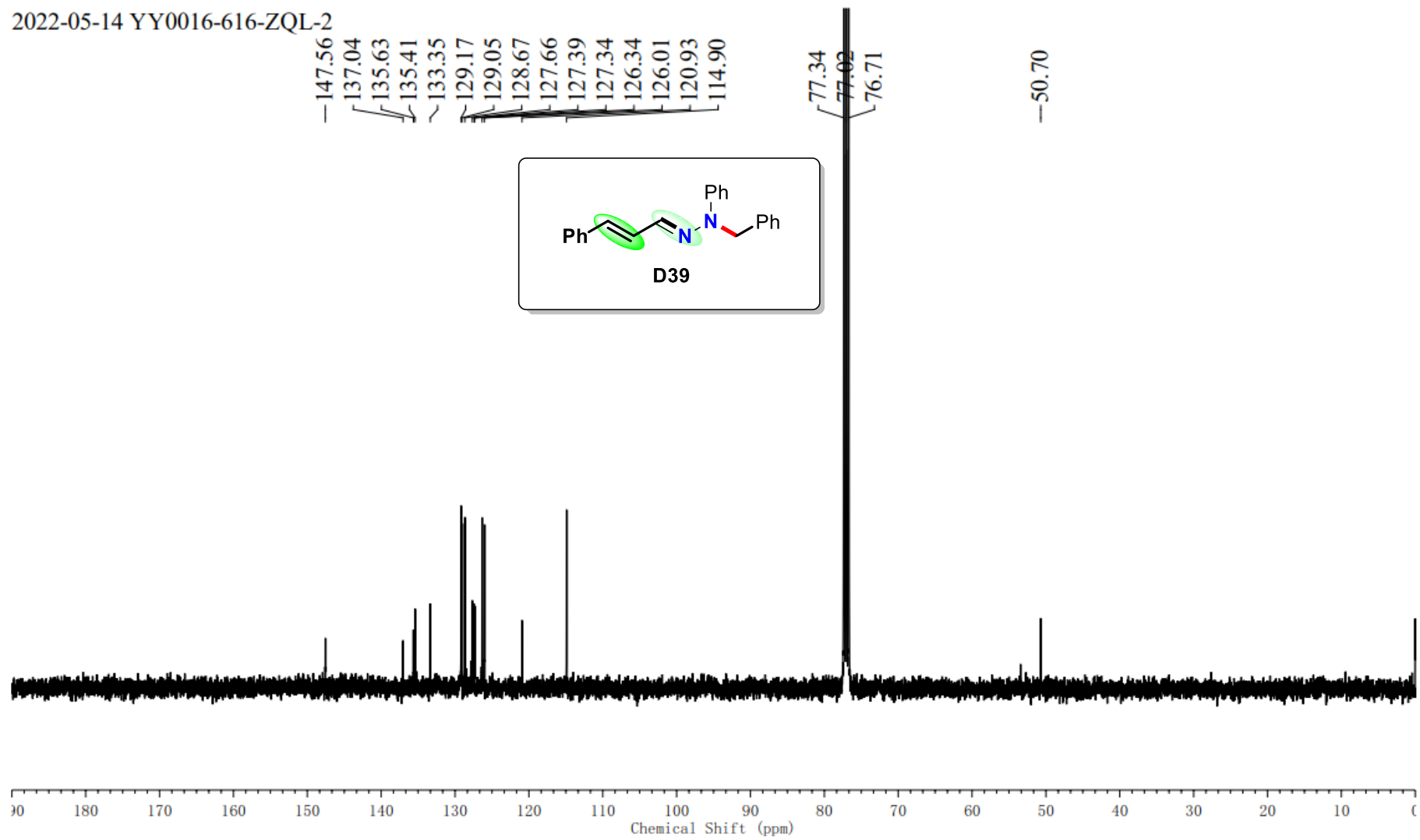
148.30 136.92 136.39 129.10 128.97 128.85 128.51 128.26 128.03 127.44 127.07 126.09 119.77 114.16 113.53 77.40 77.08 76.76 50.44 32.97 31.83 29.16 29.08 27.45 22.68 14.15



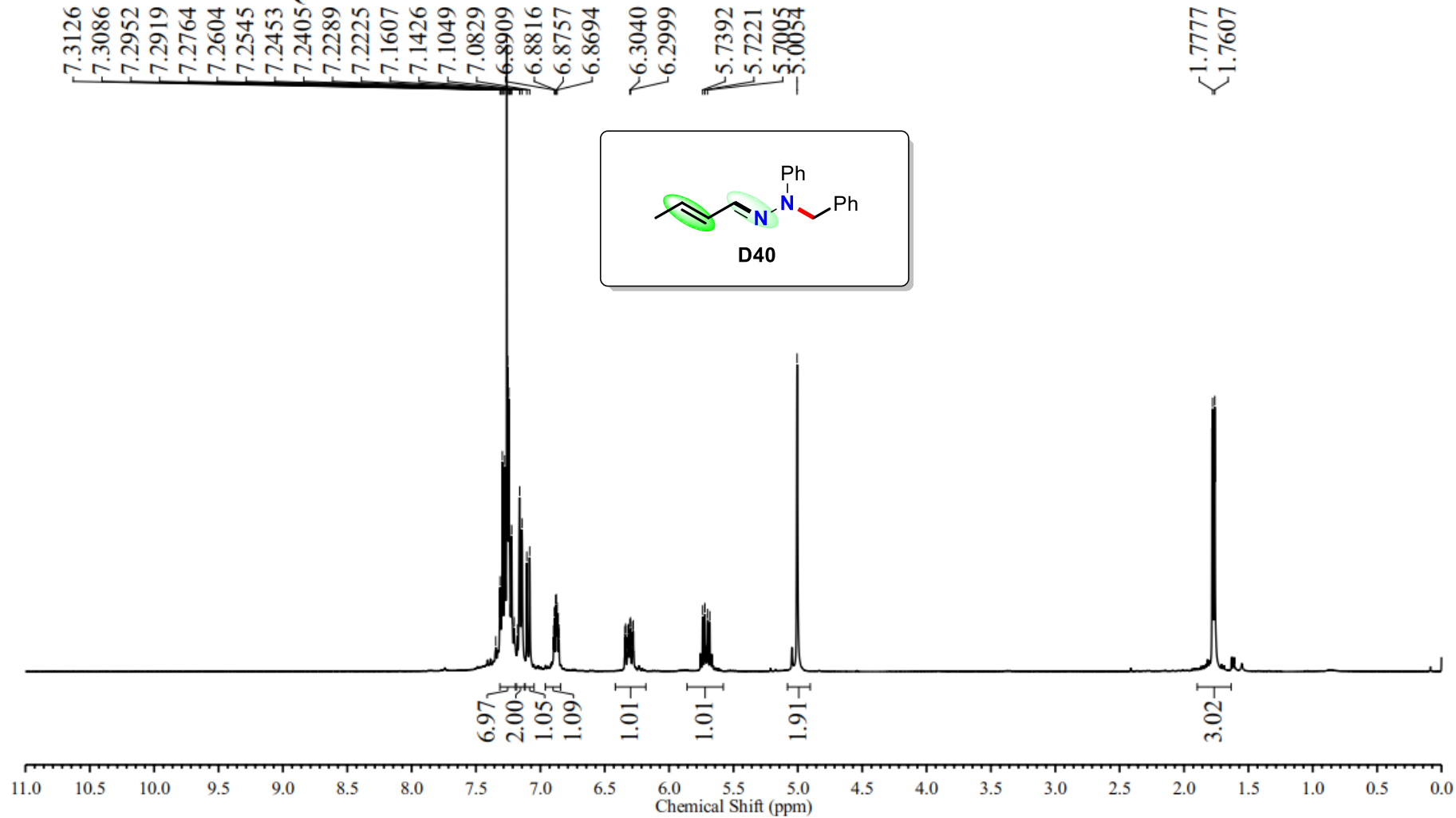
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2022-05-14 YY0016-616-ZQL-2

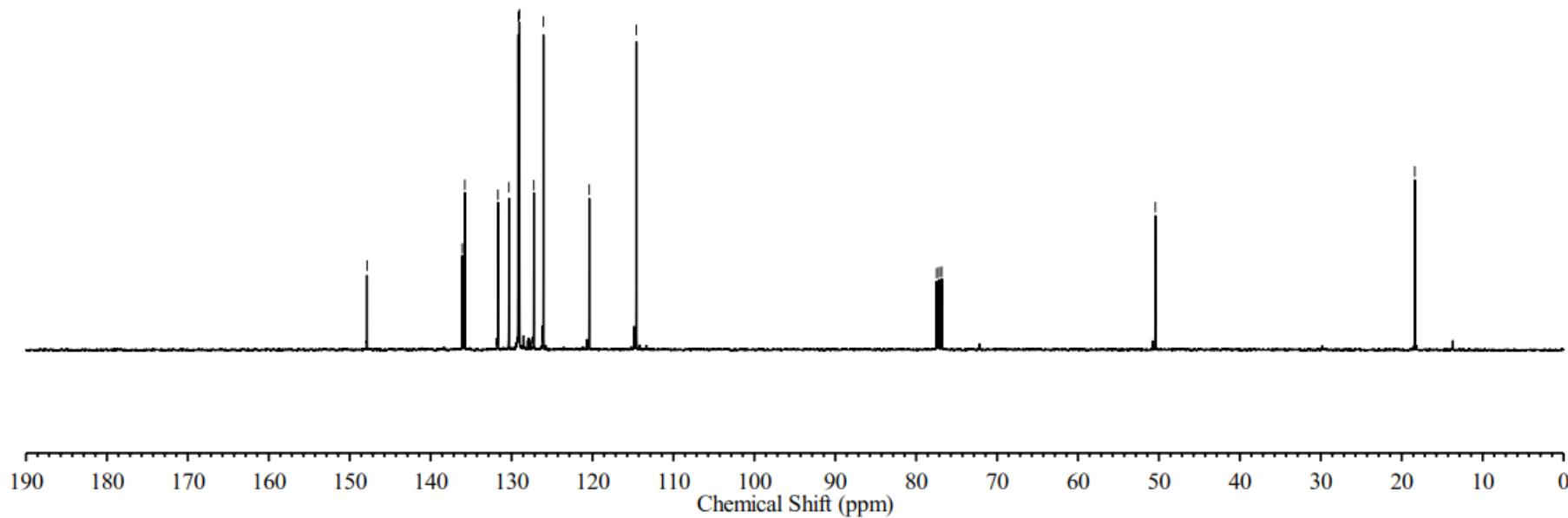
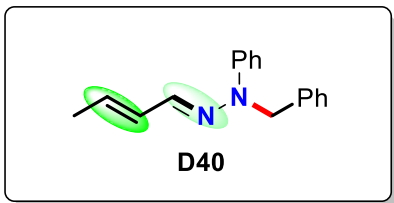


2022-6-11 YY0001-616-ZQL-1

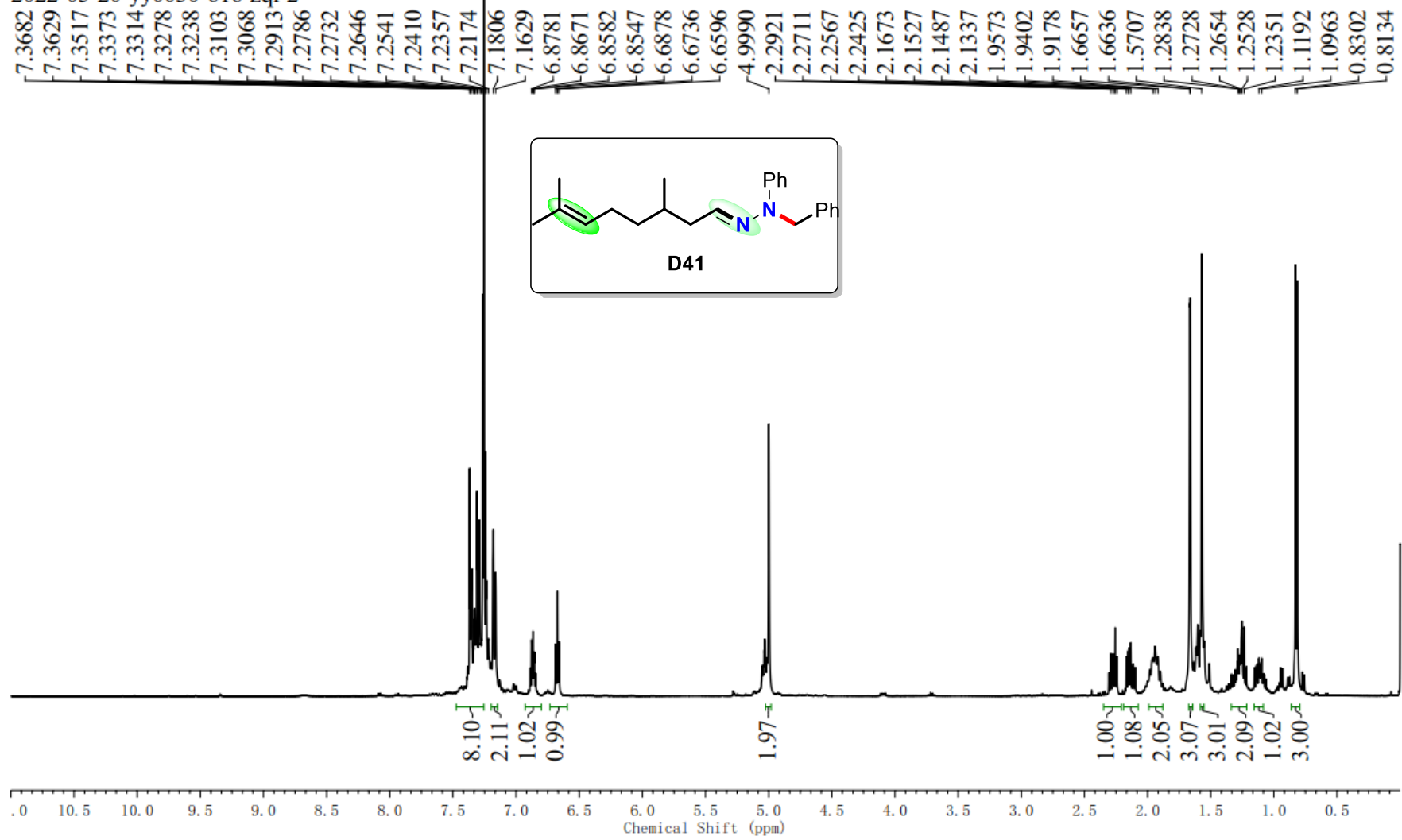


2022-6-13 YY0016-616-ZQL-10-1

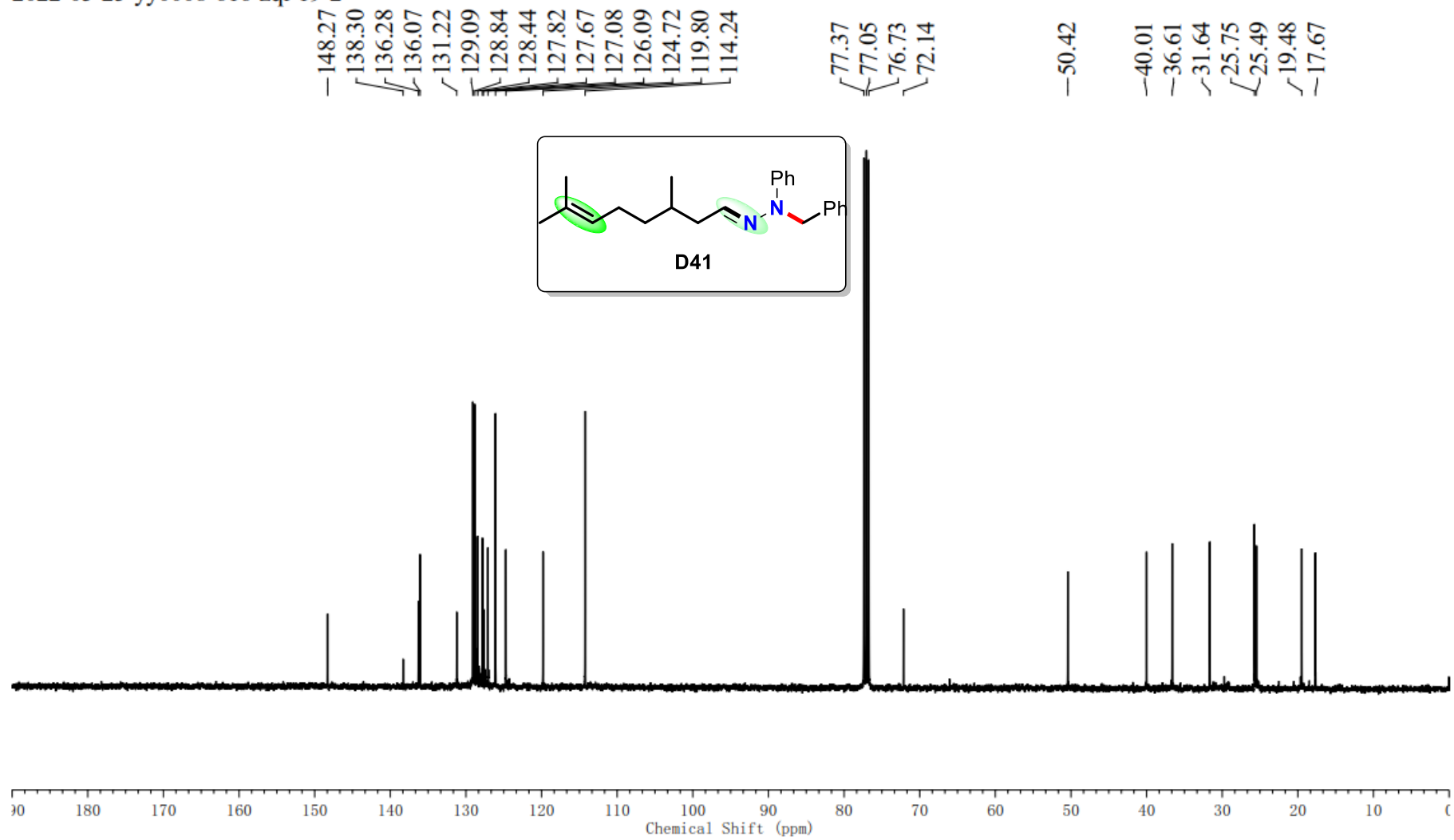
147.86
136.09
135.78
131.68
130.33
129.16
129.01
127.25
126.07
120.40
114.59
77.47
77.16
76.84
50.45
18.39



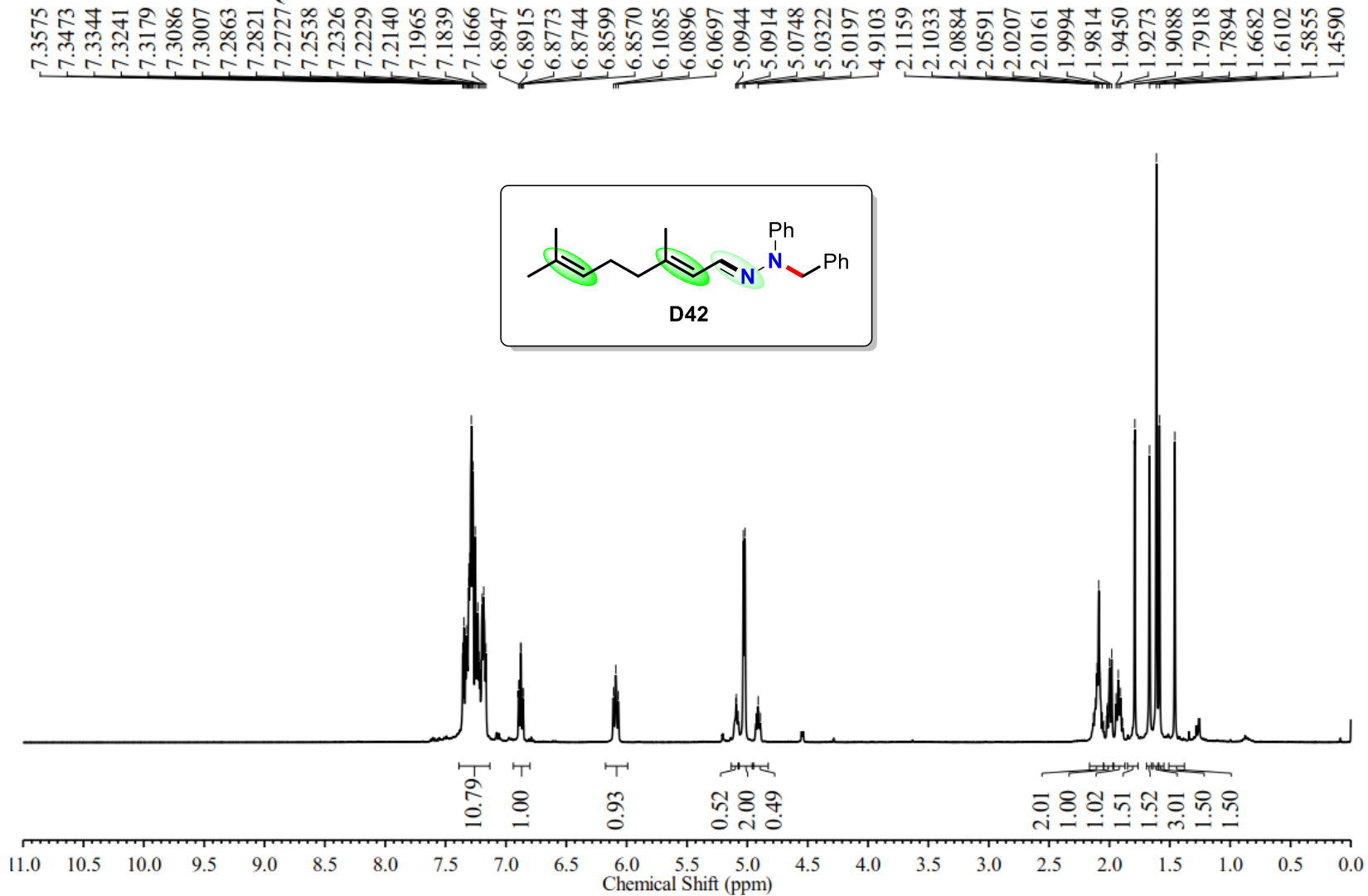
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2022-05-25-yy0008-616-zql-19-2

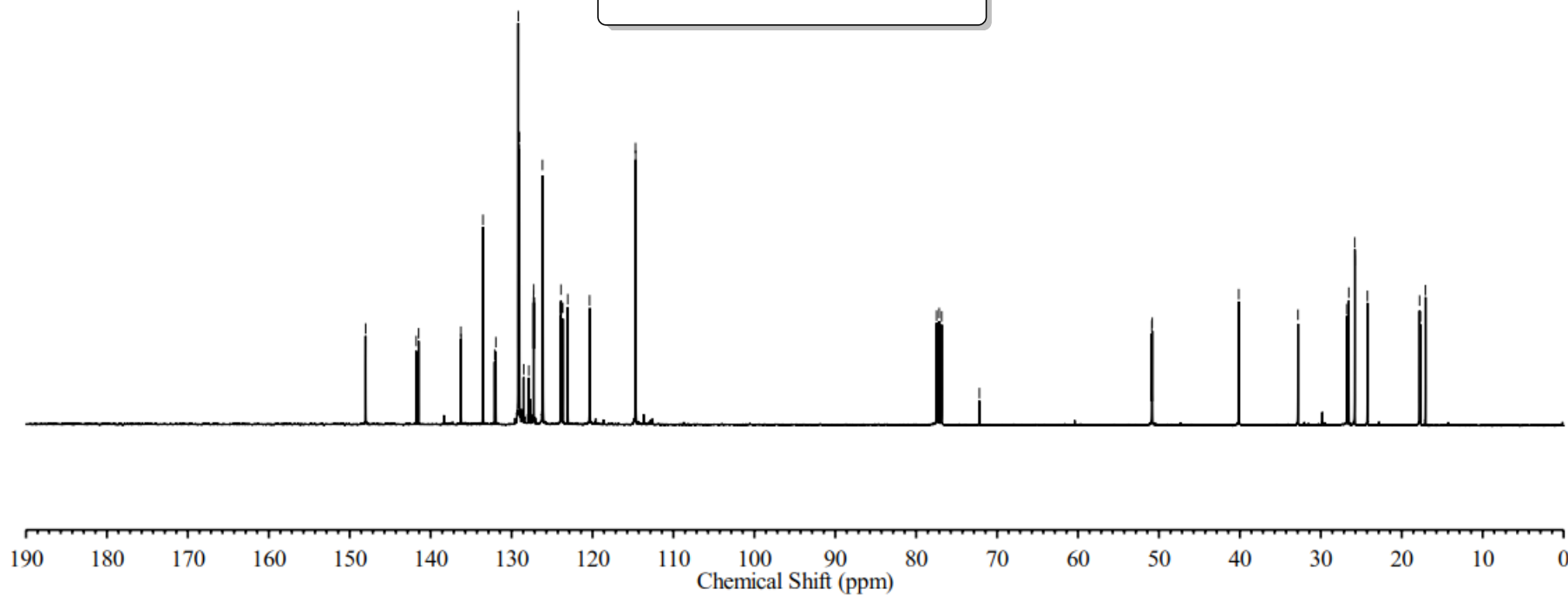
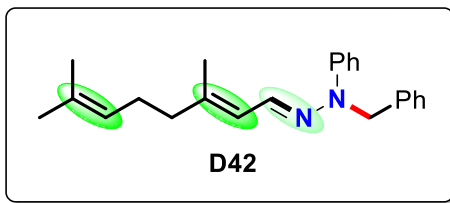


2022-6-11 YY0001-616-ZQL-2

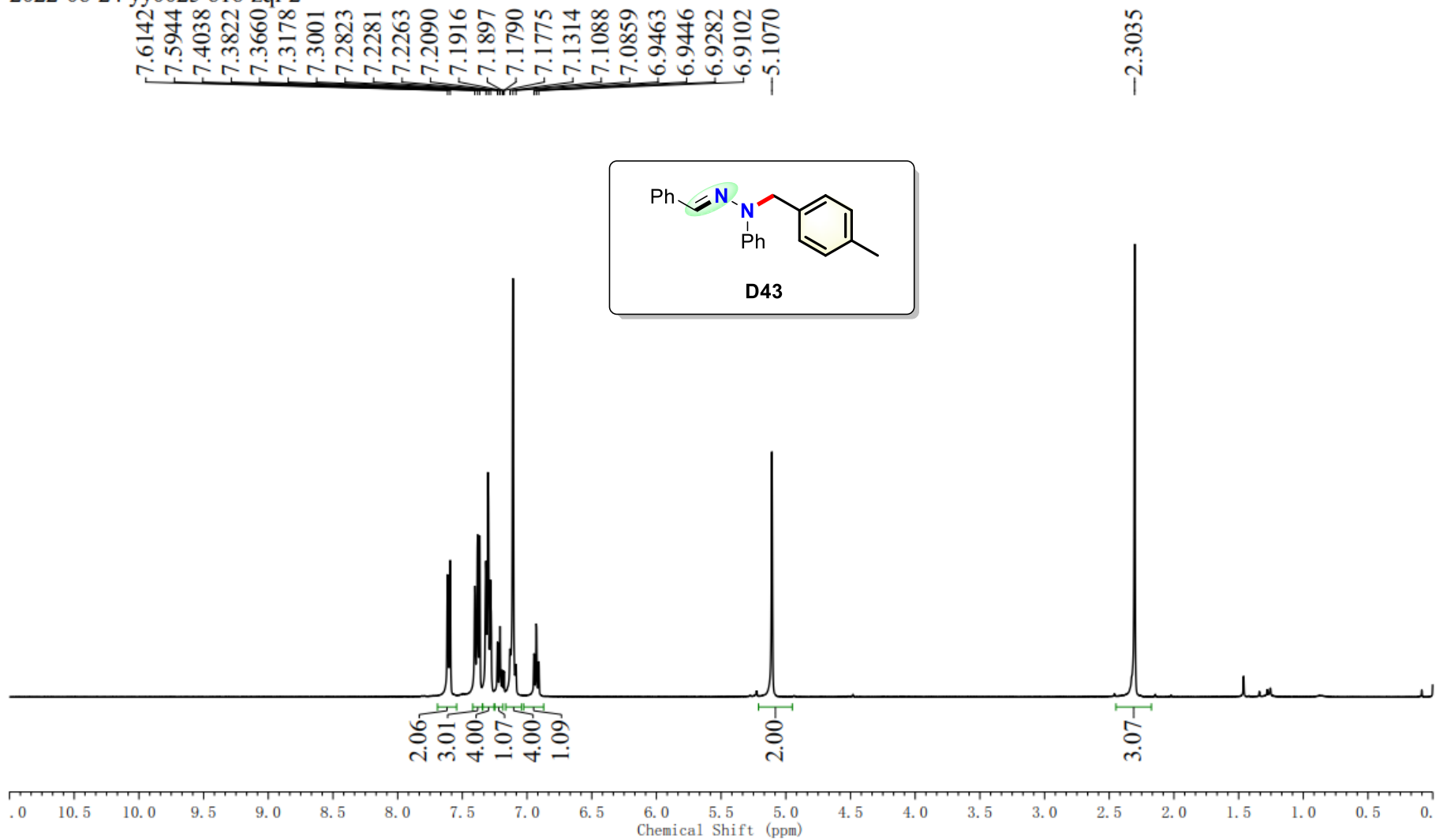


2022-6-13 YY0016-616-ZQL-10-2

148.03 141.79 141.49 136.26 136.24 133.53 132.09 131.94 129.17 129.04 129.02 128.49 127.86 127.28 127.26 126.18 126.11 123.93 123.89 123.66 123.05 120.36 120.32 114.68 114.66 77.48 77.16 76.84 72.19 50.88 50.83 40.14 32.81 26.78 26.54 25.81 25.80 24.23 17.80 17.67 17.06



2022-06-24 yy0025 616-zql-2



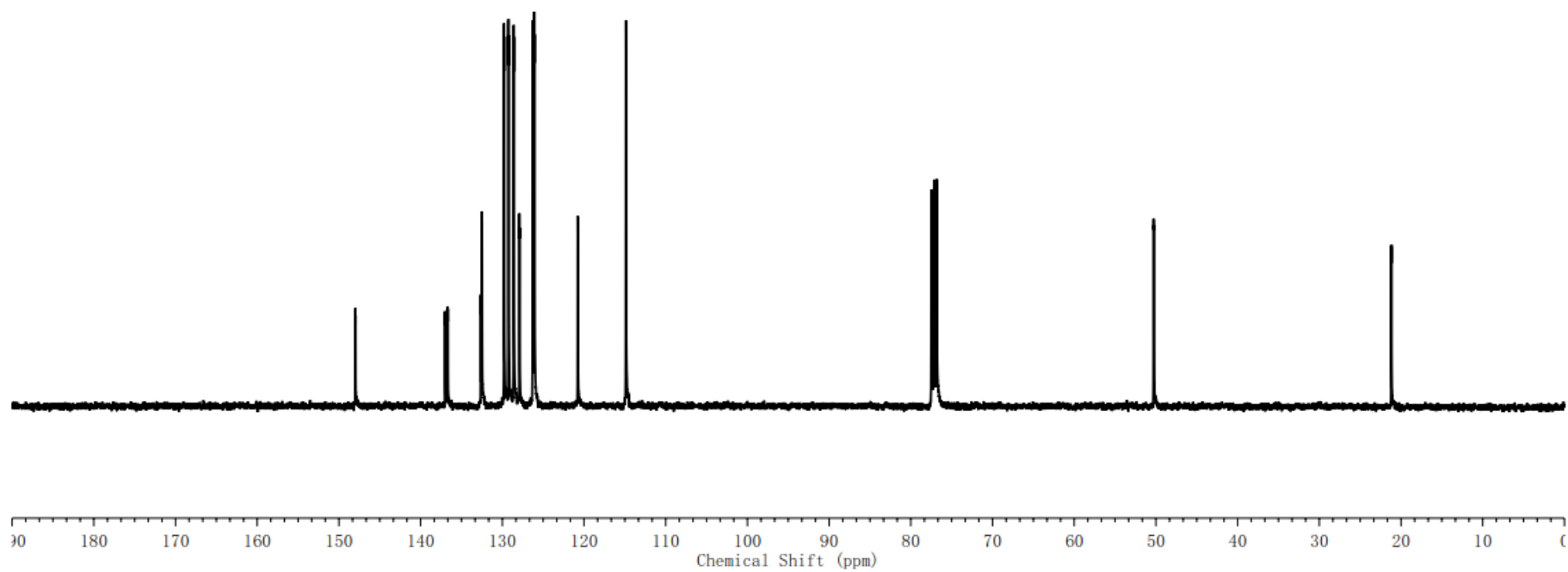
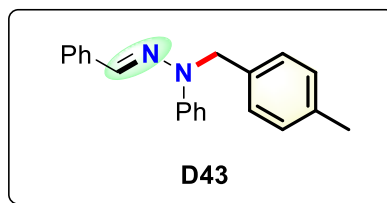
2022-06-28-yy0028-616-20220624-2

148.00
137.01
136.70
132.63
132.51
129.79
129.25
128.60
126.29
126.08
120.77
114.88

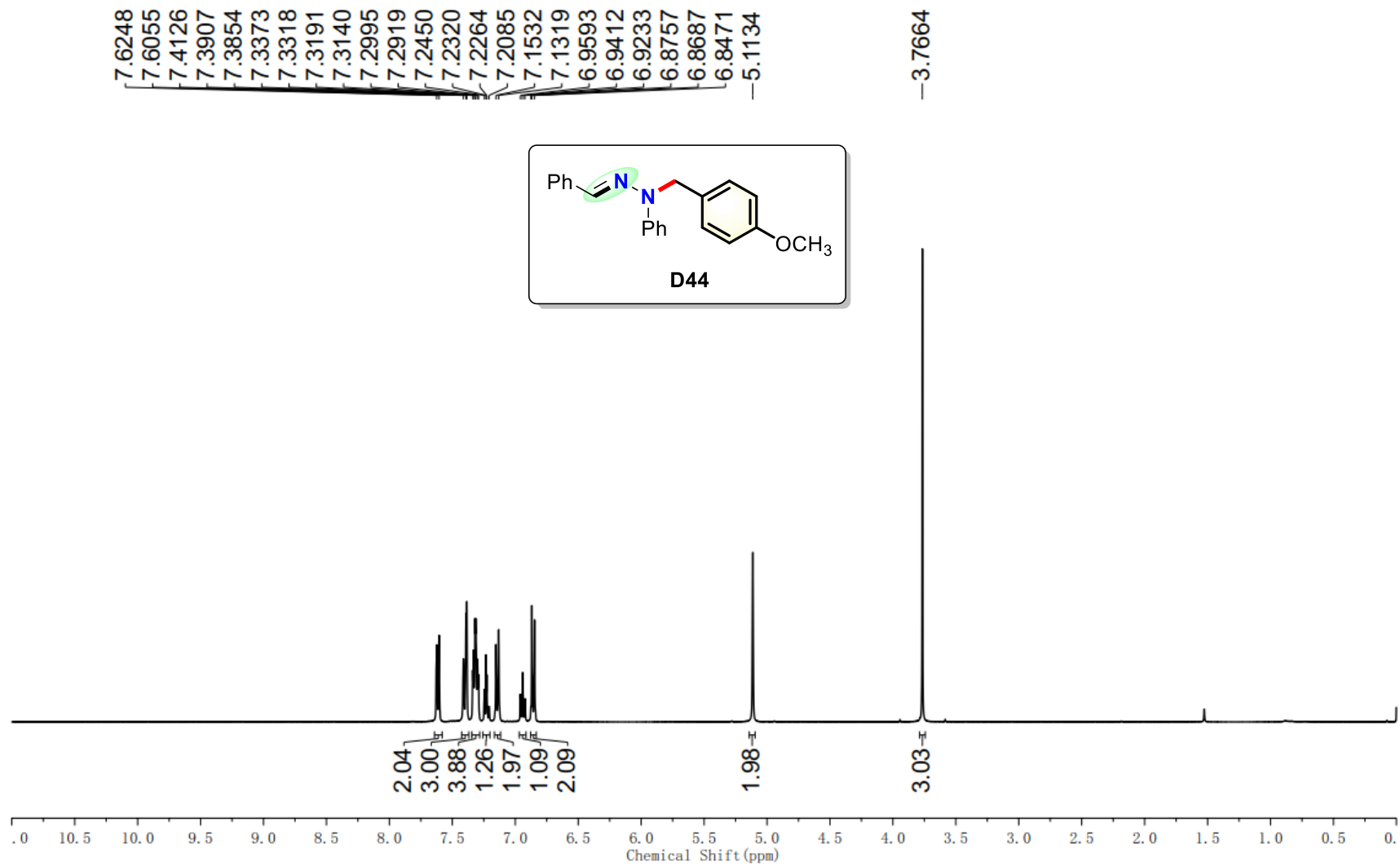
77.47
77.15
76.83

50.27

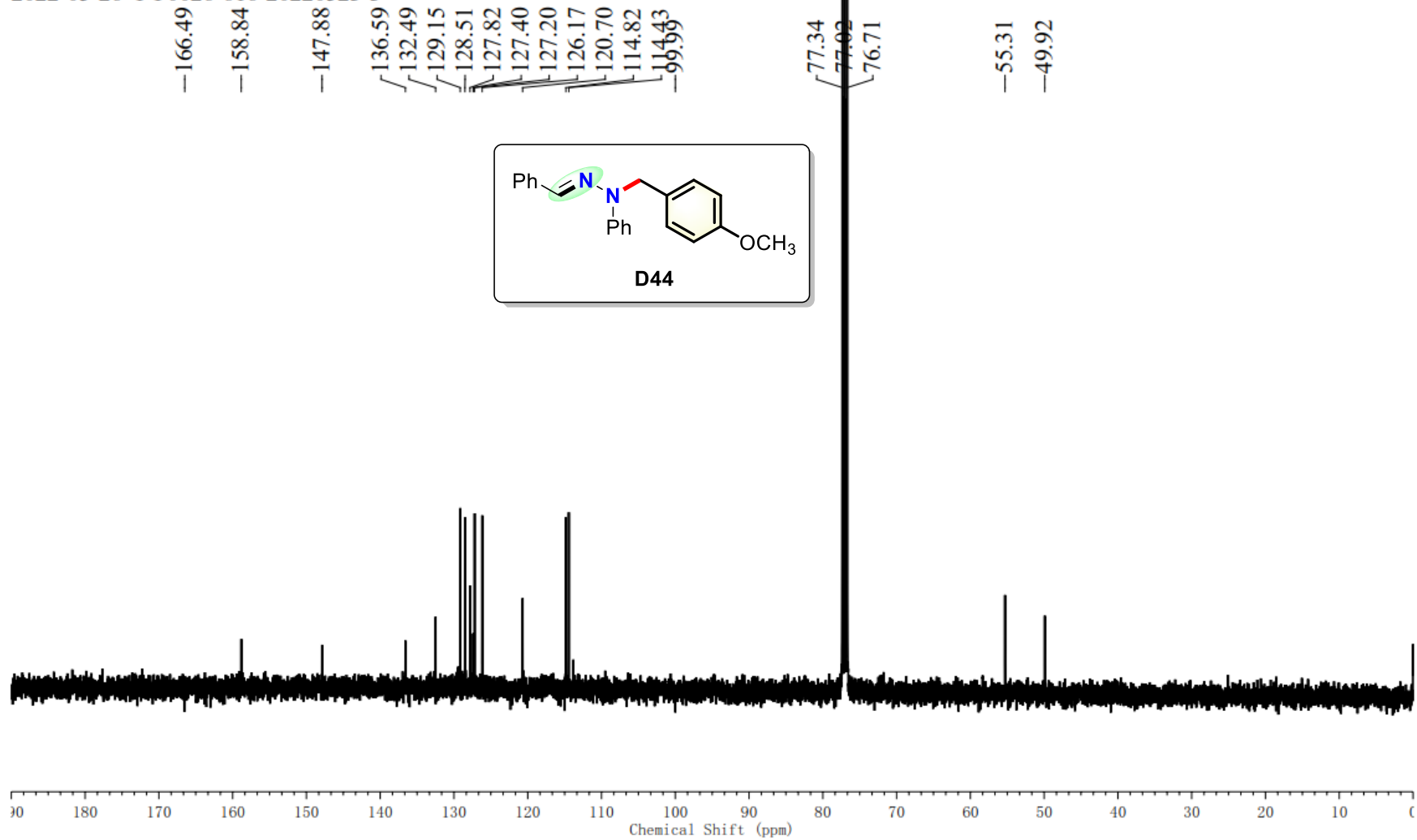
21.16



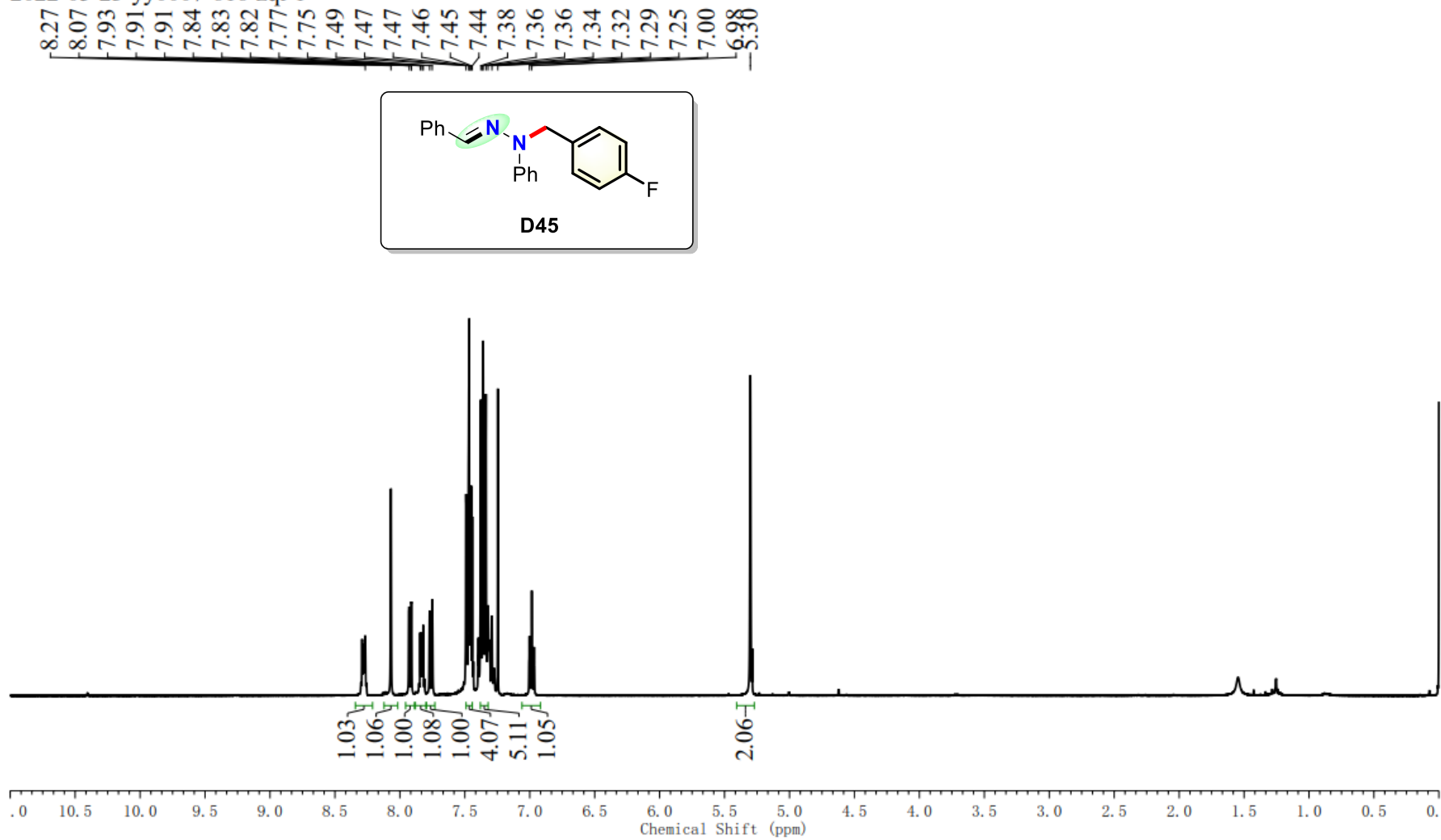
2022-11-09-yy0022-616-d7



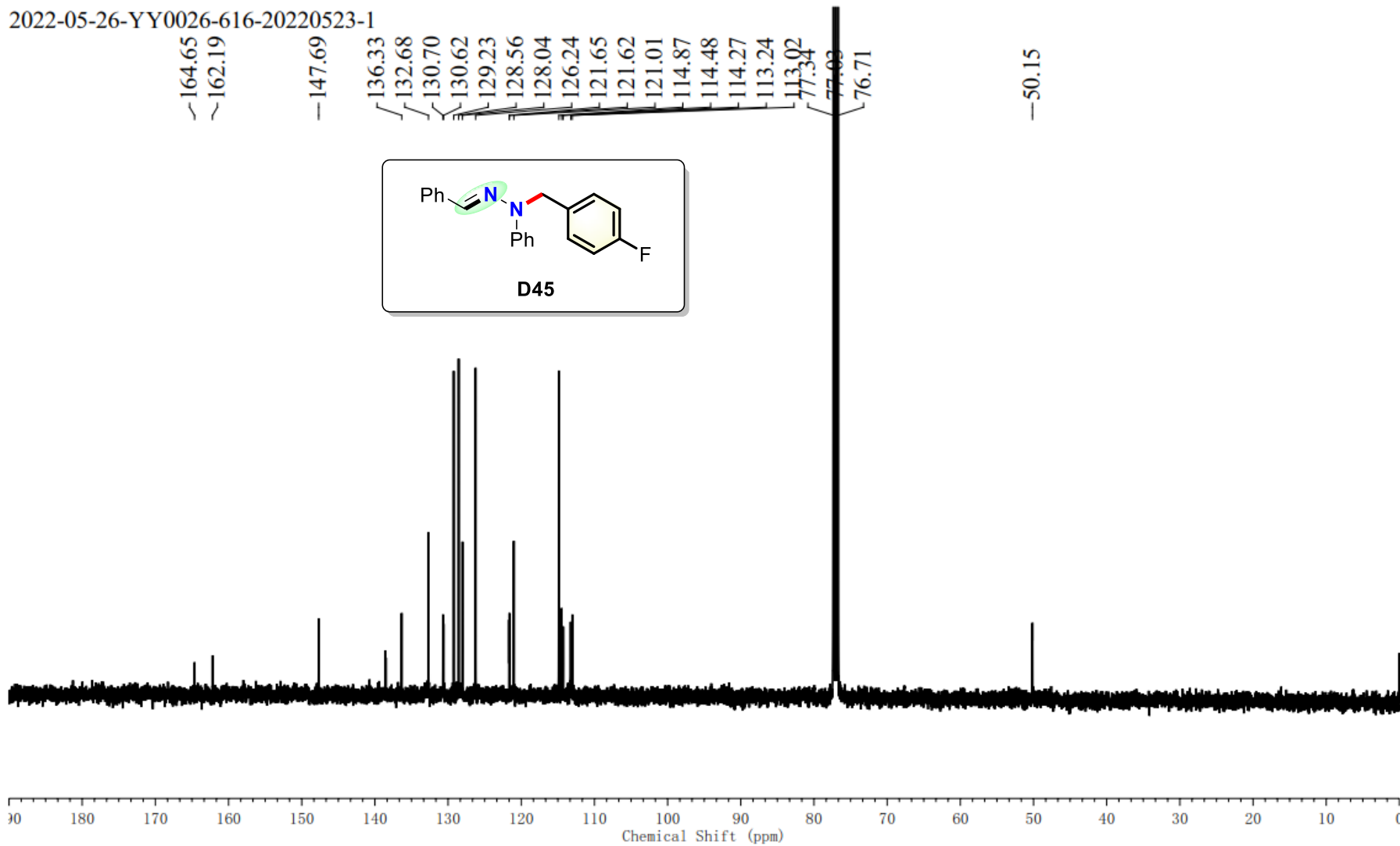
2022-05-26-YY0026-616-20220523-3



2022-05-23-yy0007-616-zql-1

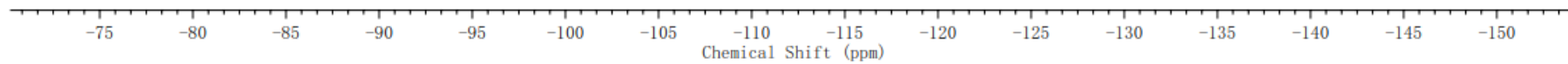
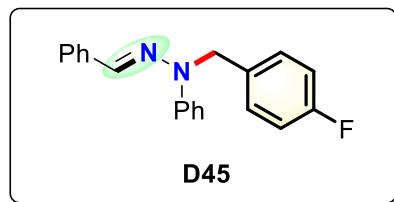


2022-05-26-YY0026-616-20220523-1



2022-05-30 YY0023 616-ZQI-20220523-1

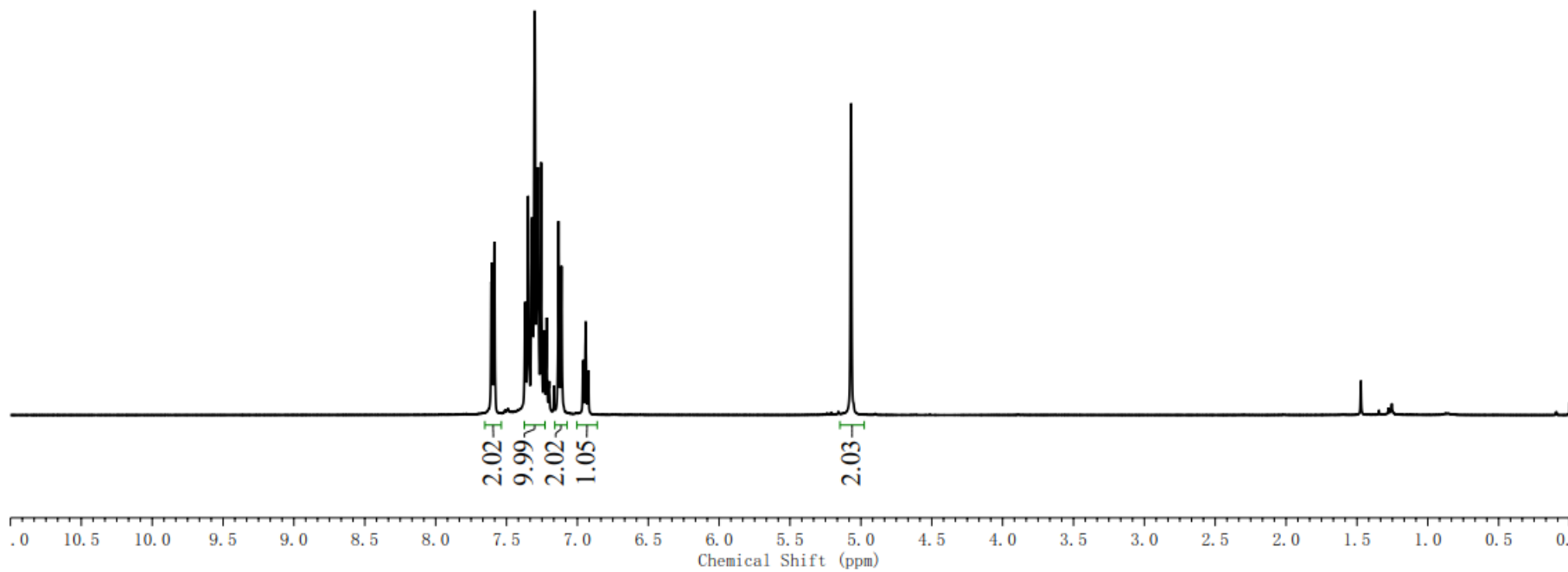
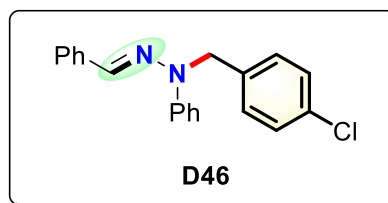
--112.0987



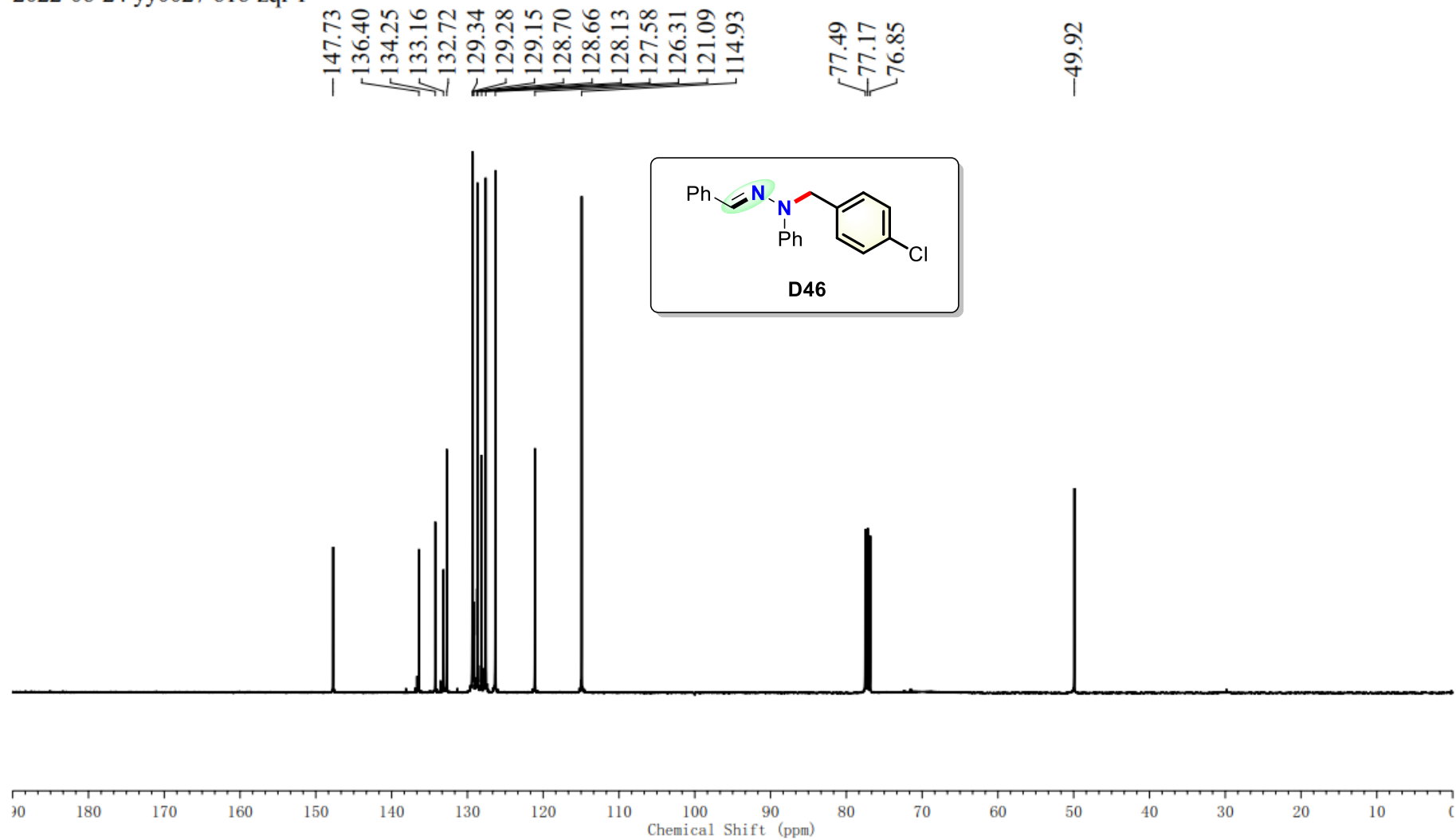
S233

2022-06-22 yy0025 61zql-01

7.6068
7.6040
7.5860
7.3702
7.3676
7.3481
7.3194
7.3136
7.3010
7.2843
7.2782
7.2565
7.2355
7.2327
7.2174
7.1989
7.1655
7.1329
7.1119
6.9593
6.9415
6.9237
5.0695

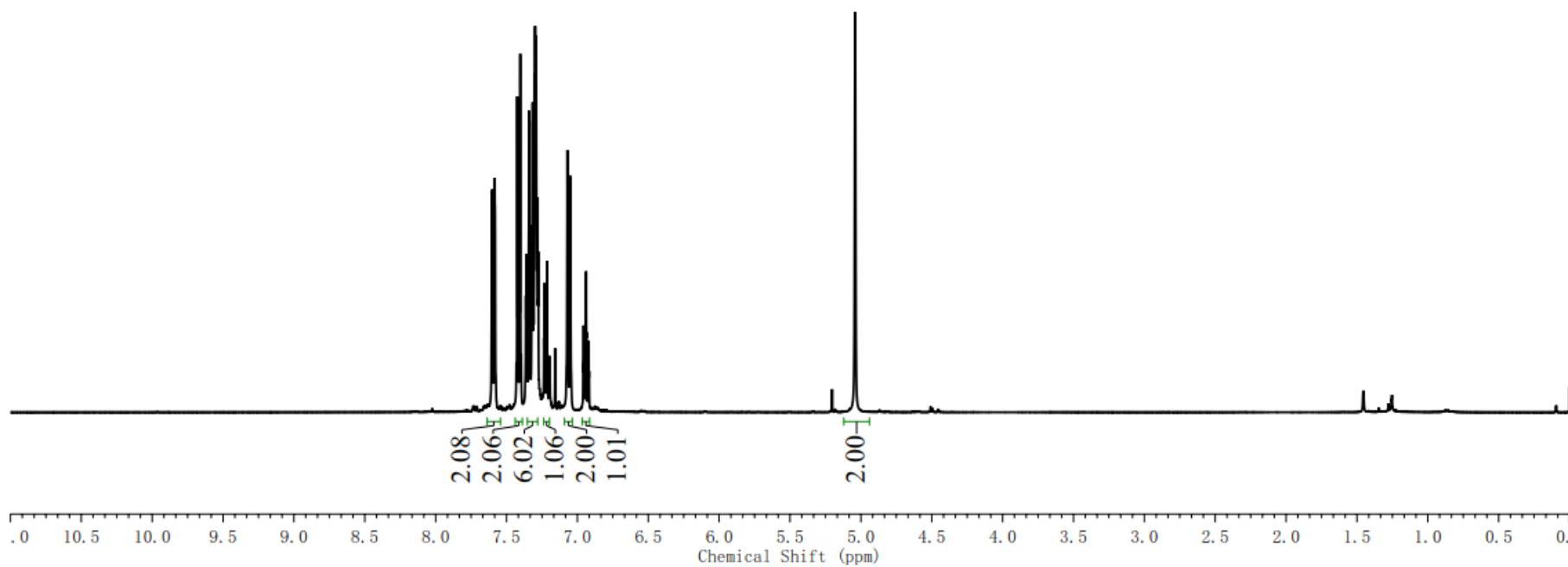
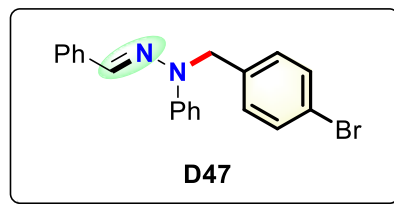


2022-06-24 yy0027 616-zql-1

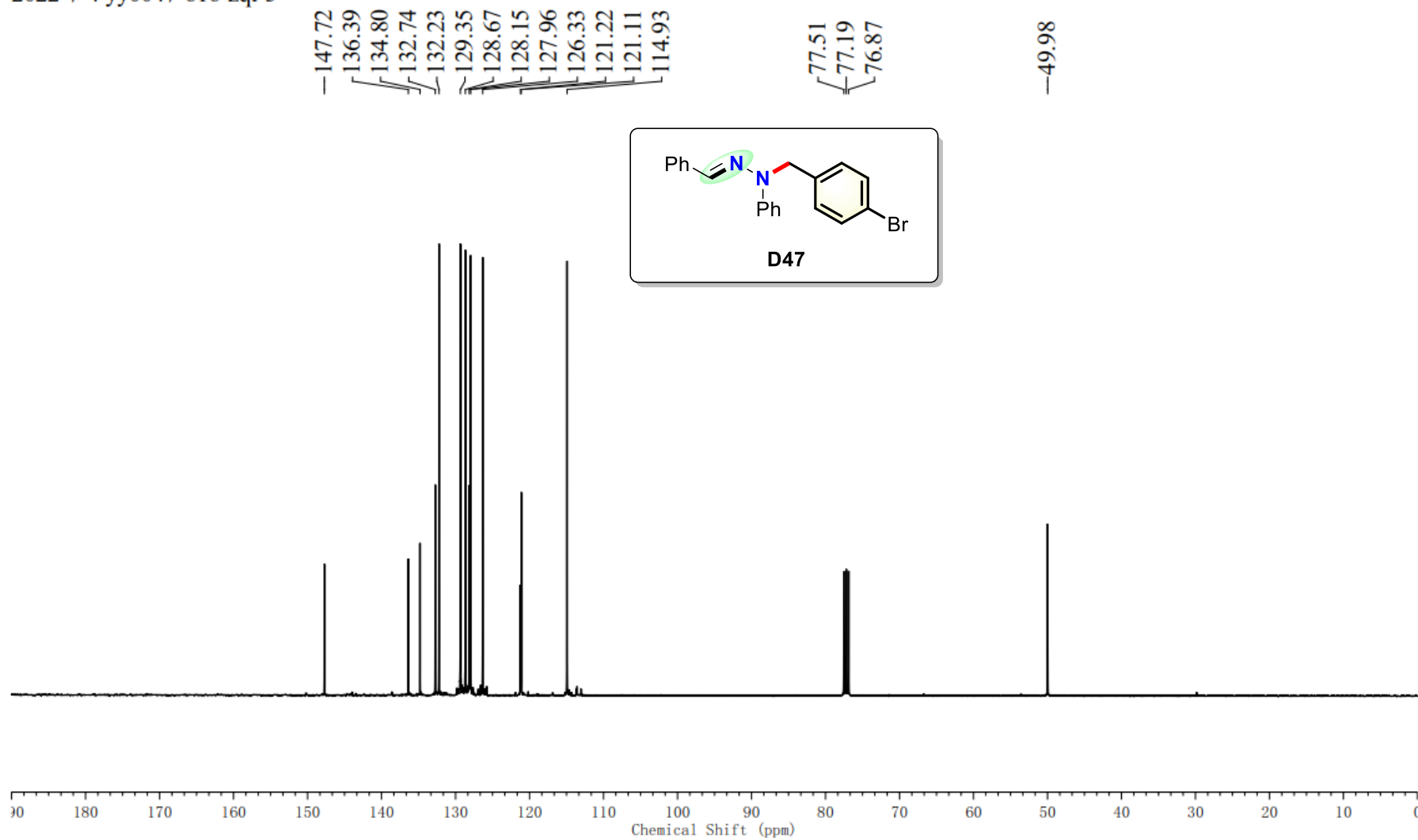


2022-07-02-yy0019-616-zql-2

7.6043
7.6007
7.5964
7.5829
7.5812
7.4243
7.4196
7.4079
7.4031
7.3970
7.3638
7.3605
7.3554
7.3467
7.3413
7.3384
7.3334
7.3199
7.3155
7.3095
7.3019
7.2979
7.2917
7.2825
7.2780
7.2755
7.2362
7.2330
7.2298
7.2198
7.2145
7.1964
7.1578
7.0707
7.0494
6.9562
6.9415
6.9385
6.9356
6.9208

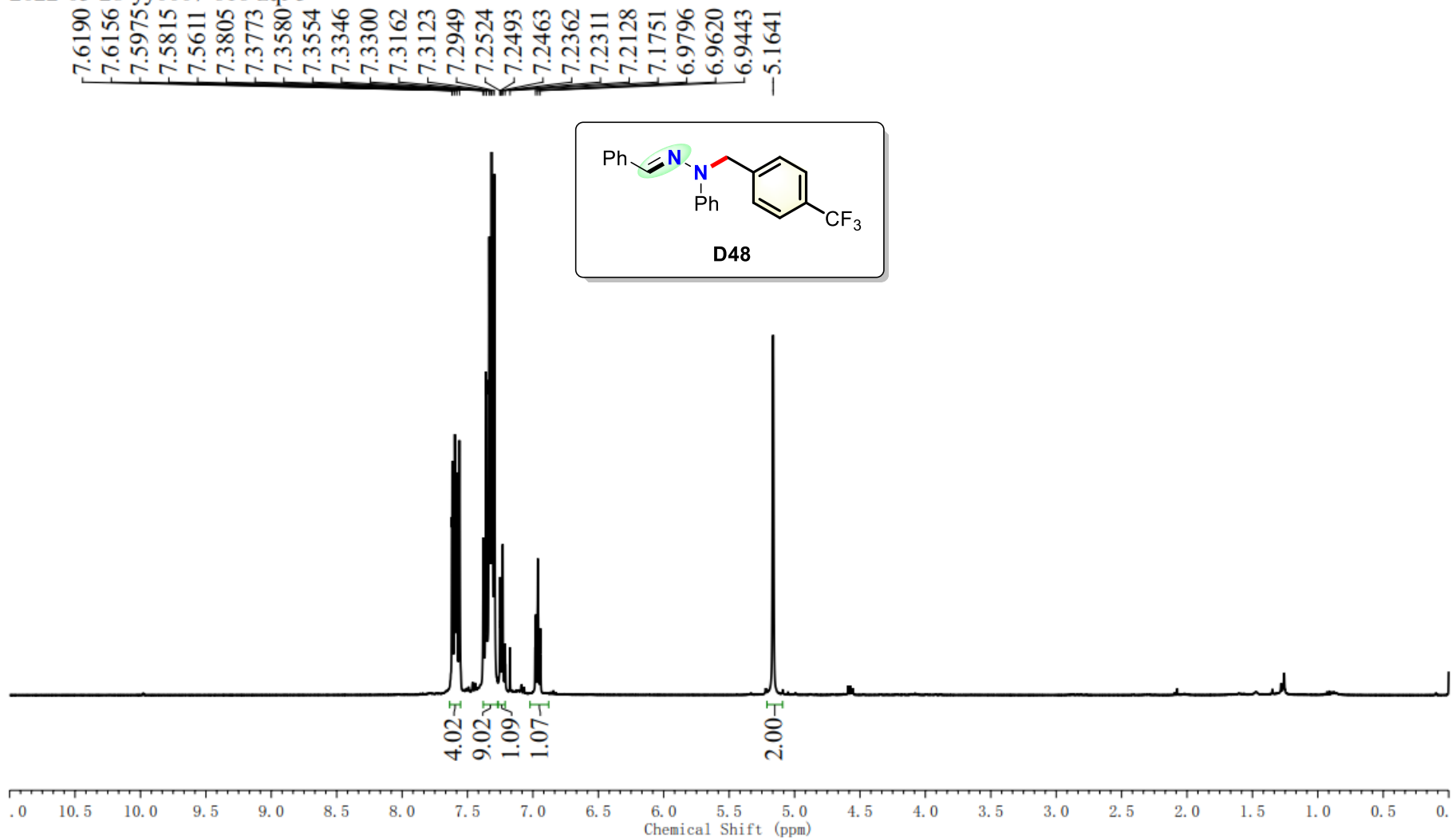


2022-7-4 yy0047-616-zql-3

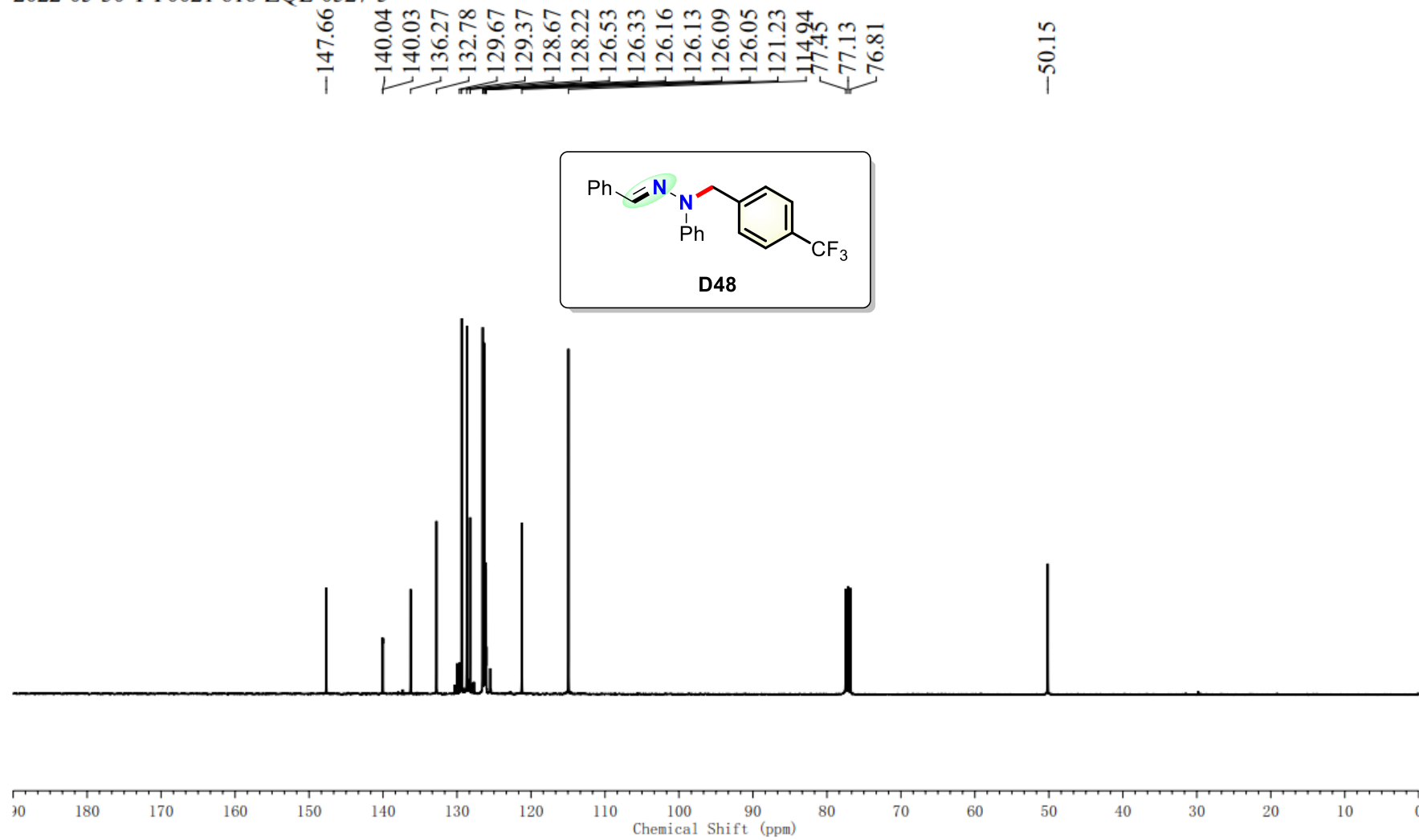


S237

2022-05-28-yy0007-616-zql-5

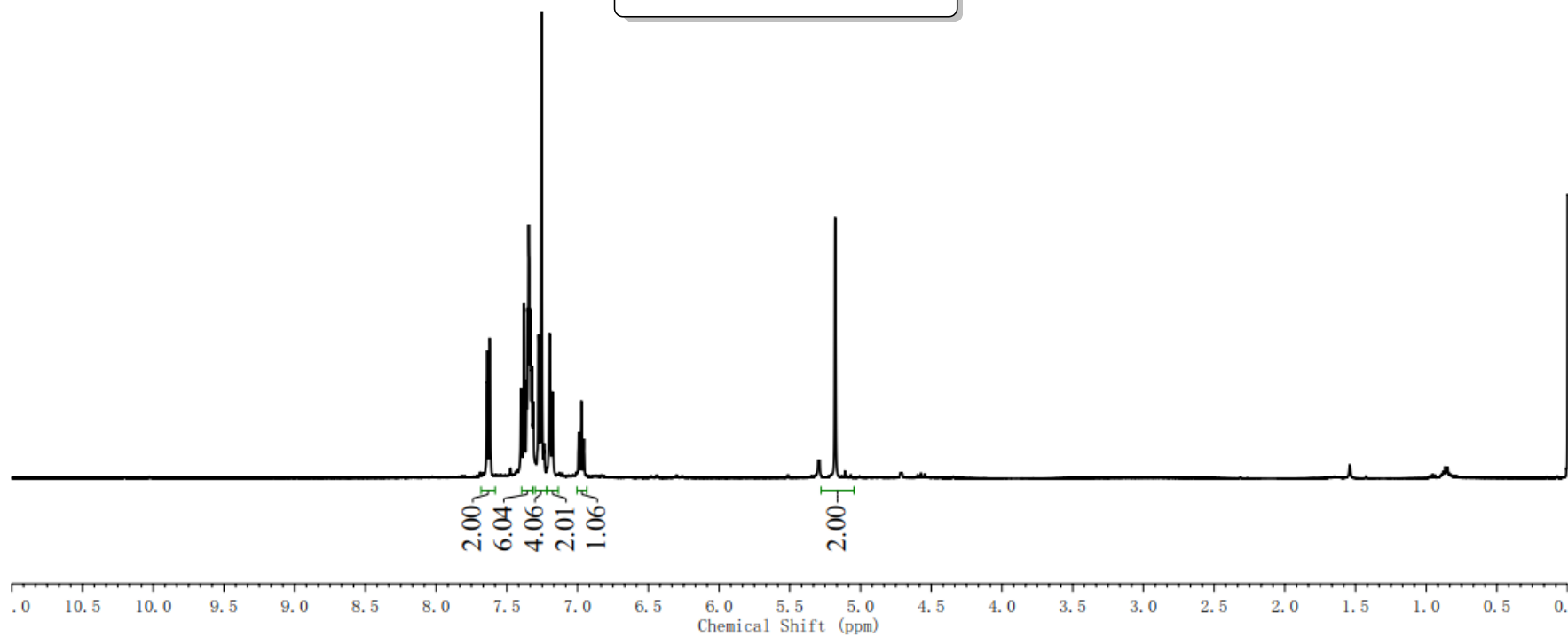
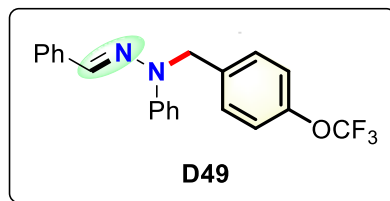


2022-05-30 YY0021 616-ZQL-0527-5

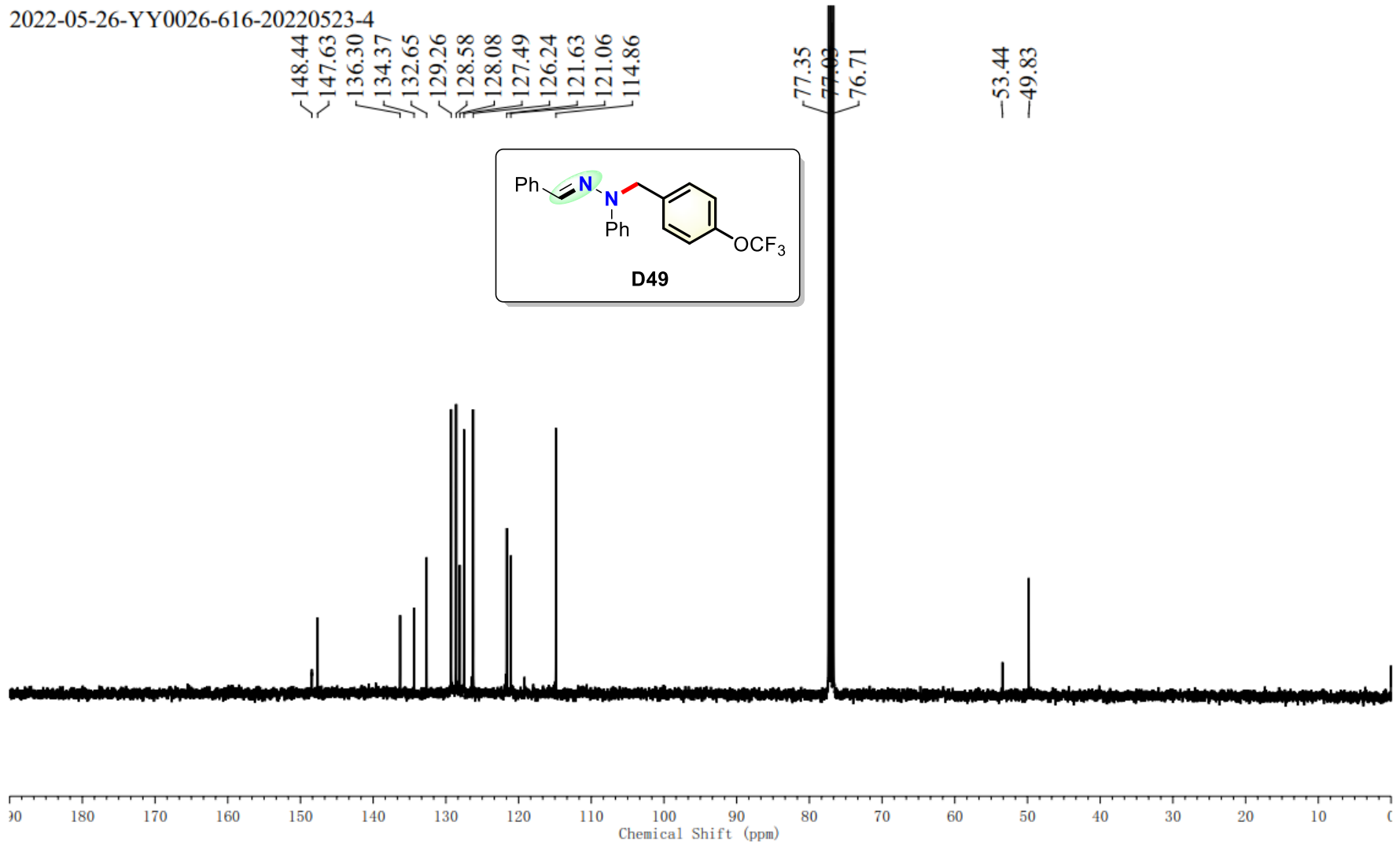


2022-05-23-yy0007-616-zql-4

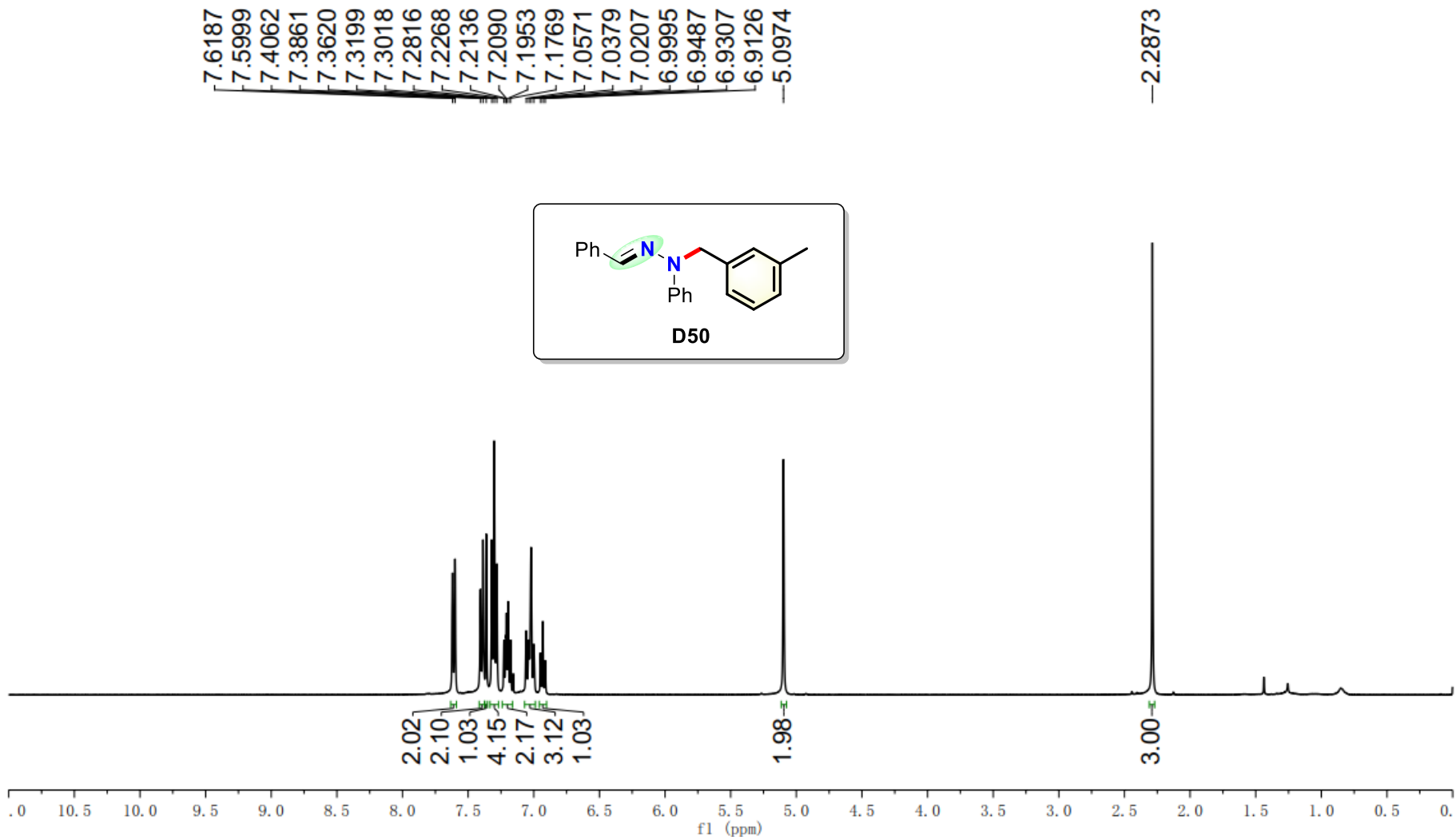
7.6380
7.6198
7.3975
7.3781
7.3757
7.3598
7.3515
7.3457
7.3339
7.3297
7.3224
7.3171
7.3119
7.2768
7.2534
7.2377
7.1977
7.1770
6.9914
6.9737
6.9559
-5.1786



2022-05-26-YY0026-616-20220523-4



2022-9-31 yy0060-616-lqq-d3



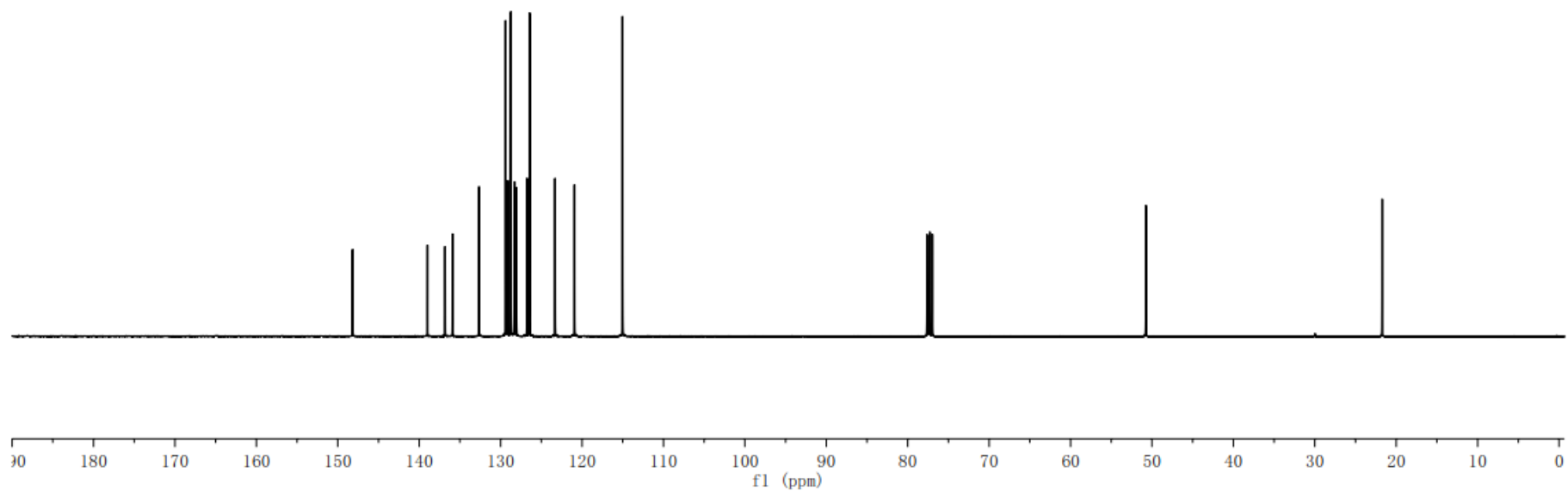
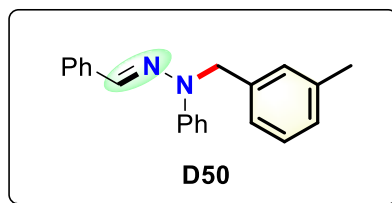
S242

2022-09-30 yy0006 616-lqq-D3

148.17
139.00
136.83
135.92
132.65
129.39
129.14
128.74
128.32
128.06
126.77
126.44
123.32
120.93
115.02

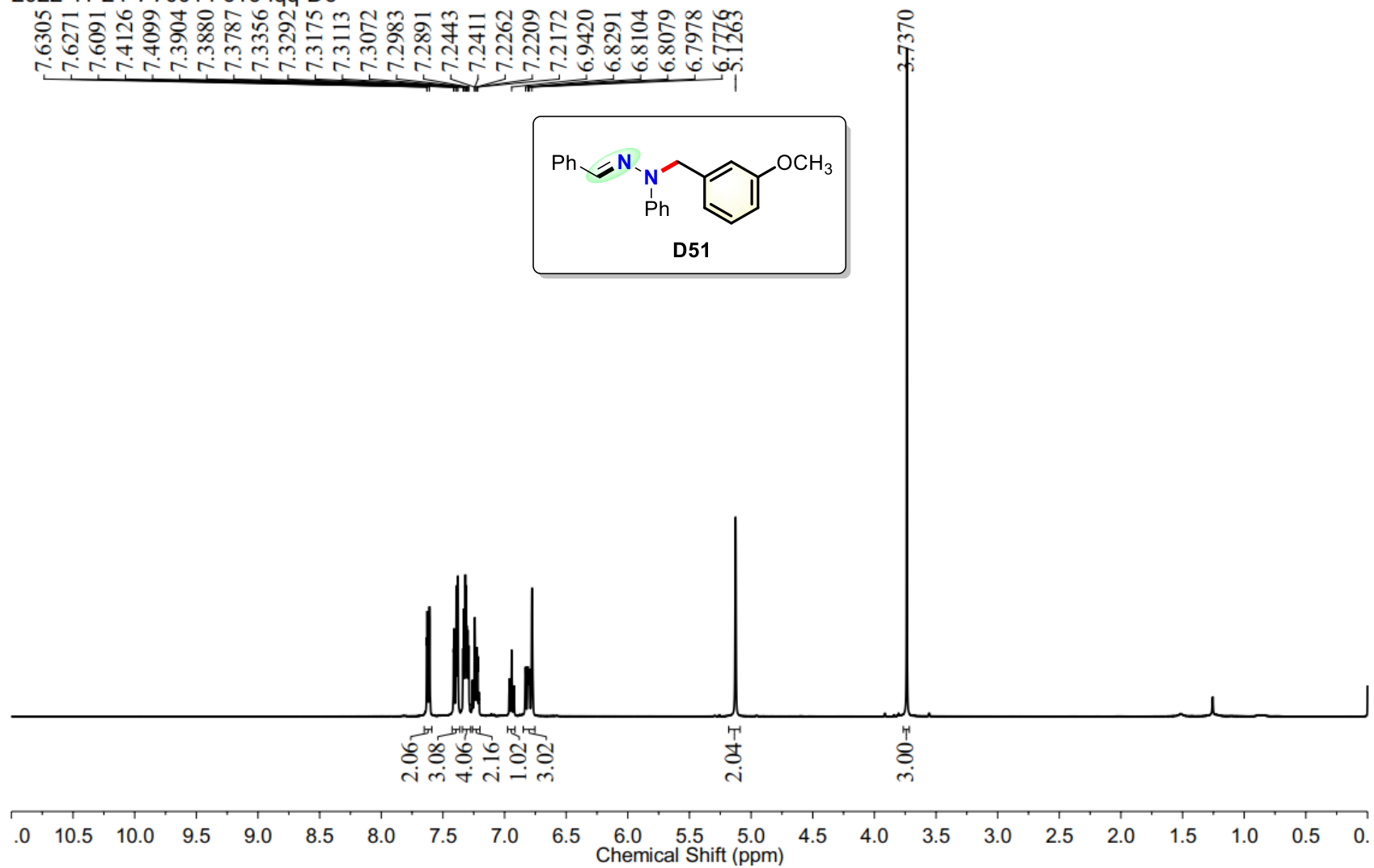
50.75

21.73



S243

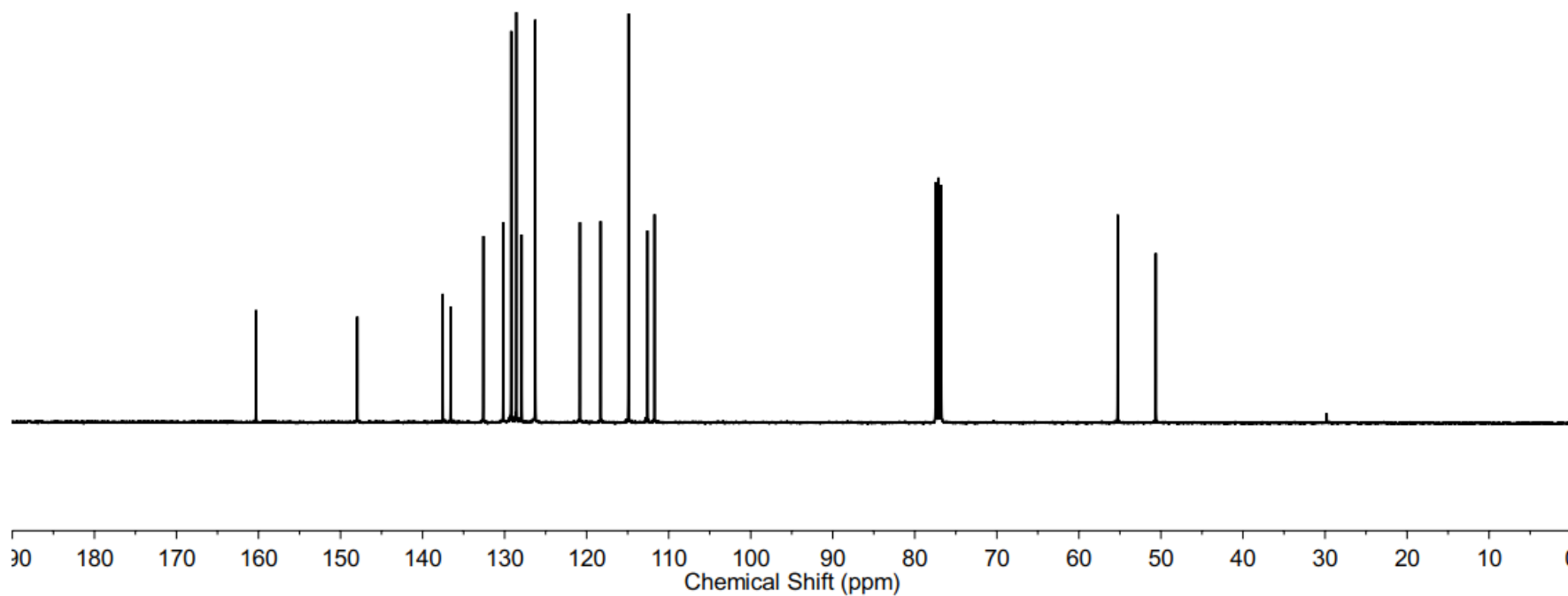
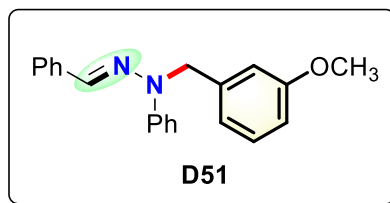
2022-11-24 YY0014 616-lqq-D6



2022-11-30-yy0010-616-d6

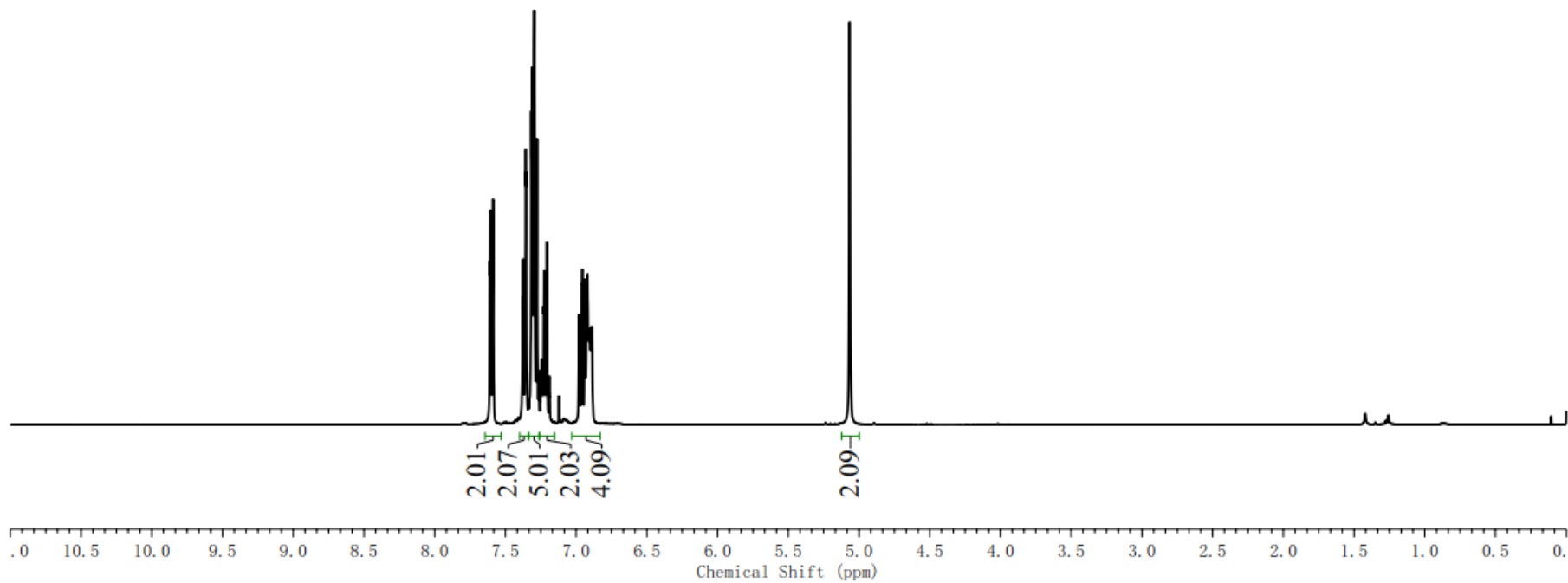
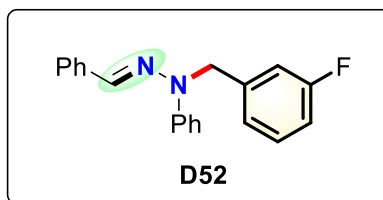
160.29
147.95
137.54
136.57
132.59
130.16
129.20
128.56
127.92
126.26
120.82
118.30
114.89
112.58
111.72

55.27
50.63

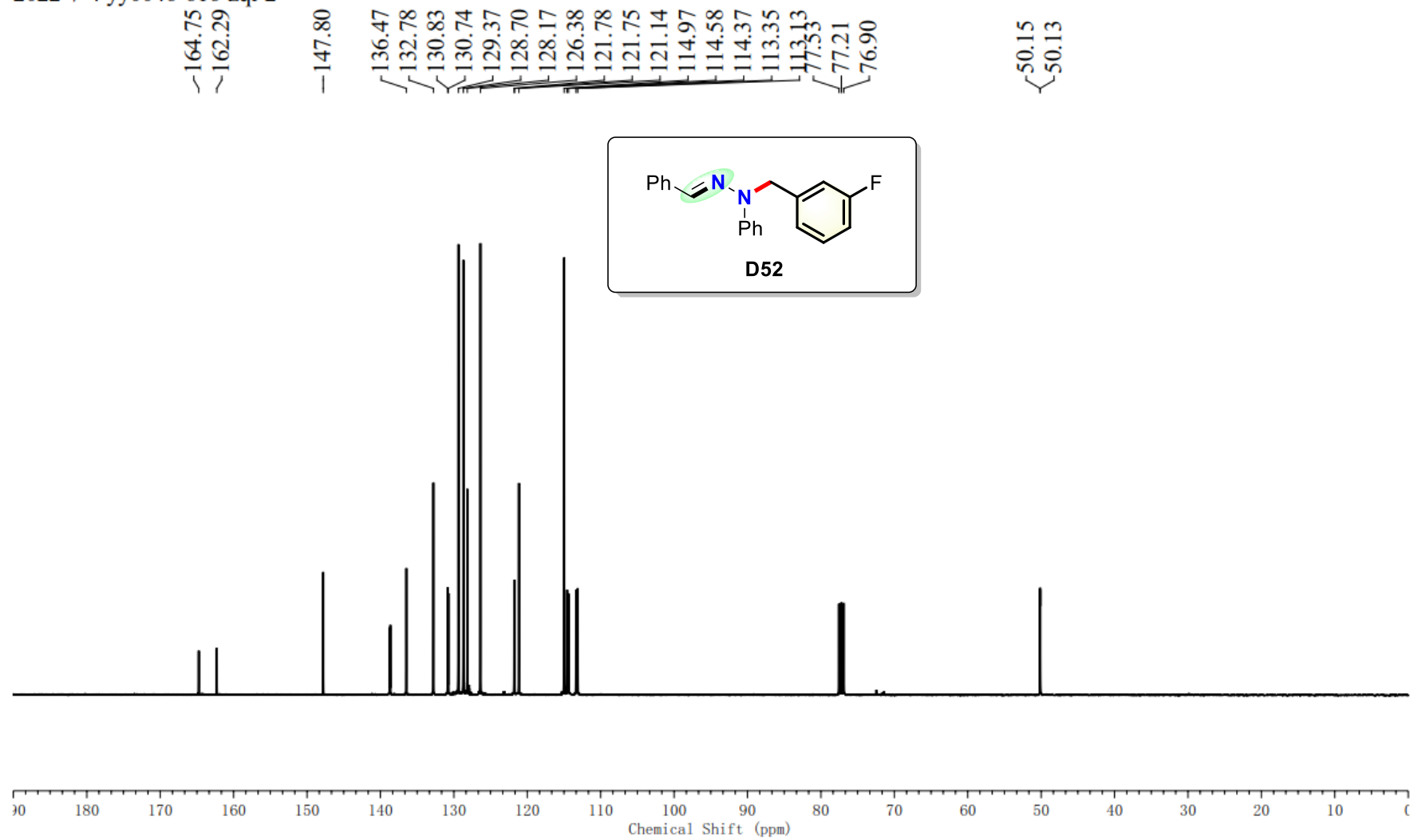


2022-07-02-yy0019-616-zql-1

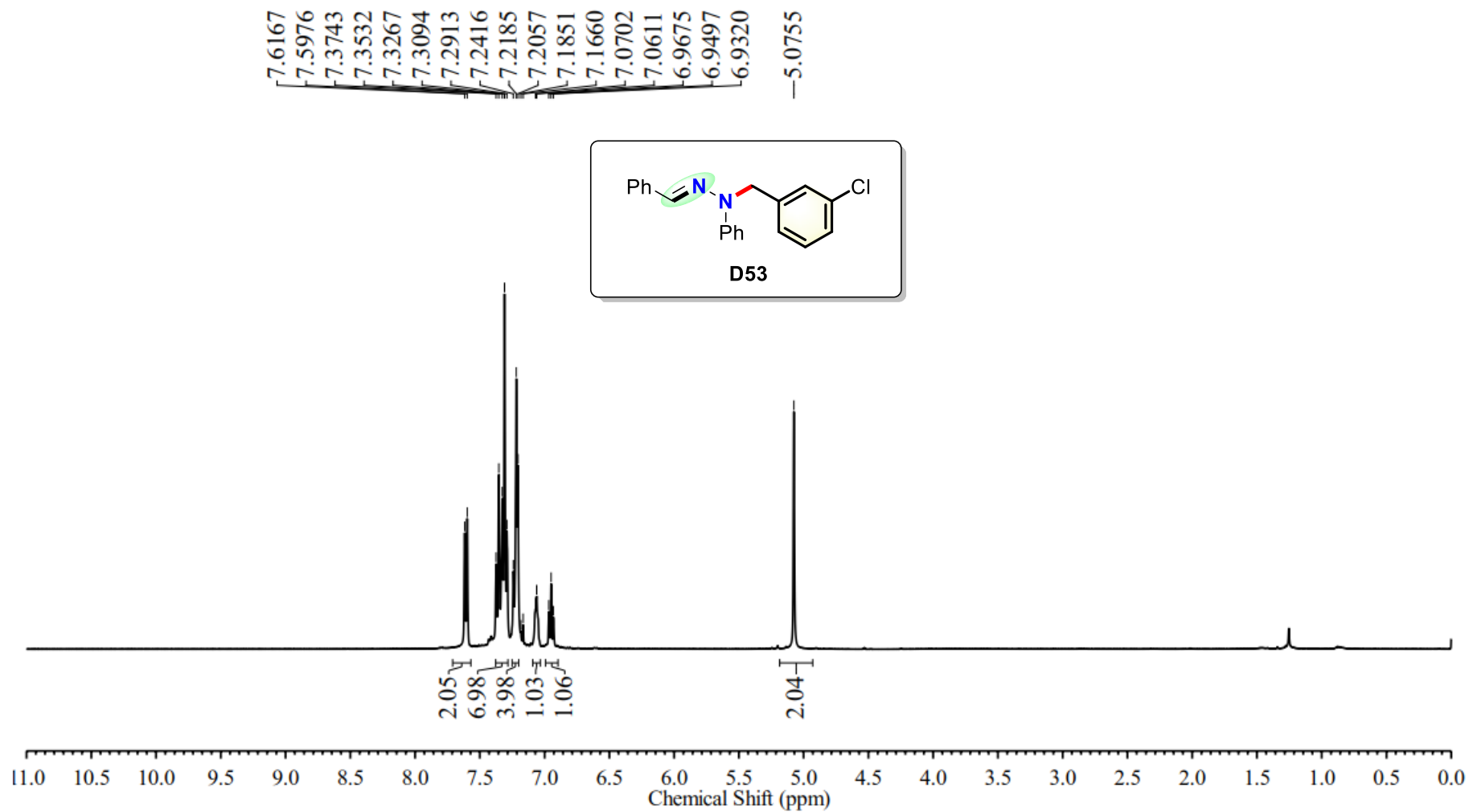
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7.6060
7.5880
7.3782
7.3753
7.3559
7.3534
7.3166
7.3102
7.2967
7.2768
7.2292
7.2260
7.2225
7.2075
6.9775
6.9582
6.9557
6.9377
6.9351
6.9230
6.9197
6.9108
6.9029
6.8943
6.8895
5.0659



2022-7-4 yy0046-616-zql-2

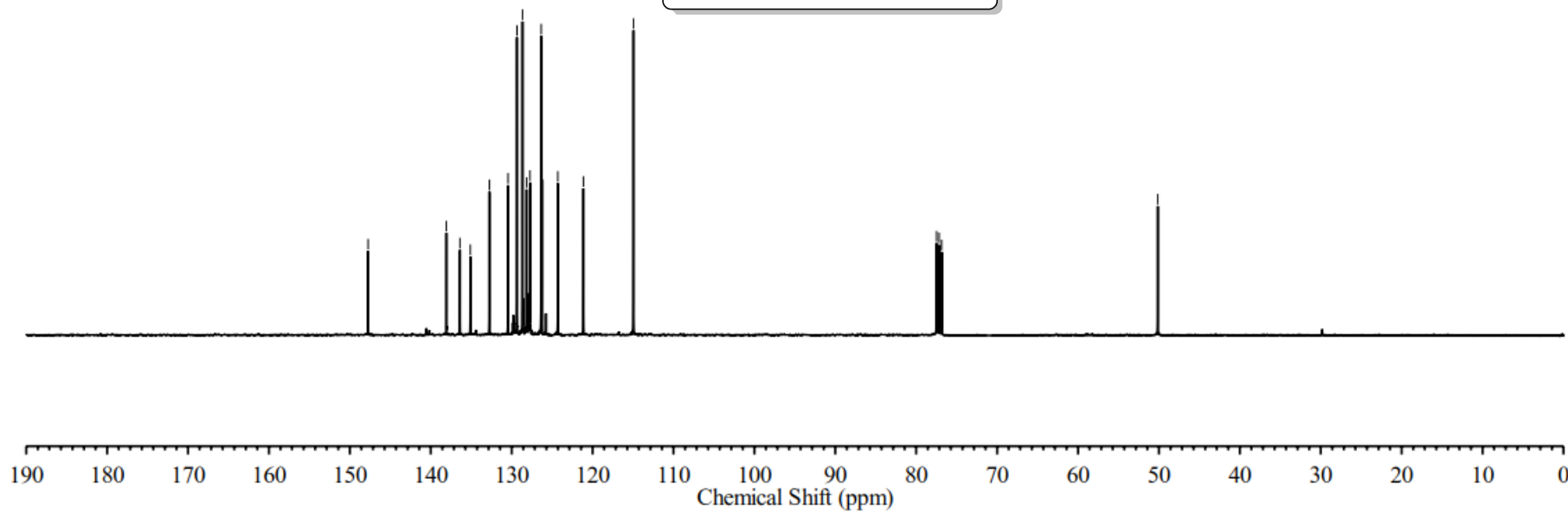
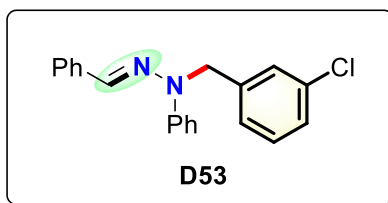


2022-6-13 YY0018-616-ZQL-13-1

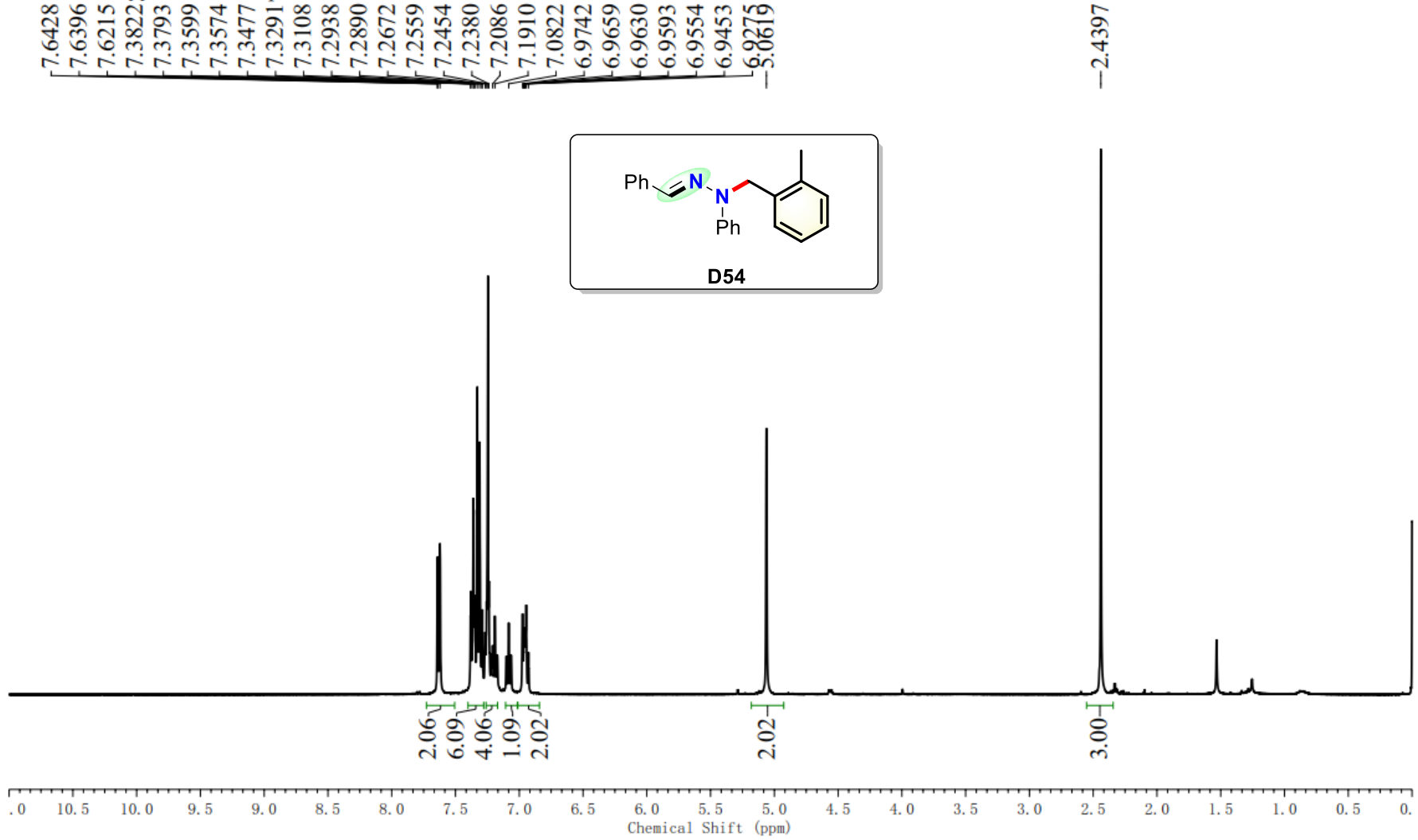


2022-6-16 yy0005-616-zql-613-1

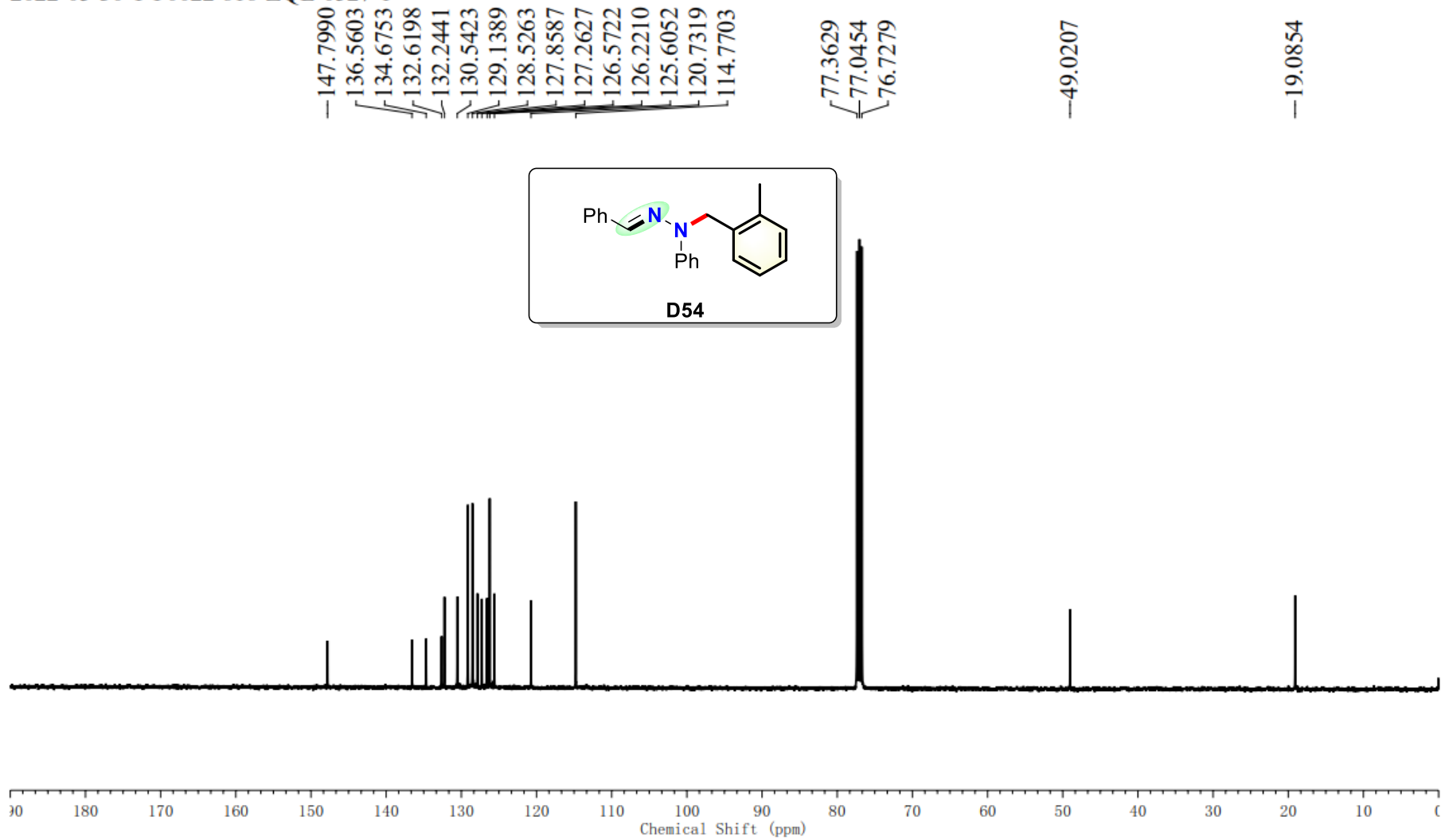
147.7425
138.0737
136.3904
135.1138
132.7456
130.4477
129.3435
128.6566
128.1467
127.7420
126.3505
126.2490
124.2994
121.1322
114.9449
77.4823
77.1647
76.8469
50.1438



2022-05-28-yy0007-616-zql-1

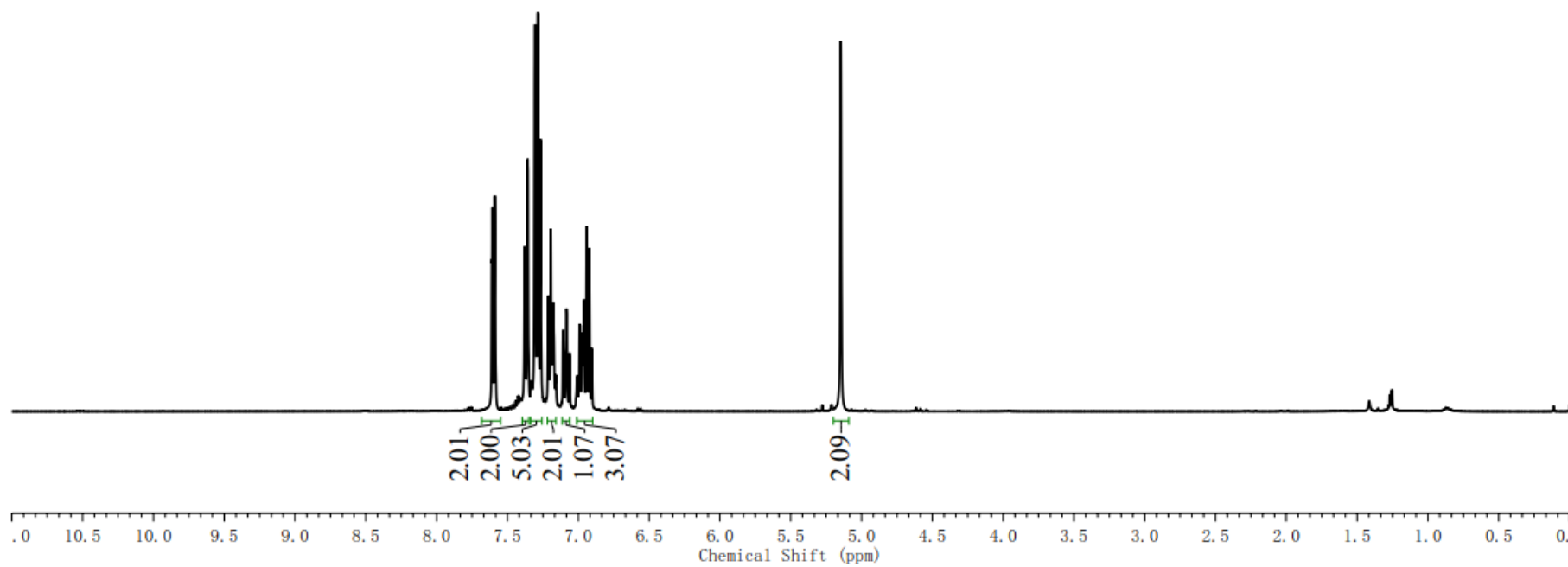
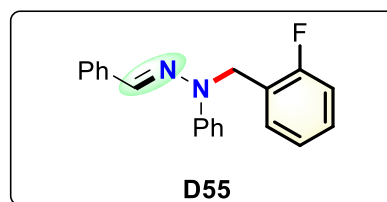


2022-05-30 YY0022 616-ZQL-0527-1



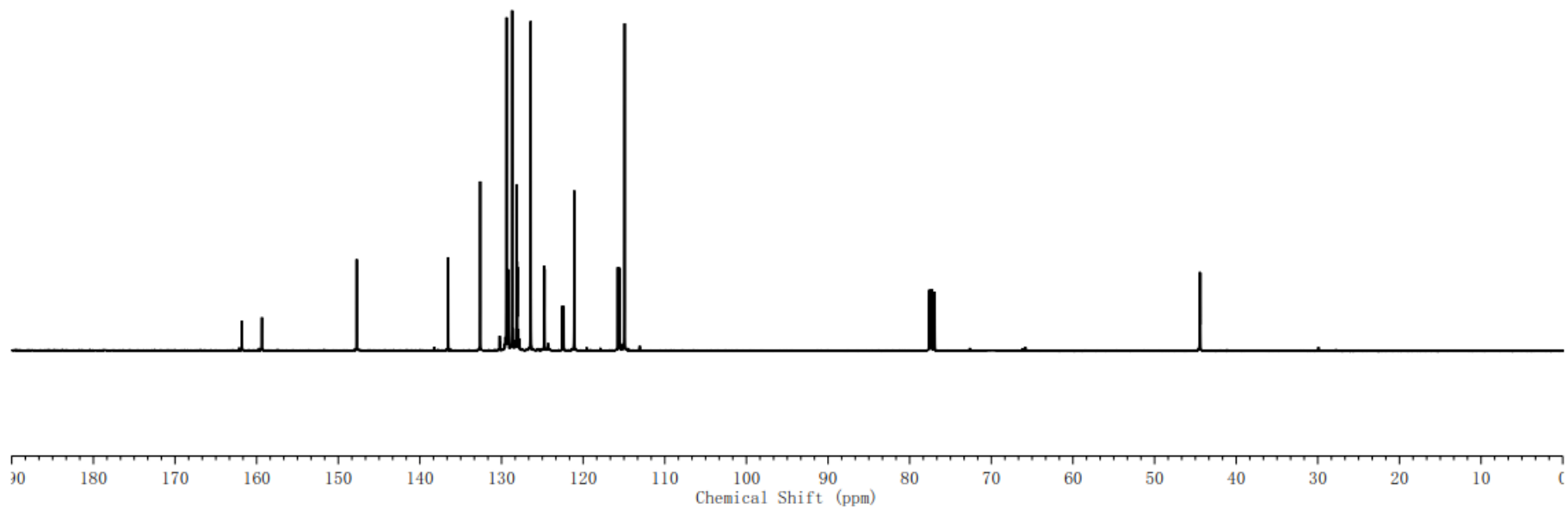
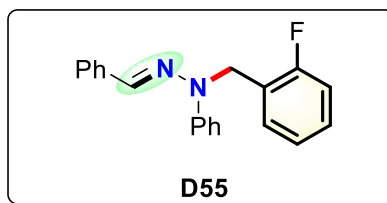
2022-07-02-yy0019-616-zql-3

7.6100
7.6067
7.5886
7.3774
7.3577
7.3055
7.2854
7.2652
7.2117
7.2089
7.1979
7.1933
7.1888
7.1751
7.1060
7.0830
7.0621
6.9901
6.9865
6.9719
6.9674
6.9601
6.9578
6.9402
6.9222
6.9042
5.1464

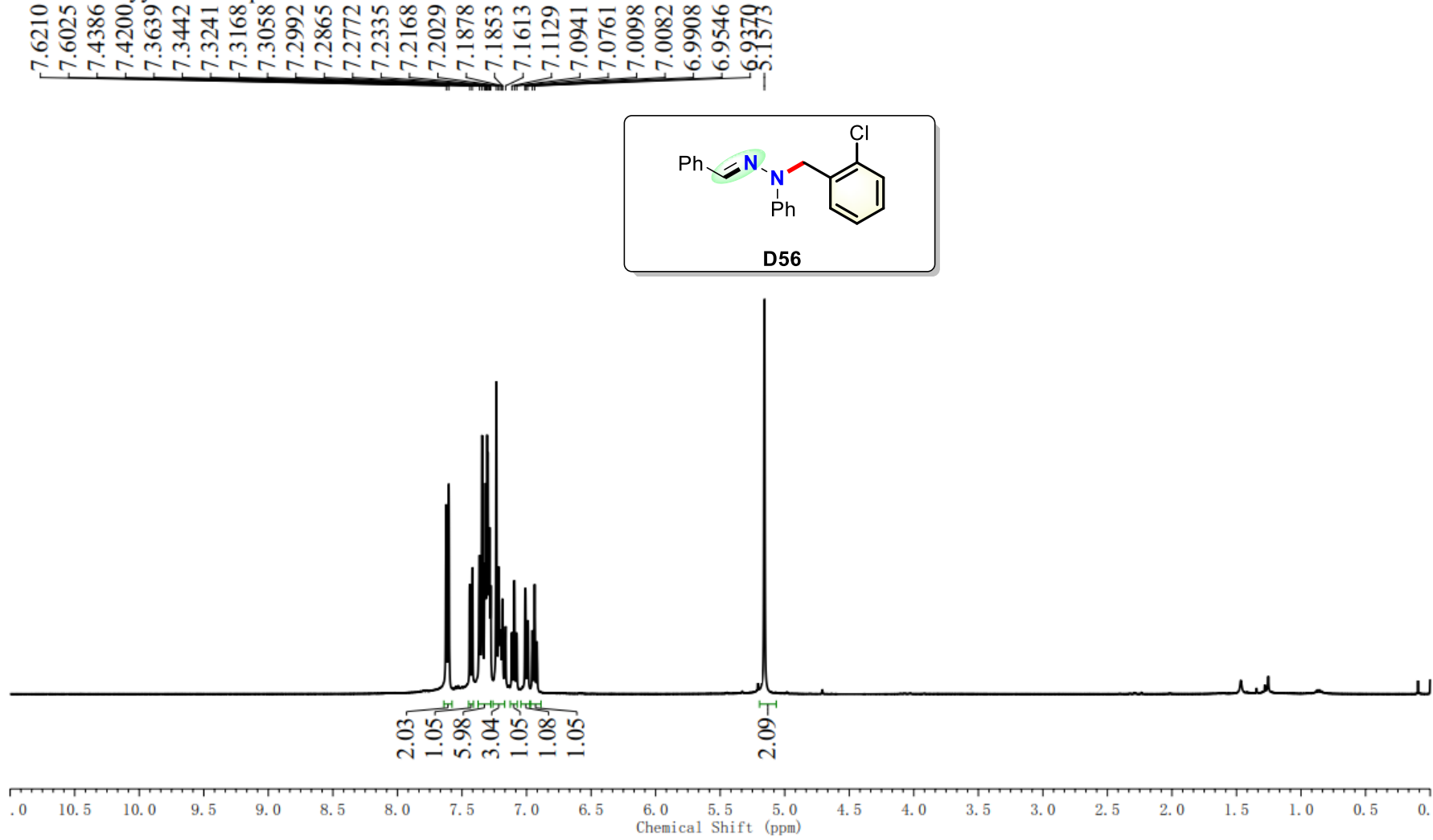


2022-7-4 yy0046-616-zql-1

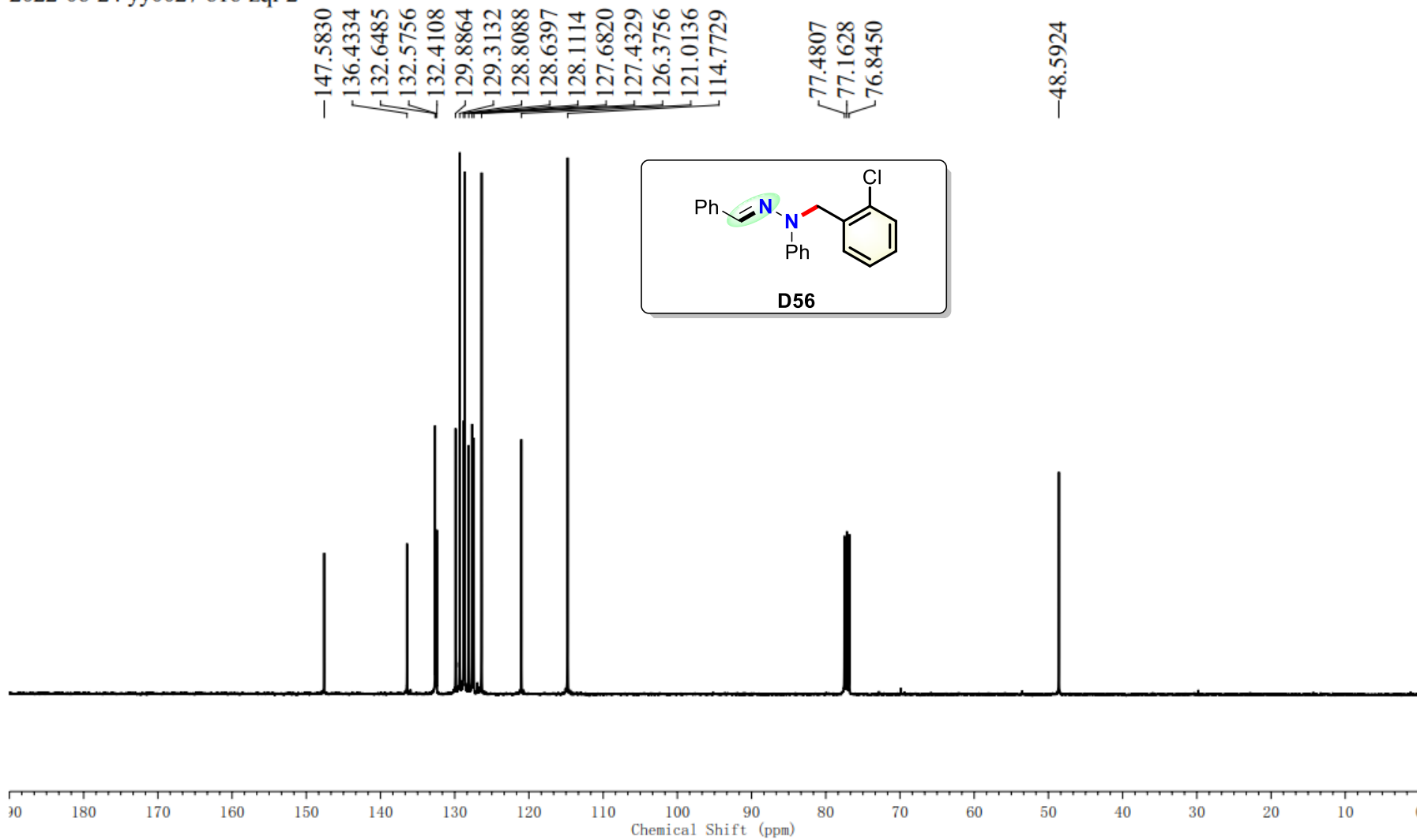
~161.7939
~159.3575
—147.7362
136.5355
132.6088
129.3860
129.2073
129.1287
128.7079
128.1576
128.0926
128.0528
126.4254
124.7587
124.7239
121.0843
115.7618
115.5574
114.9248
77.5752
77.2573
76.9395
44.4473
44.3982



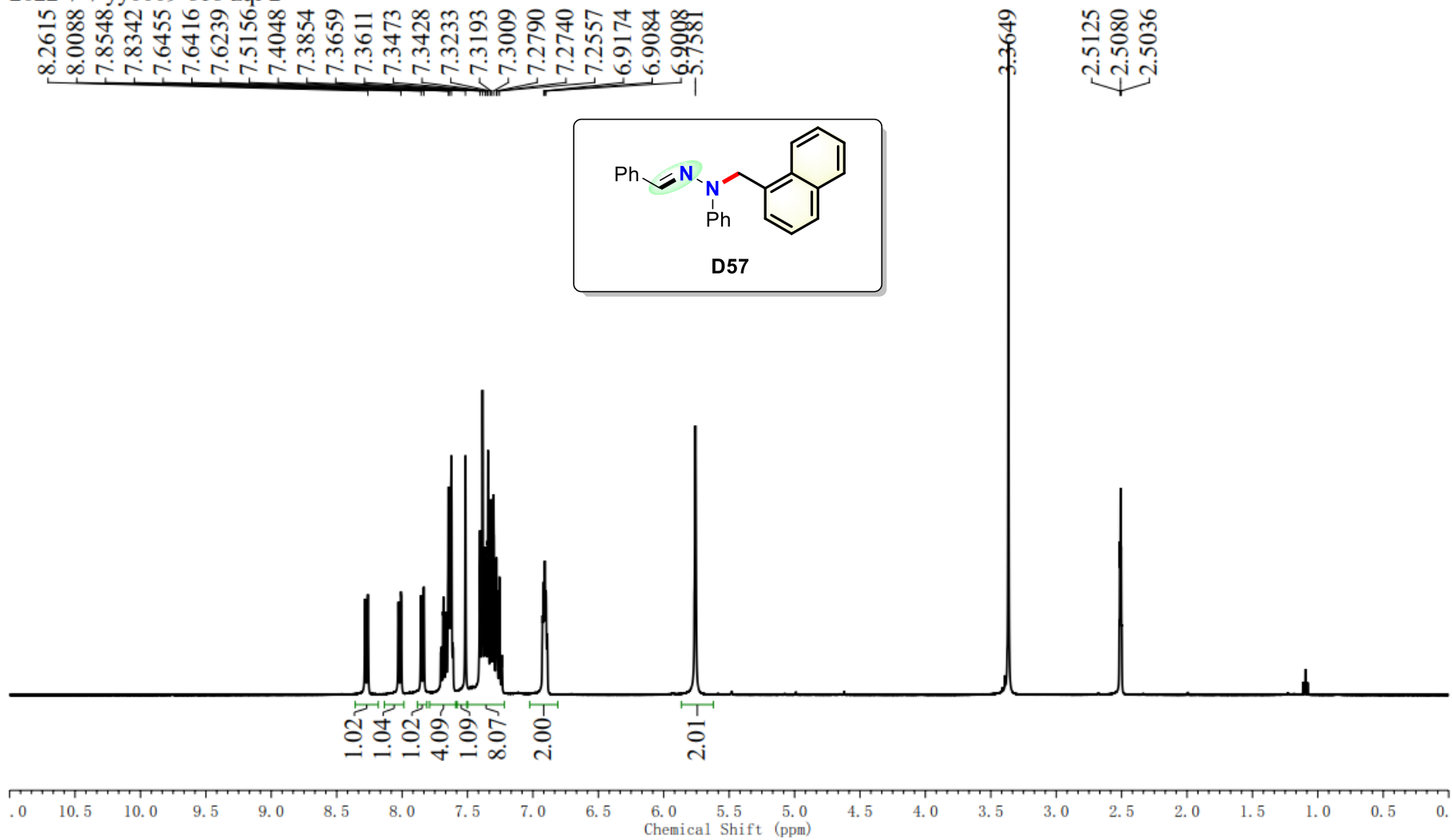
2022-06-22 yy0025 61zql-02



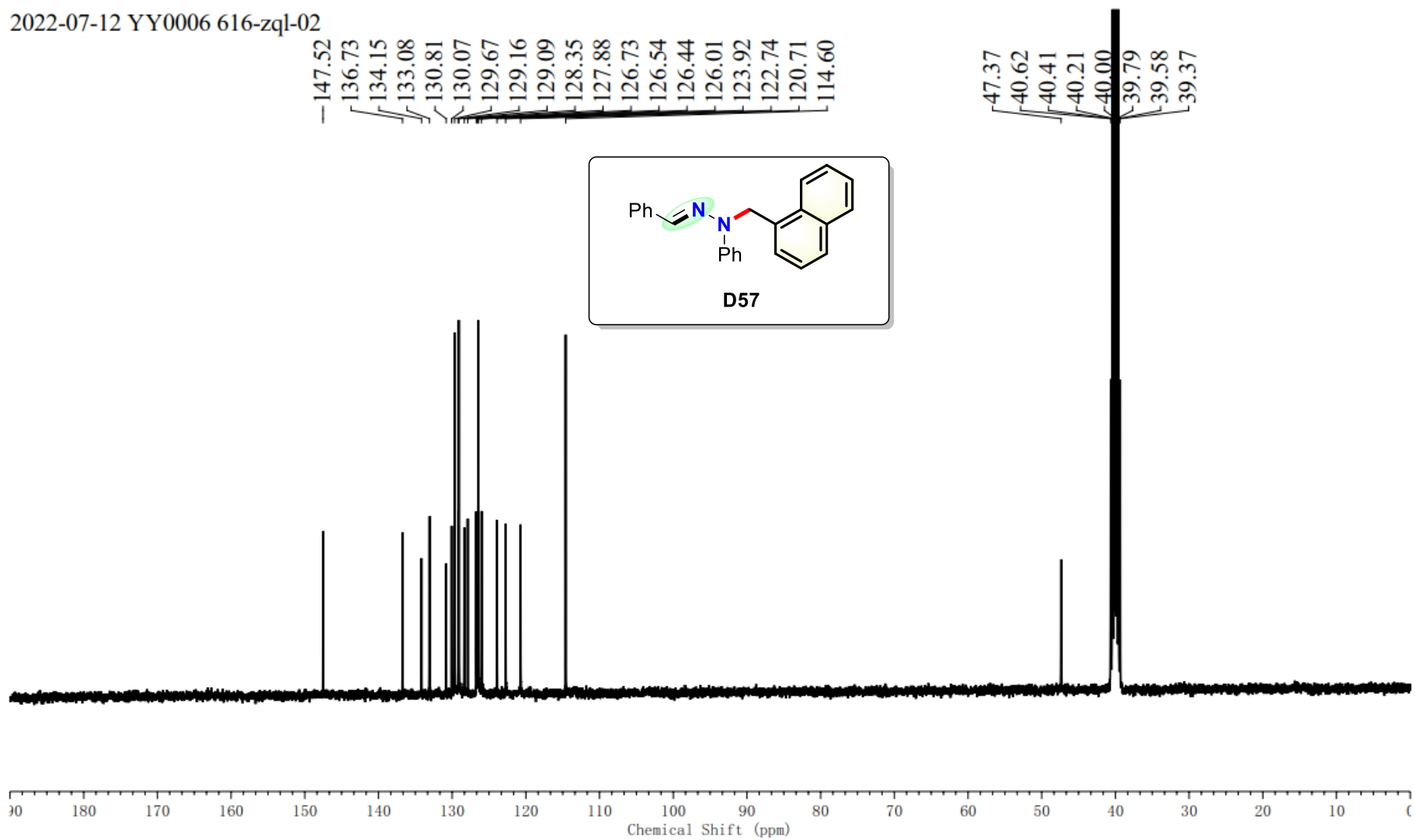
2022-06-24 yy0027 616-zql-2



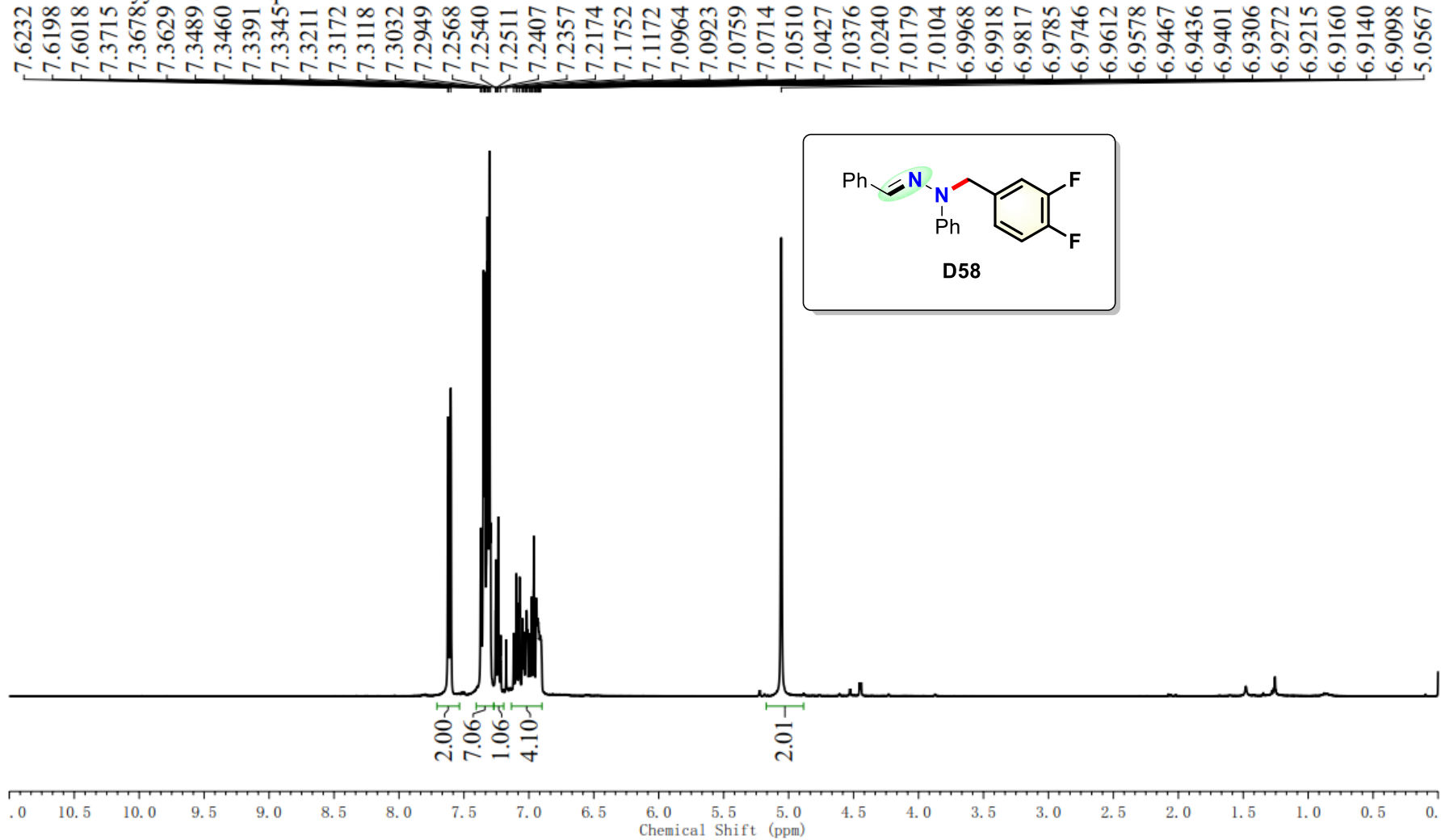
2022-7-7 yy0009-616-zql-2



2022-07-12 YY0006 616-zql-02

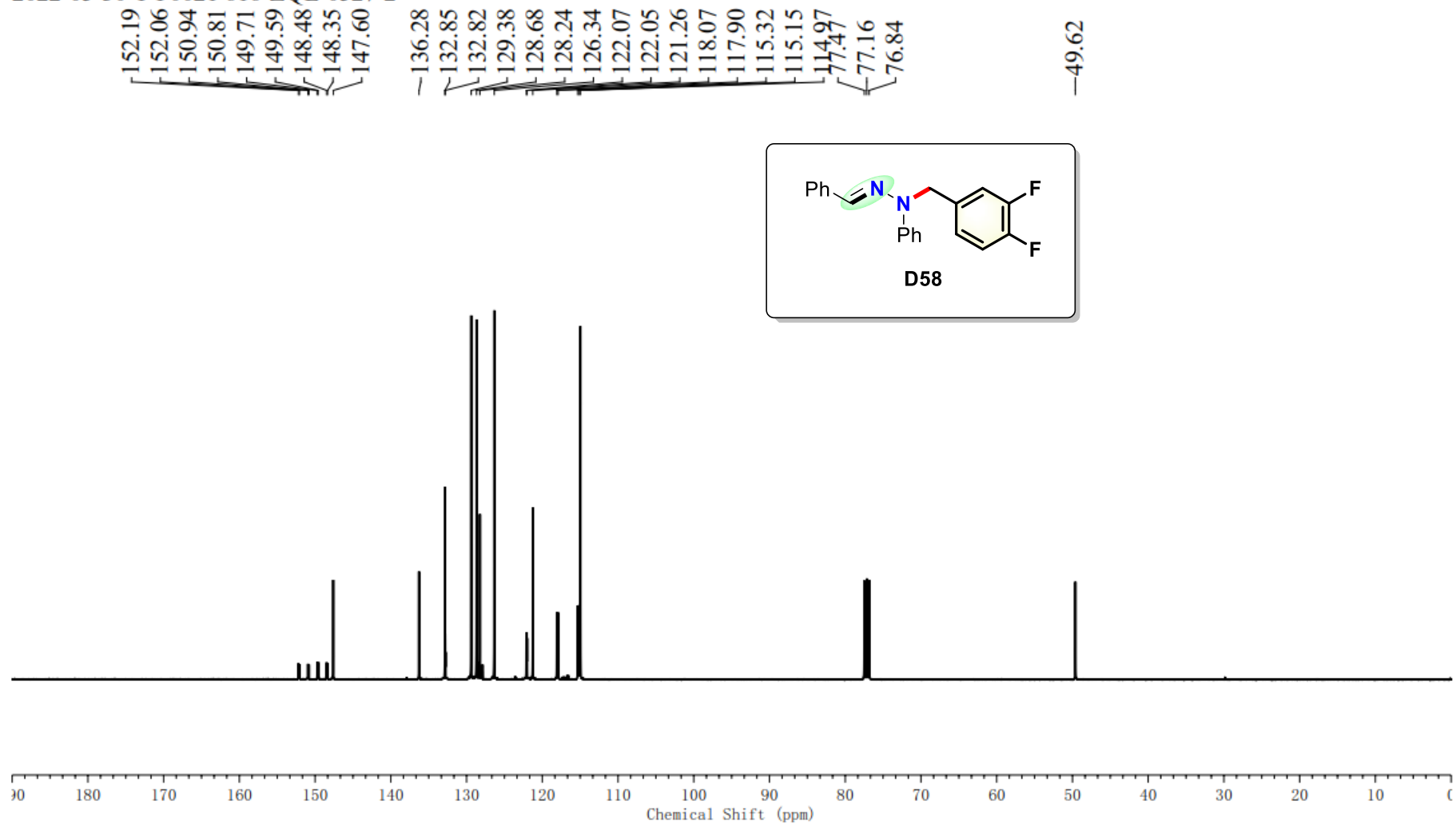


2022-05-28-yy0007-616-zq1-2

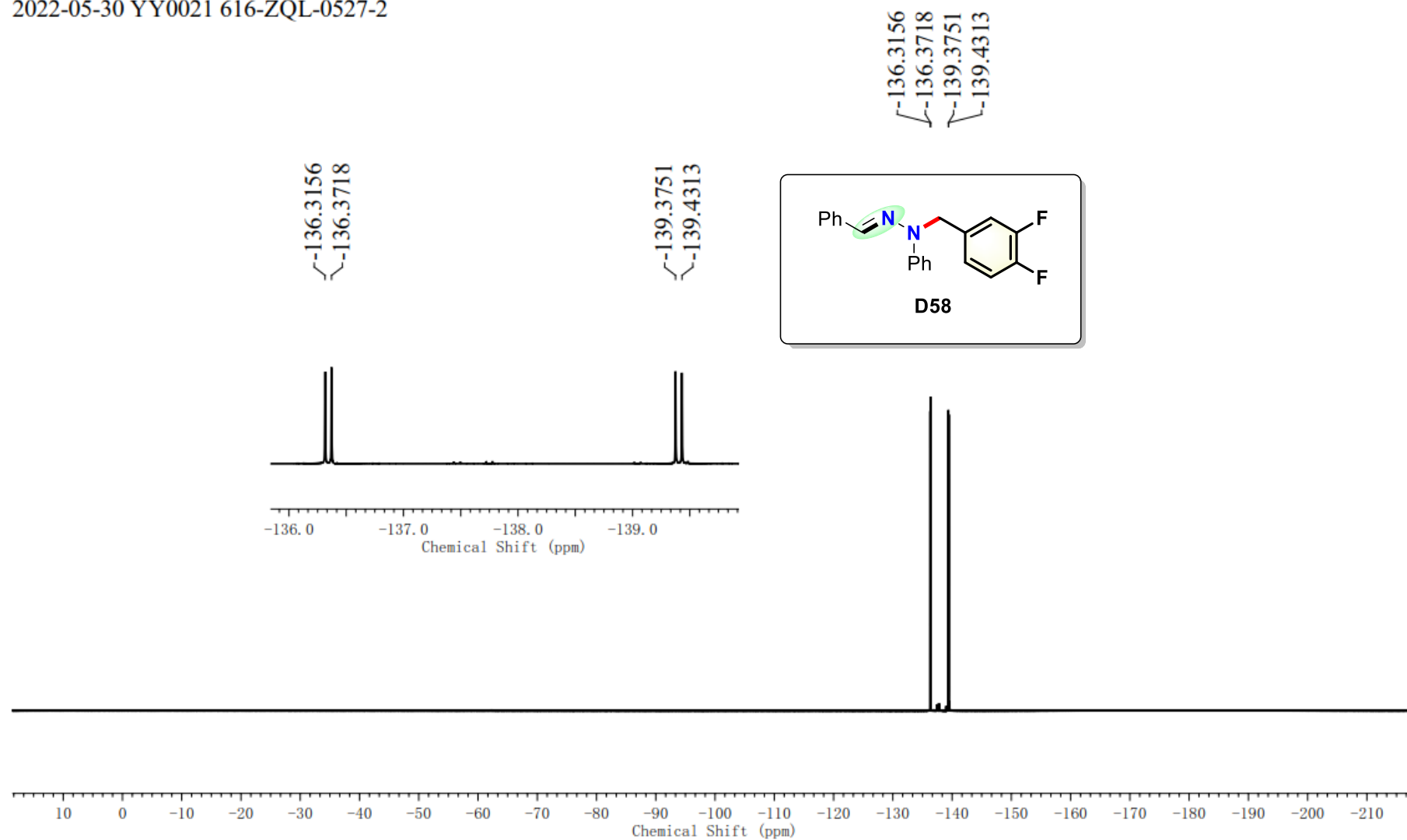


S258

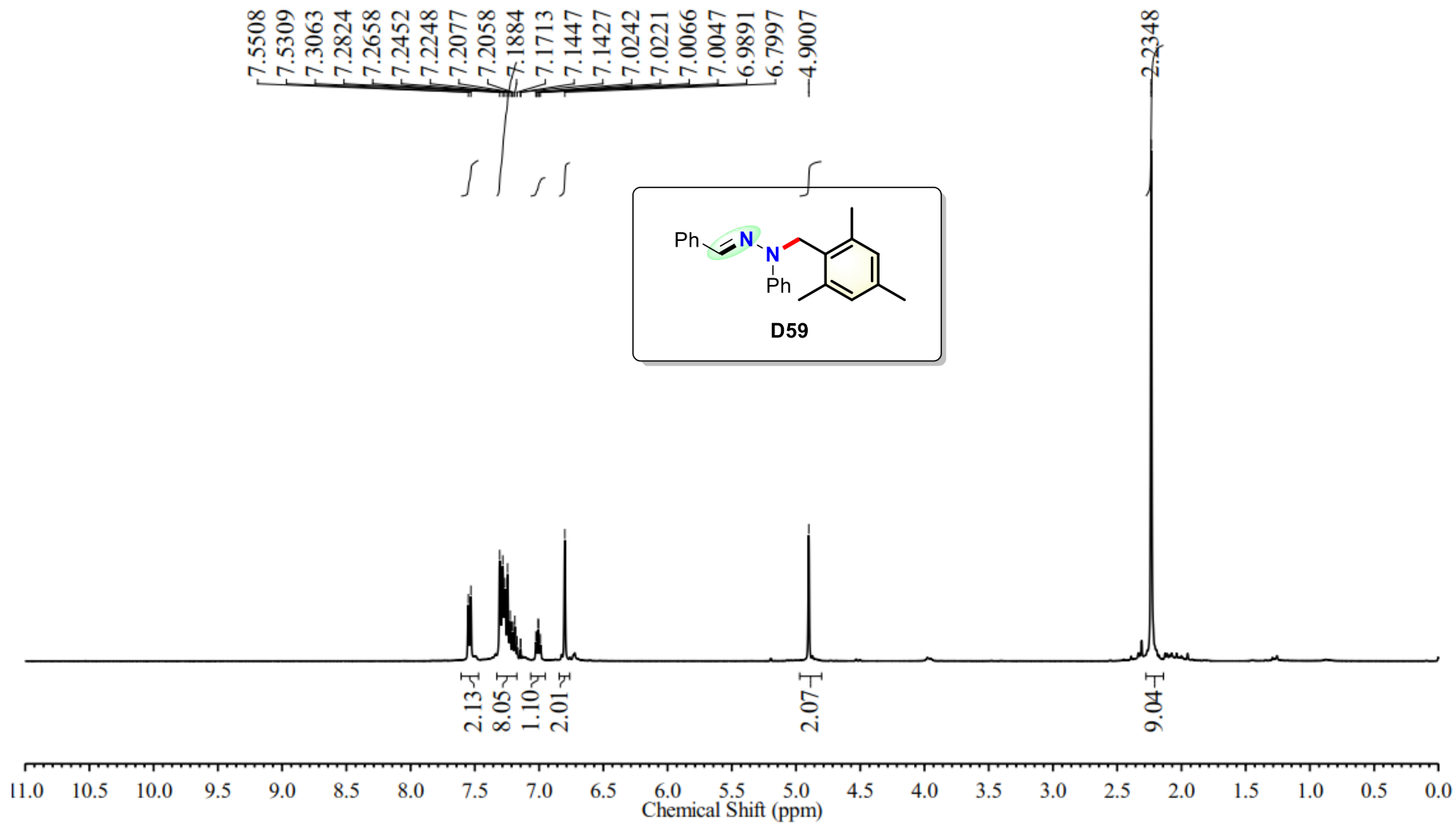
2022-05-30 YY0021 616-ZQL-0527-2



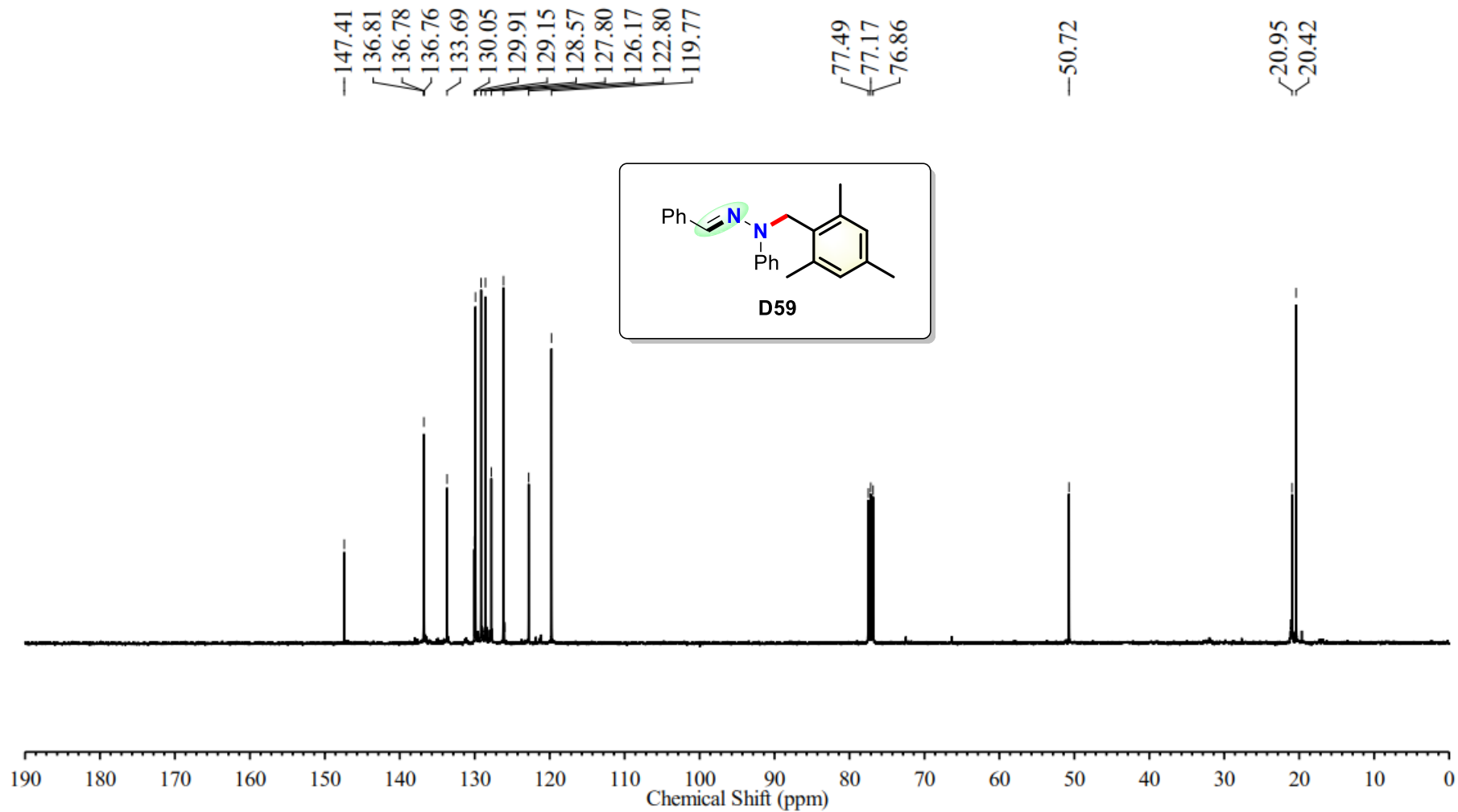
2022-05-30 YY0021 616-ZQL-0527-2



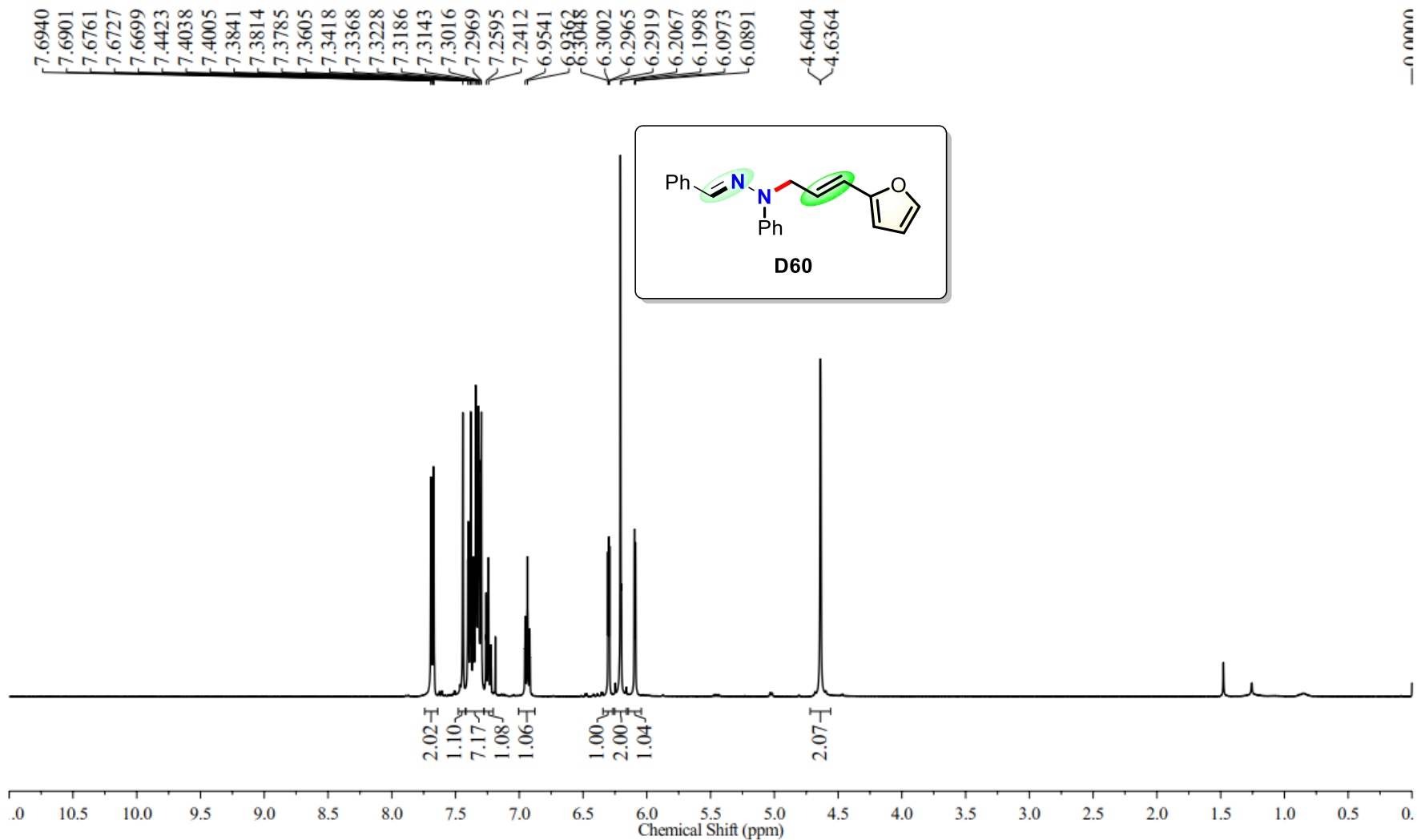
2022-06-20 yy0003-616-zql-3



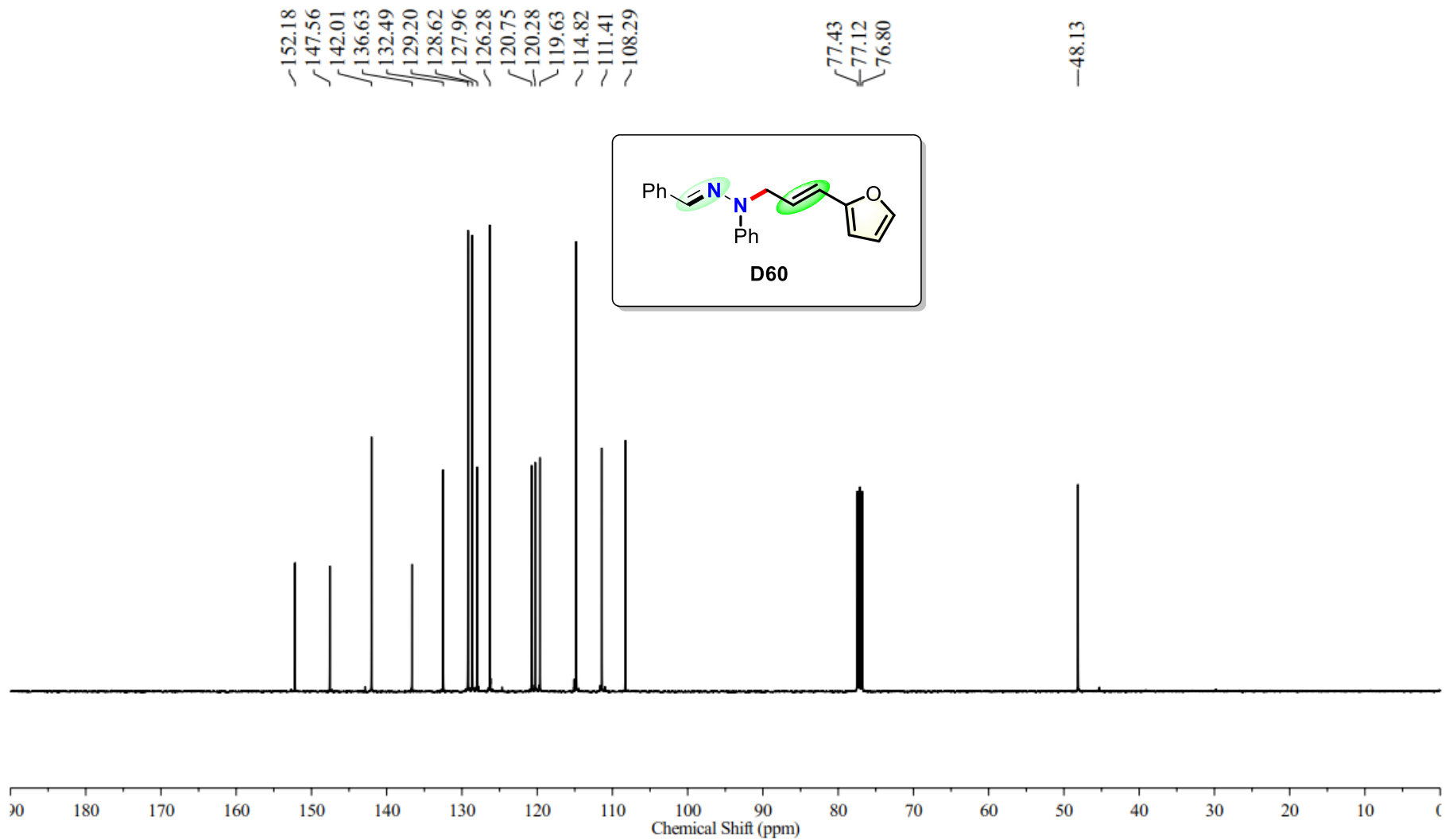
2022-06-21 yy0027 616-zql-3



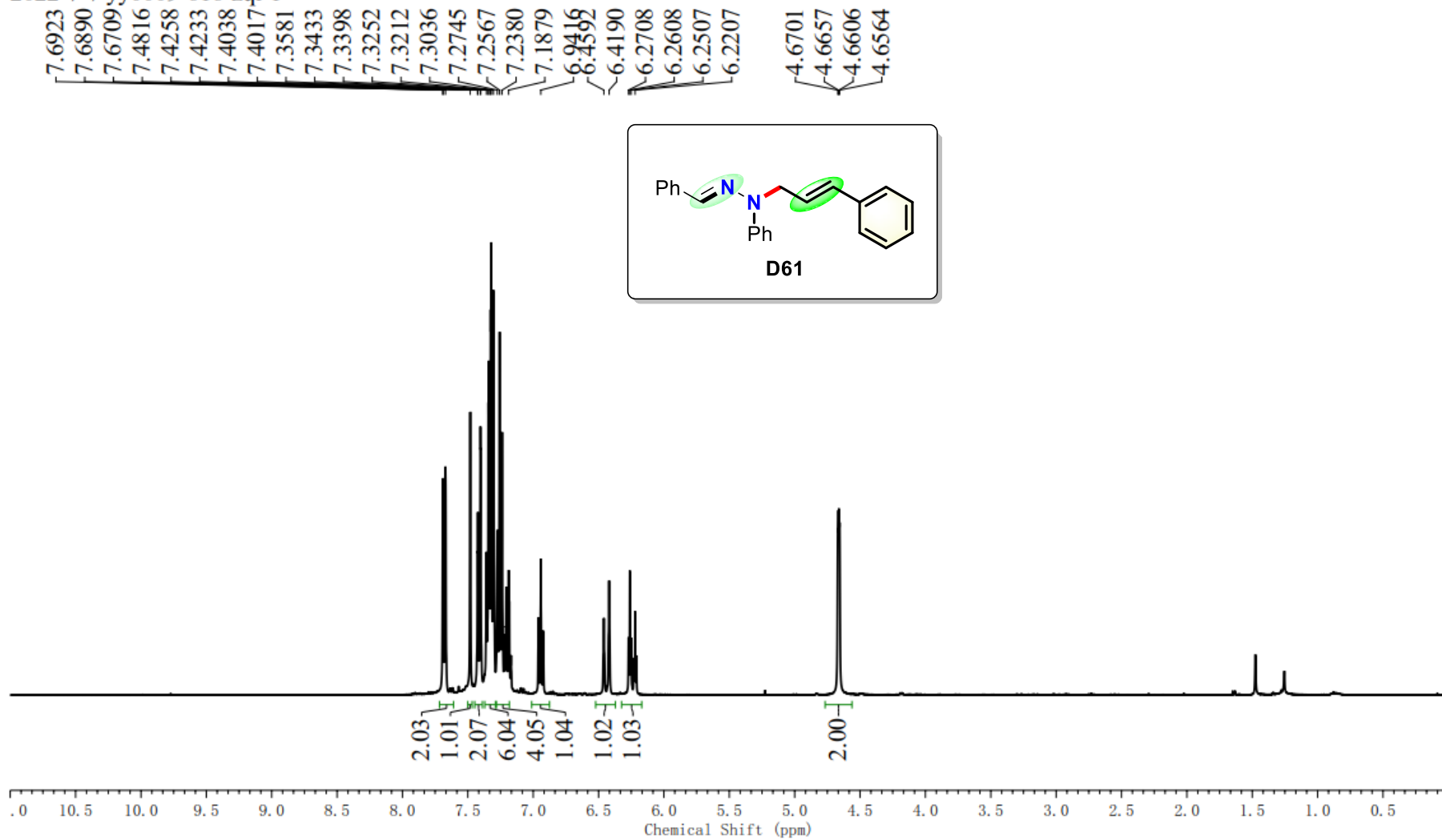
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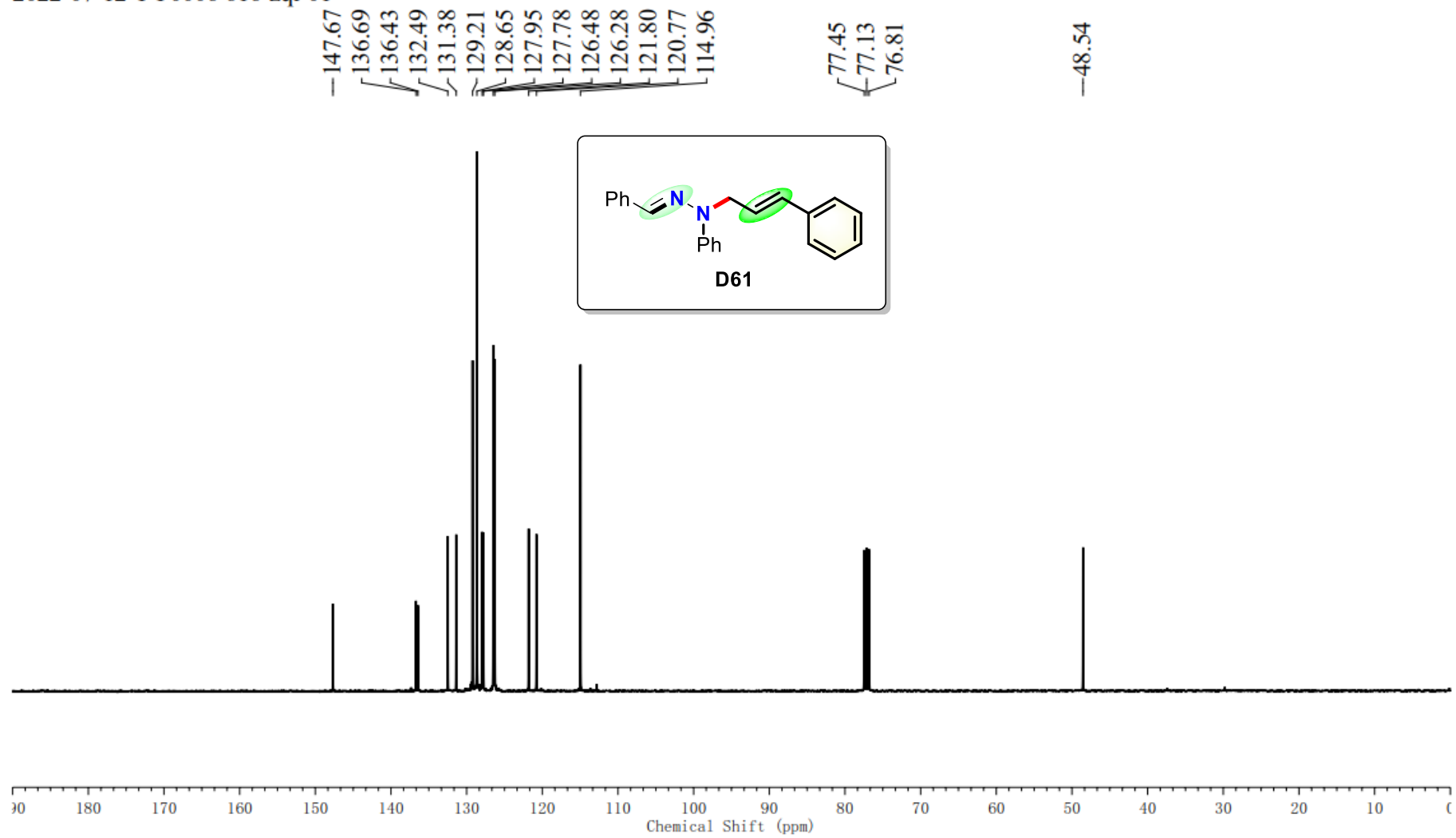
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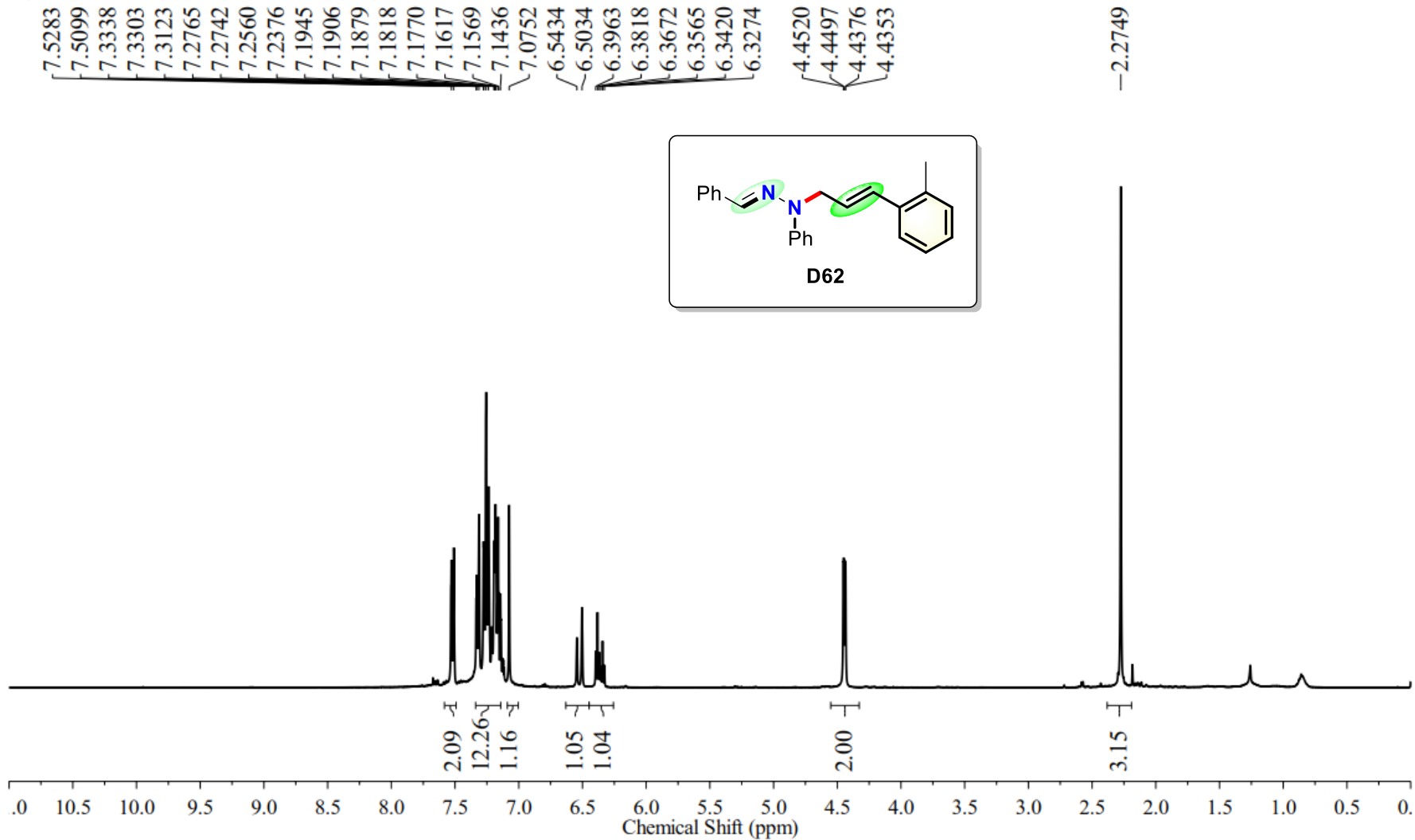
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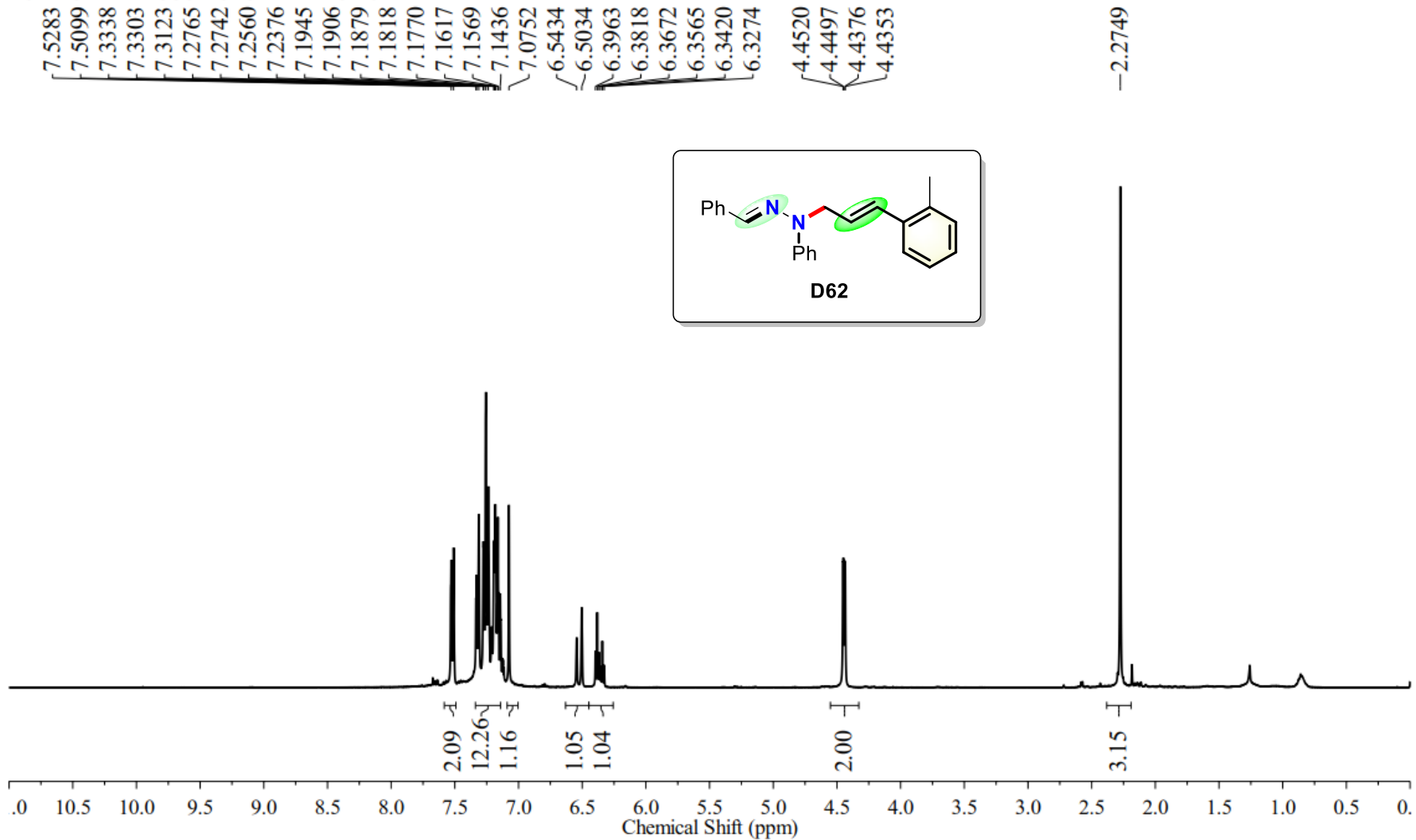
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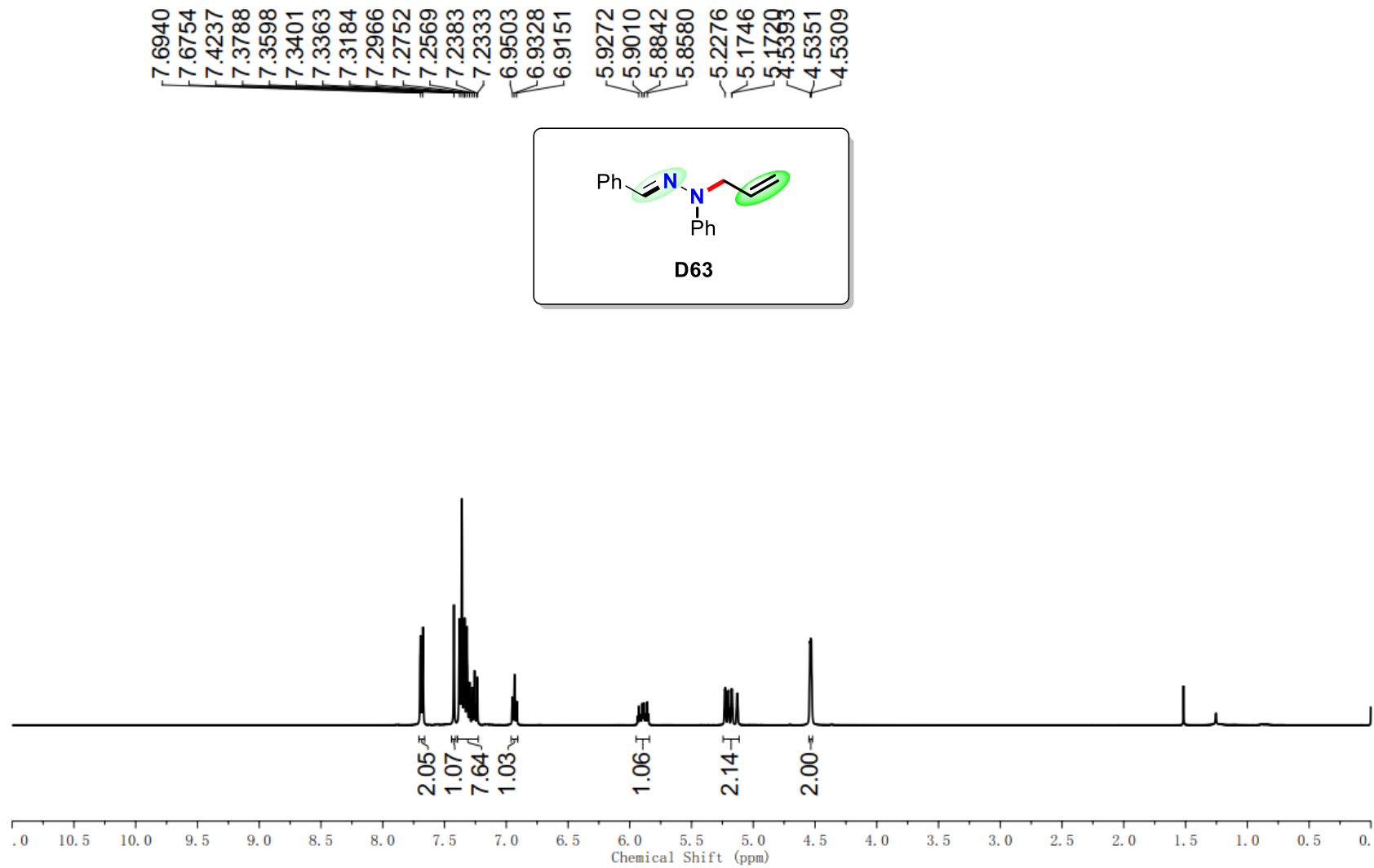
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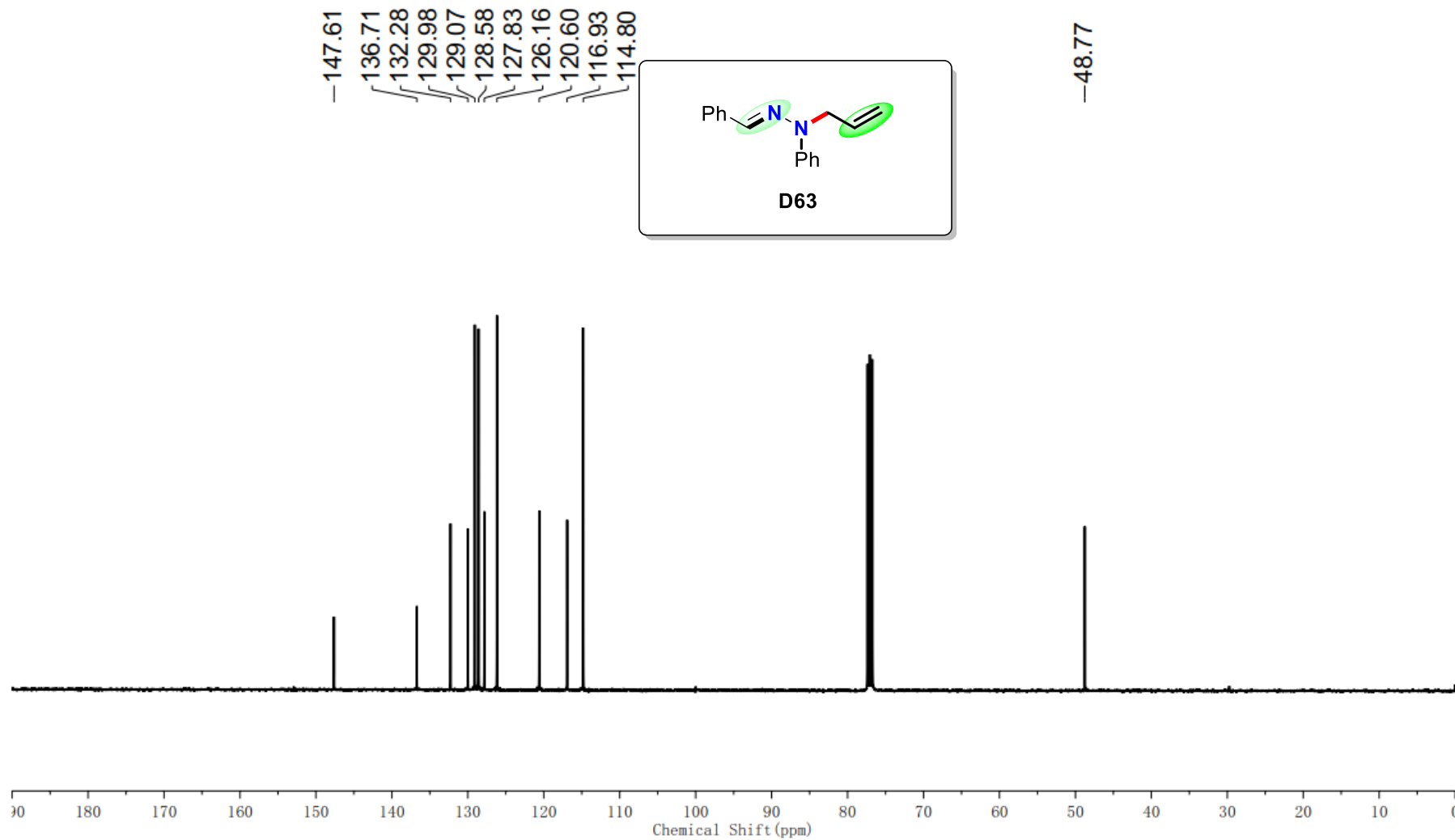
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2022-10-28 yy0029 616-lqq-26

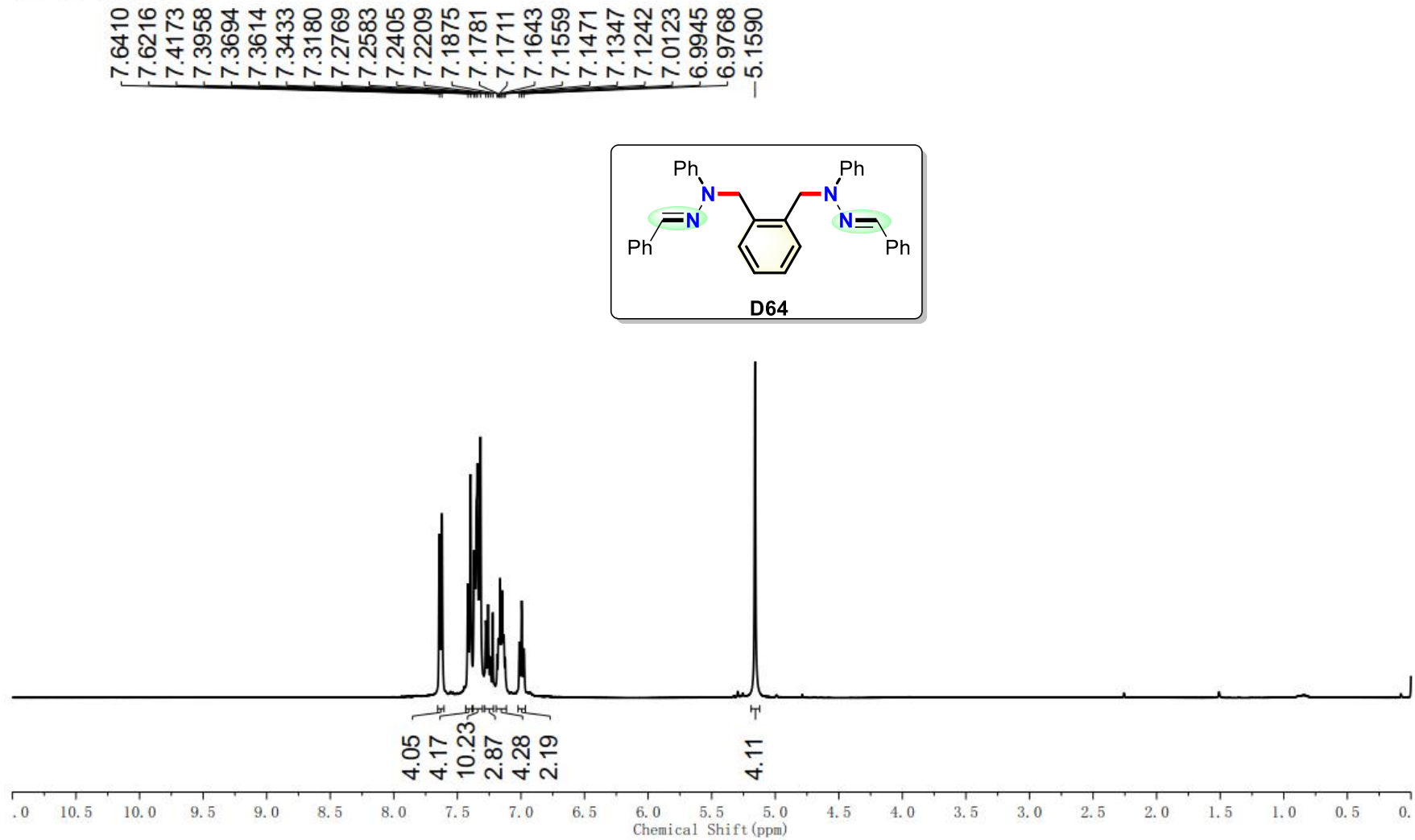


2022-11-08-yy0037-616-d26



S270

2022-11-7-yy0025-616-d24

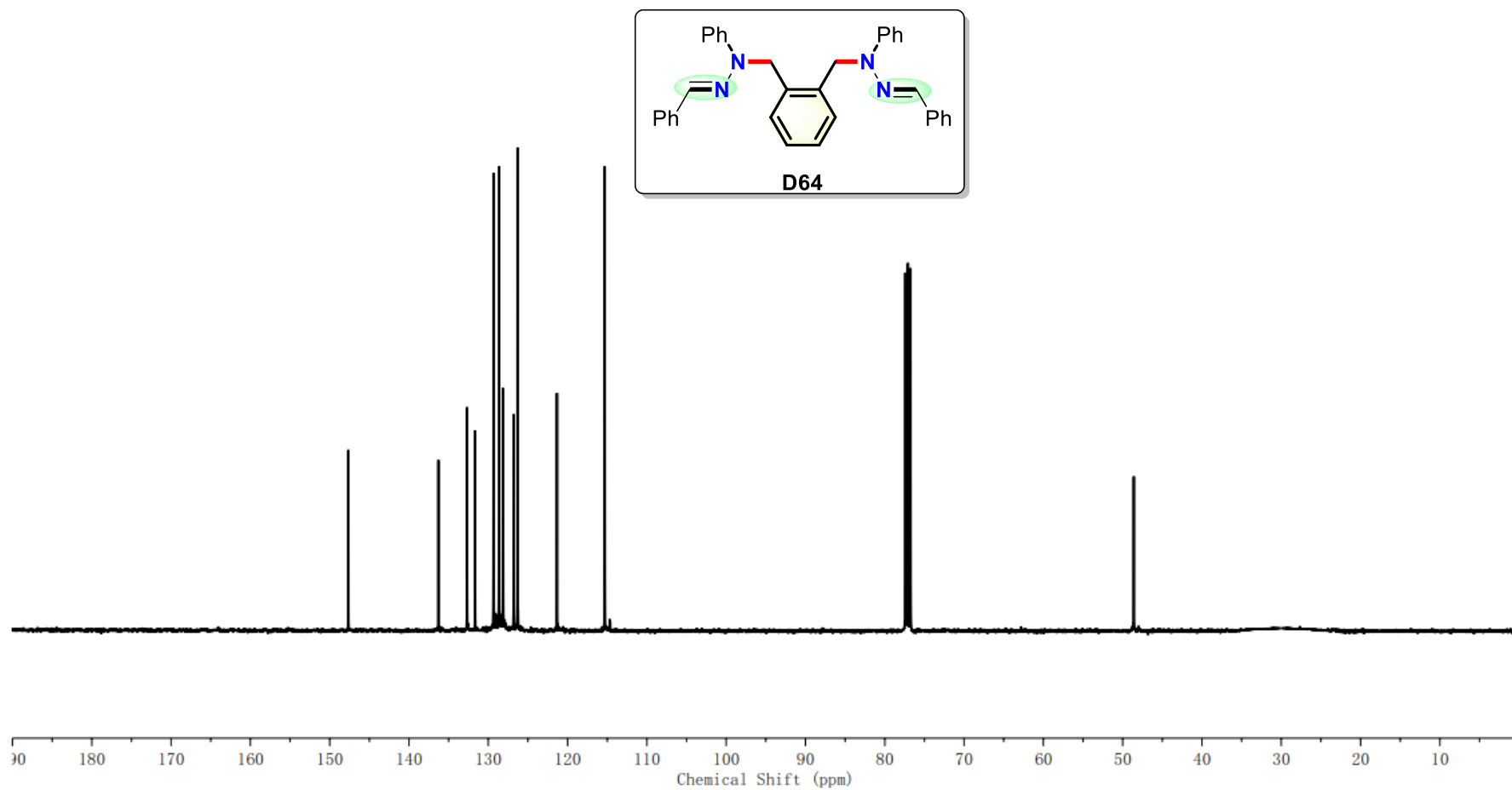
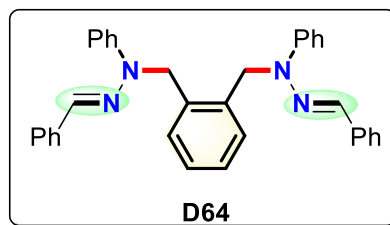


S271

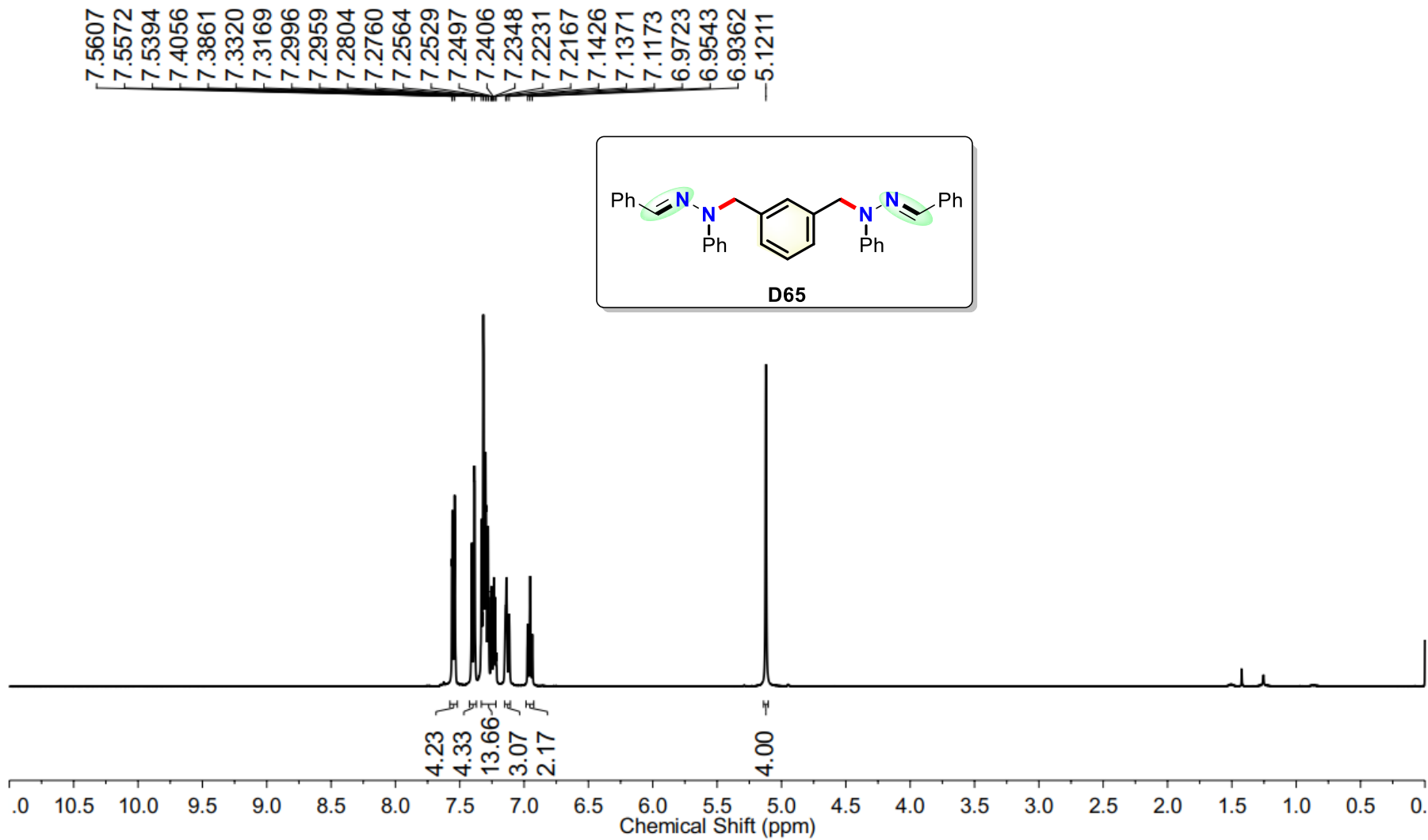
2022-11-17 yy001 616 lqq-24

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136.29
132.67
131.66
129.34
128.66
128.16
128.13
126.79
126.32
121.34
115.31

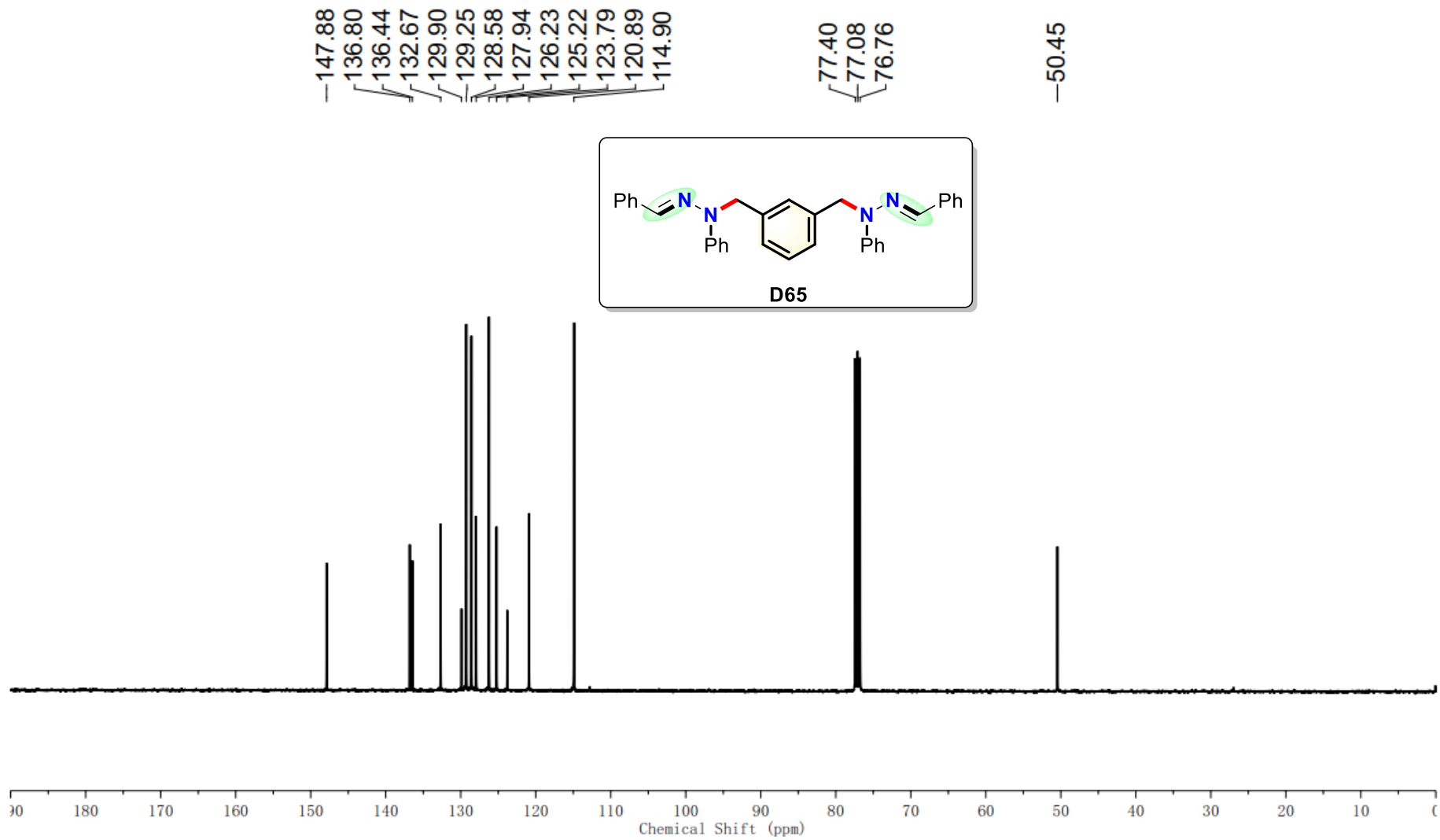
48.60



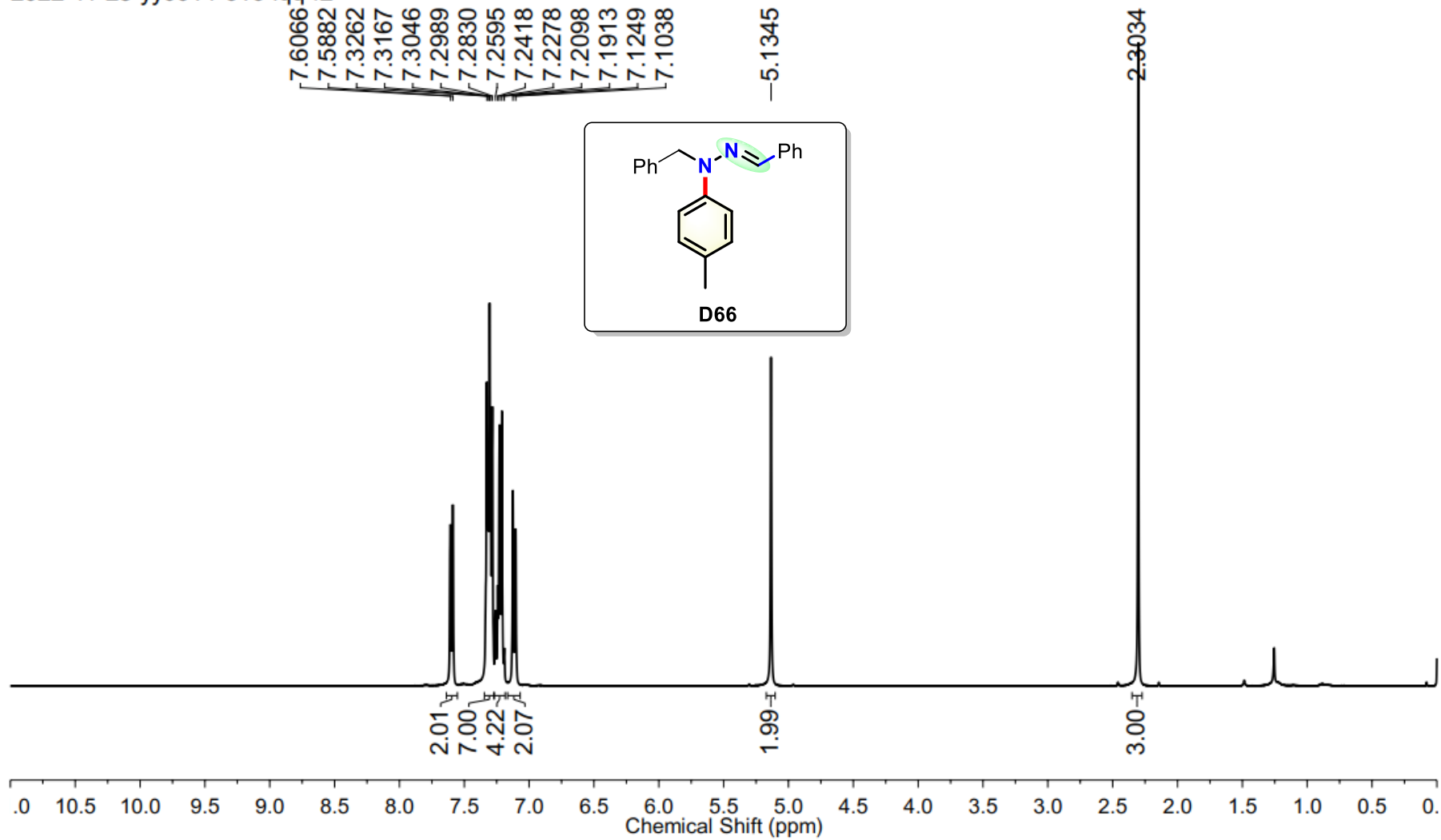
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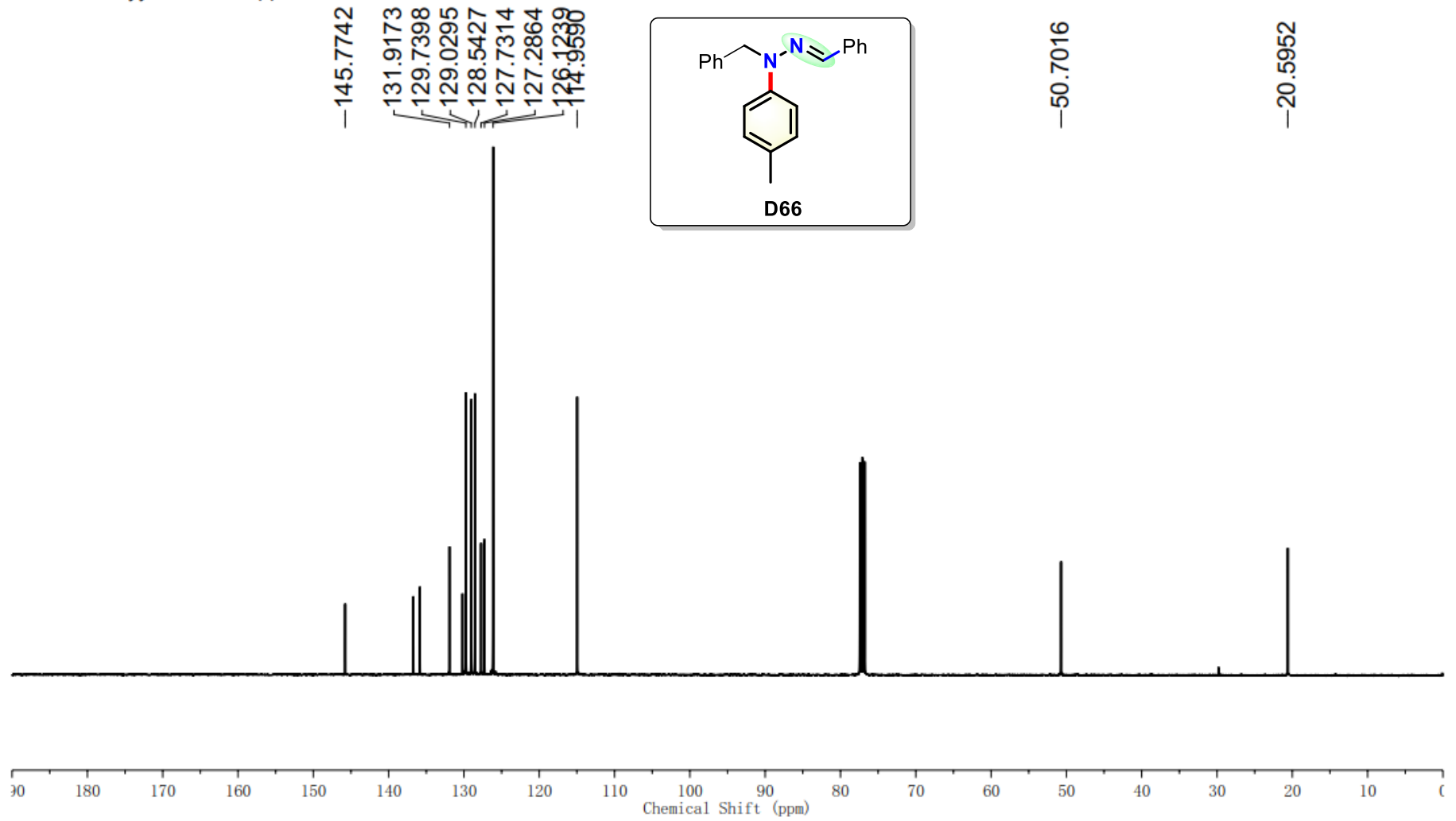
2022-12-7 yy0034 616 1qq-23



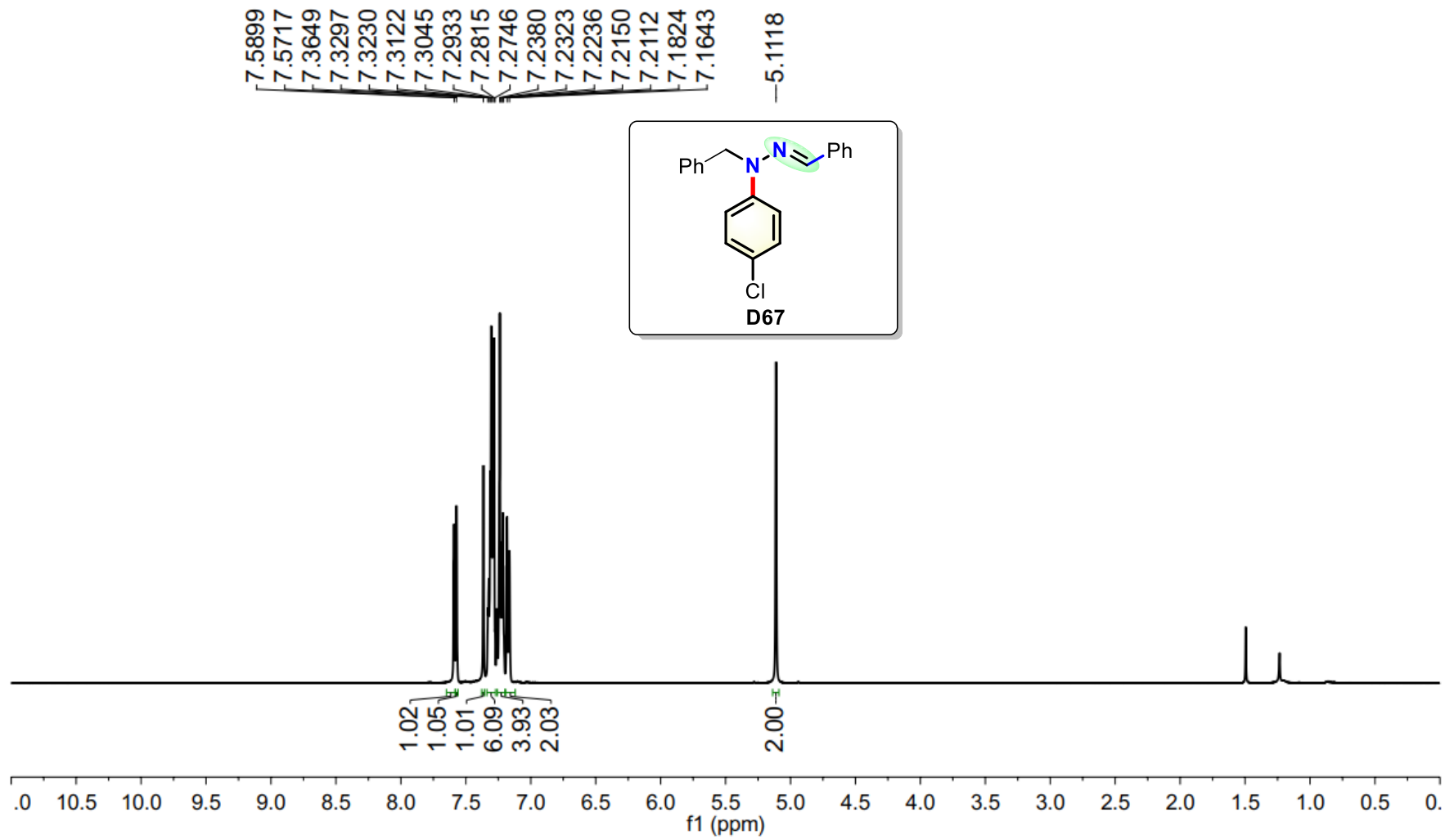
2022-11-28-yy0014-616-lqq-f2



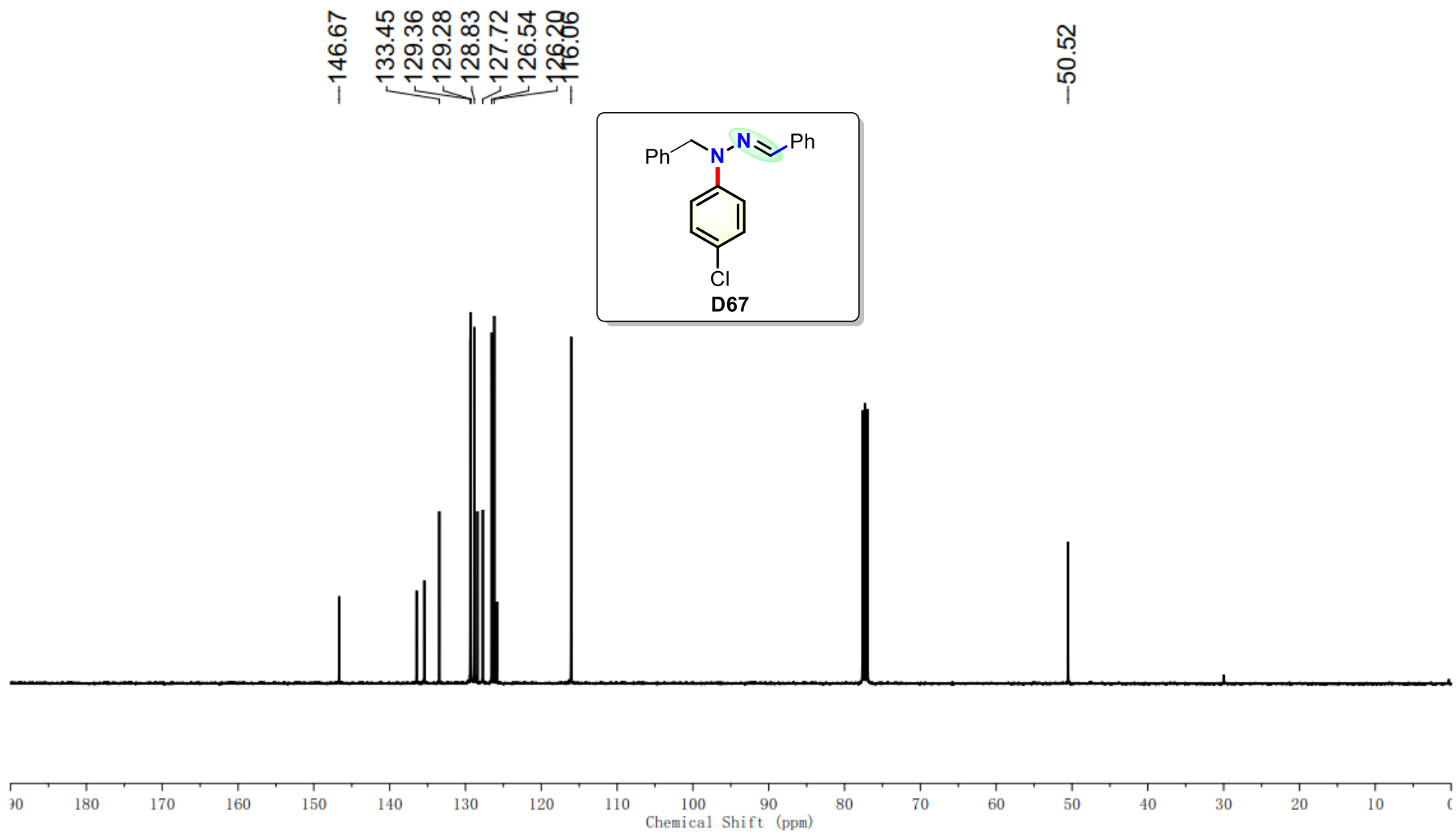
2022-12-02-yy0013-616-lqq-f2



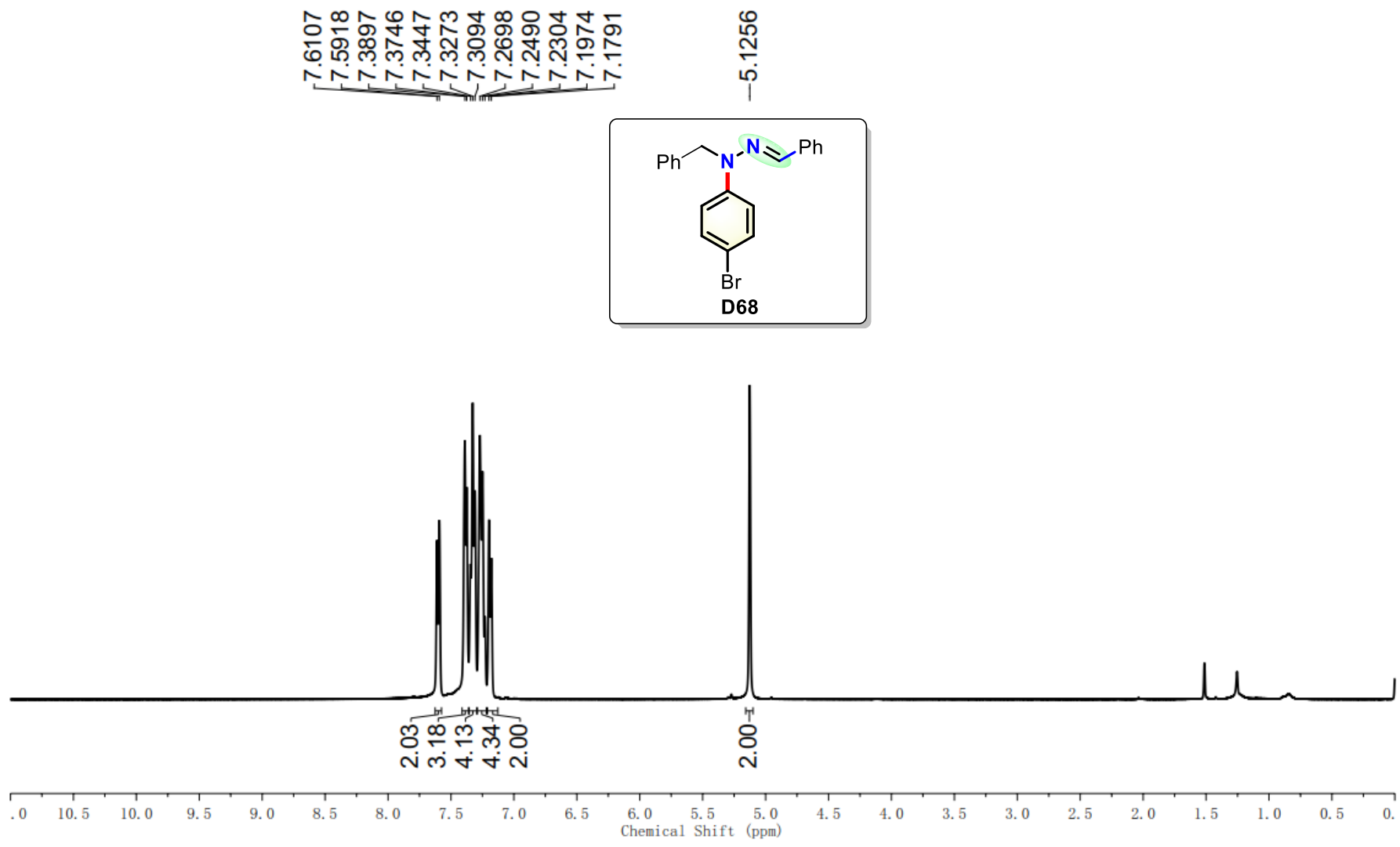
2022-10-21 YY0018 616-LQQ-F9



2022-10-26 yy0025 616 LQQ-9



2022-10-13 yy0007 616-1qq-F13

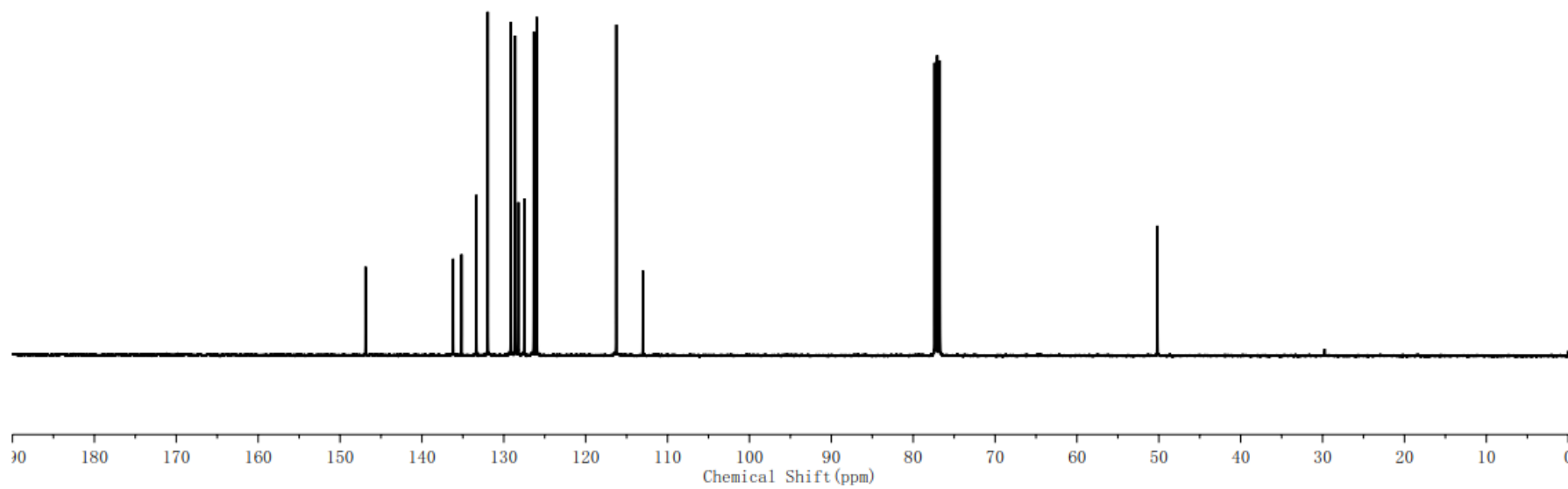
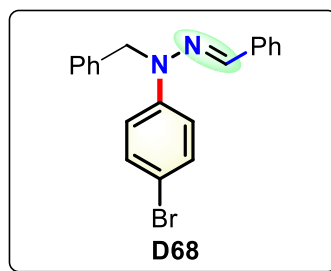


2022-11-7-yy0026-616-f13

146.87
133.35
131.96
129.13
128.60
127.50
126.33
125.97
116.26
113.01

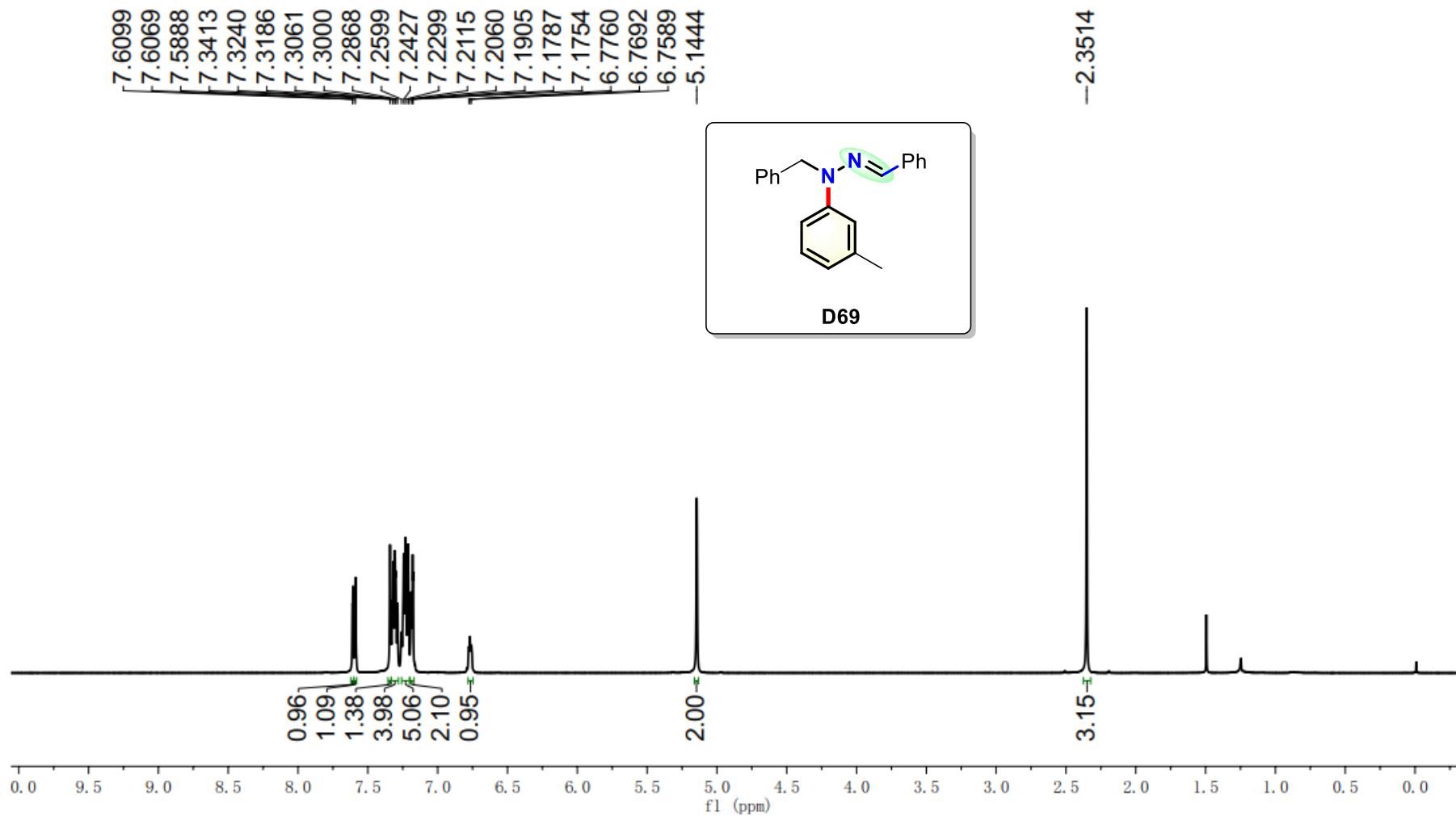
77.38
77.07
76.75

50.18

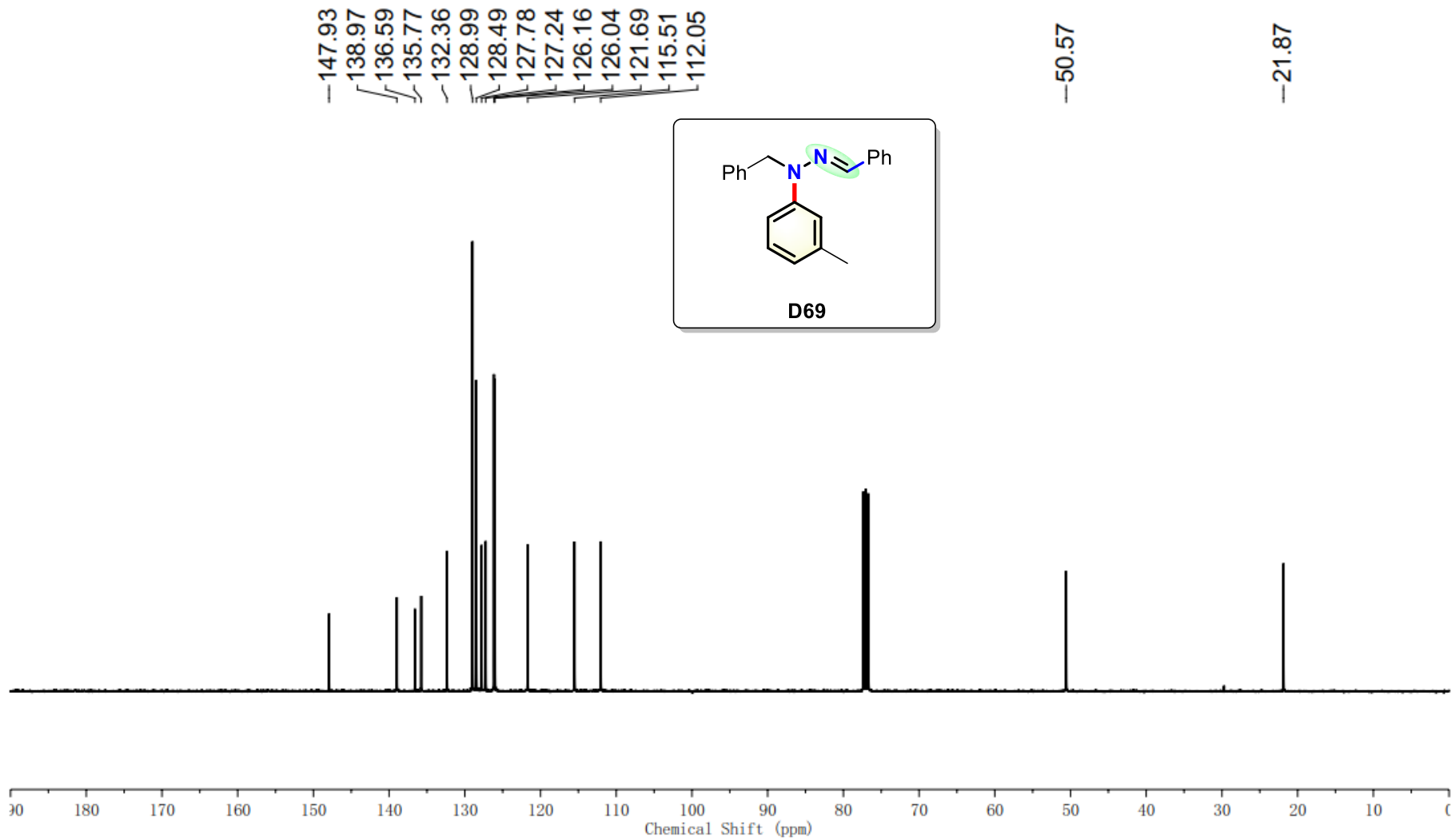


S280

2022-10-21 YY0018 616-LQ-F3

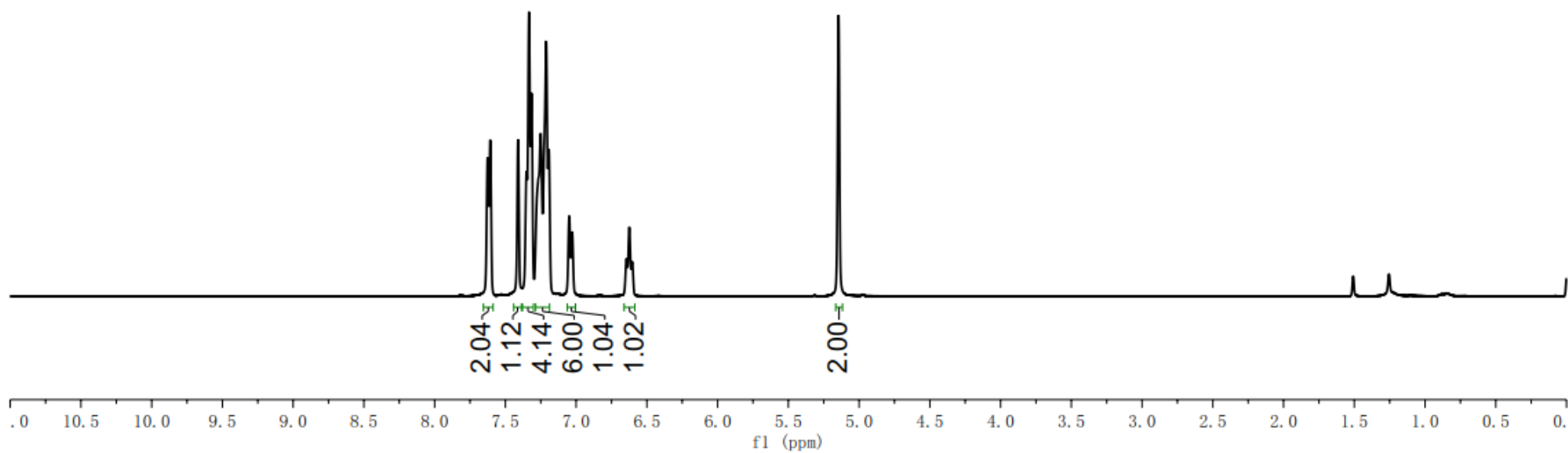
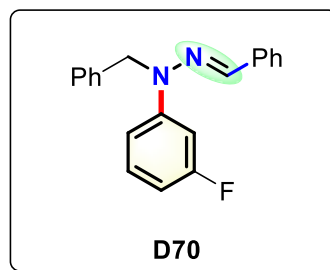


2022-10-26 yy0025 616 LQQ-3



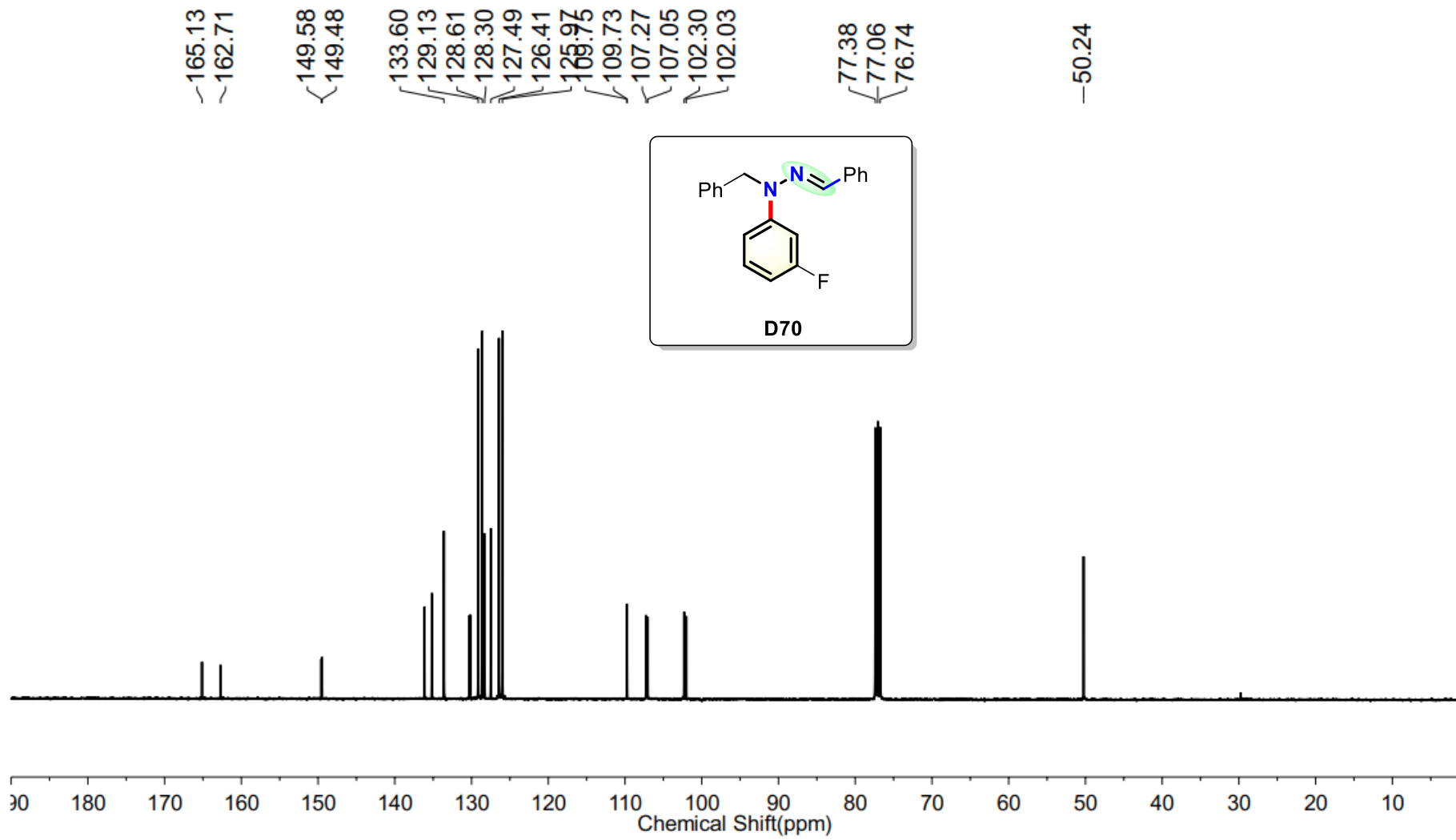
2022-10-17-YY0007-616-ZGH-f6

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7.6071
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7.3331
7.3151
7.2505
7.2118
7.1930
7.0486
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6.6439
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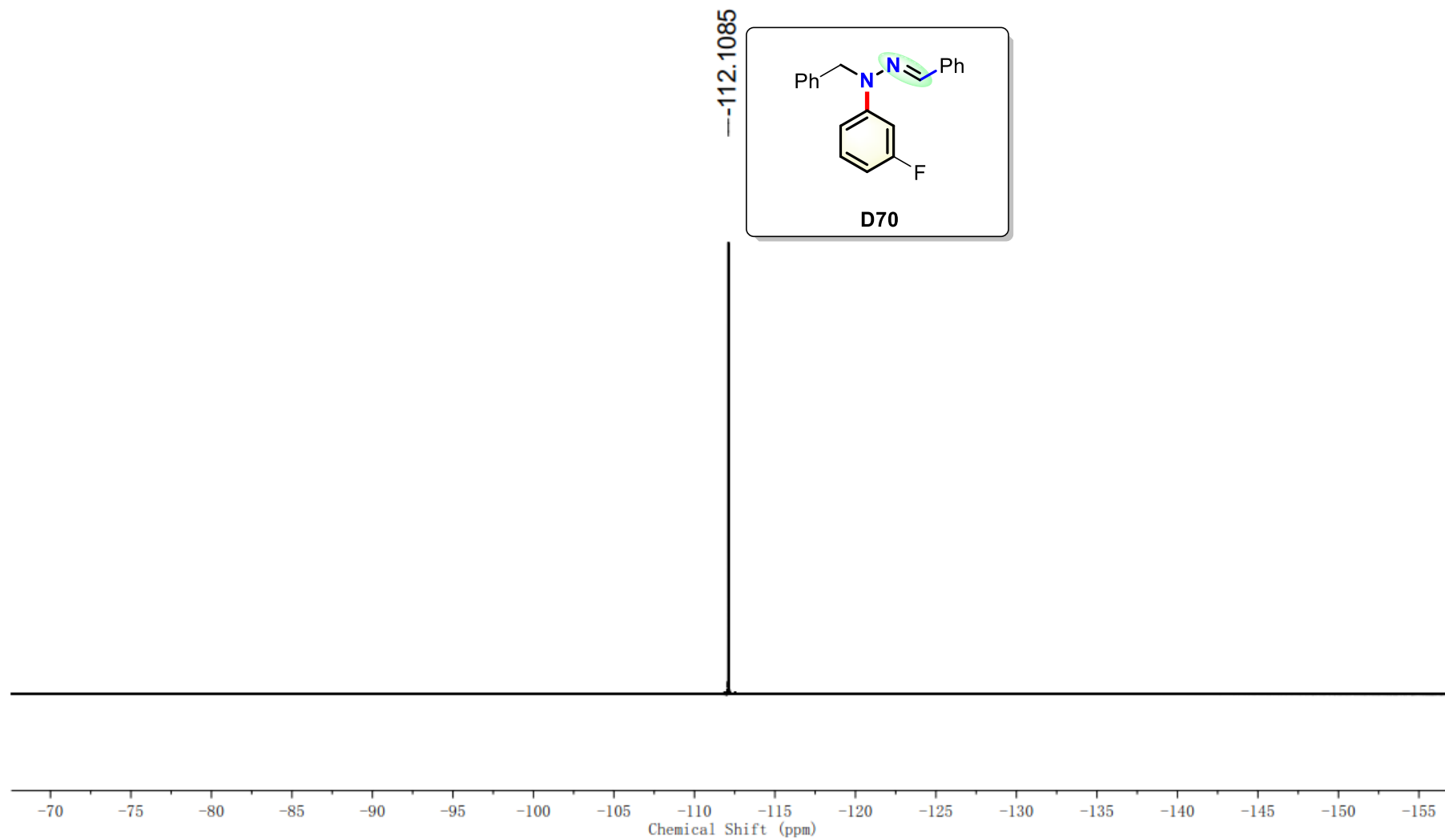


S283

2022-11-08-yy0037-616-f6

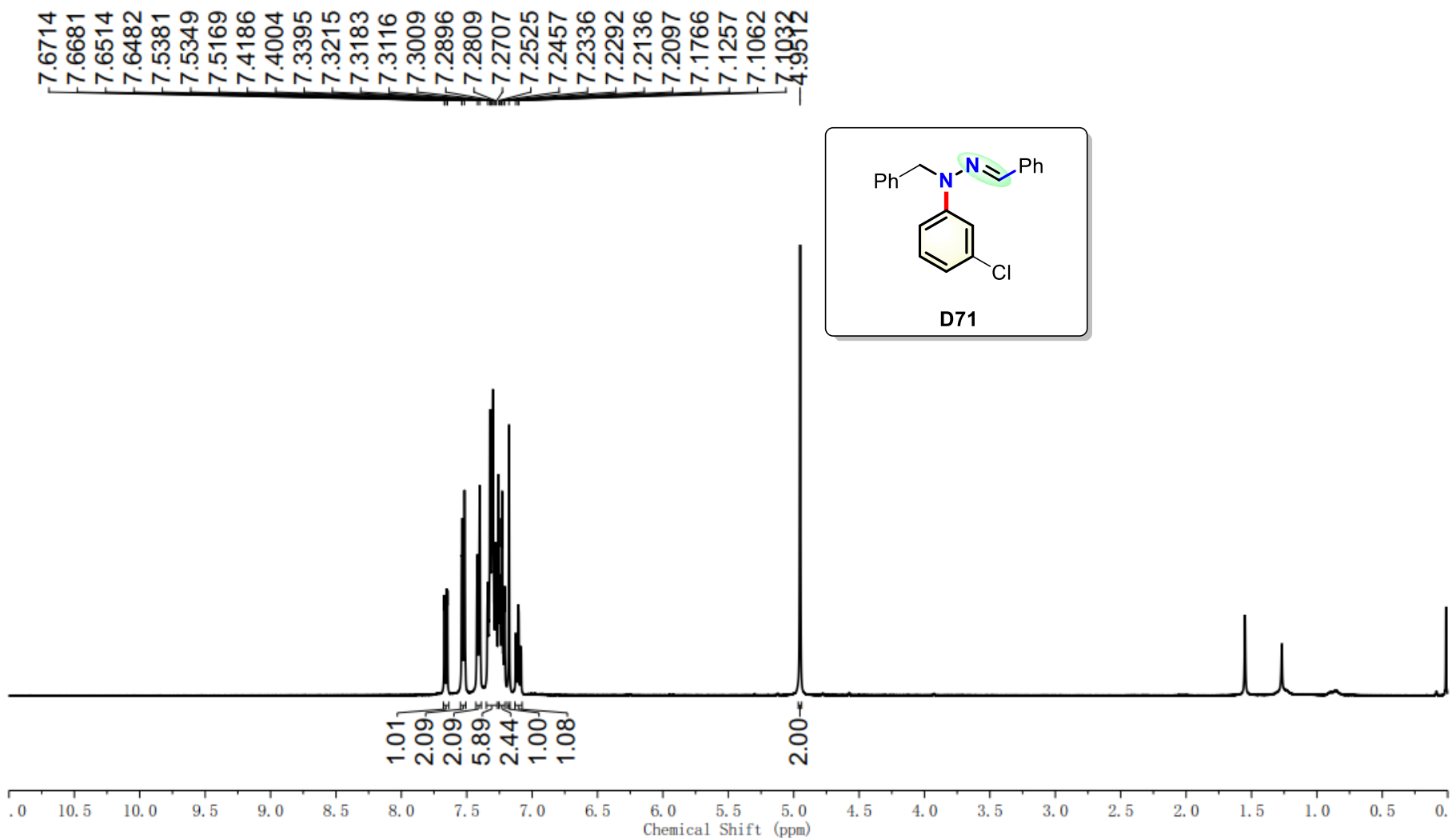


2022-12-7 yy0035 616-1qq-6



S285

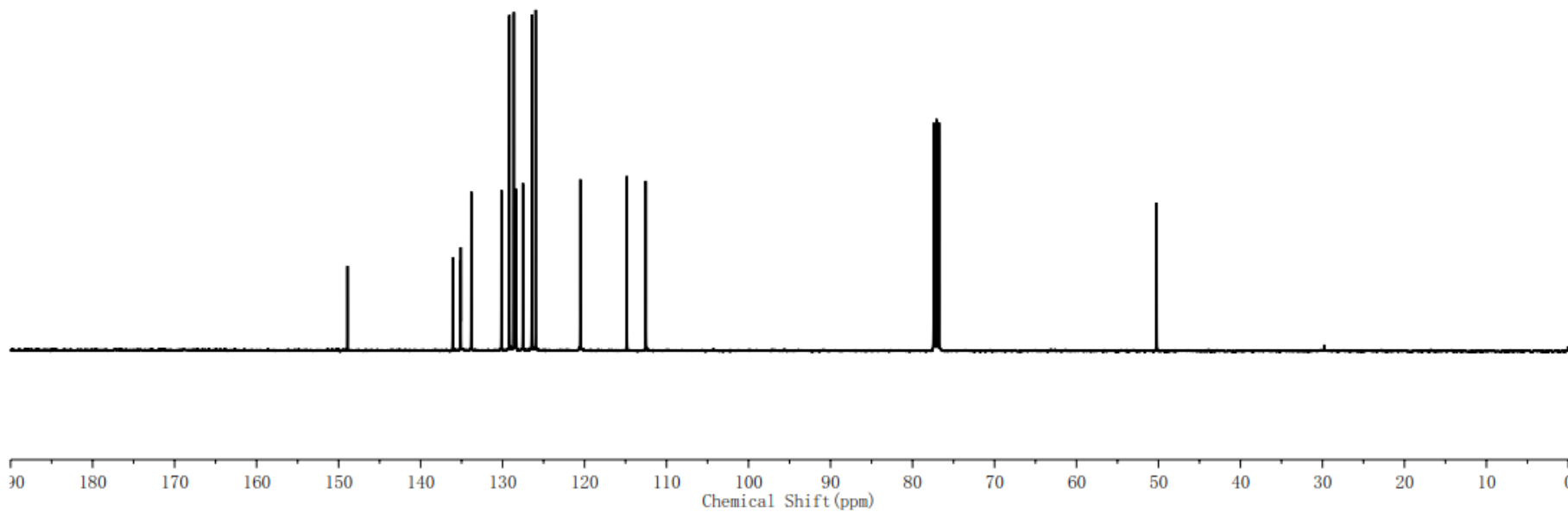
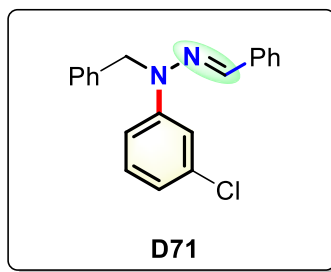
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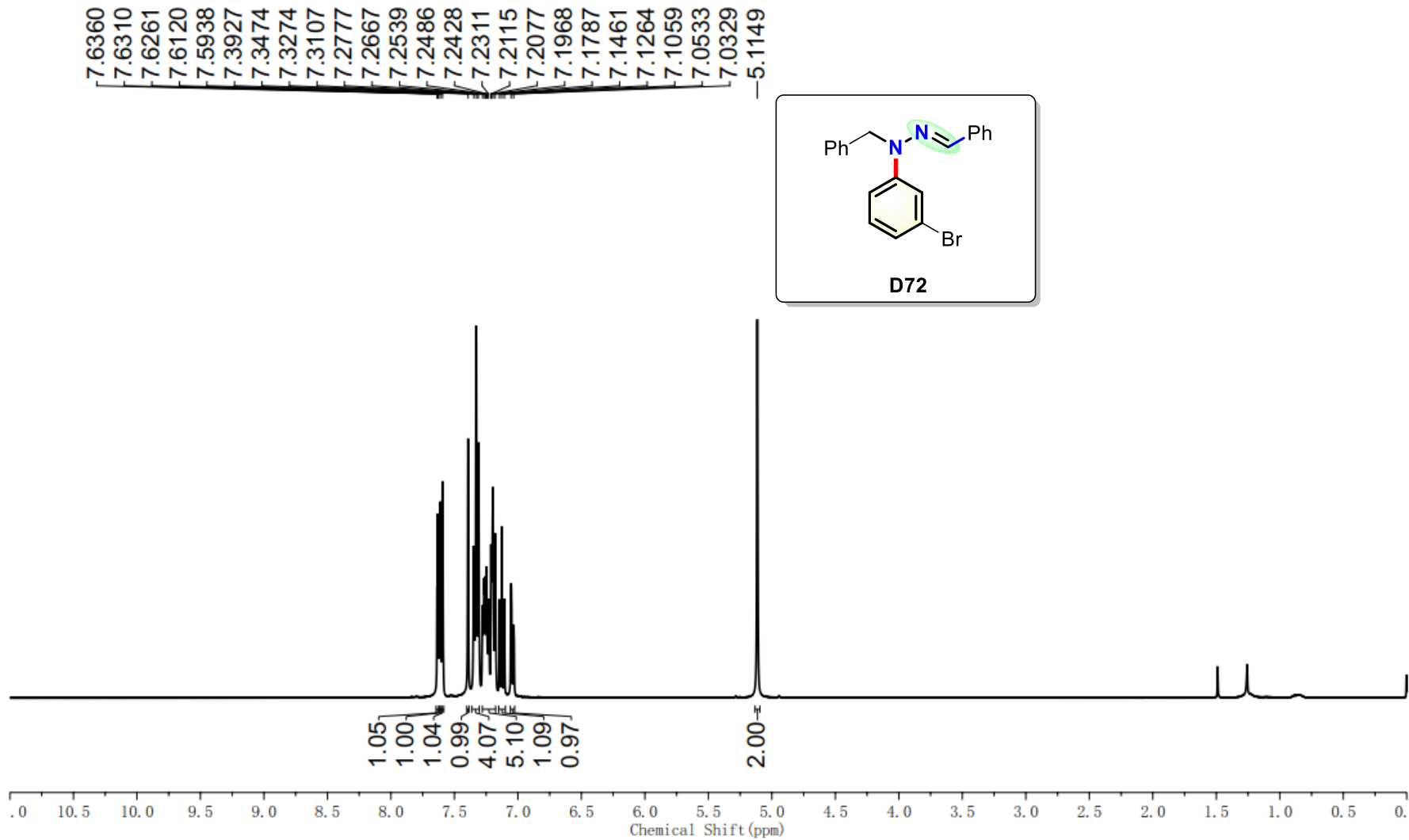
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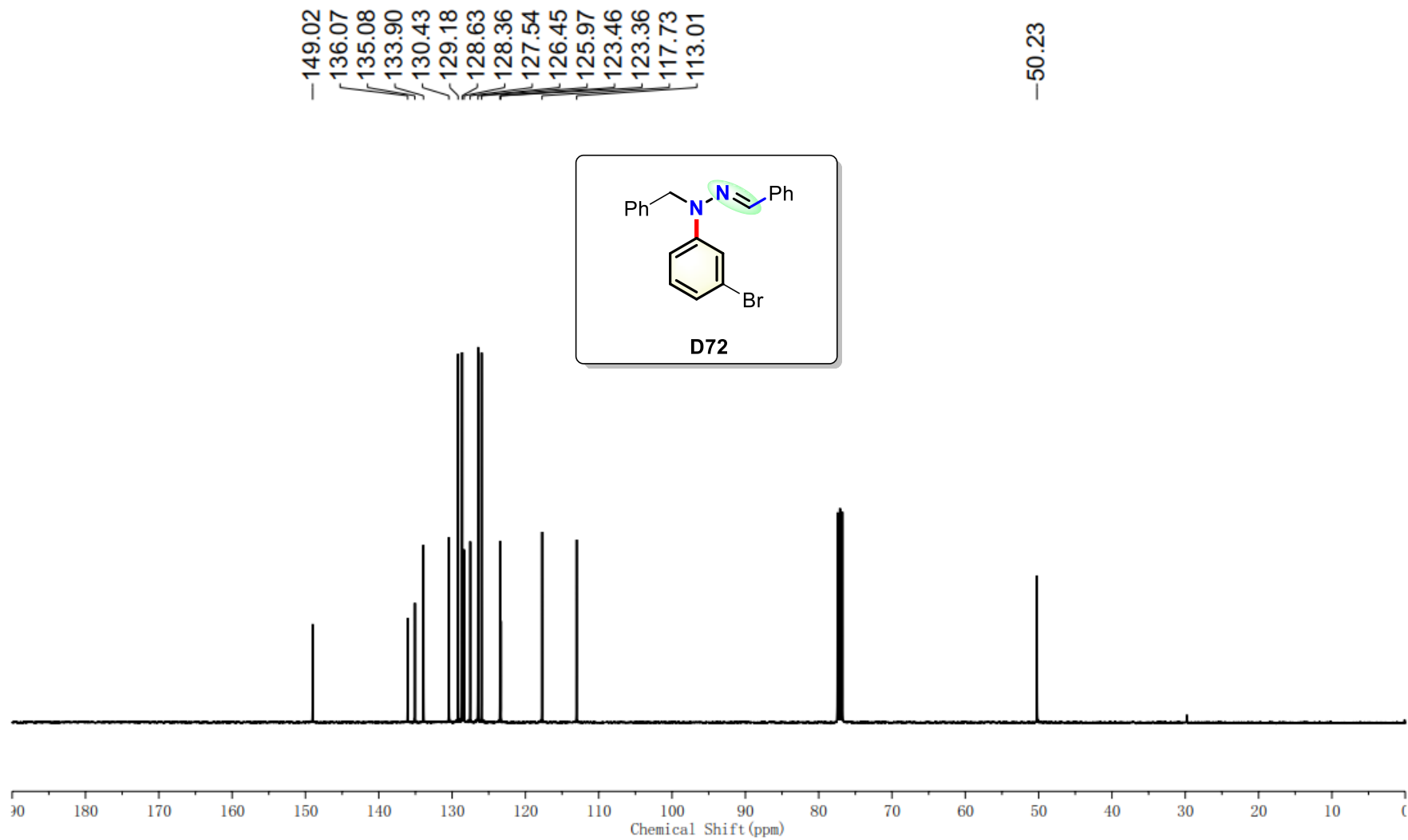
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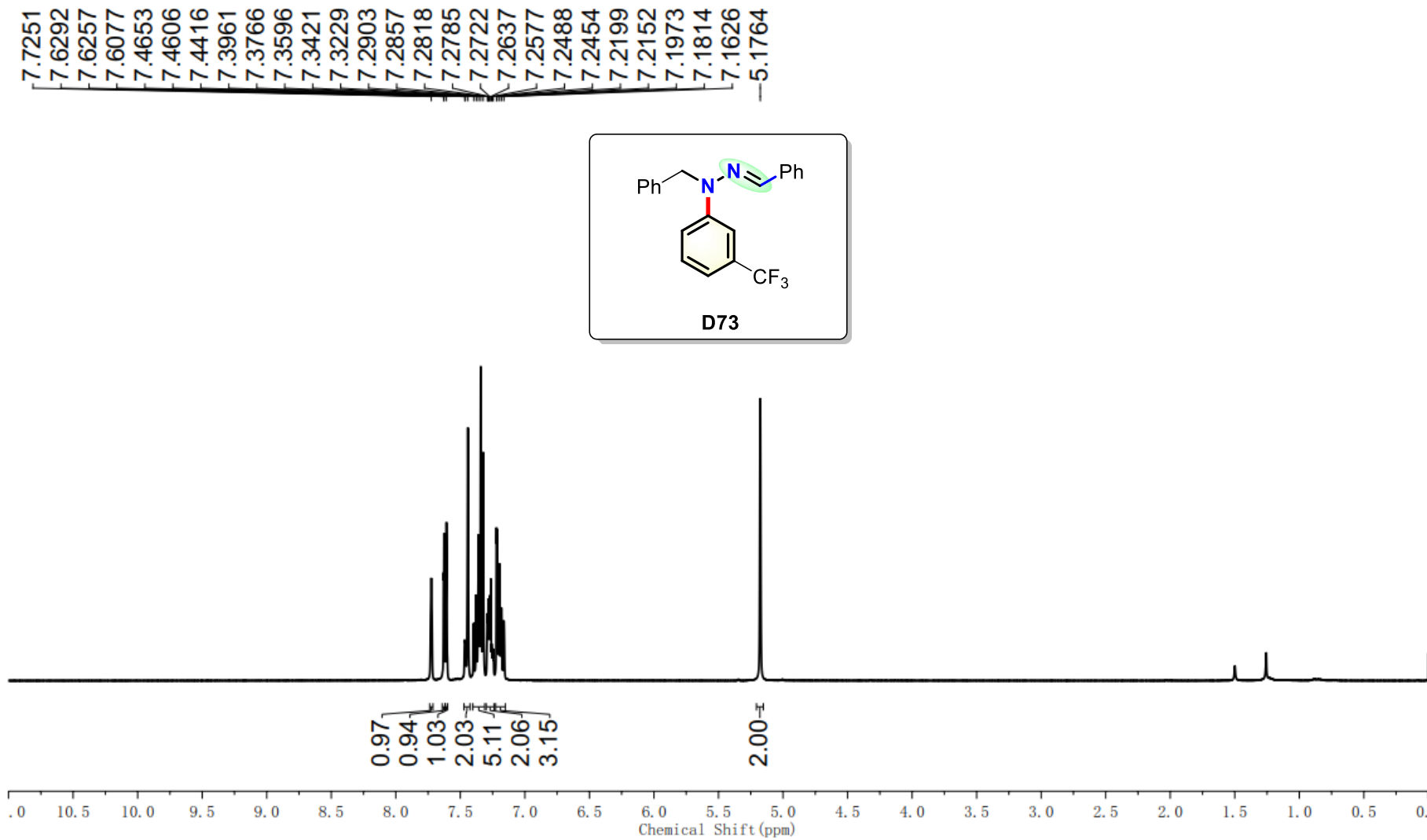
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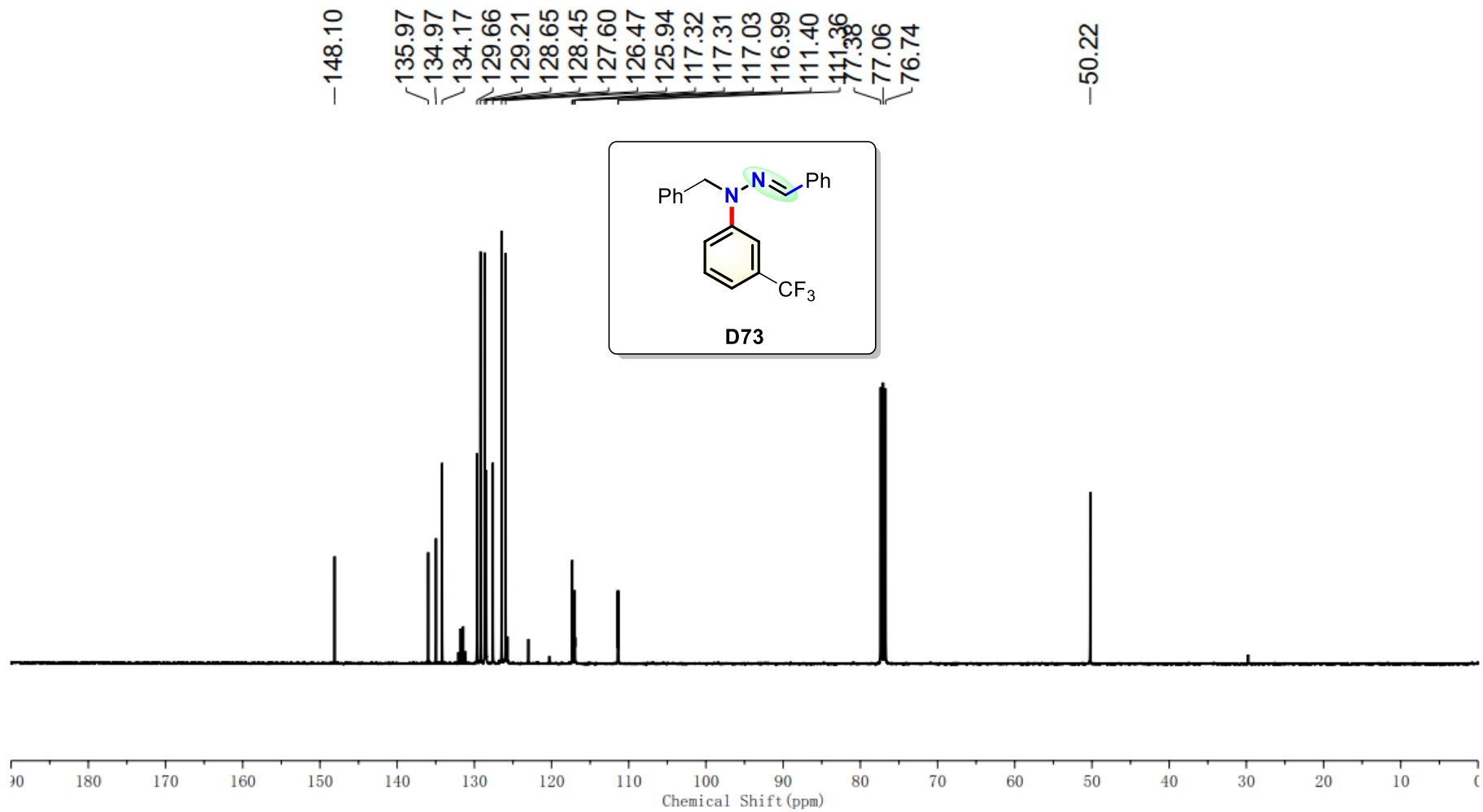
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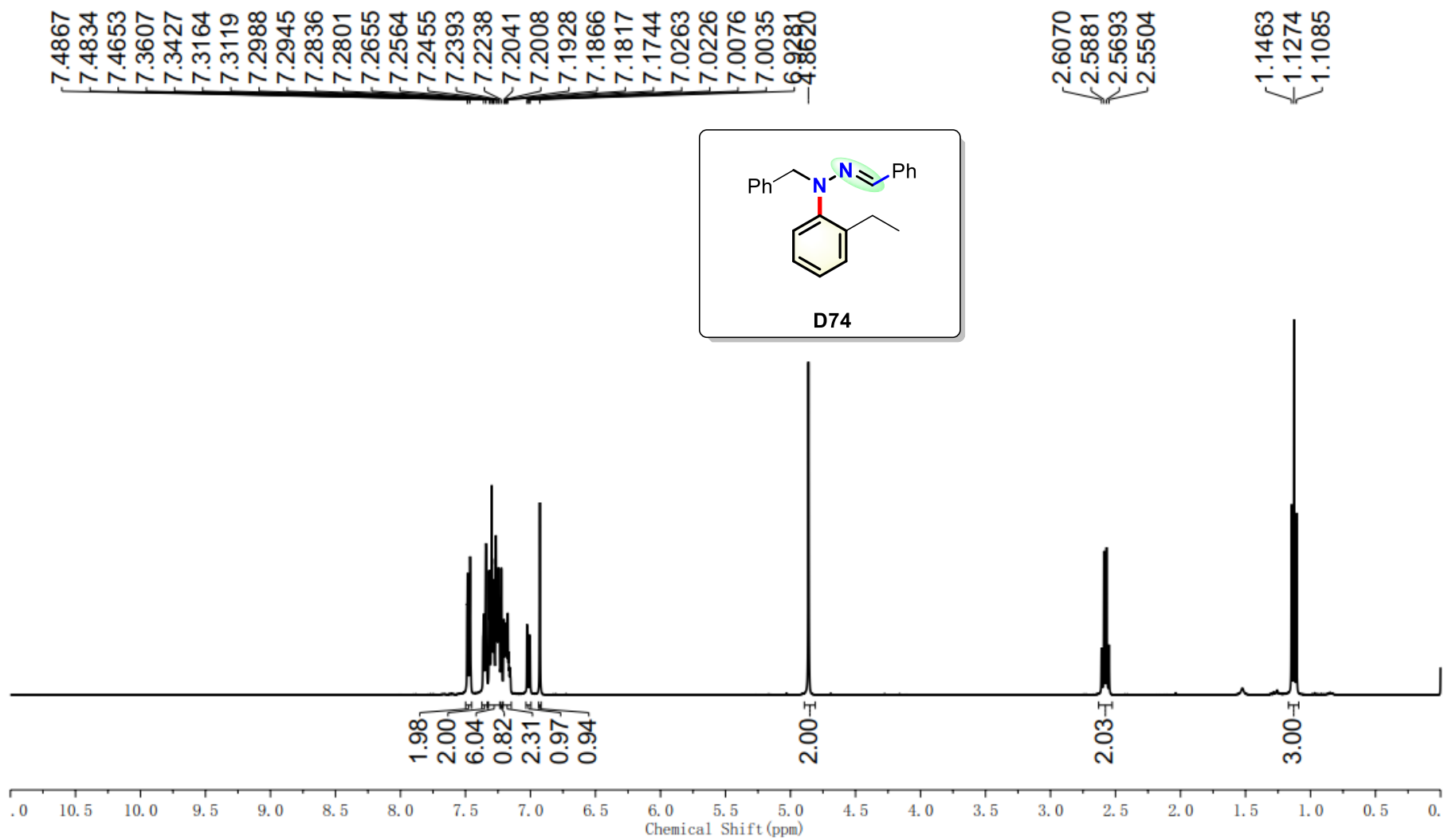
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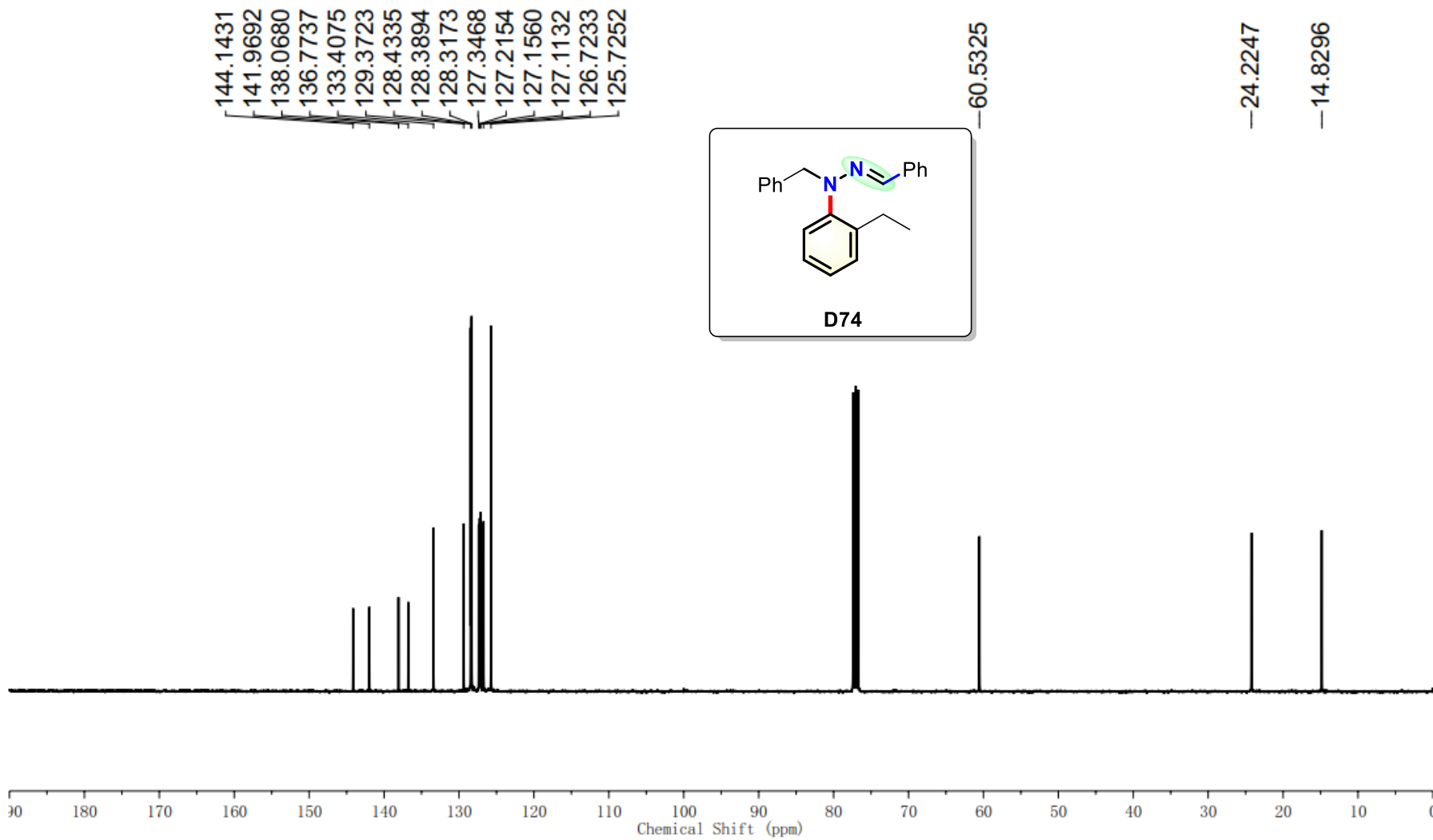
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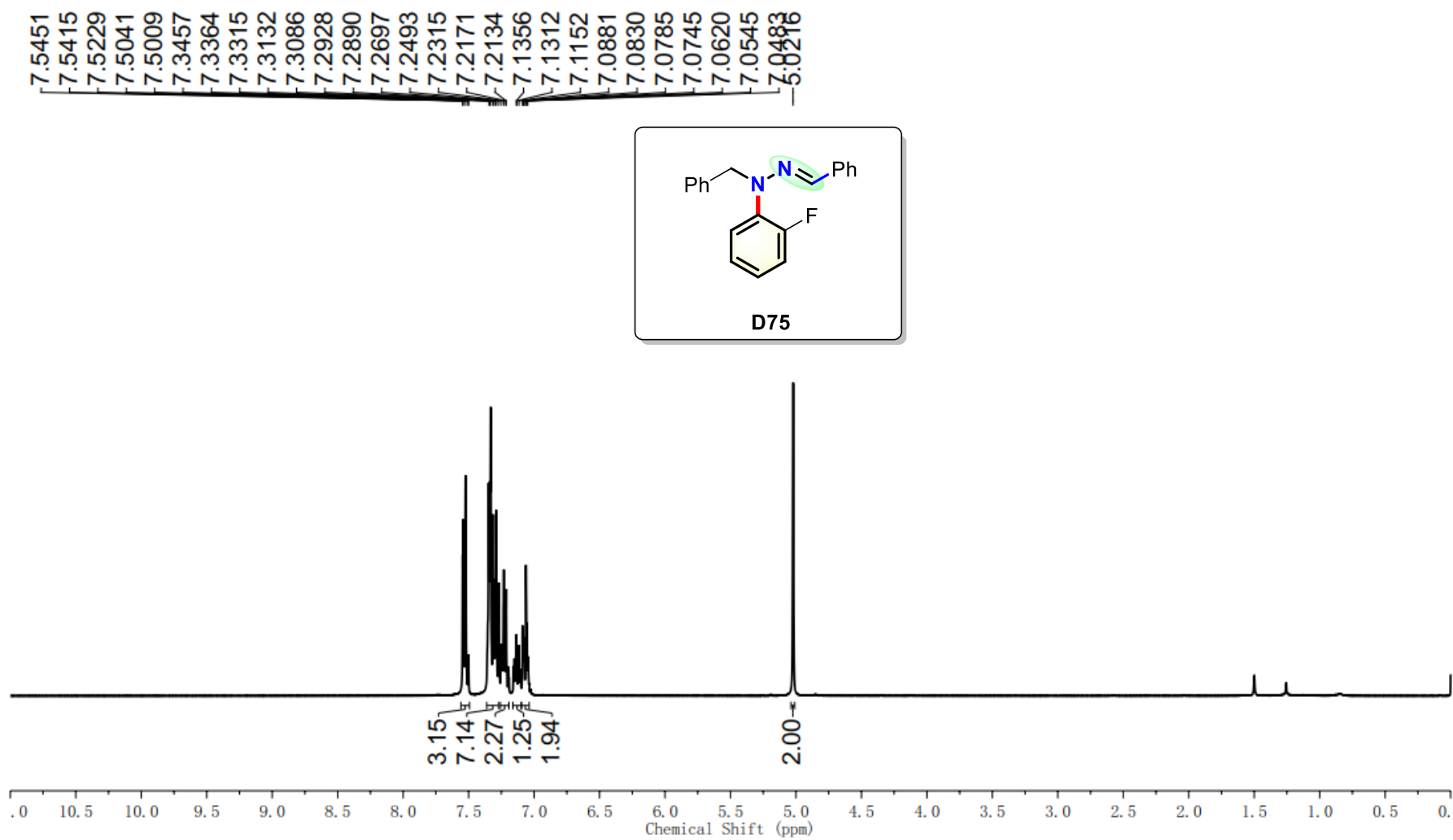
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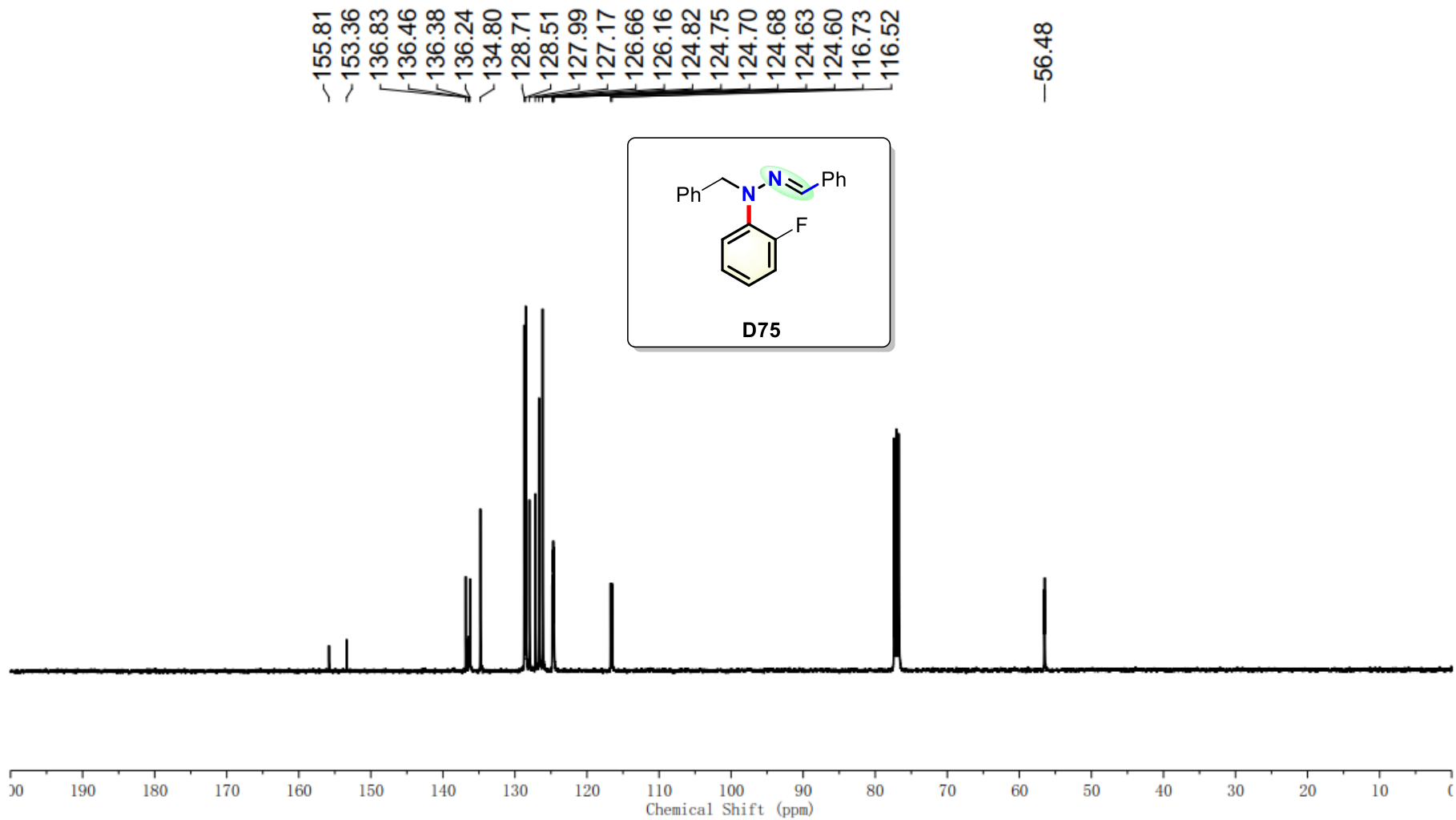
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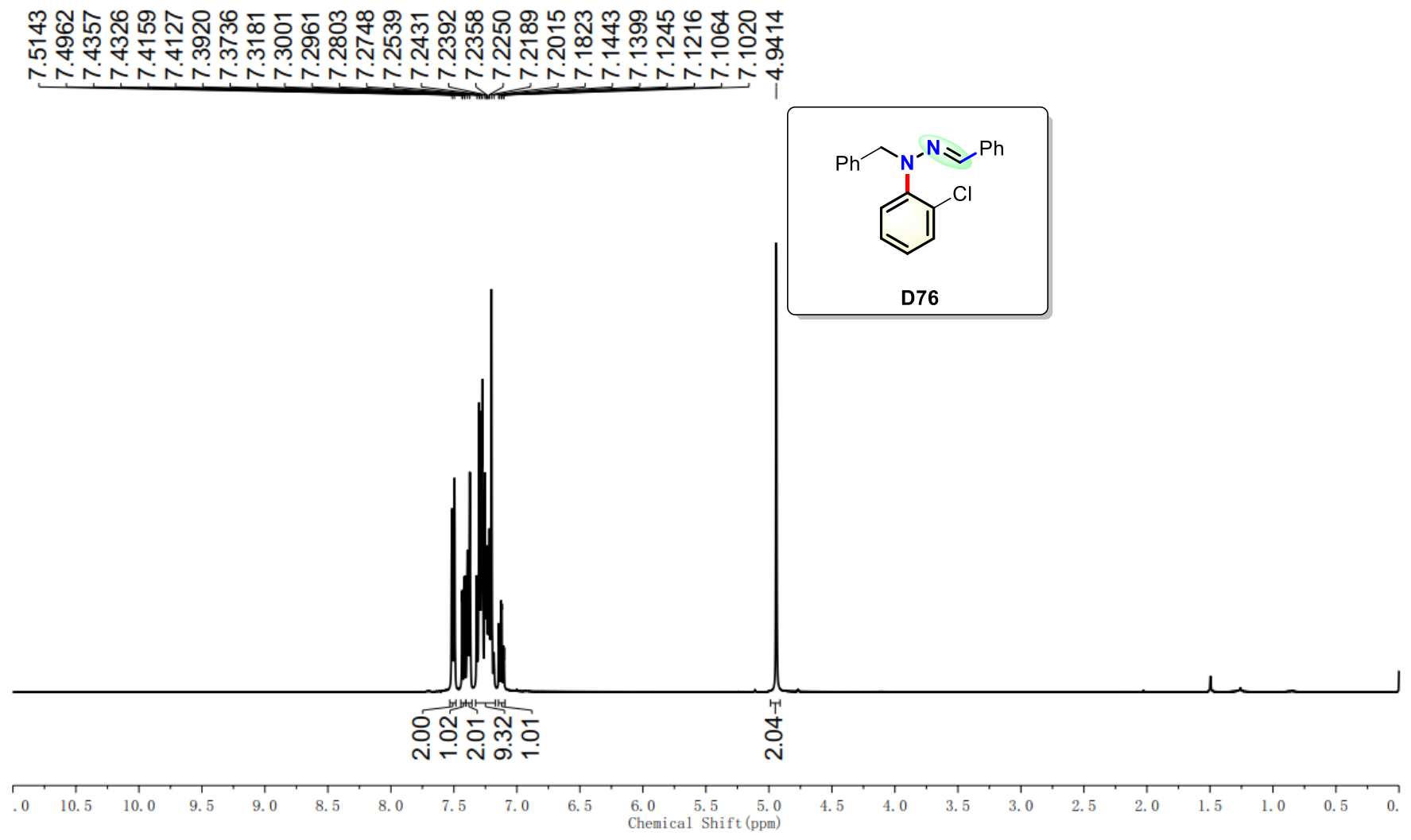
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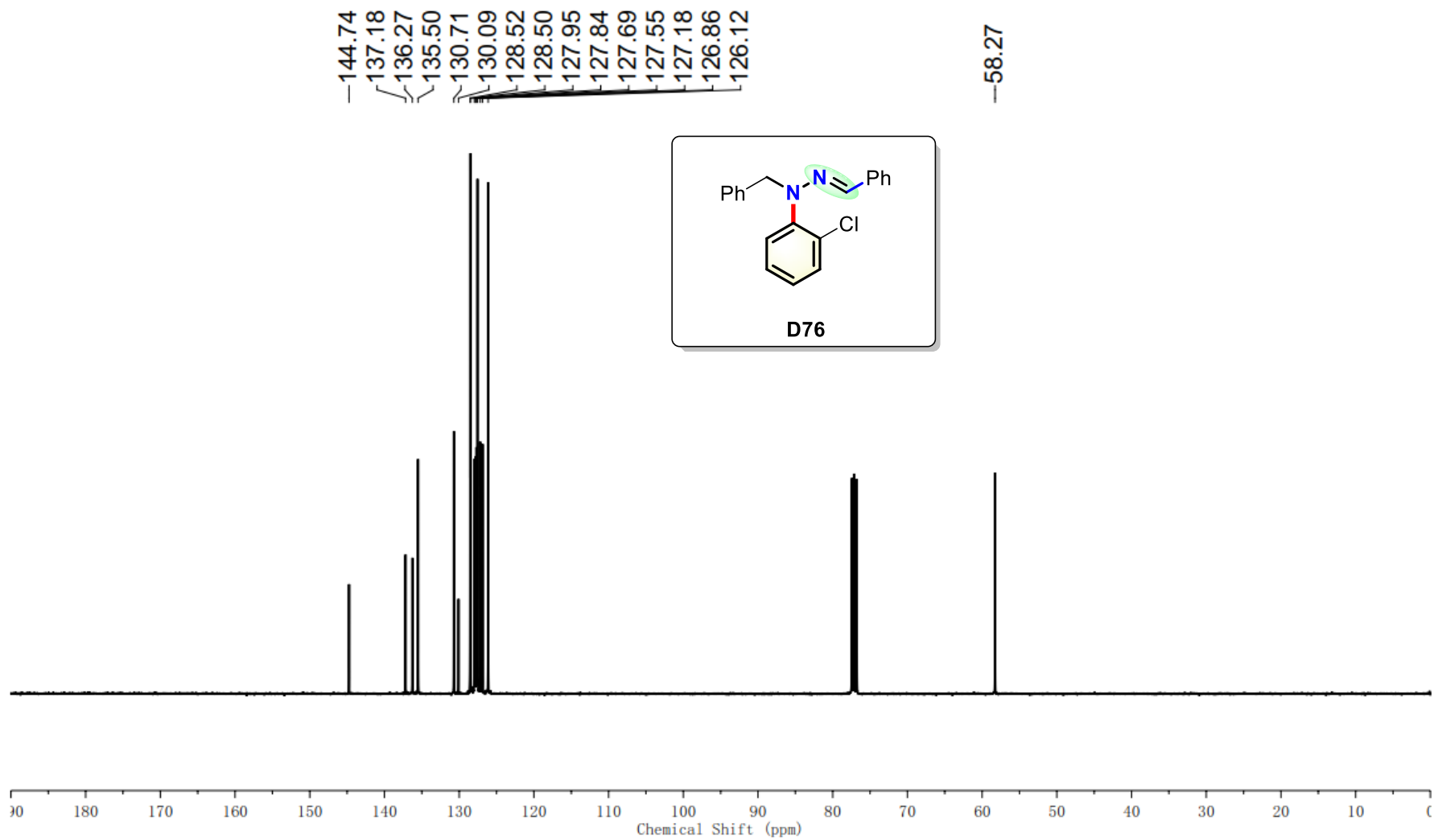
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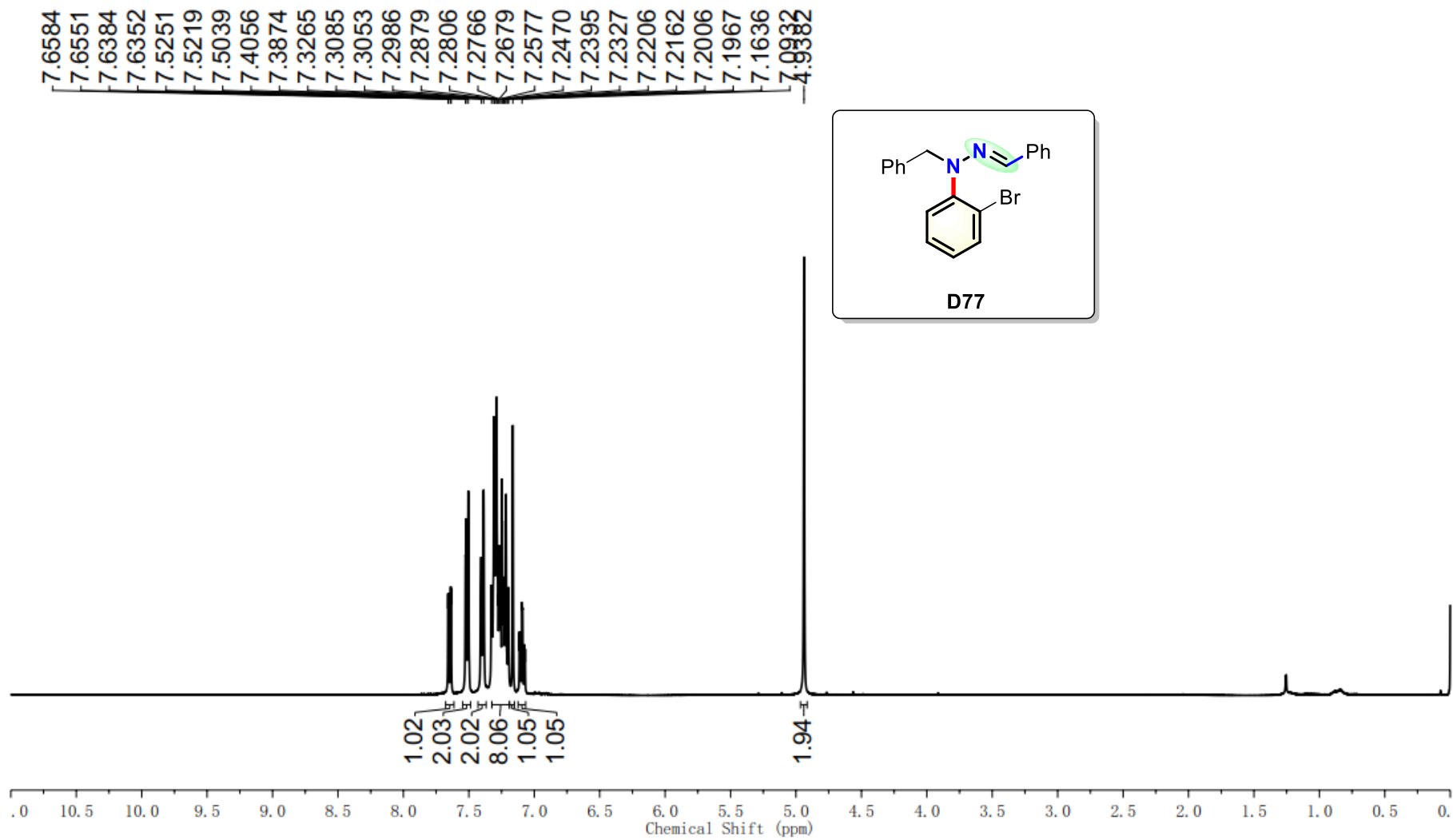
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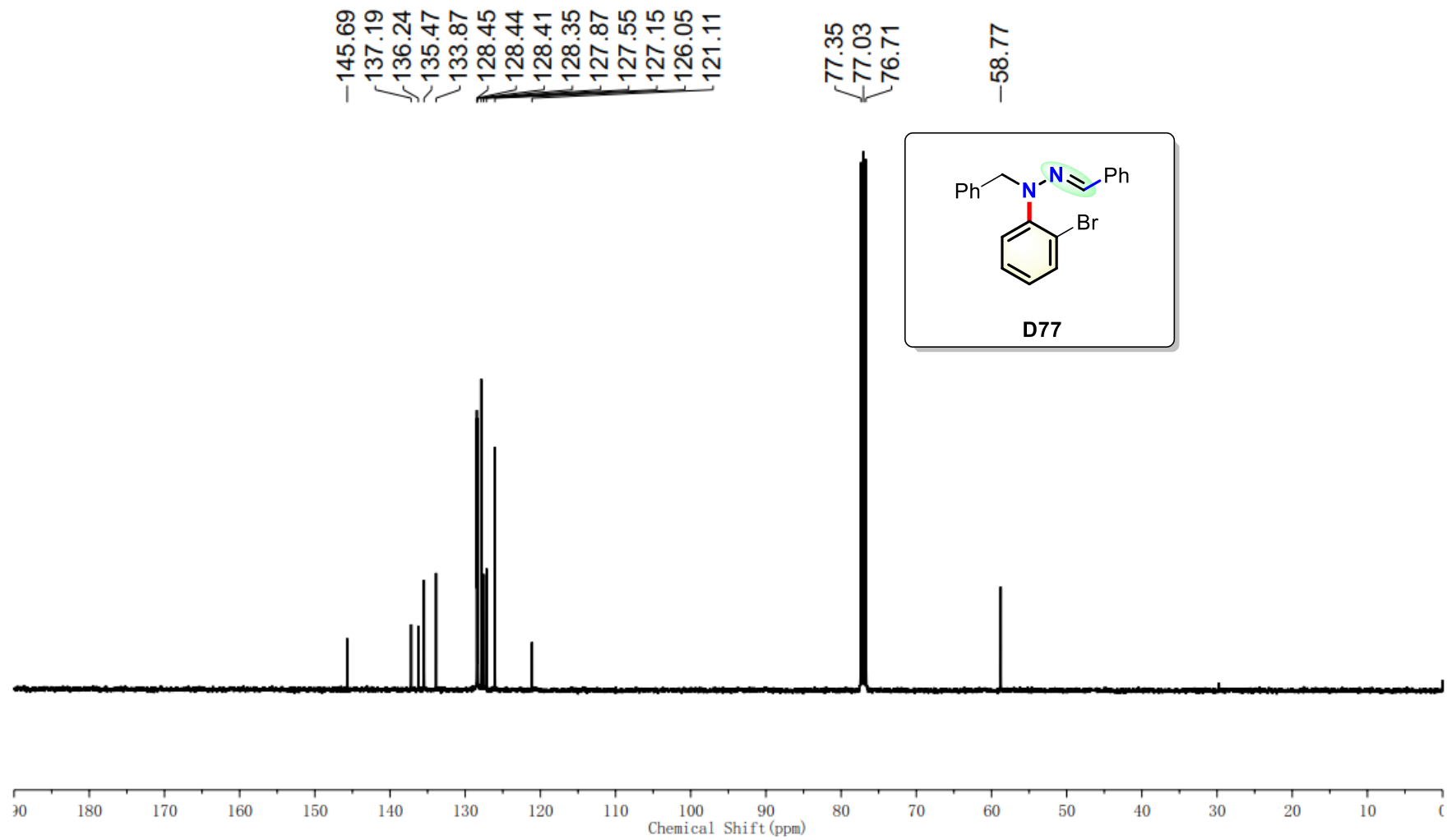
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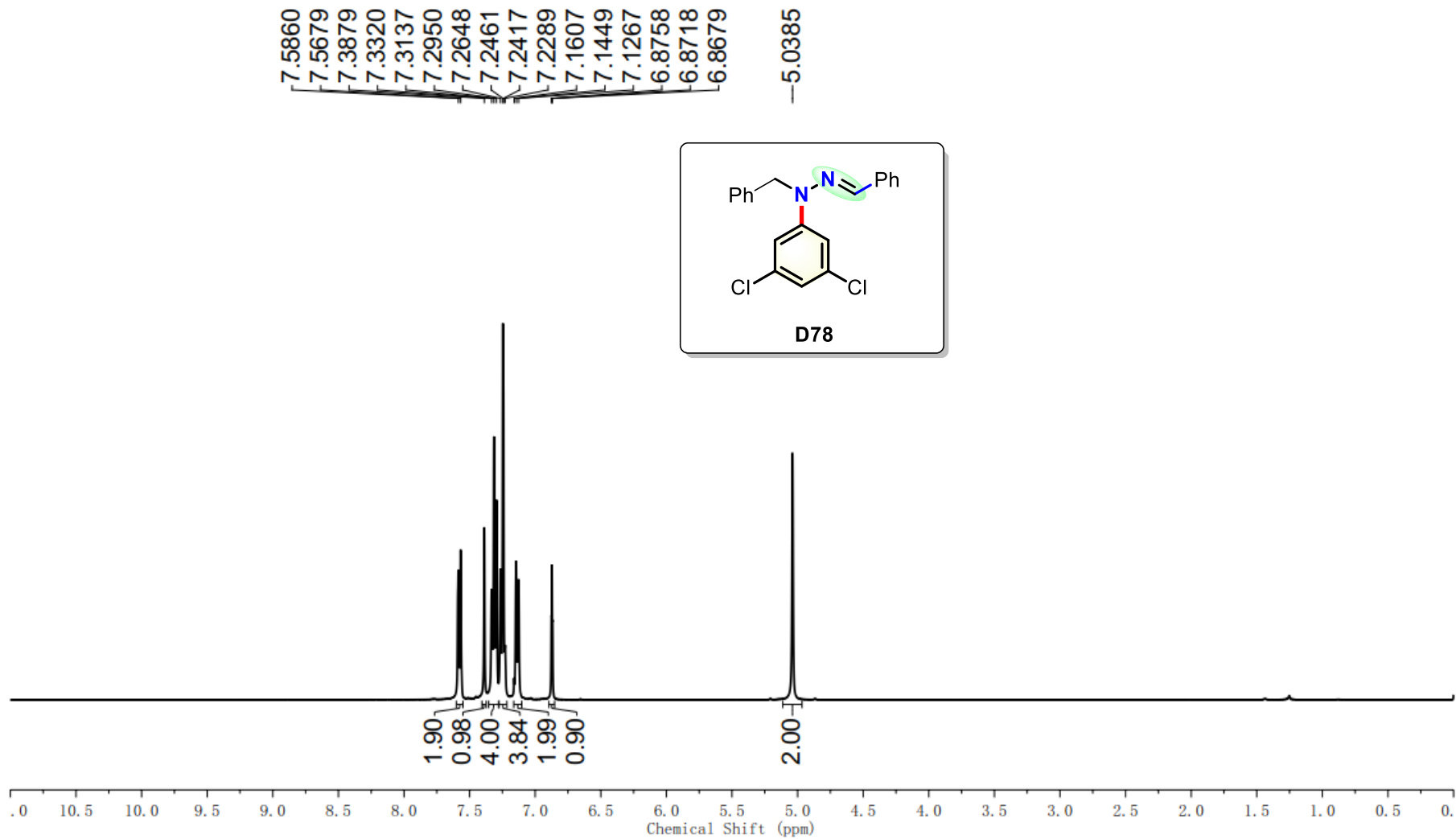
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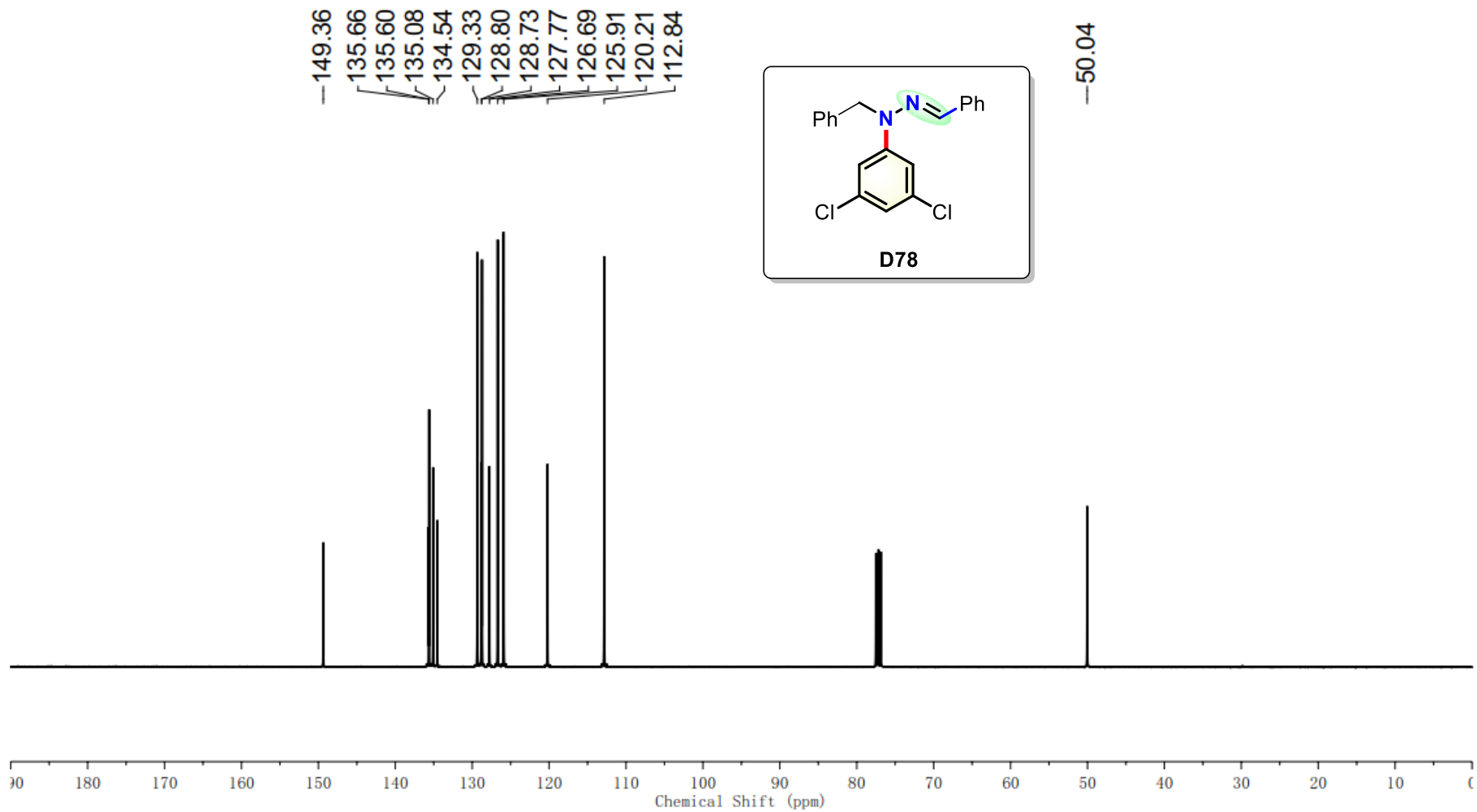
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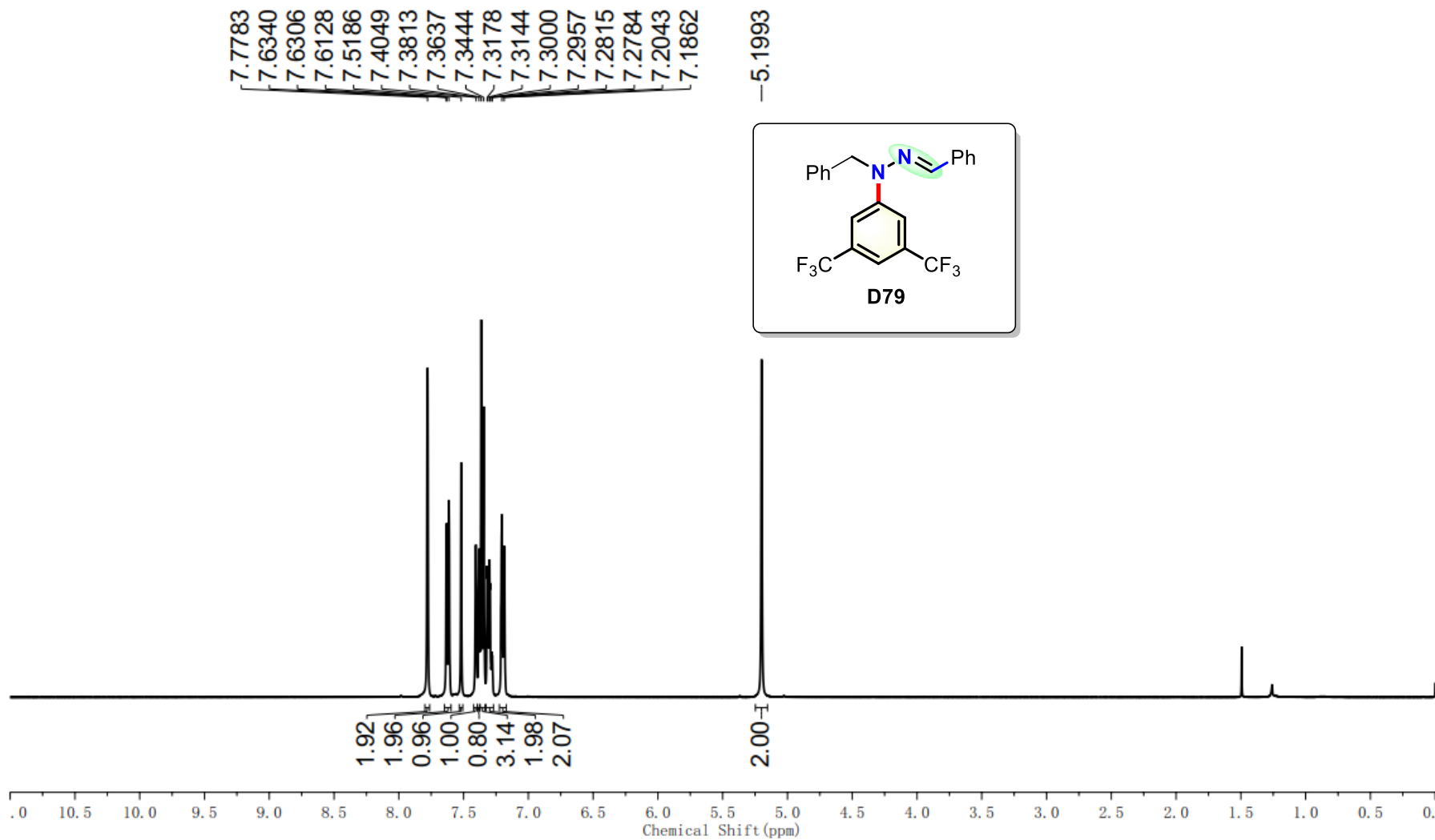
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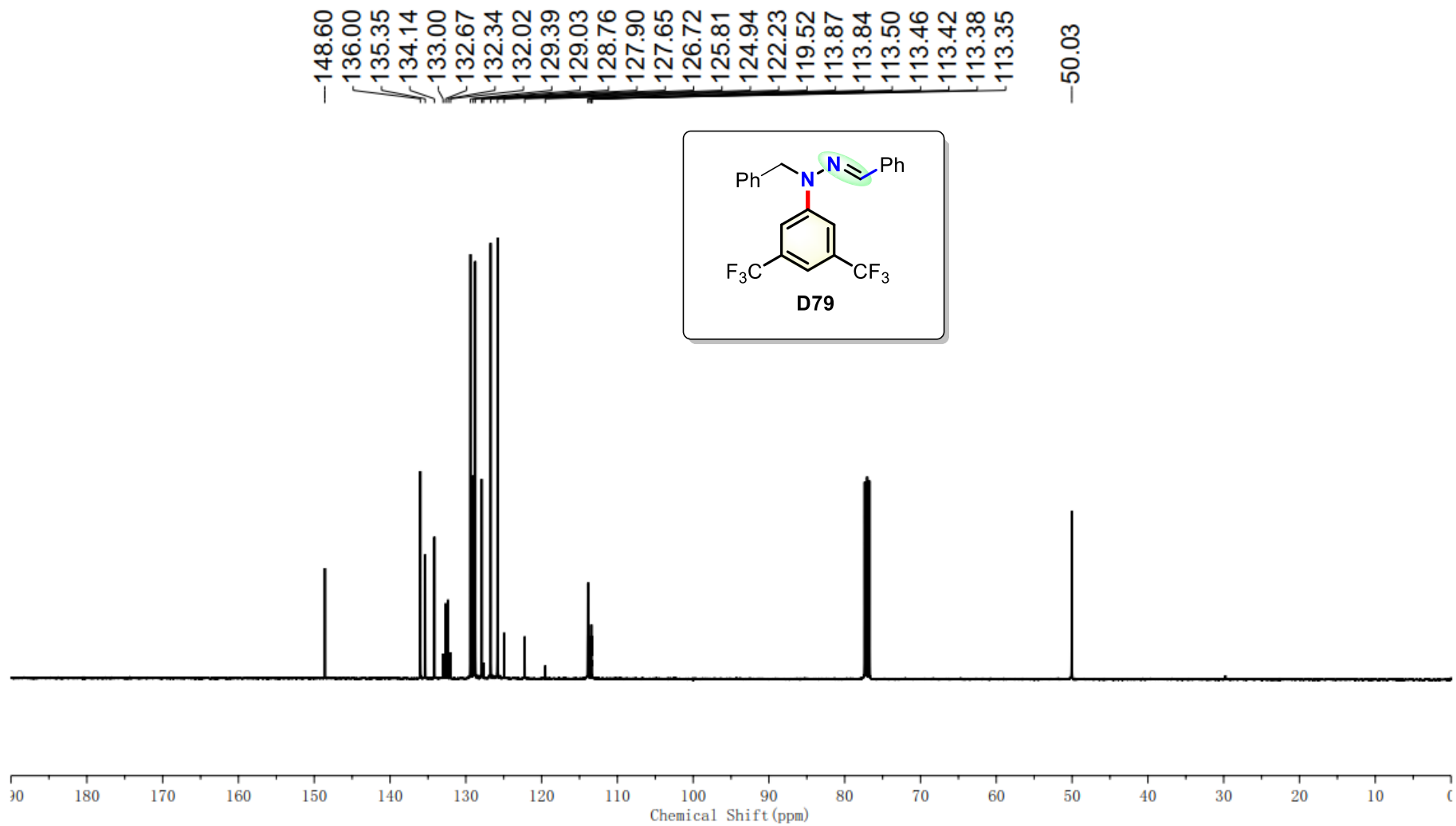
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2022-11-7-yy0025-616-f18



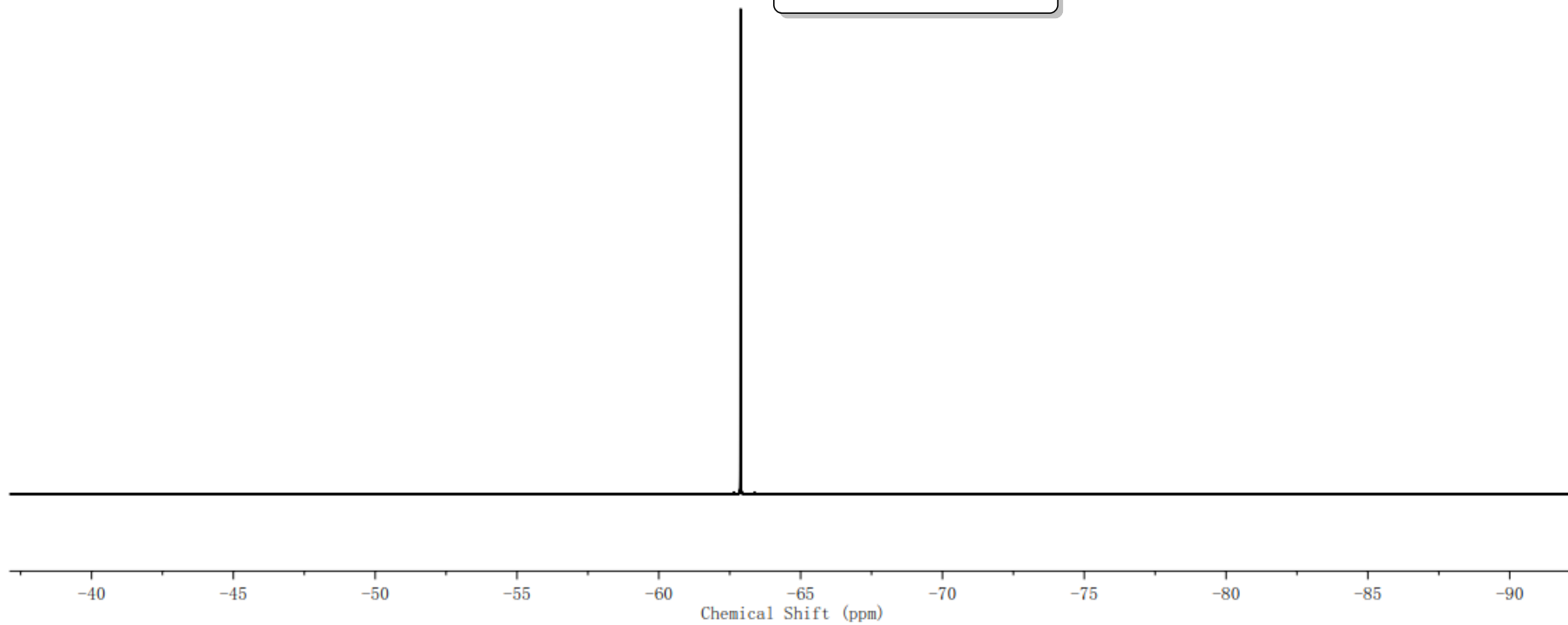
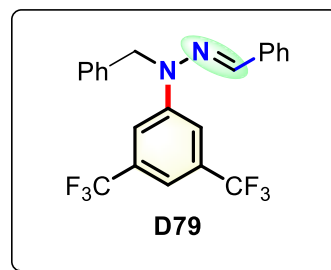
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S303

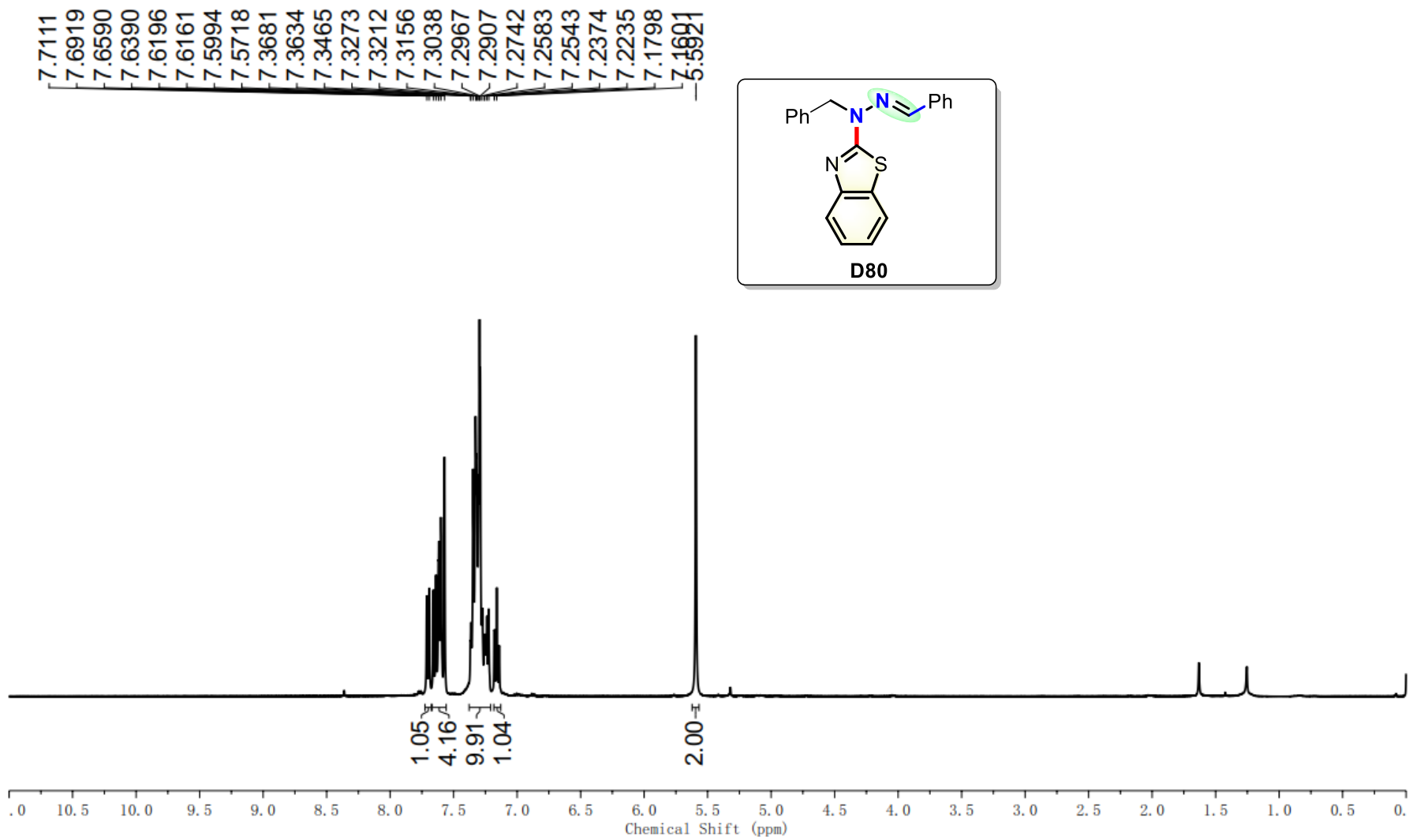
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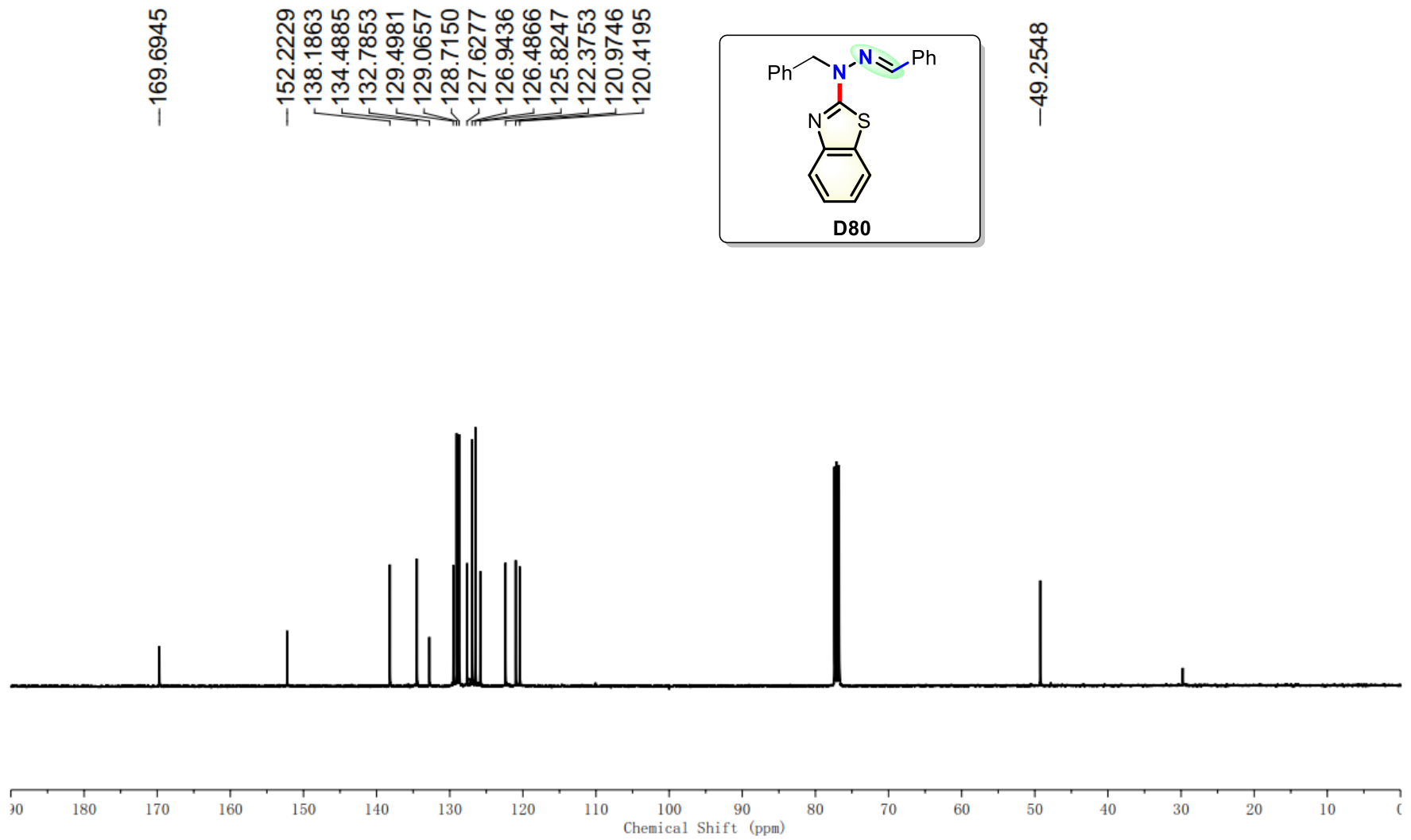
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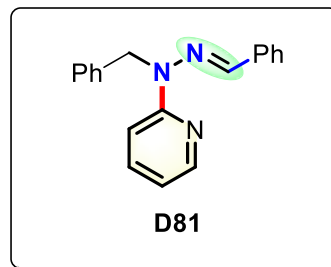
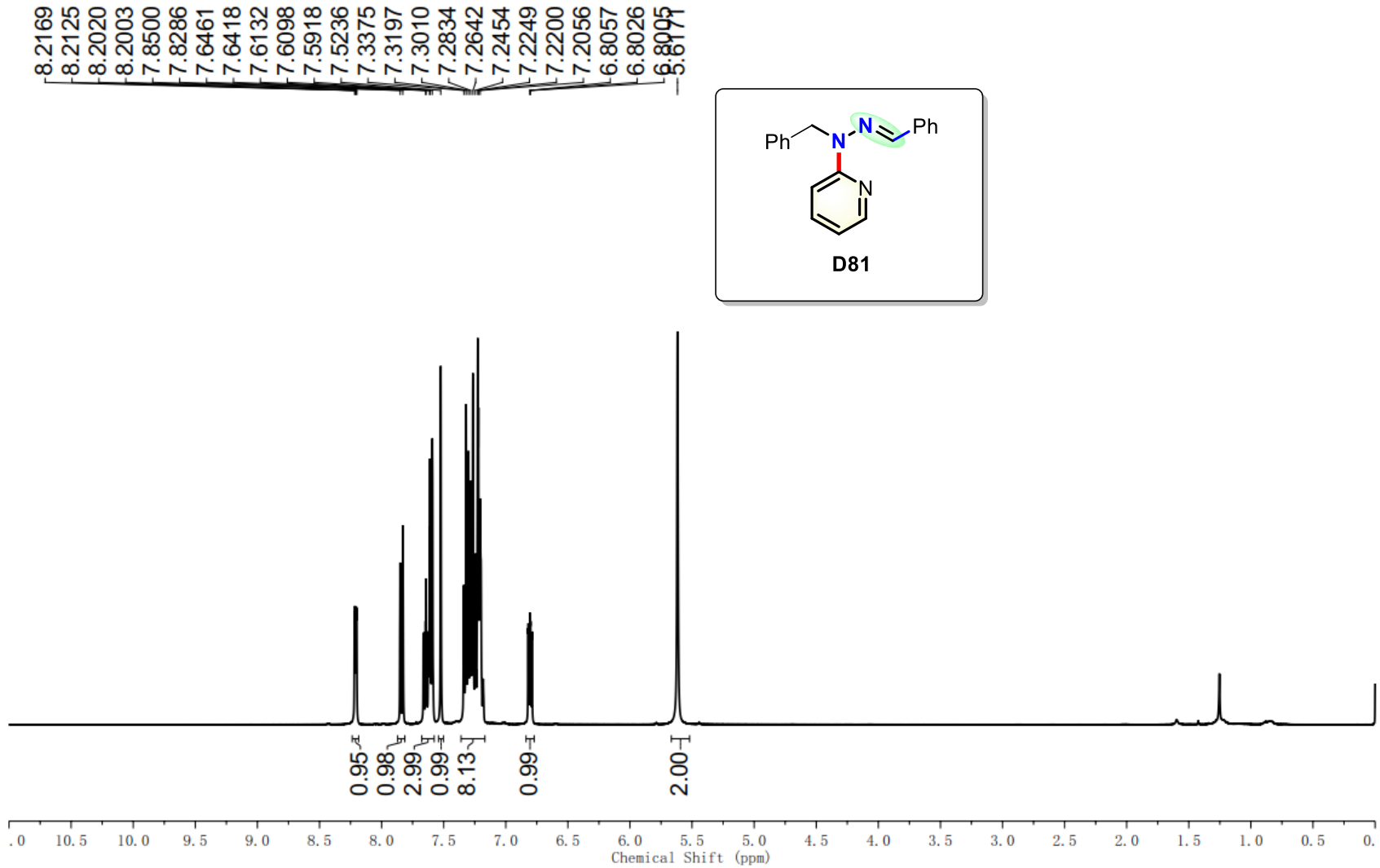


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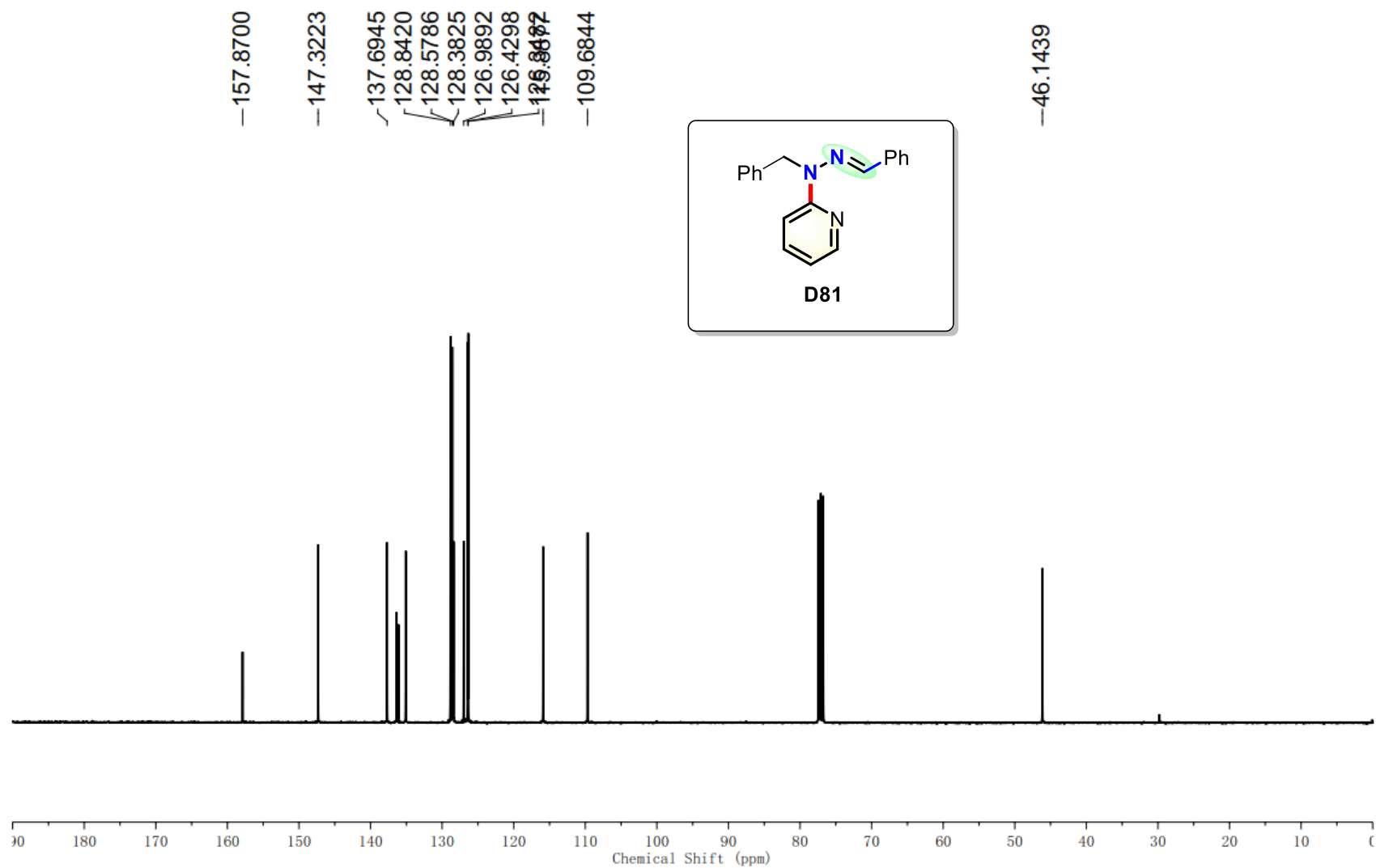
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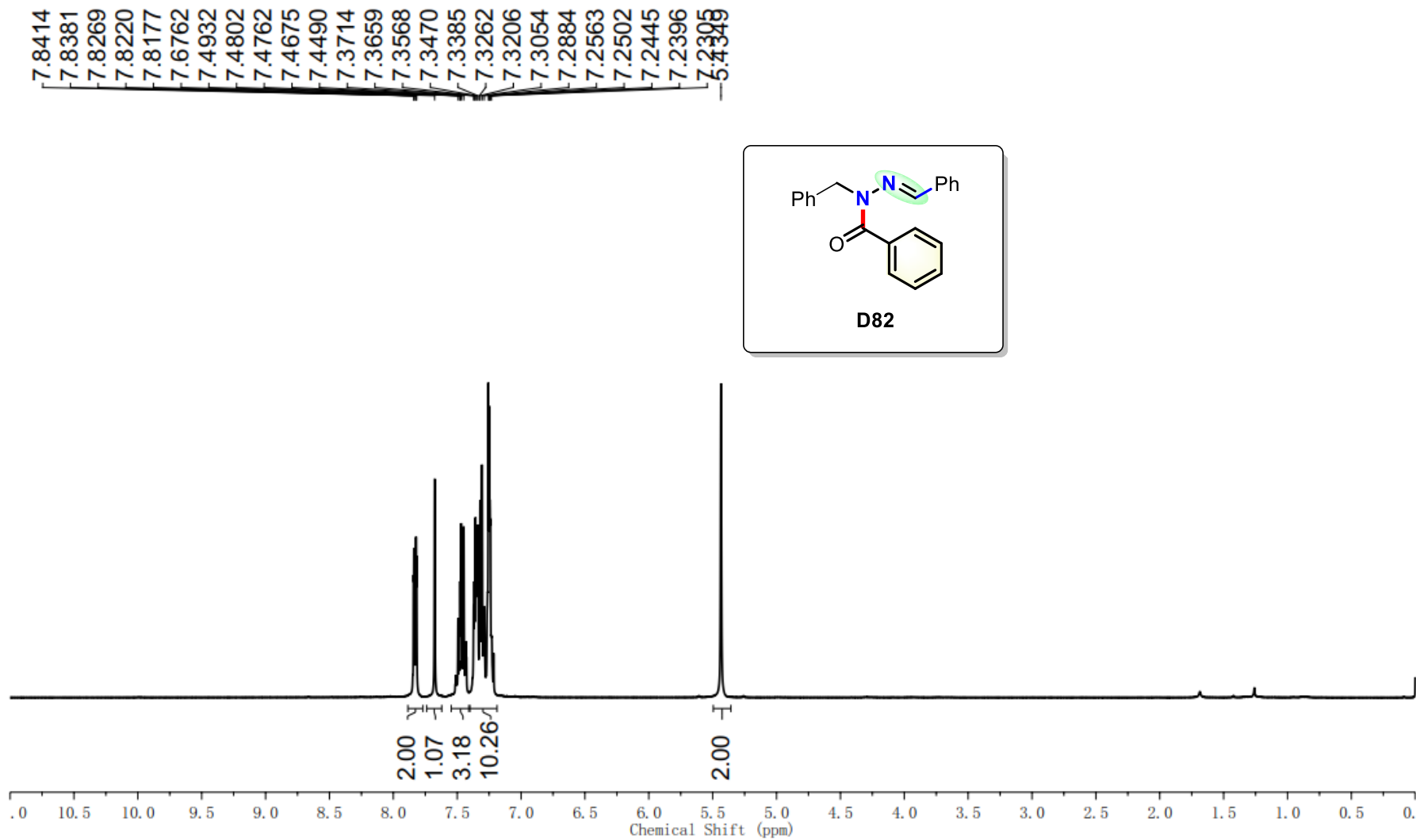


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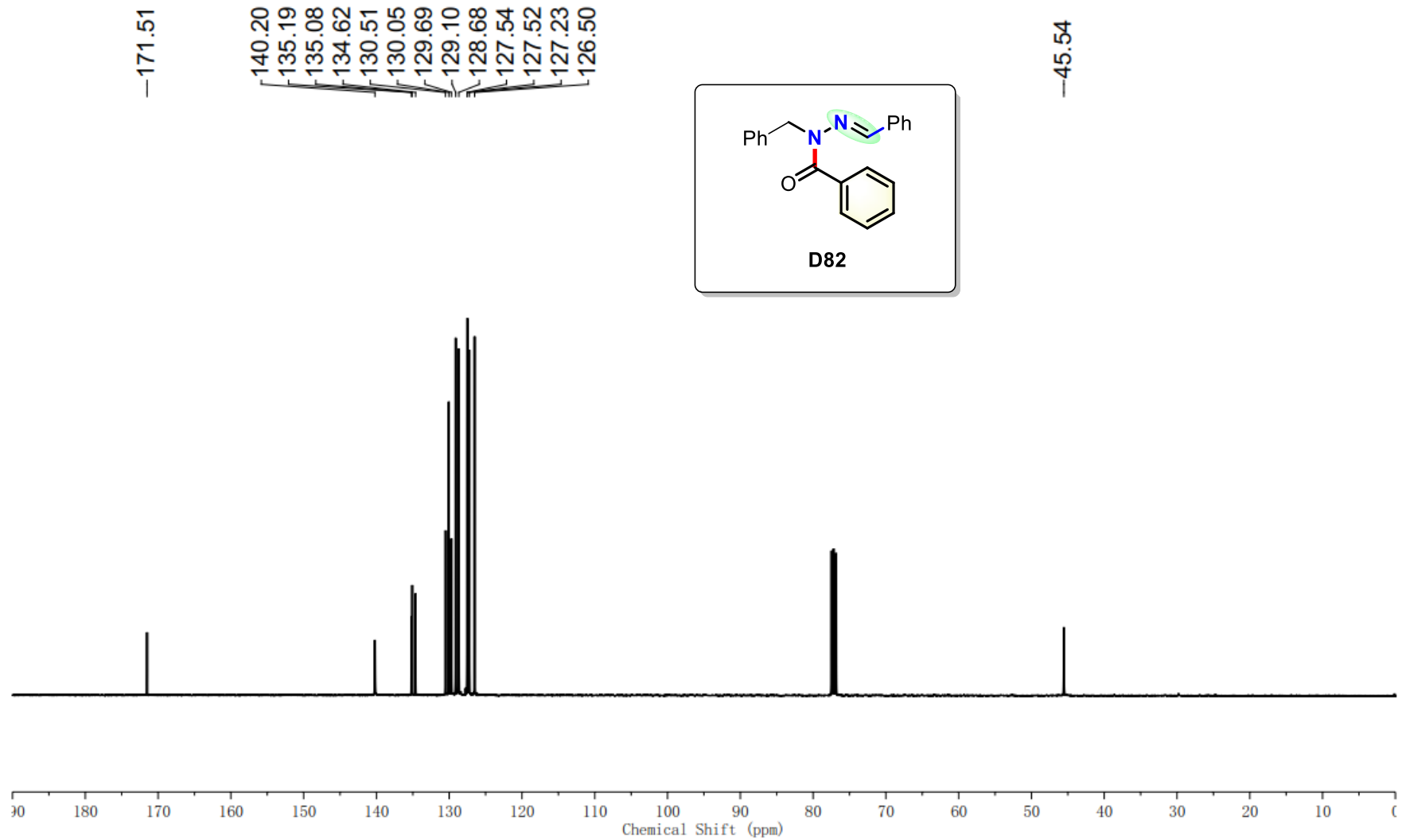


S308

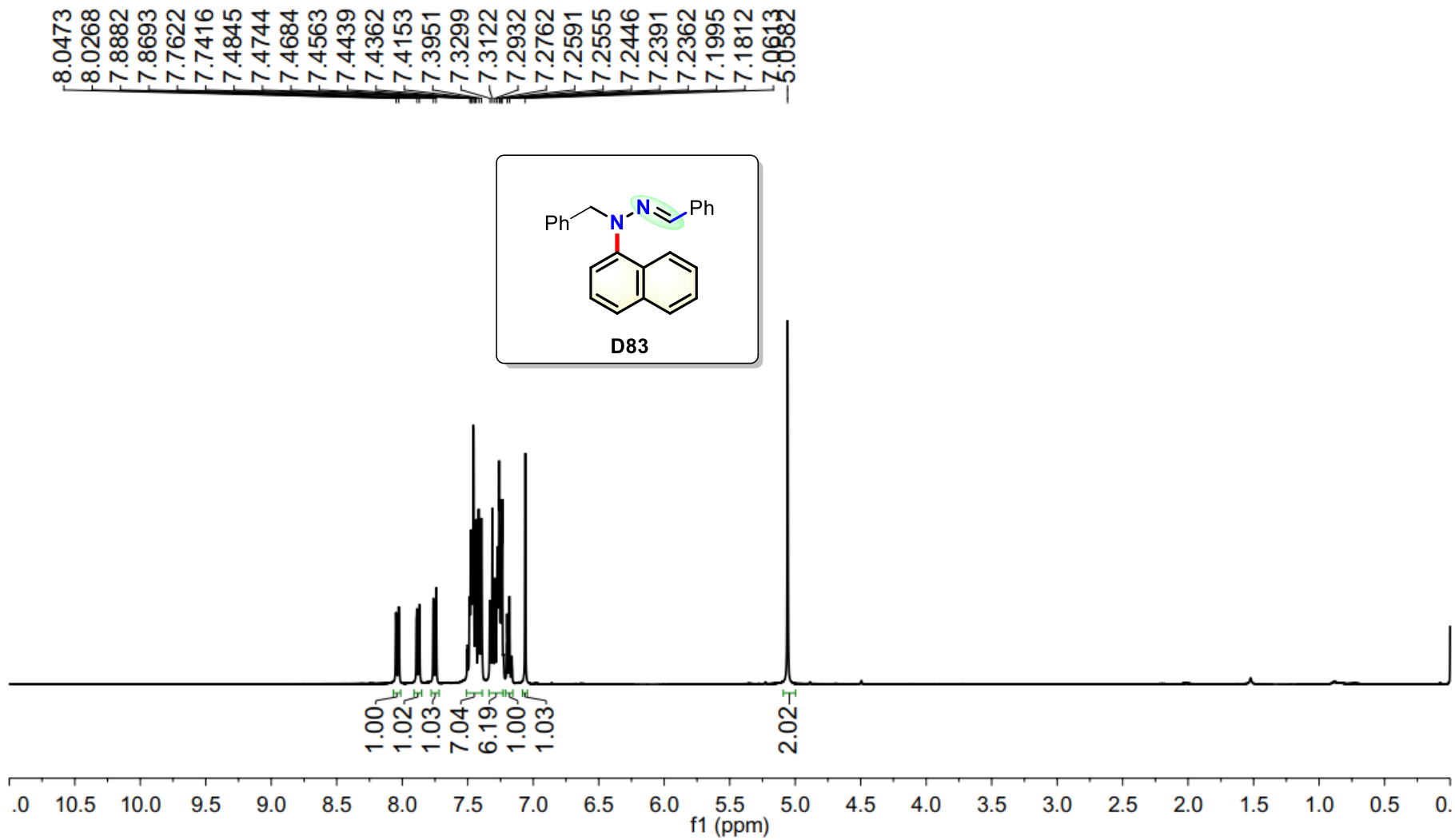
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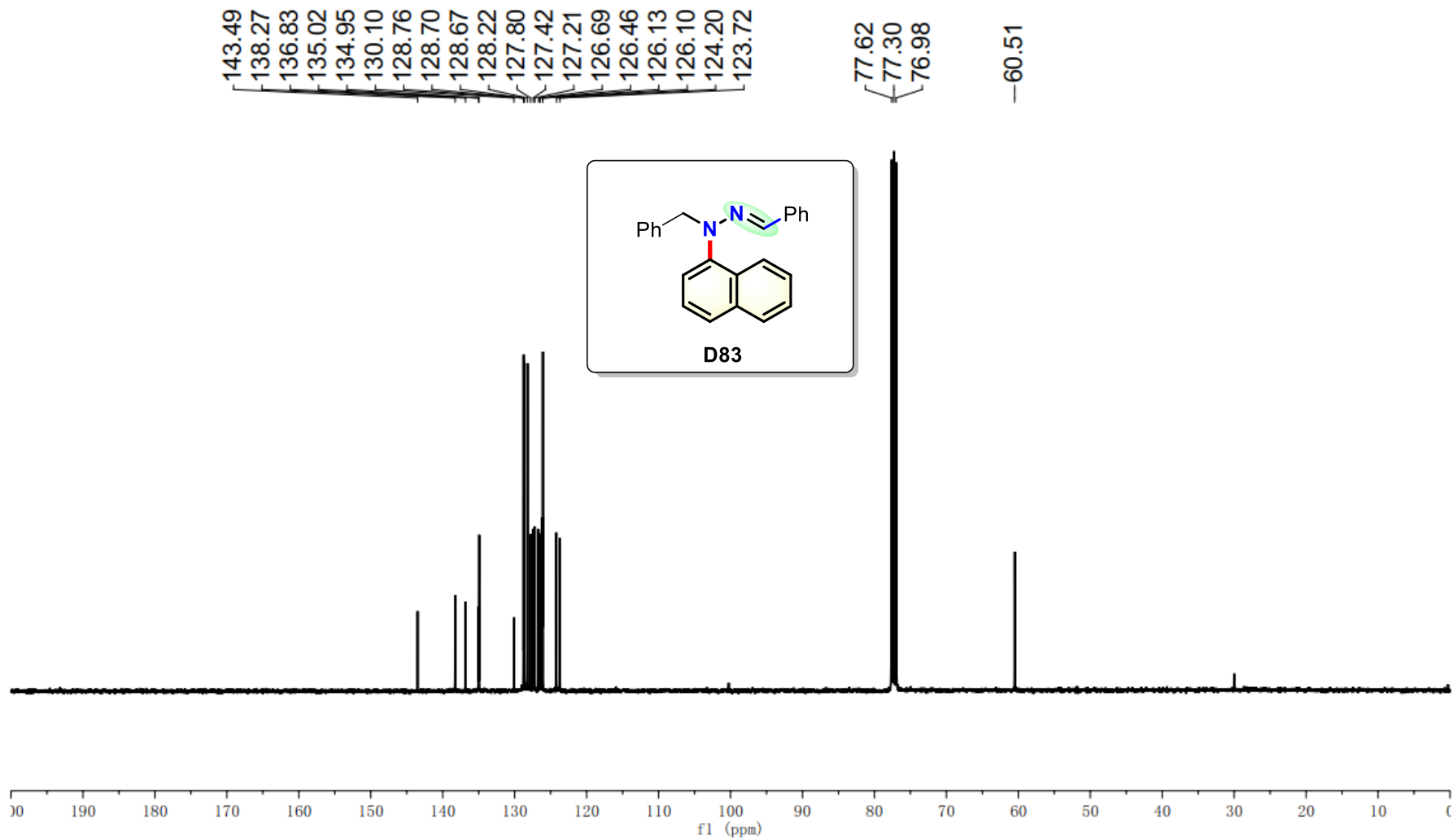
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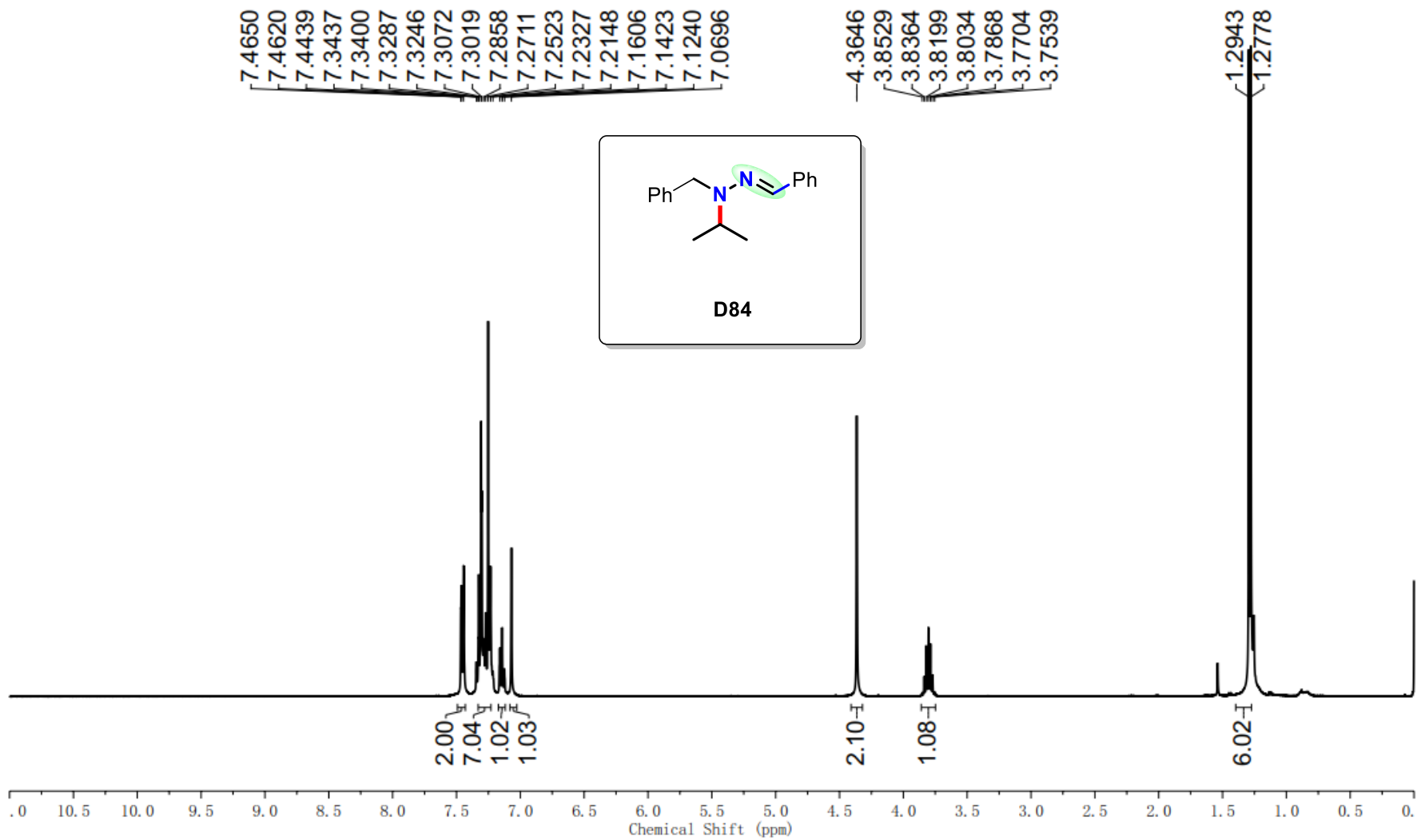


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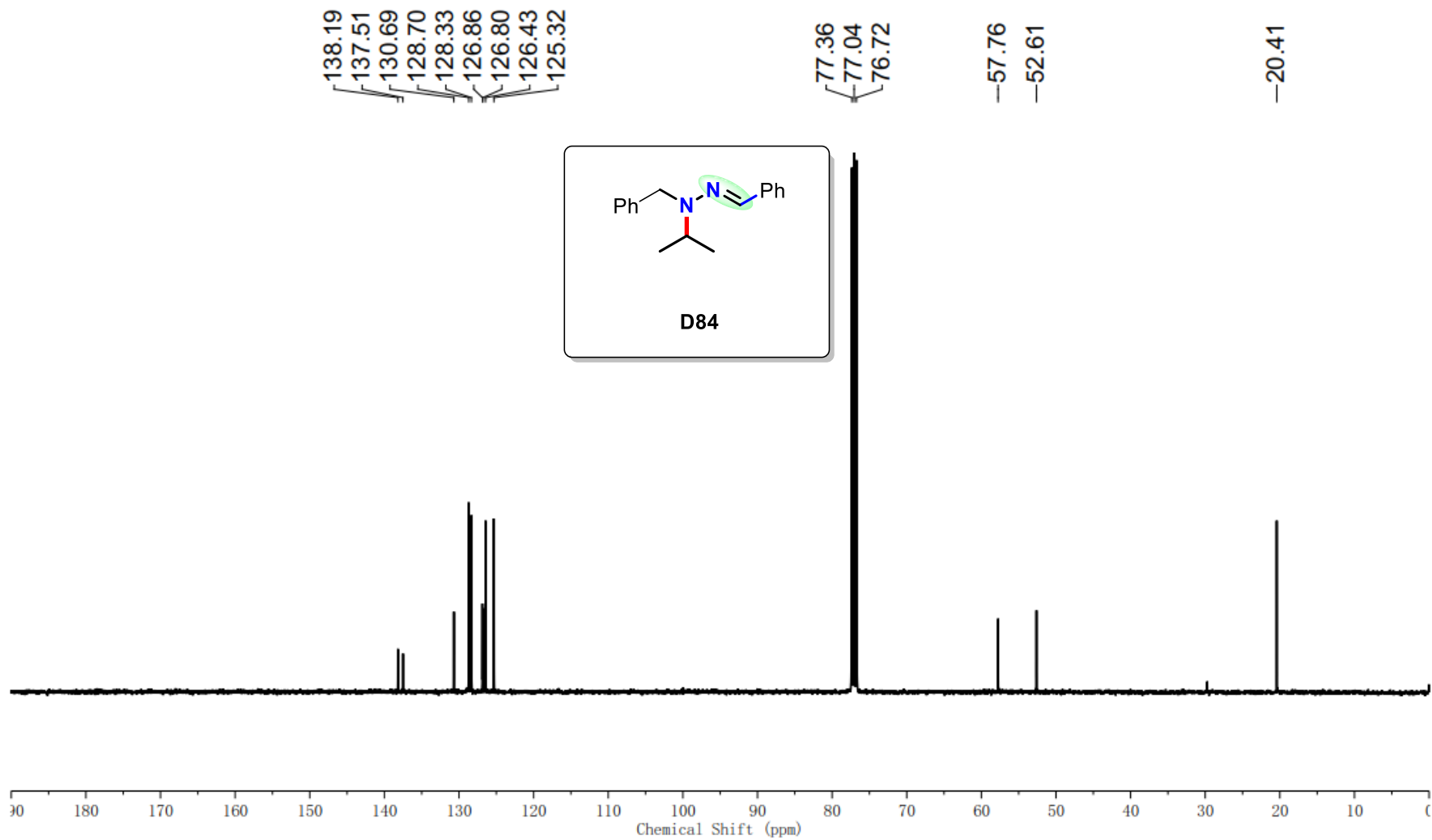


2022-10-26 yy0025 616 LQQ-20

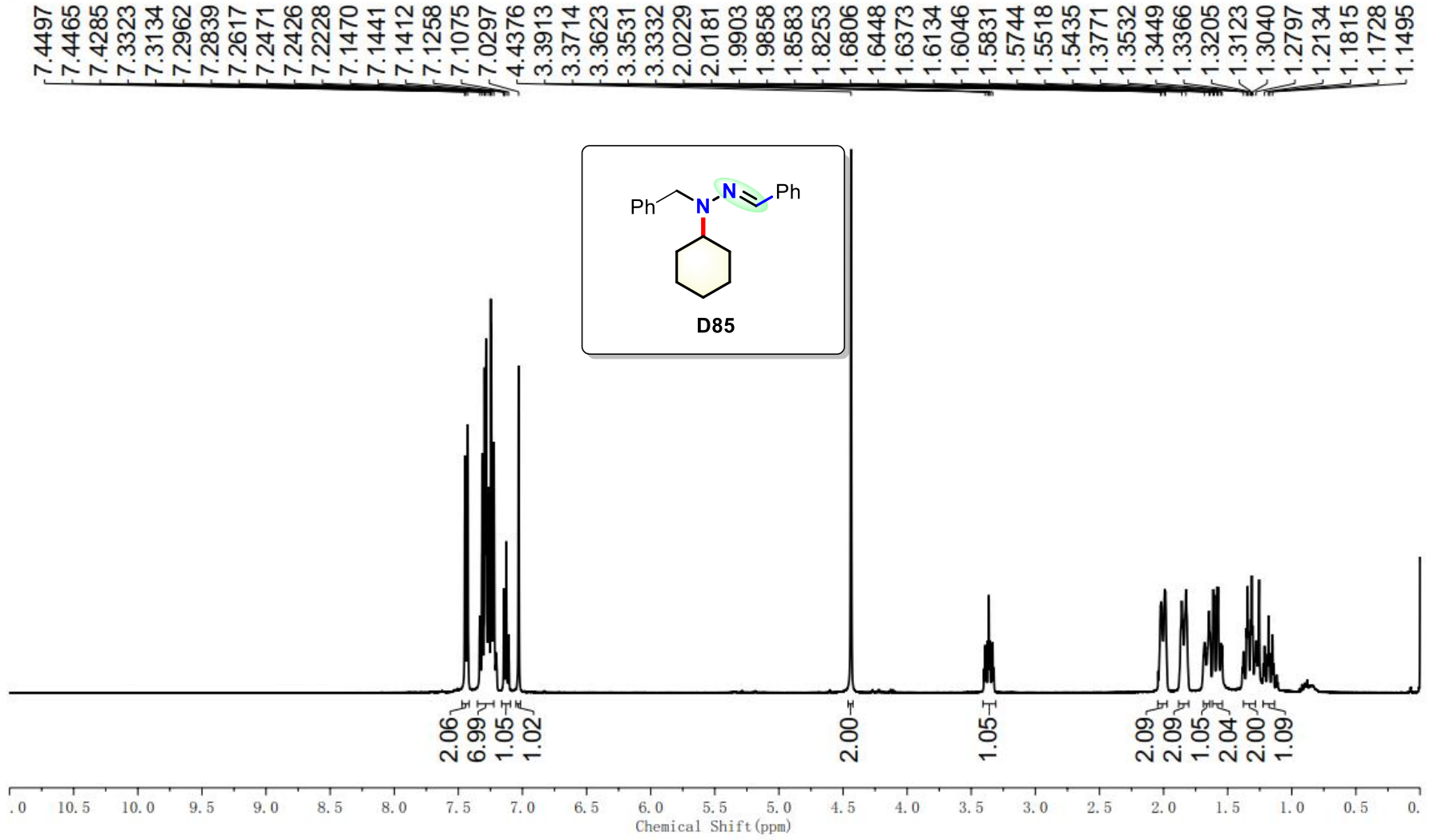




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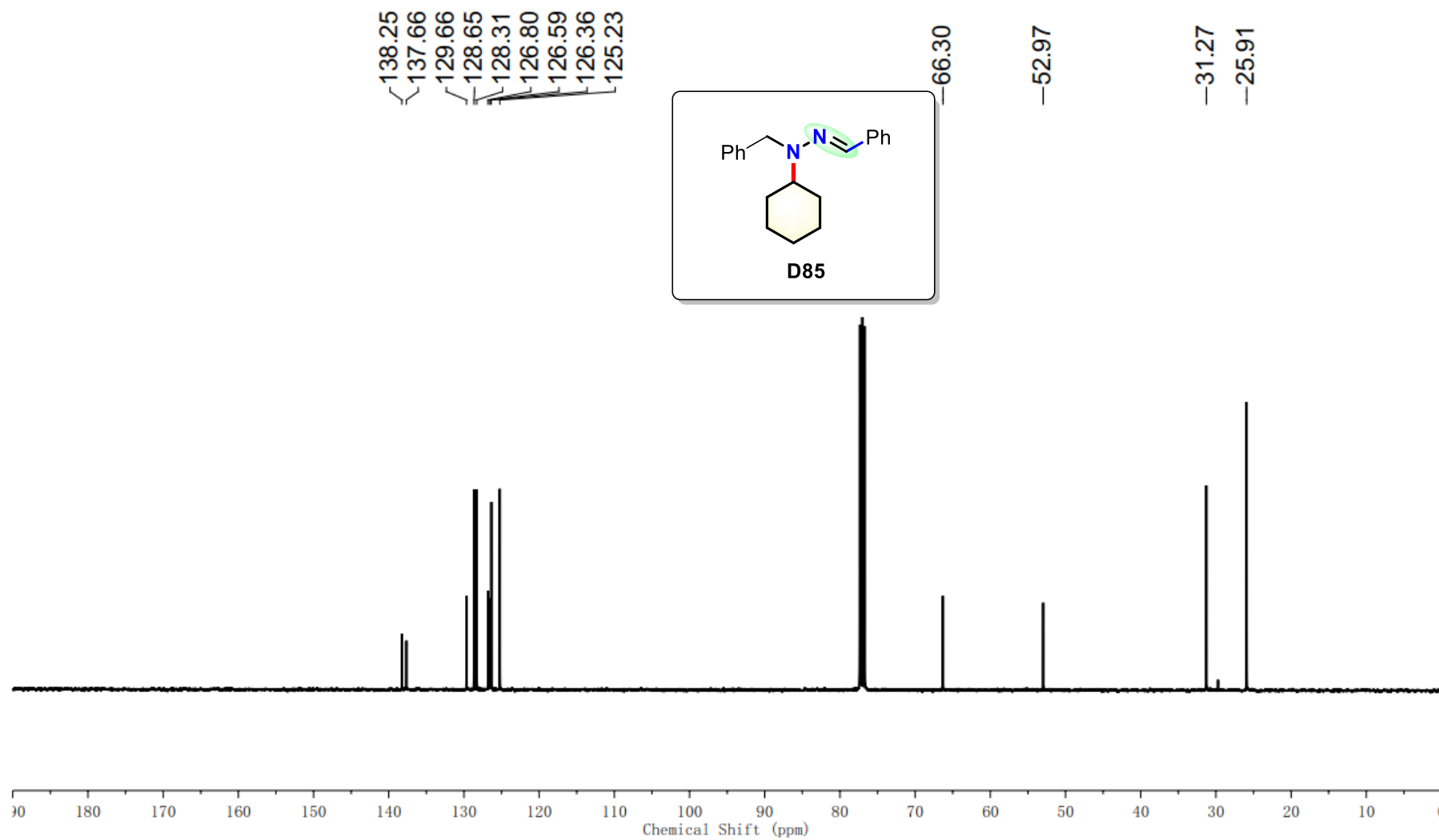


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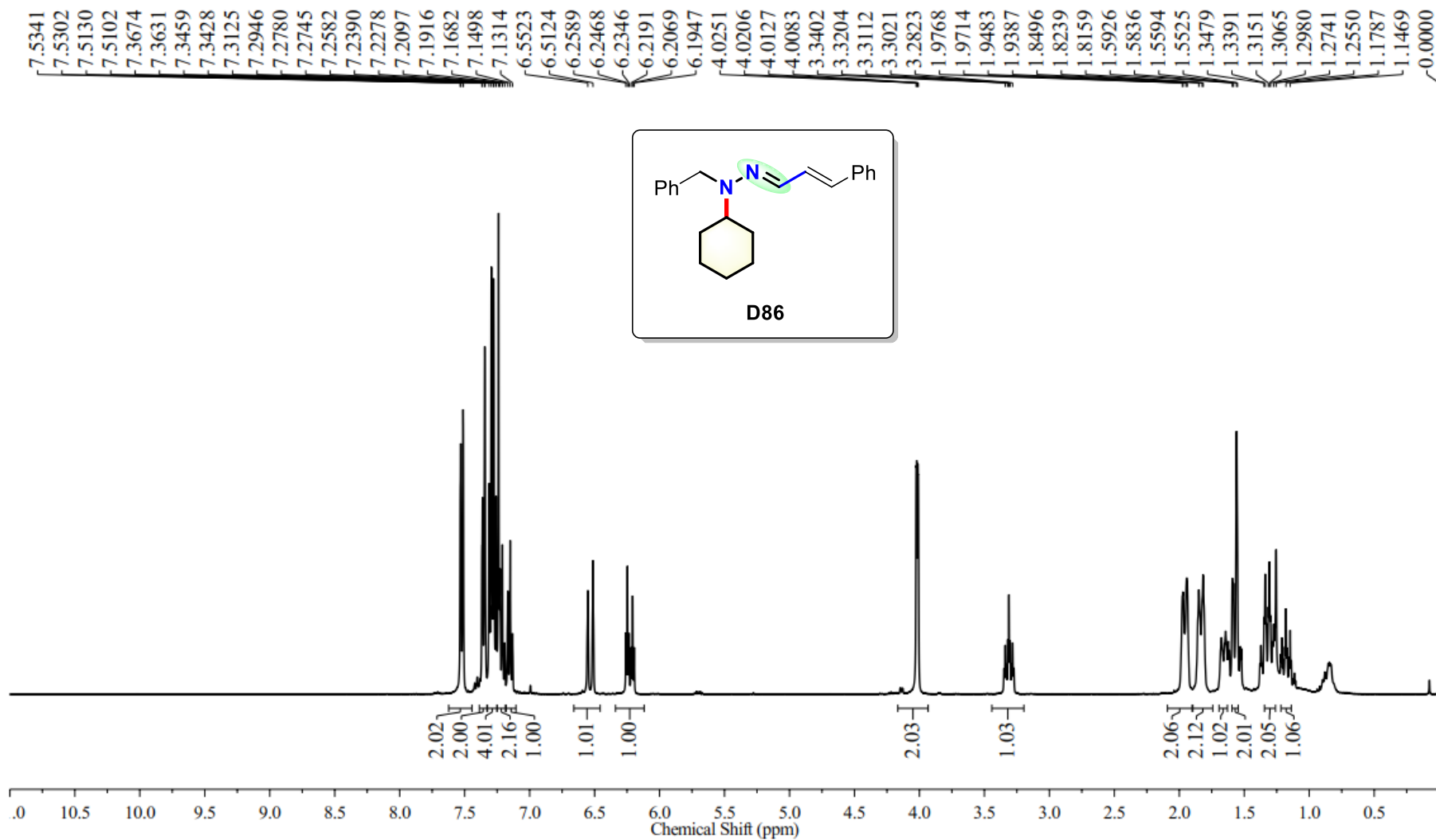


S315

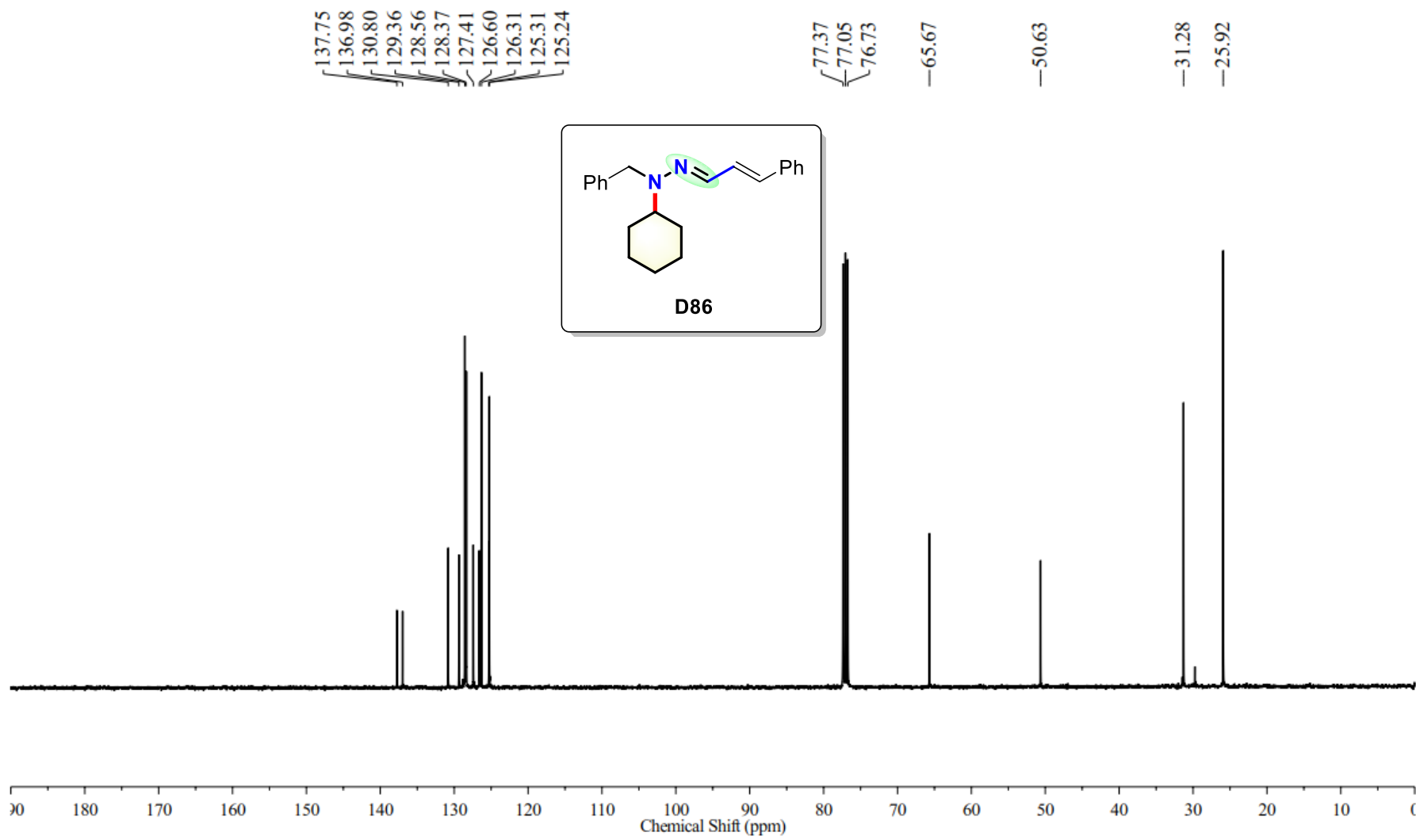
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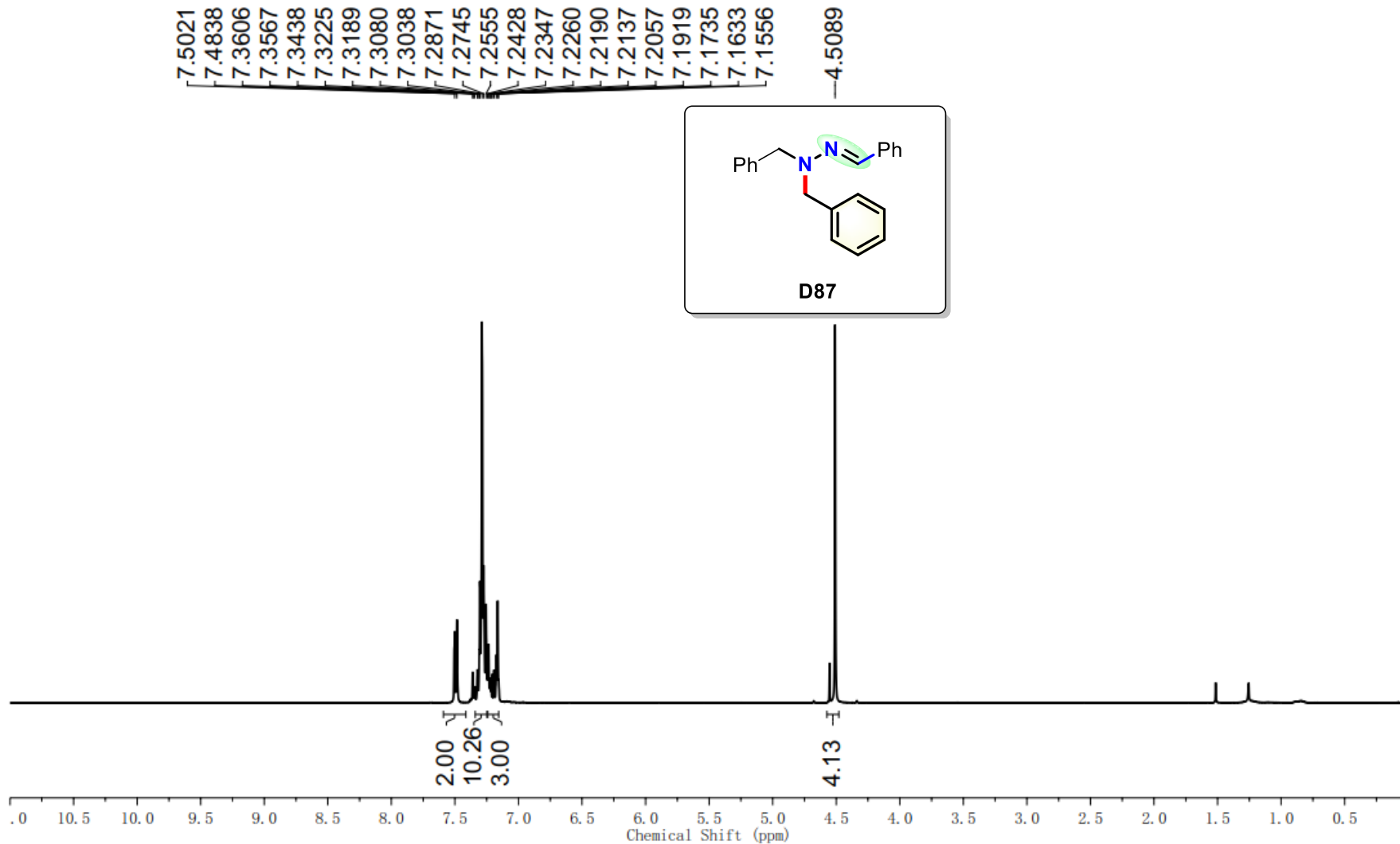
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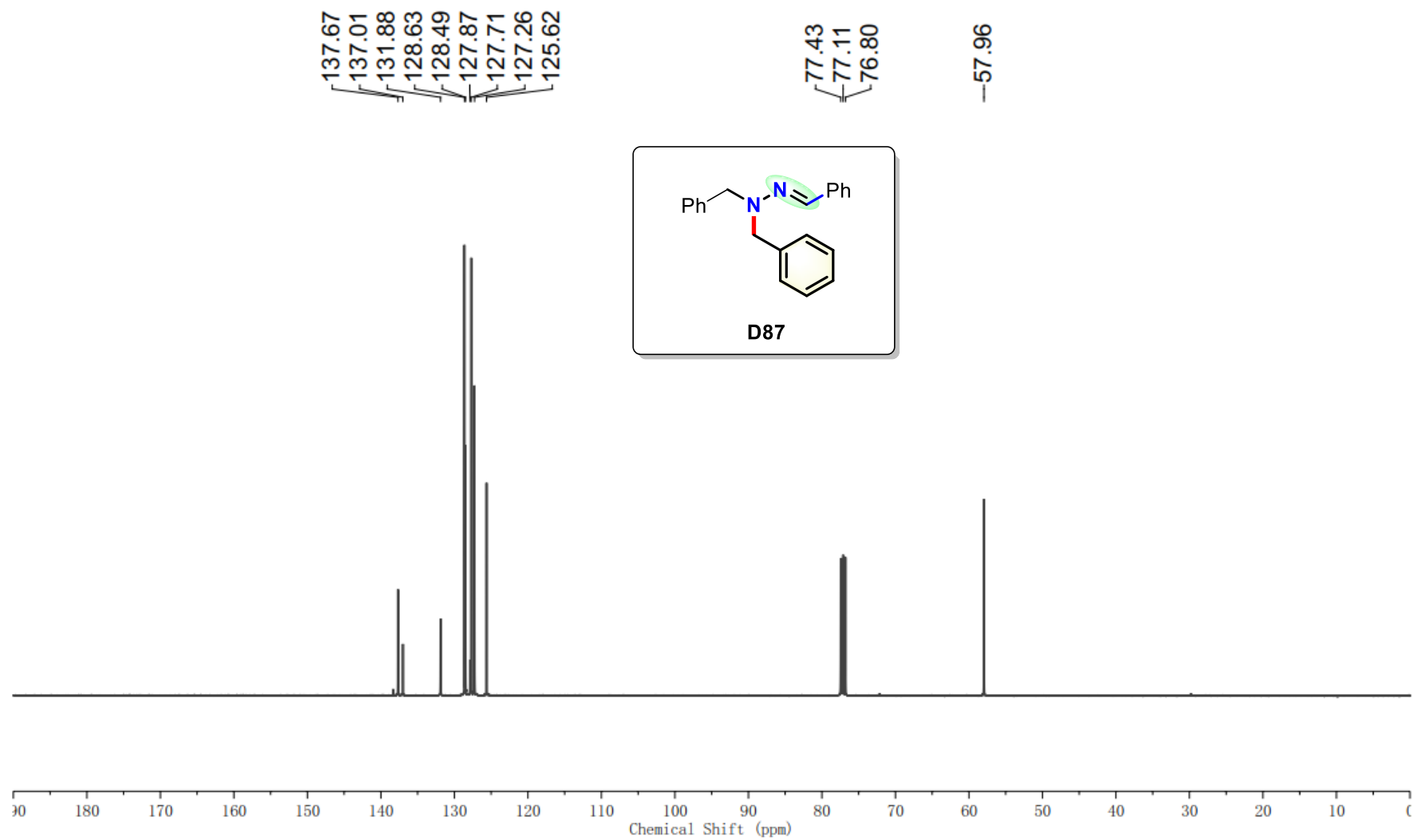
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2022-11-24 YY0014 616-lqq-F32

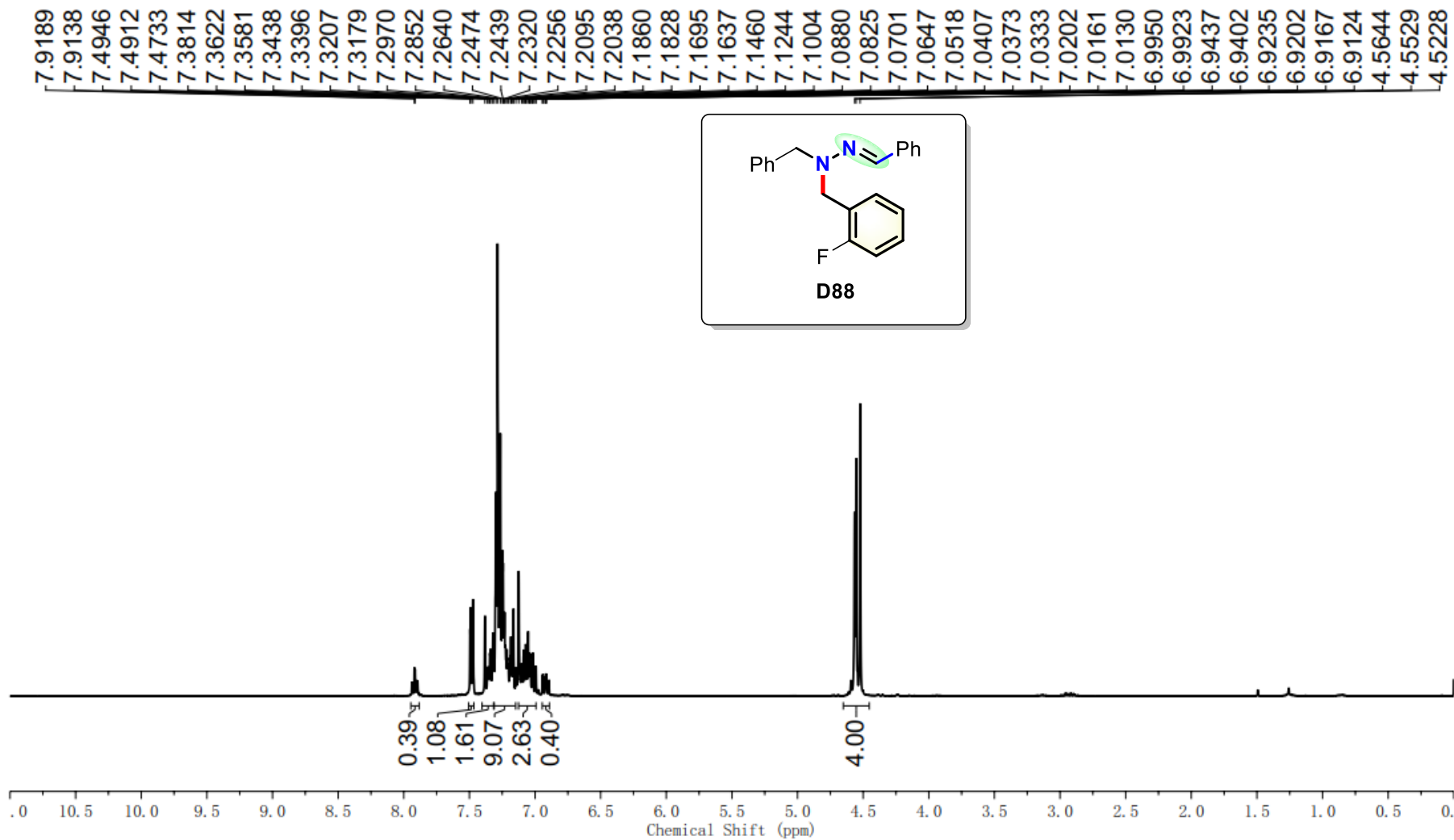


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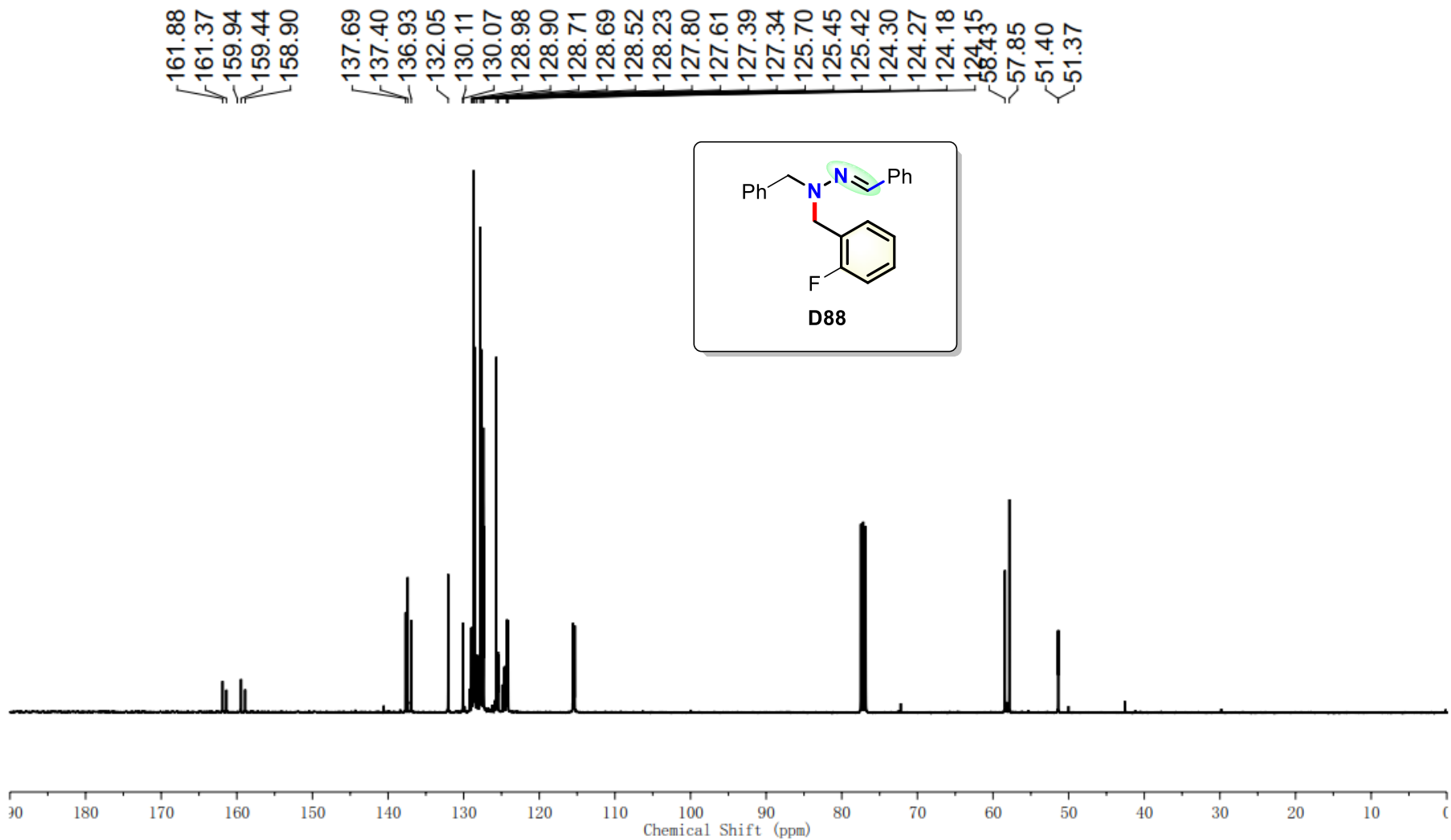


S320

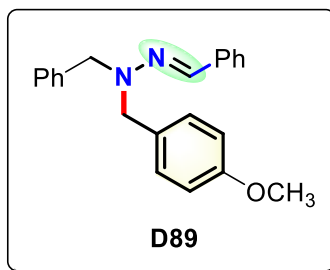
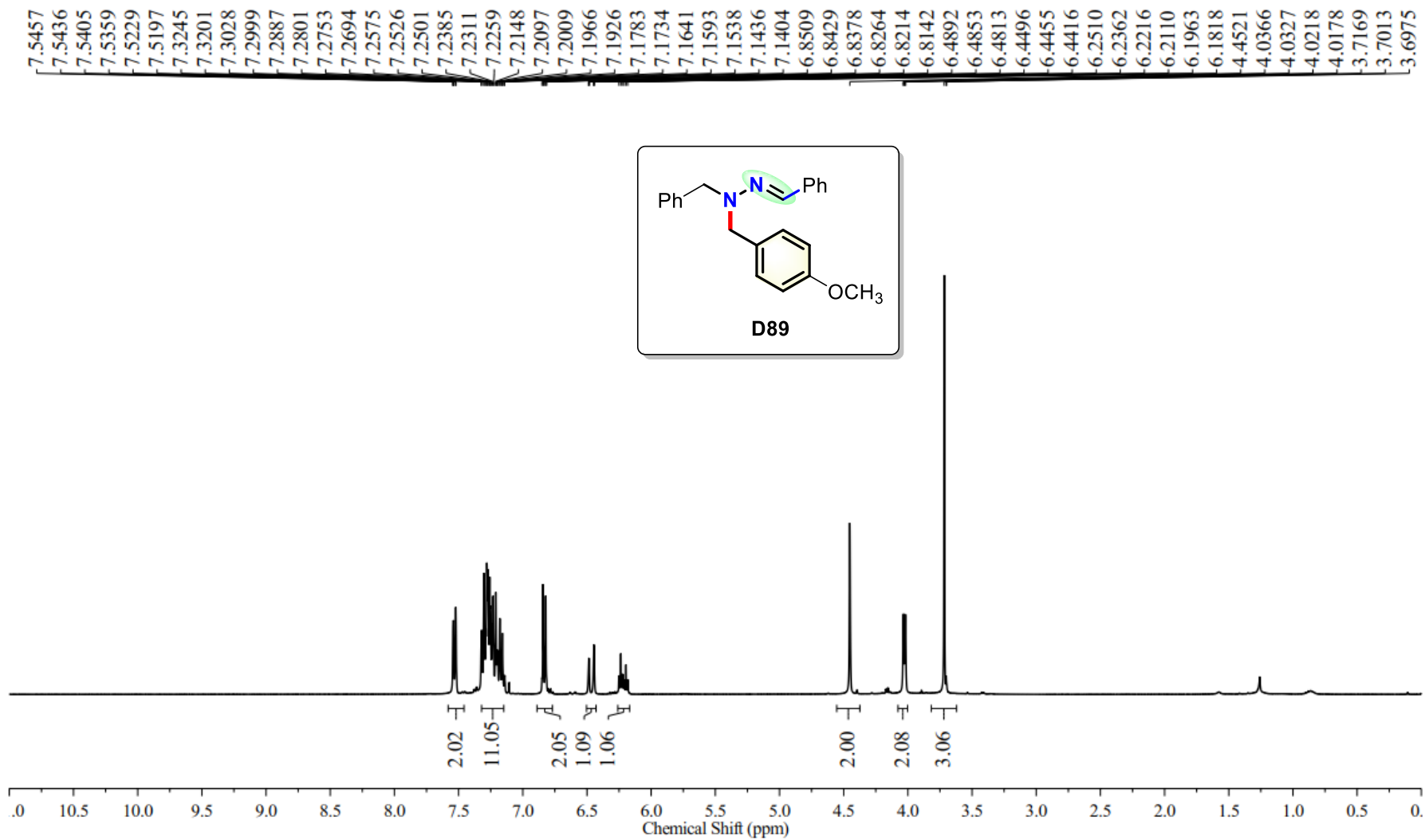
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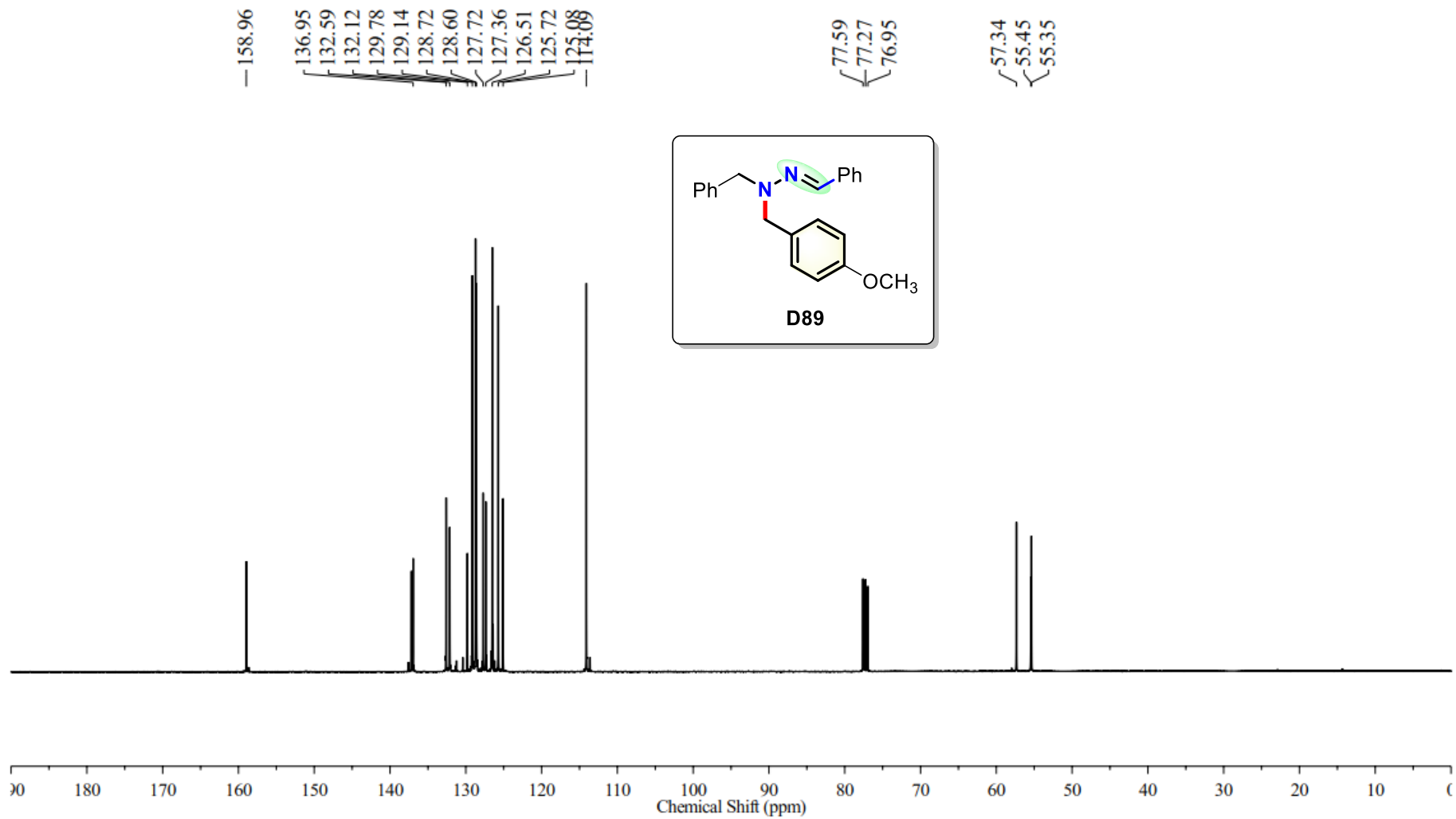
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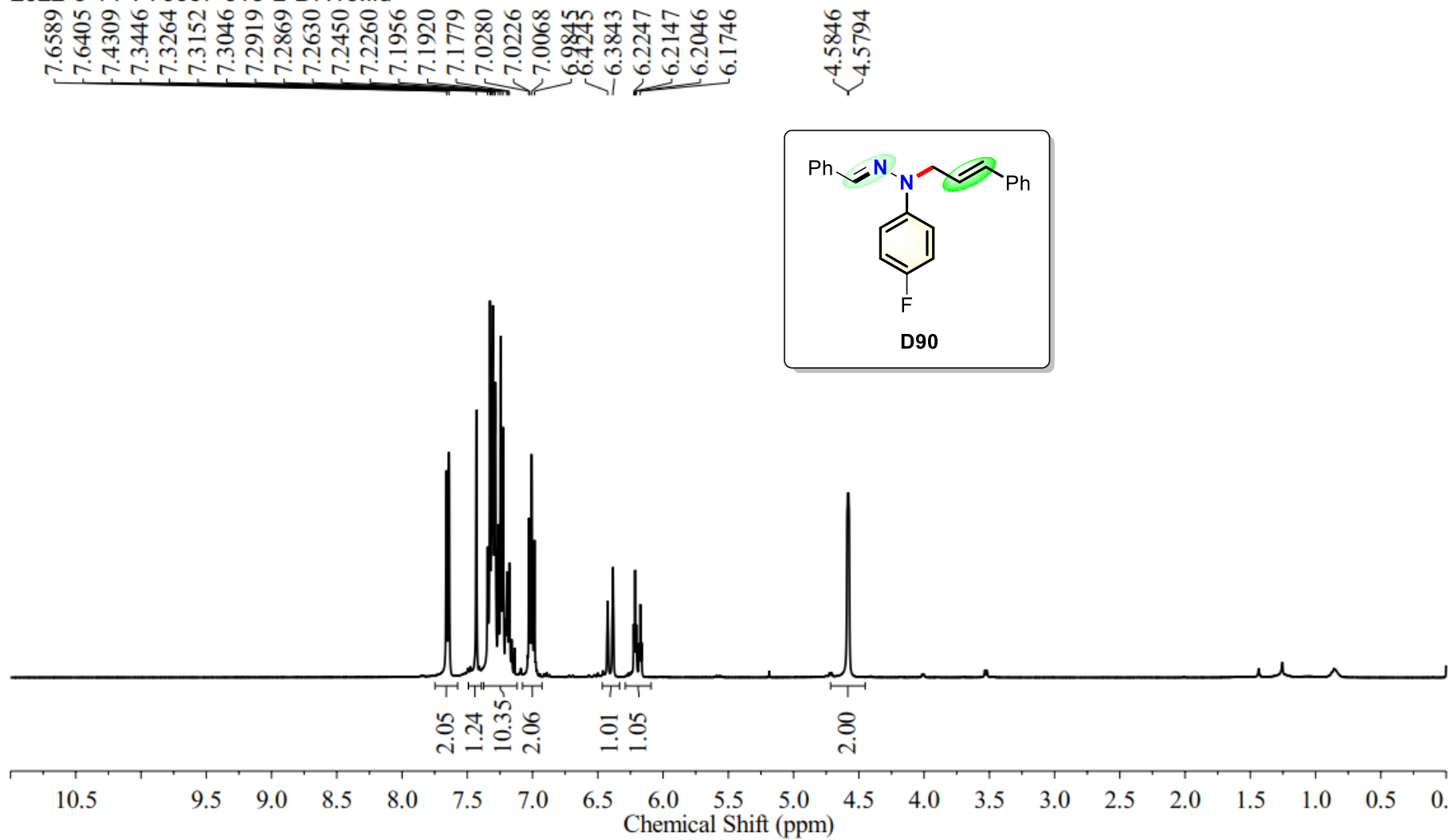
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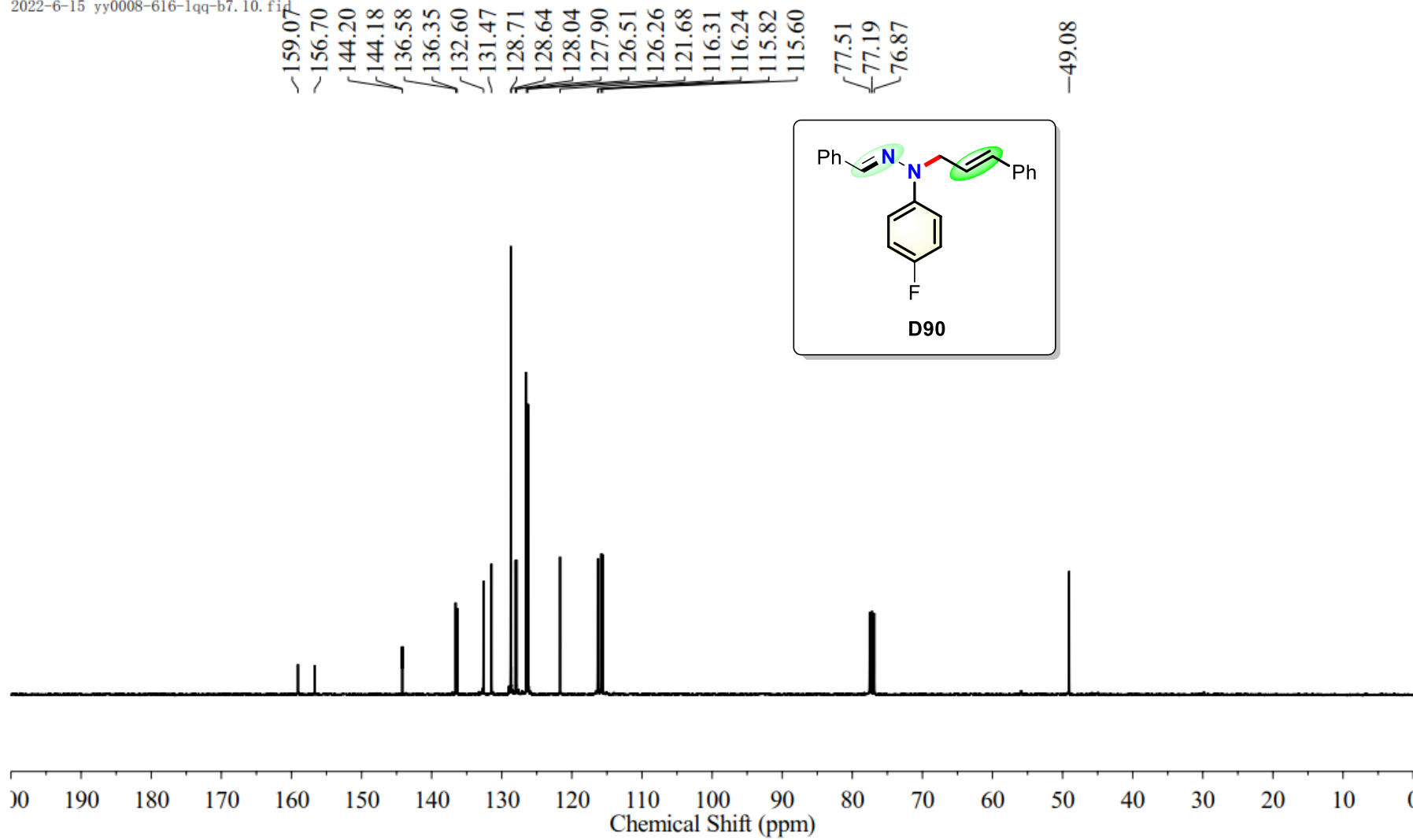
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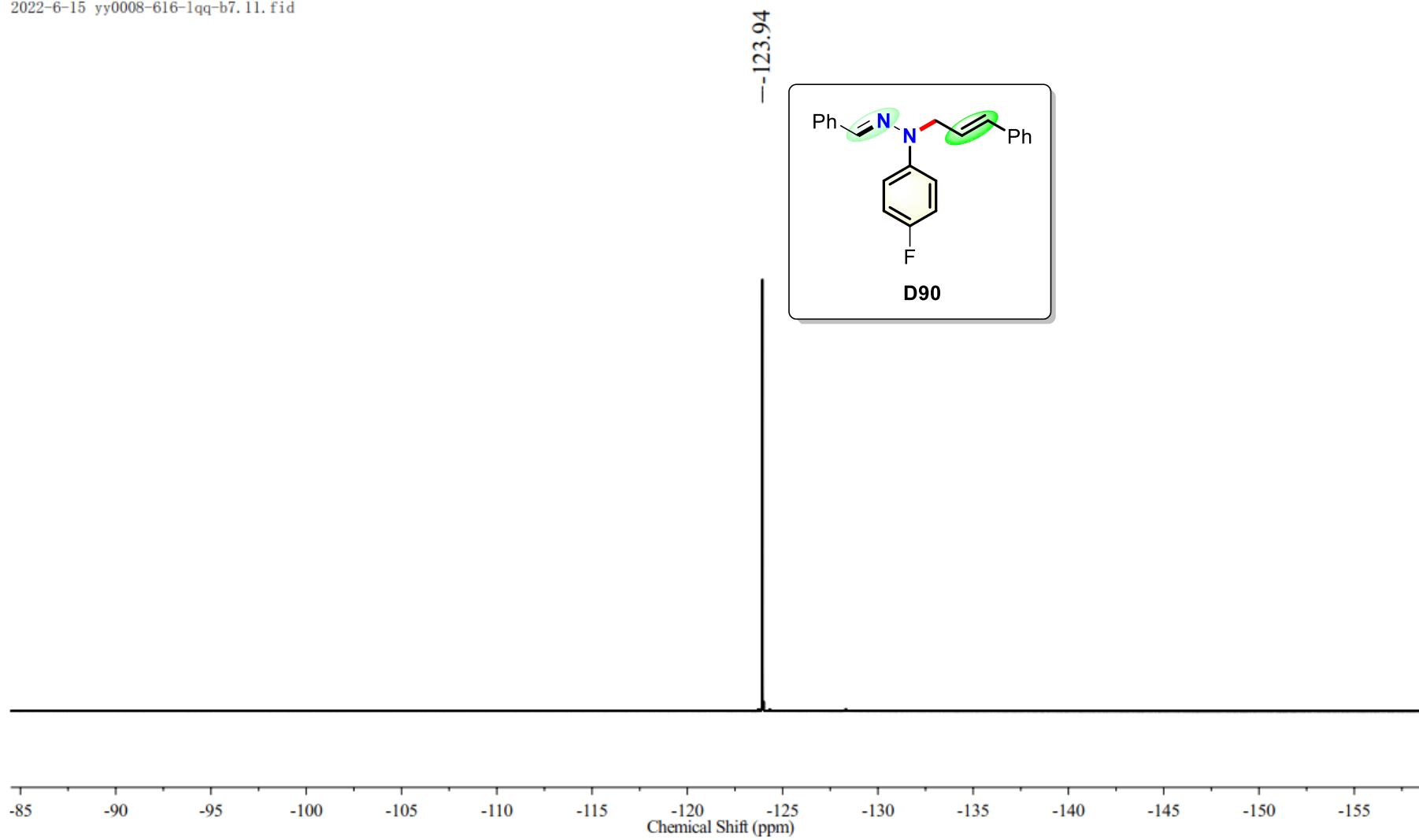


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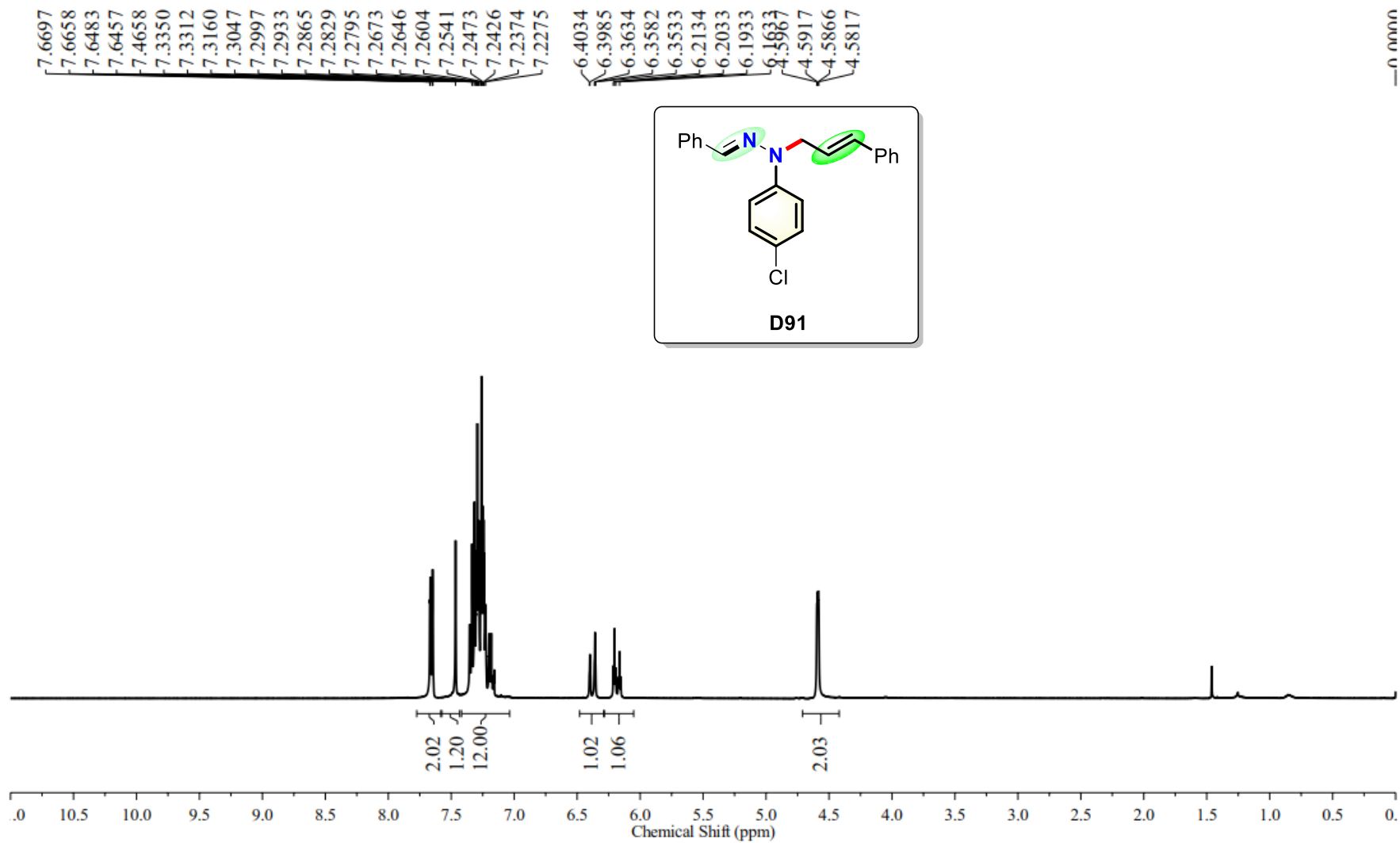


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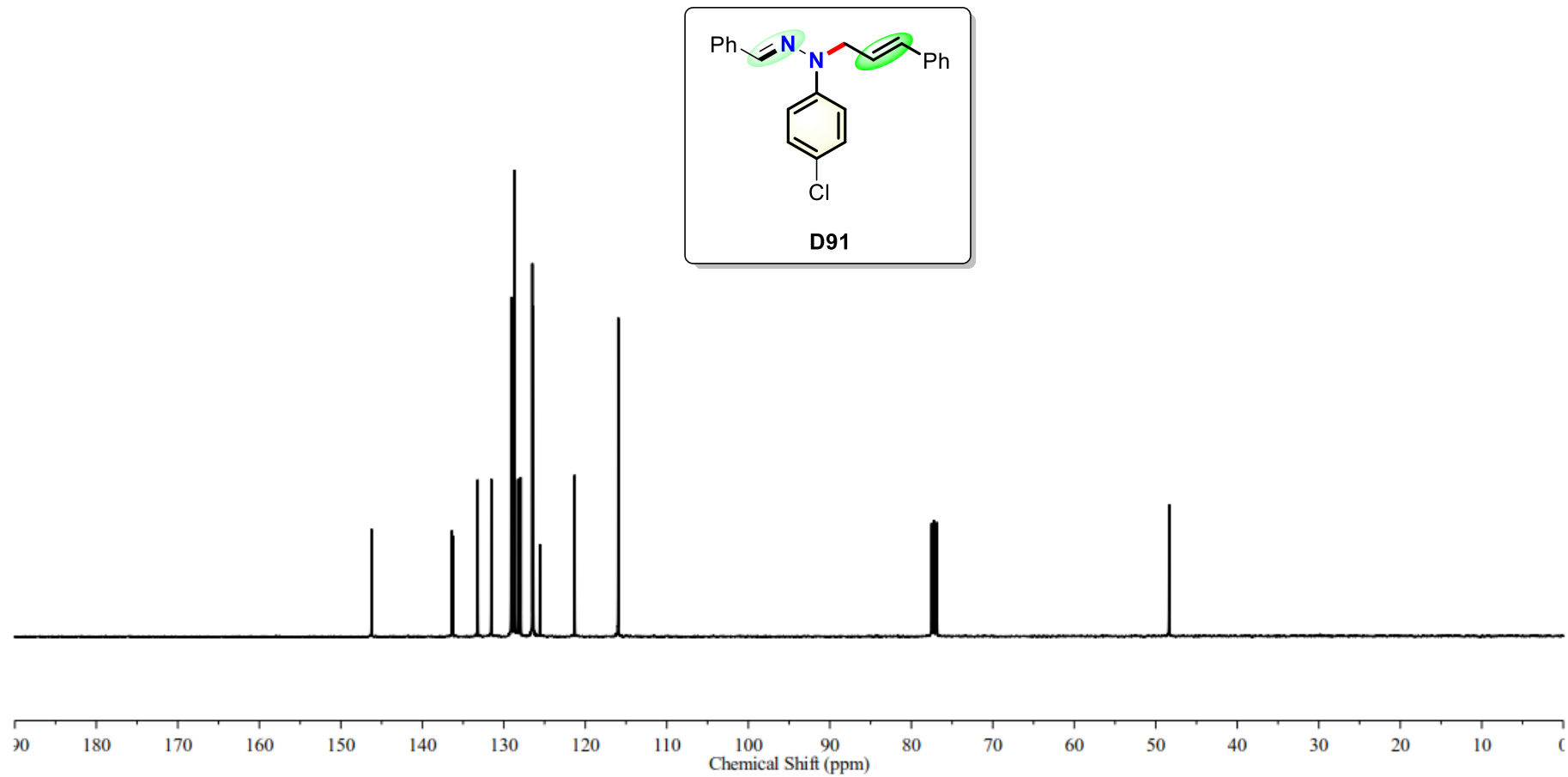
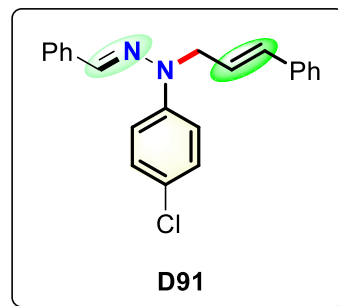


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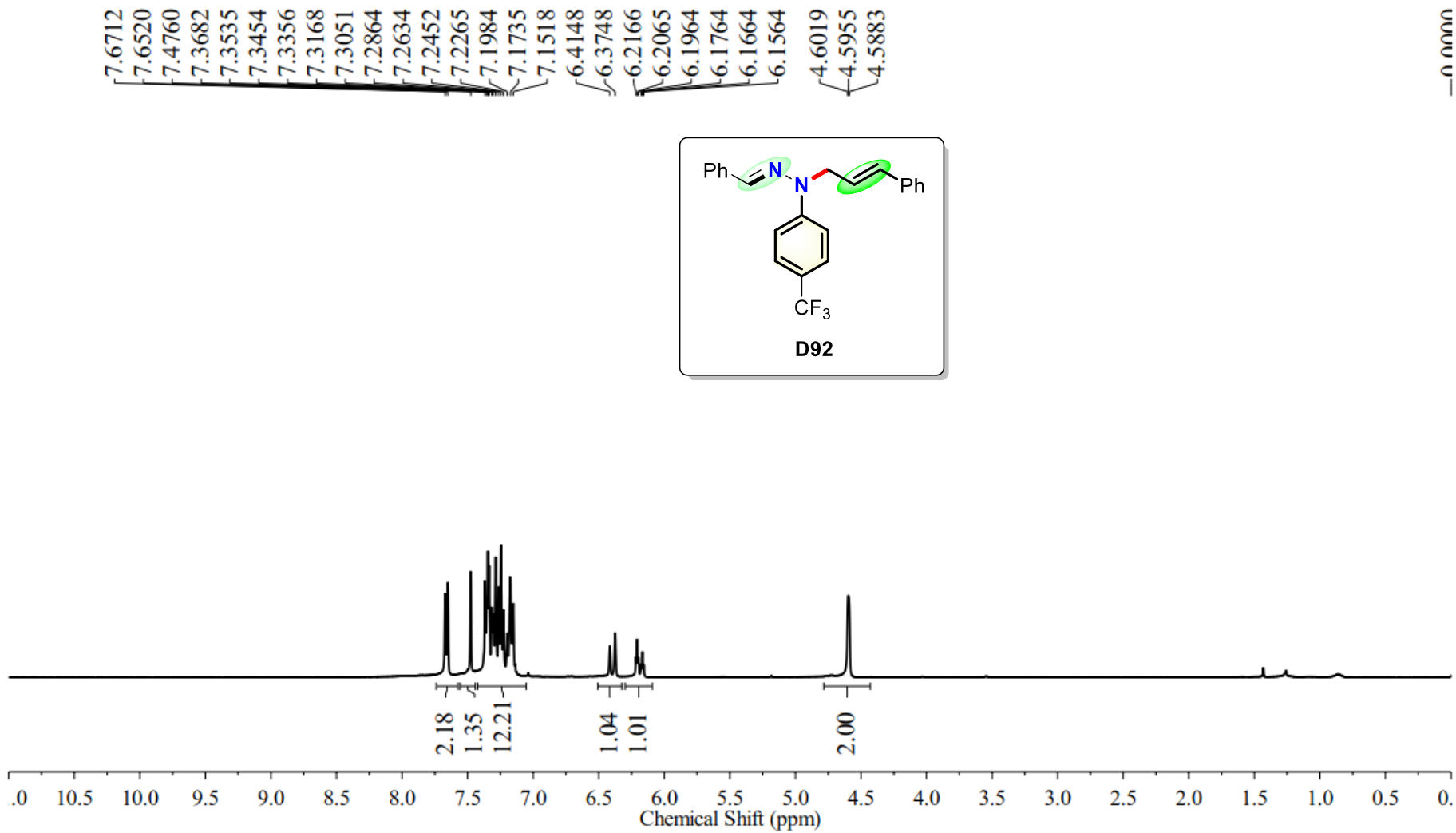
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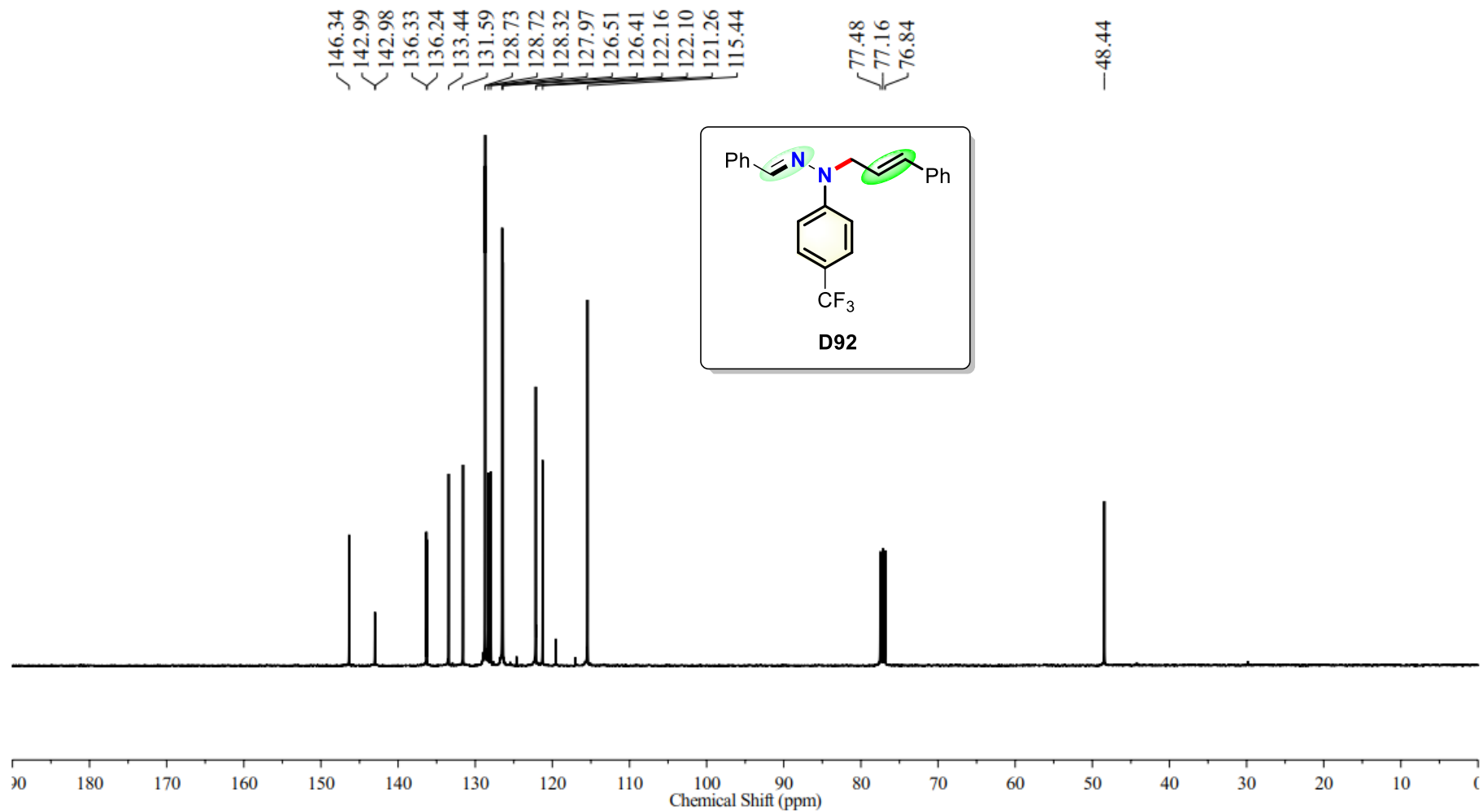
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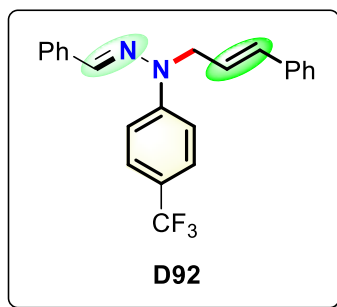
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2022-06-21 yy0025 616-lqq-12.10.fid



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--58.05

