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Supporting Information

Synthesis and characterization of bridged bis(amidato) rare earth metal amides and their applications in C-N bond formation

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Table of contents **Page**

Table 1 Details of crystallographic data.

	3·Tol	**5**·2THF	**6**·2Hex	(**7**·2Tol)$_2$	**8**·2Tol
Formula	$C_{65}H_{100}Nd_2N_6O_4Si_4$	$C_{70}H_{116}La_2N_6O_6Si_4$	$C_{74}H_{128}Nd_2N_6O_4Si_4$	$C_{152}H_{236}La_4N_{16}O_8Si_8$	$C_{76}H_{118}Nd_2N_8O_4Si_4$
Fw	1428.48	1527.87	1566.44	3193.30	1608.62
T (K)	293(2)	253(2)	293(2)	223(2)	223(2)
Crystsyst	Monoclinic	Monoclinic	Monoclinic	Monoclinic	Triclinic
Space group	C12/c1	P12/c1	P1c1	P21/n	P-1
Crystal size (mm^3)	1.40×0.80×0.80	0.50×0.40×0.40	0.75×0.60×0.40	0.50×0.30×0.20	0.30×0.20×0.20
a (Å)	20.64(4)	13.89(1)	13.84(1)	16.70(1)	12.38(1)
b (Å)	19.89(2)	11.7360(3)	11.67(1)	17.83(1)	12.68(1)
c (Å)	20.36(2)	24.69(1)	24.69(1)	27.92(1)	14.02(1)
α (°)	90	90	90	90	83.14(3)
β (°)	116.44(2)	103.73(1)	103.91(1)	104.87(1)	74.52(3)
γ (°)	90	90	90	90	72.32(3)
V (Å^3)	7482(16)	3909.4(2)	3872.0(1)	8037.9(3)	2020.3(8)

Z	4	2	2	4	1
D_{calc}(g/cm^3)	1.230	1.298	1.320	1.206	1.316
μ (mm^{-1})	1.479	1.188	1.436	1.152	1.379
F (000)	2784	1592	1588	3010	830
θ range (^{o})	2.85-26.37	3.02-25.00	3.03-25.00	3.02-26.37	2.85-25.00
Collected	16113	21353	20595	58323	17119
Unique reflns	7539	6880	11148	16402	7100
R [I > 2σ (I)]	0.0412	0.0412	0.0389	0.0456	0.0338
wR	0.1809	0.1089	0.1016	0.1190	0.0527
R_{int}	0.0412	0.0382	0.0225	0.0417	0.0256
GOF	1.091	1.083	1.071	1.059	1.093
peak/hole (e/Å^3)	1.587, -1.469	1.632, -0.861	1.131, -1.321	1.458, -1.450	0.631, -0.418

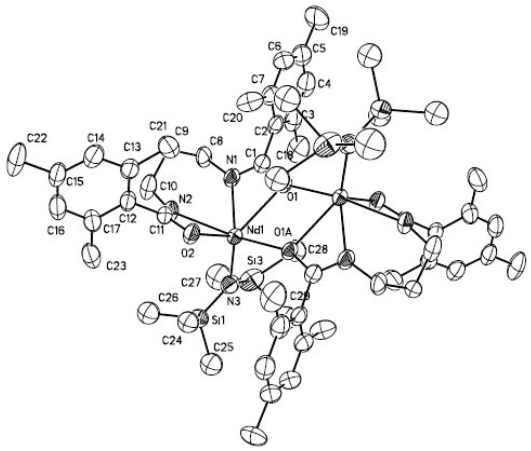


Fig. 1 Molecular structure of complex **3**·toluene. Hydrogen atoms and toluene molecules are omitted for clarity. Selected bond lengths (Å): Nd(1)-N(3) 2.312(8), Nd(1)-N(2) 2.430(8), Nd(1)-N(1) 2.491(7), Nd(1)-O(2) 2.397(6), Nd(1)-O(1) 2.554(5), Nd(1A)-O(1) 2.384(6), N(1)-C(1) 1.263(11), N(2)-C(11) 1.274(11), O(1)-C(1) 1.313(9), O(2)-C(11) 1.308(10); bond angles (°): Nd(1)-O(1)-C(1) 93.1(5), O(1)-C(1)-N(1) 117.2(8), C(1)-N(1)-Nd(1) 97.4(5), N(1)-Nd(1)-O(1) 51.7(2), Nd(1)-O(2)-C(11) 94.1(5), O(2)-C(11)-N(2) 117.5(7), C(11)-N(2)-Nd(1) 93.5(5), N(2)-Nd(1)-O(2) 54.4(2), O(1A)-Nd(1)-N(2) 144.7(2), N(3)-Nd(1)-O(1) 124.8(3), O(1)-Nd(1)-O(1A) 68.9(2), O(1)-Nd(1)-O(2) 115.6(2).

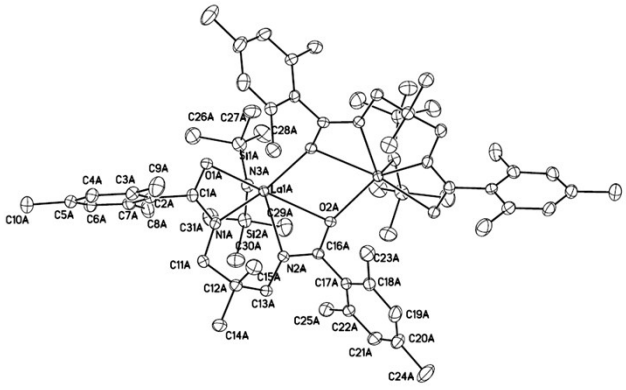


Fig. 2 Representative molecular structure of complex **5**·2THF**.** Hydrogen atoms, and THF molecules are omitted for clarity. Bond lengths (Å): La(1A)-O(1A) 2.471(3), La(1A)-

O(2A) 2.706(3), La(1A)-O(2) 2.426(3), La(1A)-N(1A) 2.492(3), La(1A)-N(2A) 2.579(3), La(1A)-N(3A) 2.360(4), O(1A)-C(1A) 1.296(5), O(2A)-C(16A) 1.320(5), N(1A)-C(1A) 1.298(5), N(1)-C(11) 1.456(5), N(2A)-C(16A) 1.462(5), N(2A)-C(16A) 1.288(5); Angles (°): O(2)-La(1)-O(2A) 67.0(1), O(1A)-La(1A)-N(1A) 53.0(1), O(2A)-La(1A)-N(2A) 49.5(1), N(1A)-La(1A)-N(2A) 67.6(1), O(1A)-C(1A)-N(1A) 117.3(4), O(2A)-C(16A)-N(2A) 116.3(4).

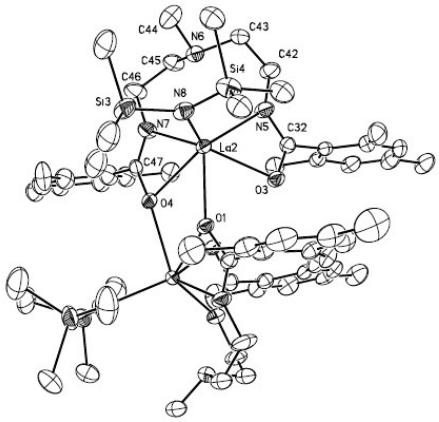


Fig. 3 Molecular structure of one independent molecule of complex **7**·2toluene**.** Hydrogen atoms and toluene molecules are omitted for clarity. Bond lengths (Å): La(2)-O(1) 2.467(3), La(2)-O(3) 2.507(3), La(2)-O(4) 2.761(3), La(2)-N(5) 2.498(4), La(2)-N(7) 2.524(4), La(2)-N(8) 2.397(4), O(3)-C(32) 1.288(6), O(4)-C(47) 1.301(5), N(5)-C(32) 1.303(6), N(7)-C(47) 1.285(6); Angles (°): O(1)-La(2)-O(4) 66.89(10), N(5)-La(2)-O(3) 52.08(12), N(7)-La(2)-O(4) 48.90(12), O(3)-C(32)-N(5) 116.0(4), N(7)-C(47)-O(4) 116.4(4).

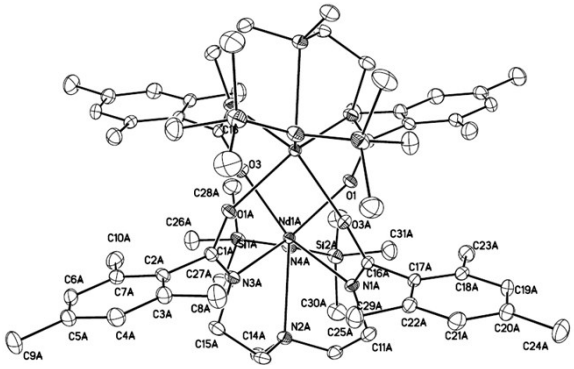


Fig. 4 Molecular structure of complex **8**·2toluene**.** Hydrogen atoms and toluene molecules are omitted for clarity. Bond lengths (Å): Nd(1A)-O(1) 2.336(2), Nd(1A)-O(3) 2.356(2), Nd(1A)-N(1A) 2.435(2), Nd(1A)-N(2A) 2.819(2), Nd(1A)-N(3A) 2.557(2), Nd(1A)-N(4A) 2.394(2), N(1A)-C(1A) 1.307(3), N(3A)-C(16A) 1.294(3), O(1A)-C(1A) 1.283(3), O(3A)-C(16A) 1.299(3); Angles (°): O(1A)-Nd(1A)-O(3A) 82.37(6), O(1A)-Nd(1A)-N(1A) 136.38(6), N(3A)-Nd(1A)-N(1A) 82.48(7), O(1A)-C(1A)-N(1A) 121.70(2).

Spectral data for the amides:

***N*-Benzyl-*N*-methylbenzamide (11a):** ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.51-7.15 (m, 10H, ArH), 4.79 (s, 1H, CH_2), 4.54 (s, 1H, CH_2), 2.97 (3H, CH_3) ppm. HRMS (ESI, m/z) calcd for $C_{15}H_{16}NONa^+$: 248.1051, found: 248.1052.

***N*-Benzyl-4-methoxy-*N*-methylbenzamide (11b):** ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.44-6.79 (m, 9H, ArH), 4.67 (s, 1H, CH_2), 4.57 (s, 1H, CH_2), 3.78 (s, 3H, CH_3O), 2.93 (s, 3H, CH_3) ppm. HRMS (ESI, m/z) calcd for $C_{16}H_{17}NO_2Na^+$: 278.1157, found: 278.1159.

***N*-Benzyl-*N*-methyl-4-fluorobenzamide (11c):** ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.50-7.02 (m, 9H, ArH), 4.72 (s, 1H, CH_2), 4.50 (s, 1H, CH_2), 3.0 (s, 1.5H, CH_3), 2.85 (s, 1.5H, CH_3) ppm. HRMS (ESI, m/z) calcd for $C_{15}H_{15}FNONa^+$: 266.0957, found: 266.0956.

***N*-Benzyl-*N*,4-dimethylbenzamide (11d):** ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.40-7.20 (m, 9H, ArH), 4.77 (s,1H, CH_2), 4.55 (s, 1H, CH_2), 3.00 (s, 1.5H, $ArCH_3$), 2.89 (s, 1.5H, $ArCH_3$), 2.37 (s, 3H, NCH_3) ppm. HRMS (ESI, m/z) calcd for $C_{16}H_{17}NONa^+$: 262.1208, found: 262.1208.

Morpholino(phenyl)methanone (11e): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.41-7.35 (m, 5H, ArH), 3.73-3.43 (m, 8H, CH_2) ppm. HRMS (ESI, m/z) calcd for $C_{11}H_{13}NO_2Na^+$: 214.0844, found: 214.0839.

Morpholino(*p*-tolyl)methanone (11f): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.31-7.19(m, 4H, ArH), 3.68(s, 8H, CH_2), 2.37(s, 3H, CH_3) ppm. HRMS (ESI, m/z) calcd for $C_{12}H_{15}NO_2Na^+$: 228.1000, found: 228.1007.

(4-Fluorophenyl)(morpholino)methanone (11g): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.38

(ddd, *J* = 8.5, 5.3, 1.6 Hz, 2H, ArH), 7.06 (td, *J*= 8.6, 1.7 Hz, 2H, ArH), 3.66-3.49(m, 3H, CH_2) ppm. HRMS (ESI, m/z) calcd for $C_{11}H_{12}FNO_2Na^+$: 232.0750, found: 232.0740.

(4-Chlorophenyl)(morpholino)methanone (11h): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.37-7.30 (m, 4H, ArH), , 3.66-3.42 (m, 8H,CH_2) ppm. HRMS (ESI, m/z) calcd for $C_{11}H_{12}ClNO_2H^+$: 226.0635, found: 226.0631.

(3-Chlorophenyl)(morpholino)methanone (11i): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.38-7.21 (m, 4H, ArH), 3.76-3.32 (m, 8H, CH_2) ppm. HRMS (ESI, m/z) calcd for $C_{11}H_{12}ClNO_2H^+$: 226.0635, found: 226.0639.

(2-Chlorophenyl)(morpholino)methanone (11j): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.40-7.25 (m, 4H, ArH), 3.89-3.72 (m, 4H, CH_2), 3.70-3.53 (m, 2H, CH_2), 3.30-3.15 (m, 2H, CH_2) ppm. HRMS (ESI, m/z) calcd for$C_{11}H_{12}ClNO_2H^+$: 226.0635, found: 226.0648.

Morpholino(4-nitrophenyl)methanone (11k):^{1}H NMR (400 MHz, $CDCl_3$)：δ 8.29-8.24 (m, 2H, ArH), 7.58-7.54 (m, 2H, ArH), 3.83-3.31 (m, 8H, CH_2) ppm. HRMS (ESI, m/z) calcd for $C_{11}H_{12}N_2O_4H^+$: 237.0875, found: 237.0873.

***N*-Benzoylpyrrolidine (11l):** ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.51 -7.35 (m, 5H, ArH), 3.62 (t, *J* = 7.0 Hz, 2H, CH_2-N), 3.39 (t, *J* = 6.6 Hz, 2H, CH_2-N), 1.97-1.90 (m, 2H, CH_2), 1.88-1.80 (m, 2H, CH_2) ppm. HRMS (ESI, m/z) calcd for $C_{11}H_{13}NONa^+$: 198.0895, found: 198.0897.

Phenyl(piperidin-1-yl)methanone (11m): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.37 (m, 5H, ArH), 3.70 (s, 2H, CH_2-N), 3.32 (s, 2H, CH_2-N),1.66-1.50 (m, 6H, CH_2) ppm. HRMS (ESI, m/z) calcd for $C_{12}H_{15}NONa^+$: 212.1051, found: 212.1055.

***N*-Phenylbenzamide (11n):** ^{1}H NMR (400 MHz, $CDCl_3$)：δ 8.02-7.06 (m, 10H, ArH) ppm. HRMS (ESI, m/z) calcd for $C_{13}H_{11}NONa^+$: 220.0738, found: 220.0728.

Indolin-1-yl(phenyl)methanone (11o): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.67-6.92 (m, 9H, ArH), 4.06 (s, 2H, CH_2), 3.10 (t, J = 8.3 Hz, 2H, CH_2) ppm. HRMS (ESI, m/z) calcd for $C_{15}H_{13}NONa^+$: 246.0895, found: 246.0889.

(2-Methylindolin-1-yl)(phenyl)methanone (11p): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.61-6.89 (m, 9H, ArH), 4.76 (s, 1H, CH), 3.43-2.65 (m, 2H, CH_2), 1.26 (s, 3H, CH_3) ppm. HRMS (ESI, m/z) calcd for $C_{16}H_{15}NONa^+$: 260.1051, found: 260.1044.

7-Methylindolin-1-yl(*p*-tolyl)methanone (11q): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.44-7.00 (m, 8H, ArH), 4.05 (s, 2H, CH_2), 3.04 (t, J = 8.2 Hz, 2H, CH_3), 2.39 (m, 2H, CH_3), 2.28 (s, 3H, CH_3) ppm. HRMS (ESI, m/z) calcd for $C_{17}H_{17}NONa^+$: 274.1208, found: 274.1210.

Piperazine-1,4-diylbis(phenylmethanone) (11r): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.37 (s, 10H, ArH), 3.60 (m, 8H, CH_2). HRMS (ESI, m/z) calcd for $C_{18}H_{18}N_2O_2Na^+$: 317.1266, found: 317.1259.

Piperazine-1,4-diylbis(*p*-tolylmethanone) (11s): ^{1}H NMR (400 MHz, DMSO)：δ 7.24-7.32 (m, 8H, ArH), 3.51 (m, 8H, CH_2), 2.33 (s, 6H, CH_3). HRMS (ESI, m/z) calcd for $C_{20}H_{22}N_2O_2Na^+$: 345.1579, found: 345.1576.

Piperazine-1,4-diylbis((4-chlorophenyl)methanone) (11t): ^{1}H NMR (400 MHz, DMSO)：δ 7.45-7.53 (m, 8H, ArH), 3.53 (m, 8H, CH_2). HRMS (ESI, m/z) calcd for $C_{18}H_{16}Cl_2N_2O_2H^+$: 363.0667, found: 363.0660.

Spectral data for the guanidines:

1,3-Diisopropyl-2-phenylguanidine (13a): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.25-6.83 (m, 5H, ArH), 3.75 (s, 2H, CH), 3.57 (s, 2H, NH), 1.14 (d, J = 6.3 Hz, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{13}H_{21}N_3H^+$: 220.1814, found: 220.1819.

2-(4-Fluorophenyl)-1,3-diisopropylguanidine (13b): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 6.94-6.74 (m, 4H, ArH), 3.73 (s, 2H, CH), 3.50 (s, 2H, NH), 1.14 (d, J = 6.3 Hz, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{13}H_{20}FN_3H^+$: 238.1720, found: 237.1716.

2-(4-Chlorophenyl)-1,3-diisopropylguanidine (13c): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.17 (d, J = 8.5 Hz, 2H, ArH), 6.75 (d, J = 8.5 Hz, 2H, ArH), 3.76-3.68 (m, 2H, CH), 3.52 (s, 1H, NH), 1.13 (d, J = 6.3 Hz, 12H, CH_3).HRMS (ESI): m/z calcd for $C_{13}H_{20}ClN_3H^+$: 254.1424, found: 254.1426.

2-(4-Bromophenyl)-1,3-diisopropylguanidine (13d): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.31 (d, J = 8.4 Hz, 2H, ArH), 6.71 (d, J= 8.4 Hz, 2H, ArH), 4.14-3.58 (m, 2H, CH), 3.52 (s, 2H, NH), 1.14 (d, J = 6.3 Hz, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{13}H_{20}BrN_3H^+$: 298.0919, found:298.0931.

1,3-Diisopropyl-2-(4-methoxyphenyl)guanidine (13e): ^{1}H NMR (400 MHz,)：δ 6.78 (q, J = 9.0 Hz, 4H, ArH), 3.66 (m, 5H, OCH_3, CH), 3.56 (s, 2H, NH), 1.14 (d, J = 6.1 Hz, CH_3).HRMS (ESI) : m/z: calcd for $C_{14}H_{23}N_3OH^+$:250.1919, found: 250.1930.

1,3-Diisopropyl-2-(*p*-tolyl)guanidine (13f): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.03 (d, J = 7.6 Hz, 2H, ArH), 6.72 (d, J = 7.7 Hz, 2H, ArH), 3.74 (s, 2H, CH), 3.53 (s, 2H, NH), 2.26 (s, 3H, $ArCH_3$), 1.14 (d, J = 6.1 Hz, 12H, CH_3). HRMS (ESI) : m/z: calcd for $C_{14}H_{23}N_3H^+$

:234.1970, found: 234.1972.

1,3-Diisopropyl-2-(*m*-tolyl)guanidine (13g): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.12-6.62 (m, 4H, ArH), 3.75-3.74 (m, 2H, CH), 3.55 (s, 2H, NH), 2.26 (s, 3H), 1.14 (d, J = 6.3 Hz, 12H, CH_3). HRMS (ESI) : m/z: calcd for $C_{14}H_{23}N_3H^+$:234.1970, found: 234.1982.

1,3-Diisopropyl-2-(*o*-tolyl)guanidine (13h): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.16 - 6.62 (m, 4H, ArH), 3.74 (s, 2H, CH), 2.12 (s, 3H, Ar-CH_3), 1.14 (d, J = 6.4 Hz, 12H, CH_3). HRMS (ESI): m/z: calcd for $C_{14}H_{23}N_3H^+$:234.1970, found: 234.1976.

1,3-Diisopropyl-2-(pyridin-2-yl)guanidine (13i): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 8.05-6.57 (m, 4H, ArH), 3.91 (m, 2H, CH), 1.22 (d, J = 6.4 Hz, 12H, CH_3). HRMS (ESI) : m/z: calcd for$C_{12}H_{20}N_4H^+$:221.1766, found: 221.1762.

1,3-Diisopropyl-2-(naphthalen-1-yl)guanidine (13j): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 8.33- 6.80 (m, 7H, ArH), 3.85 (dd, J = 11.4, 5.8 Hz, 2H, CH), 3.62 (s, 2H, NH), 1.16 (d, J = 6.1 Hz, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{17}H_{23}N_3H^+$: 270.1970, found: 270.1966.

2-(2-(*tert*-butyl)phenyl)-1,3-diisopropylguanidine (13k): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.88-5.57 (m, 4H, ArH), 3.81 (m, 2H, NH), 3.50 (d, J = 7.9 Hz, 2H, CH), 1.36 (d, J = 1.2 Hz, 9H, ^{t}Bu), 1.15 (d, J = 6.4 Hz, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{17}H_{29}N_3H^+$: 276.2440, found: 276.2449.

***N,N′′*-diisopropyl-*N′*-2,6-dimethylphenylguanidine (13l):** ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.09-6.70 (3 H, ArH), 3.31-3.09 (m, 2 H, CH), 2.11 (s, $ArCH_3$, 6 H), 1.15 (s, CH_3, 12 H). HRMS (ESI): m/z calcd for $C_{15}H_{25}N_3H^+$: 248.2127, found: 248.2127.

1,3-Diisopropyl-2-(4-nitrophenyl)guanidine (13m): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 8.09-6.86(m, 4H, ArH), 3.77 (s, 4H, CH, NH), 1.18 (s, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{13}H_{20}N_4O_2H^+$: 265.1665, found: 265.1674.

1,3-Diisopropyl-2-(4-(trifluoromethyl)phenyl)guanidine (13n): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.49-6.91(m, 4H, ArH), 3.81-3.73(m, 2H, CH), 3.60-3.59(m, 2H, NH), 1.18 (d, J = 6.4 Hz, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{14}H_{20}F_3N_3H^+$: 288.1688, found: 288.1696.

(*Z*)-*N,N*'-Diisopropylmorpholine-4-carboximidamide (13o): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 3.87-3.61 (m, 4H, CH_2), 3.57-3.42 (m, 2H, CH), 3.12-3.10 (m, 4H,CH_2), 1.13 (dd, J = 14.8, 6.3 Hz, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{11}H_{23}N_3OH^+$: 214.1919, found: 214.1914.

(*Z*)-*N,N'*-Diisopropylpyrrolidine-1-carboximidamide (13p): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 3.32 (dt, J = 12.6, 6.3 Hz, 2H, CH), 3.23-3.13 (m, 4H, CH_2), 1.83-1.63 (m, 4H, CH_2), 1.04 (d, J = 6.4 Hz, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{11}H_{23}N_3H^+$: 198.1970, found: 198.1979.

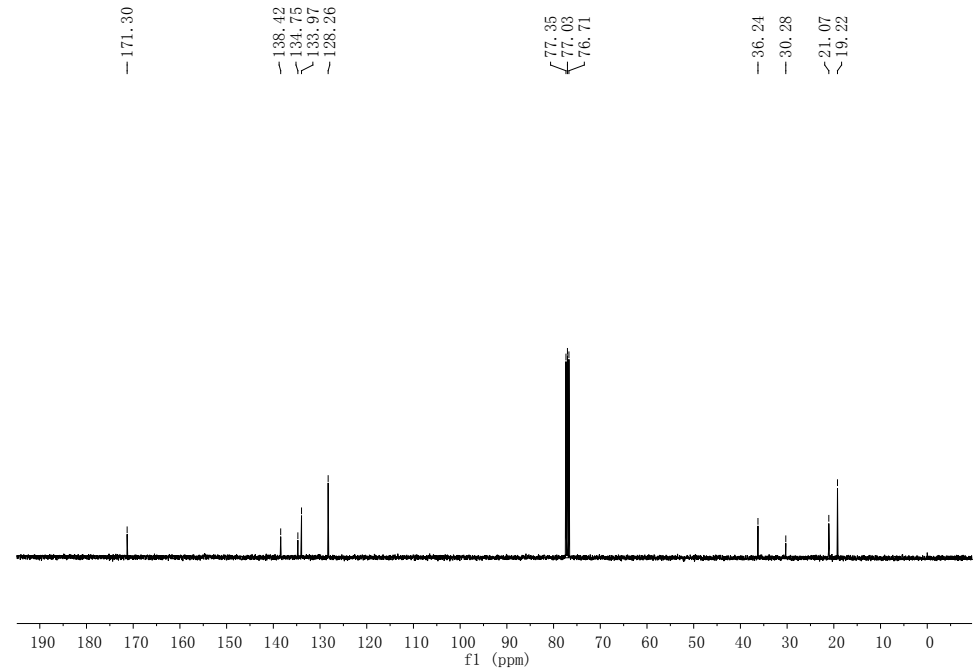
(*Z*)-*N,N'*-Diisopropylindoline-1-carboximidamide (13q): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.19-6.73 (m, 4H, ArH), 3.88-3.78 (m, 4H, CH_2), 3.45-3.44 (m, 2H, CH), 2.99-2.95 (m, 2H, CH_2), 1.15-1.13 (m, 12H, CH_3). HRMS (ESI): m/z calcd for $C_{11}H_{23}N_3H^+$: 198.1970, found: 198.1979.

(*Z*)-*N,N'*-Diisopropyl-6-methylindoline-1-carboximidamide (13r): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 7.06-6.88 (m, 3H, ArH), 4.36-2.69 (m, 6H, CH, CH_2), 2.25 (s, 3H, Ar-CH_3), 1.13 (s, 12H,CH_3). HRMS (ESI): m/z calcd for $C_{16}H_{25}N_3H^+$: 260.2127, found: 260.2134.

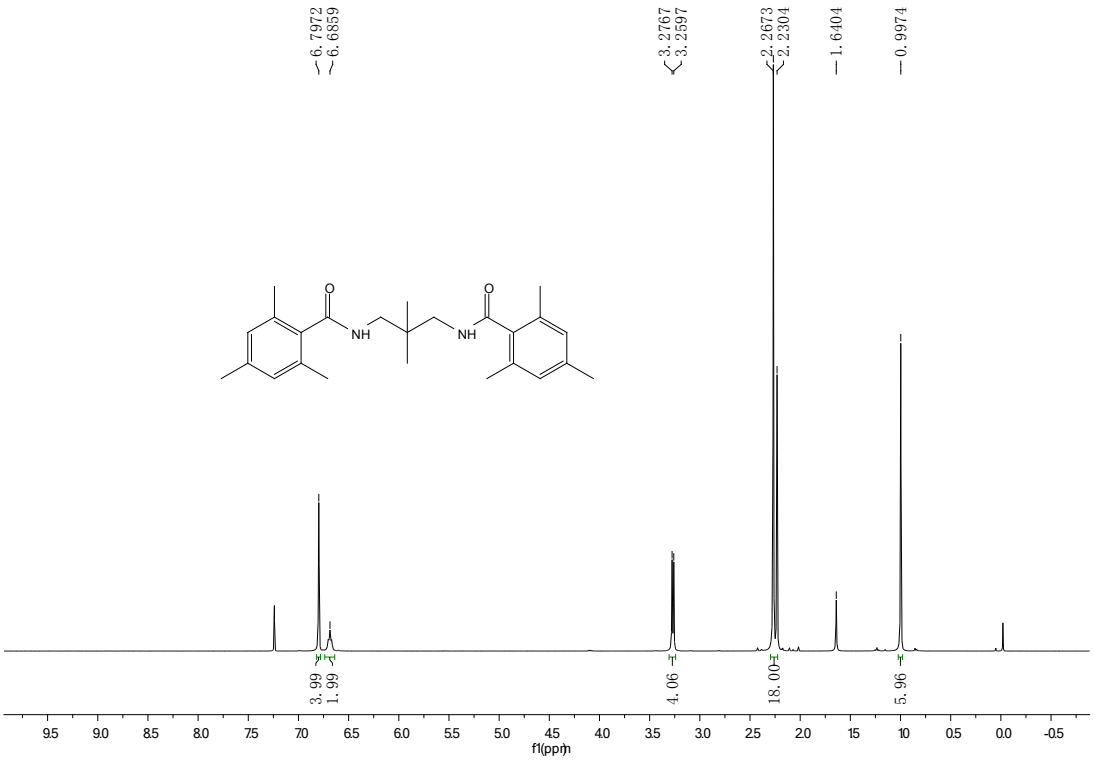
(1*Z*,4*Z*)-*N1,N'1,N4,N'4*-tetraisopropylpiperazine-1,4-bis(carboximidamide) (13s): ^{1}H NMR (400 MHz, $CDCl_3$)：δ 3.51-3.34 (m, 2H, CH), 3.31-3.17 (m, 1H, CH), 3.01 (s, 8H, CH_2), 1.03 (d, J = 5.5 Hz, 24H, CH_3).^{13}C NMR (100 MHz, $CDCl_3$)：δ 155.21, 47.78, 46.57, 45.63, 24.34, 23.17. HRMS (ESI): m/z calcd for $C_{18}H_{38}N_6H^+$: 339.3236, found: 339.3278.

The ^{1}H NMR of H_2L^1:

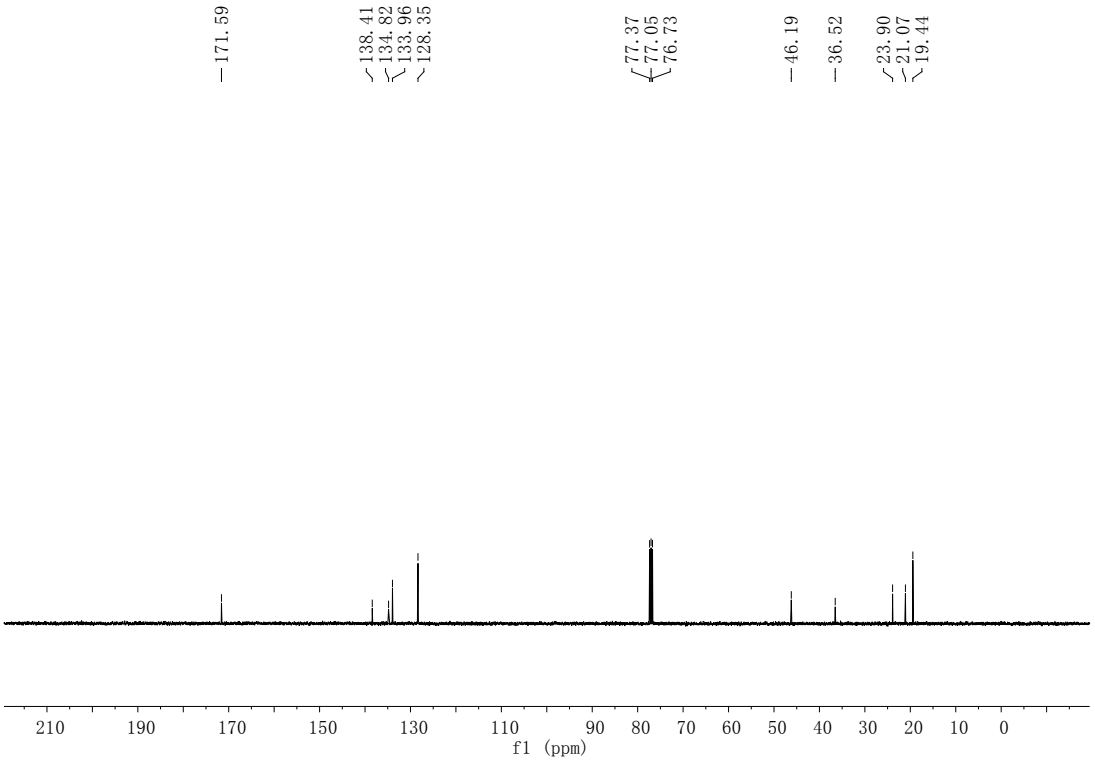
The ^{13}C NMR of H_2L^1:



The ^{1}H NMR of H_2L^2:

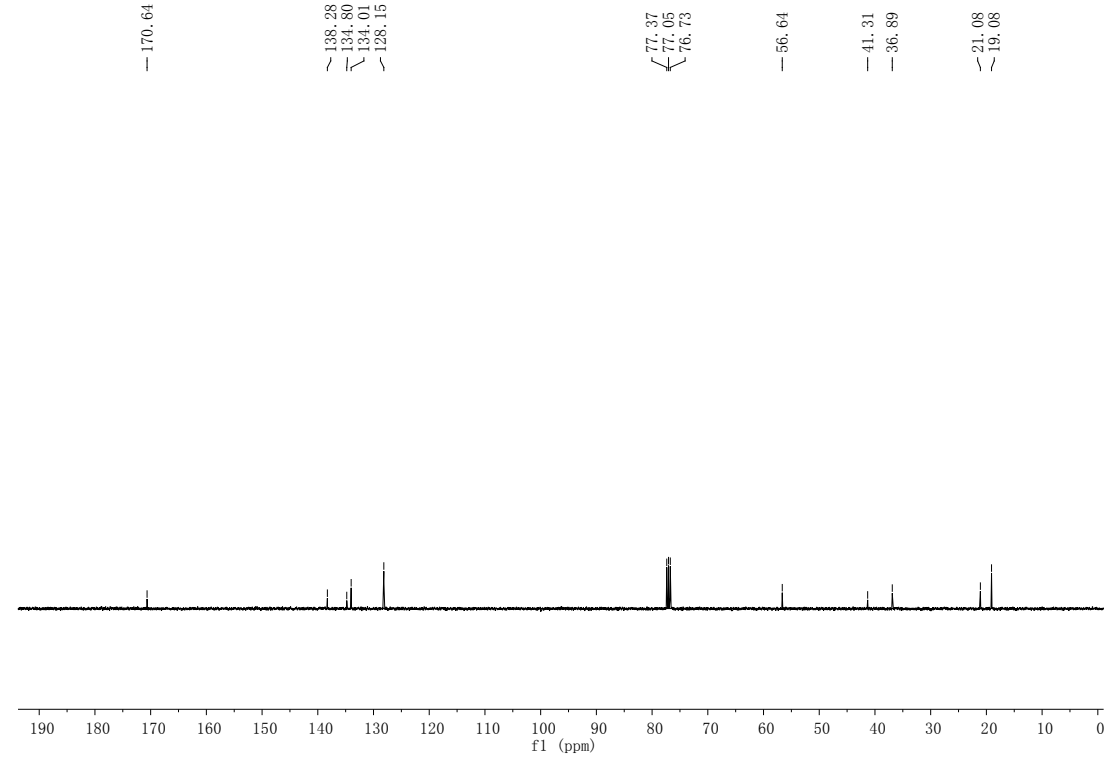


The ^{13}C NMR of H_2L^2:

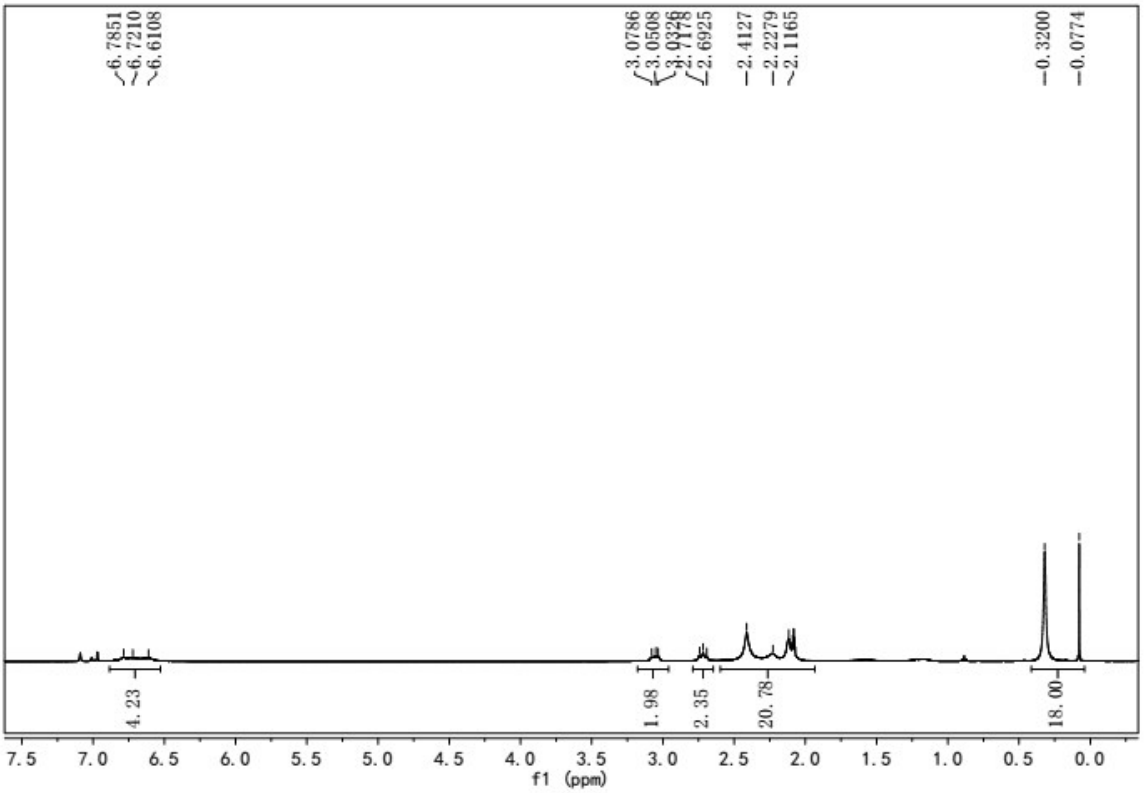


The ^{1}H NMR of H_2L^3:

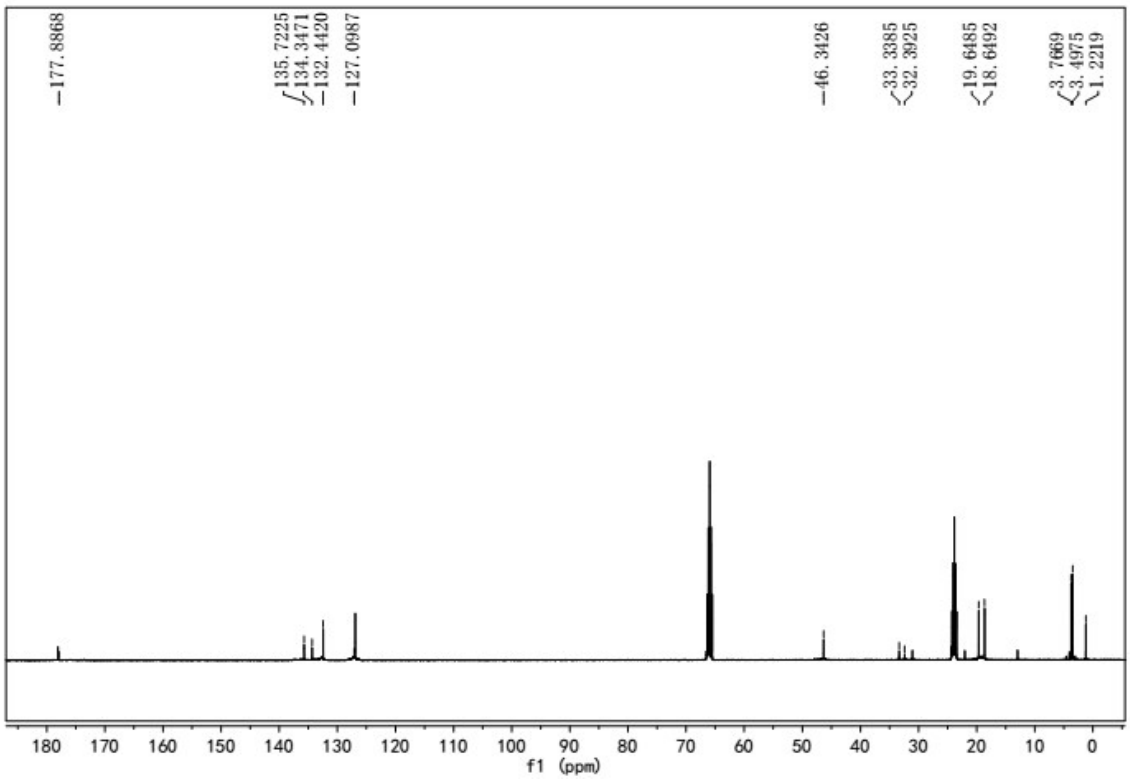
The ^{13}C NMR of H_2L^3:



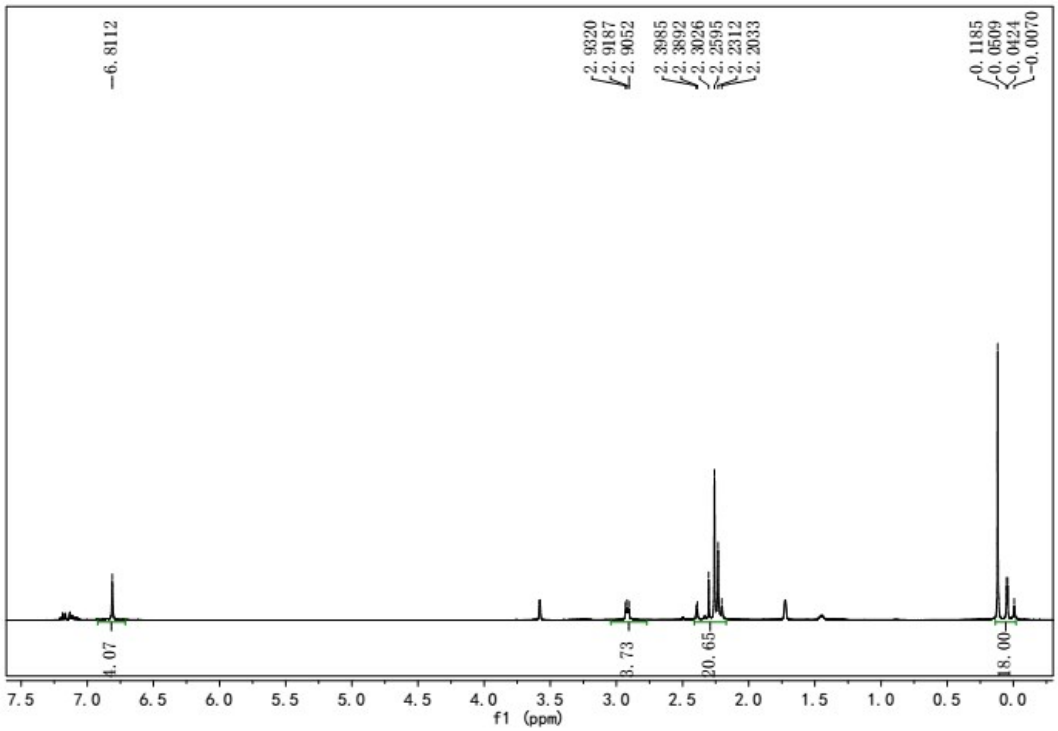
The ^{1}H NMR of complex **1:**



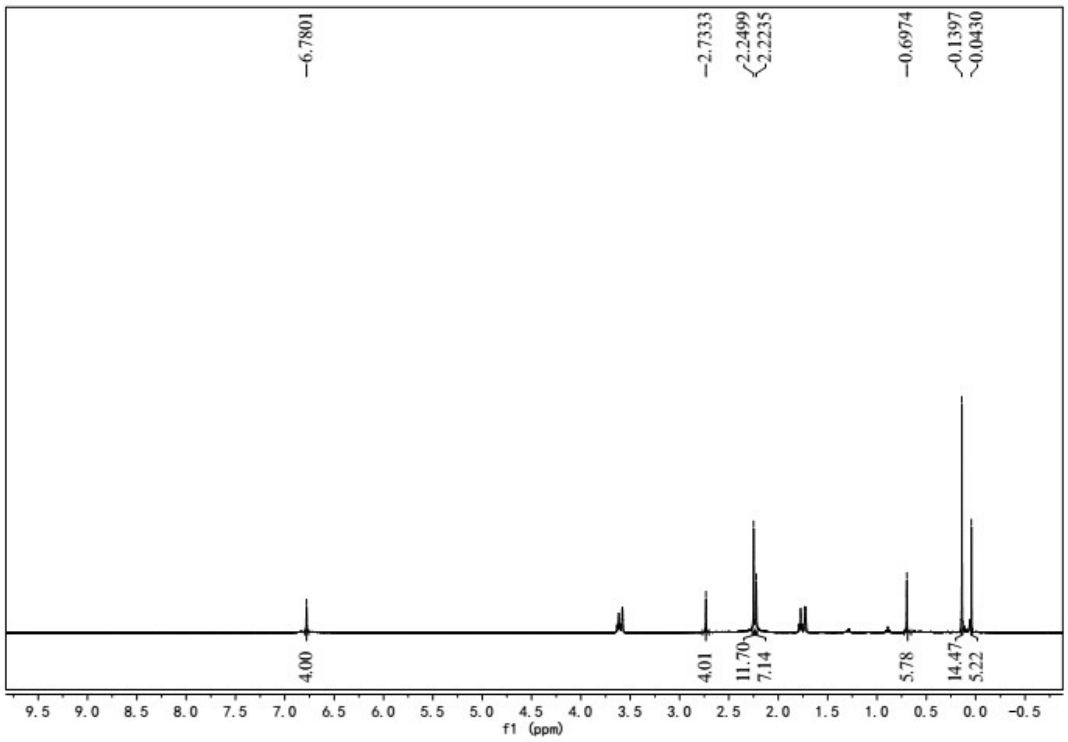
The ^{13}C NMR of complex **1:**



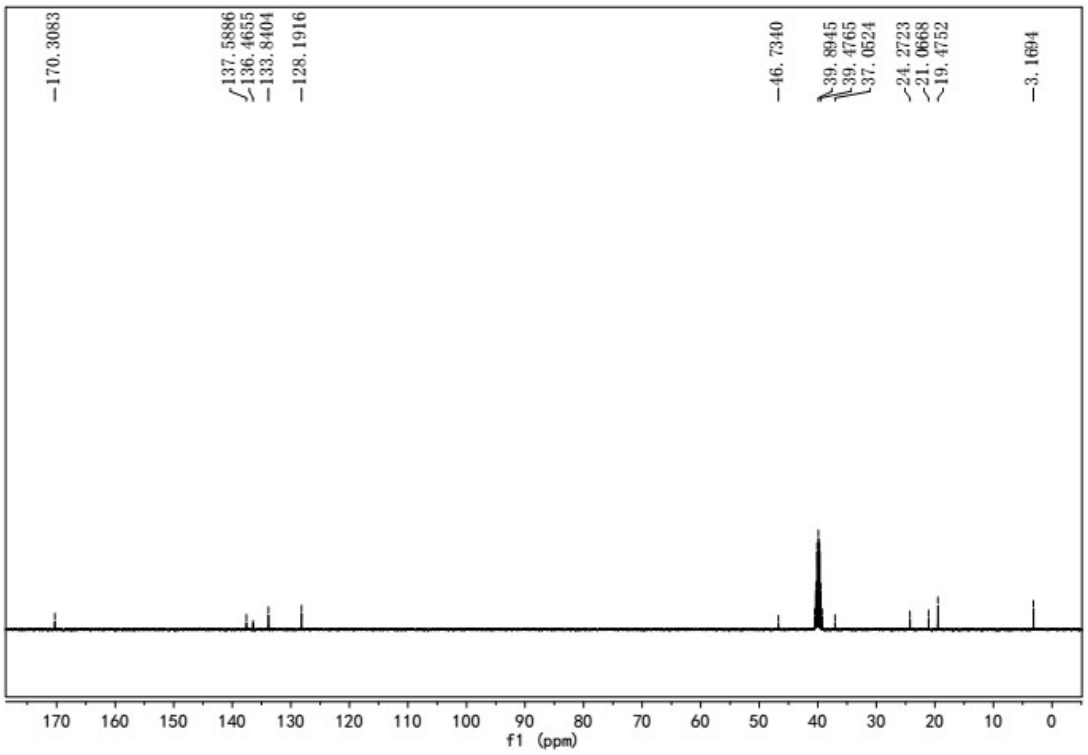
The ^{1}H NMR of complex **4:**



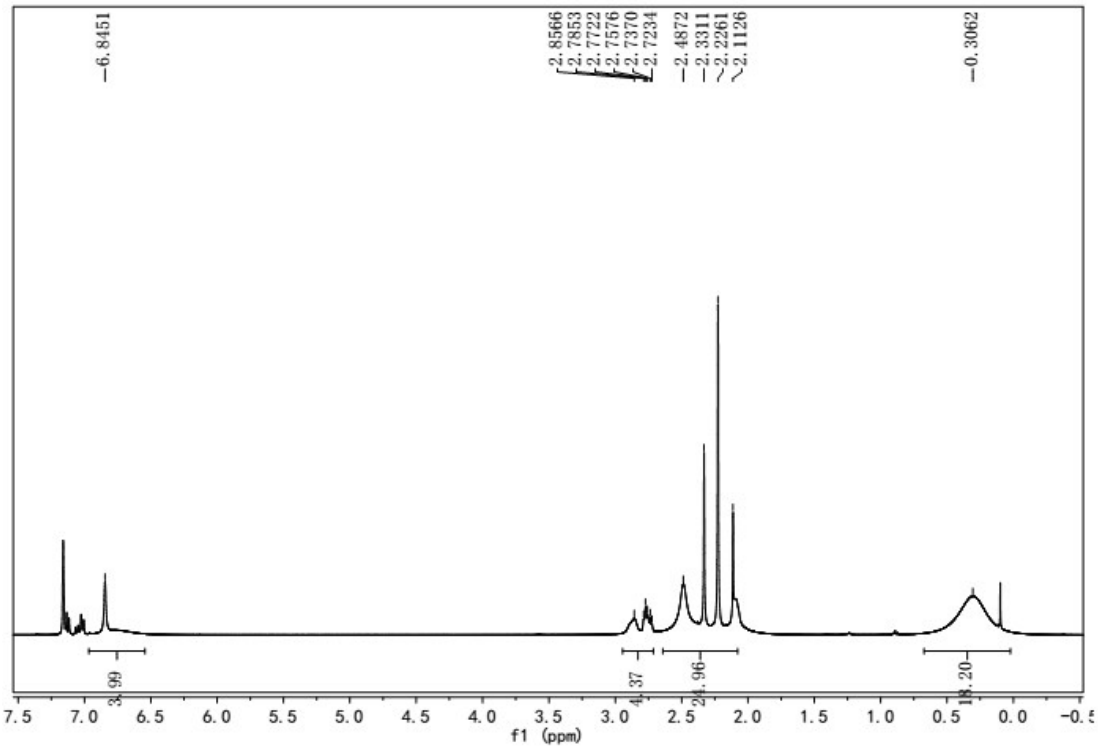
The ^{1}H NMR of complex **5:**



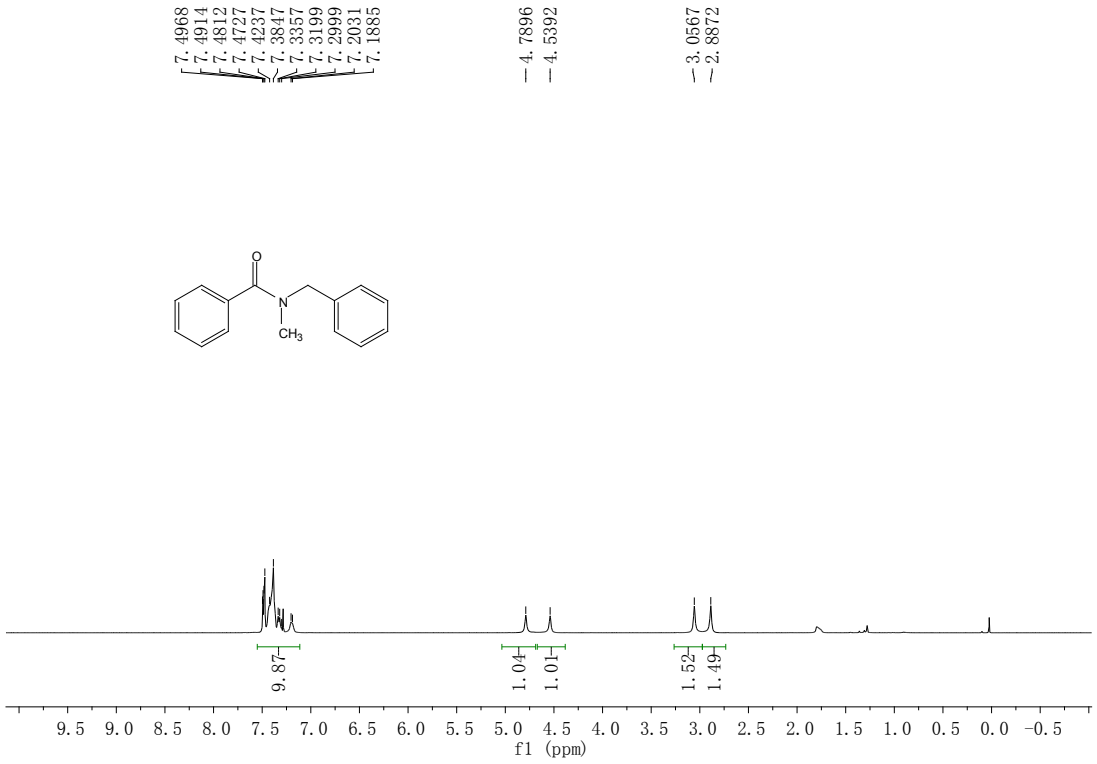
The ^{13}C NMR of complex **5:**



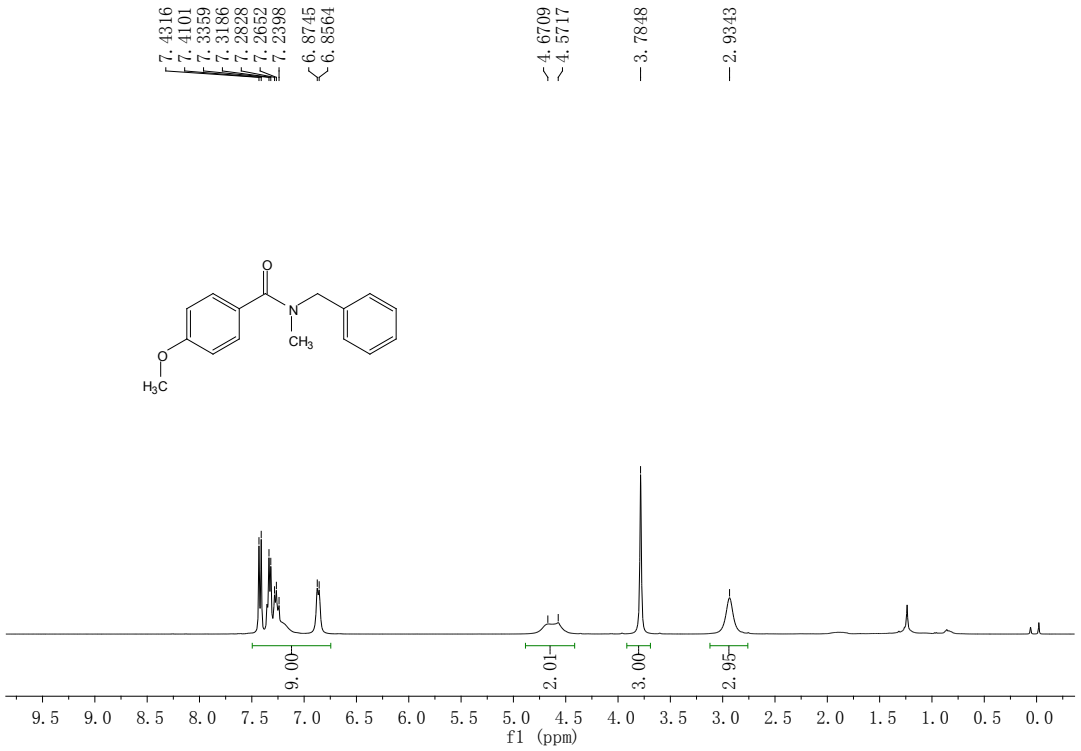
The ^{1}H NMR of complex **7:**



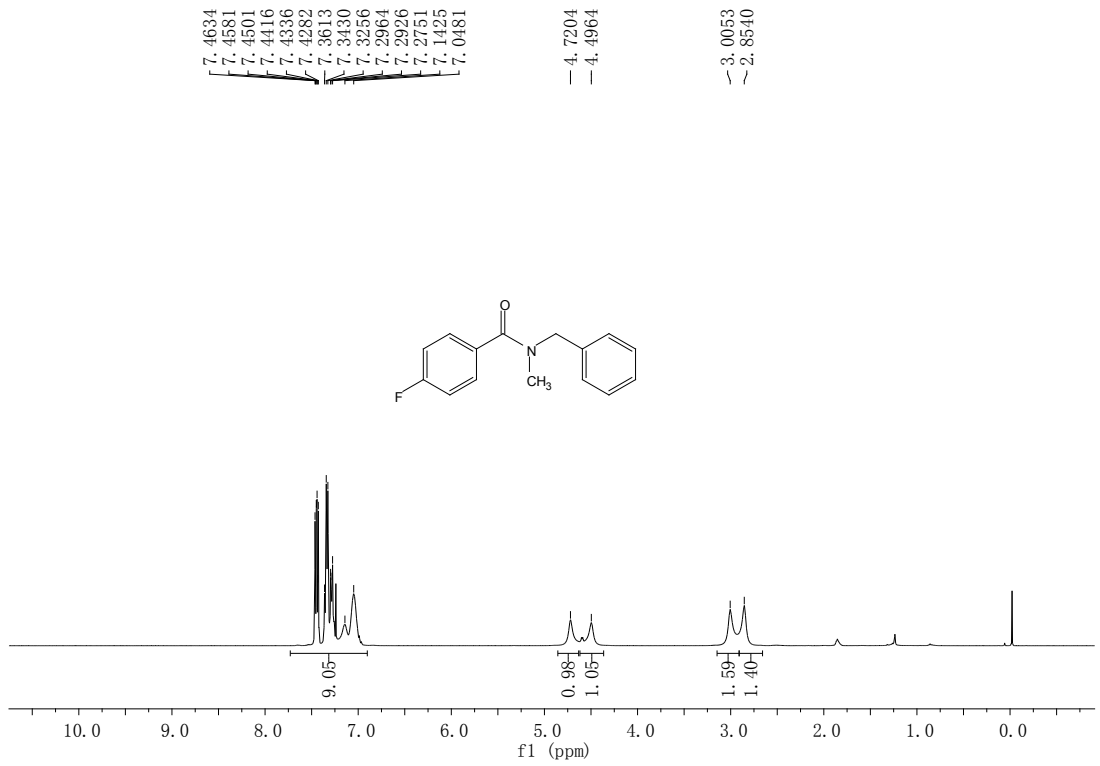
N-Benzyl-N-methylbenzamide (11a): ^{1}H NMR



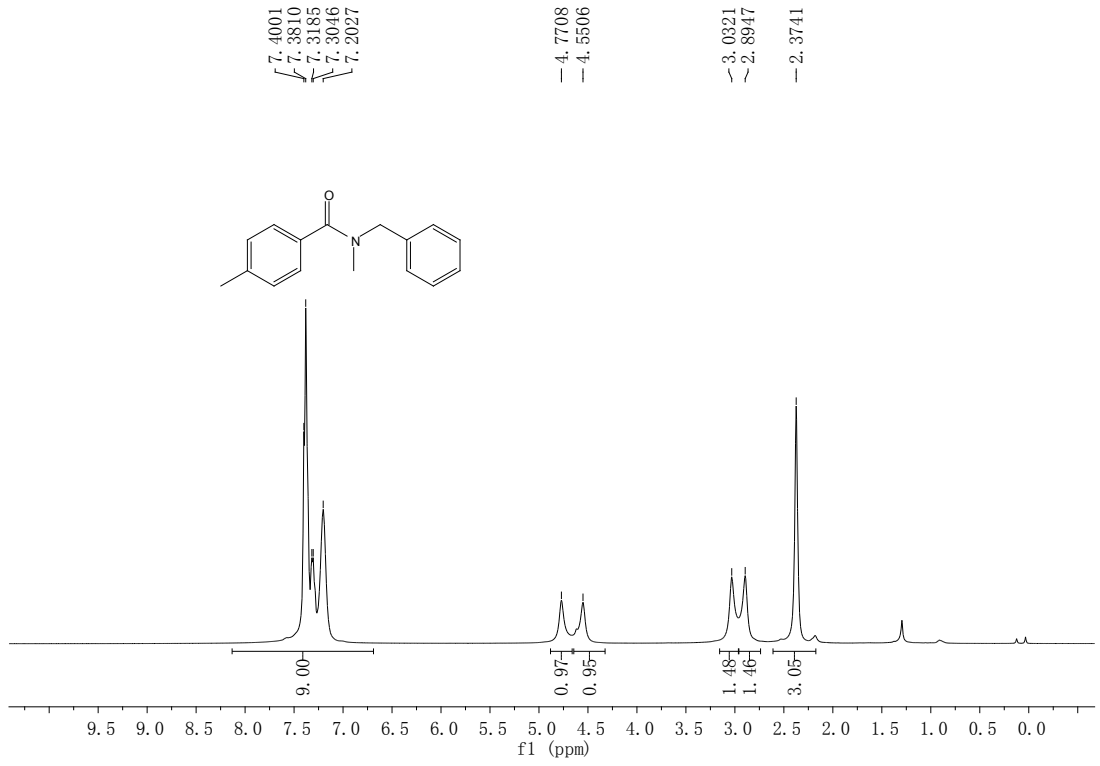
N-benzyl-4-methoxy-N-methylbenzamide (11b):



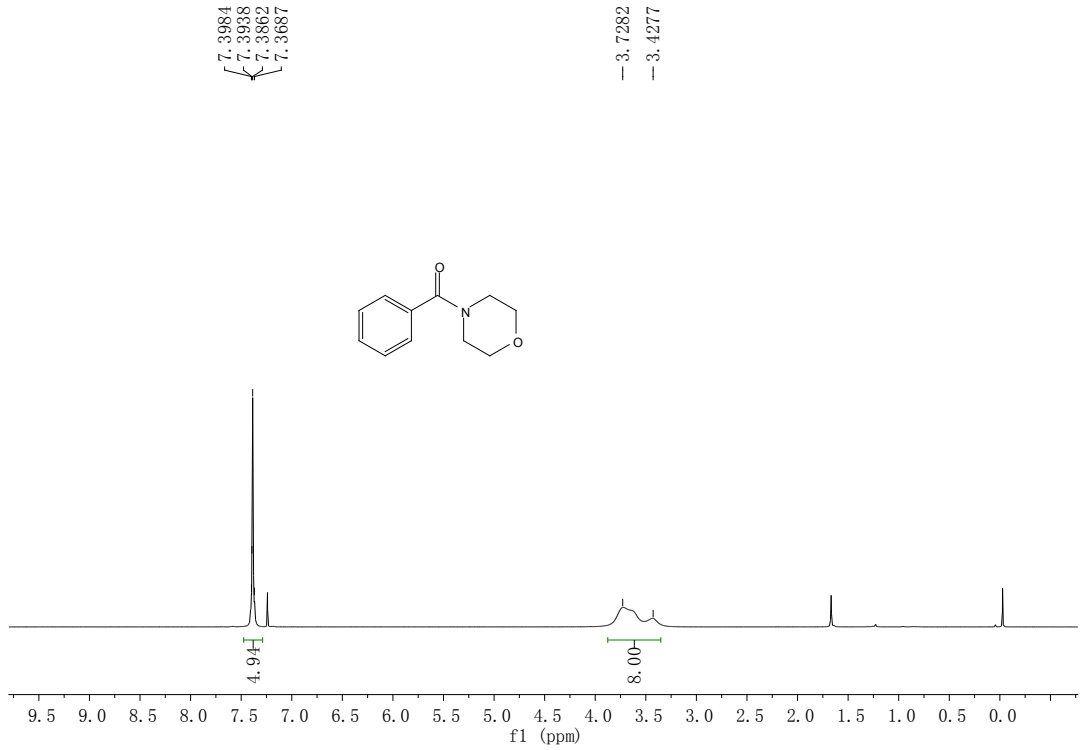
N-Benzyl-N-methyl-4-fluorobenzamide (11c):



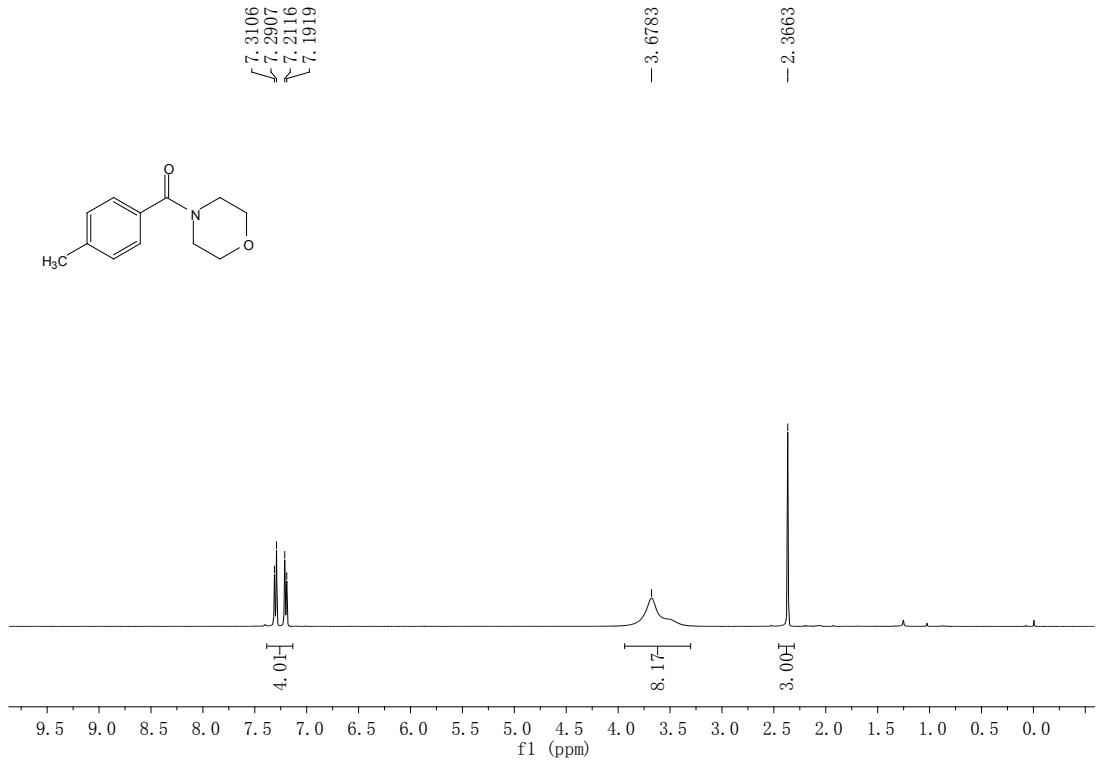
N-benzyl-N,4-dimethylbenzamide (11d):



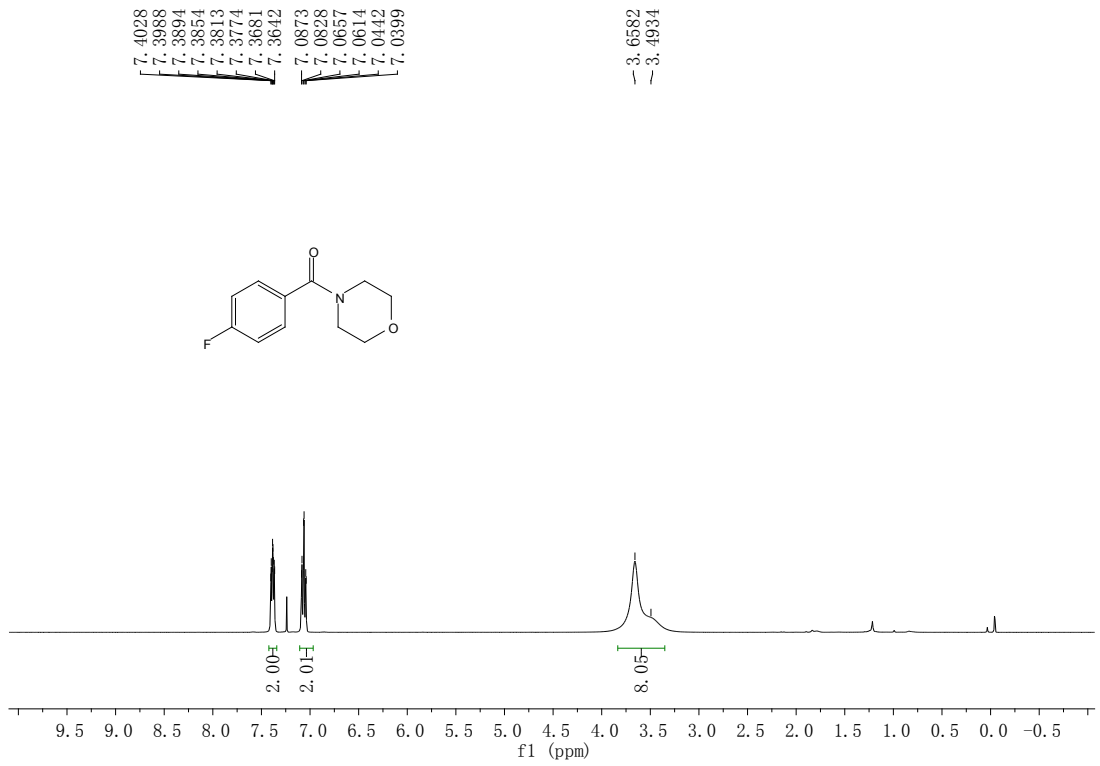
Morpholino(phenyl)methanone (11e):



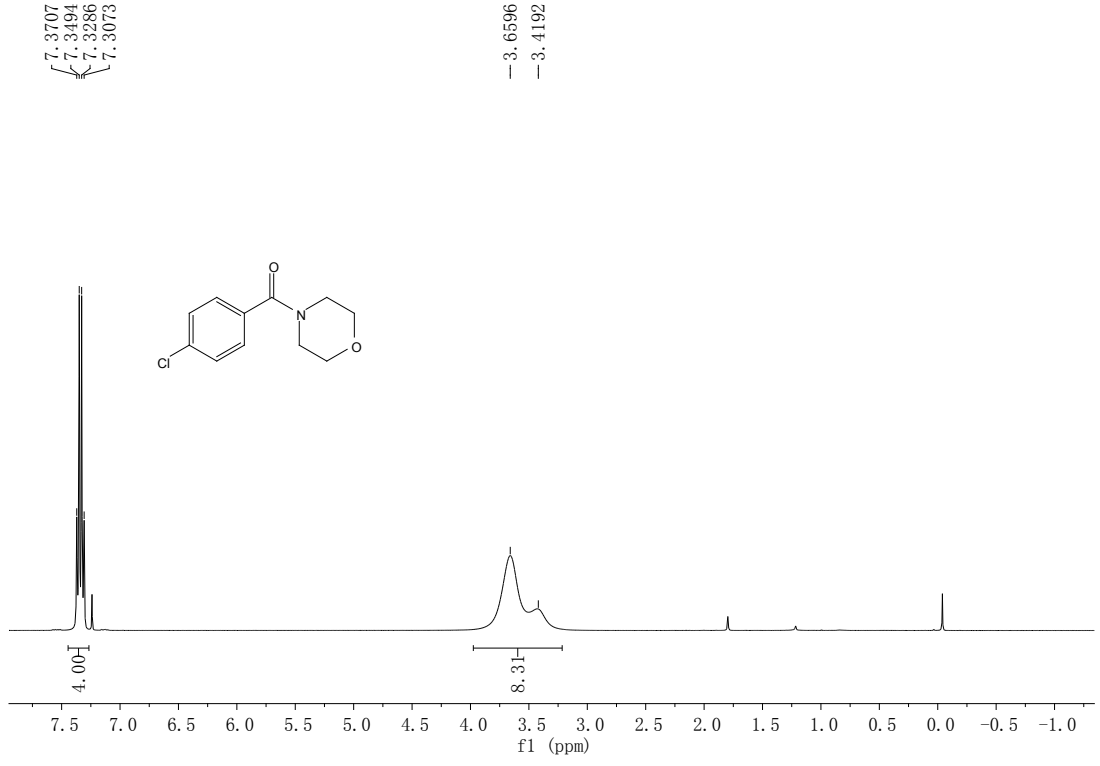
Morpholino(p-tolyl)methanone (11f):



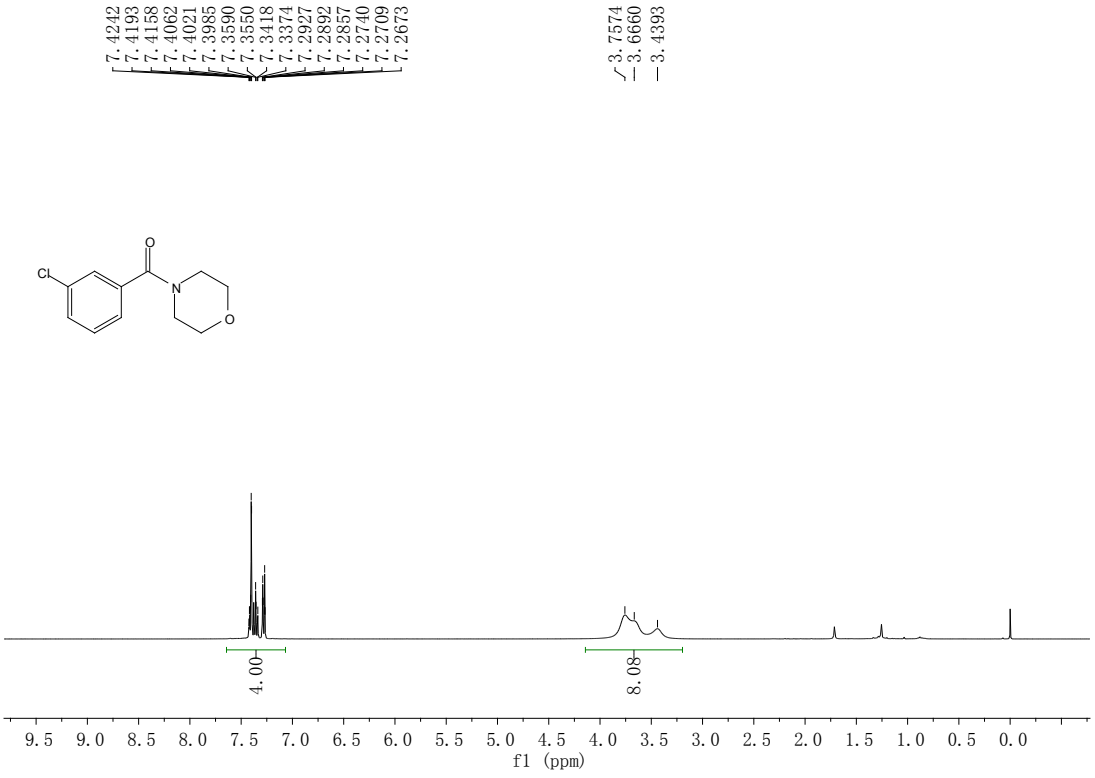
(4-Fluorophenyl)(morpholino)methanone (11g):



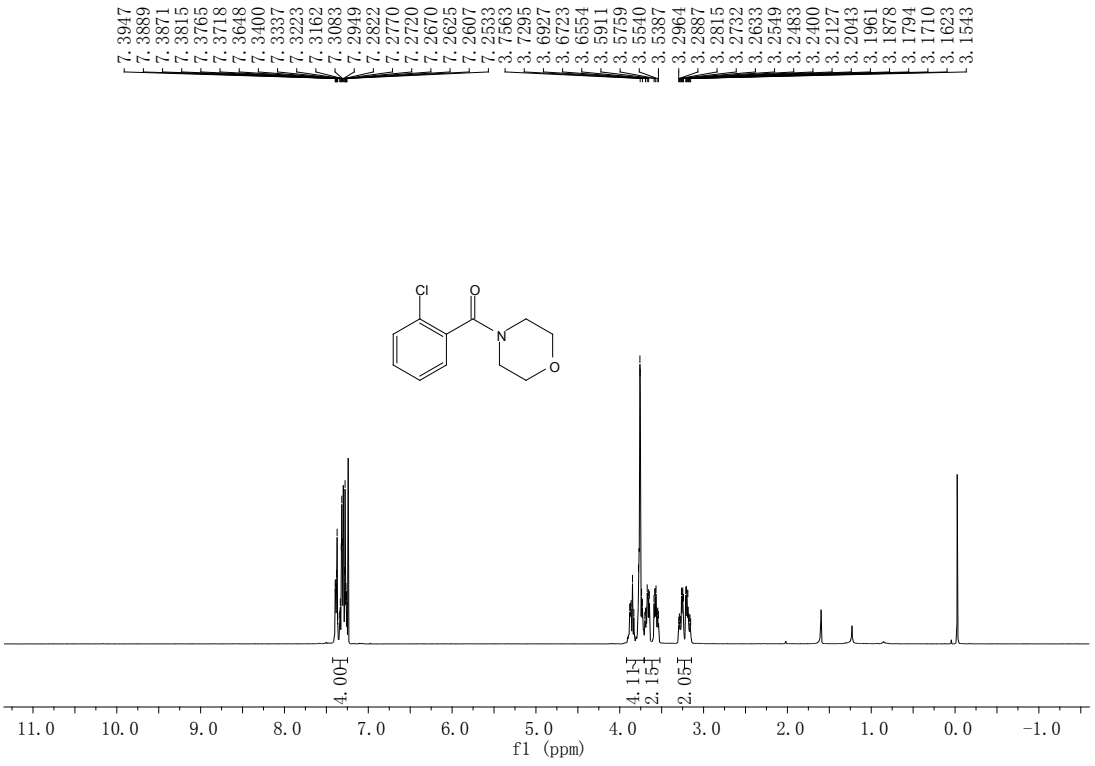
(4-chlorophenyl)(morpholino)methanone (11h):



(3-Chlorophenyl)(morpholino)methanone (11i):



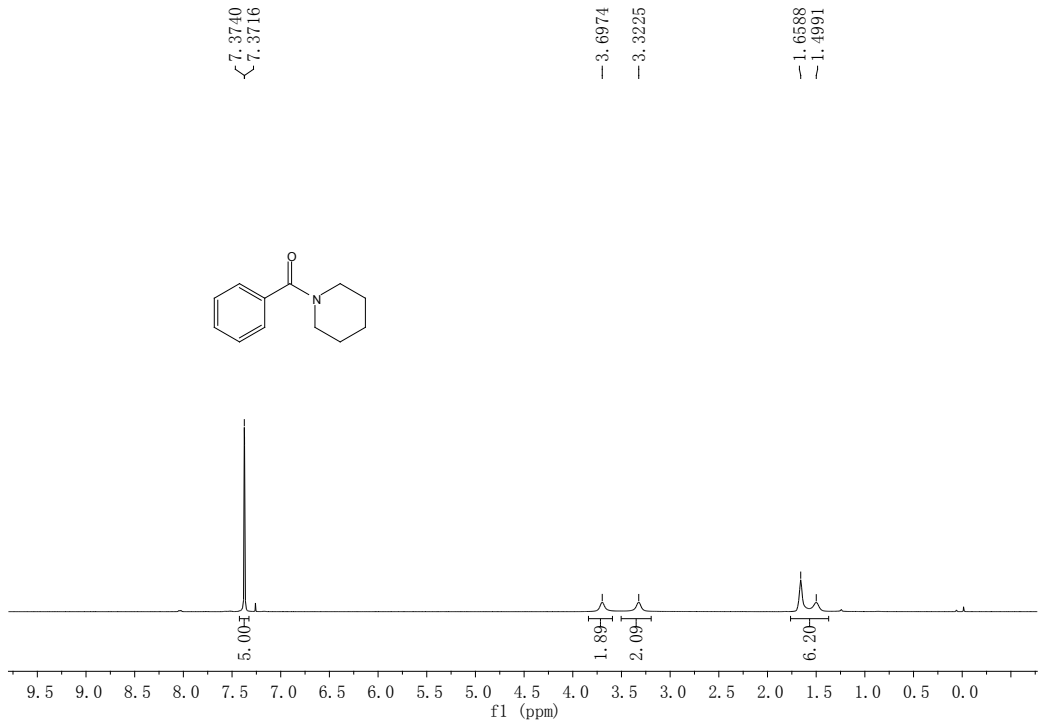
(2-Chlorophenyl)(morpholino)methanone (11j):



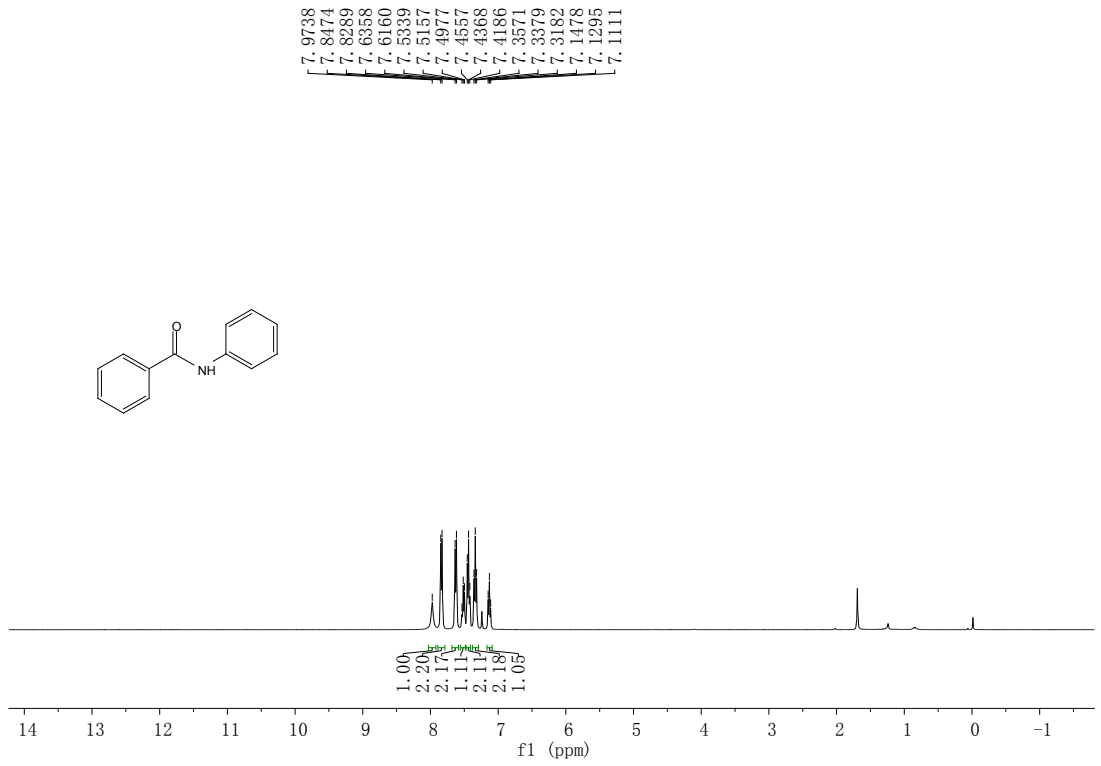
Morpholino(4-nitrophenyl)methanone (11k):

N-Benzoylpyrrolidine (11l):

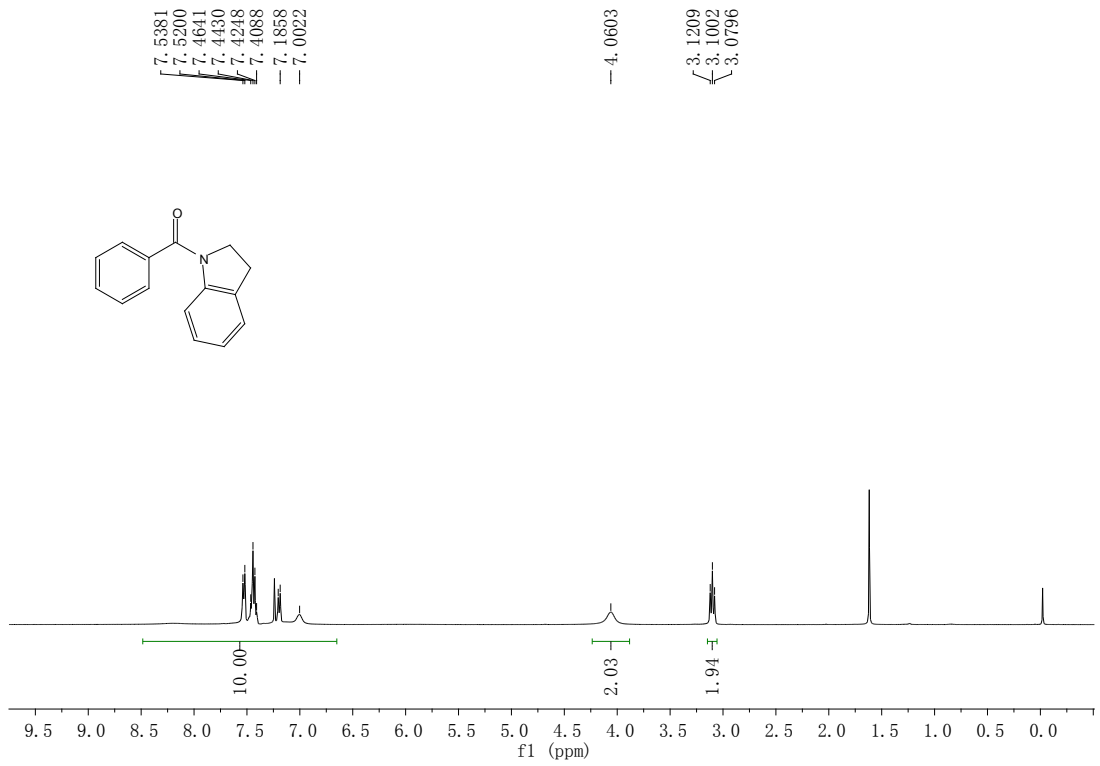
Phenyl(piperidin-1-yl)methanone (11m):



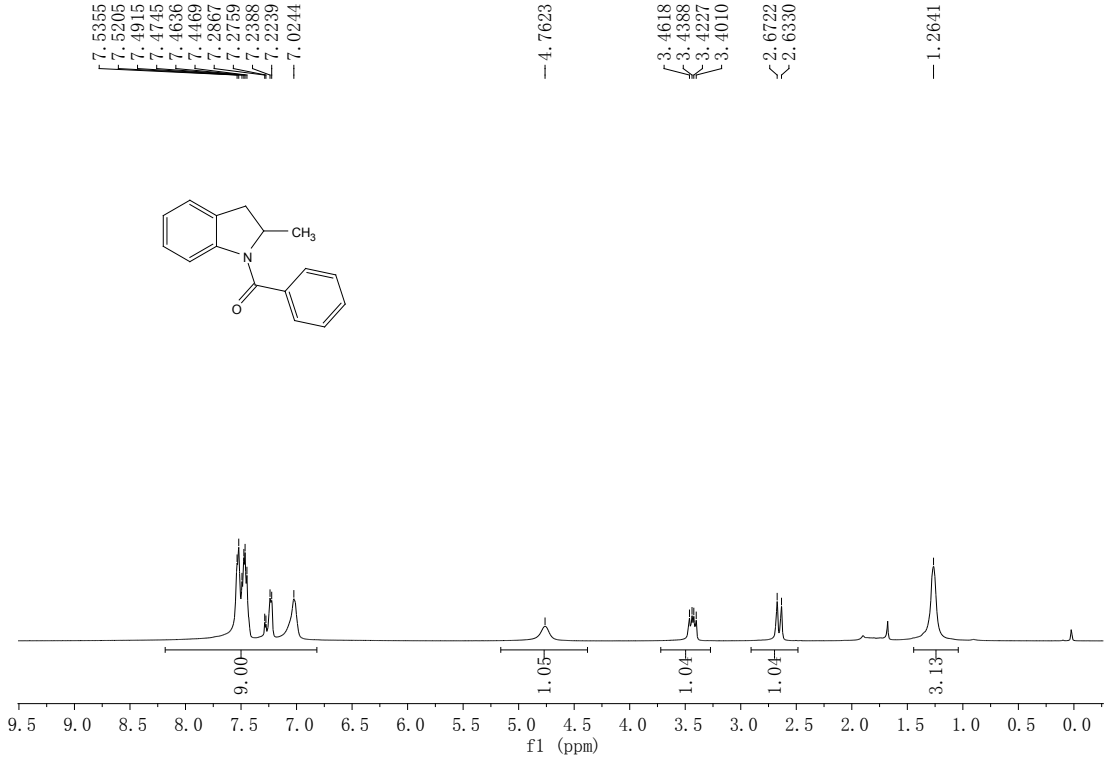
N-Phenylbenzamide (11n):



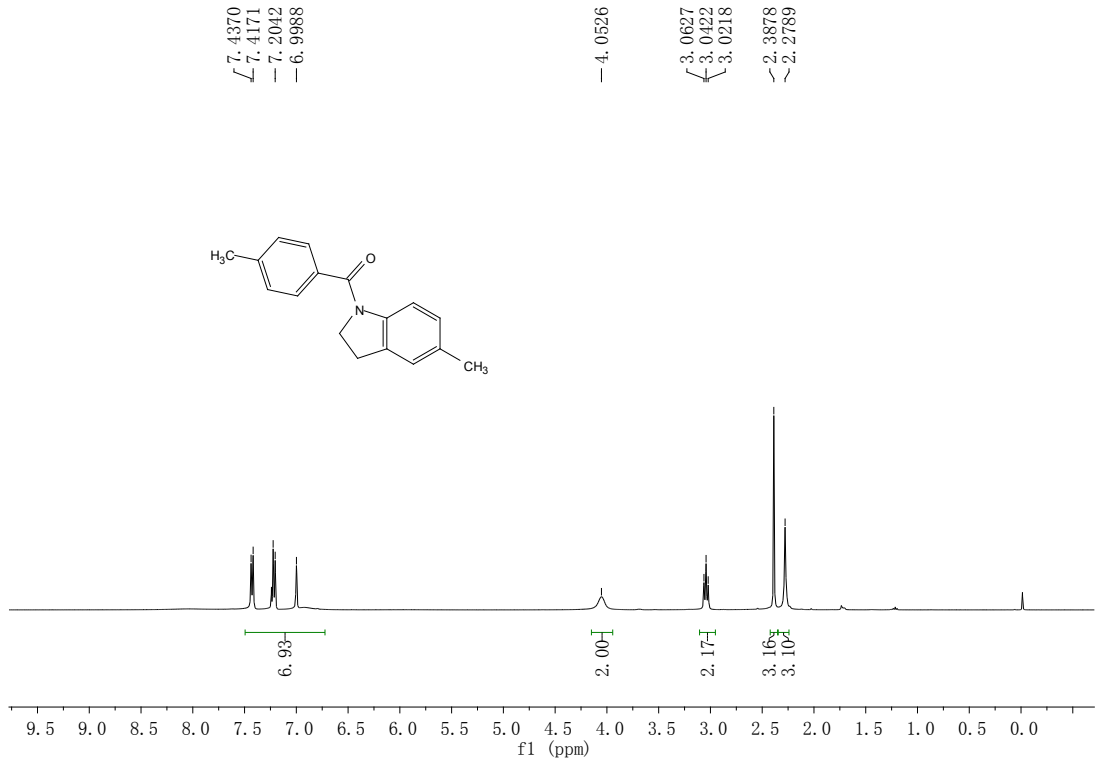
Indolin-1-yl(phenyl)methanone (11o):



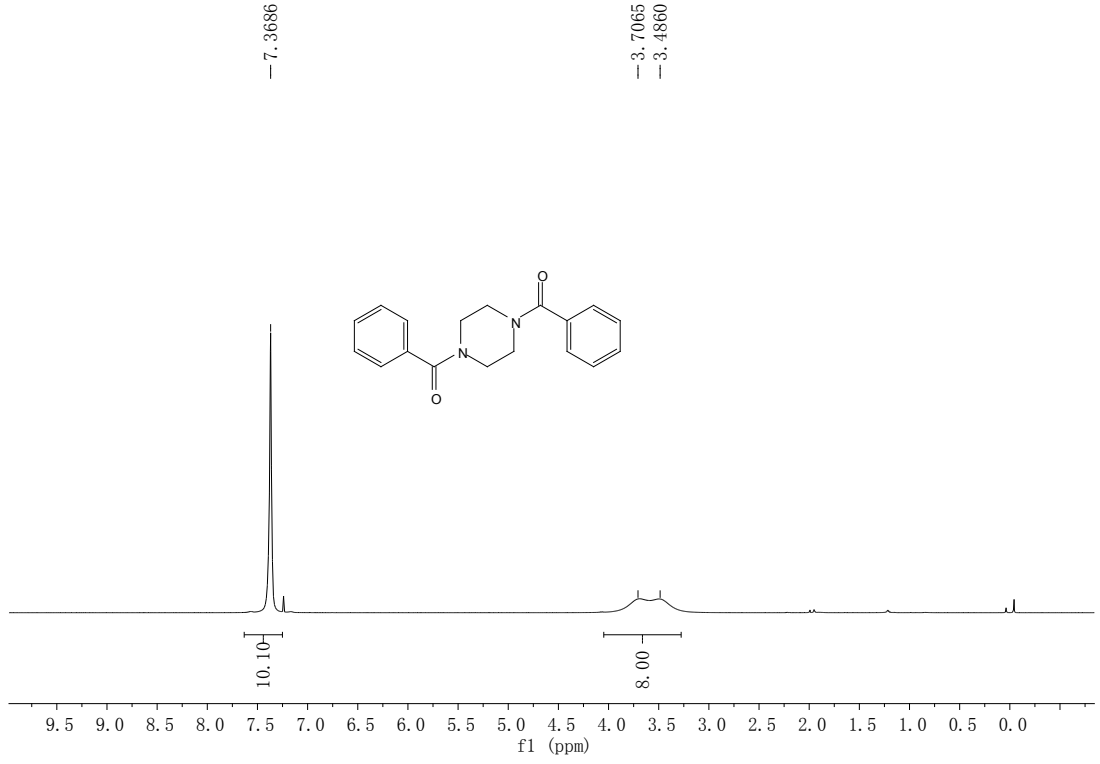
(2-Methylindolin-1-yl)(phenyl)methanone (11p):



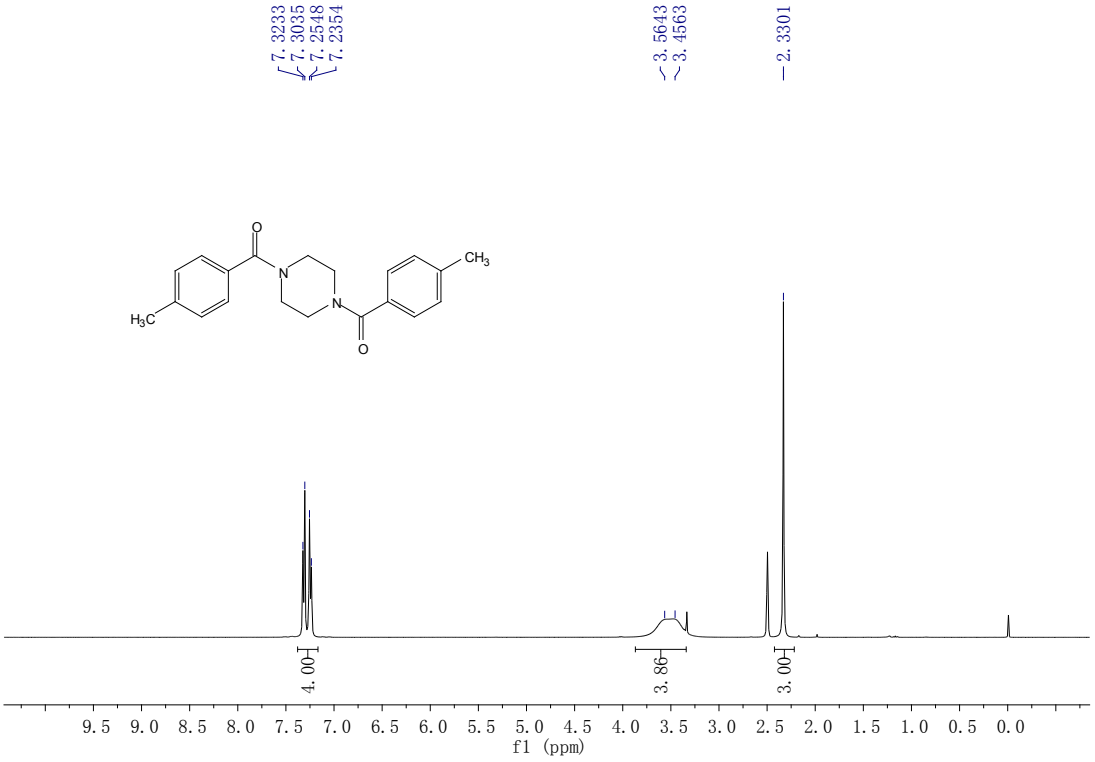
Indolin-1-yl(m-tolyl)methanone (11q):



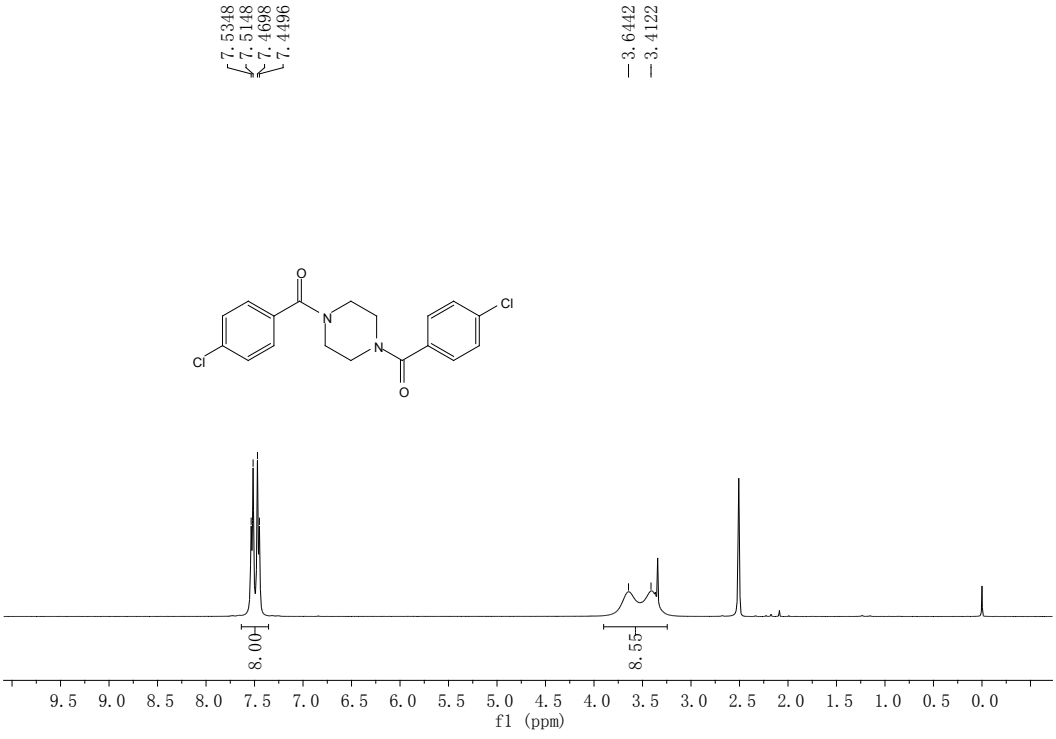
Piperazine-1,4-diylbis(phenylmethanone) (11r):



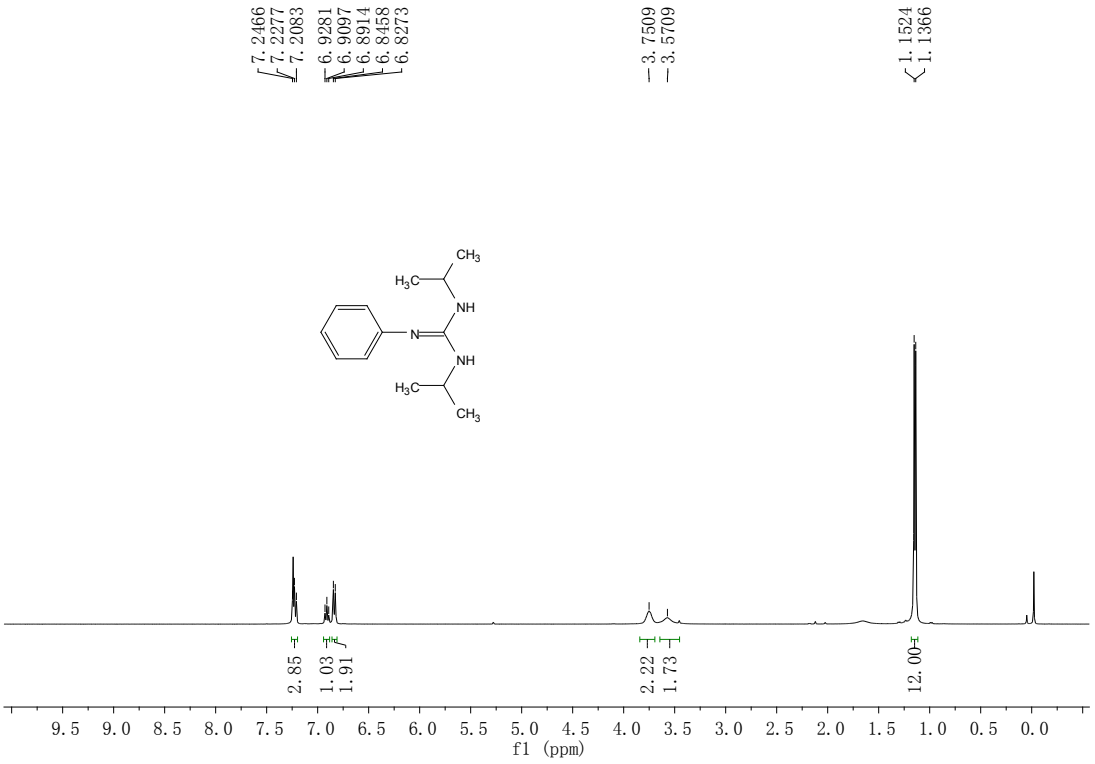
Piperazine-1,4-diylbis(p-tolylmethanone) (11s):



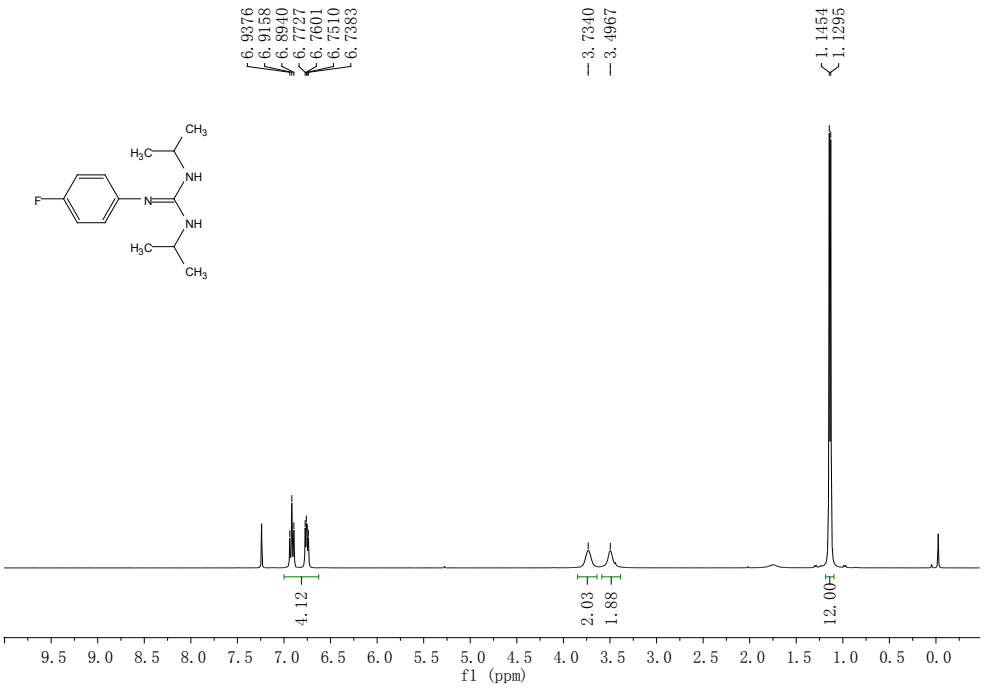
Piperazine-1,4-diylbis((4-chlorophenyl)methanone) (11t):



1,3-Diisopropyl-2-phenylguanidine (13a):

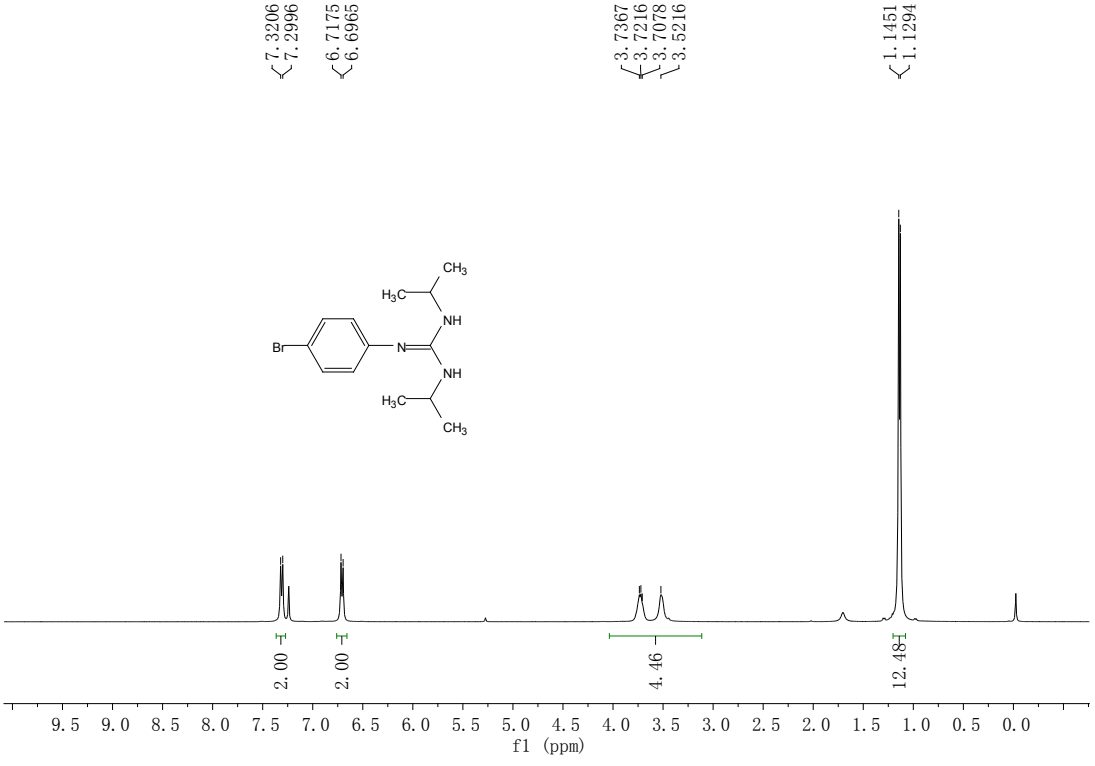


2-(4-Fluorophenyl)-1,3-diisopropylguanidine (13b):

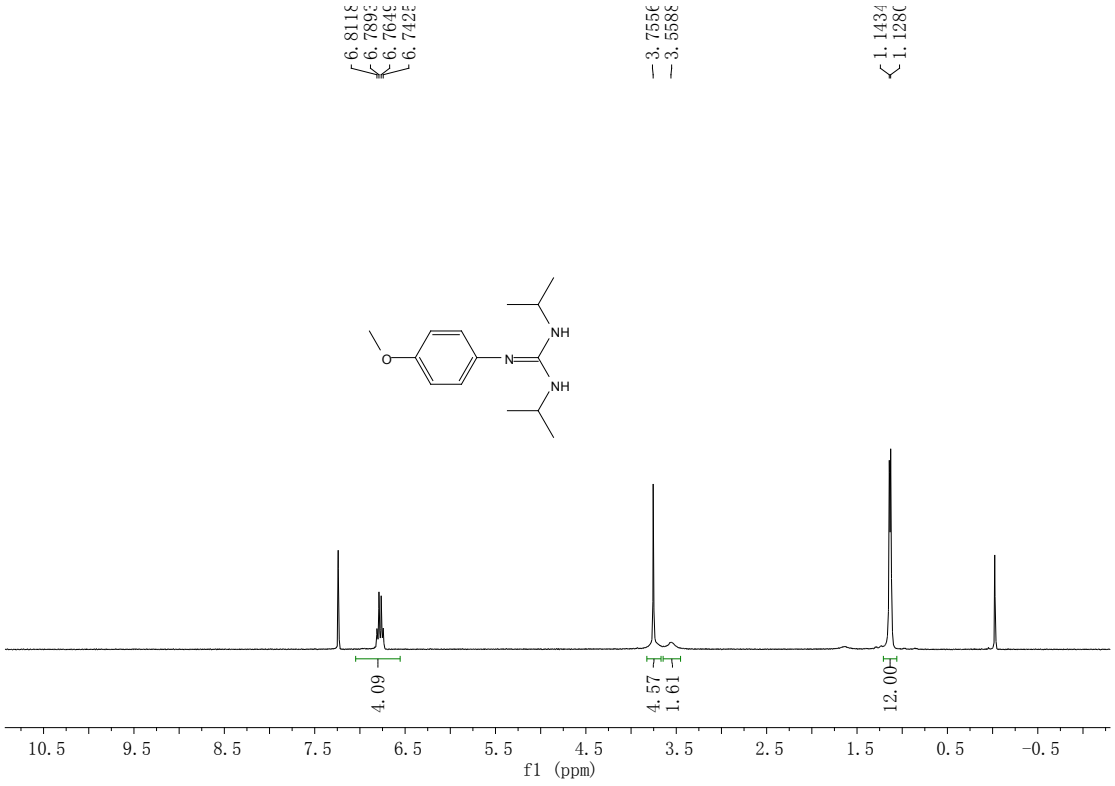


2-(4-Chlorophenyl)-1,3-diisopropylguanidine (13c):

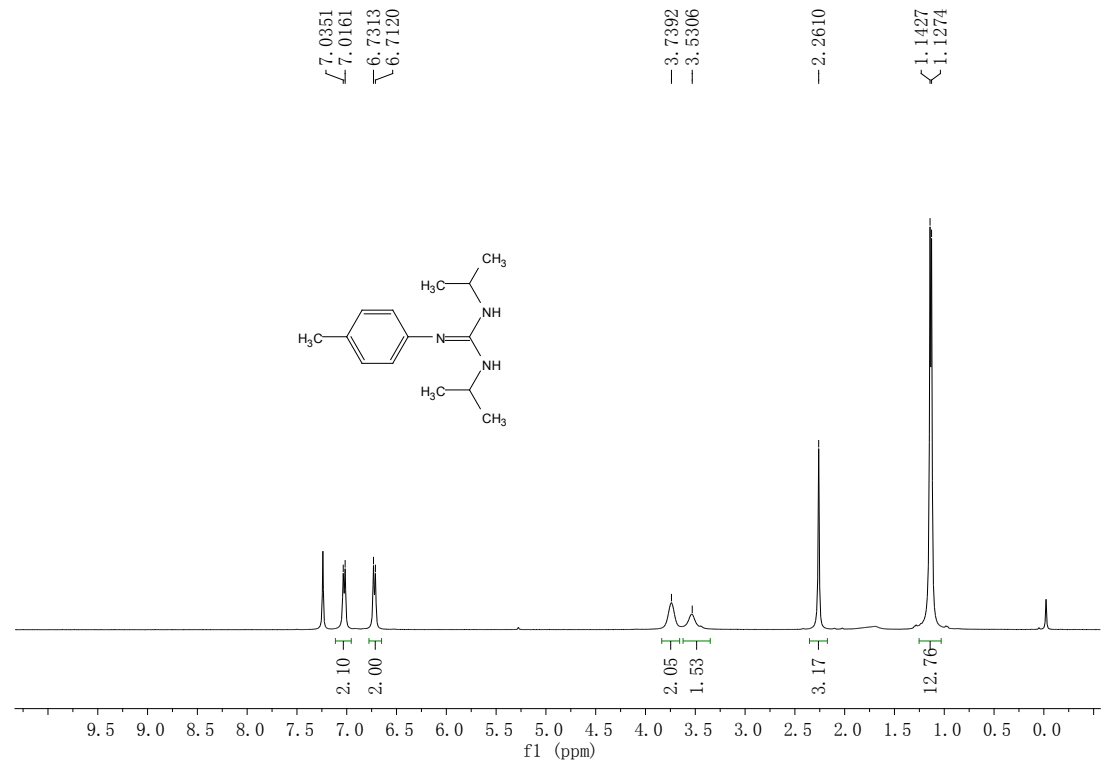
2-(4-Bromophenyl)-1,3-diisopropylguanidine (13d):



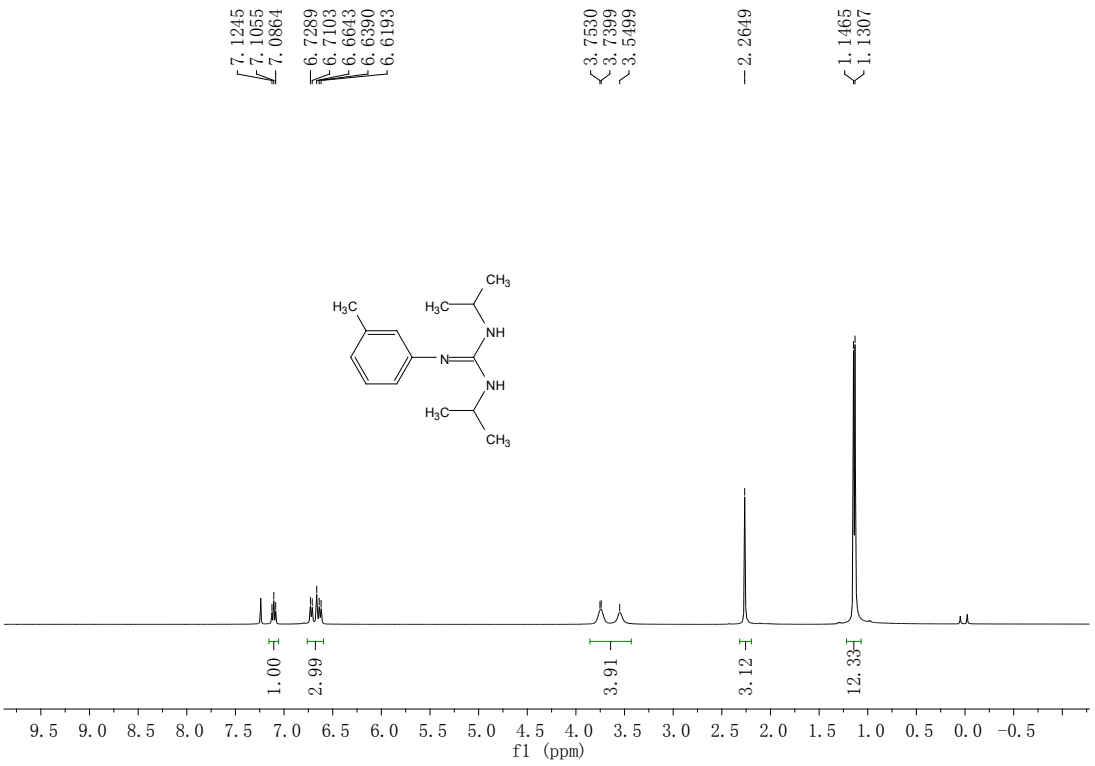
1,3-Diisopropyl-2-(4-methoxyphenyl)guanidine (13e):



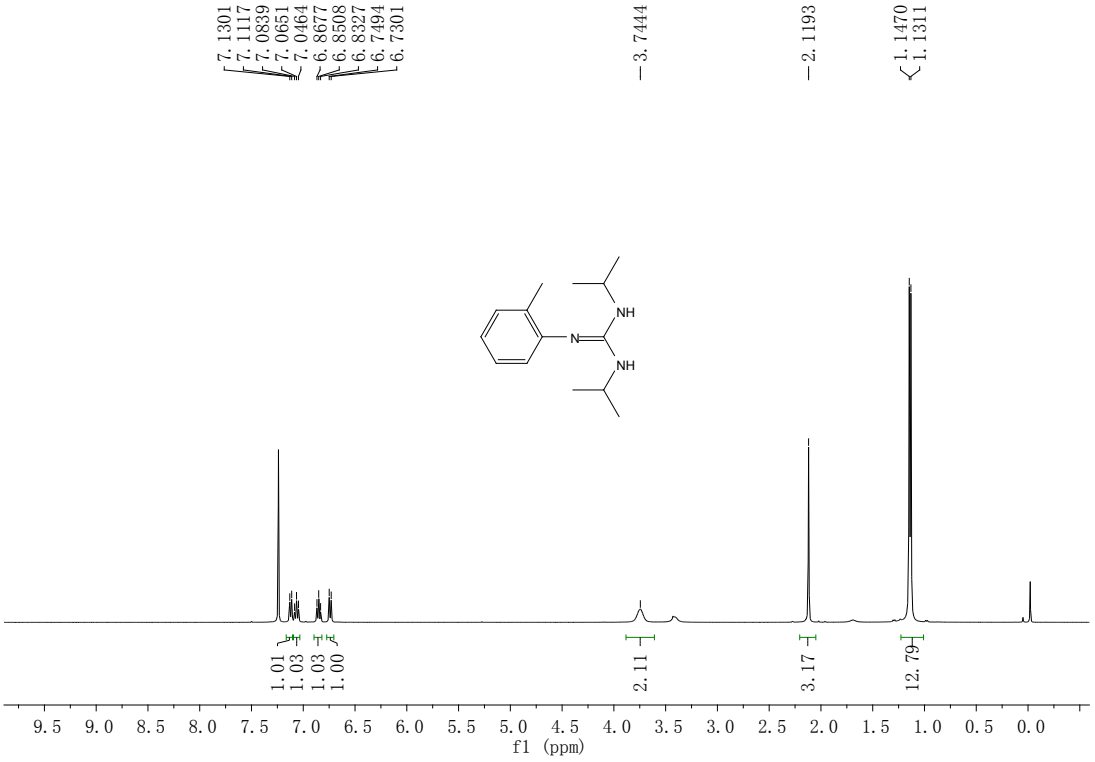
1,3-Diisopropyl-2-(p-tolyl)guanidine (13f):



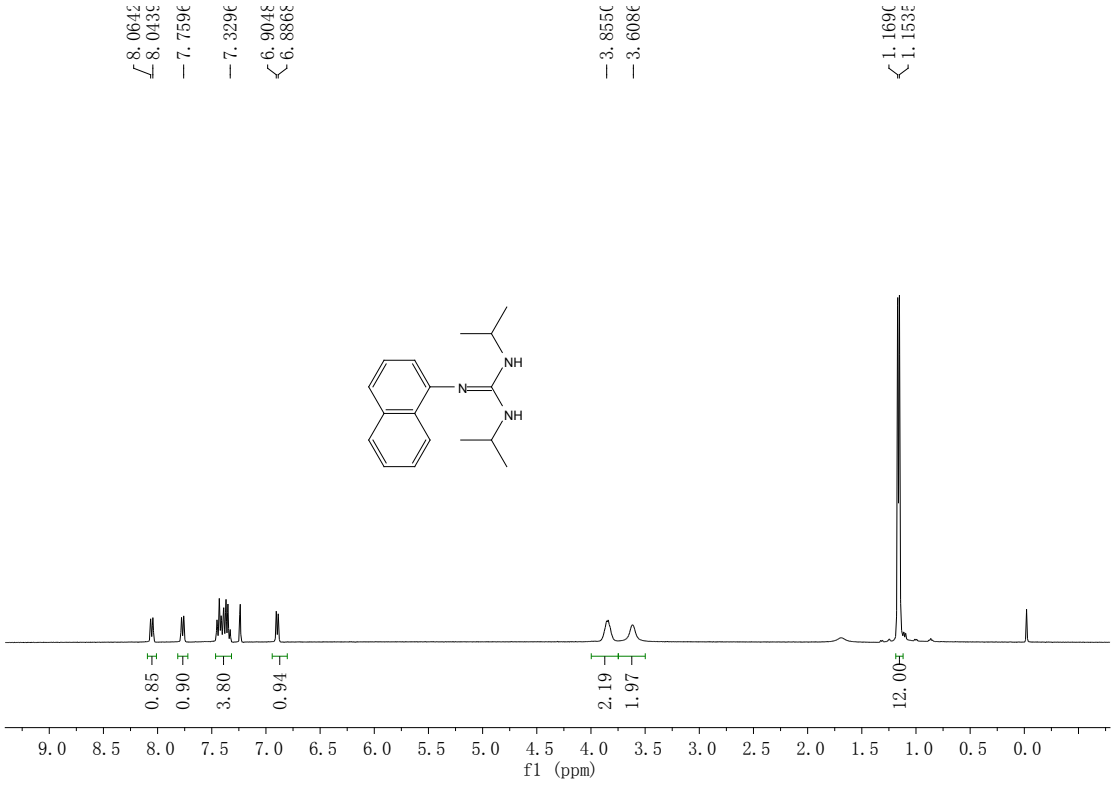
1,3-Diisopropyl-2-(m-tolyl)guanidine (13g):



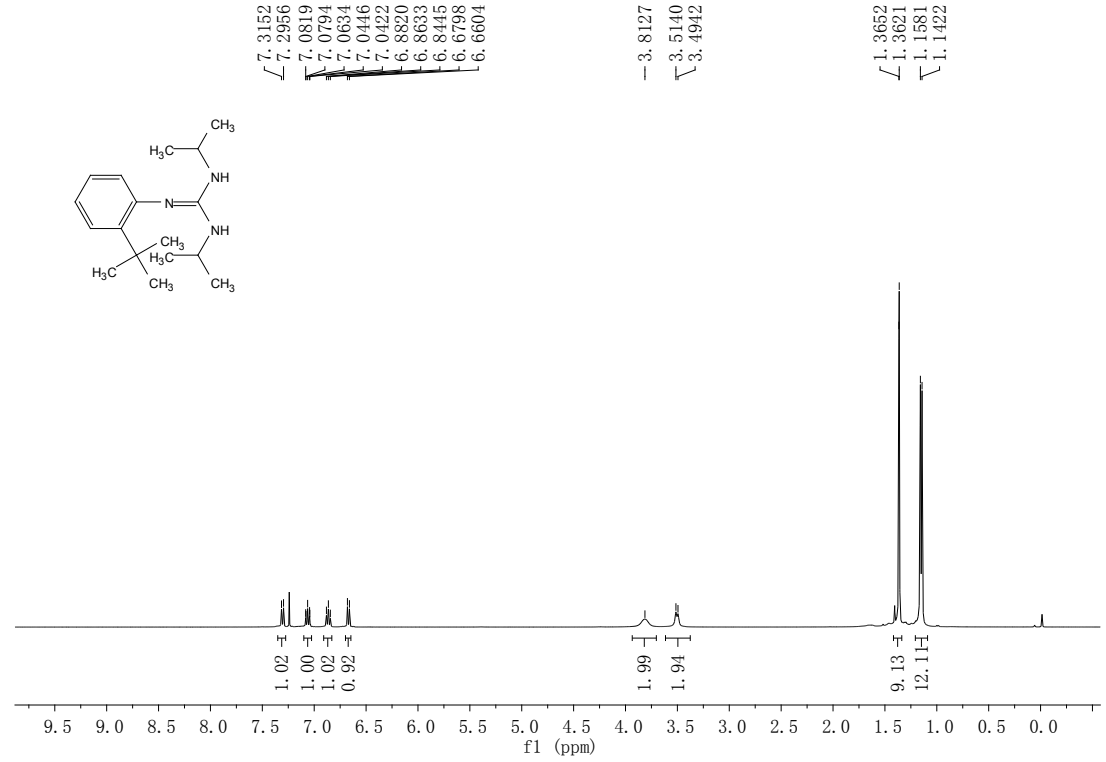
1,3-Diisopropyl-2-(o-tolyl)guanidine (13h):



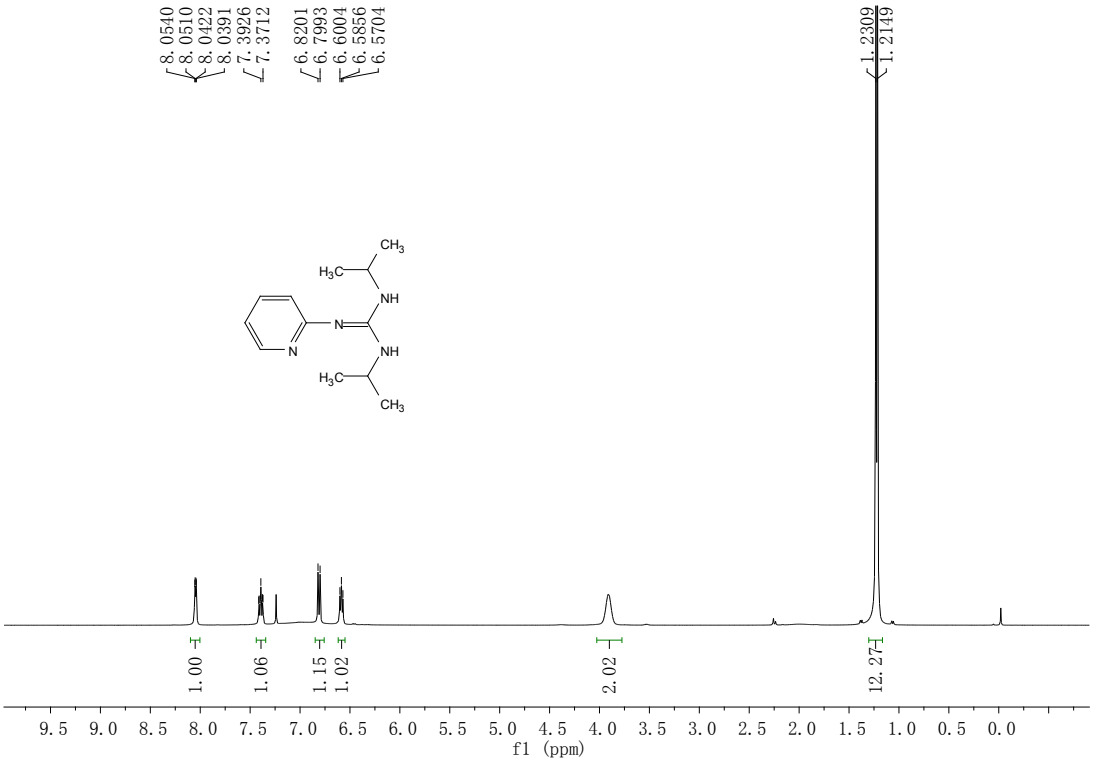
1,3-Diisopropyl-2-(naphthalen-1-yl)guanidine (13i):



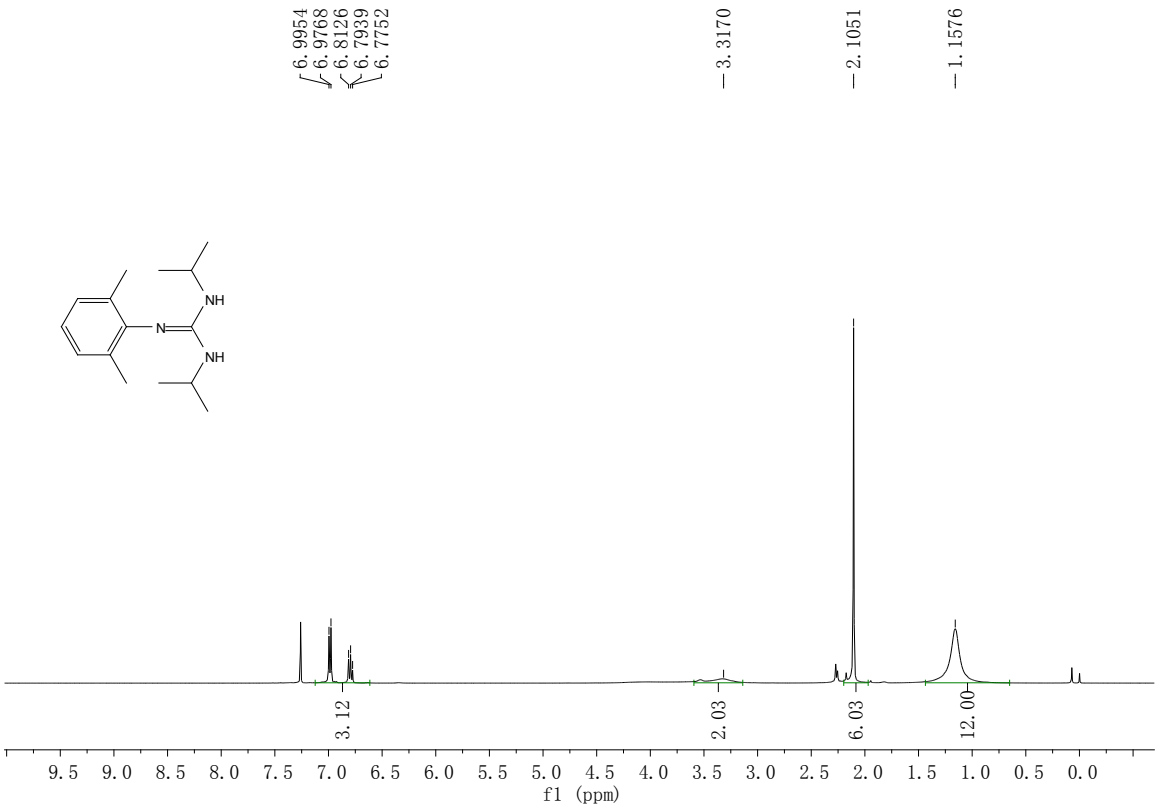
2-(2-(tert-Butyl)phenyl)-1,3-diisopropylguanidine (13j):



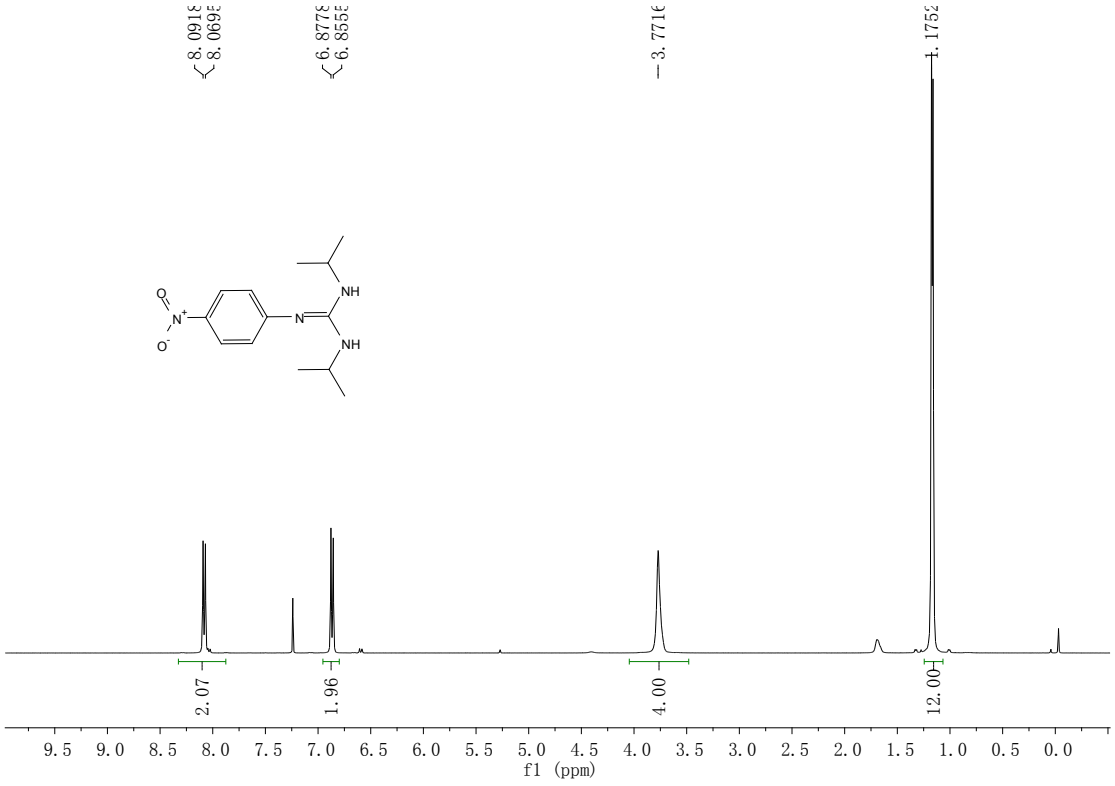
1,3-Diisopropyl-2-(pyridin-2-yl)guanidine (13k):



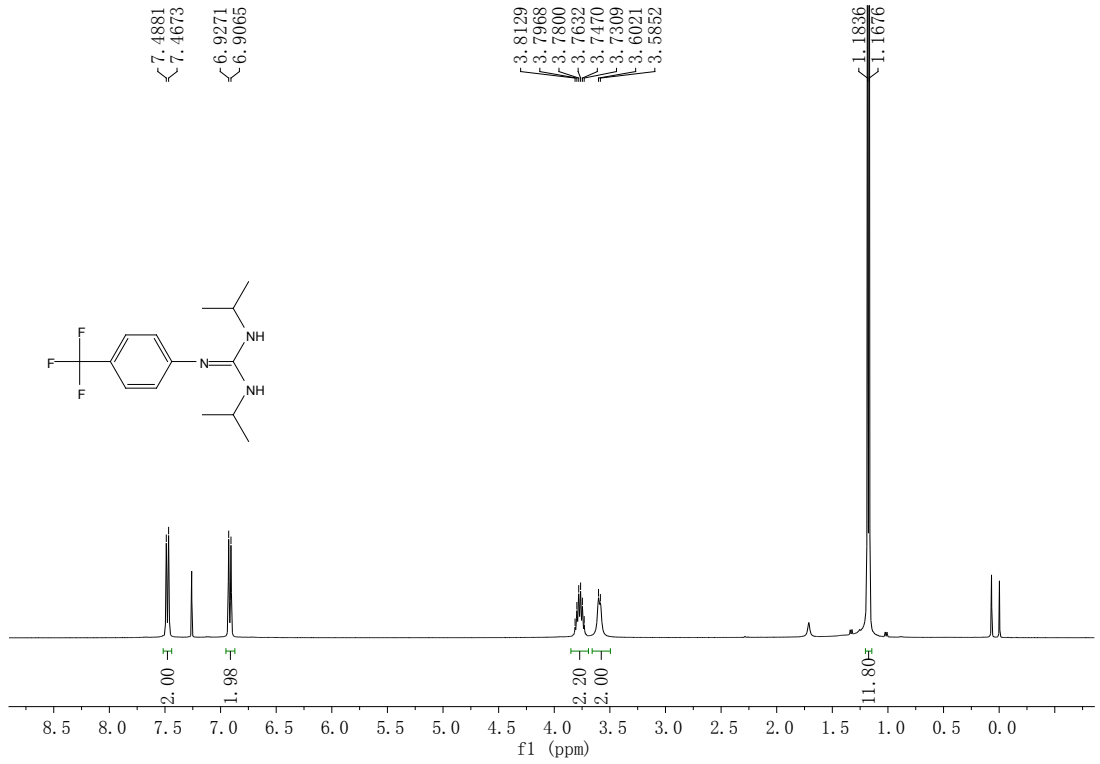
***N, N′′*-diisopropyl-*N′*-2, 6-dimethylphenylguanidine (13l):**



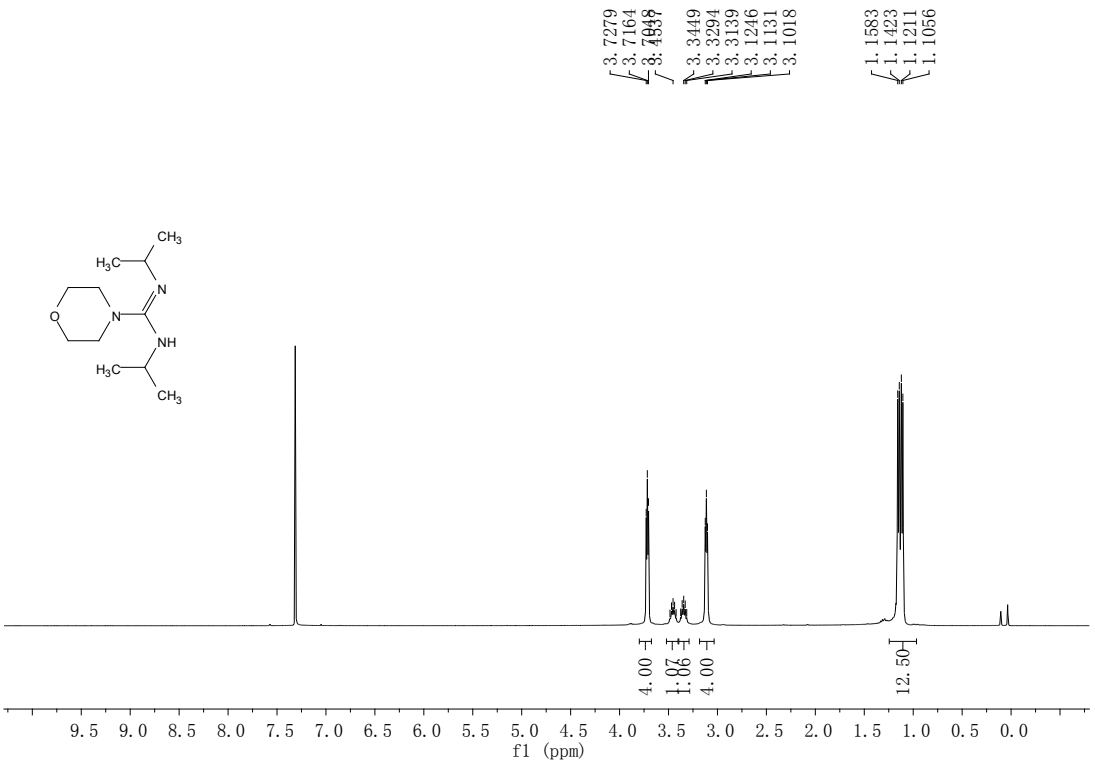
1,3-Diisopropyl-2-(4-nitrophenyl)guanidine (13m):



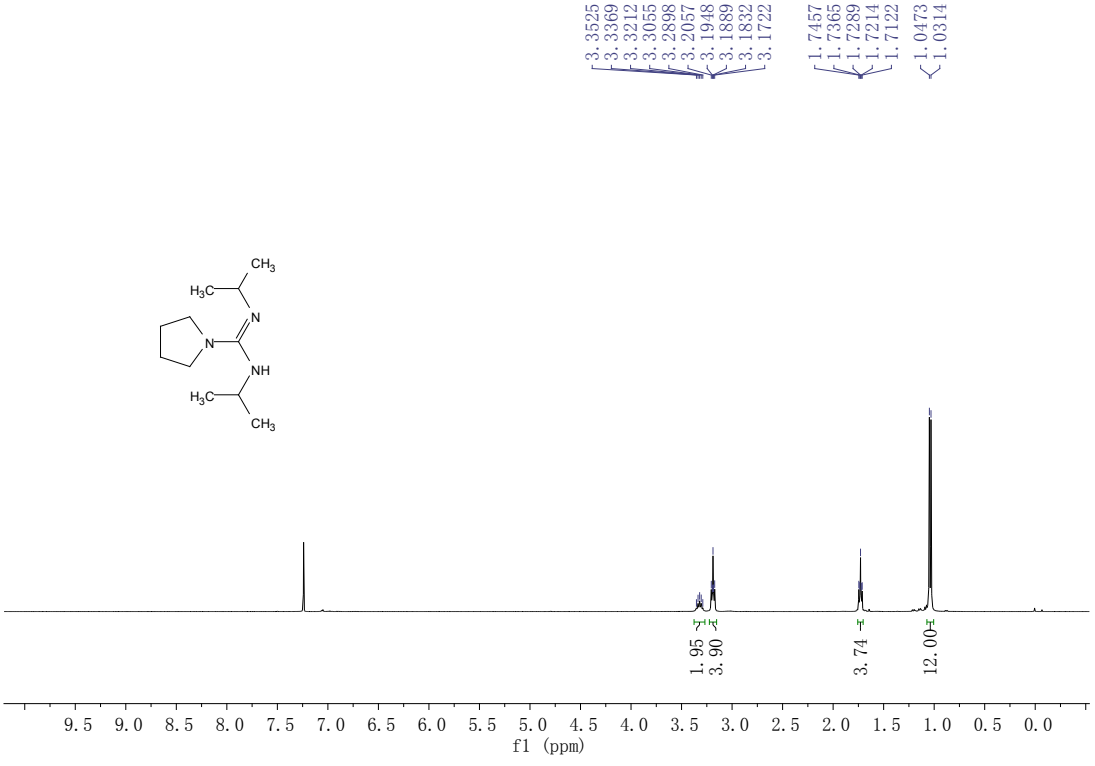
1,3-Diisopropyl-2-(4-(trifluoromethyl)phenyl)guanidine (13n):



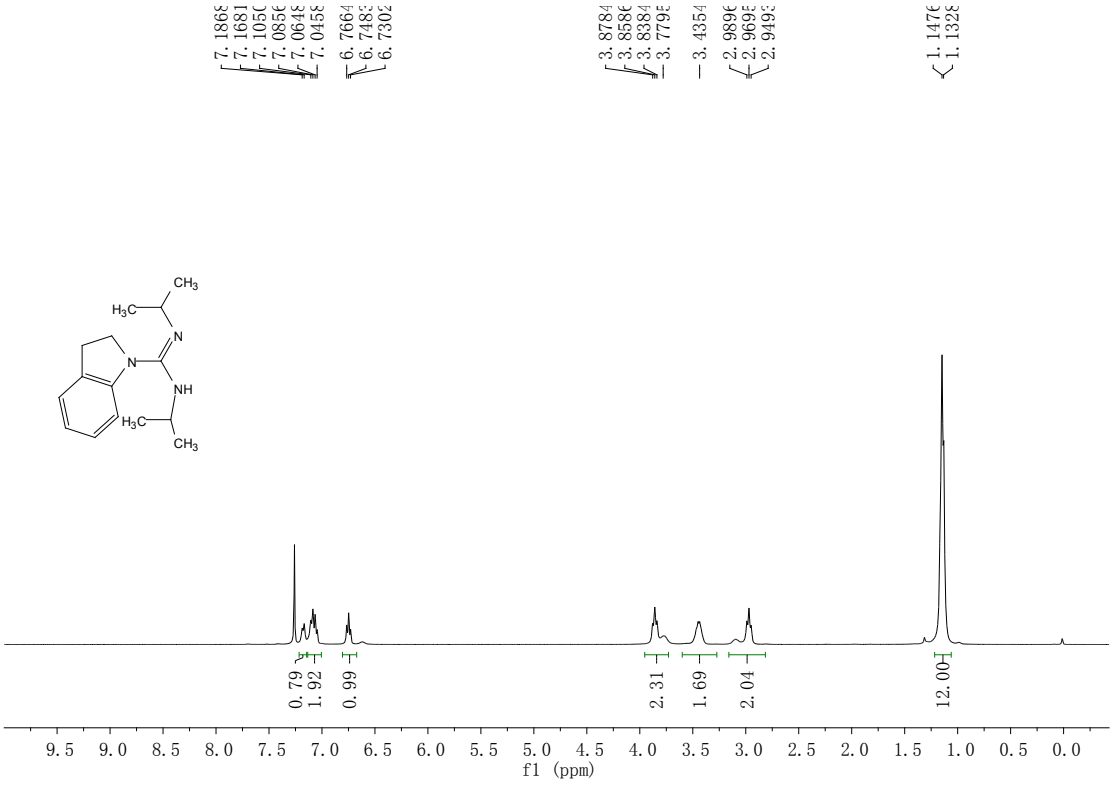
(*Z*)-N,N'-Diisopropylmorpholine-4-carboximidamide (13o):



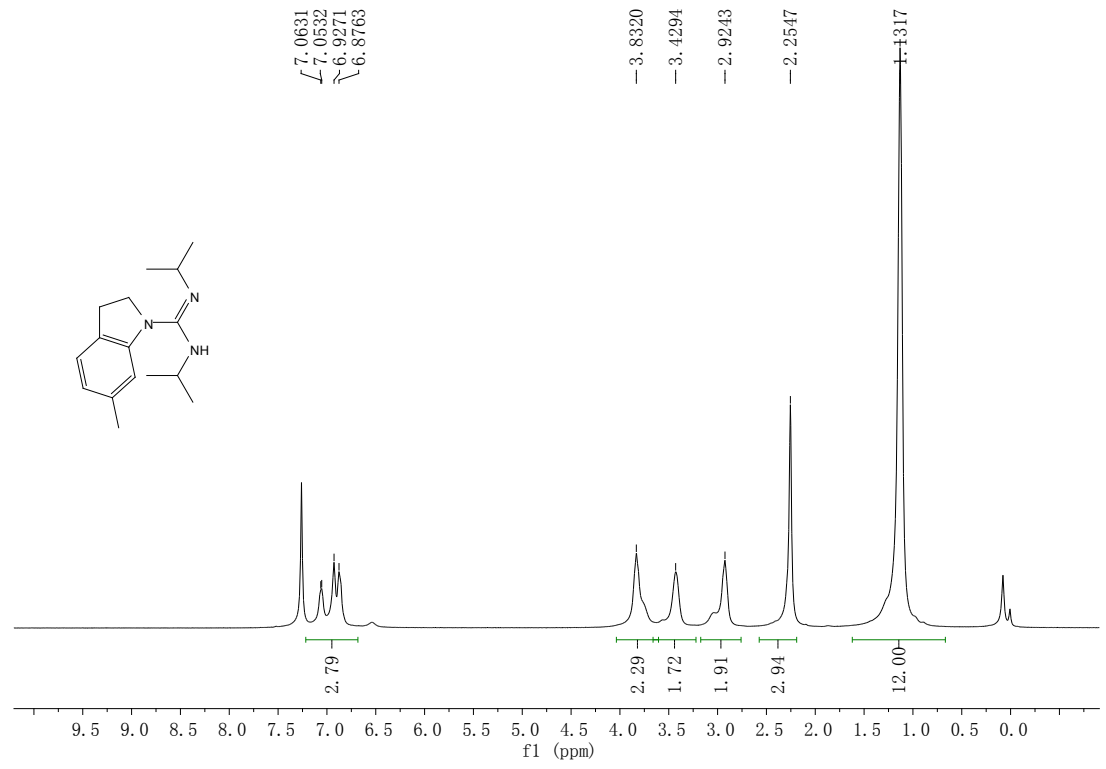
(*Z*)-N,N'-Diisopropylpyrrolidine-1-carboximidamide (13p):



(*Z*)-N,N'-Diisopropylindoline-1-carboximidamide (13q):



(*Z*)-N,N'-Diisopropyl-6-methylindoline-1-carboximidamide (13r):



(1*Z*, 4*Z*)-*N1,N'1,N4,N'4*-tetra-Isopropylpiperazine-1,4-bis(carboximidamide) (13s):