Supporting Information

Unmasking Amides: Ruthenium-Catalyzed Protodecarbonylation of *N*-Substituted Phthalimide Derivatives

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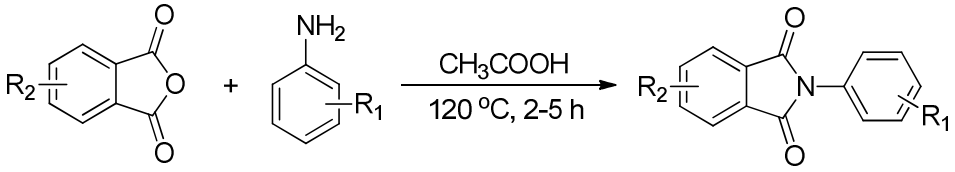
Contents

1. General Information.

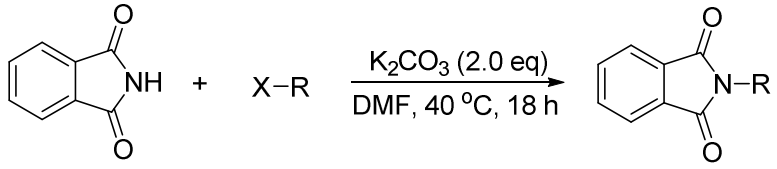
All reagents were obtained from commercial sources and used as supplied. All reactions were carried out in flame-dried glassware under argon atmosphere unless otherwise noted. Catalytic experiments were performed in Schlenk-type flasks under argon atmosphere unless otherwise noted. Organic solutions were concentrated under reduced pressure using a rotary evaporator. Thin-layer chromatography (TLC) were carried out on 0.25 mm Merck silica gel (60-F254). Flash column chromatography was performed using silica gel Silica 60 M, 0.04-0.063 mm. *N*-methyl-2-pyrrolidone (NMP) was distilled under reduced pressure and stored under molecular sieves and argon atmosphere. Technical grade petroleum ether (40-60) and ethyl acetate were used for column chromatography. $CDCl_3$ was stored under nitrogen over molecular sieves. NMR spectra were recorded on an AVANCE III 400 spectrometer. ^1H NMR spectra were referenced to residual protiated solvent (δ = 7.26 ppm for $CDCl_3$, δ = 2.50 ppm for DMSO-d_6 and δ = 2.05 ppm for acetone-d_6) and ^{13}C chemical shifts are reported relative to deuterated solvents (δ = 77.0 ppm for $CDCl_3$, δ = 39.5 ppm for DMSO-d_6 and δ = 29.8 ppm for acetone-d_6). The peak patterns are indicated as follows: s, singlet; d, doublet; t, triplet; q, quartet; m, multiplet, and br. for broad. GC-MS analyses were performed with a GCMS-QP2010S (Shimadzu) instrument with a GC-2010 equipped with a 30 m capillary column (Supelco, SLBTM-5ms, fused silica capillary column, 30 m x 0.25 mm x 0.25 mm film thickness), which was used with helium as the vector gas. The following GC conditions were used: initial temperature 80 ^oC for 2 minutes, then rate 20 ^oC/min until 280 ^oC and 280 ^oC for 28 minutes. HRMS were recorded on a Waters Q-Tof 2 mass spectrometer at the corresponding facilities of the CRMPO, Centre Régional de Mesures Physiques de l'Ouest, Université de Rennes 1.

2. Preparation of substrates 1.

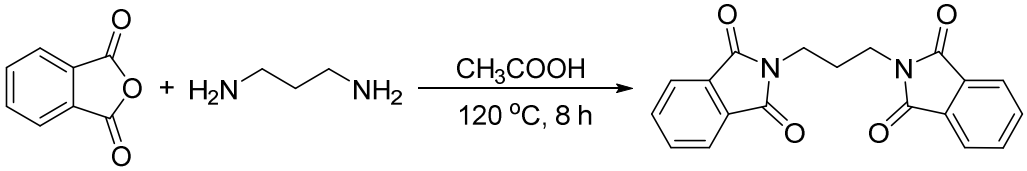
2.1. Method A: Phthalic anhydride (5 mmol, 0.74 g, 1 eq.) and the corresponding aniline (5 mmol, 1 eq.) were refluxed in acetic acid (30 mL) for 2-5 hours. Once at room temperature, water was added and the solid recovered by filtration. After drying under vacuum the desired phthalimide **1** was obtained.



2.2. Method B: Phthalimide (7 mmol, 1.03 g, 1 eq.), potassium carbonate (14 mmol, 2.59 g, 2 eq) and the corresponding alkyl or benzyl halide (14 mmol, 2 eq.) were heated at 40 °C in *N*,*N*-dimethylformamide (6 mL) for 18 hours. After solvents evaporation under vacuum, water was added to the reaction mixture followed by extraction with DCM. The combined organic phases were dried over $MgSO_4$, filtered, and concentrated in *vacuo*. The desired phthalimide **1** was purified by silica gel column chromatography with a mixture of petroleum ether and ethyl acetate as eluent.

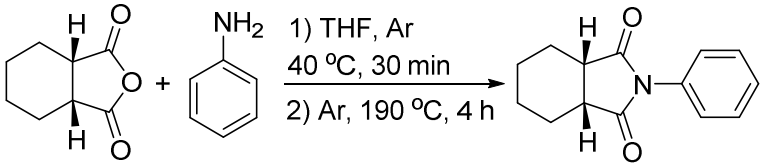


2.3. Method C: Phthalic anhydride (10 mmol, 1.48 g, 1 eq.) and 1,3-diaminopropane (5 mmol, 0.5 eq.) were refluxed in acetic acid (15 mL) for 8 hours. Once at room temperature, water was added and the solid recovered by filtration. After drying under vacuum the desired phthalimide **1** was obtained.

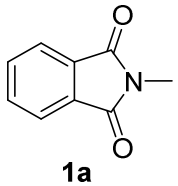


2.4. Method D: Hexahydrophthalic anhydride (10 mmol, 1.54 g, 1 eq.) and aniline (10 mmol, 1 eq.) and THF (15 mL) were added to a 100 mL round bottom flask. The solution was stirred for 30 min at 40 °C. Removal of the solvent using a rotary evaporator gave the corresponding carboxylic acid-amide as a white solid. The white solid was then heated

at 190 °C under Ar for 4 h. The desired phthalimide was purified by silica gel column chromatography with a mixture of petroleum ether and ethyl acetate as eluent.

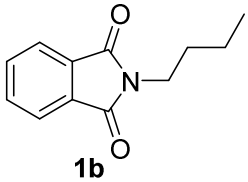


3. Characterization data of substrates 1.



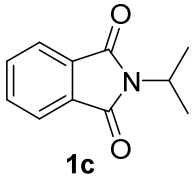
1a

***N*-Methylphthalimide (1a):** Prepared according to **Method B** starting from iodomethane in 88% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.78 (dd, J = 5.6 Hz, 3.2 Hz, 2H), 7.66 (dd, J = 5.6 Hz, 3.2 Hz, 2H), 3.13 (s, 3H) ppm. The spectral data match those previously reported.[1]



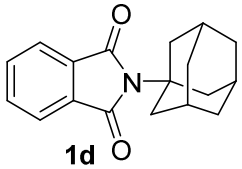
1b

***N*-Butylphthalimide (1b):** Prepared according to **Method B** starting from 1-bromobutane in 98% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.82 (dd, J = 5.6 Hz, 2.8 Hz, 2H), 7.70-7.68 (m, 2H), 3.67 (t, J = 7.6 Hz, 2H), 1.68-1.61 (m, 2H), 1.40-1.31 (m, 2H), 0.93 (t, J = 7.6 Hz, 3H) ppm. The spectral data match those previously reported.[2]



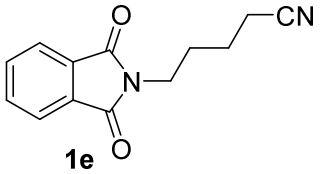
1c

***N*-isopropylphthalimide (1c):** Prepared according to **Method B** starting from 2-bromopropane in 78% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.79 (dd, J = 5.6 Hz, 3.2 Hz, 2H), 7.68 (dd, J = 5.6 Hz, 3.2 Hz, 2H), 4.57-4.47 (m, 1H), 1.48 (d, J = 7.2 Hz, 6H) ppm. The spectral data match those previously reported.[3]



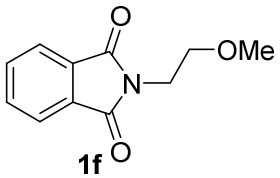
1d

***N*-(1-Adamantyl)phthalimide (1d):** Prepared according to **Method A** starting from amantadine in 35% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.75 (dd, J = 5.2 Hz, 3.2 Hz, 2H), 7.66 (dd, J = 5.2 Hz, 3.2 Hz, 2H), 2.52 (d, J = 2.8 Hz, 6H), 2.17 (s, 3H), 1.81-1.70 (m, 6H) ppm. The spectral data match those previously reported.[4]



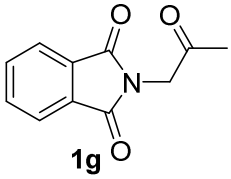
1e

5-Phthalimidovaleronitrile (1e): Prepared according to **Method B** starting from 5-bromovaleronitrile in 87% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.83 (dd, J = 5.6 Hz, 2.8 Hz, 2H), 7.72 (dd, J = 5.6 Hz, 3.2 Hz, 2H), 3.72 (t, J = 7.2 Hz, 2H), 2.42 (t, J = 7.2 Hz, 2H), 1.88-1.81 (m, 2H), 1.74-1.66 (m, 2H) ppm. The spectral data match those previously reported.[3]



1f

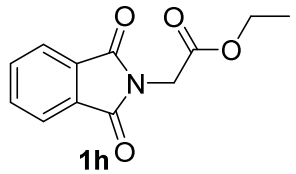
***N*-[2-Methoxyethyl]phthalimide (1f):** Prepared according to **Method A** starting from 2-methoxyethanamine in 75% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.84 (dd, J = 5.2 Hz, 2.8 Hz, 2H), 7.70 (dd, J = 5.2 Hz, 2.8 Hz, 2H), 3.89 (t, J = 6.0 Hz, 2H), 3.63 (t, J = 6.0 Hz, 2H), 3.34 (s, 3H) ppm. The spectral data match those previously reported.[5]



1g

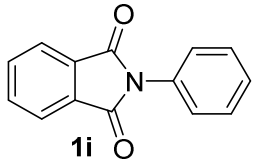
***N*-Acetonylphthalimide (1g):** Prepared according to **Method B** starting from chloroacetone in 50% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.86 (dd, J = 5.2

Hz, 2.8 Hz, 2H), 7.73 (dd, *J* = 5.2 HZ, 2.8 Hz, 2H), 4.49 (s, 2H), 2.26 (s, 3H) ppm. The spectral data match those previously reported.[6]



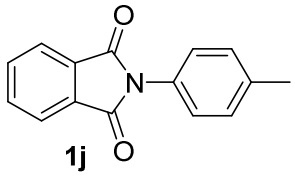
1h

Methyl phthalimidoacetate (1h): Prepared according to **Method B** starting from ethyl bromoacetate in 75% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.87 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.73 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 4.42 (s, 2H), 4.22 (q, *J* = 7.2 Hz, 2H), 1.27 (t, *J* = 7.2 Hz, 3H) ppm. The spectral data match those previously reported.[7]



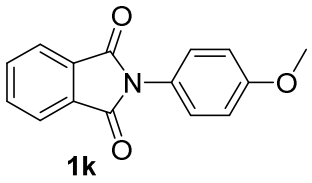
1i

***N*-Phenylphthalimide (1i):** Prepared according to **Method A** starting from aniline in 80% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.96 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.80 (dd, *J* = 5.2 Hz, 3.2 Hz, 2H), 7.52 (dd, *J* = 7.6 Hz, 7.6 Hz, 2H), 7.34-7.27 (m, 3H) ppm. The spectral data match those previously reported.[2]



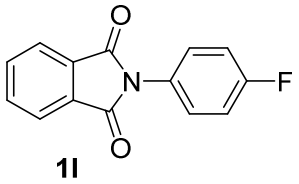
1j

***N*-(*p*-Tolyl)phthalimide (1j):** Prepared according to **Method A** starting from *p*-toluidine in 72% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.94 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.78 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.31 (s, 4H), 2.41 (s, 3H) ppm. The spectral data match those previously reported.[2]



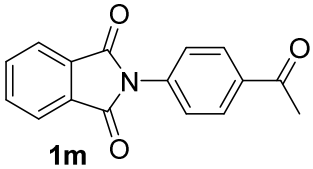
1k

***N*-*p*-Anisylphthalimide (1k):** Prepared according to **Method A** starting from *p*-anisidine in 80% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.94 (dd, *J* = 5.2 Hz, 2.8 Hz, 2H), 7.78 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.34 (d, *J* = 9.2 Hz, 2H), 7.02 (d, *J* = 9.2 Hz, 2H), 3.85 (s, 3H) ppm. The spectral data match those previously reported.[2]



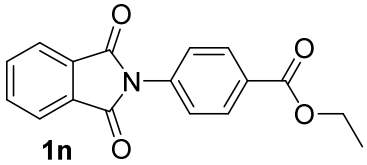
1l

***N*-(*p*-Fluorophenyl)phthalimide (1l):** Prepared according to **Method A** starting from *p*-fluoroaniline in 82% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.86 (dd, *J* = 5.2 Hz, 3.2 Hz, 2H), 7.71 (dd, *J* = 5.2 Hz, 3.2 Hz, 2H), 7.34 (dd, *J* = 9.2 Hz, 4.8 Hz, 2H), 7.11 (dd, *J* = 8.8 Hz, 8.8 Hz, 2H) ppm. ^{19}F{^{1}H} NMR (376 MHz, $CDCl_3$): δ = -113.8 ppm. The spectral data match those previously reported.[8]



1m

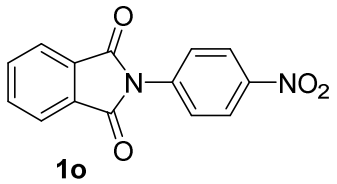
***p*-Phthalimidoacetophenone (1m):** Prepared according to **Method A** starting from *p*-aminoacetophenone in 79% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.10 (dd, *J* = 6.8 Hz, 2.0 Hz, 2H), 7.97 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.82 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.63 (dd, *J* = 6.8 Hz, 2.0 Hz, 2H), 2.64 (s, 3H) ppm. The spectral data match those previously reported.[2]



1n

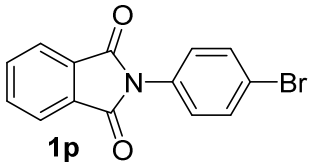
***N*-(*p*-Ethoxycarbonylphenyl)phthalimide (1n):** Prepared according to **Method A** starting from benzocaine in 50% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.19

(dd, *J* = 6.8 Hz, 1.6 Hz, 2H), 7.97 (dd, *J* = 5.2 Hz, 2.8 Hz, 2H), 7.82 (dd, *J* = 5.2 Hz, 2.8 Hz, 2H), 7.59 (dd, *J* = 6.8 Hz, 1.6 Hz, 2H), 4.41 (q, *J* = 7.2 Hz, 2H), 1.41 (t, *J* = 7.2 Hz, 3H) ppm. The spectral data match those previously reported.[9]



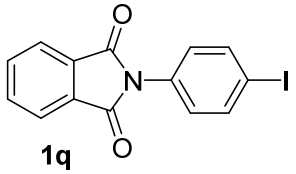
1o

***N*-(*p*-Nitrophenyl)phthalimide (1o):** Prepared according to **Method A** starting from *p*-nitroaniline in 63% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.38 (dd, *J* = 7.2 Hz, 2.0 Hz, 2H), 8.00 (dd, *J* = 5.2 Hz, 2.8 Hz, 2H), 7.85 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.78 (dd, *J* = 7.2 Hz, 2.0 Hz, 2H) ppm. The spectral data match those previously reported.[2]



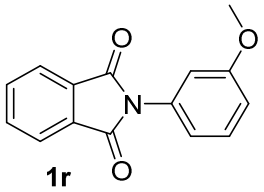
1p

***N*-*p*-Bromophenylphthalimide (1p):** Prepared according to **Method A** starting from *p*-bromoaniline in 88% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.96 (dd, *J* = 5.2 Hz, 2.8 Hz, 2H), 7.80 (dd, *J* = 5.2 Hz, 2.8 Hz, 2H), 7.63 (d, *J* = 8.4 Hz, 2H), 7.36 (d, *J* = 8.8 Hz, 2H) ppm. The spectral data match those previously reported.[10]



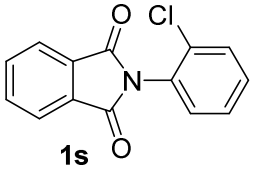
1q

***N*-*p*-Iodophenylphthalimide (1q):** Prepared according to **Method A** starting from *p*-iodoaniline in 80% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.96-7.94 (m, 2H), 7.84-7.79 (m, 4H), 7.23 (d, *J* = 8.4 Hz, 2H) ppm. The spectral data match those previously reported.[11]



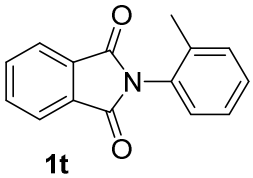
1r

***N*-(*m*-Methoxyphenyl)phthalimide (1r):** Prepared according to **Method A** starting from *m*-anisidine in 76% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.95 (dd, *J* = 5.2 Hz, 3.2 Hz, 2H), 7.78 (dd, *J* = 5.2 Hz, 3.2 Hz, 2H), 7.41 (dd, *J* = 8.0 Hz, 8.0 Hz, 1H), 7.04-7.02 (m, 1H), 6.99-6.96 (m, 1H), 6.94 (dd, *J* = 2.4 Hz, 1.2 Hz, 1H), 3.84 (s, 3H) ppm. The spectral data match those previously reported.[12]



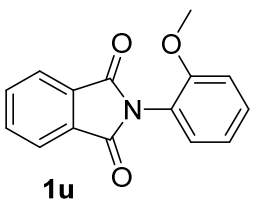
1s

***N*-(*o*-Chlorophenyl)phthalimide (1s):** Prepared according to **Method A** starting from *o*-chloroaniline in 70% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.96 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.80 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.59-7.55 (m, 1H), 7.46-7.38 (m, 2H), 7.37-7.35 (m, 1H) ppm. The spectral data match those previously reported.[13]



1t

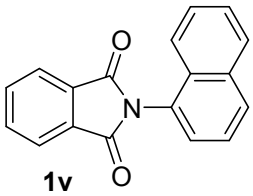
***N*-*o*-Tolylphthalimide (1t):** Prepared according to **Method A** starting from *o*-toluidine in 43% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.96 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.80 (dd, *J* = 5.6 Hz, 2.8 Hz, 2H), 7.39-7.31 (m, 3H), 7.21 (d, *J* = 7.6 Hz, 1H), 2.21 (s, 3H) ppm. The spectral data match those previously reported.[4]



1u

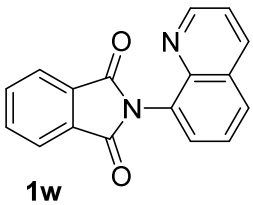
***N*-*o*-Methoxyphenylphthalimide (1u):** Prepared according to **Method A** starting from *o*-anisidine in 85% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.94 (dd, *J* = 5.6 Hz,

3.2 Hz, 2H), 7.77 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.46-7.42 (m, 1H), 7.27-7.25 (m, 1H), 7.10-7.04 (m, 2H), 3.80 (s, 3H) ppm. The spectral data match those previously reported.[14]



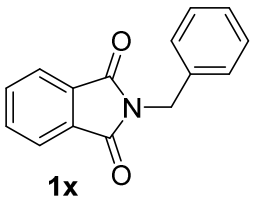
1v

***N*-1-Naphthylphthalimide (1v):** Prepared according to **Method A** starting from 1-aminonaphthalene in 79% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 8.02-7.99 (m, 3H), 7.95 (d, *J* = 7.2 Hz, 1H), 7.82 (dd, *J* = 5.6 Hz, 2.8 Hz, 2H), 7.66-7.59 (m, 2H), 7.56-7.48 (m, 3H) ppm. The spectral data match those previously reported.[15]



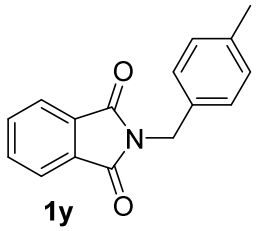
1w

***N*-8-Quinolyl-phthalimide (1w):** Prepared according to **Method A** starting from 8-aminoquinoline in 76% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 8.86 (dd, *J* = 4.4 Hz, 1.6 Hz, 1H), 8.23 (dd, *J* = 8.4 Hz, 1.6 Hz, 1H), 8.02-7.96 (m, 3H), 7.81 (dd, *J* = 5.2 Hz, 3.2 Hz, 2H), 7.76 (dd, *J* = 7.6 Hz, 1.6 Hz, 1H), 7.68 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H), 7.44 (dd, *J* = 8.0 Hz, 4.4 Hz, 1H) ppm. The spectral data match those previously reported.[16]



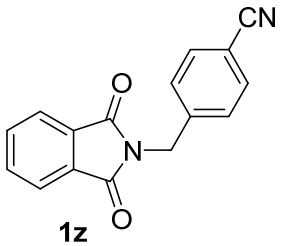
1x

***N*-Benzylphthalimide (1x):** Prepared according to **Method B** starting from benzyl bromide in 63% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.84 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.70 (dd, *J* = 5.2 Hz, 3.2 Hz, 2H), 7.43 (d, *J* = 7.2 Hz, 2H), 7.34-7.27 (m, 3H), 4.85 (s, 2H) ppm. The spectral data match those previously reported.[17]



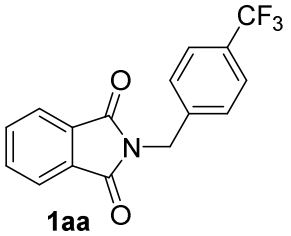
1y

***N*-[*p*-(methyl)benzyl]phthalimide (1y):** Prepared according to **Method B** starting from *p*-methylbenzyl bromide in 86% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.83 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.69 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.33 (d, *J* = 8.4 Hz, 2H), 7.12 (d, *J* = 7.6 Hz, 2H), 4.81 (s, 2H), 2.30 (s, 3H) ppm. The spectral data match those previously reported.[3]



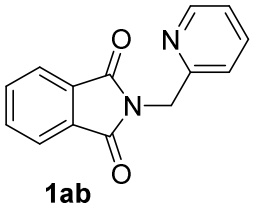
1z

***N*-[*p*-(cyano)benzyl]phthalimide (1z):** Prepared according to **Method B** starting from *p*-cyanobenzyl bromide in 86% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.86 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.73 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.61 (d, *J* = 8.0 Hz, 2H), 7.52 (d, *J* = 8.0 Hz, 2H), 4.88 (s, 2H) ppm. The spectral data match those previously reported.[14]



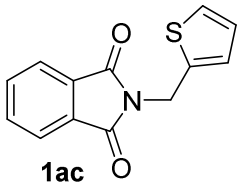
1aa

***N*-[*p*-(trifluoromethyl)benzyl]phthalimide (1aa):** Prepared according to **Method B** starting from *p*-(triflouromethyl)benzyl bromide in 77% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.85 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.72 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.58-7.52 (m, 4H), 4.89 (s, 2H) ppm. The spectral data match those previously reported.[3]



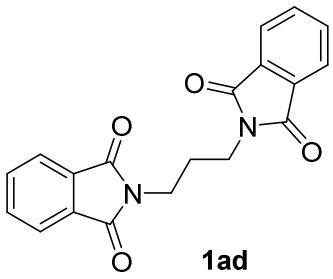
1ab

2-Phthalimidomethylpyridine (1ab): Prepared according to **Method A** starting from 2-picolylamine in 73% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.53-8.51 (m, 1H), 7.88 (dd, *J* = 5.6 Hz, 3.2 Hz, 2H), 7.73 (dd, *J* = 5.6 Hz, 2.8 Hz, 2H), 7.65-7.61 (m, 1H), 7.27 (d, *J* = 8.4 Hz, 1H), 7.15 (dd, *J* = 7.6 Hz, 4.8 Hz, 1H), 5.01 (s, 2H) ppm. The spectral data match those previously reported.[18]



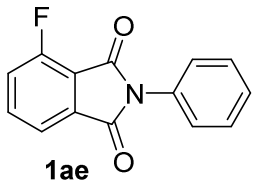
1ac

2-Phthalimidomethylthiophene (1ac): Prepared according to **Method A** starting from 2-thiophenemethylamine in 47% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.84 (dd, *J* = 5.2 Hz, 2.8 Hz, 2H), 7.69 (dd, *J* = 5.2 Hz, 2.8 Hz, 2H), 7.20 (dd, *J* = 5.2 Hz, 1.2 Hz, 1H), 7.14 (dd, *J* = 3.2 Hz, 0.4 Hz, 1H), 6.92 (dd, *J* = 5.2 Hz, 3.6 Hz, 1H), 5.01 (s, 2H) ppm. The spectral data match those previously reported.[19]



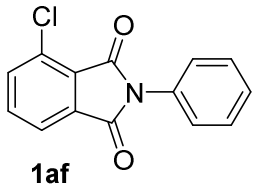
1ad

***N*,*N*-Diphthaloyl-1,3-propanediamine (1ad):** Prepared according to **Method C** starting from 1,3-diaminopropane in 86% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.83-7.79 (m, 4H), 7.71-7.67 (m, 4H), 3.75 (t, *J* = 7.2 Hz, 4H), 2.12-2.05 (m, 2H) ppm. The spectral data match those previously reported.[20]



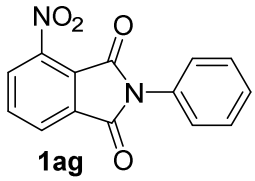
1ae

***N*-phenyl-*m*-fluorophthalimide (1ae):** Prepared according to **Method A** starting from *m*-fluorophthalic anhydride in 91% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.82-7.77 (m, 2H), 7.53-7.49 (m, 2H), 7.47-7.40 (m, 4H) ppm. $^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$): δ= -112.3 ppm. The spectral data match those previously reported.[21]



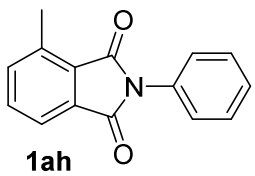
1af

***N*-phenyl-*m*-chlorophthalimide (1af):** Prepared according to **Method A** starting from *m*-chlorophthalic anhydride in 94% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.87 (dd, *J* = 4.8 Hz, 3.6 Hz, 1H), 7.71-7.70 (m, 2H), 7.53-7.48 (m, 2H), 7.45-7.41 (m, 3H) ppm. The spectral data match those previously reported.[22]



1ag

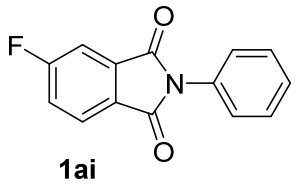
***N*-phenyl-*m*-nitrophthalimide (1ag):** Prepared according to **Method A** starting from *m*-nitrophthalic anhydride in 78% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 8.21 (dd, *J* = 7.6 Hz, 0.8 Hz, 1H), 8.15 (dd, *J* = 8.0 Hz, 0.8 Hz, 1H), 7.98 (dd, *J* = 8.0 Hz, 8.0 Hz, 1H), 7.54-7.50 (m, 2H), 7.46-7.42 (m, 3H) ppm. The spectral data match those previously reported.[23]



1ah

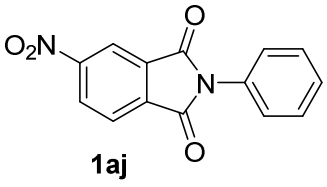
***N*-phenyl-*m*-methylphthalimide (1ah):** Prepared according to **Method A** starting from *m*-methylphthalic anhydride in 95% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.78

(d, J = 7.2 Hz, 1H), 7.63 (d, J = 7.6 Hz, 1H), 7.54-7.49 (m, 3H), 7.45-7.38 (m, 3H), 2.75 (s, 3H) ppm. The spectral data match those previously reported.[24]



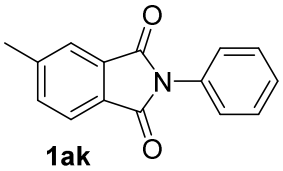
1ai

***N*-phenyl-*p*-fluorophthalimide (1ai):** Prepared according to **Method A** starting from *p*-fluorophthalic anhydride in 89% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.96 (dd, J = 8.4 Hz, 4.4 Hz, 1H), 7.62 (dd, J = 7.2 Hz, 2.0 Hz, 1H), 7.54-7.47 (m, 2H), 7.46-7.39 (m, 4H) ppm. ^{19}F{^{1}H} NMR (376 MHz, $CDCl_3$): δ = -101.1 ppm. The spectral data match those previously reported.[21]



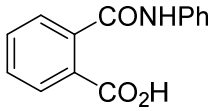
1aj

***N*-phenyl-*p*-nitrophthalimide (1aj):** Prepared according to **Method A** starting from *p*-nitrophthalic anhydride in 98% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 8.77 (dd, J = 6.0, 0.8 Hz, 1H), 8.67 (dd, J = 8.0 Hz, 2.0 Hz, 1H), 8.16 (dd, J = 8.0 Hz, 0.8 Hz, 1H), 7.56-7.52 (m, 2H), 7.48-7.43 (m, 3H) ppm. The spectral data match those previously reported.[15]



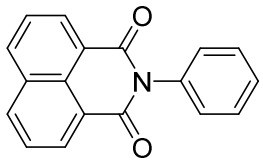
1ak

***N*-phenyl-*p*-methylphthalimide (1ak):** Prepared according to **Method A** starting from *p*-methylphthalic anhydride in 97% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.83 (d, J = 7.6 Hz, 1H), 7.75 (dd, J = 0.8 Hz, 0.8 Hz, 1H), 7.59-7.56 (m, 1H), 7.52-7.48 (m, 2H), 7.45-7.37 (m, 3H), 2.55 (s, 3H) ppm. The spectral data match those previously reported.[19]



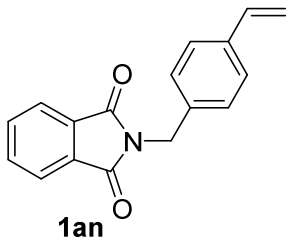
1al

***N*-Phenylphthalamic acid (1al):** Prepared according to a literature report starting from phthalic anhydride and aniline in quantitative yield.[25] ^{1}H NMR (400 MHz, DMSO-d_6): δ = 9.15 (br. s, 2H), 7.71-7.67 (m, 2H), 7.60-7.56 (m, 2H), 7.02 (dd, J = 7.3 Hz, 8.4 Hz, 2H), 6.59 (dd, J = 8.4 Hz, 1.0 Hz, 2H), 8.53 (tt, J = 7.3 Hz, 1.0 Hz, 1H) ppm. The spectral data match those previously reported.[26]



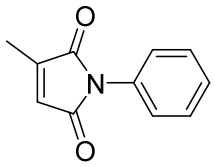
1am

2-phenyl-1*H*-benzo[*de*]isoquinoline-1,3(2*H*)-dione (1am): Prepared according to **Method A** starting from 1,8-naphthalic anhydride in 78% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 8.64 (dd, J = 7.2, 1.2 Hz, 2H), 8.26 (dd, J = 8.4 Hz, 0.8 Hz, 2H), 7.78 (dd, J = 8.0 Hz, 7.2 Hz, 2H), 7.59-7.54 (m, 2H), 7.51-7.47 (m, 1H), 7.34 (dd, J = 4.0 Hz, 1.2 Hz, 2H) ppm. The spectral data match those previously reported.[15]



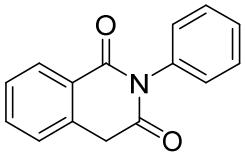
1an

***N*-(*p*-Vinylbenzyl)phthalimide (1an):** Prepared according to **Method B** starting from *p*-vinylbenzyl chloride in 77% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ= 7.83 (dd, J = 5.6 Hz, 2.8 Hz, 2H), 7.68 (dd, J = 5.6 Hz, 2.8 Hz, 2H), 7.40-7.34 (m, 4H), 6.67 (dd, J = 17.6 Hz, 11.2 Hz, 1H), 5.70 (dd, J = 17.6 Hz, 0.8 Hz, 1H), 5.22 (dd, J = 10.8 Hz, 0.8 Hz, 1H), 4.82 (s, 2H) ppm. The spectral data match those previously reported.[27]



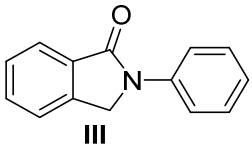
I

3-Methyl-1-phenylmaleimide (I): Prepared according to **Method A** starting from methylmaleic anhydride in 77% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.48-7.43 (m, 2H), 7.36-7.32 (m, 3H), 6.46 (q, *J* = 2.0 Hz, 1H), 2.15 (d, *J* = 2.0 Hz, 3H) ppm. The spectral data match those previously reported.[28]



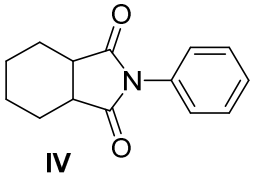
II

***N*-Phenylhomophthalimide (II):** Prepared according to **Method A** starting from homophthalic anhydride in 95% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.25 (dd, *J* = 8.0 Hz, 0.8 Hz, 1H), 7.67-7.63 (m, 1H), 7.54-7.43 (m, 4H), 7.35 (d, *J* = 7.2 Hz, 1H), 7.23-7.20 (m, 2H), 4.23 (s, 2H) ppm. The spectral data match those previously reported.[29]



III

***N*-Phenylphthalimidine (III):** Prepared according to a literature report[30] in 98% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.93 (d, *J* = 7.6 Hz, 1H), 7.89-7.86 (m, 2H), 7.62-7.58 (m, 1H), 7.53-7.49 (m, 2H), 7.43 (dd, *J* = 8.4 Hz, 7.2 Hz, 2H), 7.18 (dd, *J* = 7.2 Hz, 7.2 Hz, 1H), 4.87 (s, 2H) ppm. The spectral data match those previously reported.[31]



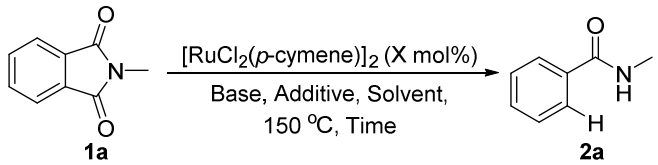
IV

Hexahydro-*N*-phenylphthalimide (IV): Prepared according to **Method D** in 86% isolated yield. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.48-7.44 (m, 2H), 7.39-7.35 (m, 1H), 7.30-7.27 (m, 2H), 3.06-3.00 (m, 2H), 1.95-1.85 (m, 4H), 1.53-1.50 (m, 4H) ppm. The spectral data match those previously reported.[32]

4. Reaction optimization.

4.1. General procedure: $[RuCl_2(p\text{-cymene})]_2$ (0.004 mmol, 2.5 mg, 0.01 eq.), potassium carbonate (1.2 mmol, 165.8 mg, 3 eq.), distilled water (0.6 mmol, 10.8 mg, 10.8 μL, 1.5 eq.), substrate **1** (0.4 mmol, 1 eq.) and *N*-methyl-2-pyrrolidone (2.0 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 ºC during six hours. Then, the reaction mixture was cooled down to room temperature and diluted with water (20 mL). Then, HCl (1 M) was added until pH reached *ca.* 7. The aqueous phase was extracted with ethyl acetate (3 x 20 mL) and the combined organic layer was dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the desired product **2** was purified by column chromatography with a mixture of petroleum ether and ethyl acetate as the eluent.

4.2. Screening of reaction conditions (Table S1).



1a **2a**

Entry[a]	X	Base	Additive	Solvent	Time (h)	Yield(%)[b]
1	5	K_2CO_3	–	NMP	24	56
2	5	K_2CO_3	KOAc	NMP	24	56
3	5	K_2CO_3	H_2O	NMP	24	99 (93)
4[c]	5	K_2CO_3	H_2O	NMP	24	35
5[d]	5	K_2CO_3	H_2O	NMP	24	trace
6	5	Na_2CO_3	H_2O	NMP	24	17
7	5	$KHCO_3$	H_2O	NMP	24	34
8	5	NEt_3	H_2O	NMP	24	12
9	5	NaOH	H_2O	NMP	24	82
10	5	K_2CO_3	H_2O	DMF	24	16
11	5	K_2CO_3	H_2O	^{t}AmOH	24	47
12	5	K_2CO_3	H_2O	Toluene	24	0
13	5	K_2CO_3	–	H_2O	24	trace
14[e]	5	K_2CO_3	H_2O	NMP	24	57
15	10	K_2CO_3	H_2O	NMP	24	79
16	2.5	K_2CO_3	H_2O	NMP	24	99
17	1	K_2CO_3	H_2O	NMP	24	99
18	0.5	K_2CO_3	H_2O	NMP	24	74
19	1	K_2CO_3	H_2O	NMP	14	99
20	***1***	***K_2CO_3***	***H_2O***	***NMP***	***6***	***99 (93)***
21	1	K_2CO_3	H_2O	NMP	3	64
22	1	K_2CO_3	H_2O	NMP	1	31
23[f]	1	K_2CO_3	H_2O	NMP	6	69
24[g]	1	K_2CO_3	H_2O	NMP	6	18
25[h]	1	K_2CO_3	H_2O	NMP	6	10

[a] Reaction conditions: **1a** (0.2 mmol), catalysts (1 mol%), Base (300 mol%) and Additive (150 mol%) are stirred in 1 mL of solvents for 6 h under Ar. [b] Determined by ^{1}H NMR spectroscopy against an internal standard (1,3,5-trimethoxybenzene). The isolated yield is shown in parentheses. [c] $RuCl_3 \bullet nH_2O$ as the catalyst. [d] $Ru_3(CO)_{12}$ as the catalyst. [e] 2 equivalent K_2CO_3. [f] Reaction performed at 130 °C. [g] Undisitillted solvent. [h] Under air.

5. Scale-up experiment.

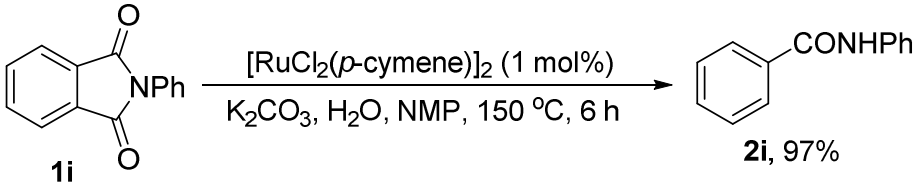


Fig. S1. Reaction studied for scale-up experiments.

$[RuCl_2(p\text{-cymene})]_2$ (0.055 mmol, 33.6 mg, 0.01 eq.), potassium carbonate (16.5 mmol, 2.28 g, 3 eq.), distilled water (8.3 mmol, 0.15 g, 0.15 mL, 1.5 eq.), substrate **1i** (5.5 mmol, 1.23 g, 1 eq.) and *N*-methyl-2-pyrrolidone (20 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 °C during six hours. Then, the reaction mixture was cooled down to room temperature and water (200 mL) was added. Then, HCl (1 M) was added until pH reached *ca.* 7. The aqueous layer was extracted with ethyl acetate and the combined organic phases were dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the desired product **2i** was obtained in 97% yield (1.05 g) by column chromatography with a mixture of petroleum ether and ethyl acetate as the eluent.

6. Mechanistic investigations.

6.1. GC-Gas analysis of the reaction mixture. $[RuCl_2(p\text{-cymene})]_2$ (0.01 mmol, 6.1 mg, 0.01 eq.), potassium carbonate (3 mmol, 414.6 mg, 3 eq.), distilled water (1.5 mmol, 27 mg, 27 µL, 1.5 eq.), substrate **1a** (1 mmol, 161 mg, 1 eq.) and *N*-methyl-2-pyrrolidone (5 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 ºC during six hours. Then, the reaction mixture was cooled down to room temperature and analysed by GC-gas analysis (see details below) indicating the major presence of H_2 and CO_2 besides traces of CH_4 and CO. Air (O_2 and N_2) was observed because the analysis could not be done under completely argon atmosphere. TLC and ^{1}H NMR spectroscopy analysis of the reaction mixture indicated the full conversion of **1a** into **2a**.

The same study was performed in a reaction lacking the substrate **1a**.

Analysis method: Gas phase chromatography apparatus µGC 3000 SRA.
Column chromatography: Molecular sieves 5A-30m.
Oven temperature: 100 ºC.
Vector gas: Helium.
Detector: Cathetometer.

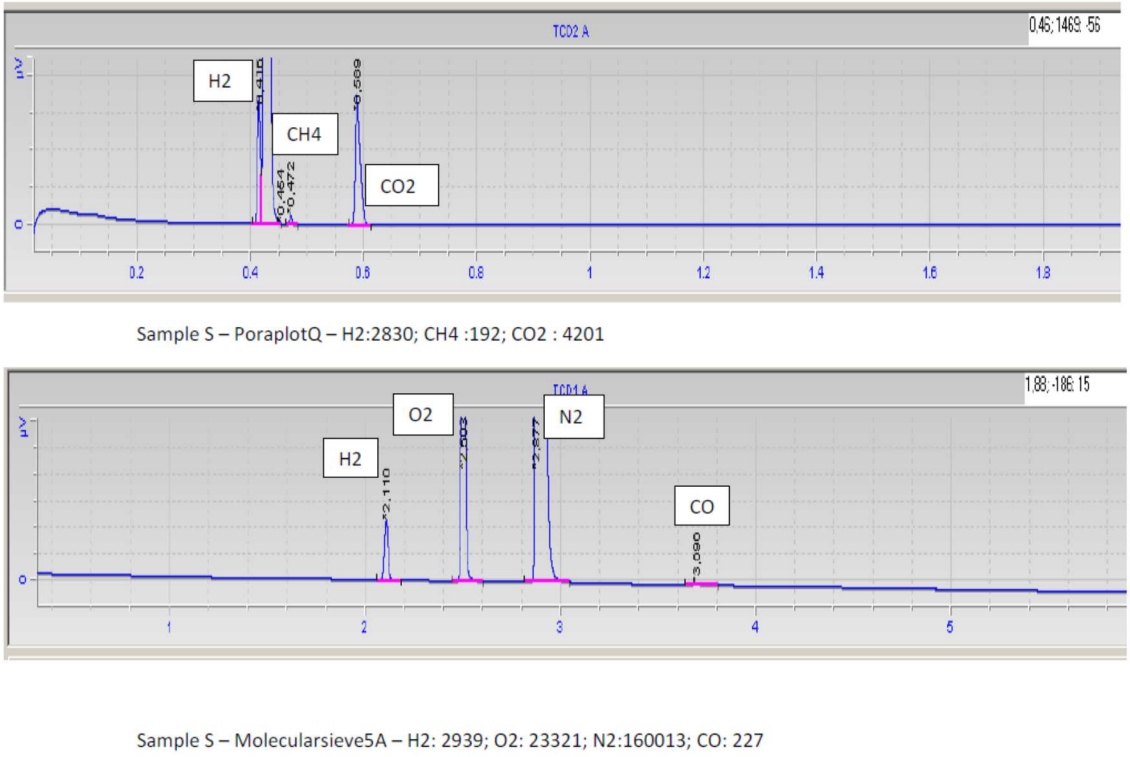


Fig. S2. GC-Gas spectrum (using two columns) of the reaction mixture.

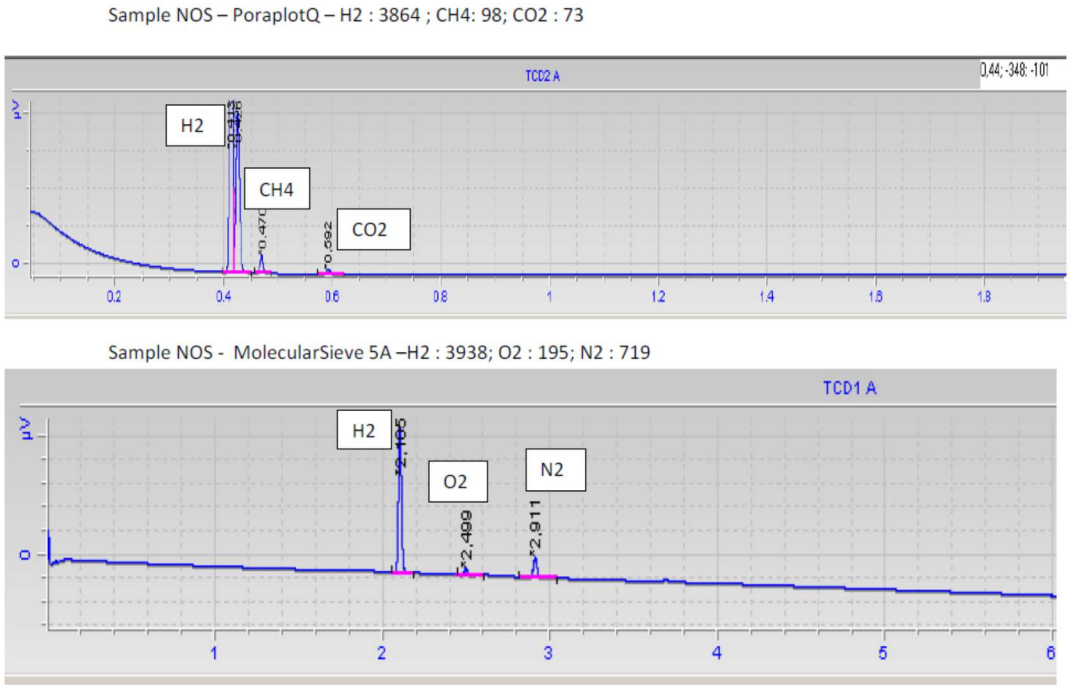


Fig. S3. GC-Gas spectrum (using two columns) of a reaction performed without substrate **1a**.

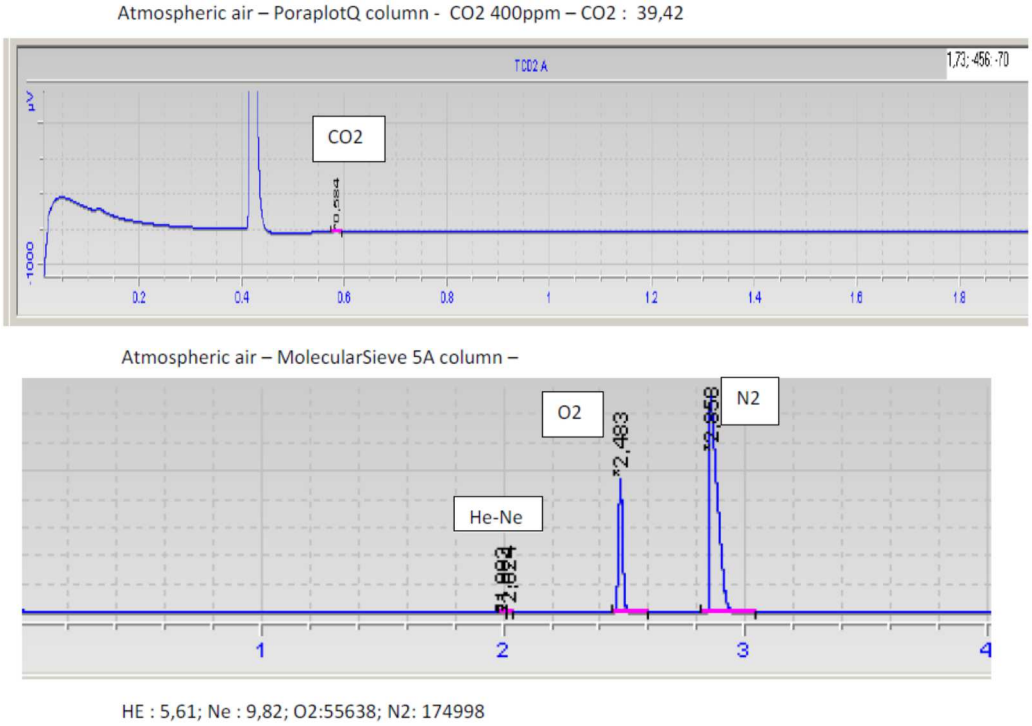


Fig. S4. GC-Gas spectrum (using two columns) of a blank analysis.

6.2. Deuteration experiments. [$RuCl_2$(*p*-cymene)]$_2$ (0.001 mmol, 0.6 mg, 0.01 eq.), potassium carbonate (0.3 mmol, 41.5 mg, 3 eq.), substrate **1a** (0.1 mmol, 16 mg, 1 eq.), *N*-methyl-2-pyrrolidone (0.9 mL) and D_2O (0.1 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 °C during six hours. Then, the reaction mixture was cooled down to room temperature and water (10 mL) was added. Then, HCl (1 M) was added until pH reached *ca.* 7. The aqueous phase was extracted with ethyl acetate (3 x 10 mL) and the combined organic layer was dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the reaction mixture was analysed without any further purification by ^{1}H NMR spectroscopy (using 1,3,5-trimethoxybenzene as internal standard) indicating the exclusive presence of product **2a**-*d* in 63% yield (see spectra below).

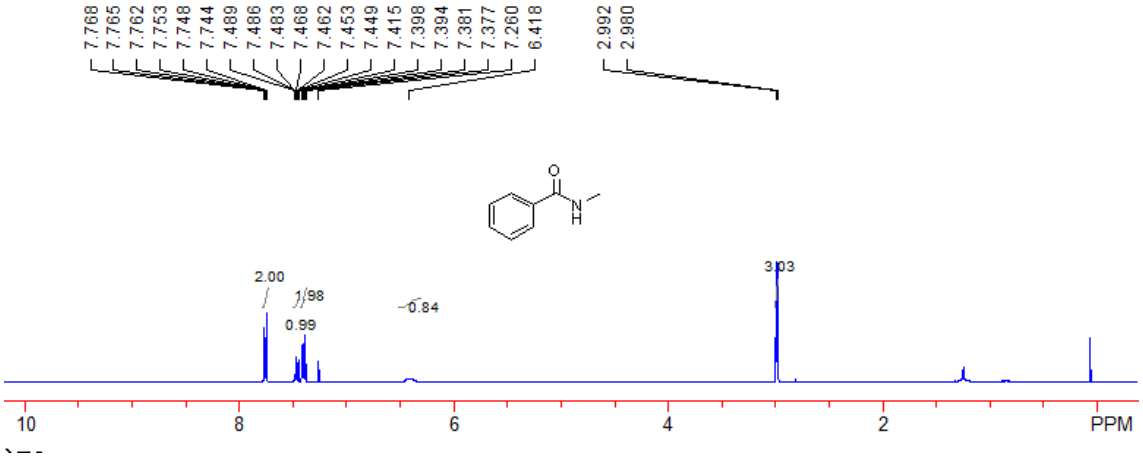


Fig. S5. ^{1}H NMR ($CDCl_3$) spectrum of **2a** in a reaction without using D_2O.

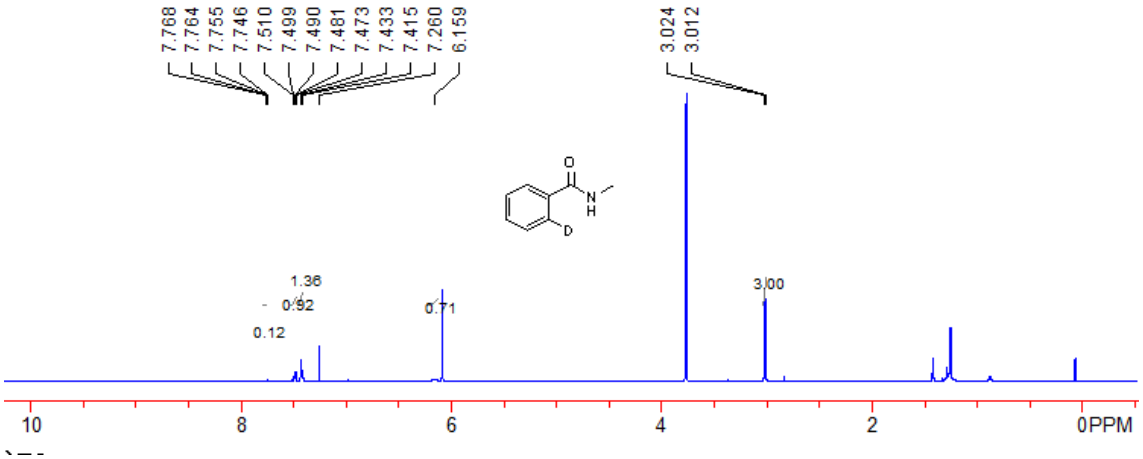


Fig. S6. ^{1}H NMR ($CDCl_3$) spectrum of **2a**-*d* showing the disappearance of the peak at 7.77-7.74 ppm.

6.3. Hydrogenation experiments. $[RuCl_2(p\text{-cymene})]_2$ (0.002 mmol, 1.2 mg, 0.01 eq.), potassium carbonate (0.6 mmol, 83 mg, 3 eq.), distilled water (0.3 mmol, 5.4 mg, 5.4 µL, 1.5 eq.), substrate **1a** (0.2 mmol, 32 mg, 1 eq.) and *N*-methyl-2-pyrrolidone (1 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. Then, the reaction mixture was flushed with vaccum/H_2 over 3 cycles. The Schlenk tube was connected to a balloon filled with H_2 and the reaction mixture was stirred at 150 °C during six hours. Then, the reaction mixture was cooled down to room temperature and water (10 mL) was added. Then HCl (1 M) was added until pH reached *ca.* 7. The aqueous phase was extracted with ethyl acetate (3 x 10 mL) and the combined organic phases were dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, ^1H NMR spectroscopy analysis (using 1,3,5-trimethoxybenzene as internal standard) indicated <10% formation of **2a** and >90% presence of **1a**.

6.4. TEMPO experiments. $[RuCl_2(p\text{-cymene})]_2$ (0.002 mmol, 1.2 mg, 0.01 eq.), potassium carbonate (0.6 mmol, 83 mg, 3 eq.), distilled water (0.3 mmol, 5.4 mg, 5.4 µL, 1.5 eq.), substrate **1a** (0.2 mmol, 32 mg, 1 eq.), TEMPO (0.2 mmol, 31 mg, 1 eq.) and *N*-methyl-2-pyrrolidone (1 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere and the reaction mixture was stirred at 150 °C during six hours. Then, the reaction mixture was cooled down to room temperature and analysed by GC-MS indicating the presence of [**1a** + (2 x TEMPO)] at *m/z* = 474 besides the main presence of **1a** (see spectra below). The reaction mixture was further diluted with water (10 mL) and HCl (1 M) was added until pH reached *ca.* 7. Then, the aqueous phase was extracted with ethyl acetate (3 x 10 mL) and the combined organic phases were dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the desired product **2a** (24% isolated yield) was purified by column chromatography with a mixture of petroleum ether and ethyl acetate as the eluent.

GC-MS results:

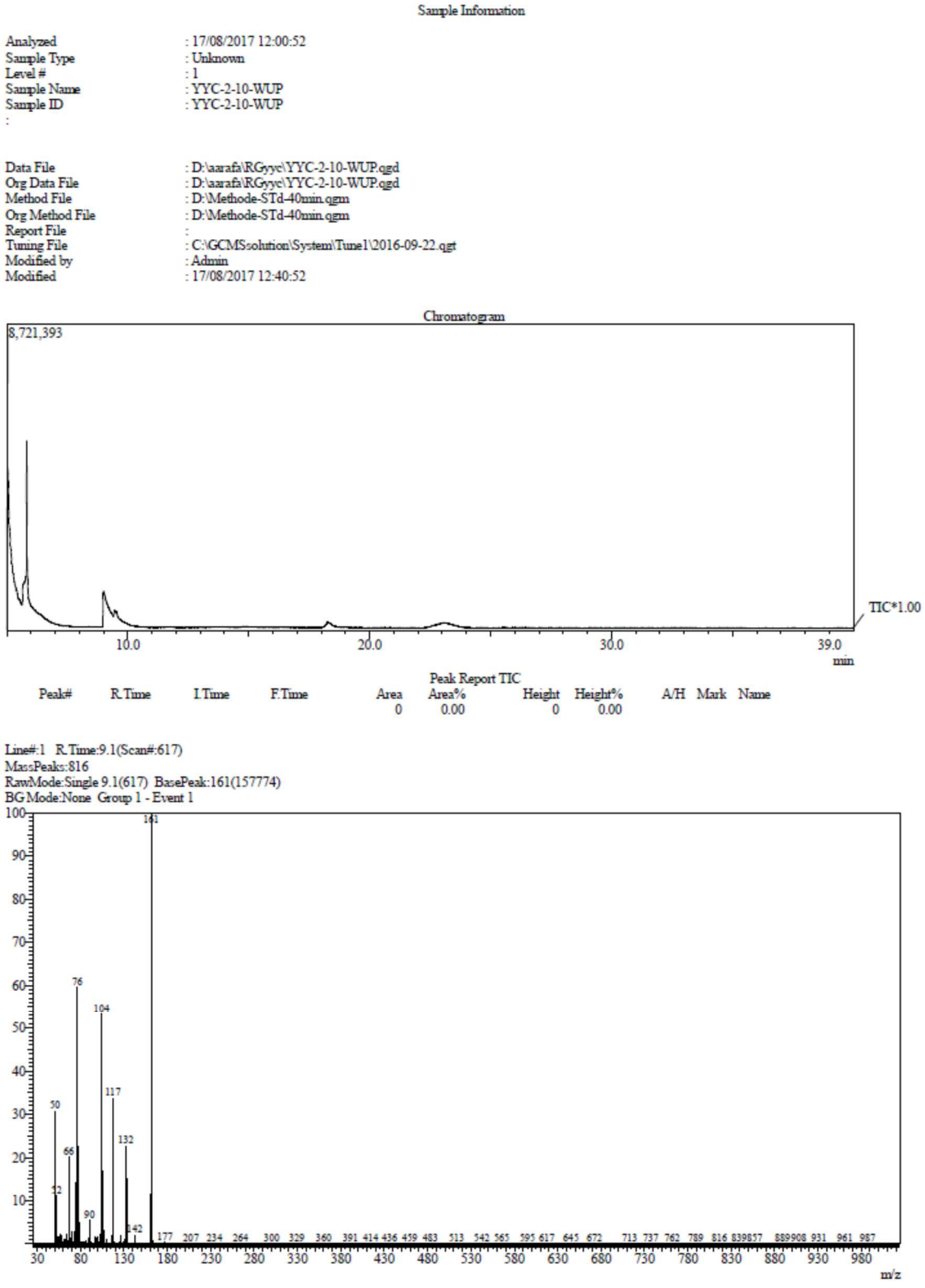


Fig. S7. GC-MS peak belonging to starting material **1a**.

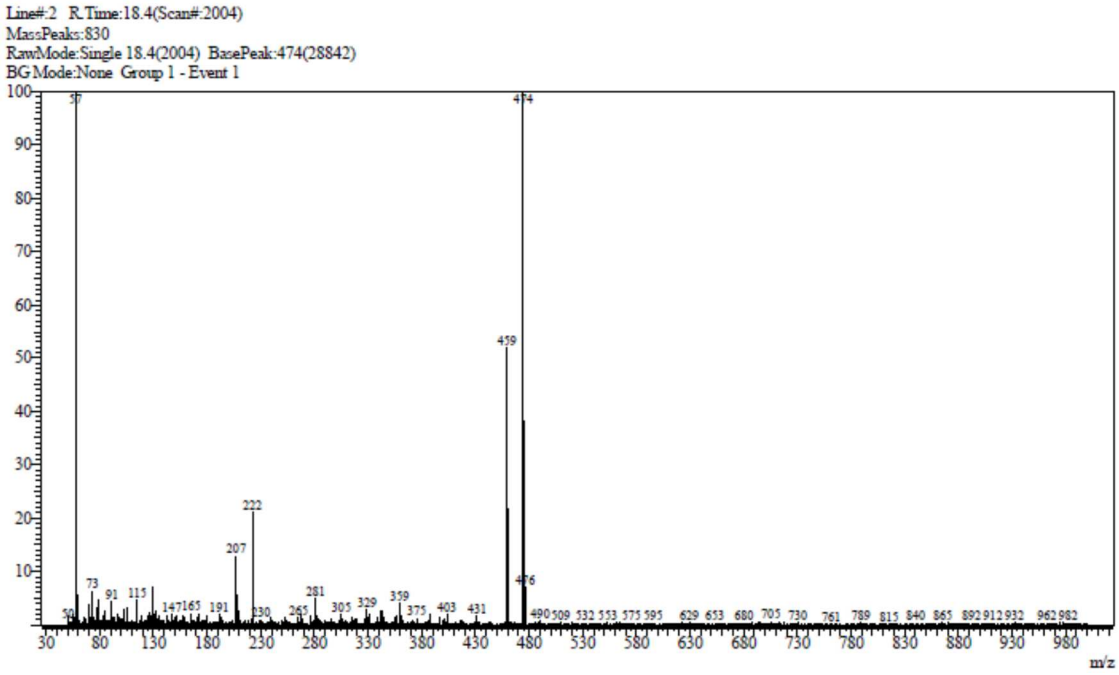


Fig. S8. GC-MS peak belonging to [**1a** + (2 x TEMPO)].

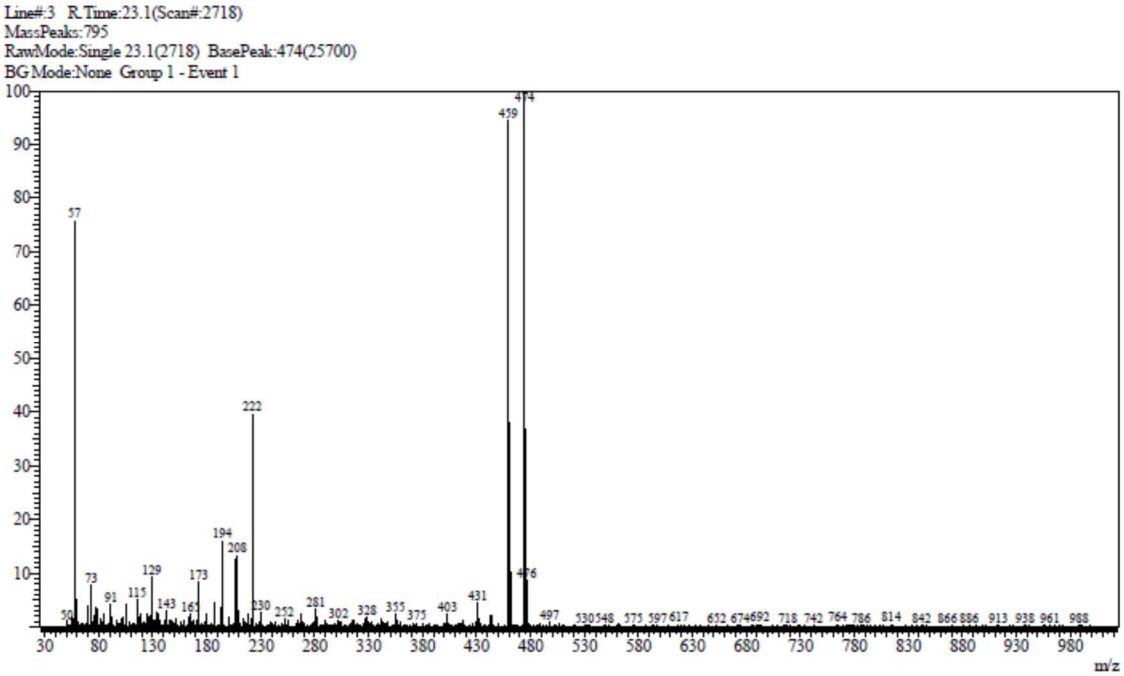


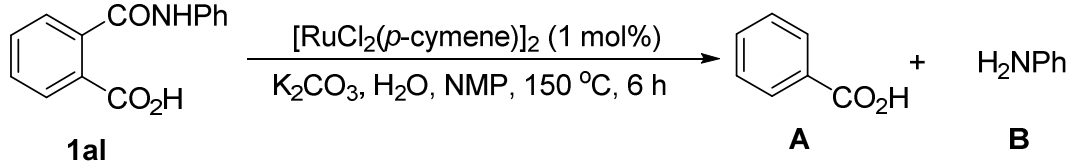
Fig. S9. GC-MS peak belonging to [**1a** + (2 x TEMPO)].

6.5. Mercury tests.

Test A: $[RuCl_2(p\text{-cymene})]_2$ (0.002 mmol, 1.2 mg, 0.01 eq.), potassium carbonate (0.6 mmol, 83 mg, 3 eq.), distilled water (0.3 mmol, 5.4 mg, 5.4 µL, 1.5 eq.), substrate **1a** (0.2 mmol, 32 mg, 1 eq.), 1 drop of mercury and *N*-methyl-2-pyrrolidone (1 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 ºC during six hours. Then, the reaction mixture was cooled down to room temperature and water (10 mL) was added. Then HCl (1 M) was added to the mixture until pH reached *ca.* 7. The aqueous phase was extracted with ethyl acetate (3 x 10 mL) and the combined organic layer was dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the desired product **2a** was obtained in 92% isolated yield by column chromatography with a mixture of petroleum ether and ethyl acetate as the eluent.

Test B: $[RuCl_2(p\text{-cymene})]_2$ (0.002 mmol, 1.2 mg, 0.01 eq.), potassium carbonate (0.6 mmol, 83 mg, 3 eq.), distilled water (0.3 mmol, 5.4 mg, 5.4 µL, 1.5 eq.), substrate **1a** (0.2 mmol, 32 mg, 1 eq.), and *N*-methyl-2-pyrrolidone (1 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 ºC during during one hour. Then, at 150 ºC and under argon atmosphere, a drop of mercury was added to the reaction mixture, which was further stirred at 150 ºC during five hours (total reaction time was six hours). Then, the reaction mixture was cooled down to room temperature and water (10 mL) was added. Then HCl (1 M) was added to the mixture until pH reached *ca.* 7. The aqueous phase was extracted with ethyl acetate (3 x 10 mL) and the combined organic layer was dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the desired product **2a** was obtained in 88% isolated yield by column chromatography with a mixture of petroleum ether and ethyl acetate as the eluent.

6.6. Study on a plausible intermediate (1al).



1al **A** **B**

$[RuCl_2(p\text{-cymene})]_2$ (0.002 mmol, 1.2 mg, 0.01 eq.), potassium carbonate (0.6 mmol, 83 mg, 3 eq.), distilled water (0.3 mmol, 5.4 mg, 5.4 µL, 1.5 eq.), substrate **1al** (0.2 mmol, 48.2 mg, 1 eq.) and *N*-methyl-2-pyrrolidone (1.0 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 °C during six hours. Then, the reaction mixture was cooled down to room temperature and diluted with water (10 mL). Then, HCl (1 M) was added until pH reached *ca.* 7. The aqueous phase was extracted with ethyl acetate (3 x 10 mL) and the combined organic layer was dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the crude mixture was analysed by ^1H NMR spectroscopy indicating the exclusive formation of benzoic acid together with aniline (see spectrum below).

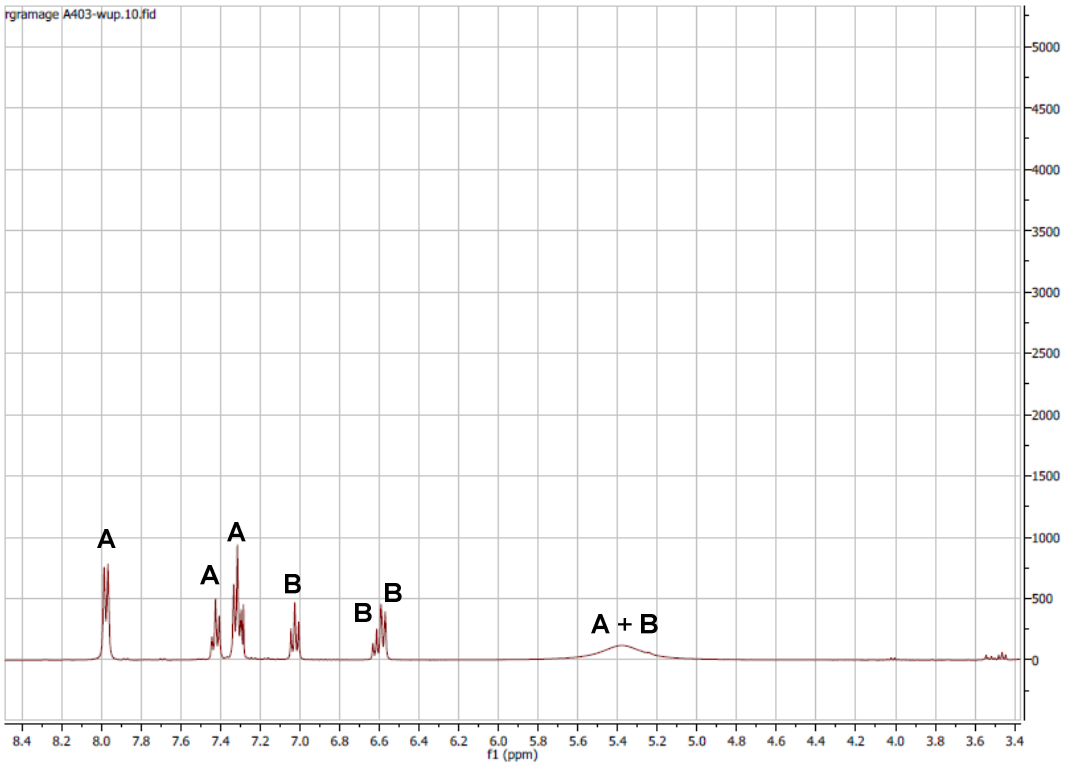
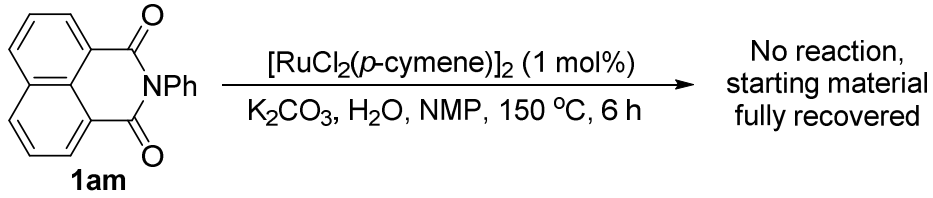


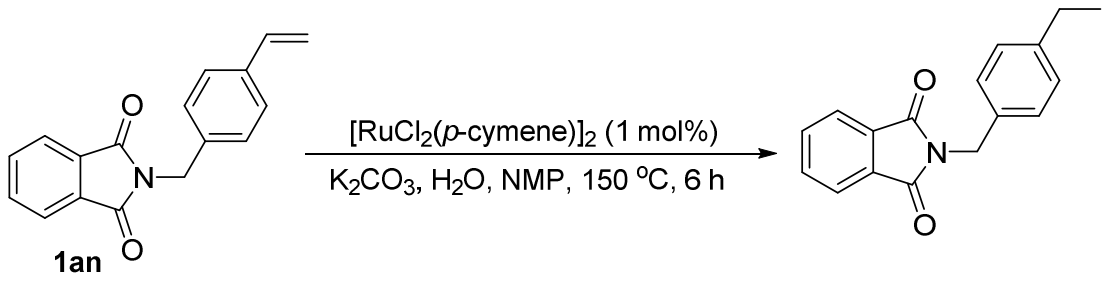
Fig. S10. 1H NMR spectrum of the reaction mixture after catalysis using substrate **1al**.

6.7. Study of the size of the phthalimide ring (1am).



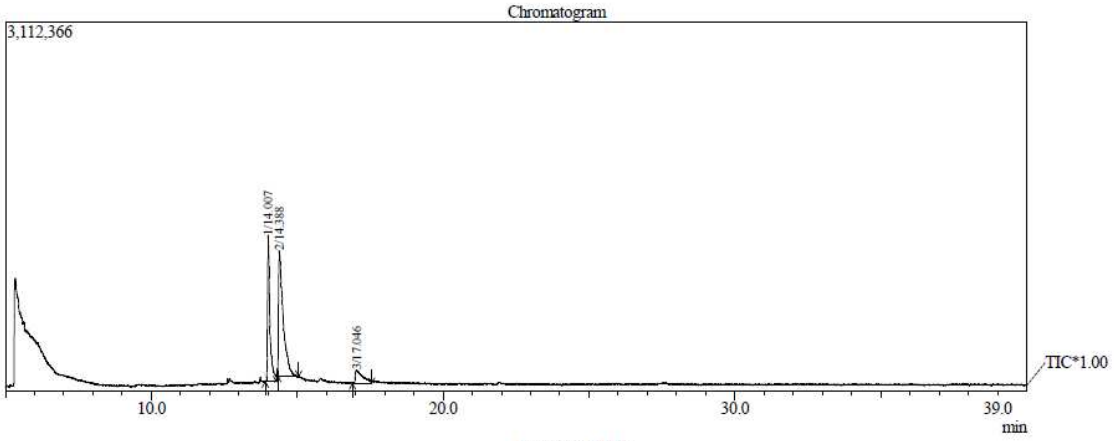
$[RuCl_2(p\text{-cymene})]_2$ (0.004 mmol, 2.5 mg, 0.01 eq.), potassium carbonate (1.2 mmol, 165.8 mg, 3 eq.), distilled water (0.6 mmol, 10.8 mg, 10.8 µL, 1.5 eq.), substrate **1am** (0.4 mmol, 109.3 mg, 1 eq.) and *N*-methyl-2-pyrrolidone (2.0 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 °C during six hours. Then, the reaction mixture was cooled down to room temperature and diluted with water (20 mL). Then, HCl (1 M) was added until pH reached *ca.* 7. The aqueous phase was extracted with ethyl acetate (3 x 20 mL) and the combined organic layer was dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the crude mixture was analysed by ^1H NMR spectroscopy indicating the exclusive presence of starting material **1am**.

6.8. Indirect evidence of hydrogen formation in the catalysis.



$[RuCl_2(p\text{-cymene})]_2$ (0.004 mmol, 2.5 mg, 0.01 eq.), potassium carbonate (1.2 mmol, 165.8 mg, 3 eq.), distilled water (0.6 mmol, 10.8 mg, 10.8 µL, 1.5 eq.), substrate **1an** (0.4 mmol, 105.3 mg, 1 eq.) and *N*-methyl-2-pyrrolidone (2.0 mL) were introduced in a flame-dried Schlenk tube under argon atmosphere. The reaction mixture was stirred at 150 °C during six hours. Then, the reaction mixture was cooled down to room temperature and diluted with water (20 mL). Then, HCl (1 M) was added until pH reached *ca.* 7. The aqueous phase was extracted with ethyl acetate (3 x 20 mL) and the combined organic layer was dried over anhydrous $MgSO_4$, filtered, and concentrated in *vacuo*. After solvents evaporation, the crude mixture was analysed by GC-MS indicating the presence of the starting material **1am** and its hydrogenated version at *m/z* = 265 (see GC-MS spectrum below).

Data File	: D:\aarafa\RGyyc\YYC-2-28-RE-2.qgd
Org Data File	: D:\aarafa\RGyyc\YYC-2-28-RE-2.qgd
Method File	: D:\Methode-STd-40min.qgm
Org Method File	: D:\Methode-STd-40min.qgm
Report File	:
Tuning File	: C:\GCMSsolution\System\Tune1\2016-09-22.qgt
Modified by	: Admin
Modified	: 14/09/2017 19:14:42



Peak Report TIC

Peak#	R.Time	I.Time	F.Time	Area	Area%	Height	Height%	A/H	Mark	Name
1	14.007	13.913	14.293	7083312	33.88	1236219	51.38	5.73	MI	
2	14.388	14.340	15.053	11719222	56.05	1058282	43.98	11.07	MI	
3	17.046	16.893	17.547	2107170	10.08	111552	4.64	18.89	MI	
				20909704	100.00	2406053	100.00			

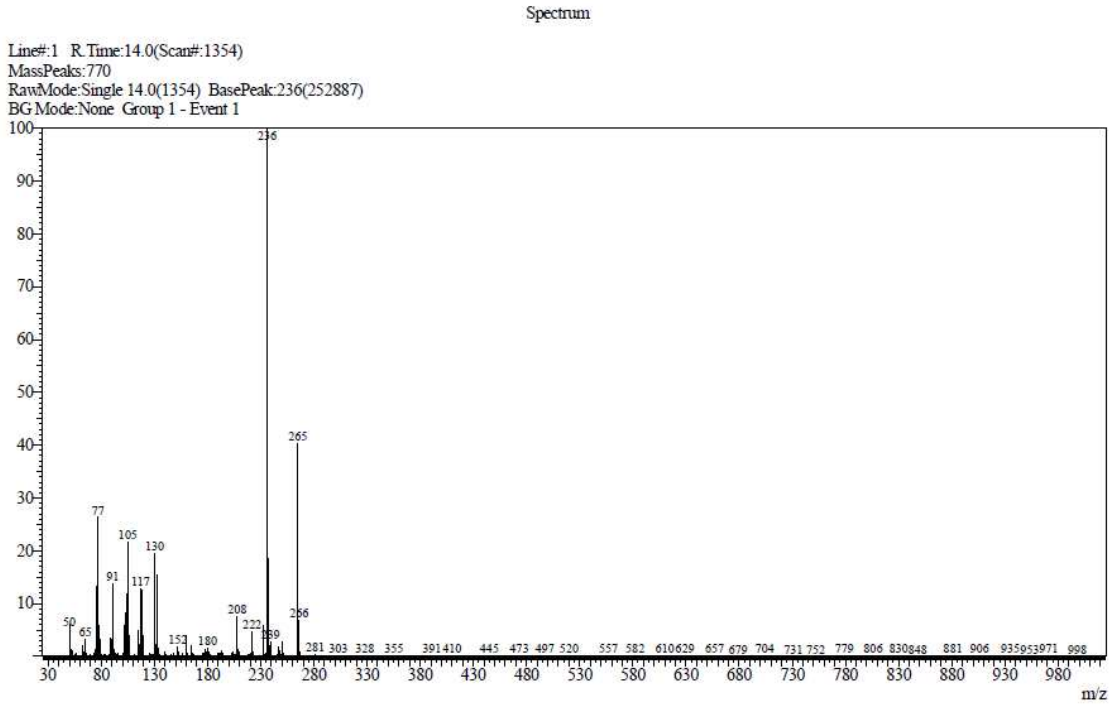
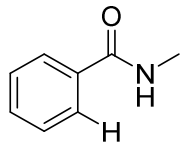


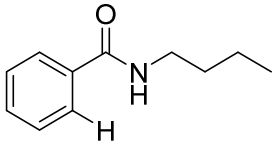
Fig. S11. GC-MS peak belonging to the hydrogenated starting material.

7. Characterization data of products 2-3.



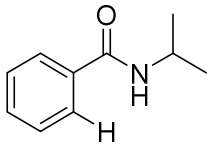
2a

***N*-Methylbenzamide (2a):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 5:1 to 2:1, *v*/*v*) in 93% yield (50.3 mg) as a colourless solid using petroleum ether. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.77-7.74 (m, 2H), 7.49-7.45 (m, 1H), 7.42-7.38 (m, 2H), 6.42 (br. s, 1H), 2.98 (d, *J* = 4.8 Hz, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 168.3, 134.5, 131.2, 128.3, 126.8, 26.7 ppm. GC: t_R = 8.7 min; MS (EI): *m/z* = 134 (M^+, 48), 105 (100), 77 (91), 51 (34). The spectral data match those previously reported.[33]



2b

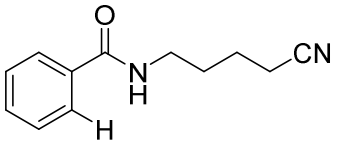
***N*-Butylbenzamide (2b):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 88% yield (62.4 mg) as a yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.76 (d, *J* = 7.6 Hz, 2H), 7.46 (dd, *J* = 7.2 Hz, 7.2 Hz, 1H), 7.39 (dd, *J* = 7.6 Hz, 7.6Hz, 2H), 6.36 (br. s, 1H), 3.43 (td, *J* = 6.8 Hz, 6.4 Hz, 2H), 1.62-1.54 (m, 2H), 1.43-1.34 (m, 2H), 0.93 (t, *J* = 7.2 Hz, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 167.5, 134.7, 131.0, 128.2, 126.8, 39.7, 31.6, 20.0, 13.6 ppm. GC: t_R = 9.9 min; MS (EI): *m/z* = 177 (M^+, 8), 105 (100), 77 (41). The spectral data match those previously reported.[34]



2c

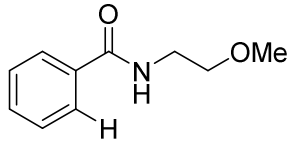
***N*-Isopropylbenzamide (2c):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 87% yield (56.8 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.76-7.73 (m, 2H), 7.50-7.46 (m, 1H), 7.44-7.39 (m, 2H), 5.94

(br. s, 1H), 4.33-4.25 (m, 1H), 1.26 (d, J = 6.4 Hz, 6H) ppm. $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ = 166.7, 135.0, 131.2, 128.5, 126.8, 41.9, 22.8 ppm. GC: t_R = 8.7 min; MS (EI): *m/z* = 163 (M^+, 25), 105 (100), 77 (39). The spectral data match those previously reported.[35]



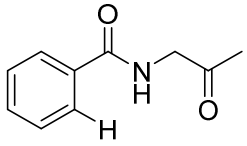
2e

***N*-(*p*-Cyanobutyl)benzamide (2e):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 91% yield (73.6 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.75 (dd, J = 8.0 Hz, 1.2 Hz, 2H), 7.48-7.43 (m, 1H), 7.39-7.35 (m, 2H), 6.80 (br. s, 1H), 3.45-3.40 (m, 2H), 2.35 (t, J = 6.8 Hz, 2H), 1.76-1.63 (m, 4H) ppm. $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ = 167.7, 134.2, 131.4, 128.4, 126.8, 119.5, 38.6, 28.6, 22.6, 16.6 ppm. HRMS (ESI) calcd. for $[M+Na]^+$ $C_{12}H_{14}N_2ONa$ 225.09983, found 225.0996 (1 ppm). The spectral data match those previously reported.[36]



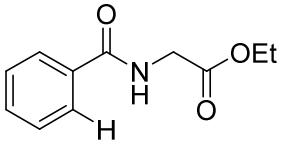
2f

***N*-(2-Methoxyethyl)benzamide (2f):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 98% yield (70.3 mg) as a colourless oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.76-7.74 (m, 2H), 7.46-7.42 (m, 1H), 7.39-7.34 (m, 2H), 6.76 (br. s, 1H), 3.62-3.58 (m, 2H), 3.52-3.50 (m, 2H), 3.33 (s, 3H) ppm. $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ = 167.4, 134.4, 131.2, 128.3, 126.8, 71.0, 58.6, 39.5 ppm. HRMS (ESI) calcd. for $[M+Na]^+$ $C_{10}H_{13}NO_2Na$ 202.08385, found 202.0837 (1 ppm). The spectral data match those previously reported.[37]



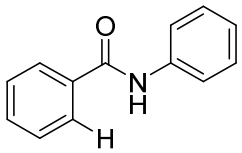
2g

***N*-(2-Oxopropyl)benzamide (2g):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 73% yield (51.2 mg) as a yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.80 (d, *J* = 6.8 Hz, 2H), 7.50-7.47 (m, 1H), 7.41 (dd, *J* = 7.6 Hz, 7.6 Hz, 2H), 7.04 (br. s, 1H), 4.31 (d, *J* = 4.4 Hz, 2H), 2.23 (s, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 203.0, 167.2, 133.6, 131.7, 128.5, 127.0, 50.2, 27.3 ppm. GC: t_R = 11.2 min; MS (EI): *m/z* = 177 (M^+, 10), 135 (45), 105 (100), 77 (55). The spectral data match those previously reported.[38]



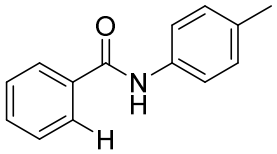
2h

Ethyl benzamidoacetate (2h): Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 50% yield (41.4 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.82-7.80 (m, 2H), 7.54-7.49 (m, 1H), 7.46-7.42 (m, 2H), 6.69 (br. s, 1H), 4.29-4.23 (m, 4H), 1.31 (t, *J* = 7.2 Hz, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 170.1, 167.4, 133.7, 131.8, 128.6, 127.0, 61.6, 41.9, 14.1 ppm. GC: t_R = 10.9 min; MS (EI): *m/z* = 207 (M^+, 7), 105 (100), 77 (35). The spectral data match those previously reported.[39]



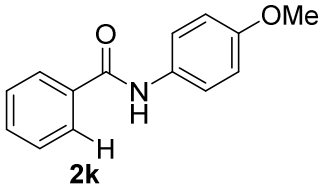
2i

***N*-Phenylbenzamide (2i):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 98% yield (77.1 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.89-7.85 (m, 3H), 7.65 (d, *J* = 7.6 Hz, 2H), 7.57-7.52 (m, 1H), 7.49-7.45 (m, 2H), 7.37 (dd, *J* = 7.6 Hz, 7.6 Hz, 2H), 7.18-7.13 (m, 1H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 165.8, 137.9, 135.0, 131.8, 129.1, 128.8, 127.0, 124.6, 120.2 ppm. GC: t_R = 12.0 min; MS (EI): *m/z* = 197 (M^+, 28), 105 (100), 77 (55). The spectral data match those previously reported.[40]



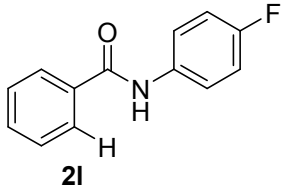
2j

***N*-(*p*-Tolyl)benzamide (2j):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 96% yield (81.4 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.86 (d, *J* = 7.2 Hz, 3H), 7.52 (d, *J* = 8.4 Hz, 3H), 7.48-7.44 (m, 2H), 7.16 (d, *J* = 8.0 Hz, 2H), 2.34 (s, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 165.6, 135.3, 135.1, 134.2, 131.7, 129.5, 128.7, 127.0, 120.3, 20.9 ppm. GC: t_R = 12.7 min; MS (EI): *m/z* = 211 (M^+, 20), 105 (100), 77 (79). The spectral data match those previously reported.[41]



2k

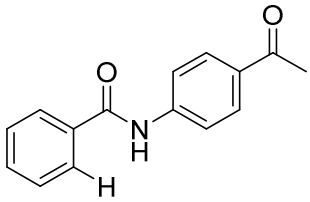
***N*-(*p*-Methoxyphenyl)benzamide (2k):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 96% yield (87.1 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.86 (d, *J* = 7.2 Hz, 2H), 7.80 (br. s, 1H), 7.55-7.52 (m, 3H), 7.47 (dd, *J* = 7.2 Hz, 7.2 Hz, 2H), 6.90 (dd, *J* = 6.8 Hz, 2.0 Hz, 2H), 3.81 (s, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 165.6, 156.6, 135.0, 131.7, 131.0, 128.7, 127.0, 122.1, 114.2, 55.5 ppm. GC: t_R = 13.6 min; MS (EI): *m/z* = 227 (M^+, 21), 105 (100), 77 (59). The spectral data match those previously reported.[41]



2l

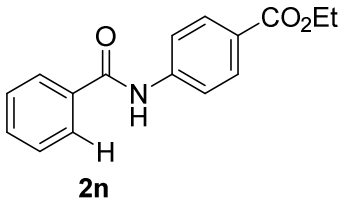
***N*-(*p*-Fluorophenyl)benzamide (2l):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 96% yield (82.3 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.79 (d, *J* = 7.6 Hz, 2H), 7.72 (br. s, 1H), 7.55-7.51 (m, 2H), 7.48 (d, *J* = 7.2 Hz, 1H), 7.44-7.40 (m, 2H), 7.00 (dd, *J* = 8.4 Hz, 8.4 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6): δ = 165.5, 158.3 (d, $J_{C\text{-}F}$ = 239.0 Hz), 135.5 (d, $J_{C\text{-}F}$ = 2.3 Hz), 134.8, 131.6, 128.4, 127.6, 122.2 (d, $J_{C\text{-}F}$ = 7.7 Hz), 115.2 (d, $J_{C\text{-}}$

$_F$ = 22.2 Hz) ppm. $^{19}F\{^1H\}$ NMR (376 MHz, $CDCl_3$): δ = -117.6 ppm. GC: t_R = 12.0 min; MS (EI): *m/z* = 215 (M^+, 12), 105 (100), 77 (80), 51 (28). The spectral data match those previously reported.[42]



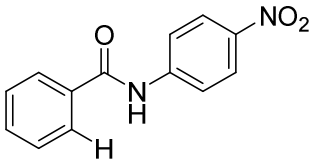
2m

4-Benzamidoacetophenone (2m): Isolated by column chromatography (SiO_2, petroleum ether/dichloromethane, 4:1 to 1:2, *v*/*v*) in 98% yield (93.4 mg) as a colourless solid. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.00 (dd, *J* = 6.8 Hz, 2.0 Hz, 3H), 7.90-7.88 (m, 2H), 7.77 (dd, *J* = 6.8 Hz, 2.0 Hz, 2H), 7.59 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H), 7.53-7.49 (m, 2H), 2.60 (s, 3H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 196.9, 165.7, 142.2, 134.5, 133.2, 132.3, 129.8, 128.9, 127.1, 119.2, 26.4 ppm. GC: t_R = 15.6 min; MS (EI): *m/z* = 239 (M^+, 8), 105 (100), 77 (69). The spectral data match those previously reported.[43]



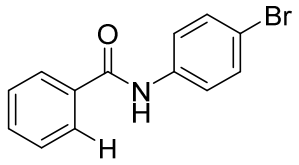
2n

Ethyl *p*-[(phenylcarbonyl)amino]benzoate (2n): Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 94% yield (100.9 mg) as a colourless solid. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.37 (br. s, 1H), 8.01 (dd, *J* = 6.8 Hz, 6.8 Hz, 2H), 7.85 (dd, *J* = 7.2 Hz, 7.2 Hz, 2H), 7.75 (dd, *J* = 7.2 Hz, 7.2 Hz, 2H), 7.52 (dd, *J* = 7.2 Hz, 7.2 Hz, 1H), 7.43 (dd, *J* = 7.2 Hz, 7.2 Hz, 2H), 4.34 (q, *J* = 7.2 Hz, 2H), 1.38 (t, *J* = 7.2 Hz, 3H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 166.2, 166.0, 142.2, 134.5, 132.1, 130.7, 128.7, 127.1, 126.0, 119.2, 60.9, 14.3 ppm. GC: t_R = 15.8 min; MS (EI): *m/z* = 269 (M^+, 19), 105 (100), 77 (38). The spectral data match those previously reported.[44]



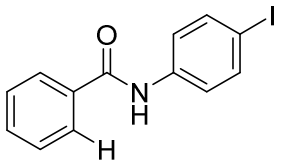
2o

***N*-(*p*-Nitrophenyl)benzamide (2o):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 80% yield (77.5 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.27 (d, *J* = 9.2 Hz, 2H), 8.05 (br. s, 1H), 7.87 (dd, *J* = 8.4 Hz, 7.6 Hz, 4H), 7.62 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H), 7.54 (dd, *J* = 7.6 Hz, 7.6 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (100 MHz, acetone-d_6): δ = 167.0, 146.4, 144.1, 135.5, 133.0, 129.4, 128.6, 125.5, 120.6 ppm. GC: t_R = 16.3 min; MS (EI): *m/z* = 242 (M^+, 10), 105 (100), 77 (53). The spectral data match those previously reported.[45]



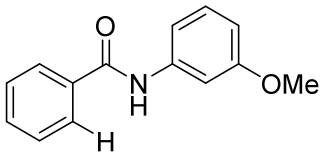
2p

***N*-(*p*-Bromophenyl)benzamide (2p):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) as a mixture of **2p**:**2i** in a ratio 74:26 according to ^{1}H NMR spectroscopy analysis. GC: t_R = 14.6 min; MS (EI): *m/z* = 275 (M^+, 10), 105 (100), 77 (45), 51 (10). The spectral data of **2p** match those previously reported.[46]



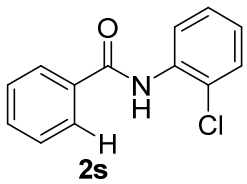
2q

***N*-(*p*-Iodophenyl)benzamide (2q):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) as a mixture of **2q**:**2i** in a ratio 24:76 according to ^{1}H NMR spectroscopy analysis. GC: t_R = 14.6 min; MS (EI): *m/z* = 323 (M^+, 30), 105 (100), 77 (50), 51 (10). The spectral data of **2q** match those previously reported.[47]



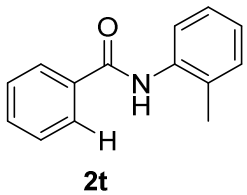
2r

***N*-(*m*-Methoxyphenyl)benzamide (2r):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 97% yield (88.5 mg) as a brown solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.22 (br. s, 1H), 7.83 (d, *J* = 7.6 Hz, 2H), 7.49 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H), 7.43-7.38 (m, 3H), 7.21 (dd, *J* = 8.0 Hz, 8.0 Hz, 1H), 7.14 (d, *J* = 7.6 Hz, 1H), 6.69 (dd, *J* = 8.0 Hz, 2.0 Hz, 1H), 3.77 (s, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 166.0, 160.1, 139.2, 134.8, 131.7, 129.6, 128.6, 127.0, 112.5, 110.4, 105.9, 55.2 ppm. GC: t_R = 13.4 min; MS (EI): *m/z* = 227 (M^+, 19), 105 (100), 77 (66). The spectral data match those previously reported.[48]



2s

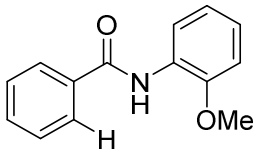
***N*-(*o*-Chlorophenyl)benzamide (2s):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 63% yield (58.3 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.48 (dd, *J* = 8.4 Hz, 0.8 Hz, 1H), 8.37 (br. s, 1H), 7.84 (d, *J* = 7.6 Hz, 2H), 7.50 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H), 7.45-7.41 (m, 2H), 7.33 (dd, *J* = 8.0 Hz, 0.8 Hz, 1H), 7.25 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H), 7.02-6.97 (m, 1H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 165.2, 134.6, 134.5, 132.1, 128.9, 128.8, 127.8, 127.0, 124.7, 123.0, 121.5 ppm. GC: t_R = 12.3 min; MS (EI): *m/z* = 231 (M^+, 5), 196 (27), 105 (100), 77 (77), 51 (27). The spectral data match those previously reported.[49]



2t

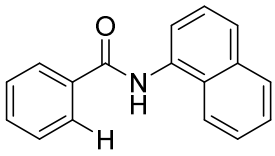
***N*-*o*-Tolylbenzamide (2t):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 89% yield (75.1 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.93-7.87 (m, 3H), 7.74 (br. s, 1H), 7.58-7.54 (m, 1H), 7.51-7.47 (m, 2H), 7.27-7.22 (m, 2H), 7.14-7.10 (m, 1H), 2.33 (s, 3H) ppm. ^{13}C{^{1}H} NMR (100

MHz, $CDCl_3$): δ = 165.7, 135.7, 134.8, 131.7, 130.5, 129.7, 128.7, 127.0, 126.7, 125.4, 123.4, 17.7 ppm. GC: t_R = 12.3 min; MS (EI): *m/z* = 211 (M^+, 28), 105 (100), 77 (50). The spectral data match those previously reported.[50]



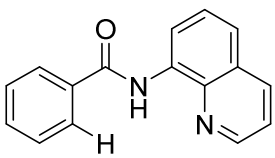
2u

***N*-(*o*-Methoxyphenyl)benzamide (2u):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 78% yield (70.9 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.55 (dd, *J* = 7.6 Hz, 1.6 Hz, 2H), 7.90 (dd, *J* = 6.8 Hz, 1.6 Hz, 2H), 7.57-7.48 (m, 3H), 7.11-7.01 (m, 2H), 6.93 (dd, *J* = 8.0 Hz, 1.2 Hz, 1H), 3.93 (s, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 165.2, 148.1, 135.3, 131.6, 128.7, 127.8, 127.0, 123.8, 121.2, 119.8, 109.9, 55.8 ppm. GC: t_R = 12.8 min; MS (EI): *m/z* = 227 (M^+, 29), 105 (100), 77 (46). The spectral data match those previously reported.[48]



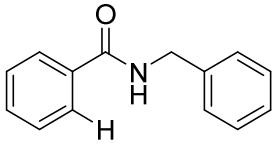
2v

***N*-(1-Naphthalenyl)benzamide (2v):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 92% yield (90.7 mg) as a pink solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.33 (br. s, 1H), 7.96-7.93 (m, 3H), 7.89-7.87 (m, 2H), 7.73 (d, *J* = 8.4 Hz, 1H), 7.57 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H), 7.51-7.45 (m, 5H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 166.3, 134.7, 134.1, 132.4, 131.8, 128.73, 128.70, 127.6, 127.2, 126.3, 126.1, 126.0, 125.6, 121.4, 120.8 ppm. GC: t_R = 16.0 min; MS (EI): *m/z* = 247 (M^+, 8), 105 (100), 77 (62). The spectral data match those previously reported.[35]



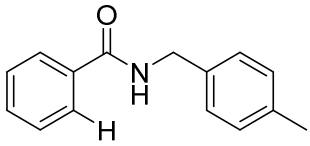
2w

***N*-8-Quinolinylbenzamide (2w):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 33% yield (32.7 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 10.75 (br. s, 1H), 8.95 (dd, *J* = 7.6 Hz, 1.6 Hz, 1H), 8.85 (dd, *J* = 4.0 Hz, 1.6 Hz, 1H), 8.18 (dd, *J* = 8.0 Hz, 1.6 Hz, 1H), 8.09 (dd, *J* = 8.0 Hz, 1.6 Hz, 2H), 7.62-7.53 (m, 5H), 7.47 (dd, *J* = 8.4 Hz, 4.0 Hz, 1H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 165.4, 148.2, 138.8, 136.4, 135.1, 134.6, 131.8, 128.8 (x 2), 128.0, 127.4, 127.3, 121.6, 116.5 ppm. GC: t_R = 15.6 min; MS (EI): *m/z* = 248 (M^+, 15), 105 (100), 77 (72). The spectral data match those previously reported.[18]



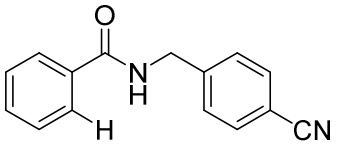
2x

***N*-Benzylbenzamide (2x):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 90% yield (76.2 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.81-7.78 (m, 2H), 7.52-7.48 (m, 1H), 7.44-7.40 (m, 2H), 7.57-7.46 (m, 4H), 7.32-7.27 (m, 1H), 6.52 (br. s, 1H), 4.64 (d, *J* = 5.6 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 167.4, 138.3, 134.2, 131.2, 128.4, 128.2, 127.5, 127.1, 126.9, 43.7 ppm. GC: t_R = 12.5 min; MS (EI): *m/z* = 211 (M^+, 47), 105 (100), 77 (57). The spectral data match those previously reported.[51] Crystals suitable for X-ray diffraction studies were grown by slow diffusion of *n*-heptane into a concentrated solution of **2x** in dichloromethane at room temperature.



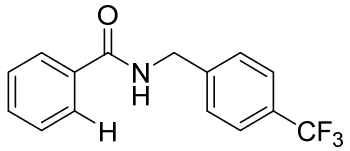
2y

***N*-*p*-Methylbenzylbenzamide (2y):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 98% yield (88.4 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.68 (d, *J* = 7.2 Hz, 2H), 7.36 (dd, *J* = 7.2 Hz, 7.2 Hz, 1H), 7.28 (dd, *J* = 7.6 Hz, 7.6 Hz, 2H), 7.11 (d, *J* = 8.0 Hz, 2H), 7.03 (d, *J* = 8.0 Hz, 2H), 6.65 (br. s, 1H), 4.45 (d, *J* = 5.6 Hz, 2H), 2.23 (s, 3H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 167.3, 137.1, 135.2, 134.4, 131.3, 129.3, 128.4, 127.8, 126.9, 43.7, 21.0 ppm. GC: t_R = 12.9 min; MS (EI): *m/z* = 225 (M^+, 24), 105 (100), 77 (60). The spectral data match those previously reported.[52]



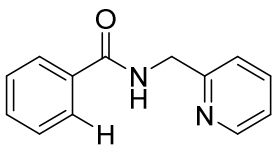
2z

***N*-[(*p*-Cyanophenyl)methyl]benzamide (2z):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 90% yield (58.2 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.79 (d, *J* = 7.2 Hz, 2H), 7.58 (dd, *J* = 6.8 Hz, 1.6 Hz, 2H), 7.54-7.49 (m, 1H), 7.44-7.40 (m, 4H), 6.89 (br. s, 1H), 4.66 (d, *J* = 6.0 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 167.6, 143.9, 133.7, 132.4, 131.9, 128.6, 128.1, 127.0, 118.7, 111.2, 43.4 ppm. GC: t_R = 15.2 min; MS (EI): *m/z* = 236 (M^+, 18), 105 (100), 77 (72), 51 (30). The spectral data match those previously reported.[53]



3a

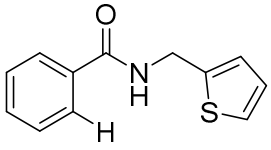
***N*-[[*p*-(Trifluoromethyl)phenyl]methyl]benzamide (3a):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 99% yield (110.9 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.80-7.78 (m, 2H), 7.56 (d, *J* = 8.4 Hz, 2H), 7.53-7.48 (m, 1H), 7.43-7.39 (m, 4H), 6.84 (br. s, 1H), 4.65 (d, *J* = 6.0 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 167.6, 142.4, 134.0, 131.7, 129.7 (q, $J_{C\text{-}F}$ = 32.9 Hz), 128.6, 127.8, 127.0, 125.6 (q, $J_{C\text{-}F}$ = 3.8 Hz), 124.0 (q, $J_{C\text{-}F}$ = 270.4 Hz), 43.4 ppm. ^{19}F{^{1}H} NMR (376 MHz, $CDCl_3$): δ = -62.5 ppm. GC: t_R = 12.3 min; MS (EI): *m/z* = 279 (M^+, 10), 105 (100), 77 (64). The spectral data match those previously reported.[54]



3b

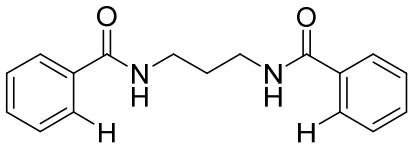
***N*-*o*-Picolylbenzamide (3b):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 88% yield (74.7 mg) as a colourless solid. ^{1}H NMR

(400 MHz, $CDCl_3$): δ = 8.54 (d, *J* = 4.4 Hz, 1H), 7.88-7.85 (m, 2H), 7.68-7.64 (m, 2H), 7.51-7.46 (m, 1H), 7.44-7.40 (m, 2H), 7.30 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H), 7.19 (dd, *J* = 7.2 Hz, 1.2 Hz, 1H), 4.74 (d, *J* = 4.8 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 167.3, 156.2, 148.9, 136.8, 134.3, 131.4, 128.5, 127.0, 122.4, 122.1, 44.7 ppm. GC: t_R = 12.7 min; MS (EI): *m/z* = 212 (M^+, 4), 107 (100), 77 (76), 51 (44). The spectral data match those previously reported.[55]



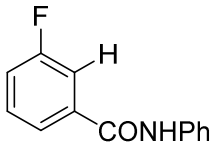
3c

***N*-(2-Thienylmethyl)benzamide (3c):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 60% yield (52.3 mg) as a colourless solid. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.79-7.77 (m, 2H), 7.51-7.47 (m, 1H), 7.41 (dd, *J* = 7.2 Hz, 7.2 Hz, 2H), 7.23 (dd, *J* = 5.2 Hz, 0.8 Hz, 1H), 7.03 (d, *J* = 6.8 Hz, 1H), 6.96 (dd, *J* = 5.2 Hz, 3.6 Hz, 1H), 6.62 (br. s, 1H), 4.80 (d, *J* = 5.6 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 167.1, 140.8, 134.1, 131.6, 128.5, 127.0, 126.9, 126.2, 125.3, 38.8 ppm. GC: t_R = 12.6 min; MS (EI): *m/z* = 217 (M^+, 42), 105 (100), 77 (44). The spectral data match those previously reported.[56]



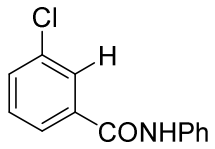
3d

***N*,*N'*-Trimethylenebis(benzamide) (3d):** Isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 93% yield (106.2 mg) as a colourless solid. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.88-7.86 (m, 4H), 7.52-7.48 (m, 2H), 7.45-7.41 (m, 4H), 7.29 (br. s, 2H), 3.58-3.53 (m, 4H), 1.84-1.78 (m, 2H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6): δ = 166.2, 134.6, 131.0, 128.2, 127.1, 37.0, 29.3 ppm. GC: t_R = 19.3 min; MS (EI): *m/z* = 282 (M^+, 8), 134 (27), 105 (100), 77 (46). The spectral data match those previously reported.[57]



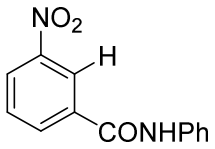
3e

***m*-Fluoro-*N*-phenylbenzamide (3e):** Starting from **1ae** and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 93% yield (80.1 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.88 (br. s, 1H), 7.64-7.56 (m, 4H), 7.48-7.43 (m, 1H), 7.41-7.35 (m, 2H), 7.27-7.24 (m, 1H), 7.22-7.14 (m, 1H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 164.2 (d, $J_{C\text{-}F}$ = 36.8 Hz), 161.6, 137.6, 137.2 (d, $J_{C\text{-}F}$ = 6.9 Hz), 130.4 (d, $J_{C\text{-}F}$ = 7.7 Hz), 129.1, 124.8, 122.4 (d, $J_{C\text{-}F}$ = 3.0 Hz), 120.3, 118.8 (d, $J_{C\text{-}F}$ = 21.4 Hz), 114.5 (d, $J_{C\text{-}F}$ = 23.0 Hz) ppm. ^{19}F{^{1}H} NMR (376 MHz, $CDCl_3$): δ = -111.3 ppm. GC: t_R = 12.6 min; MS (EI): *m/z* = 215 (M^+, 30), 123 (100), 95 (52), 75 (15). The spectral data match those previously reported.[58]



3f

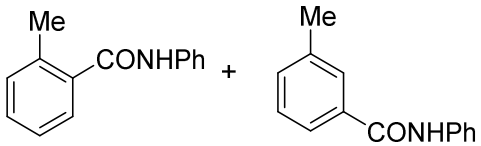
***m*-Chloro-*N*-phenylbenzamide (3f):** Starting from **1af** and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 65% yield (59.9 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.93 (br. s, 1H), 7.83 (s, 1H), 7.72 (d, *J* = 7.6 Hz, 1H), 7.62 (d, *J* = 7.6 Hz, 2H), 7.50 (d, *J* = 8.0 Hz, 1H), 7.41-7.34 (m, 3H), 7.16 (dd, *J* = 7.6 Hz, 7.6 Hz, 1H) ppm. ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 164.5, 137.6, 136.7, 134.9, 131.8, 130.0, 129.1, 127.4, 125.1, 124.9, 120.4 ppm. GC: t_R = 13.6 min; MS (EI): *m/z* = 231 (M^+, 15), 139 (100), 111 (48), 75 (27). The spectral data match those previously reported.[59]



3g

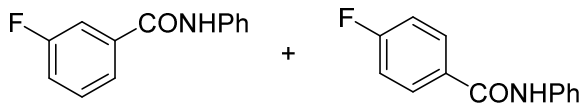
***m*-Nitro-*N*-phenylbenzamide (3g):** Starting from **1ag** and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 87% yield (87.1 mg) as a colourless solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.69 (s, 1H), 8.39 (d, *J* = 8.0

Hz, 1H), 8.25 (d, *J* = 7.6 Hz, 1H), 8.01 (br. s, 1H), 7.71-7.64 (m, 3H), 7.39 (dd, *J* = 7.6 Hz, 7.6 Hz, 2H), 7.20 (dd, *J* = 7.2 Hz, 7.2 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 163.3, 148.2, 137.2, 136.6, 133.4, 130.1, 129.2, 126.4, 125.3, 121.8, 120.5 ppm. GC: t_R = 15.2 min; MS (EI): *m/z* = 242 (M^+, 43), 212 (26), 150 (100), 120 (79), 104 (45), 92 (51), 65 (30). The spectral data match those previously reported.[60]



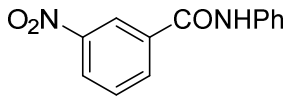
***o*-3h** ***m*-3h**

o*- and *m*-methyl-*N*-phenylbenzamide (3h):** Starting from **1ah** and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 92% yield (77.8 mg) as a mixture of isomers in a 38:62 ratio of ***o*-3h**:m*-3h** according to ^1H NMR spectroscopy analysis. GC: t_R = 12.9 min (both isomers appear together); MS (EI): *m/z* = 211 (M^+, 10), 119 (100), 91 (50), 65 (30). The spectral data match those previously reported.[61]



***m*-3i** ***p*-3i**

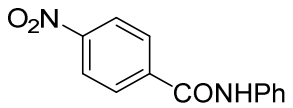
m*- and *p*-Fluoro-*N*-phenylbenzamide (3i):** Starting from **1ai** and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 82% yield (70.9 mg) as a mixture of isomers in a 59:41 ratio of ***m*-3i**:p*-3i** according to ^{19}F NMR spectroscopy analysis. GC: t_R = 12.1 min (both isomers appear together); MS (EI): *m/z* = 215 (M^+, 25), 123 (100), 95 (40), 75 (10). The spectral data match those previously reported.[47,62]



***m*-3j**

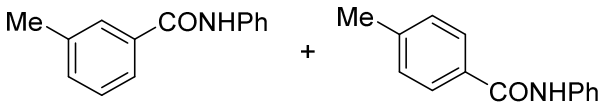
***m*-Nitro-*N*-phenylbenzamide (*m*-3j):** Starting from **1aj** and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 22% yield (21.3 mg) as a colourless solid. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.69 (s, 1H), 8.39 (d, *J* = 8.0 Hz, 1H), 8.25 (d, *J* = 7.6 Hz, 1H), 8.01 (br. s, 1H), 7.71-7.64 (m, 3H), 7.39 (dd, *J* = 7.6

Hz, 7.6 Hz, 2H), 7.20 (dd, J = 7.2 Hz, 7.2 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 163.3, 148.2, 137.2, 136.6, 133.4, 130.1, 129.2, 126.4, 125.3, 121.8, 120.5 ppm. GC: t_R = 15.2 min; MS (EI): *m/z* = 242 (M^+, 43), 212 (26), 150 (100), 120 (79), 104 (45), 92 (51), 65 (30). The spectral data match those previously reported.[60]



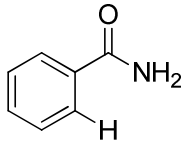
***p*-3j**

***p*-Nitro-*N*-phenylbenzamide (*p*-3j):** Starting from **1aj** and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 27% yield (25.7 mg) as a colourless solid. ^1H NMR (400 MHz, acetone-d_6): δ = 9.82 (br. s, 1H), 8.37 (d, J = 8.8 Hz, 2H), 8.24 (d, J = 8.8 Hz, 2H), 7.84 (d, J = 8.0 Hz, 2H), 7.38 (dd, J = 8.0 Hz, 8.0 Hz, 2H), 7.16 (dd, J = 7.6 Hz, 7.6 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, acetone-d_6): δ = 164.7, 150.6, 142.0, 139.9, 129.8, 129.6, 125.1, 124.4, 121.2 ppm. GC: t_R = 15.2 min; MS (EI): *m/z* = 242 (M^+, 42), 212 (27), 150 (100), 120 (78), 104 (45), 92 (51), 65 (30). The spectral data match those previously reported.[61]



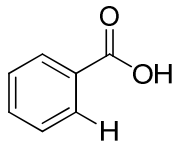
***m*-3k** ***p*-3k**

m*- and *p*-methyl-*N*-phenylbenzamide (3k):** Starting from **1ak** and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 10:1 to 2:1, *v*/*v*) in 98% yield (89.0 mg) as a mixture of isomers in a 57:43 ratio of ***m*-3k**:p*-3k** according to ^1H NMR spectroscopy analysis. GC: t_R = 12.9 min (both isomers appear together); MS (EI): *m/z* = 211 (M^+, 10), 119 (100), 91 (50), 65 (30). The spectral data match those previously reported.[61,63]



Benzamide: Starting from phthalimide and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 5:1 to 1:1, *v*/*v*) in 17% yield (8.3 mg) as a colourless solid. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.83-7.80 (m, 2H), 7.54-7.50 (m, 1H), 7.46-7.42 (m,

2H), 6.26 (br. s, 2H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 169.7, 133.4, 131.9, 128.6, 127.3 ppm. GC: t_R = 8.8 min; MS (EI): *m/z* =121 (M^+, 46), 105 (62), 77 (100), 51 (51). The spectral data match those previously reported.[64]



Benzoic acid: Starting from phthalic anhydride and isolated by column chromatography (SiO_2, petroleum ether/ethyl acetate, 4:1 to 1:2, *v*/*v*) in 51% yield (25.1 mg) as a colourless solid. ^1H NMR (400 MHz, $CDCl_3$): δ = 12.08 (br. s, 1H), 8.16-8.13 (m, 2H), 7.65-7.61 (m, 1H), 7.52-7.47 (m, 2H) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 172.5, 133.8, 130.2, 129.3, 128.5 ppm. GC: t_R = 7.0 min; MS (EI): *m/z* =122 (M^+, 71), 105 (94), 77 (100), 51 (76). The spectral data match those previously reported.[65]

8. References.

[1] S. Takebayashi, J. M. John, S. H. Bergens, *J. Am. Chem. Soc.* **2010**, *132*, 12832-12834.

[2] J.-C. Hsieh, C.-H. Cheng, *Chem. Commun.* **2005**, *36*, 4554-4556.

[3] G. Ding, C. Li, Y. Shen, B. Lu, Z. Zhang, X. Xie, *Adv. Synth. Catal.* **2016**, *358*, 1241-1250.

[4] S. Liu, Q. Deng, W. Fang, J.-F. Gong, M.-P. Song, M. Xua, T. Tu, *Org. Chem. Front.* **2014**, *1*, 1261-1265.

[5] L. Zhang, Y. Li, L.-Y. Jin, F. Liang, *RSC Adv.* **2015**, *5*, 65600-65603.

[6] P. Gupta, B. A. Shah, R. Parshad, G. N. Qazi, S. C. Taneja, *Green Chem.* **2007**, *9*, 1120-1125.

[7] P. Dawar, M. B. Raju, R. A. Ramakrishna, *Tetrahedron Lett.* **2011**, *52,* 4262-4265.

[8] L. K. Rasmussen, M. Begtrup, T. Ruhland, *J. Org. Chem.* **2004**, *69*, 6890-6893.

[9] V. Kumar, G. S. Banker, *Int. J. Pharm.* **1992**, *79*, 61-65.

[10] R. Shrestha, P. Mukherjee, Y. Tan, Z. C. Litman, J. F. Hartwig, *J. Am. Chem. Soc.* **2013**, *135*, 8480-8483.

[11] S. A. Worlikar, R. C. Larock, *J. Org. Chem.* **2008**, *73*, 7175-7180.

[12] B. Nammalwar, N. P. Muddala, F. M. Watts, R. A. Bunce, *Tetrahedron* **2015**, *71*, 9101-9111.

[13] C. D. Chu, Y. H. Qi, W. Hao, *Catal. Commun.* **2007**, *8*, 1527-1530.

[14] S. L. Yedage, D. S. D'silva, B. M. Bhanage, *RSC Adv.* **2015**, *5*, 80441-80449.

[15] H. J. Kim, J. Kim, S. H. Cho, S. Chang, *J. Am. Chem. Soc.* **2011**, *133*, 16382-16385.

[16] B. Khan, A. A. Khan, R. Kant, D. Koley, *Adv. Synth. Catal.* **2016**, *358*, 3753-3758.

[17] M. A. Ali, S. K. Moromi, A. S. Touchy, K. Shimizu, *ChemCatChem* **2016**, *8*, 891-894.

[18] S. Inoue, H. Shiota, Y. Fukumoto, N. Chatani, *J. Am. Chem. Soc.* **2009**, *131*, 6898-6899.

[19] M. Wang, J. Lu, J. Ma, Z. Zhang, F. Wang, *Angew. Chem. Int. Ed.* **2015**, *54*, 14061-14065.

[20] M. V. de Almeida, F. M. Teixeira, M. V. N. de Souza, G. W. Amarante, C. C. de S. Alves, S. H. Cardoso, A. M. Mattos, A. P. Ferreira, H. C. Teixeira, *Chem. Pharm. Bull.* **2007**, *55*, 223-226.

[21] S. D. Sarkar, L. Ackermann, *Chem. Eur. J.* **2014**, *20*, 13932-13936.

[22] F. J. Williams, P. E. Donahue, *J. Org. Chem.* **1977**, *42*, 3414-3419.

[23] Y. Shibata, K. Sasaki, Y. Hashimoto, S. Iwasaki, *Chem. Pharm. Bull.* **1996**, *44*, 156-162.

[24] X.-F. Dong, J. Fan, X.-Y. Shi, K.-Y. Liu, P.-M. Wang, J.-F. Wei, *J. Organomet. Chem.* **2015**, *779*, 55-61.

[25] A. M. Al-Azzawi, M. S. A. Al-Razzak, *Int. J. Res. Pharm. Chem.* **2013**, *3*, 682-690.

[26] W. S. Sun, Y. S. Park, J. Yoo, K. D. Park, S. H. Kim, J.-H. Kim, H.-J. Park, *J. Med. Chem.* **2003**, *46*, 5619-5627.

[27] X. Sun, X. Li, S. Song, Y. Zhu, Y.-F. Liang, N. Jiao, *J. Am. Chem. Soc.* **2015**, *137*, 6059-6066.

[28] N. Matuszak, G. G. Muccioli, G. Labar, D. M. Lambert, *J. Med. Chem.* **2009**, *52*, 7410-7420.

[29] C.-Y. Cheng, H.-B. Tsai, M.-S. Lin, *J. Heterocyclic Chem.* **1995**, *32*, 73-77.

[30] S. Wang, H. Huang, C. Bruneau, C. Fischmeister, *ChemSusChem* DOI: 10.1002/cssc.201701299.

[31] V. Kumar, S. Sharma, U. Sharma, B. Singh, N. Kumar, *Green Chem.* **2012**, *14*, 3410-3414.

[32] K. Kaminski, B. Wiklik, J. Obniska, *Arch. Pharm. Chem. Life Sci.* **2014**, *347*, 840-852.

[33] A. B. Charette, M. Grenon, A. Lemire, M. Pourashraf, J. Martel, *J. Am. Chem. Soc.* **2001**, *123*, 11829-11833.

[34] L. Ackermann, A. V. Lygin, N. Hofmann, *Angew. Chem. Int. Ed.* **2011**, *50*, 6379-6382.

[35] L. Zhang, S. Su, H. Wu, S. Wang, *Tetrahedron*, **2009**, *65*, 10022-10024.

[36] F. Foubelo, F. Lloret, M. Yus, *Anal. Quimica* **1995**, *91*, 260-266.

[37] T. Higuchi, R. Tagawa, A. Iimuro, S. Akiyama, H. Nagae, K. Mashima, *Chem. Eur. J.* **2017**, *23*, 12795-12804.

[38] (a) U. Bratusek, S. Recnik, J. Svete, L. Golic, B. Stanovnik *Heterocycles* **2002**, *57*, 2045-2064. (b) O. A. Egorova, H. Seo, Y. Kim, D. Moon, Y. M. Rhee, K. H. Ahn, *Angew. Chem. Int. Ed.* **2011**, *50*, 11446-11450.

[39] W.-J. Yoo, C.-J. Li, *J. Am. Chem. Soc.* **2006**, *128*, 13064-13065.

[40] T. B. Halima, J. K. Vandavasi, M. Shkoor, S. G. Newman, *ACS Catal.* **2017**, *7*, 2176-2180.

[41] C. K. Lee, J. S. Yu, Y. R. Ji, *J. Heterocyclic Chem.* **2002**, *39*, 1219-1223.

[42] S. Ueda, H. Nagasawa, *J. Org. Chem.* **2009**, *74*, 4272-4277.

[43] W.-T. Xu, B. Huang, J.-J. Dai, J. Xu, H.-J. Xu, *Org. Lett.* **2016**, *18*, 3114-3117.

[44] K. Inamoto, M. Shiraishi, K. Hiroya, T. Doi, *Synthesis* **2010**, *18*, 3087-3090.

[45] P. Wójcik, V. Rosar, A. Gniewek, B. Milani, A. M. Trzeciak, *J. Mol. Catal. A: Chemical* **2016**, *425*, 322-331.

[46] K. Sasaki, D. Crich, *Org. Lett.* **2011**, 13, 2256-2259.

[47] Z. Wang, W. Fan, G.-J. Deng, W. Zhou, *Tetrahedron Lett.* **2015**, *56*, 5449-5452.

[48] H. Xu, C. Wolf, *Chem. Commun.* **2009**, 1715-1717.

[49] G. Evindar, R. A. Batey, *J. Org. Chem.* **2006**, *71*, 1802-1808.

[50] T. Miura, Y. Takahashi, M. Murakami, *Chem. Commun.* **2007**, 3577-3579.

[51] L. U. Nordstrøm, H. Vogt, R. Madsen, *J. Am. Chem. Soc.* **2008**, *130*, 17672-17673.

[52] X. Cui, Y. Zhang, F. Shi, Y. Deng, *Chem. Eur. J.* **2011**, *17*, 1021-1028.

[53] G. A. Molander, M.-A. Hiebel, *Org. Lett.* **2010**, *12*, 4876-4879.

[54] P. Biallas, A. P. Häring, S. F. Kirsch, *Org. Biomol. Chem.* **2017**, *15*, 3184-3187.

[55] H. Sheng, R. Zeng, W. Wang, S. Luo, Y. Feng, J. Liu, W. Chen, M. Zhu, Q. Guo, *Adv. Synth. Catal.* **2017**, *359*, 302-313.

[56] V. Rajkumar, Naveen, S. A. Babu, *ChemistrySelect* **2016**, *6*, 1207-1219.

[57] Y. Zhu, C. Li, A. O. Biying, M. Sudarmadji, A. Chen, D. T. Tuan, A. M. Seayad, *Dalton Trans.* **2011**, *40*, 9320-9325.

[58] R. Sharma, R. A. Vishwakarma, S. B. Bharate, *Adv. Synth. Catal.* **2016**, *358*, 3027-3033.

[59] J. R. Martinelli, D. A. Watson, D. M. M. Freckmann, T. E. Barder, S. L. Buchwald, *J. Org. Chem.* **2008**, *73*, 7102-7107.

[60] M. A. Ali, P. Saha, T. Punniyamurthy, *Synthesis* **2010**, *6*, 908-910.

[61] Y. Wang, D. Zhu, L. Tang, S. Wang, Z. Wang, *Angew. Chem. Int. Ed.* **2011**, *50*, 8917-8921.

[62] G. Hong, D. Mao, X. Zhu, S. Wu, L. Wang, *Org. Chem. Front.* **2015**, *2*, 985-989.

[63] G. Meng, P. Lei, M. Szostak, *Org. Lett.* **2017**, *19*, 2158-2161.

[64] C. Wu, X. Xin, Z.-M. Fu, L.-Y. Xie, K.-J. Liu, Z. Wang, W. Li, Z.-H. Yuan, W.-M. He, *Green Chem.* **2017**, *19*, 1983-1989.

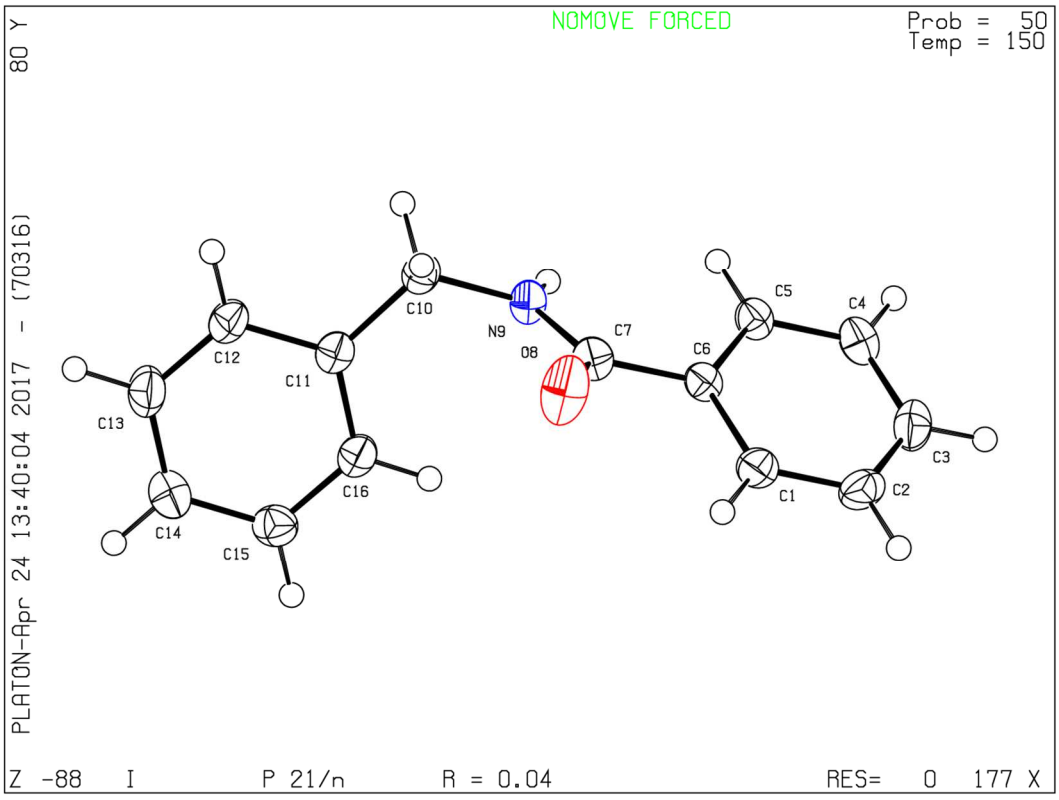
[65] T. Hattori, H. Okami, T. Ichikawa, S. Mori, Y. Sawama, Y. Monguchi, H. Sajiki, *Adv. Synth. Catal.* DOI: 10.1002/adsc.201700774.

9. X-ray crystallographic data for 2x (CCDC-1577926).

(C_{14} H_{13} N O); M = 211.25. APEXII, Bruker-AXS diffractometer, Mo-Kα radiation (λ = 0.71073 Å), T = 150 K; monoclinic $P\,2_1/n$ (I.T.#14), a = 7.9741(8), b = 24.748(3), c = 5.7767(5) Å, β = 90.056(5) °, V = 1140.00(19) Å^3. Z = 4, d = 1.231 g.cm^{-3}, μ = 0.078 mm^{-1}. The structure was solved by dual-space algorithm using the *SHELXT* program [1], and then refined with full-matrix least-square methods based on F^2 (*SHELXL*) [2]. All non-hydrogen atoms were refined with anisotropic atomic displacement parameters. Except nitrogen linked hydrogen atoms that were introduced in the structural model through Fourier difference maps analysis, H atoms were finally included in their calculated positions. A final refinement on F^2 with 2544 unique intensities and 149 parameters converged at ωR(F^2) = 0.1120 (R(F) = 0.0439) for 2221 observed reflections with I > 2σ(I).

[1] G. M. Sheldrick, Acta Cryst. A71 (2015) 3-8.

[2] Sheldrick G.M., Acta Cryst. C71 (2015) 3-8.



9.1. Structural data for 2x.

Empirical formula	$C_{14}\ H_{13}\ N\ O$
Formula weight	211.25
Temperature	150 *K*
Wavelength	0.71073 Å
Crystal system, space group	monoclinic, $P\ 2_1/n$
Unit cell dimensions	a = 7.9741(8) Å, α = 90 ° b = 24.748(3) Å, β = 90.056(5) ° c = 5.7767(5) Å, γ = 90 °
Volume	1140.00(19) $Å^3$
Z, Calculated density	4, 1.231 ($g.cm^{-3}$)
Absorption coefficient	0.078 mm^{-1}
F(000)	448
Crystal size	0.460 x 0.330 x 0.210 mm
Crystal color	colourless
Theta range for data collection	3.039 to 27.473 °
h_min, h_max	-10, 10
k_min, k_max	-32, 21
l_min, l_max	-6, 7
Reflections collected / unique	7868 / 2544 [R(int)[a] = 0.0330]
Reflections [I>2σ]	2221
Completeness to theta_max	0.998
Absorption correction type	multi-scan
Max. and min. transmission	0.984 , 0.892
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	2544 / 0 / 149
[b]S (Goodness-of-fit)	1.069
Final *R* indices [I>2σ]	$R1^c$ = 0.0439, $wR2^d$ = 0.1120
R indices (all data)	$R1^c$ = 0.0519, $wR2^d$ = 0.1171
Largest diff. peak and hole	0.187 and -0.206 $e^-.Å^{-3}$

$^a R_{int} = \sum |F_o^2 - < F_o^2 >| / \sum [F_o^2]$

$^b S = \{\sum [w(F_o^2 - F_c^2)^2] / (n - p)\}^{1/2}$

$^c R1 = \sum | |F_o| - |F_c| | / \sum |F_o|$

$^d wR2 = \{\sum [w(F_o^2 - F_c^2)^2] / \sum [w(F_o^2)^2]\}^{1/2}$

$w = 1 / [\sigma(F_o^2) + aP^2 + bP]$ where $P = [2F_c^2 + MAX(F_o^2, 0)] /3$

9.2. Atomic coordinates, site occupancy (%) and equivalent isotropic displacement parameters (Å2). U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

Atom	x	y	z	occ.	U(eq)
C1	0.3255(2)	0.34549(6)	0.3349(3)	1	0.0285(4)
H1	0.391412	0.327271	0.222448	1	0.034
C2	0.2761(2)	0.39830(7)	0.2968(3)	1	0.0336(4)
H2	0.308436	0.416411	0.158844	1	0.040
C3	0.1793(2)	0.42470(7)	0.4605(3)	1	0.0341(4)
H3	0.146982	0.461188	0.435725	1	0.041
C4	0.1295(2)	0.39827(7)	0.6599(3)	1	0.0311(4)
H4	0.061139	0.416355	0.769875	1	0.037
C5	0.1794(2)	0.34538(6)	0.6992(3)	1	0.0255(4)
H5	0.145689	0.327260	0.836479	1	0.031
C6	0.27898(19)	0.31889(6)	0.5372(3)	1	0.0226(4)
C7	0.3465(2)	0.26299(6)	0.5745(3)	1	0.0259(4)
O8	0.47488(17)	0.24814(5)	0.4742(3)	1	0.0487(4)
N9	0.26593(18)	0.23075(5)	0.7208(2)	1	0.0242(3)
H9	0.169(3)	0.2412(9)	0.790(4)	1	0.050
C10	0.3270(2)	0.17662(6)	0.7724(3)	1	0.0266(4)
H10A	0.298256	0.167825	0.934790	1	0.032
H10B	0.450836	0.176616	0.759846	1	0.032
C11	0.25799(19)	0.13267(6)	0.6173(3)	1	0.0236(4)
C12	0.2780(2)	0.07900(7)	0.6840(3)	1	0.0308(4)
H12	0.332086	0.070949	0.826492	1	0.037
C13	0.2205(2)	0.03725(7)	0.5463(3)	1	0.0371(5)
H13	0.235951	0.000845	0.593934	1	0.044
C14	0.1402(2)	0.04842(7)	0.3390(3)	1	0.0352(4)
H14	0.101047	0.019839	0.243381	1	0.042
C15	0.1178(2)	0.10152(7)	0.2726(3)	1	0.0309(4)
H15	0.061415	0.109434	0.131743	1	0.037
C16	0.1768(2)	0.14343(6)	0.4097(3)	1	0.0273(4)
H16	0.161586	0.179772	0.361142	1	0.033

9.3. Anisotropic displacement parameters ($Å^2$). The anisotropic displacement factor exponent takes the form: $-2\pi^2$ [h^2 a^{*2} U_{11} + ... + 2 h k a* b* U_{12}].

Atom	U11	U22	U33	U23	U13	U12
C1	0.0312(9)	0.0295(9)	0.0248(9)	0.0005(7)	0.0045(8)	-0.0074(7)
C2	0.0381(10)	0.0357(9)	0.0271(9)	0.0081(8)	-0.0022(8)	-0.0105(8)
C3	0.0338(9)	0.0232(8)	0.0453(11)	0.0027(8)	-0.0055(9)	-0.0024(7)
C4	0.0298(9)	0.0266(8)	0.0369(10)	-0.0065(8)	0.0029(8)	-0.0001(7)
C5	0.0249(8)	0.0243(8)	0.0274(8)	-0.0017(7)	0.0046(8)	-0.0046(7)
C6	0.0205(8)	0.0238(7)	0.0236(8)	-0.0016(6)	0.0040(7)	-0.0055(6)
C7	0.0224(8)	0.0277(8)	0.0276(9)	-0.0013(7)	0.0113(7)	-0.0022(6)
O8	0.0417(8)	0.0381(7)	0.0664(10)	0.0105(7)	0.0407(8)	0.0091(6)
N9	0.0231(7)	0.0216(6)	0.0280(8)	0.0008(5)	0.0119(6)	0.0003(5)
C10	0.0280(8)	0.0245(8)	0.0273(9)	0.0031(7)	0.0038(7)	0.0011(7)
C11	0.0199(7)	0.0248(8)	0.0261(8)	0.0028(6)	0.0077(7)	0.0013(6)
C12	0.0338(9)	0.0259(8)	0.0327(9)	0.0049(7)	-0.0035(8)	0.0044(7)
C13	0.0436(11)	0.0227(8)	0.0449(11)	0.0010(8)	-0.0025(10)	0.0039(8)
C14	0.0370(10)	0.0296(9)	0.0389(11)	-0.0051(8)	0.0000(9)	0.0003(7)
C15	0.0306(9)	0.0341(9)	0.0280(9)	-0.0014(8)	0.0009(8)	0.0039(7)
C16	0.0292(8)	0.0252(8)	0.0275(9)	0.0035(7)	0.0071(8)	0.0045(7)

9.4. Bond lengths [Å].

C1	- C2	= 1.383(2)	C1	- C6	= 1.392(2)
C1	- H1	= 0.9500	C2	- C3	= 1.385(3)
C2	- H2	= 0.9500	C3	- C4	= 1.383(3)
C3	- H3	= 0.9500	C4	- C5	= 1.387(2)
C4	- H4	= 0.9500	C5	- C6	= 1.392(2)
C5	- H5	= 0.9500	C6	- C7	= 1.500(2)
C7	- O8	= 1.233(2)	C7	- N9	= 1.328(2)
N9	- C10	= 1.456(2)	N9	- H9	= 0.91(2)
C10	- C11	= 1.513(2)	C10	- H10A	= 0.9900
C10	- H10B	= 0.9900	C11	- C16	= 1.388(3)
C11	- C12	= 1.392(2)	C12	- C13	= 1.382(3)
C12	- H12	= 0.9500	C13	- C14	= 1.385(3)
C13	- H13	= 0.9500	C14	- C15	= 1.380(2)
C14	- H14	= 0.9500	C15	- C16	= 1.387(2)
C15	- H15	= 0.9500	C16	- H16	= 0.9500

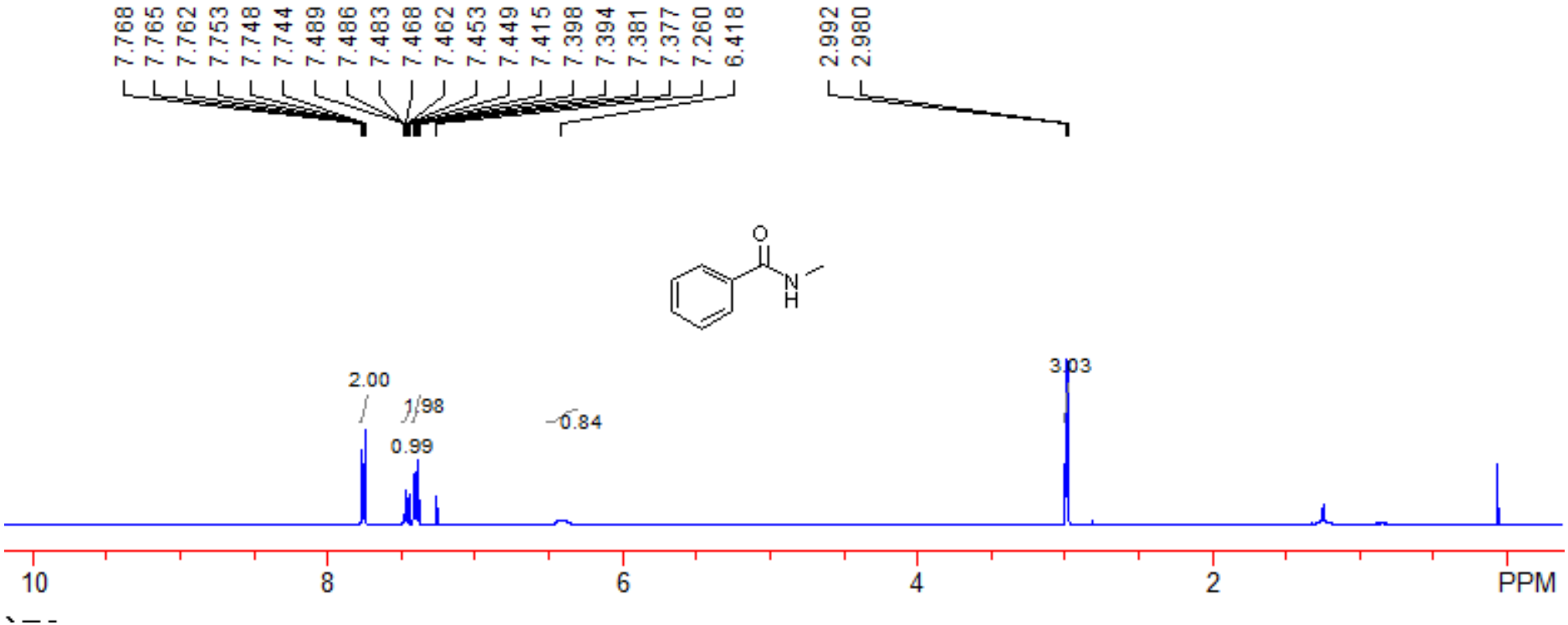
9.5. Angles [°].

C2	- C1	- C6	= 120.33(16)
C2	- C1	- H1	= 119.80
C6	- C1	- H1	= 119.80
C1	- C2	- C3	= 119.72(17)
C1	- C2	- H2	= 120.10
C3	- C2	- H2	= 120.10
C4	- C3	- C2	= 120.41(16)
C4	- C3	- H3	= 119.80
C2	- C3	- H3	= 119.80
C3	- C4	- C5	= 120.03(17)
C3	- C4	- H4	= 120.00
C5	- C4	- H4	= 120.00
C4	- C5	- C6	= 119.86(16)
C4	- C5	- H5	= 120.10
C6	- C5	- H5	= 120.10
C1	- C6	- C5	= 119.62(15)
C1	- C6	- C7	= 117.47(14)
C5	- C6	- C7	= 122.85(15)
O8	- C7	- N9	= 121.47(16)
O8	- C7	- C6	= 120.37(15)
N9	- C7	- C6	= 118.15(14)
C7	- N9	- C10	= 121.40(14)
C7	- N9	- H9	= 121.60(14)
C10	- N9	- H9	= 117.00(14)
N9	- C10	- C11	= 114.75(14)
N9	- C10	- H10A	= 108.60
C11	- C10	- H10A	= 108.60
N9	- C10	- H10B	= 108.60
C11	- C10	- H10B	= 108.60
H10A	- C10	- H10B	= 107.60
C16	- C11	- C12	= 118.40(16)
C16	- C11	- C10	= 122.89(14)
C12	- C11	- C10	= 118.71(15)
C13	- C12	- C11	= 121.06(17)
C13	- C12	- H12	= 119.50
C11	- C12	- H12	= 119.50
C12	- C13	- C14	= 120.07(16)
C12	- C13	- H13	= 120.00
C14	- C13	- H13	= 120.00
C15	- C14	- C13	= 119.31(17)
C15	- C14	- H14	= 120.30
C13	- C14	- H14	= 120.30
C14	- C15	- C16	= 120.65(17)
C14	- C15	- H15	= 119.70
C16	- C15	- H15	= 119.70
C15	- C16	- C11	= 120.49(15)
C15	- C16	- H16	= 119.80
C11	- C16	- H16	= 119.80

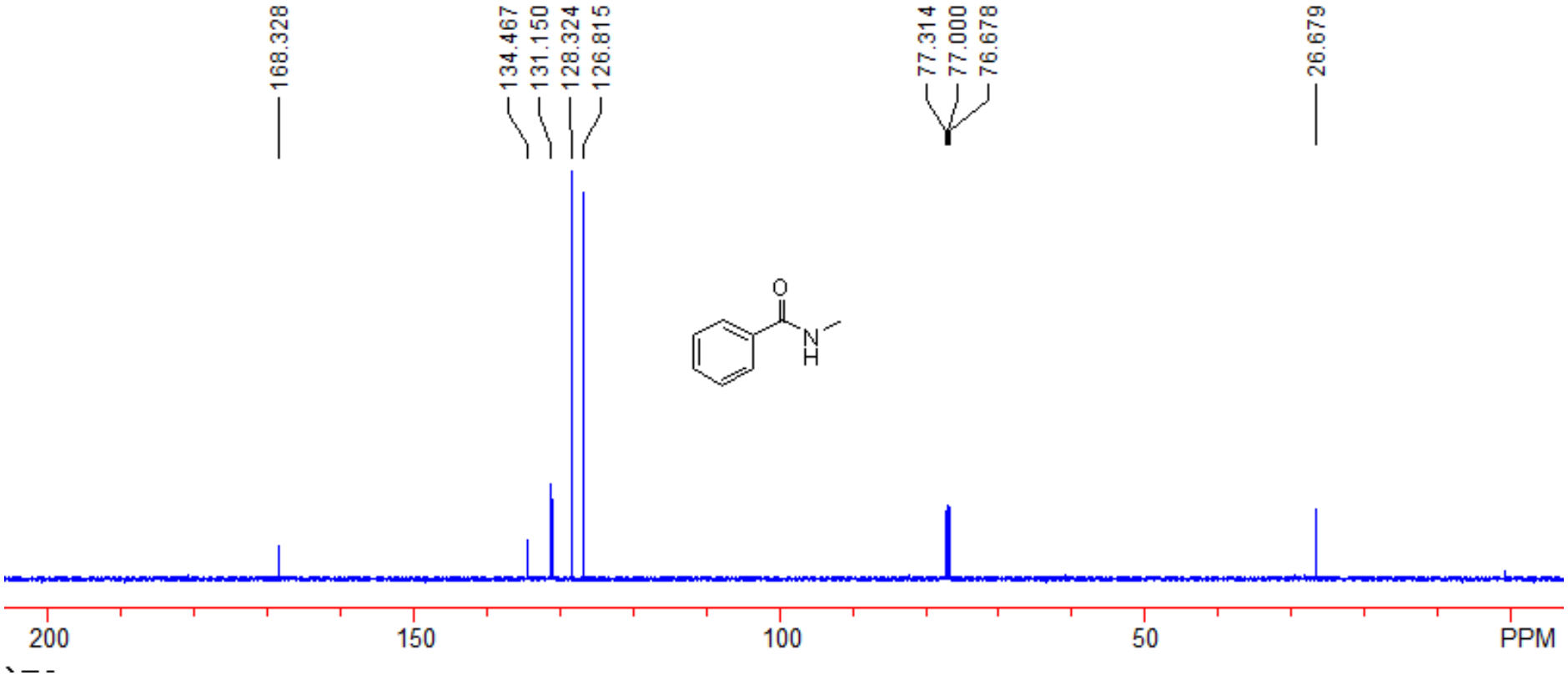
9.6. Torsion angles [°].

C6	- C1	- C2	- C3	=	-0.30(3)
C1	- C2	- C3	- C4	=	-1.10(3)
C2	- C3	- C4	- C5	=	1.40(3)
C3	- C4	- C5	- C6	=	-0.30(3)
C2	- C1	- C6	- C5	=	1.30(3)
C2	- C1	- C6	- C7	=	-175.97(16)
C4	- C5	- C6	- C1	=	-1.00(3)
C4	- C5	- C6	- C7	=	176.11(16)
C1	- C6	- C7	- O8	=	22.30(2)
C5	- C6	- C7	- O8	=	-154.94(18)
C1	- C6	- C7	- N9	=	-158.93(15)
C5	- C6	- C7	- N9	=	23.90(2)
O8	- C7	- N9	- C10	=	1.00(3)
C6	- C7	- N9	- C10	=	-177.80(14)
C7	- N9	- C10	- C11	=	-90.94(19)
N9	- C10	- C11	- C16	=	15.00(2)
N9	- C10	- C11	- C12	=	-165.61(15)
C16	- C11	- C12	- C13	=	0.80(3)
C10	- C11	- C12	- C13	=	-178.63(16)
C11	- C12	- C13	- C14	=	-0.50(3)
C12	- C13	- C14	- C15	=	-0.40(3)
C13	- C14	- C15	- C16	=	0.90(3)
C14	- C15	- C16	- C11	=	-0.70(3)
C12	- C11	- C16	- C15	=	-0.20(2)
C10	- C11	- C16	- C15	=	179.18(15)

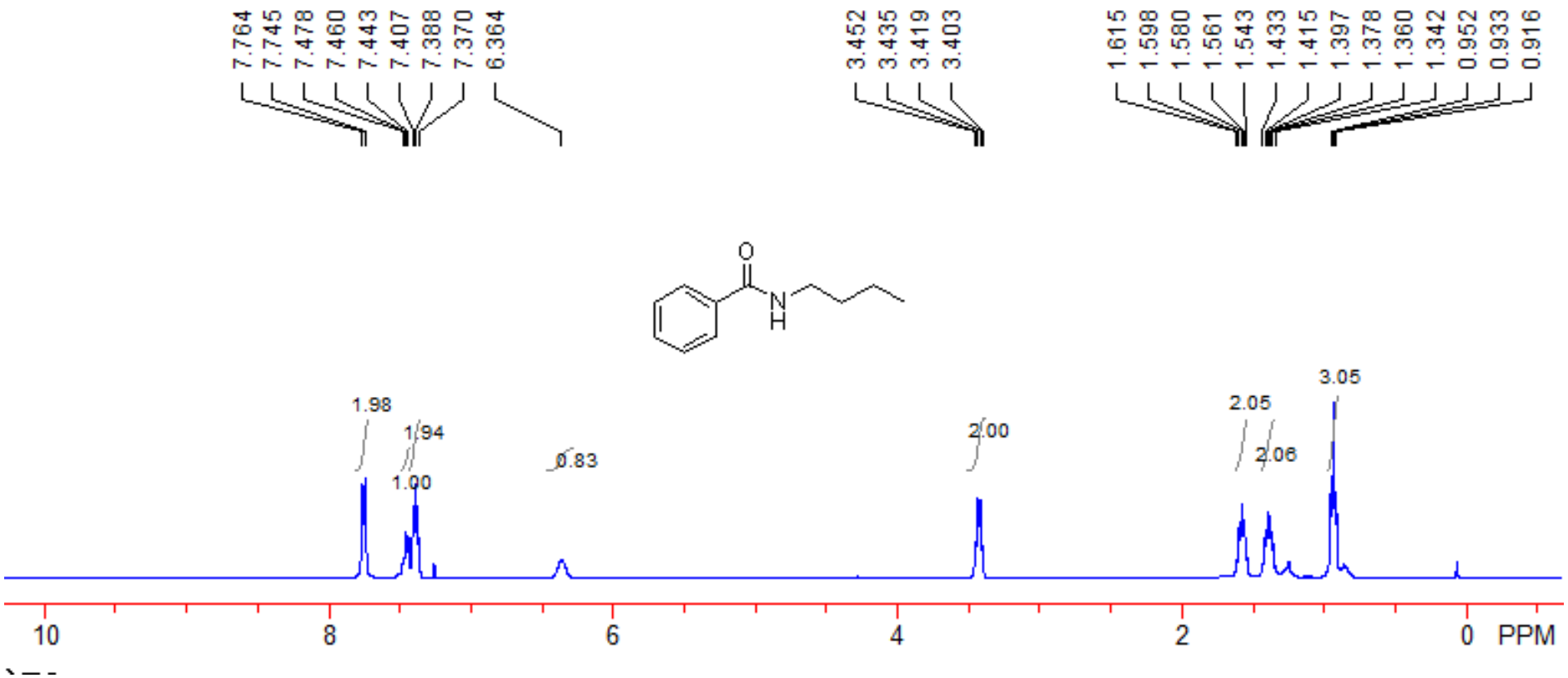
10. NMR spectra of products 2-3.



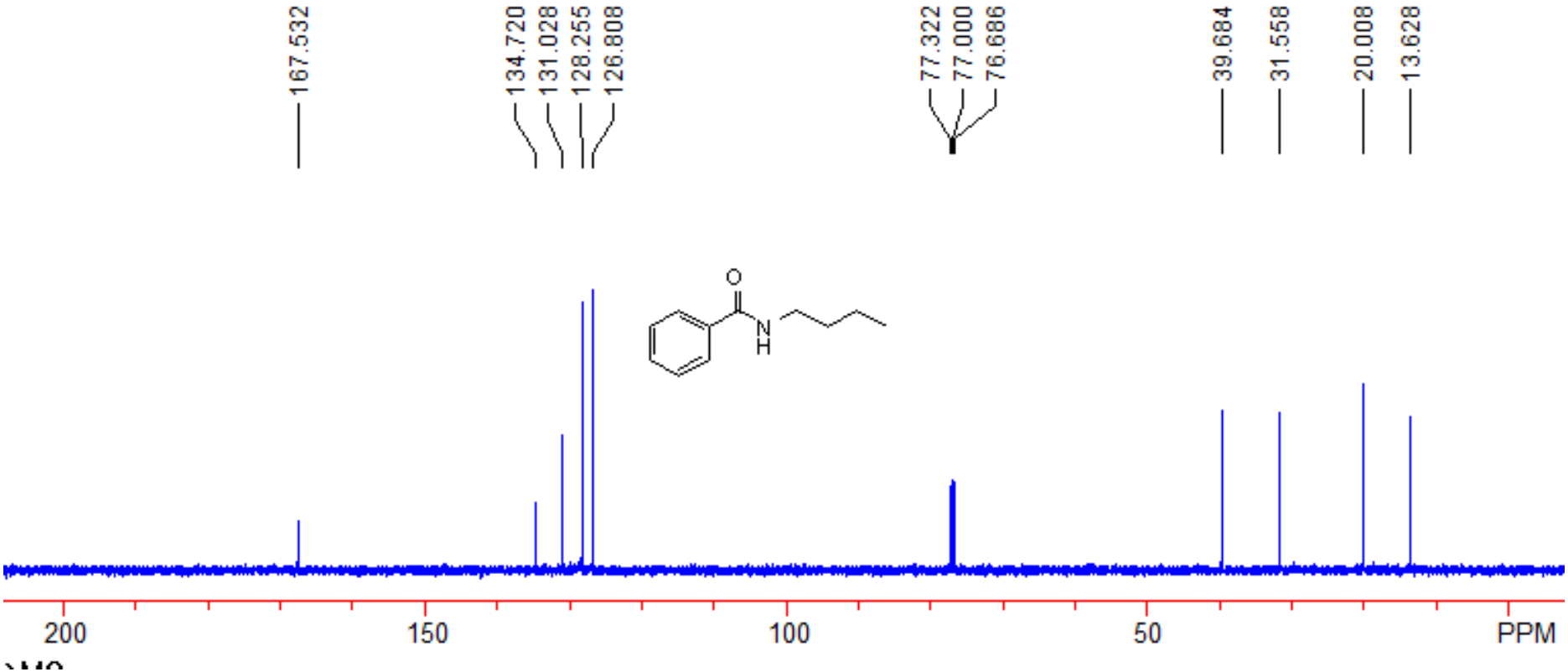
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2a**.



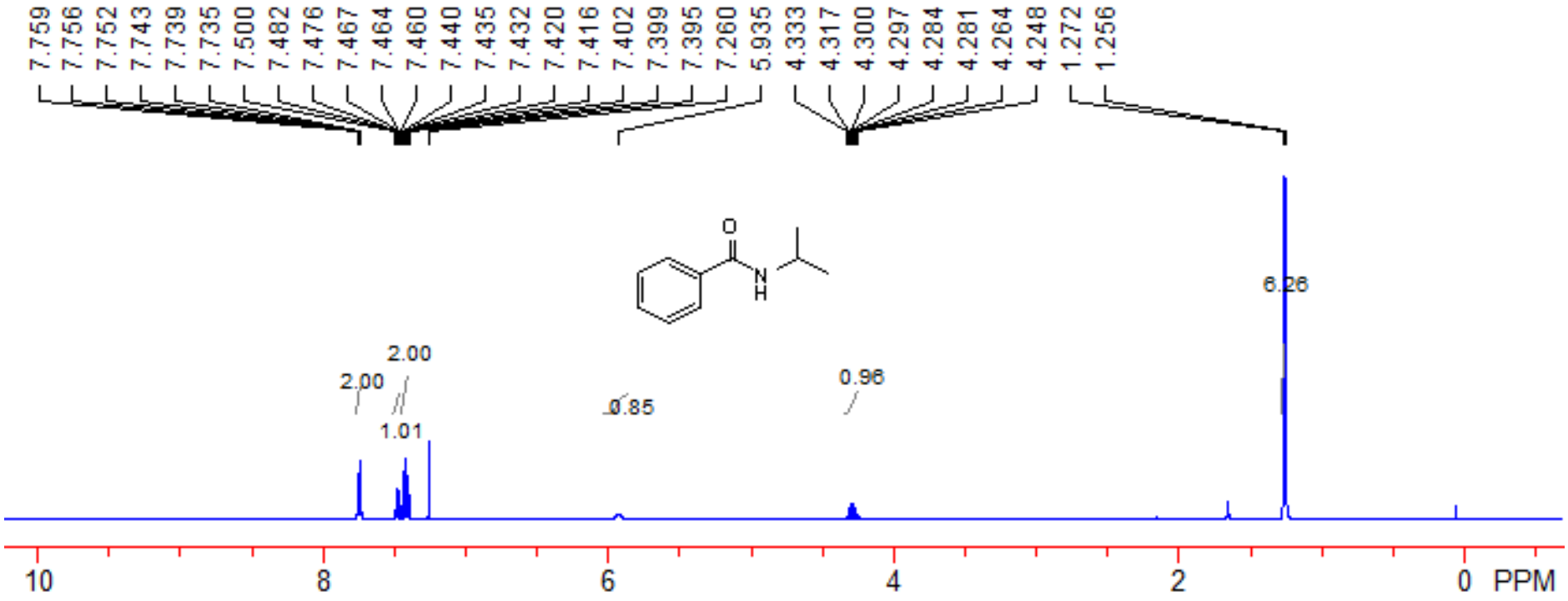
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2a**.



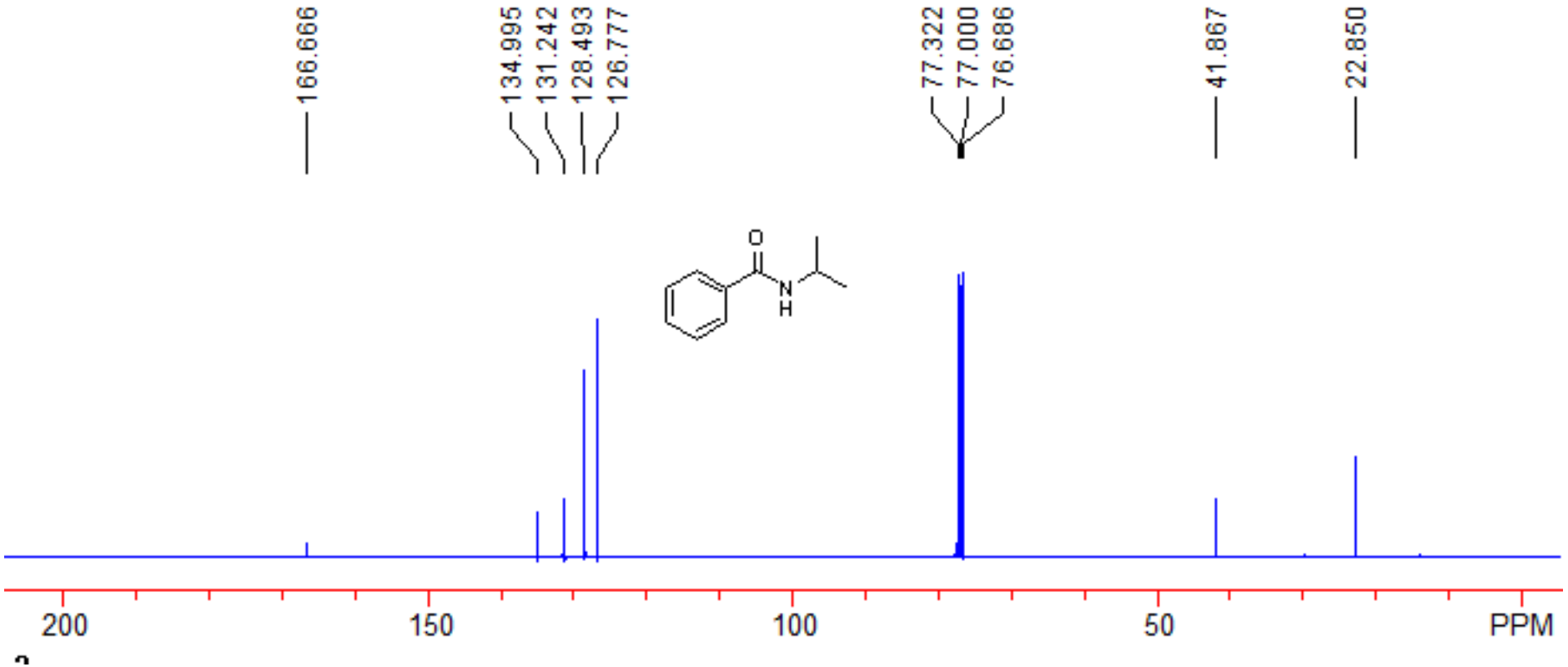
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2b**.



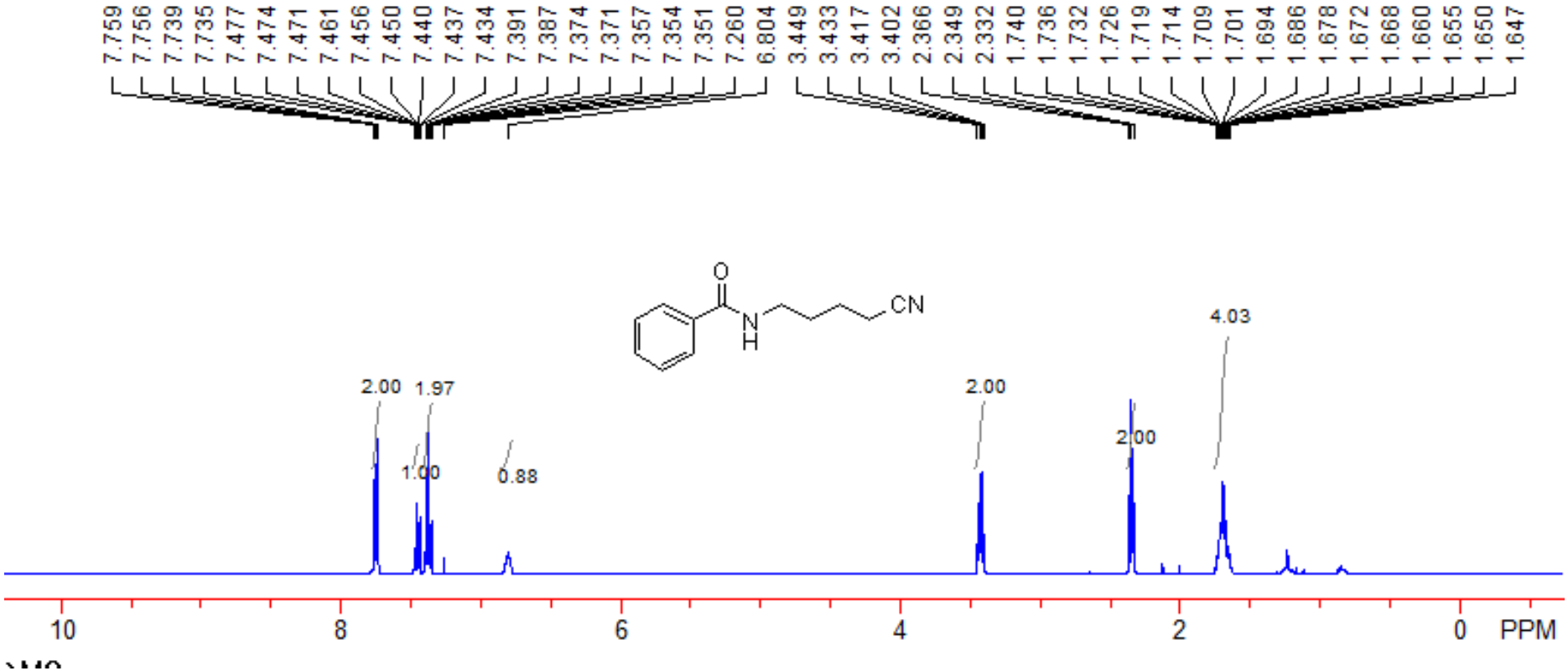
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2b**.



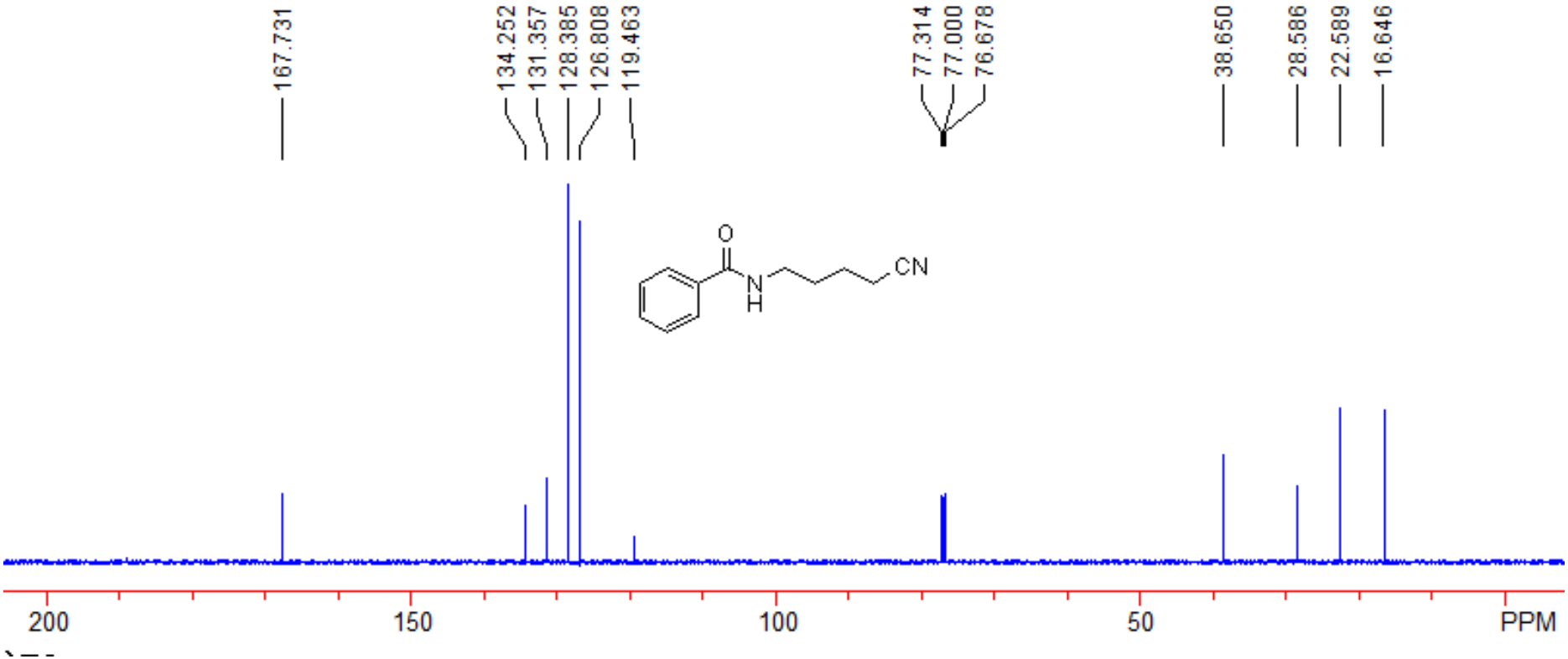
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2c**.



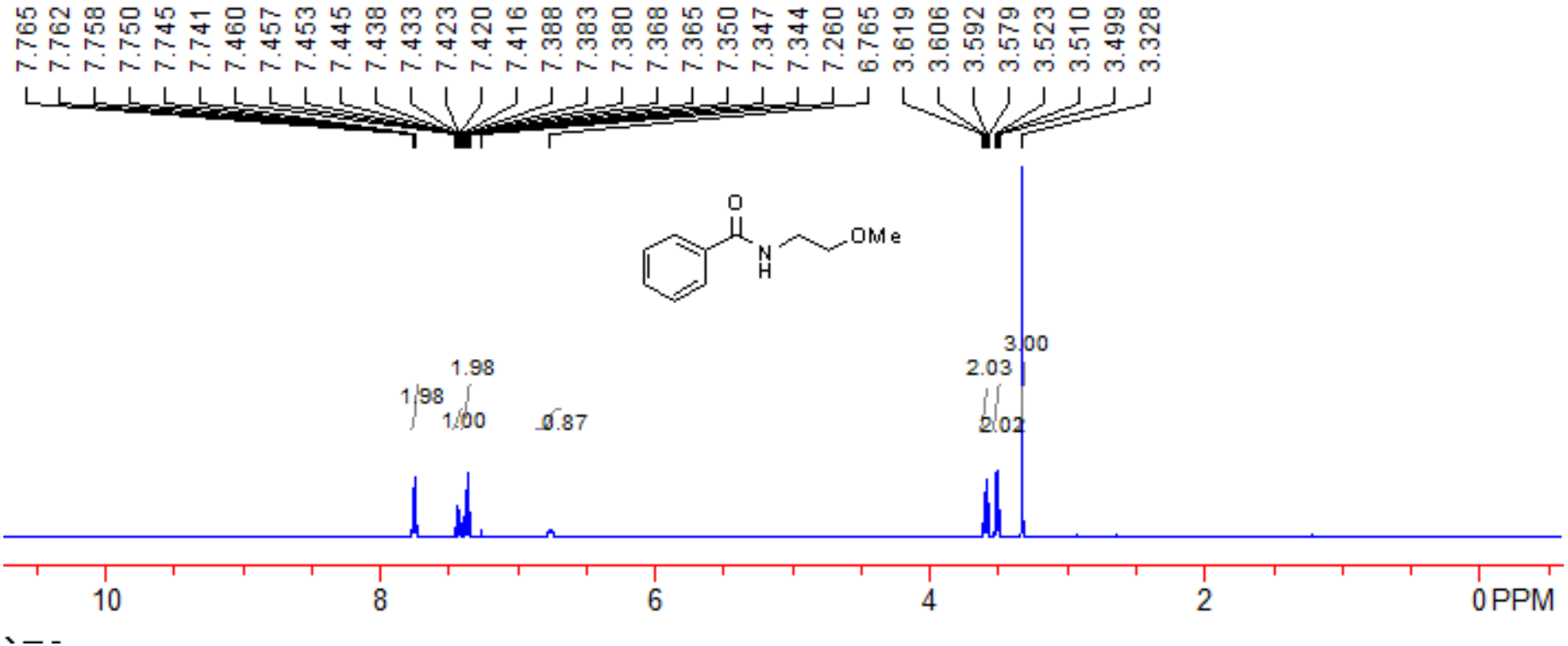
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2c**.



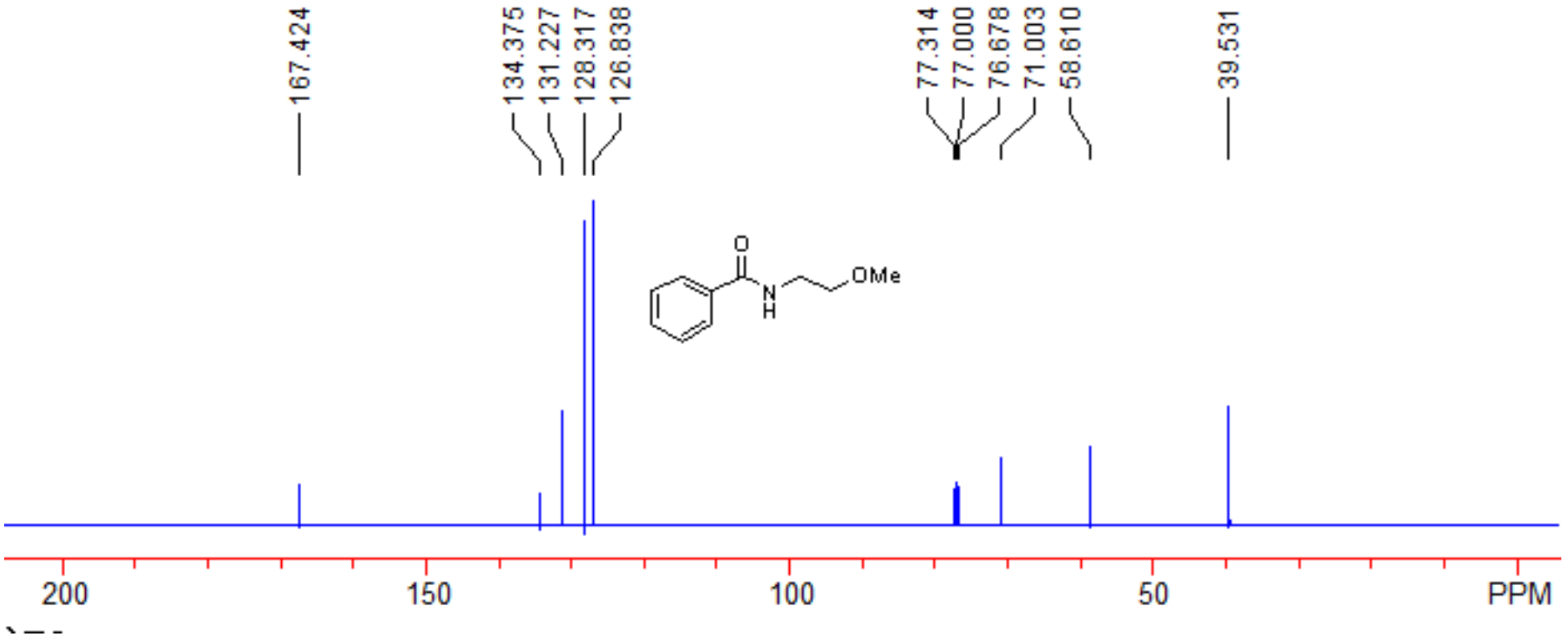
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2e**.



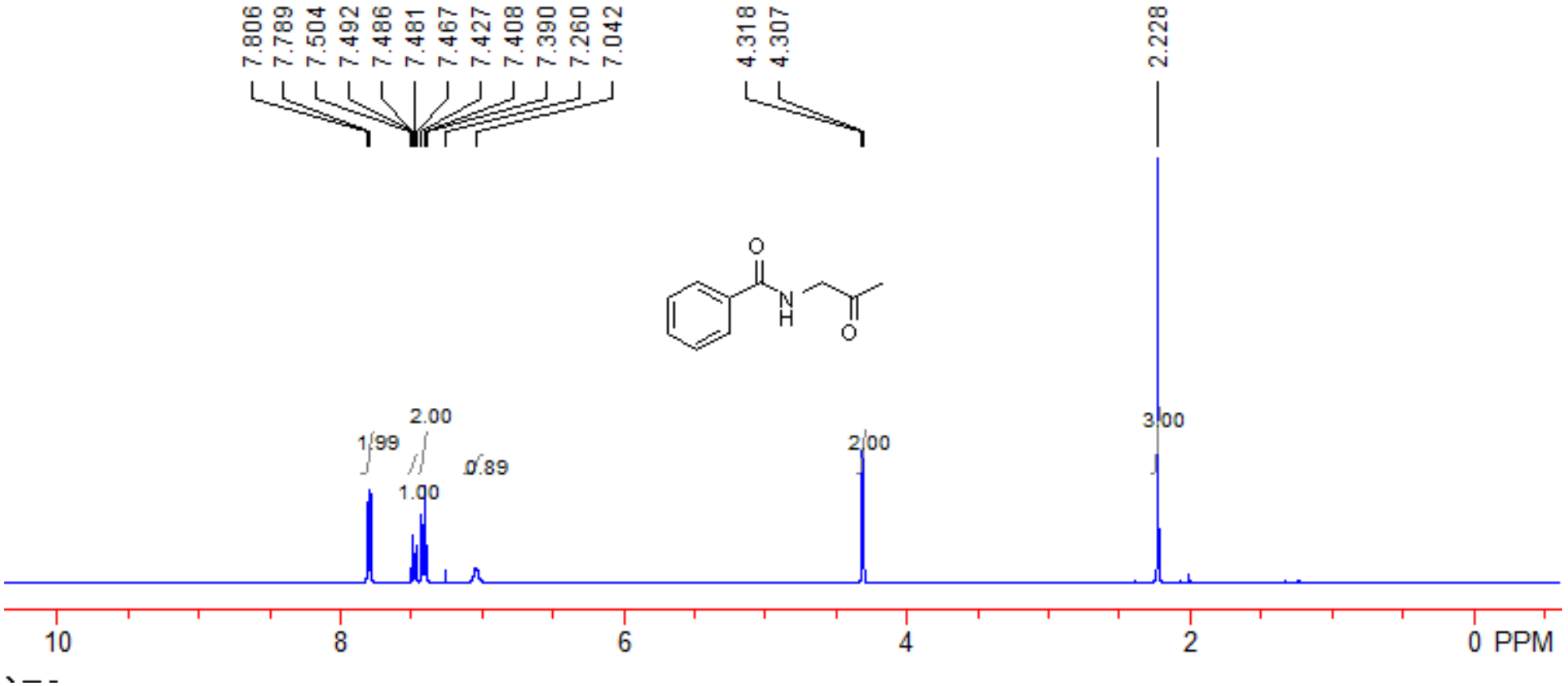
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2e**.



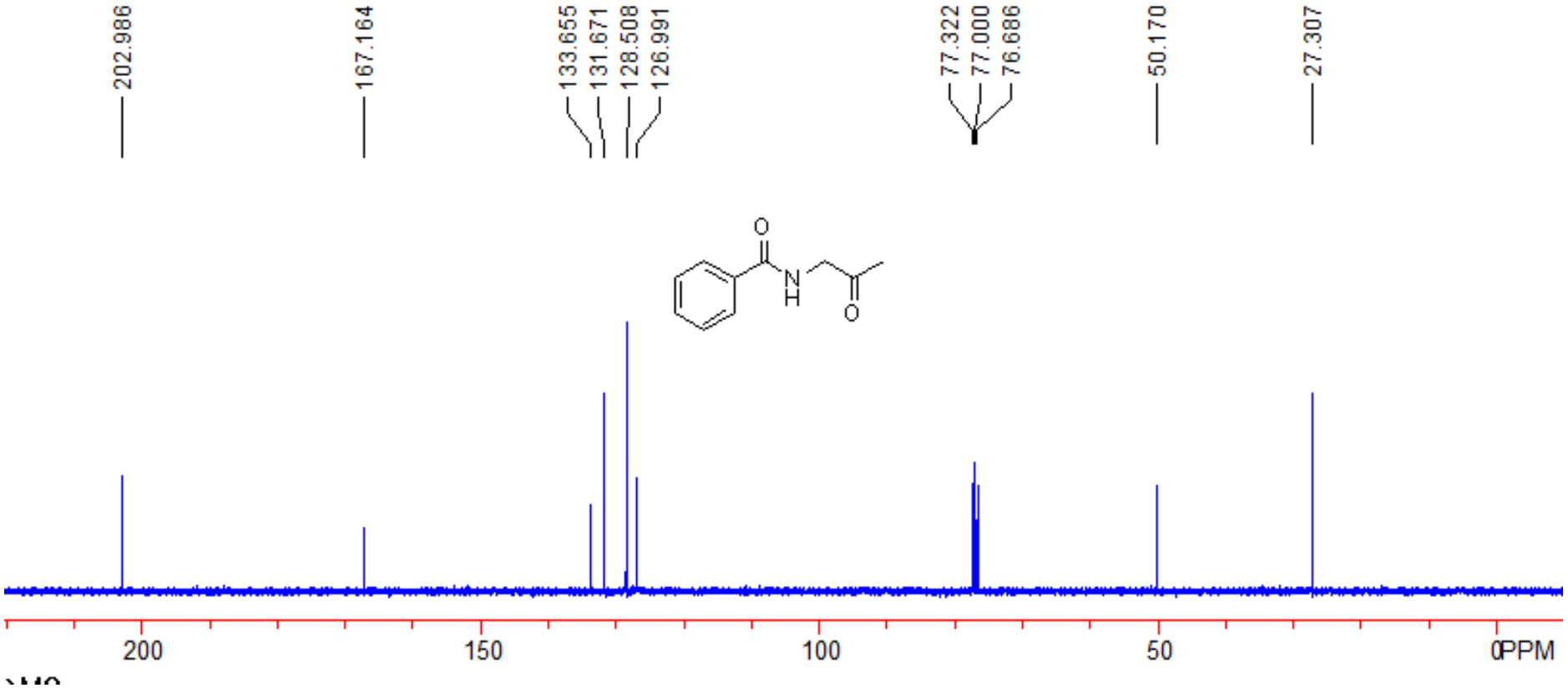
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2f**.



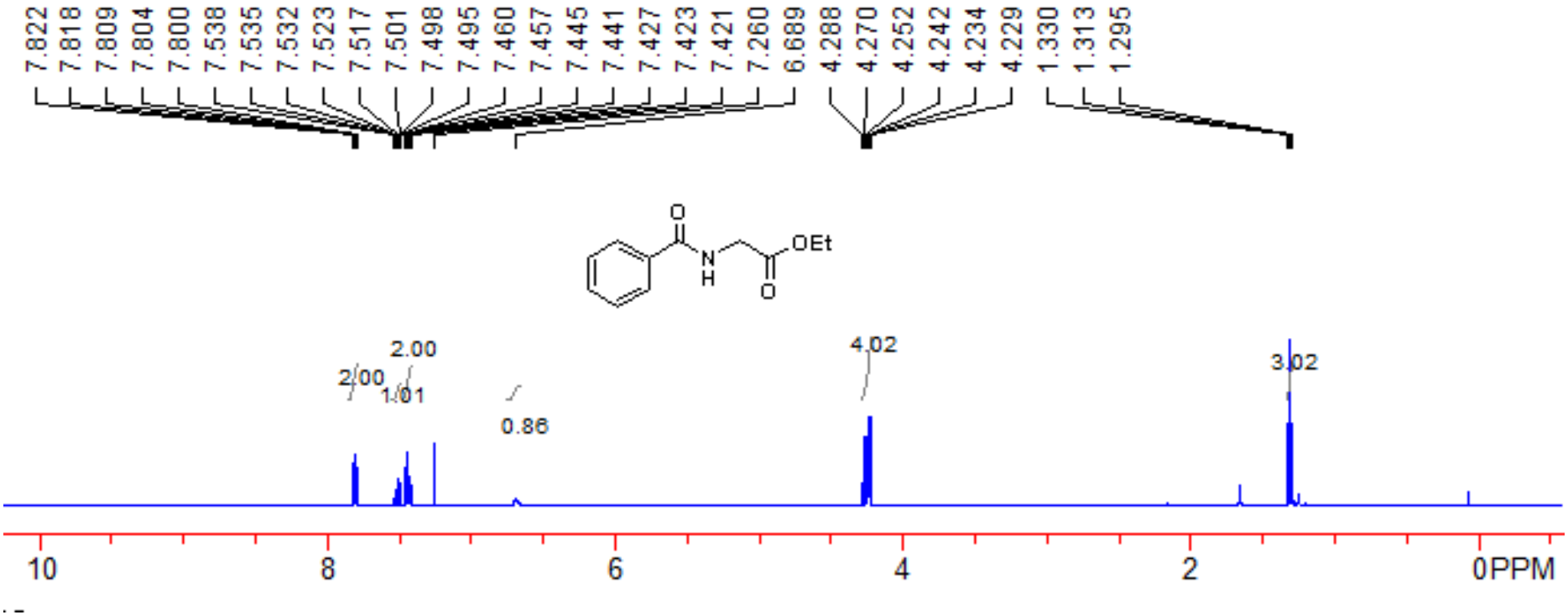
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2f**.



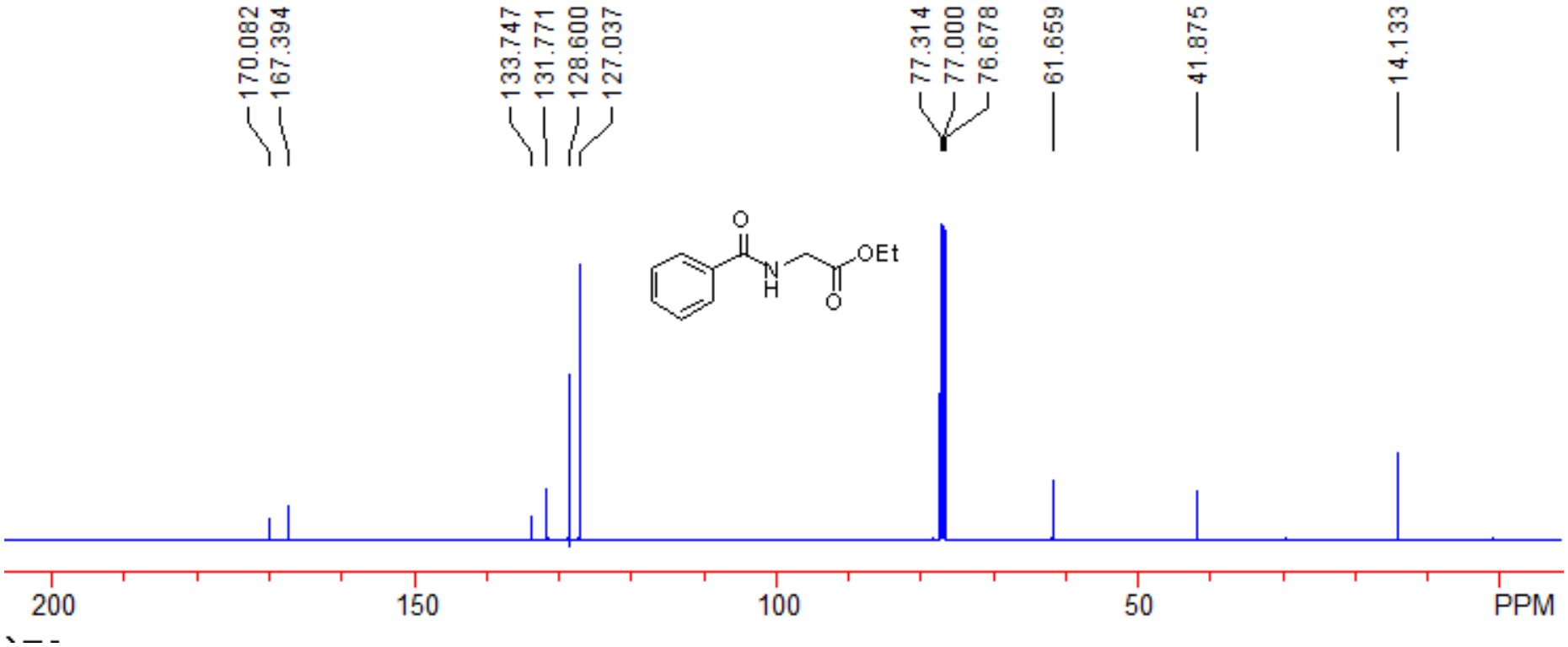
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2g**.



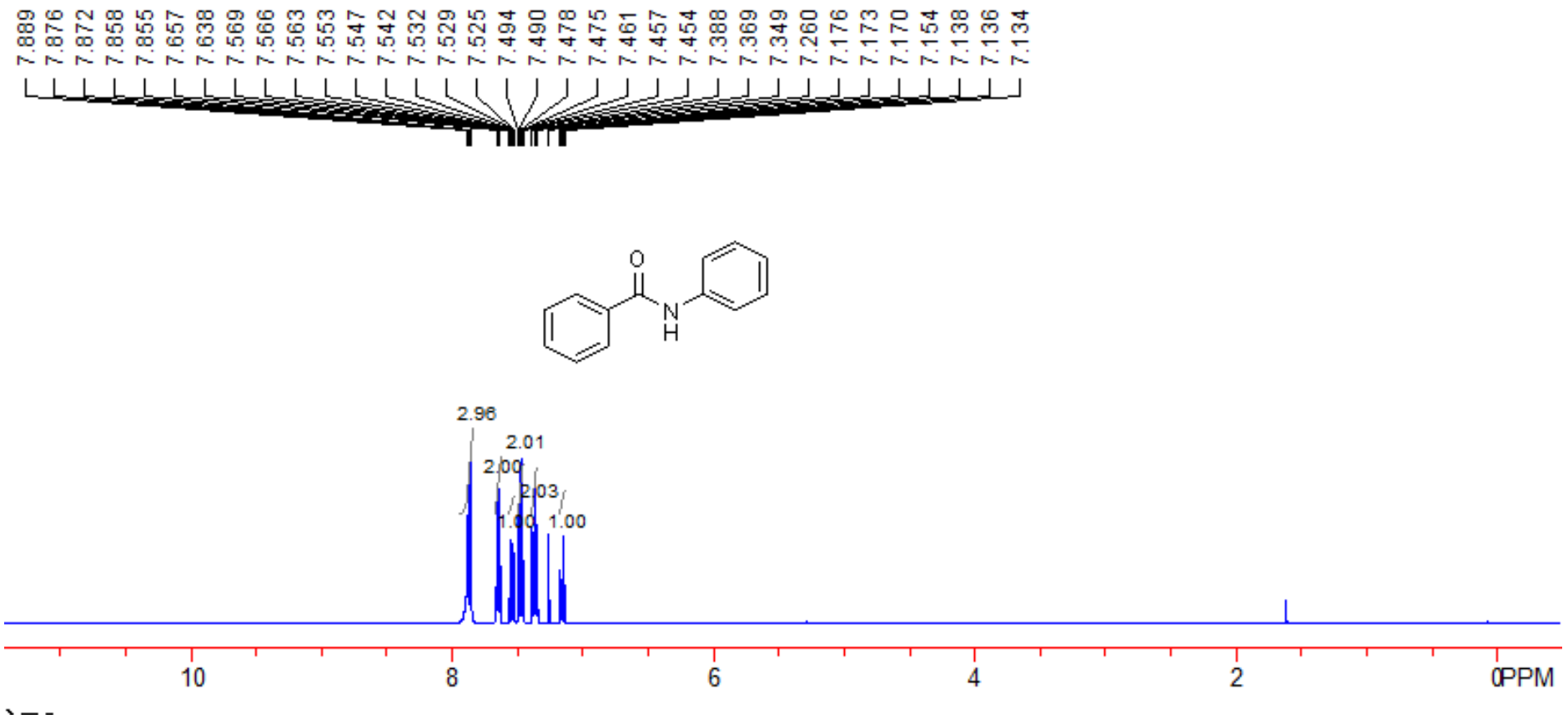
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2g**.



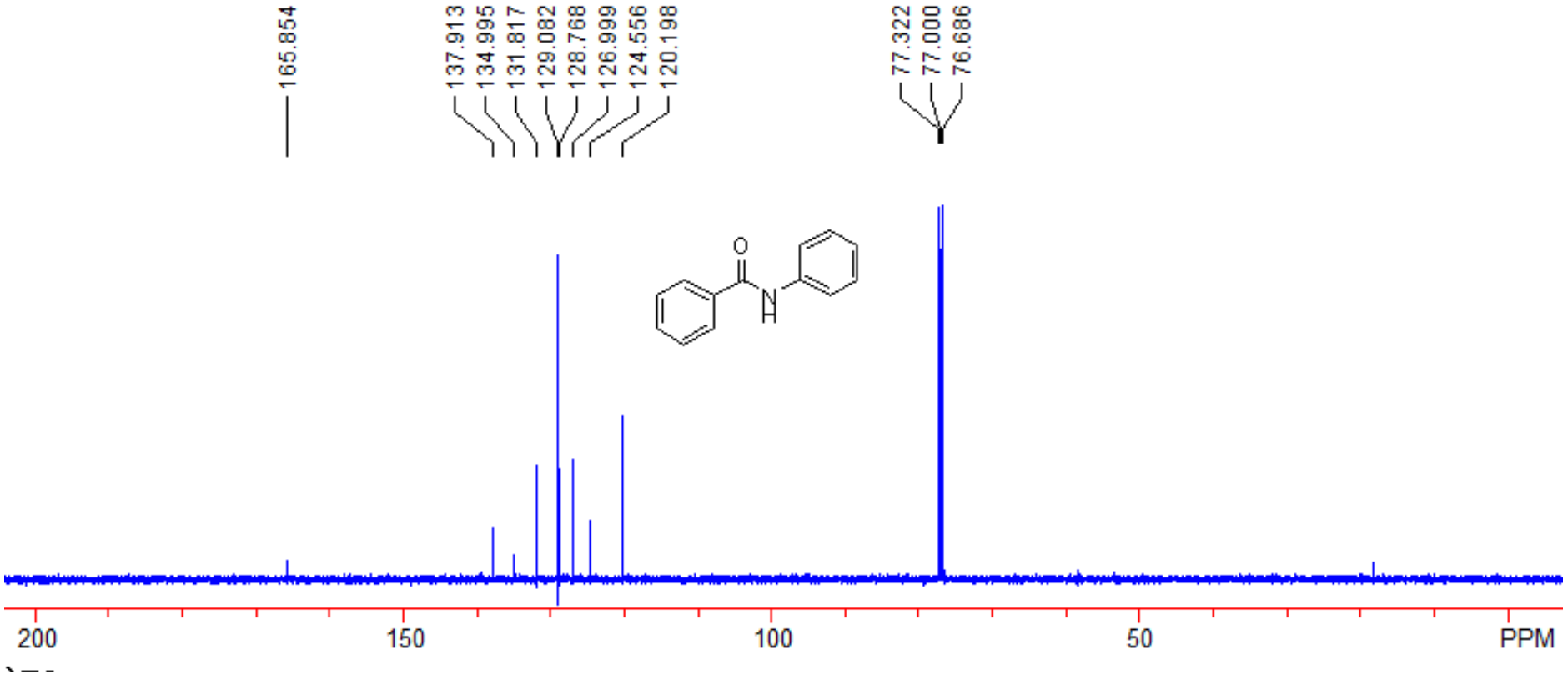
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2h**.



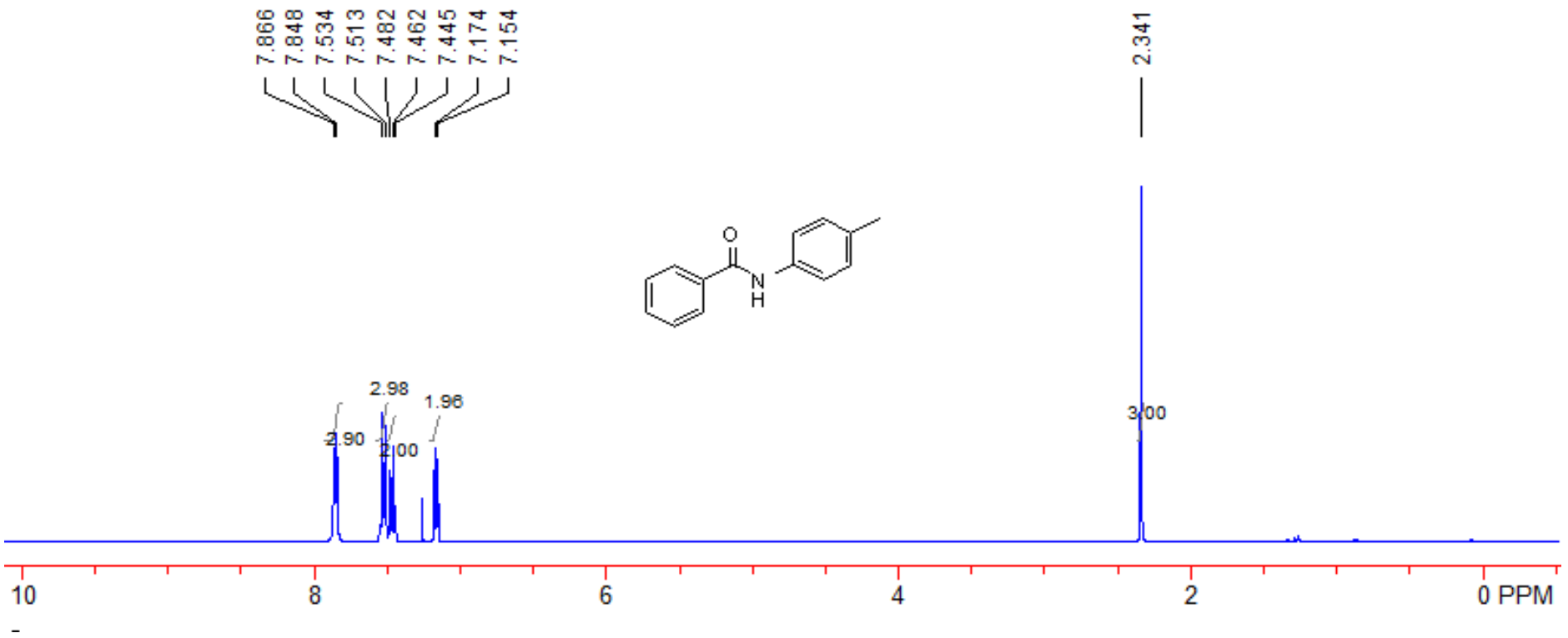
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2h**.



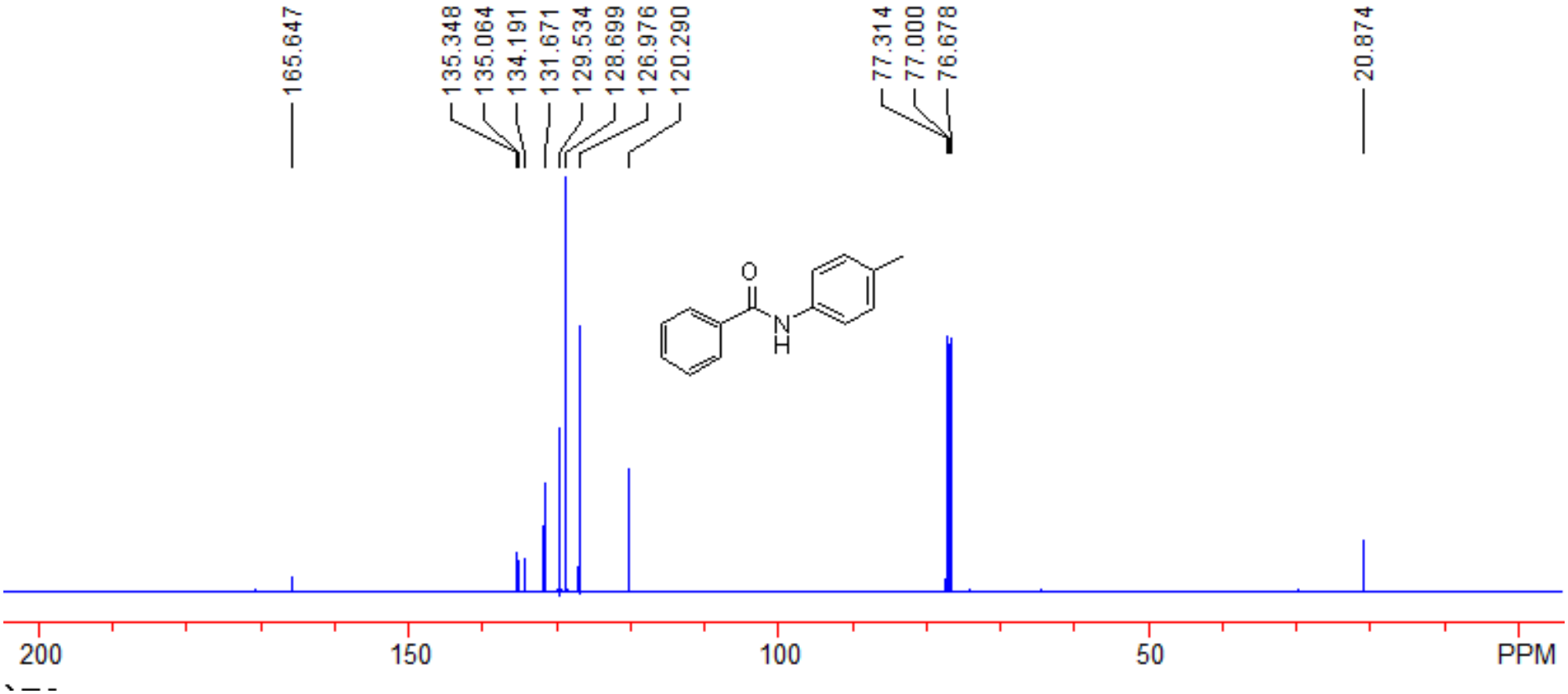
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2i**.



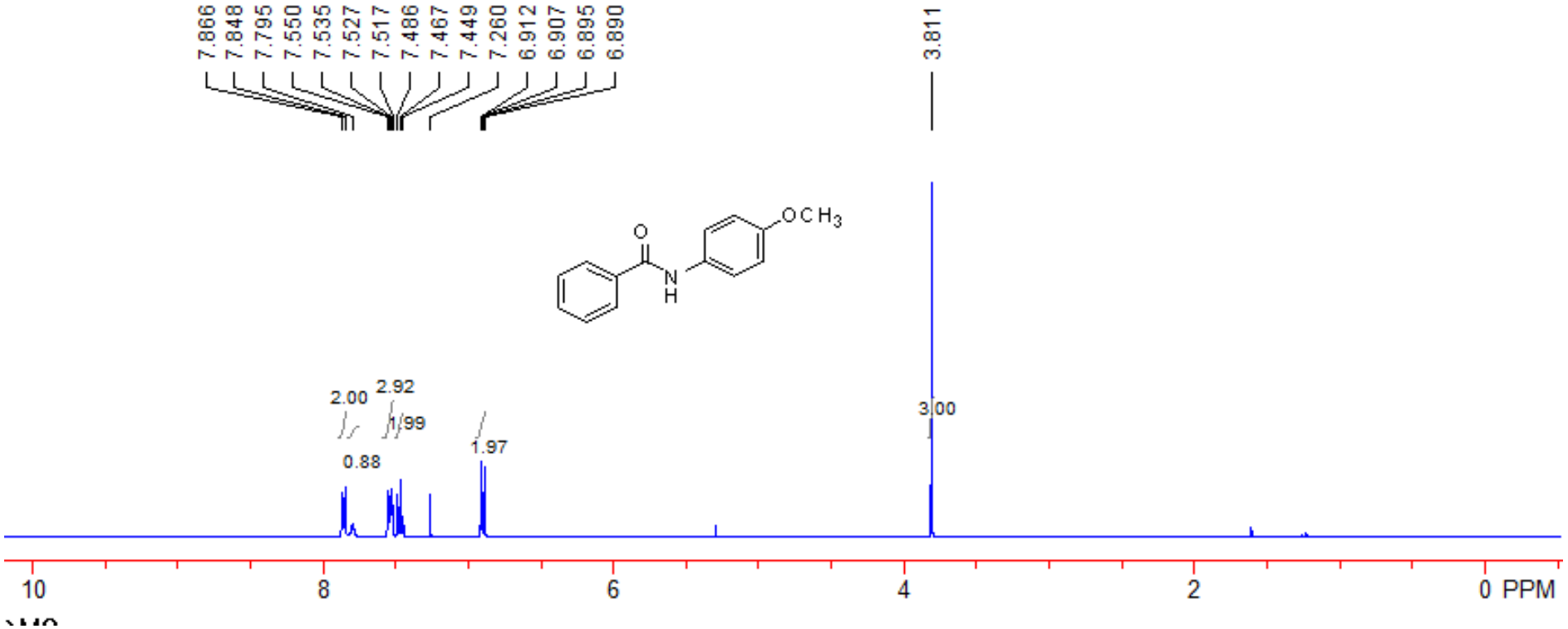
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2i**.



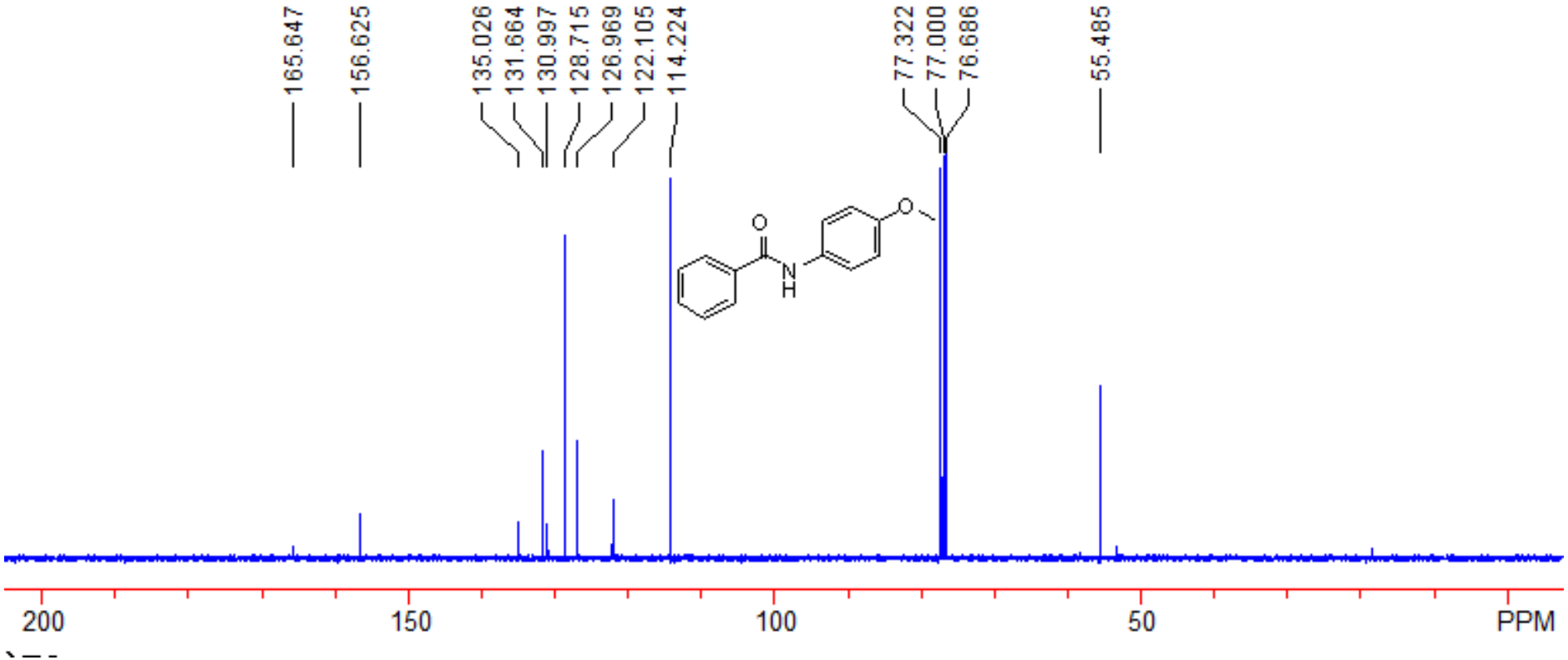
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2j**.



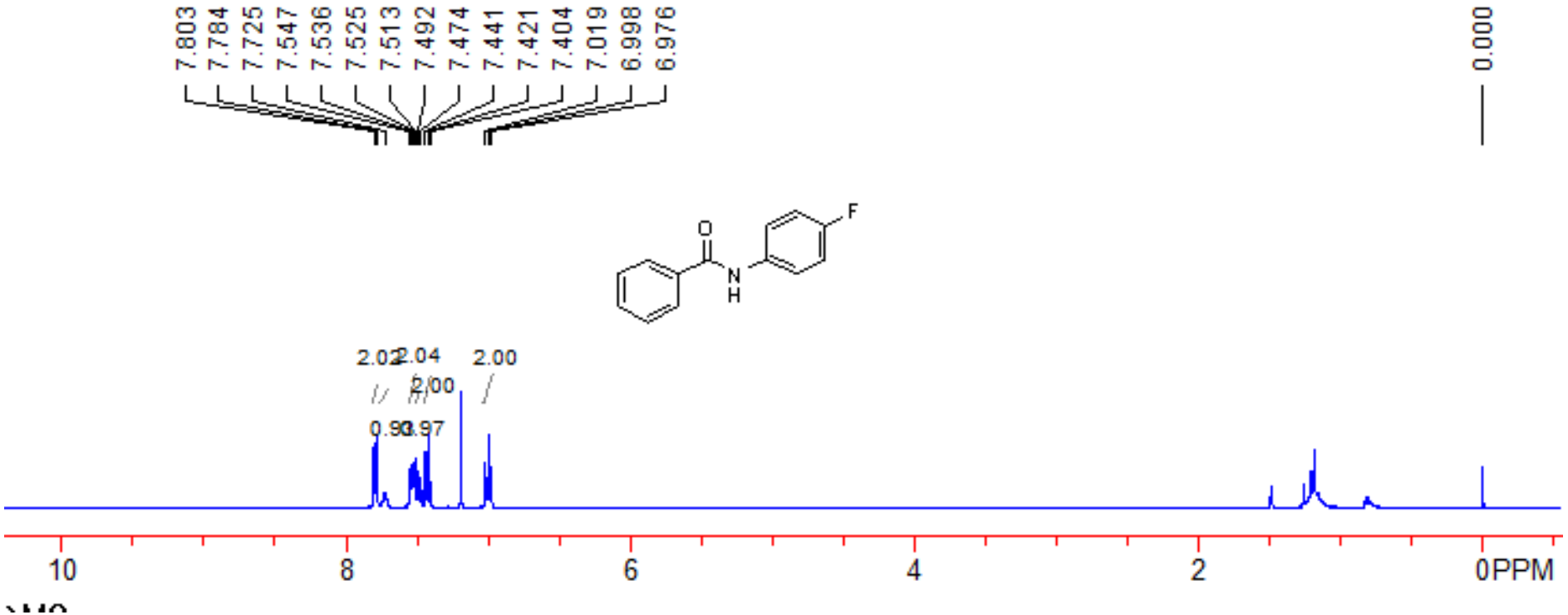
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2j**.



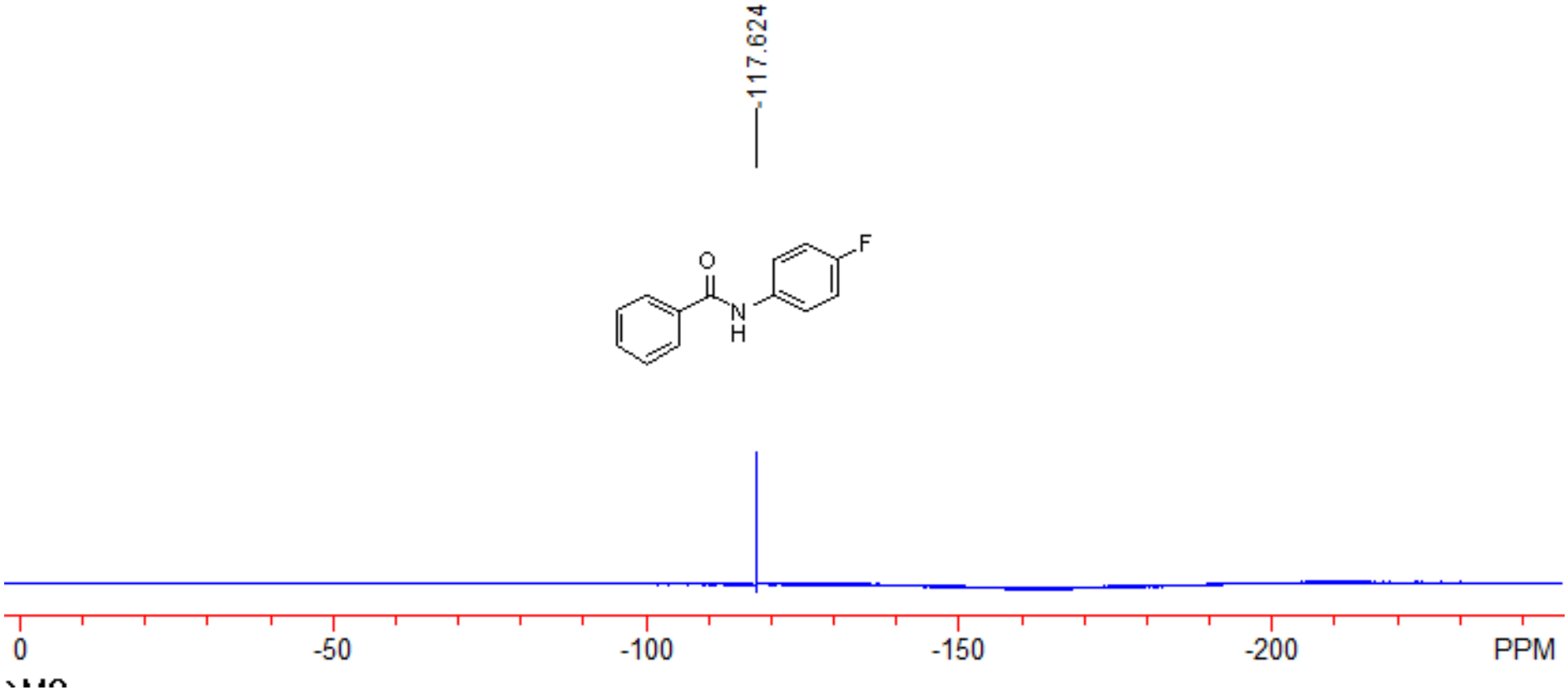
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2k**.



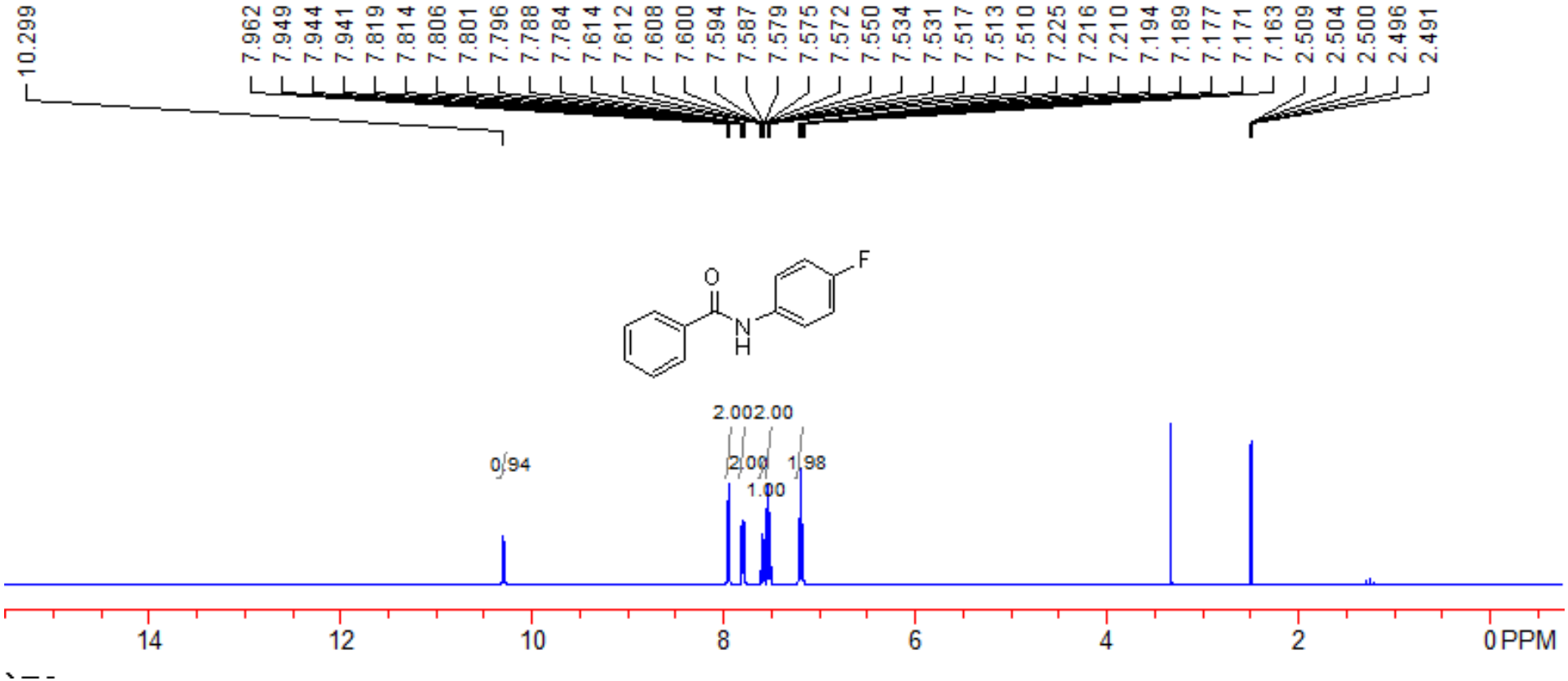
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2k**.



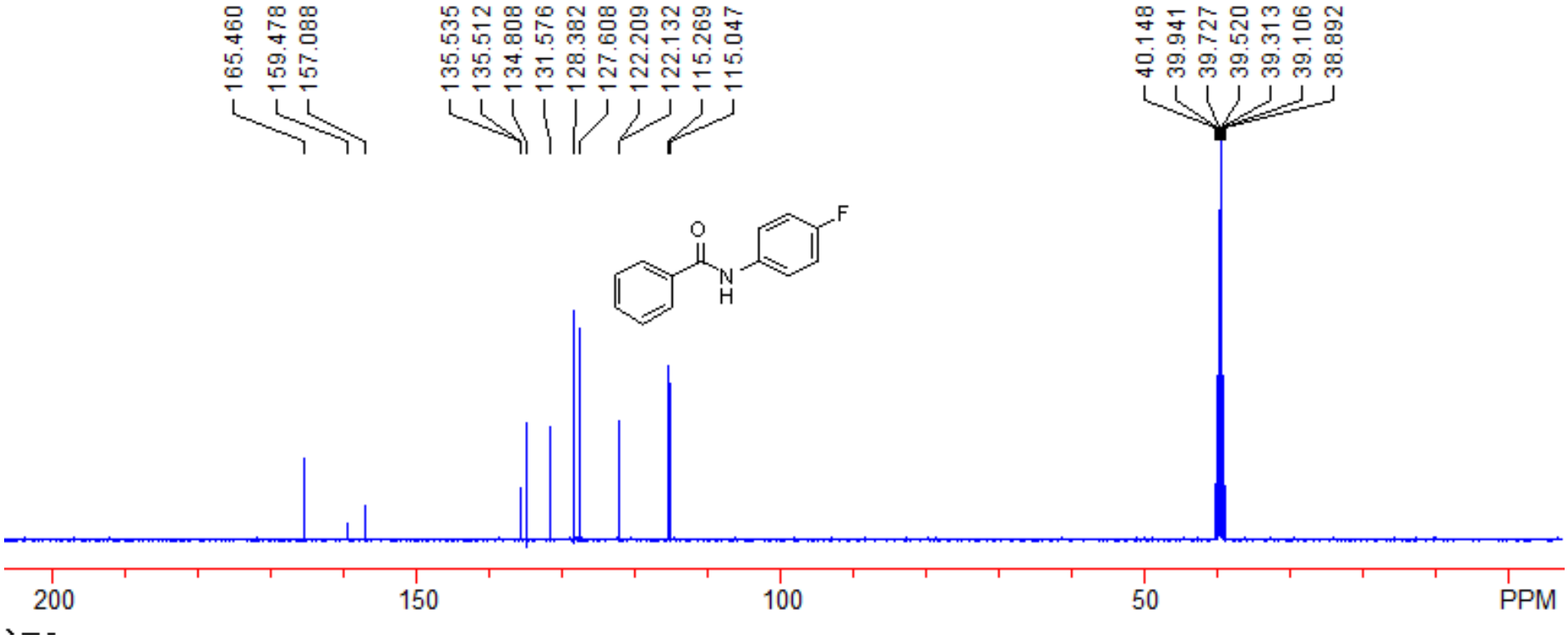
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2l**.



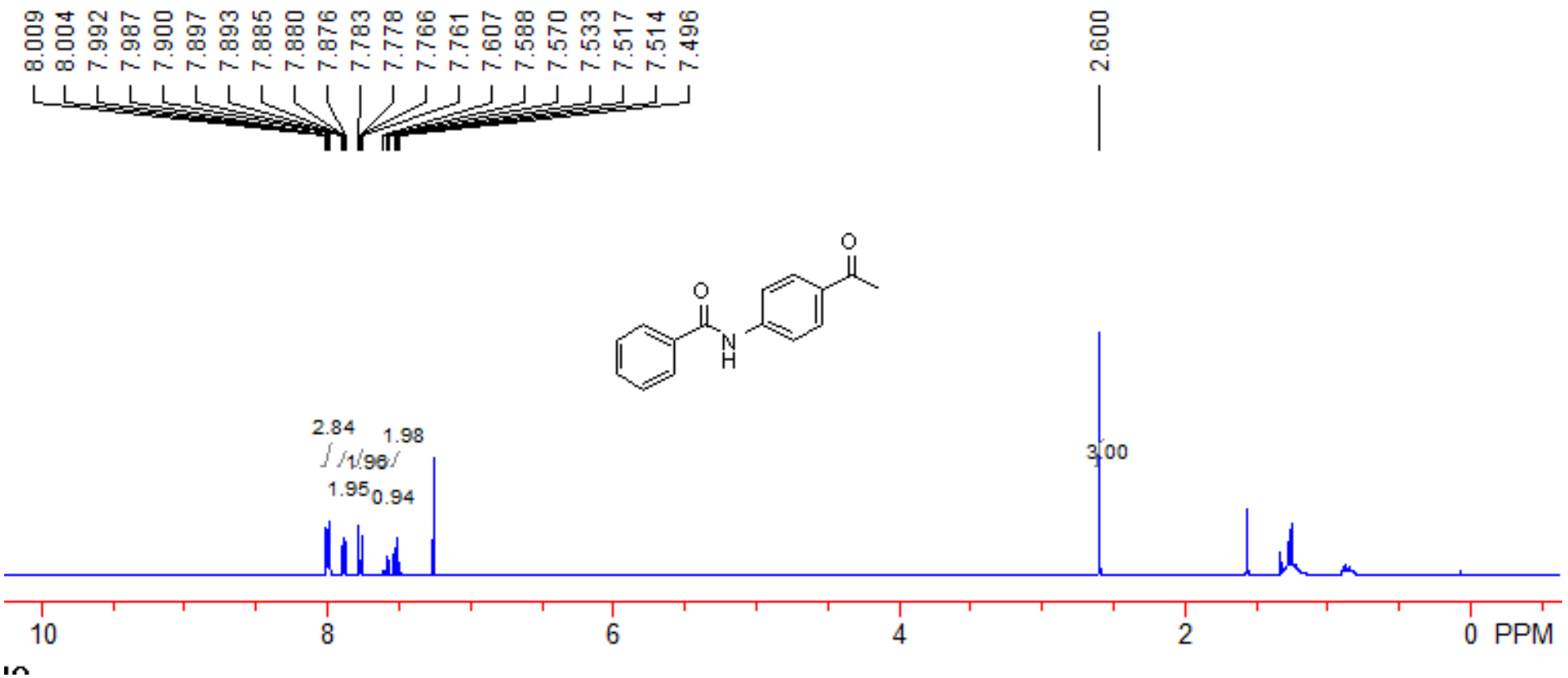
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) spectrum of **2l**.



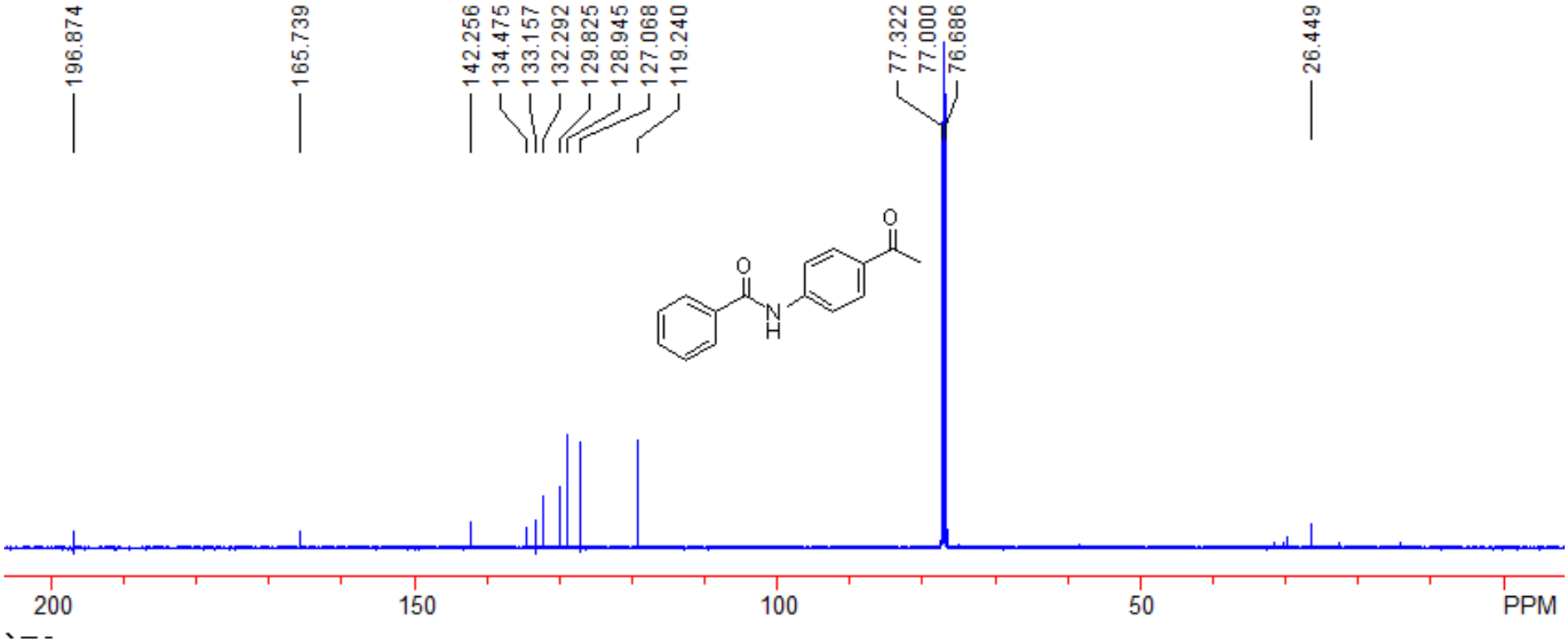
^{1}H NMR (400 MHz, DMSO-d_6) spectrum of **2l**.



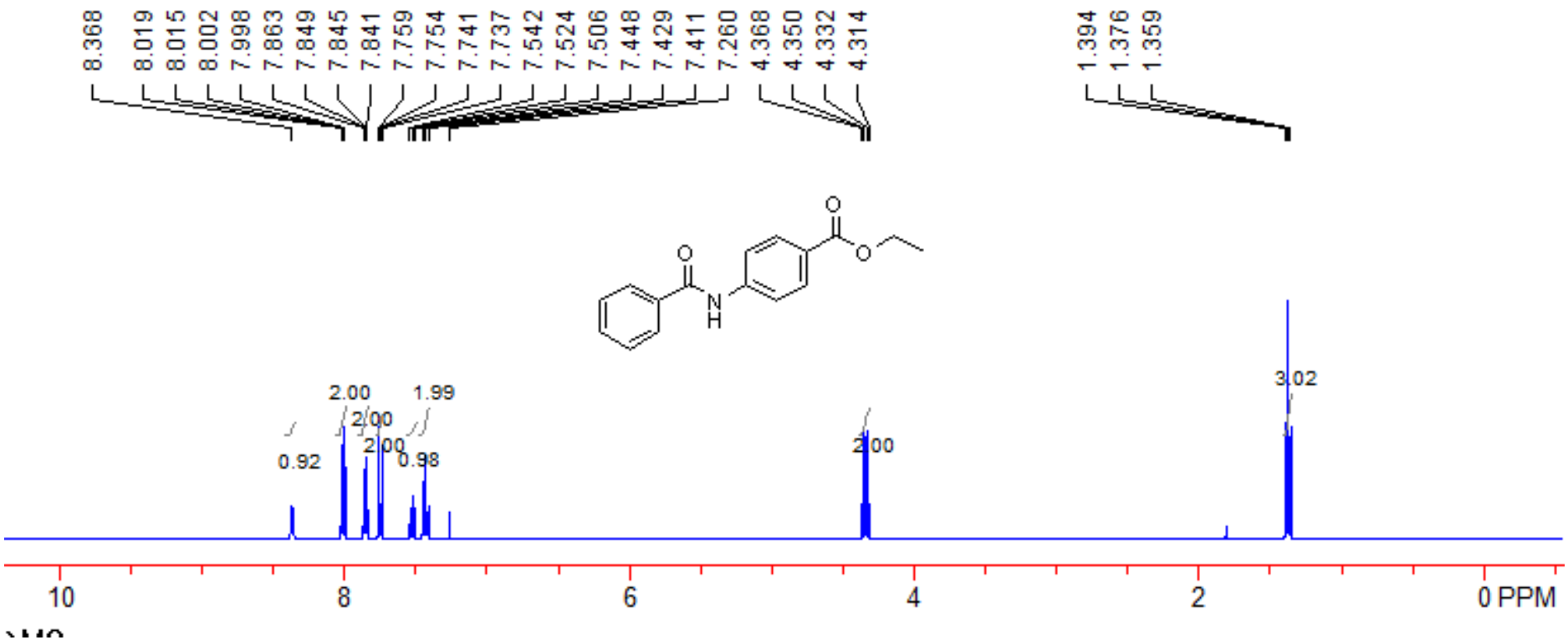
$^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) spectrum of **2l**.



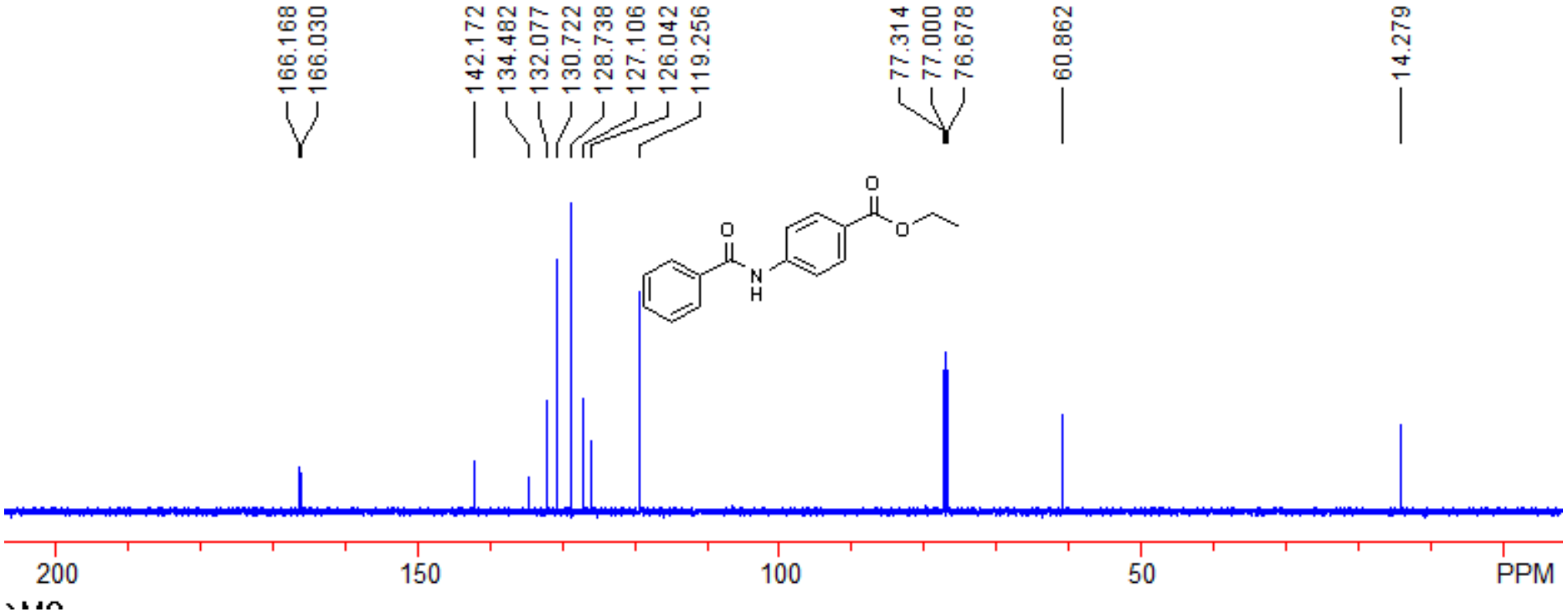
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2m**.



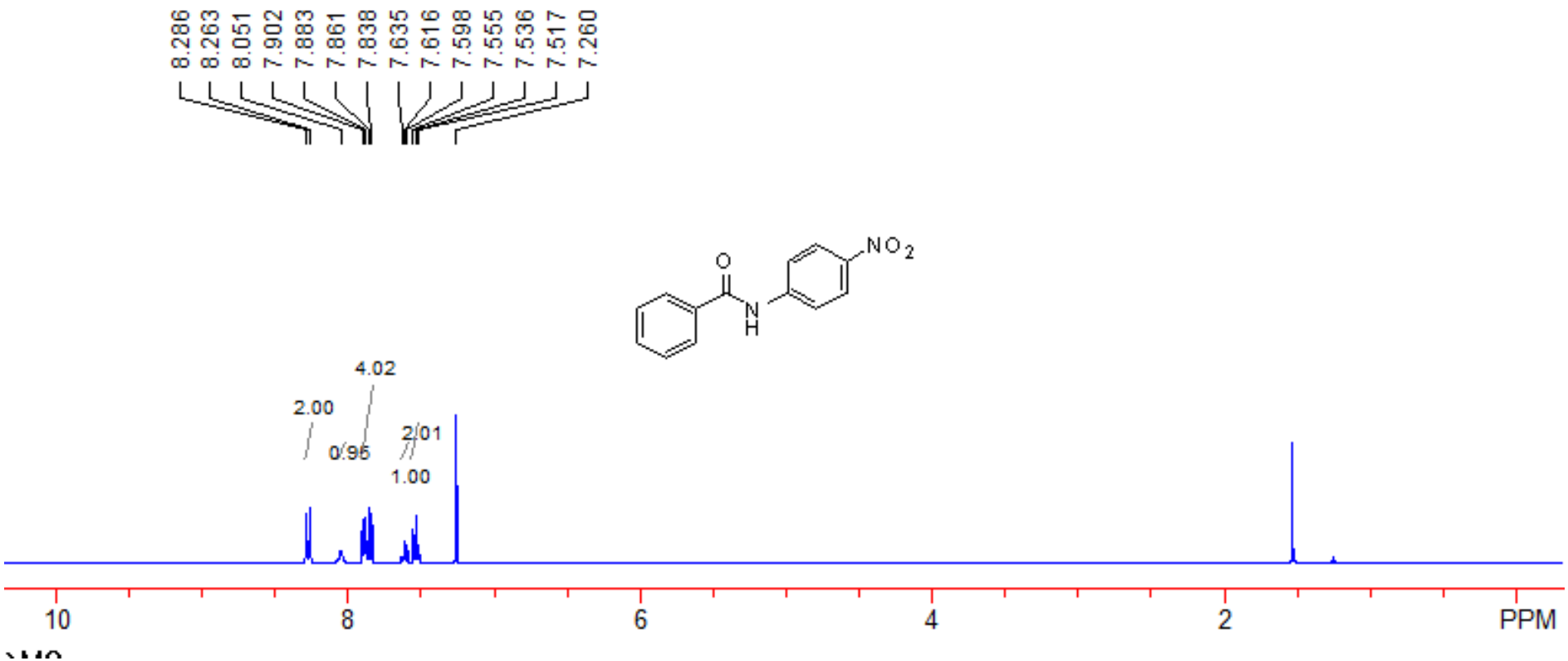
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2m**.



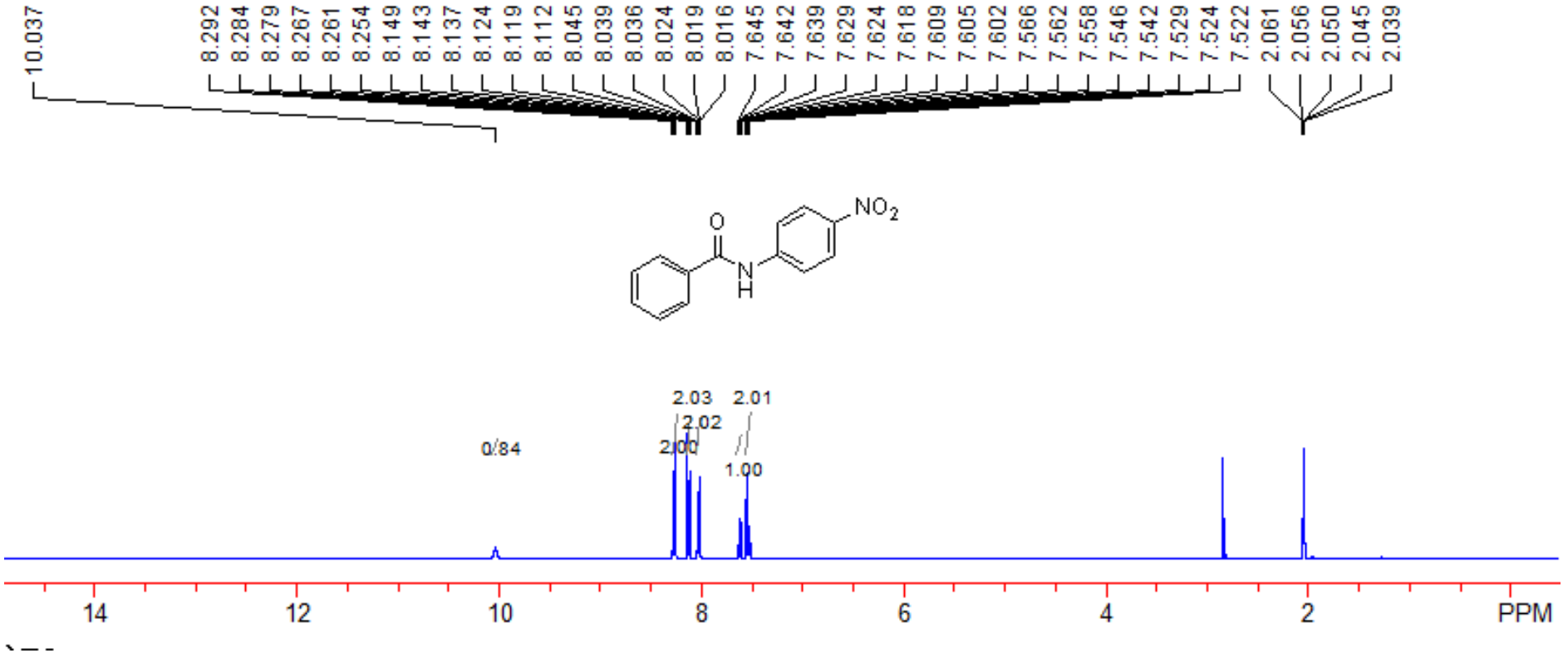
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2n**.



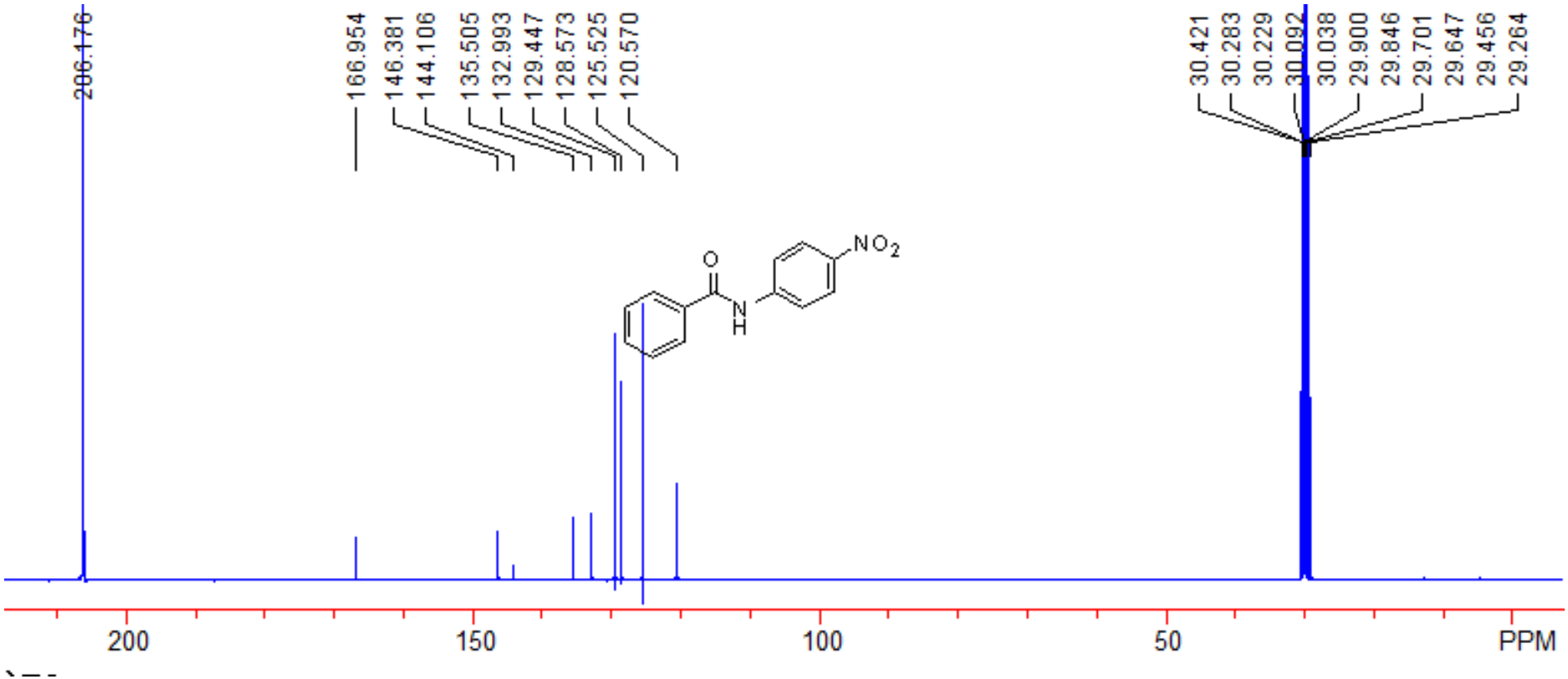
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2n**.



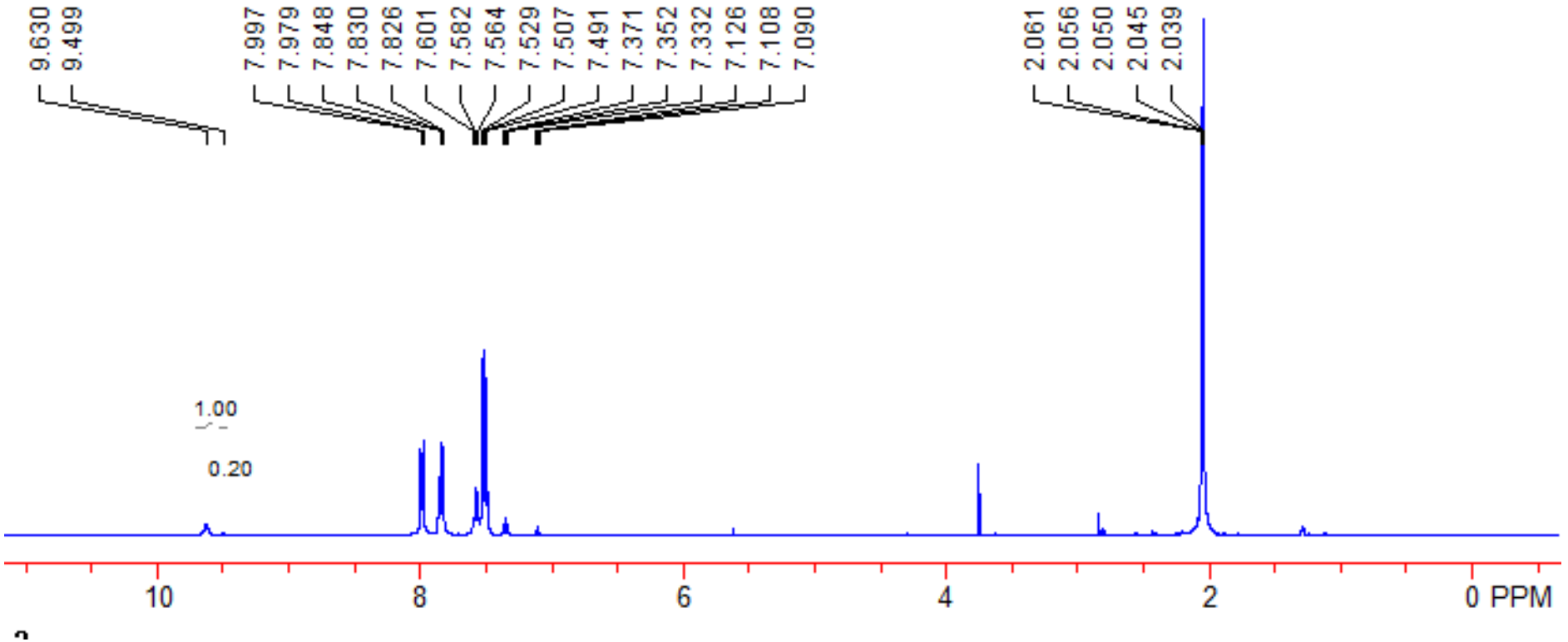
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2o**.



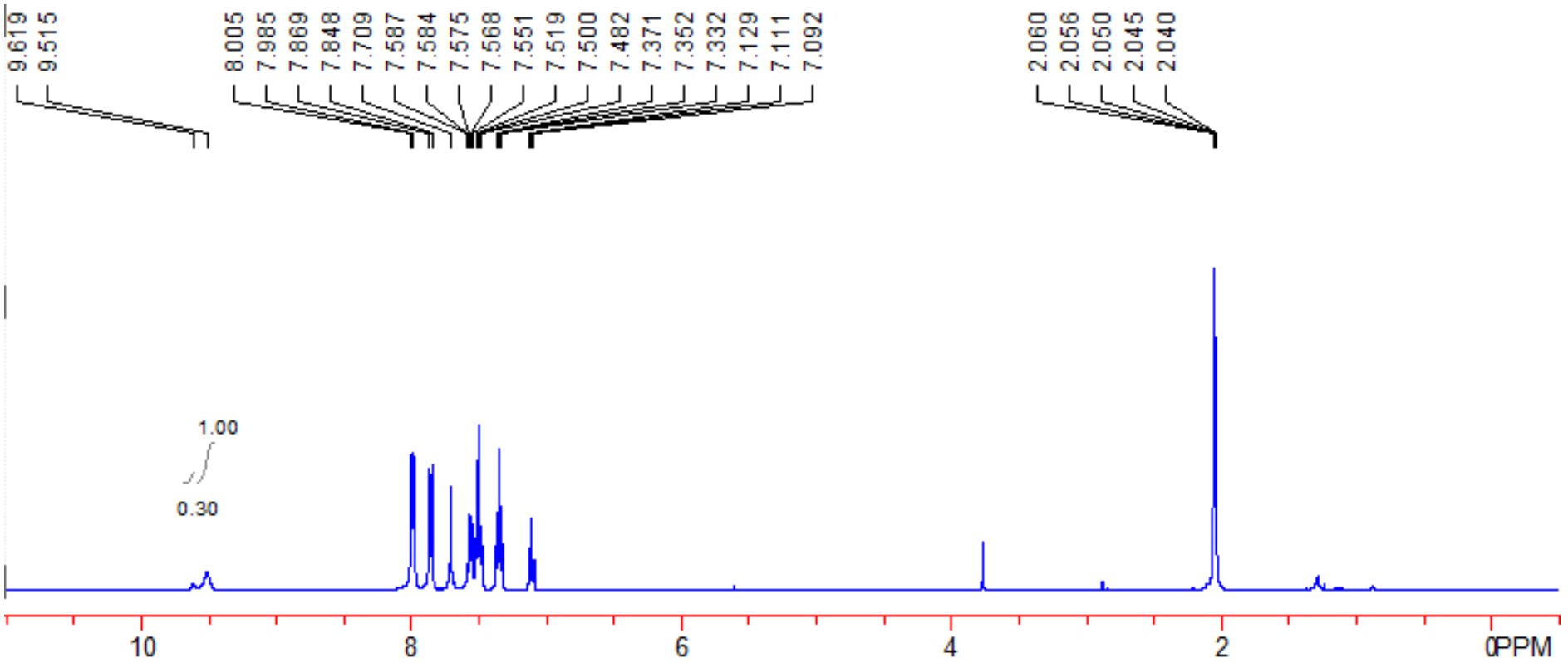
^{1}H NMR (400 MHz, acetone-d_6) spectrum of **2o**.



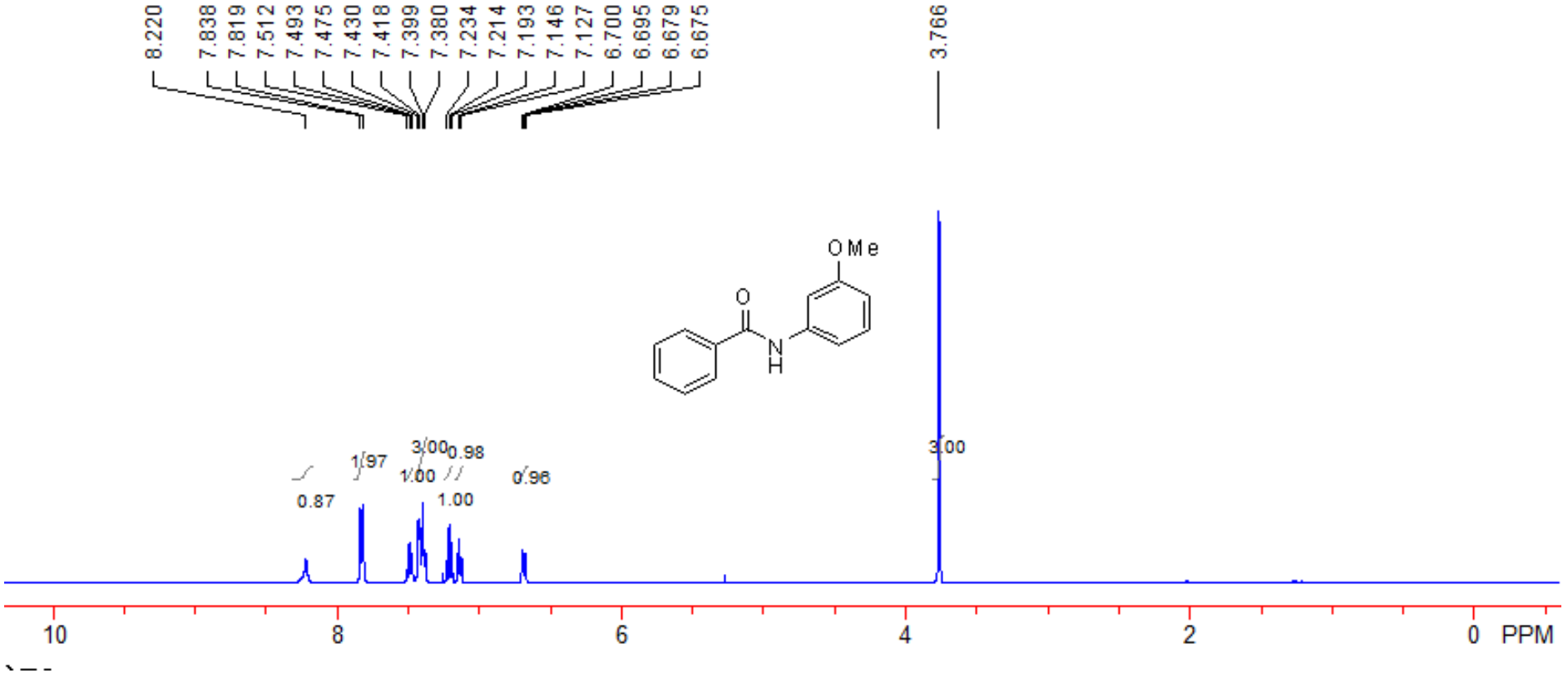
$^{13}C\{^{1}H\}$ NMR (100 MHz, acetone-d_6) spectrum of **2o**.



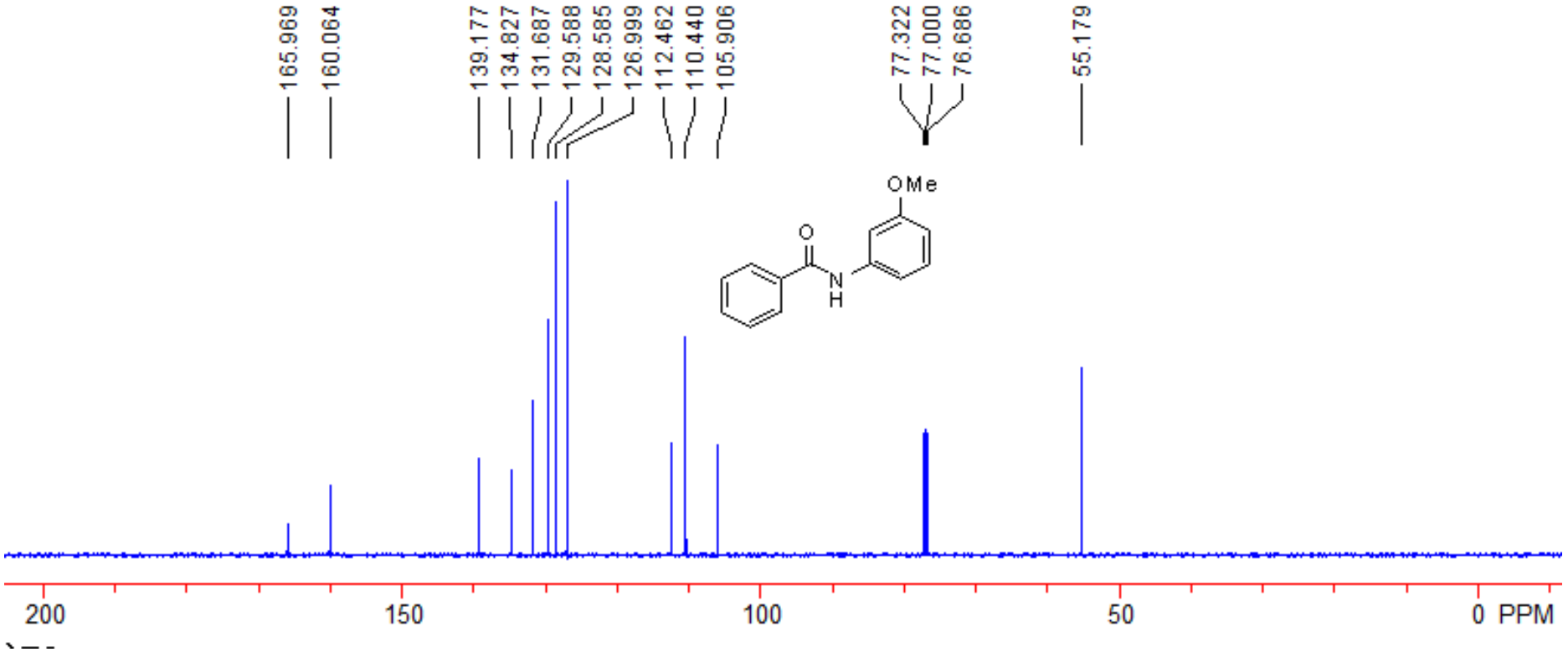
^{1}H NMR (400 MHz, acetone-d_6) spectrum of **2p** containing of 26% of **2i**.



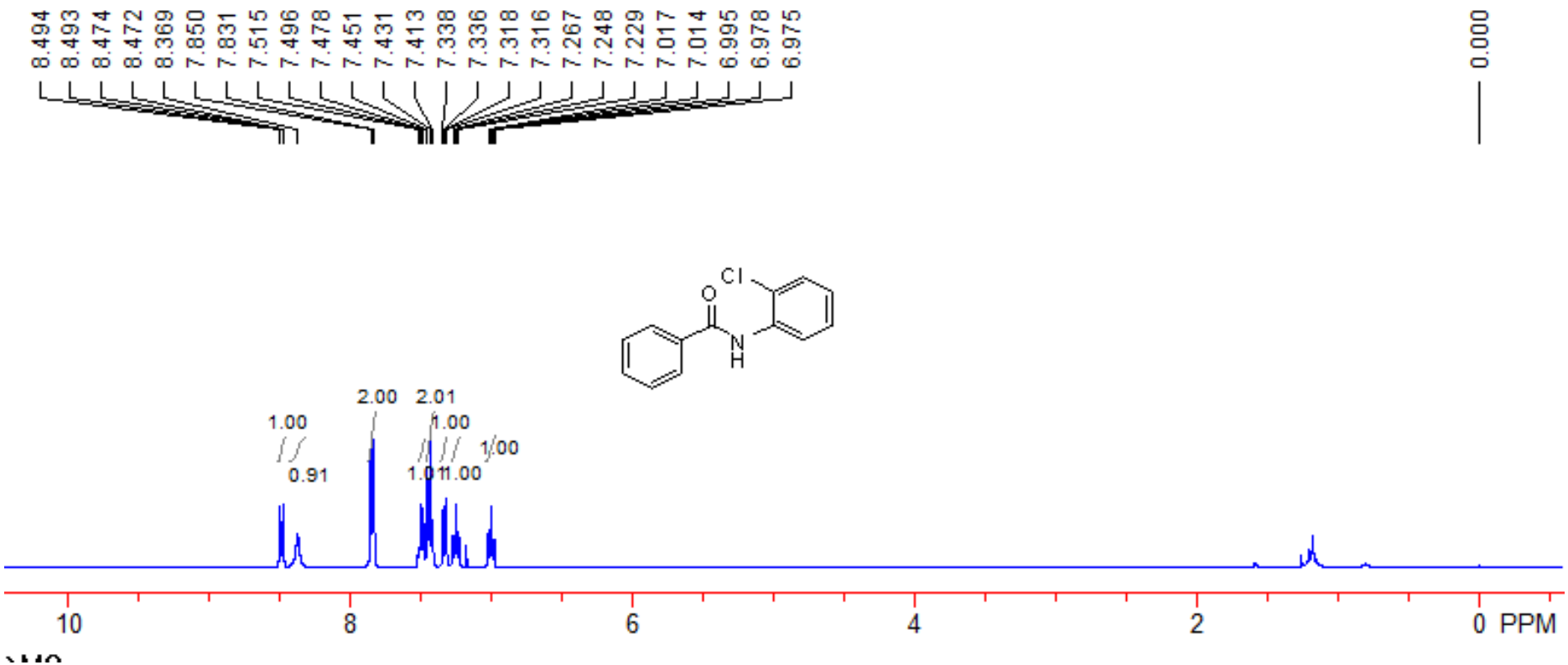
^{1}H NMR (400 MHz, acetone-d_6) spectrum of **2q** containing of 76% of **2i**.



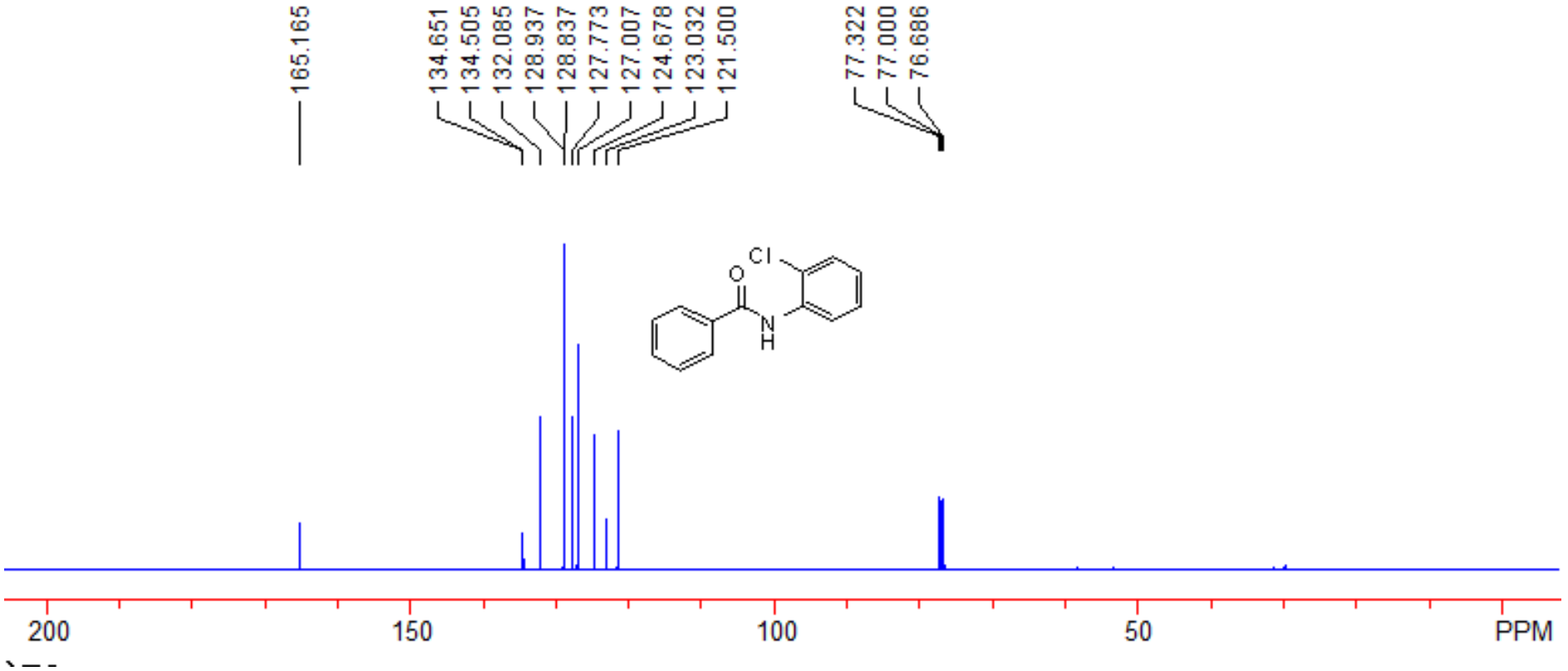
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2r**.



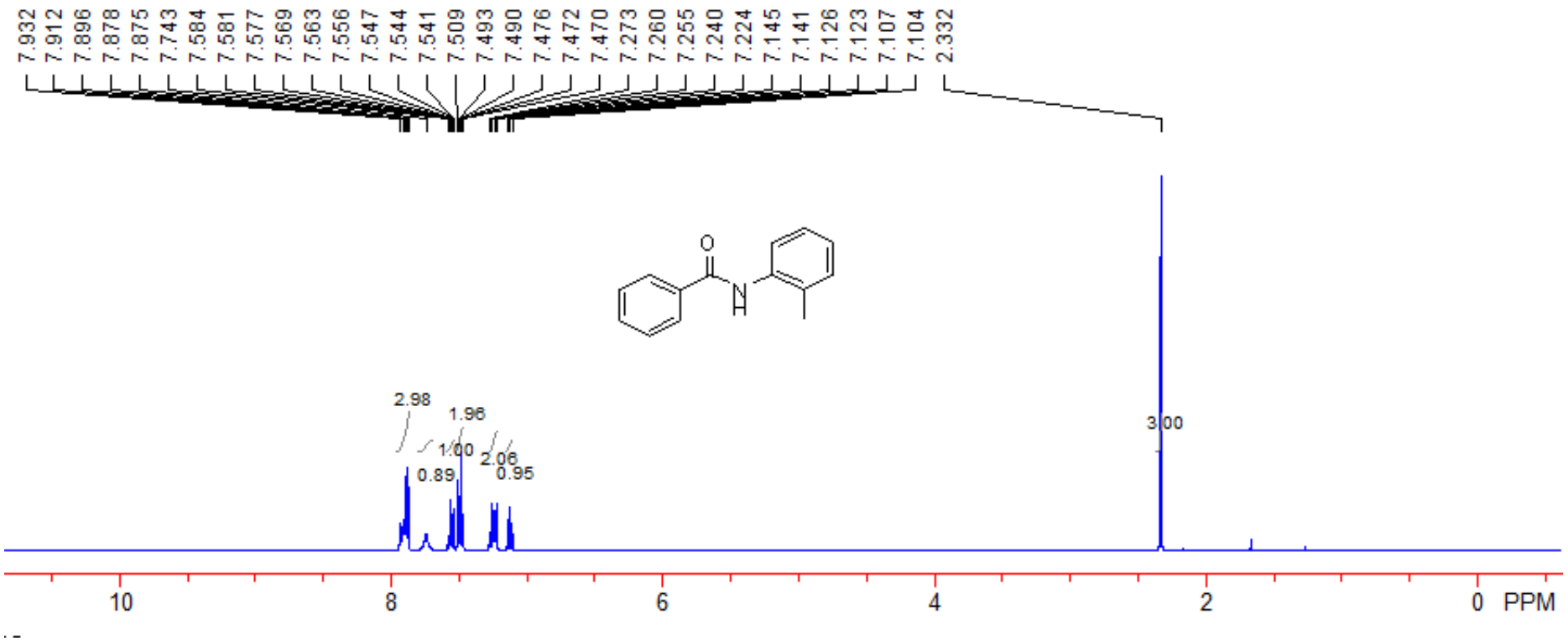
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2r**.



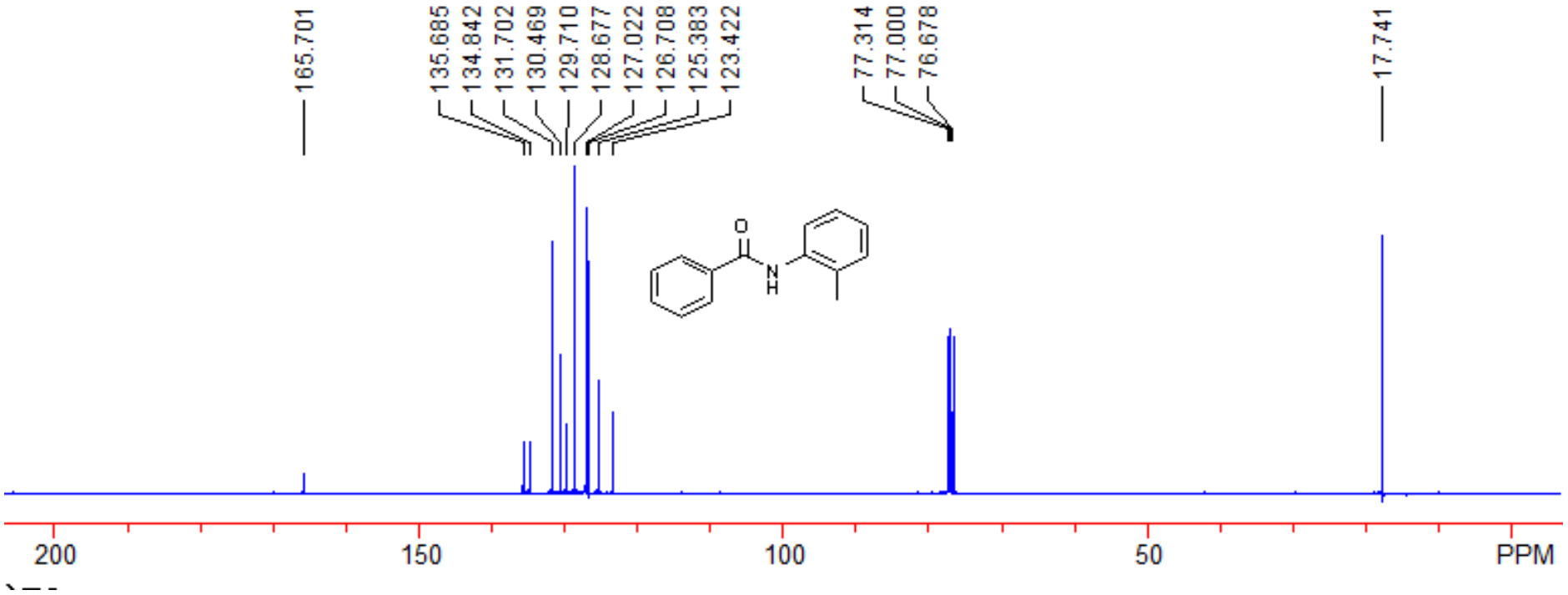
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2s**.



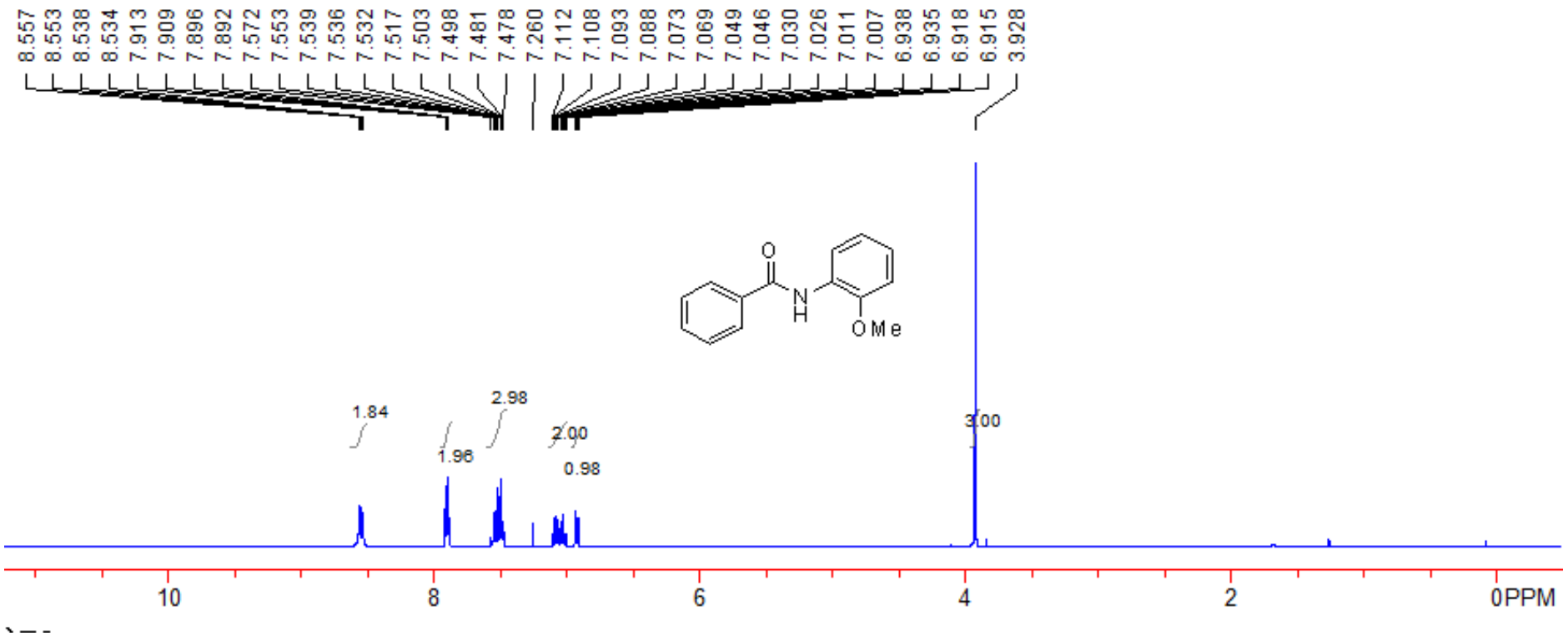
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2s**.



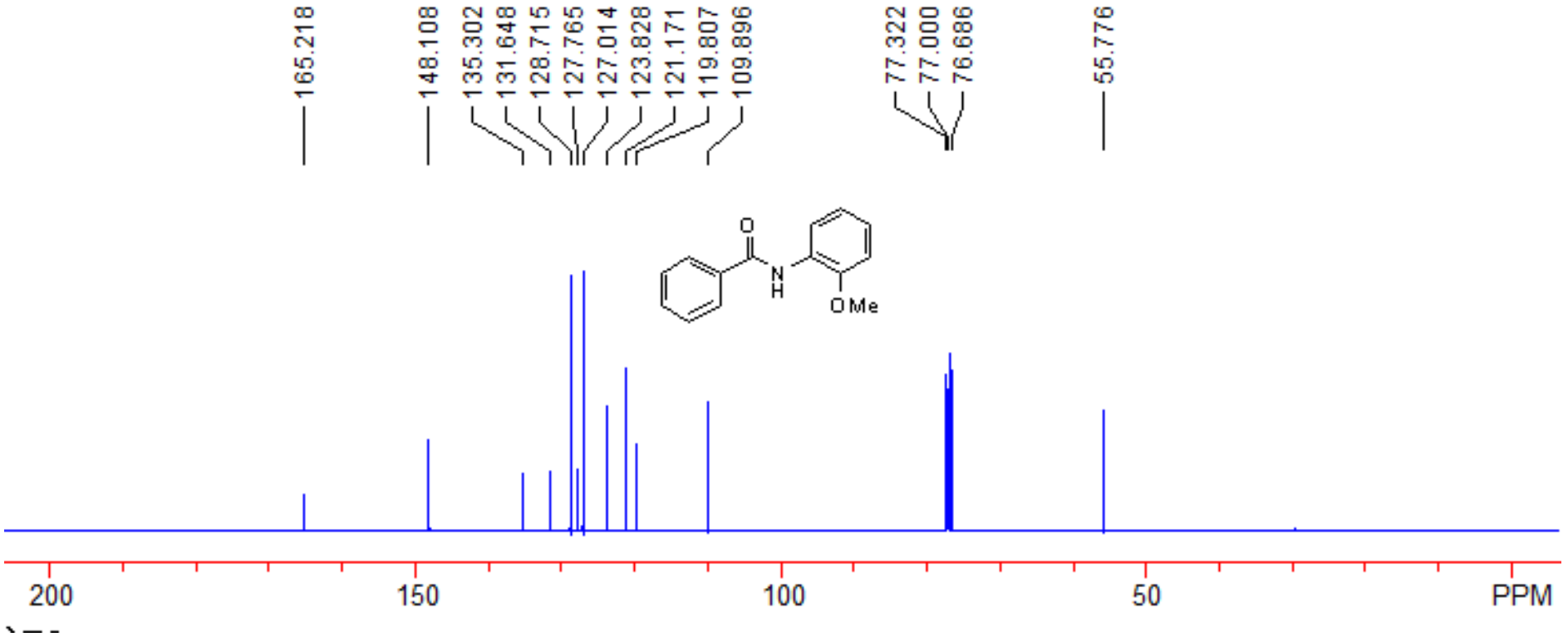
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2t**.



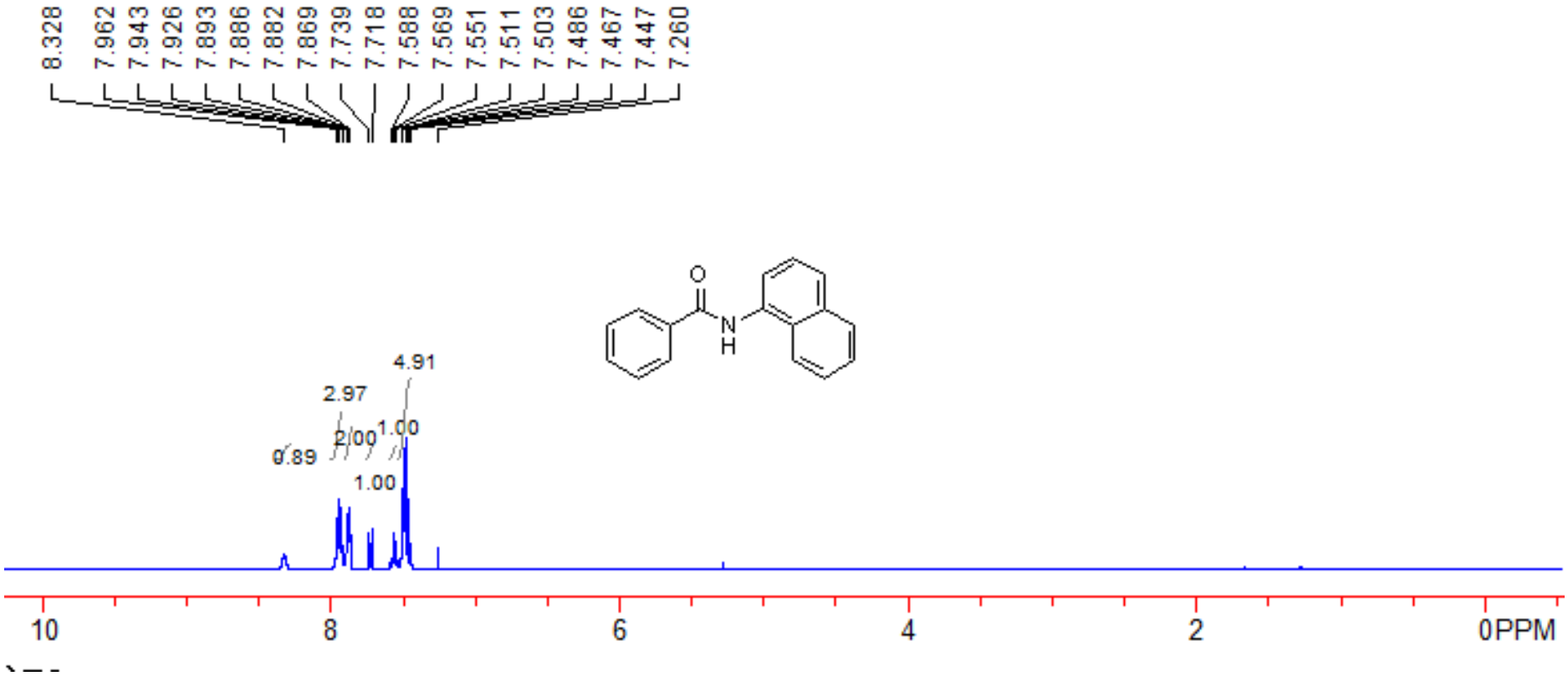
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2t**.



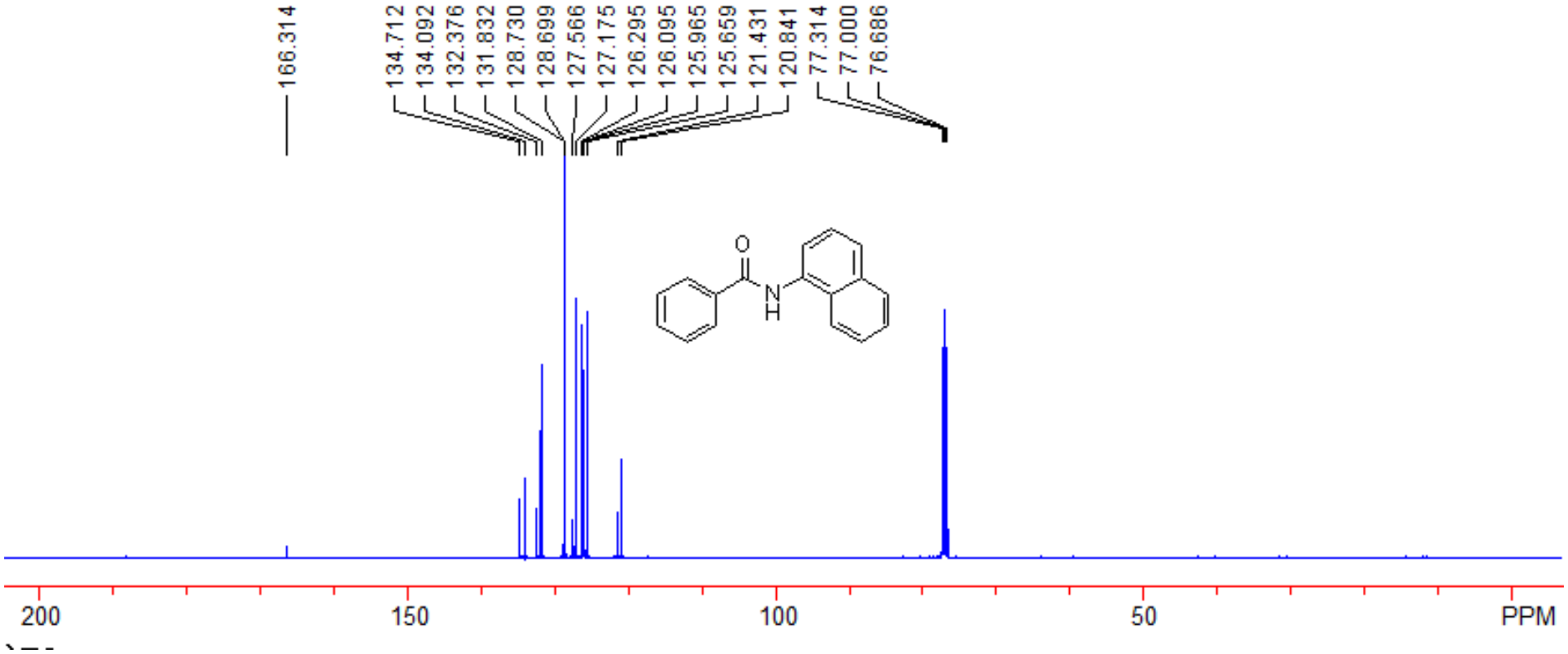
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2u**.



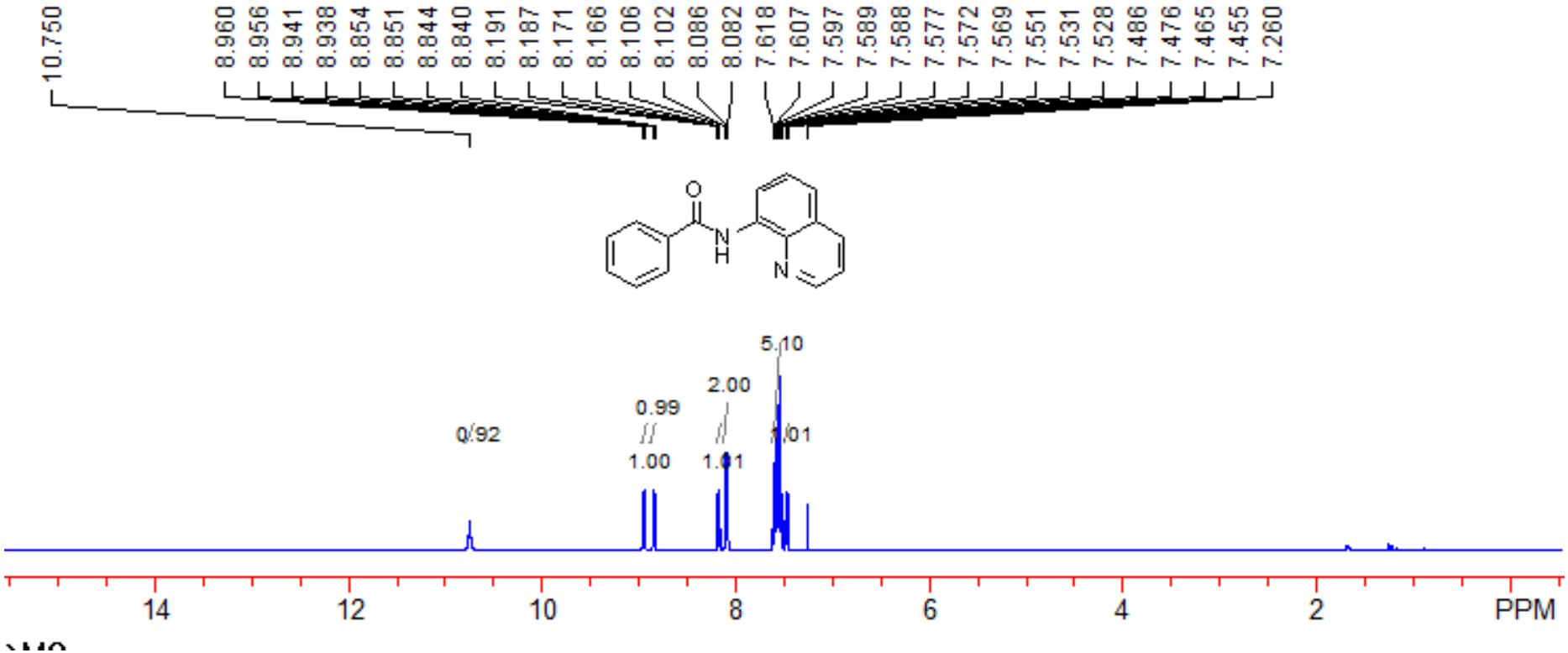
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2u**.



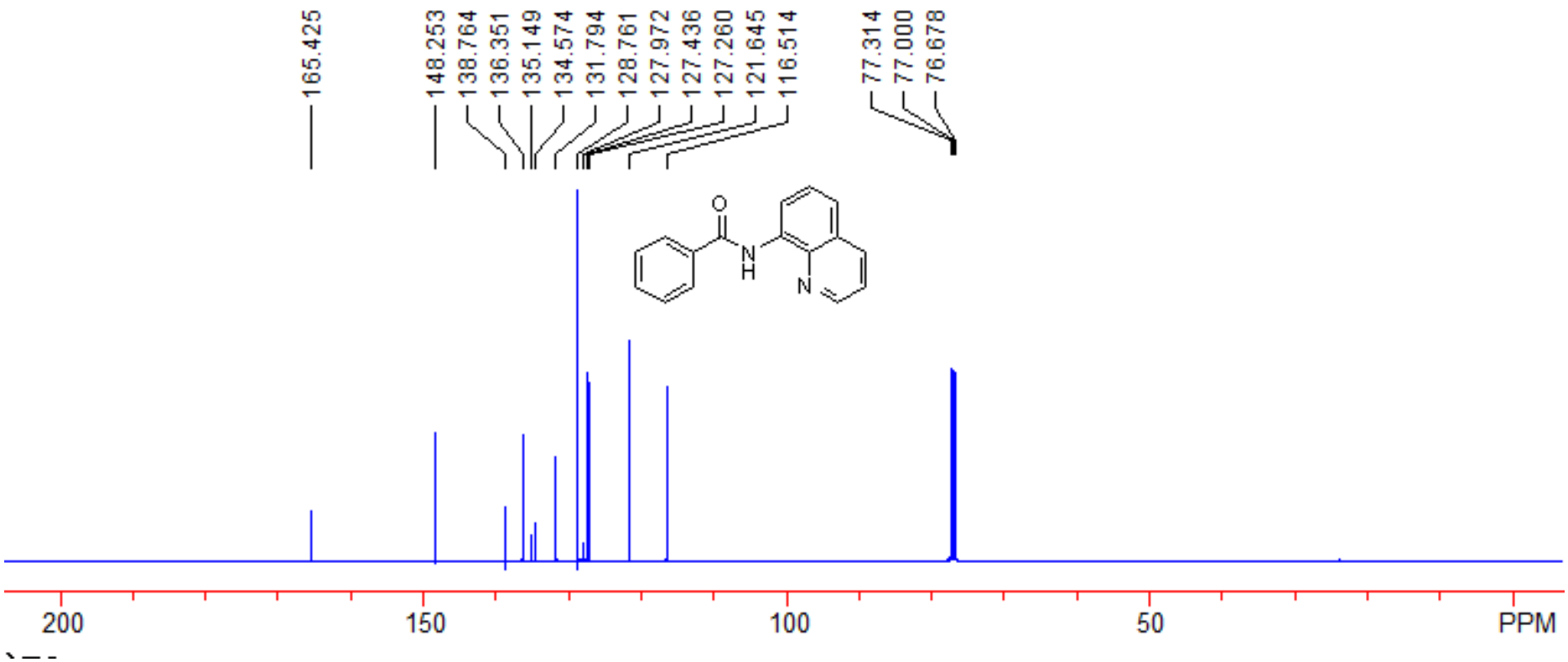
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2v**.



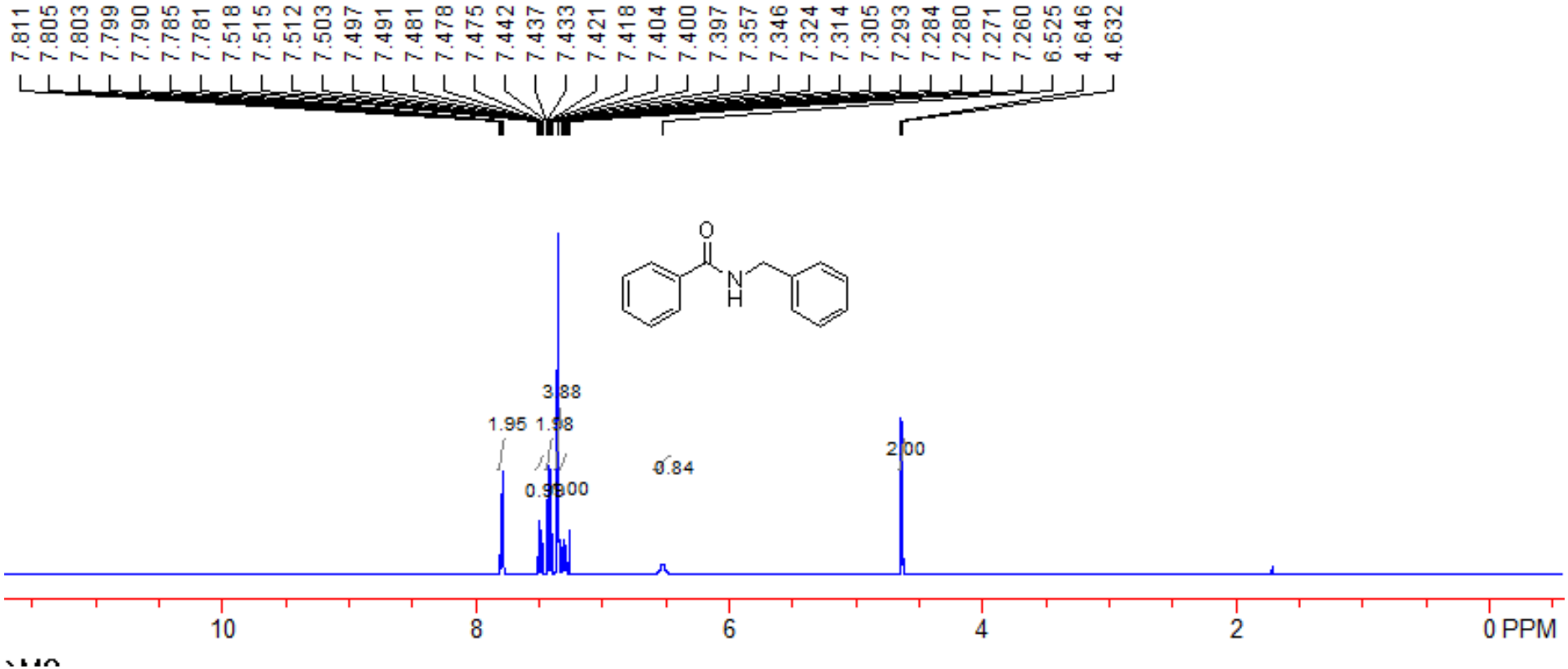
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2v**.



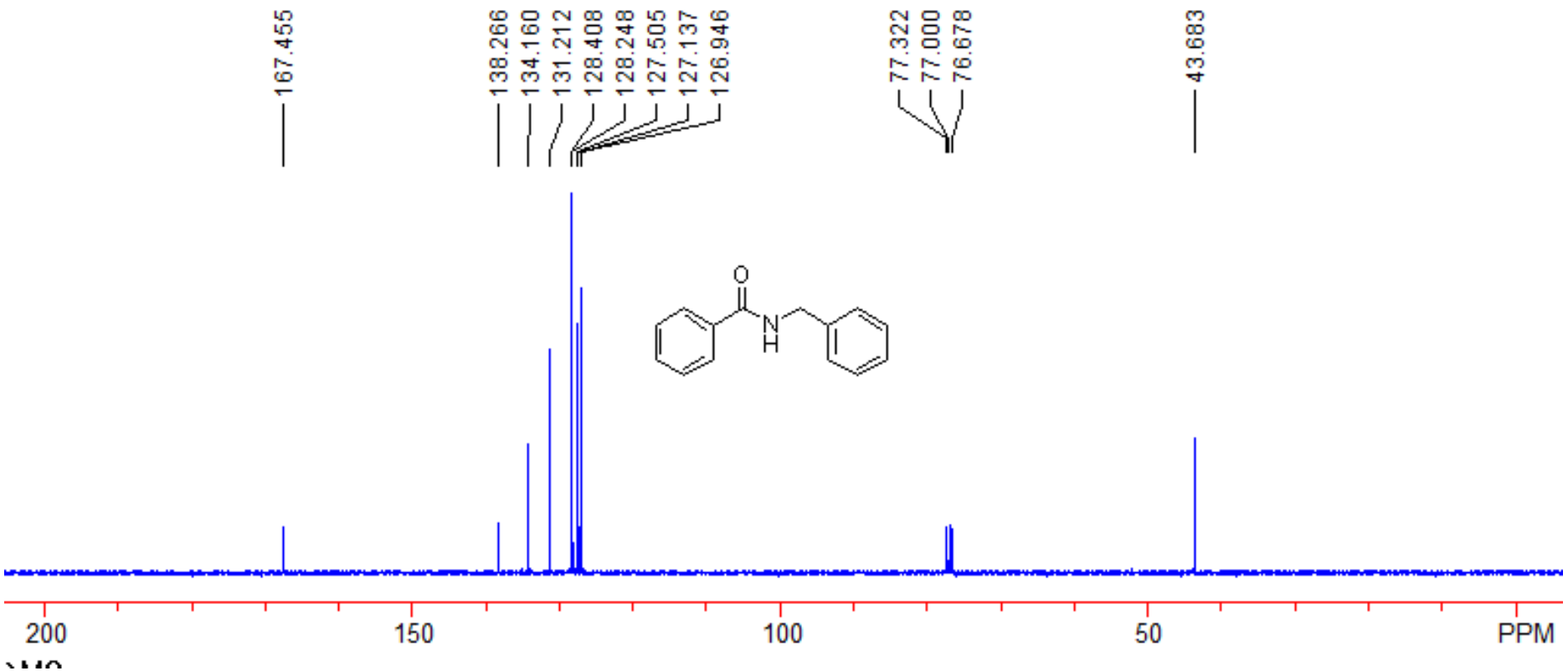
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2w**.



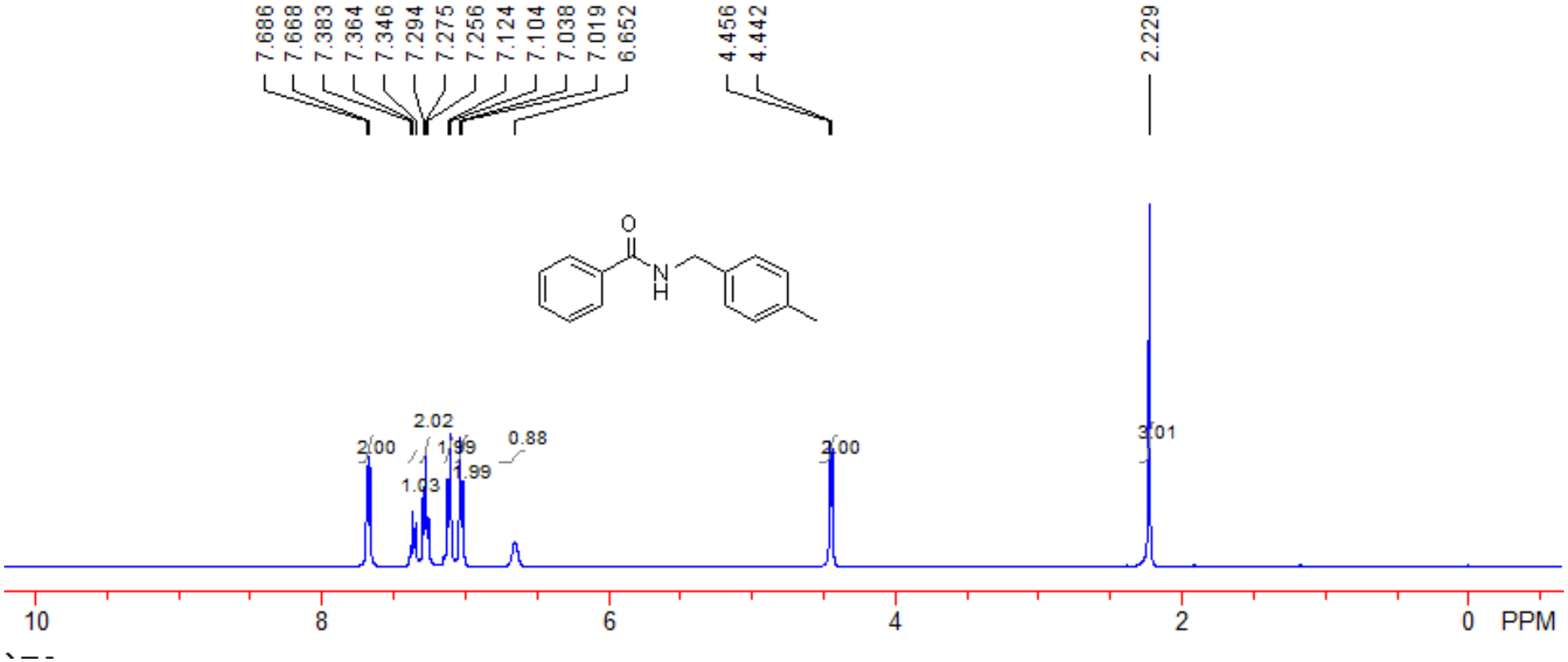
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2w**.



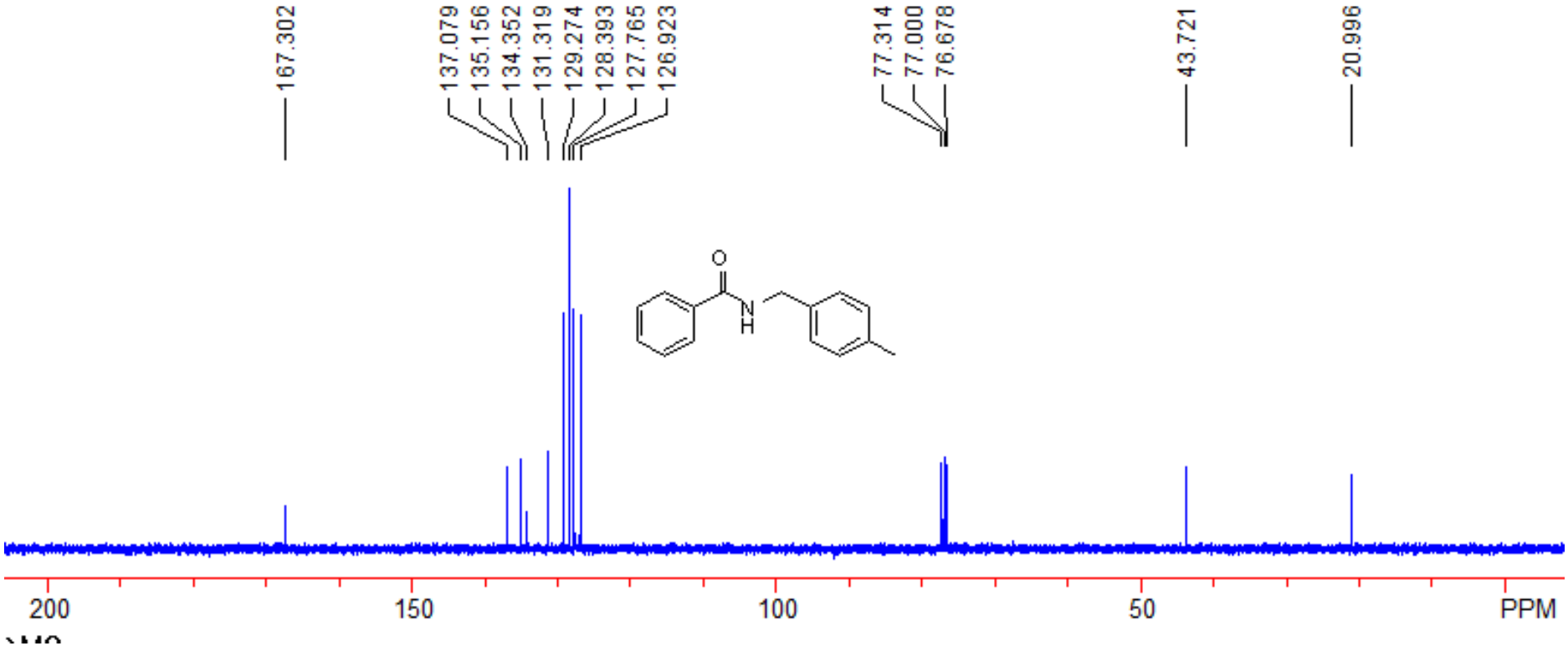
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2x**.



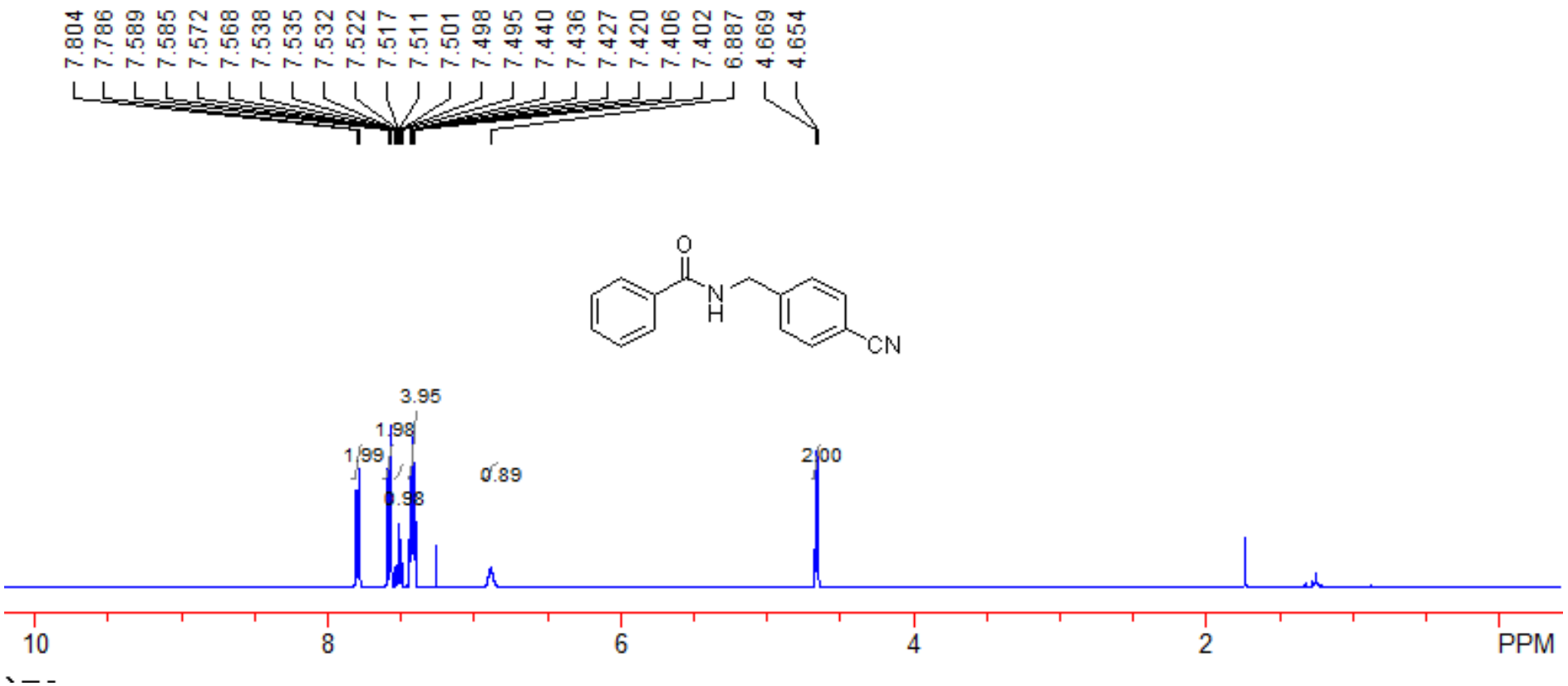
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2x**.



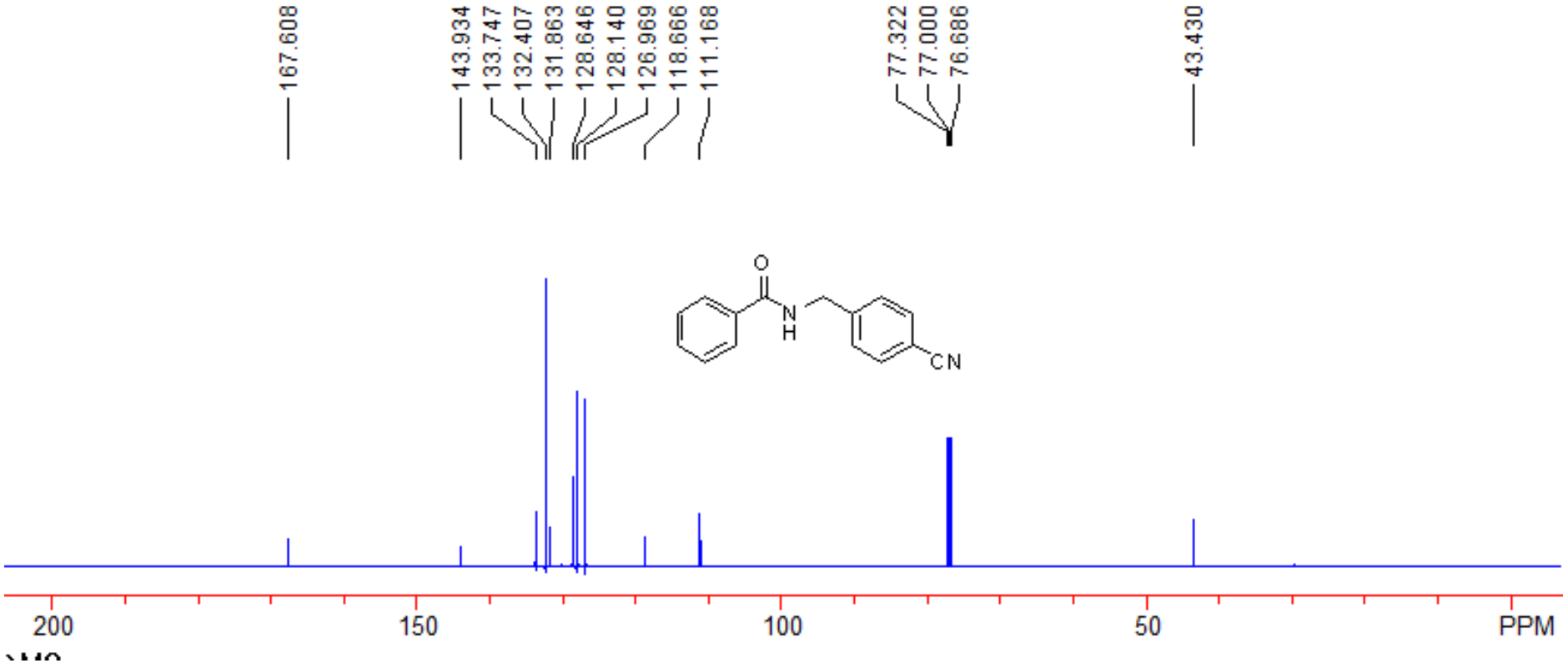
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2y**.



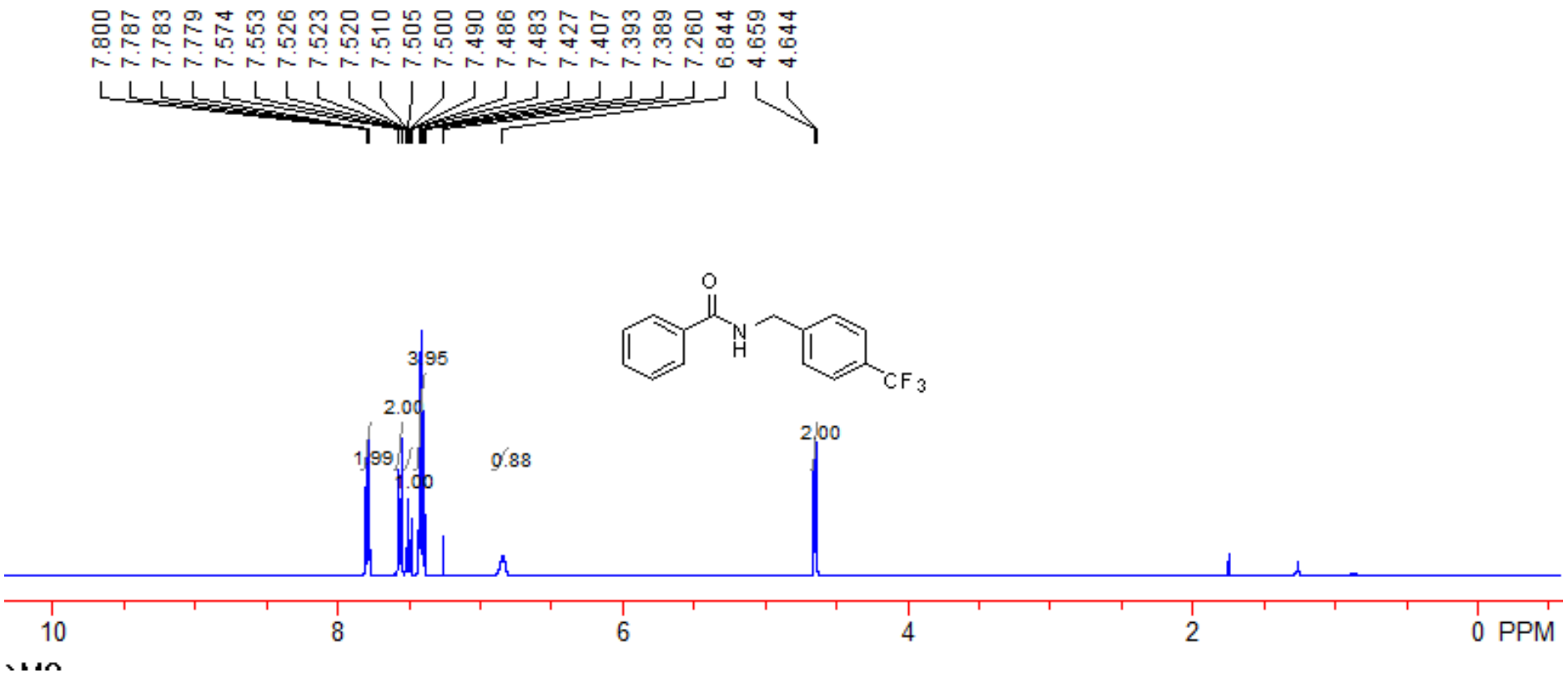
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2y**.



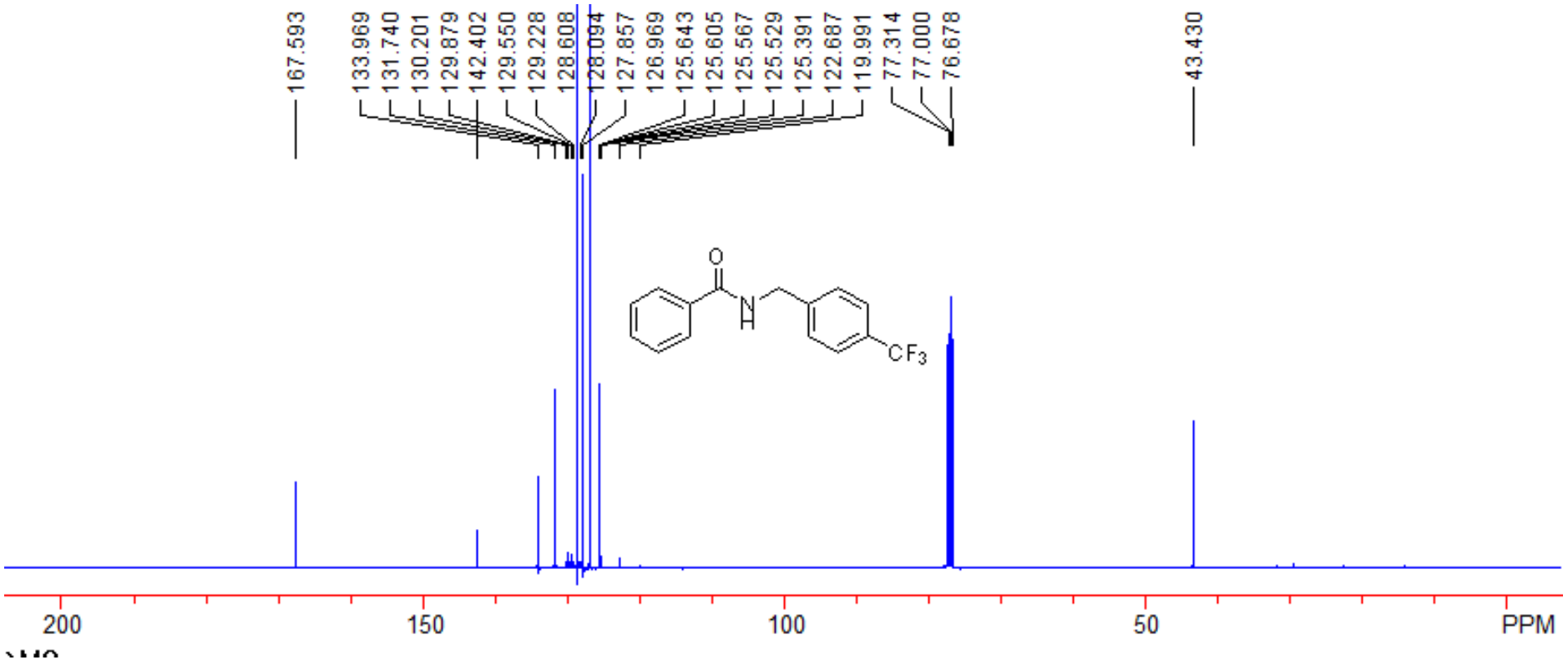
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2z**.



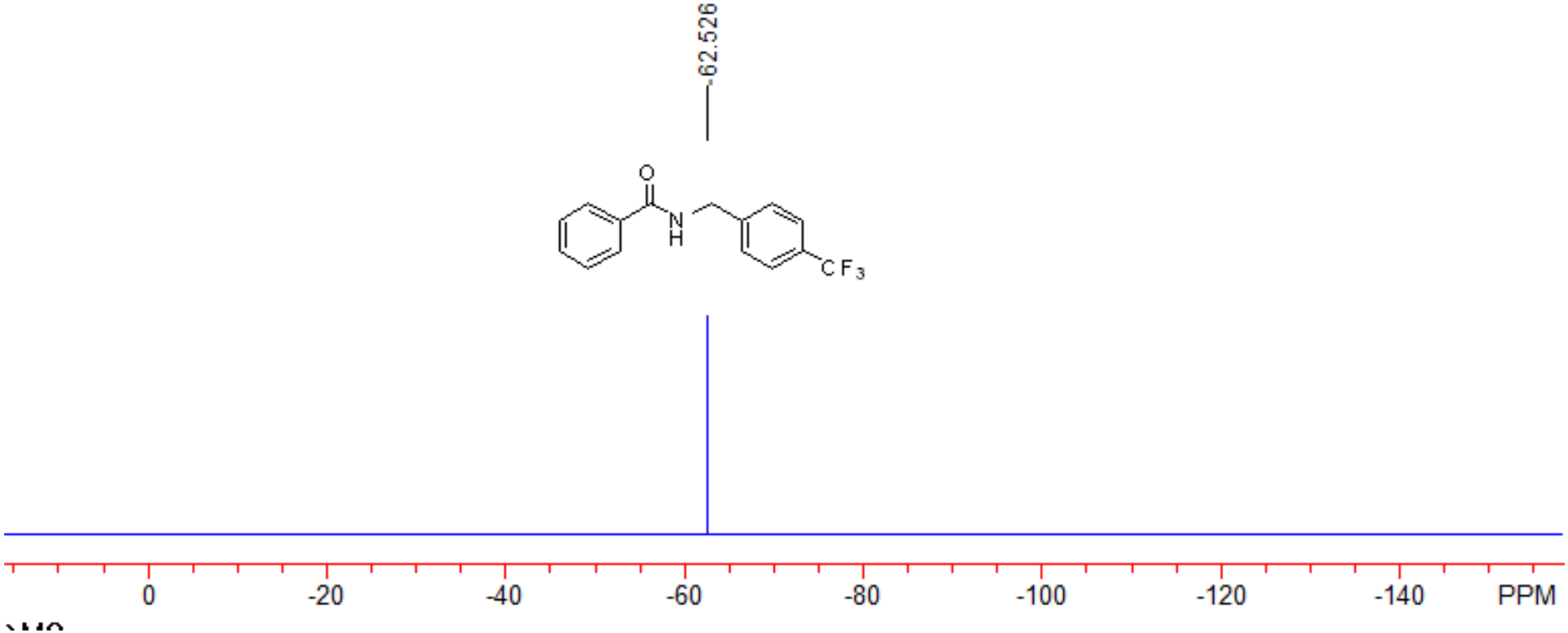
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2z**.



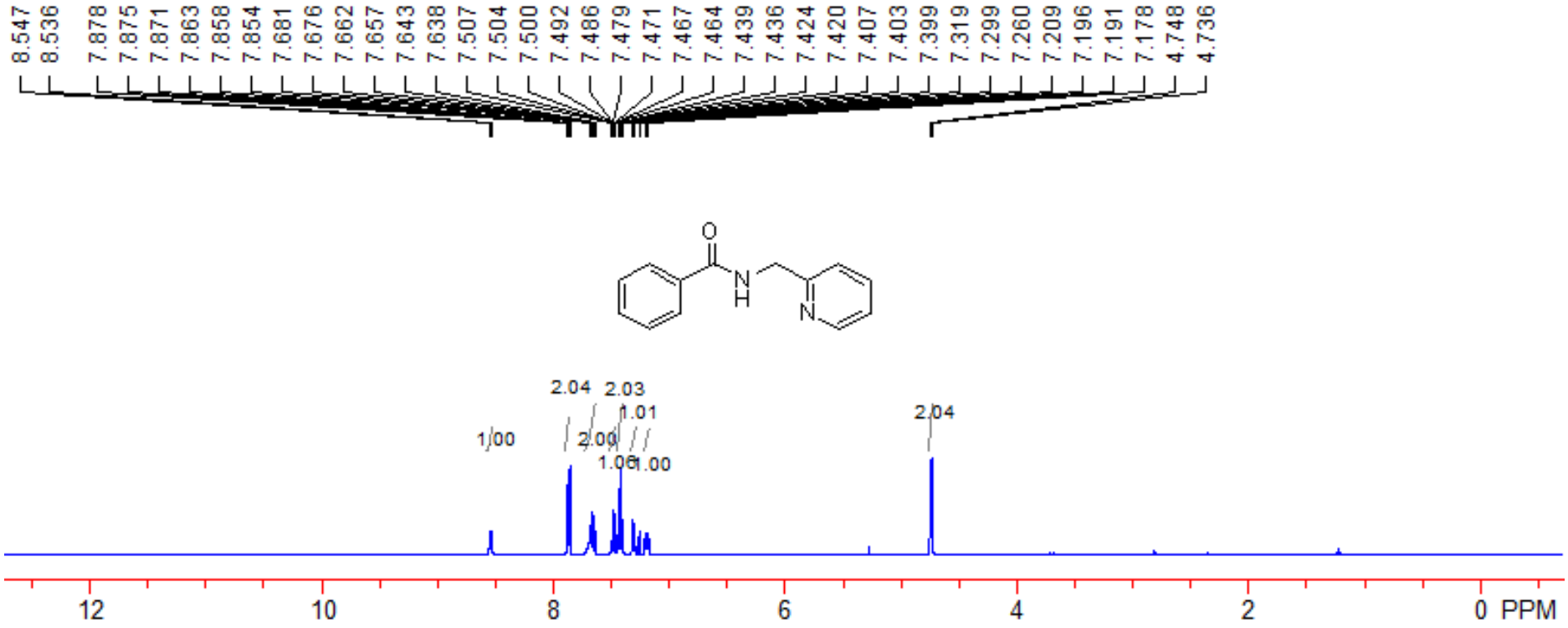
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3a**.



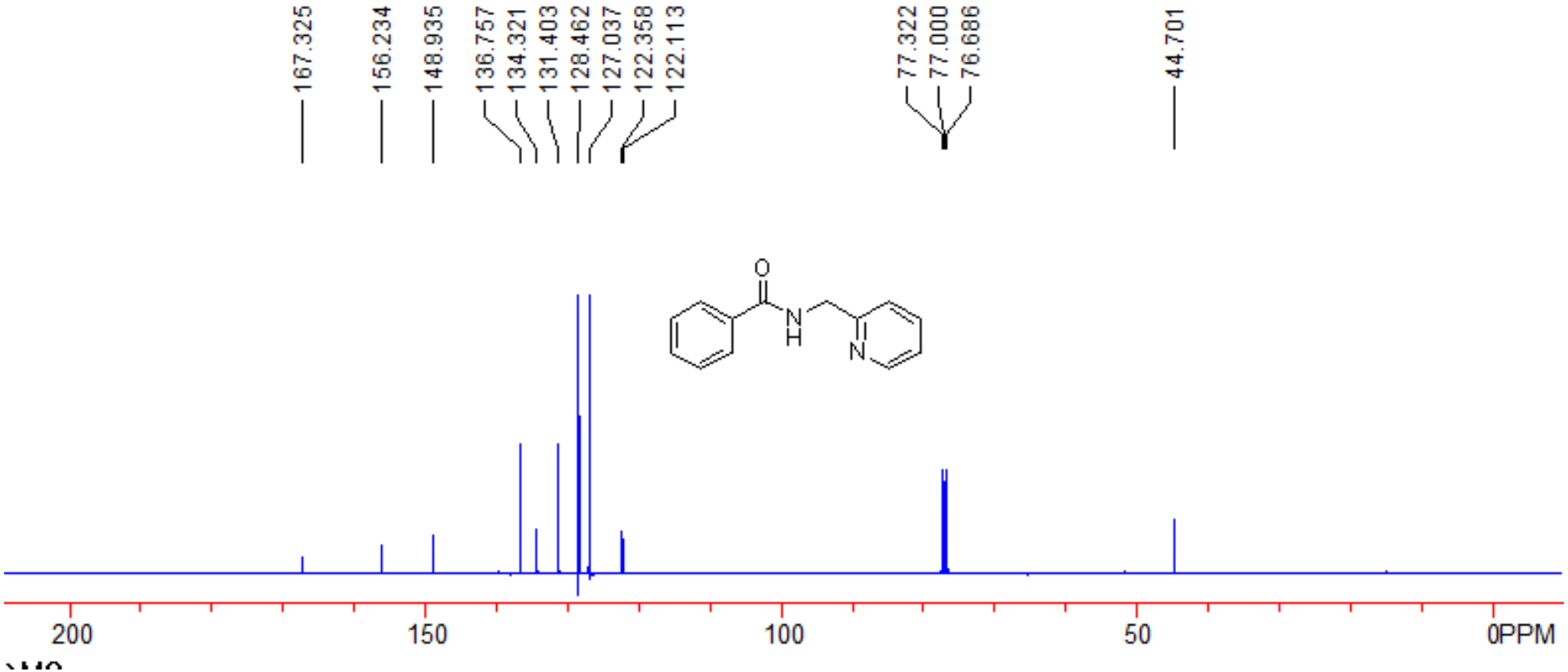
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **3a**.



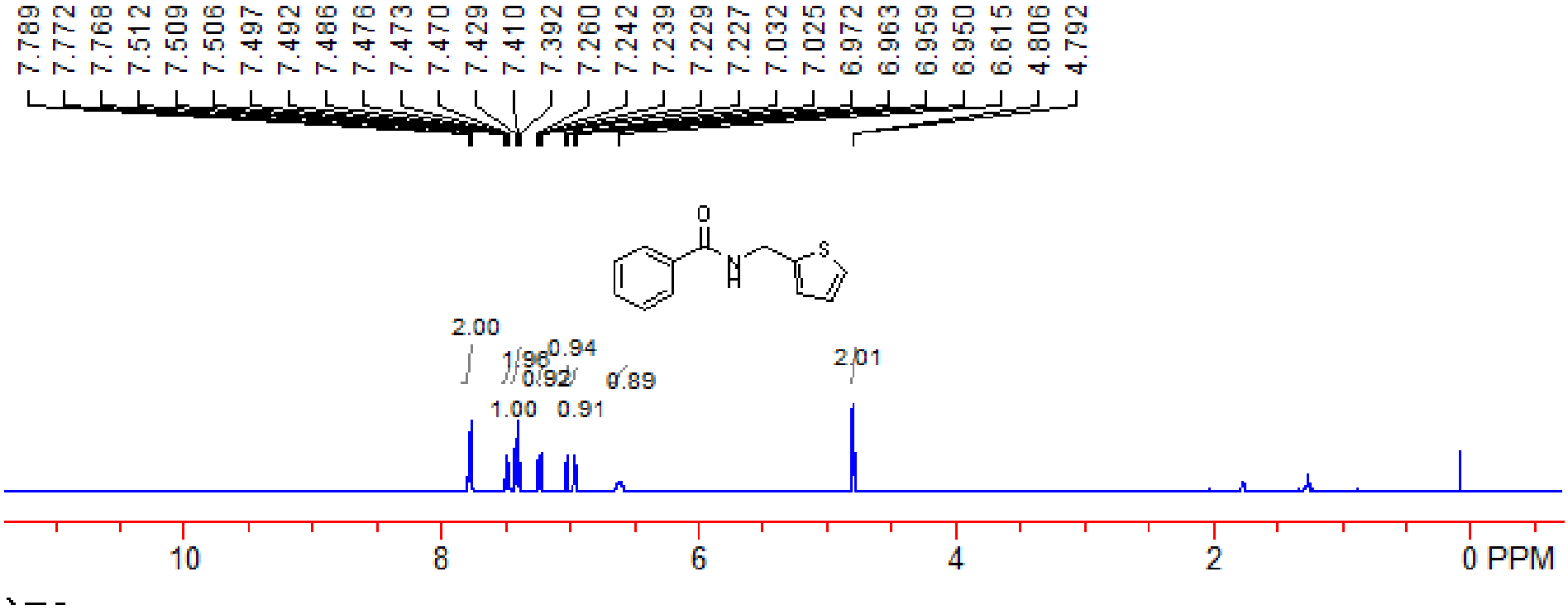
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) spectrum of **3a**.



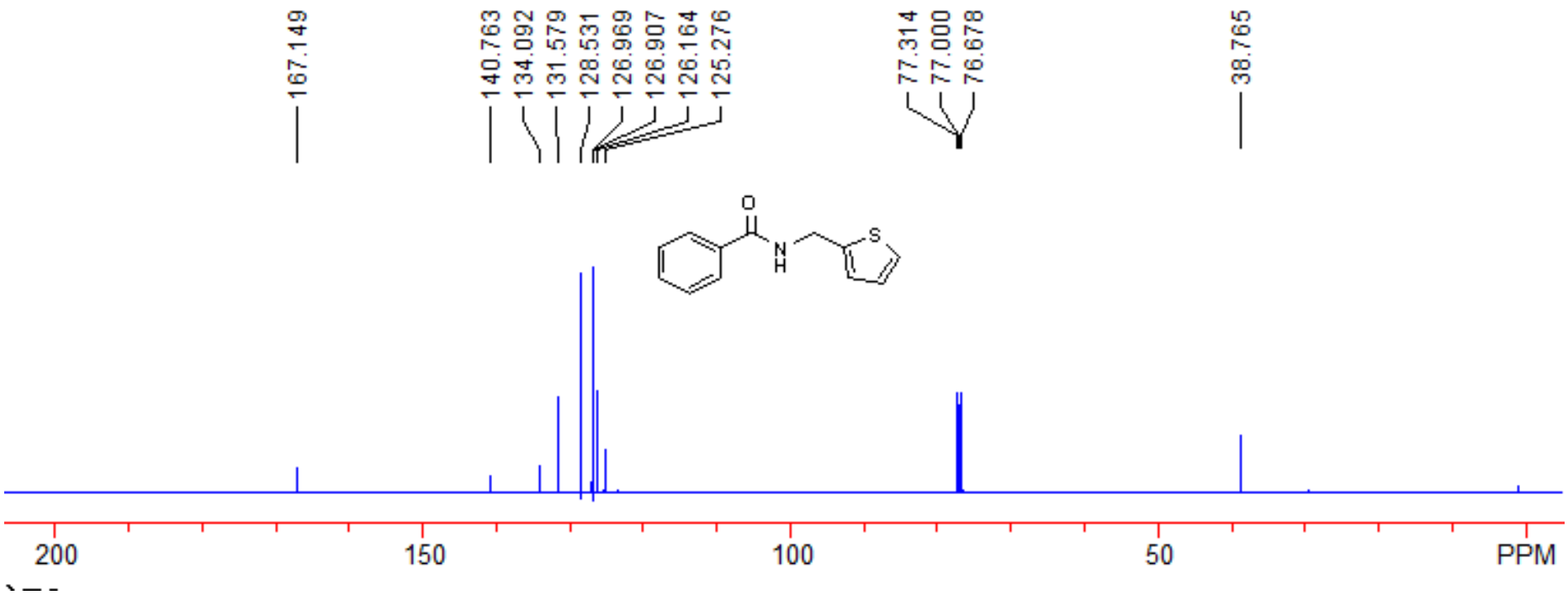
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3b**.



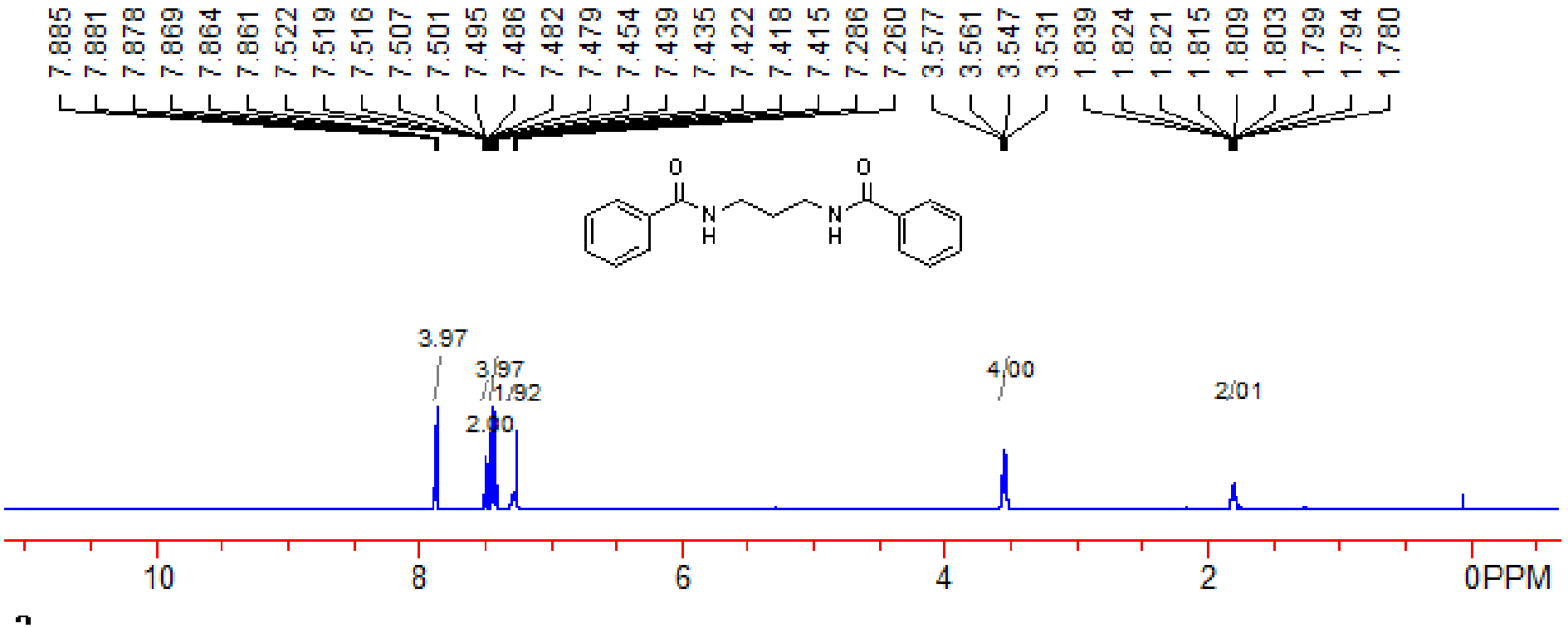
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **3b**.



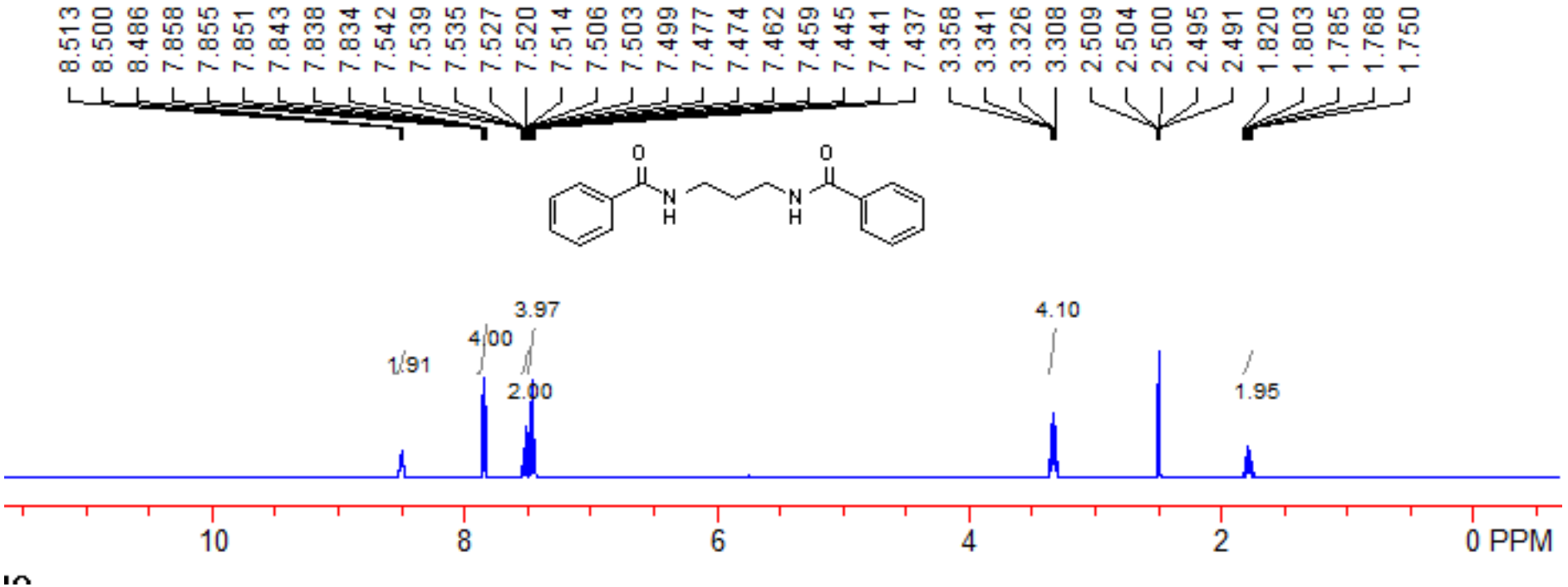
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3c**.



$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **3c**.

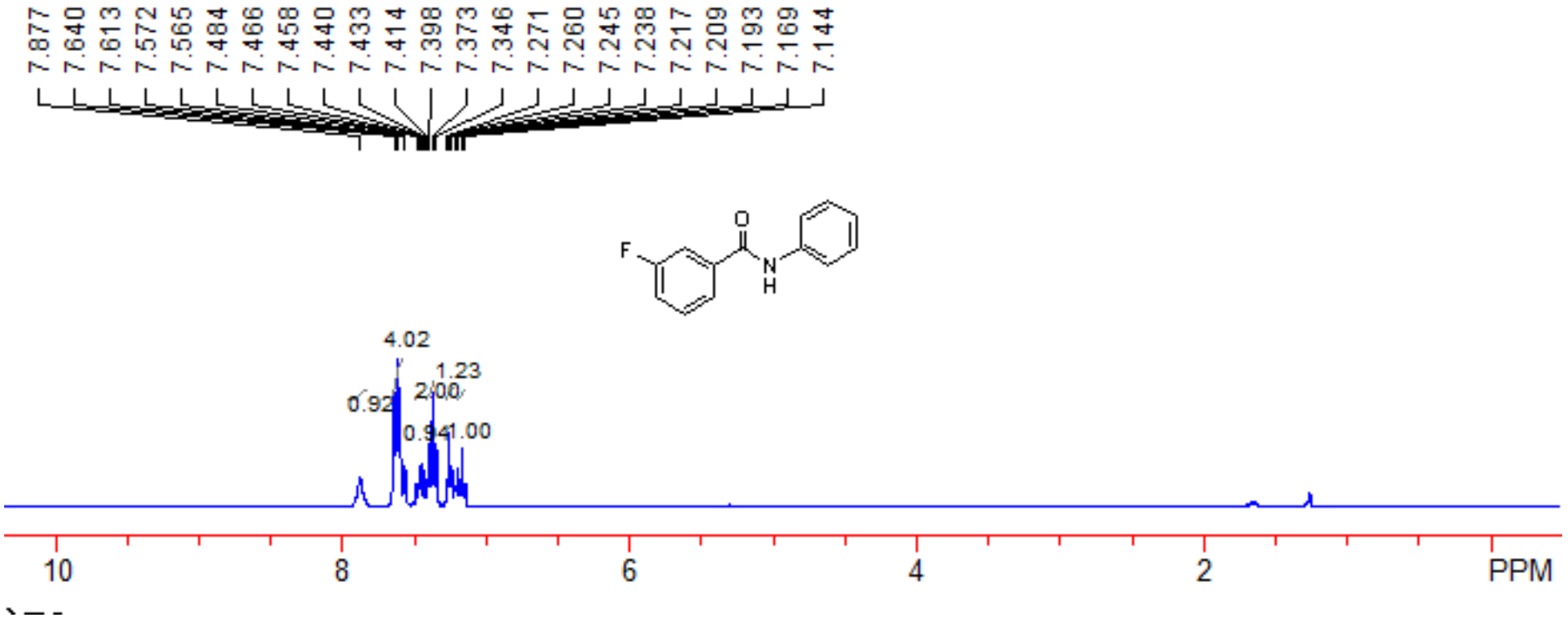


^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3d**.

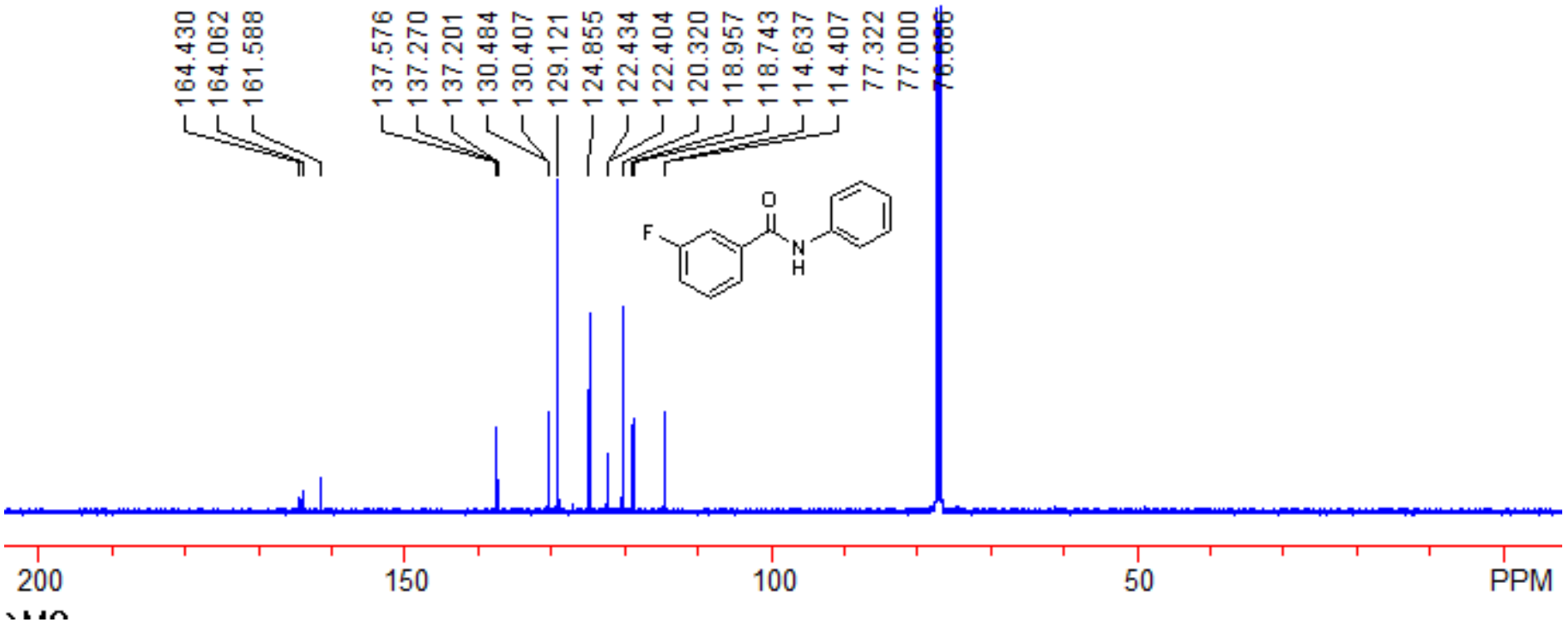


^{1}H NMR (400 MHz, DMSO-d_6) spectrum of **3d**.

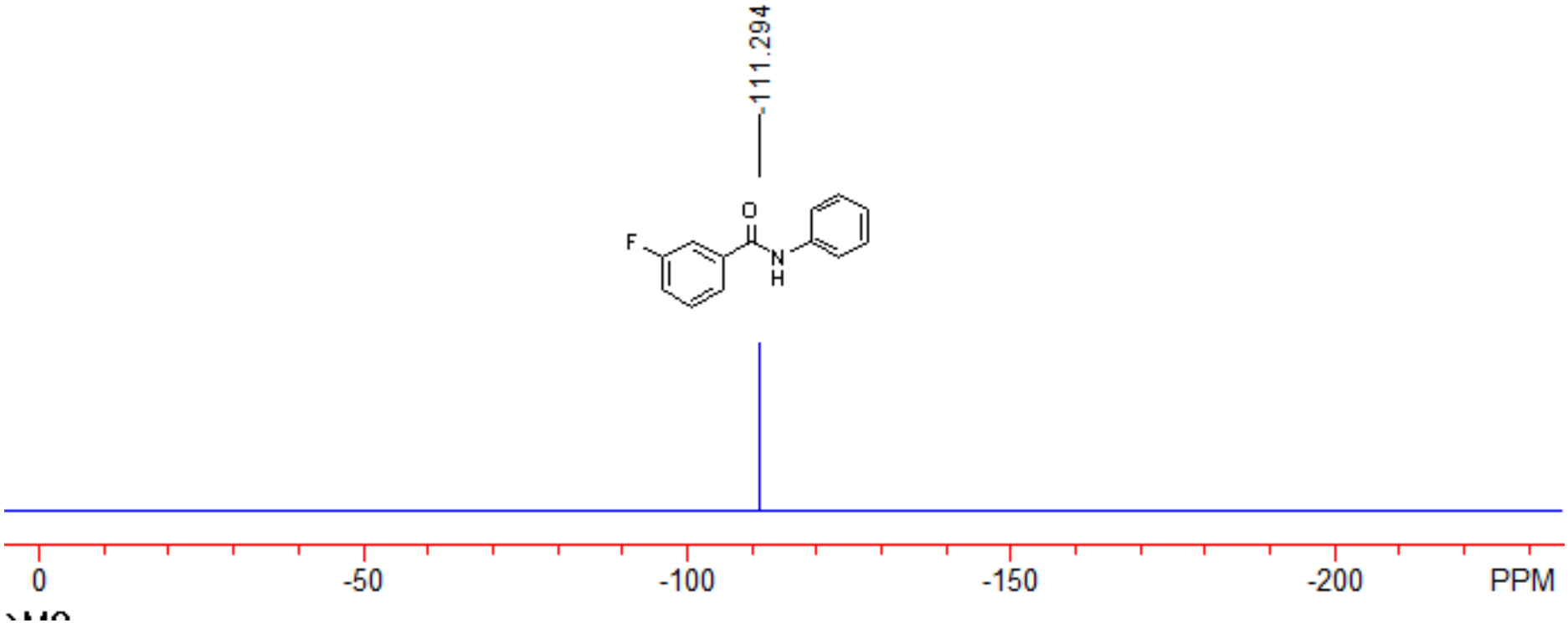
$^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) spectrum of **3d**.



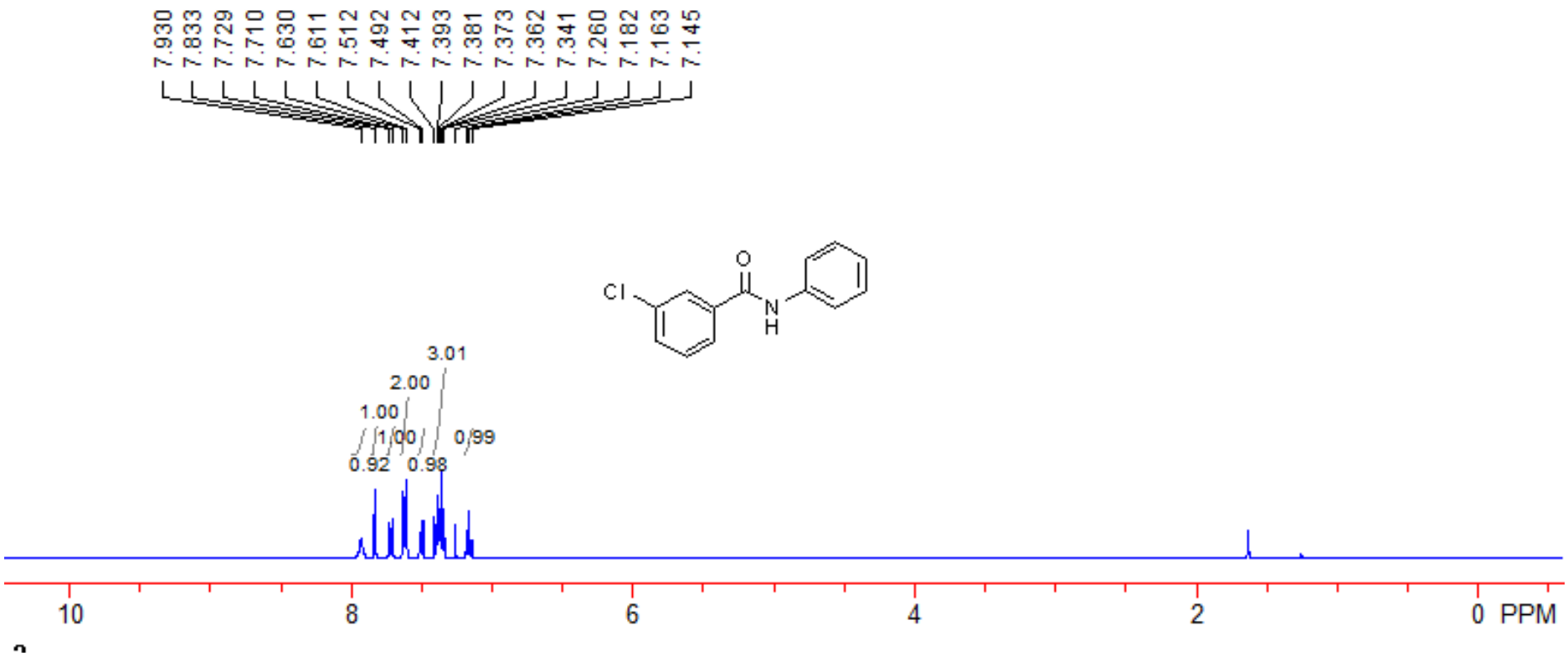
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3e**.



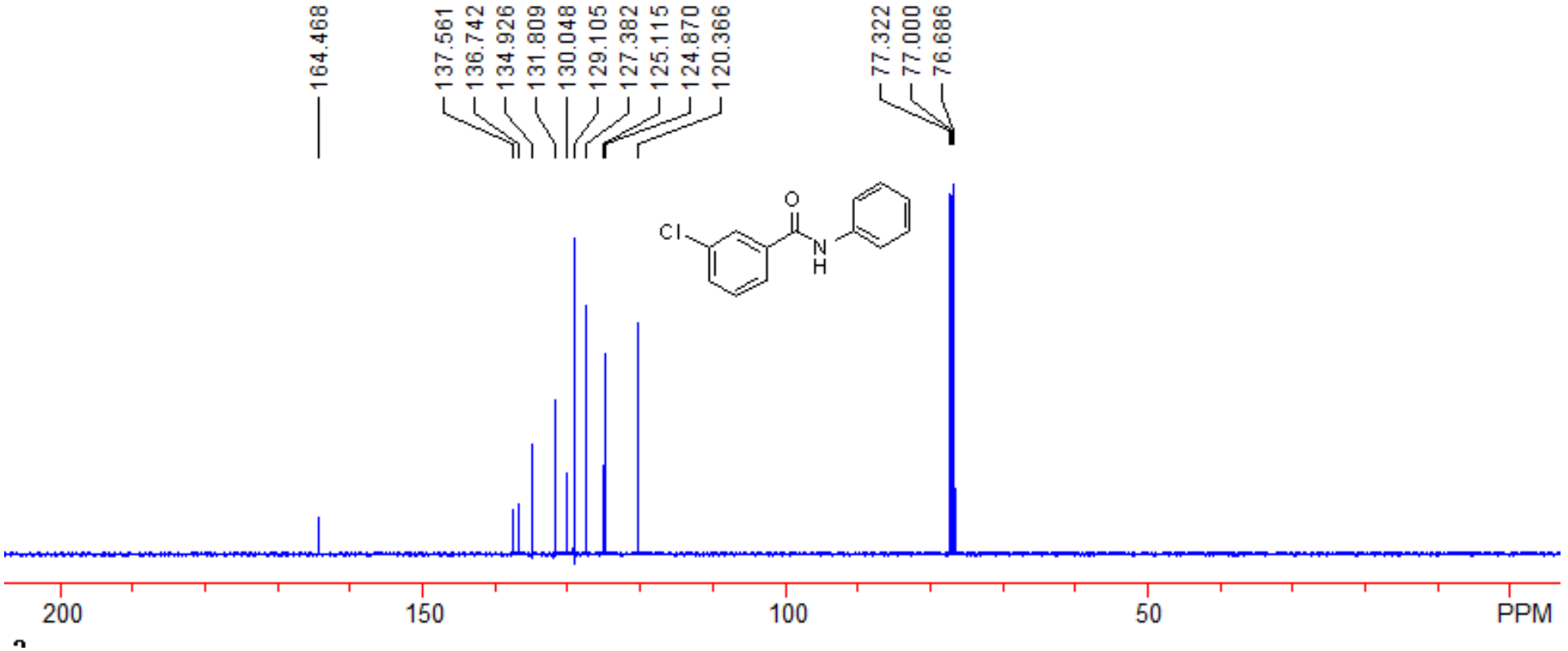
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **3e**.



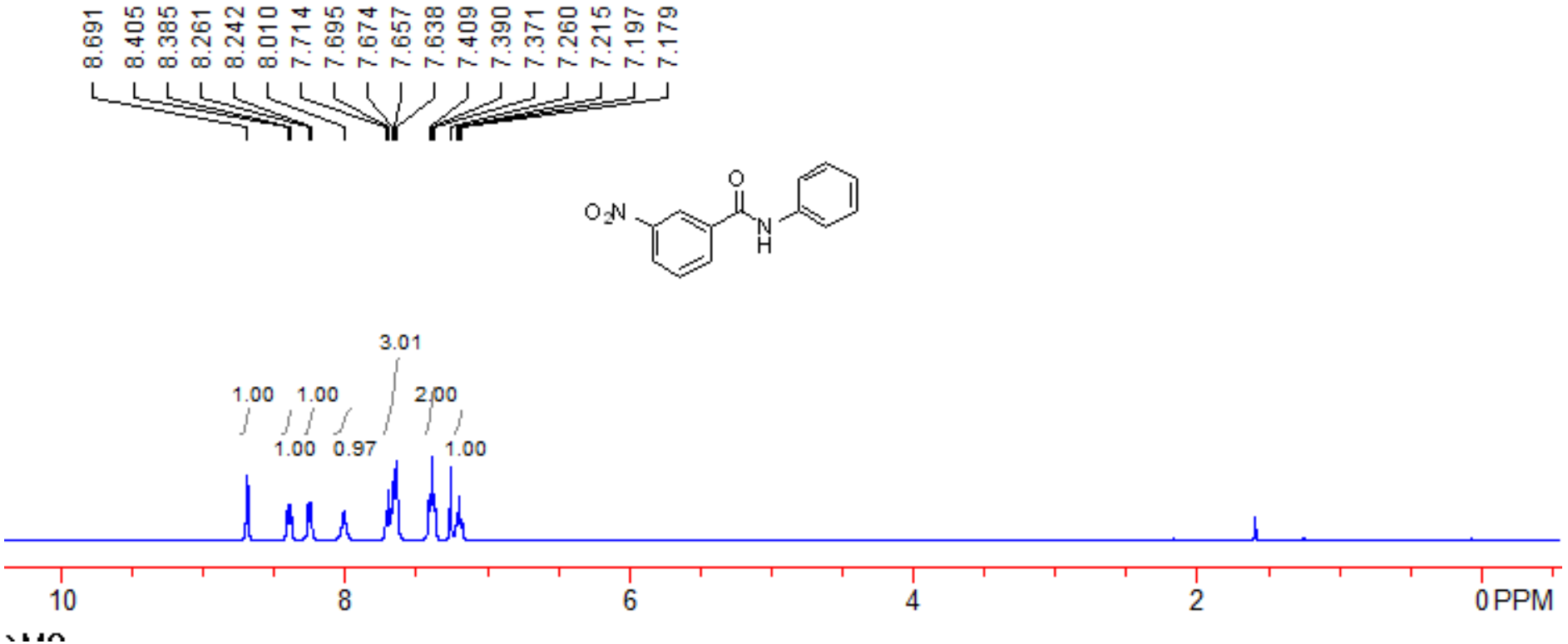
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) spectrum of **3e**.



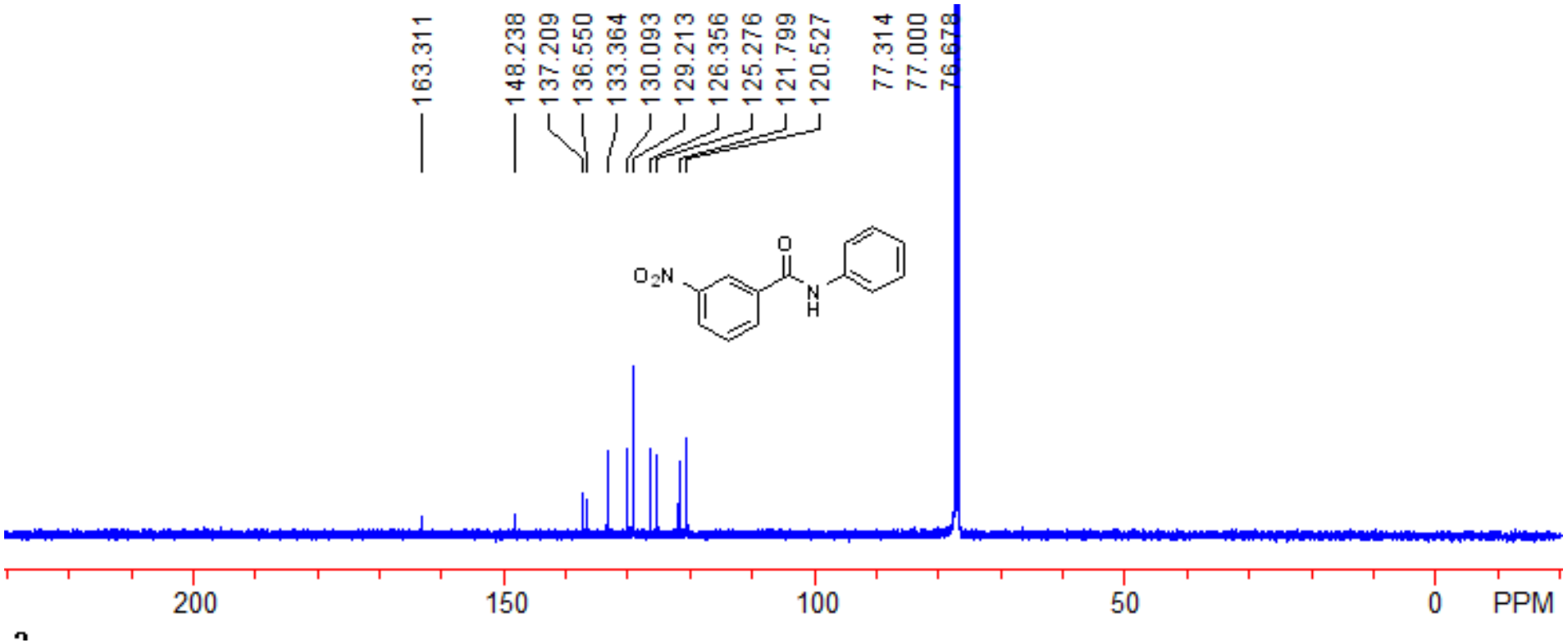
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3f**.



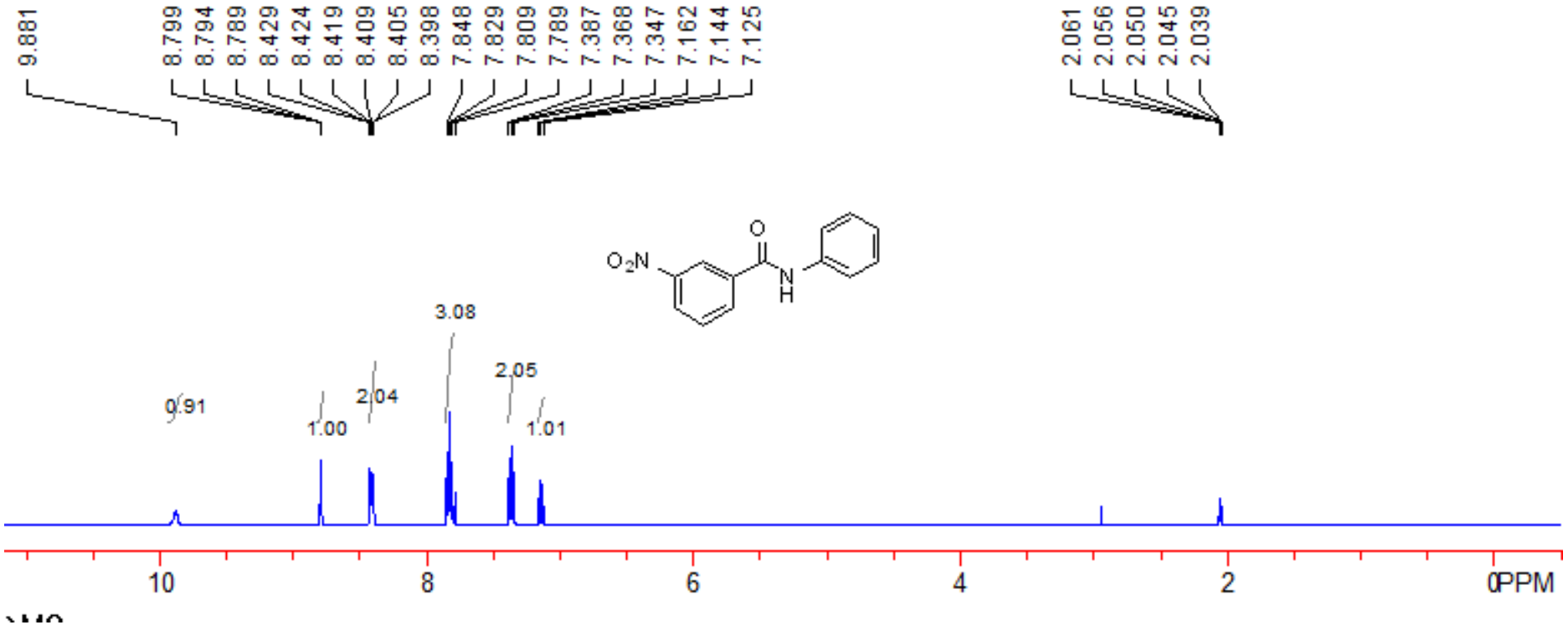
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **3f**.



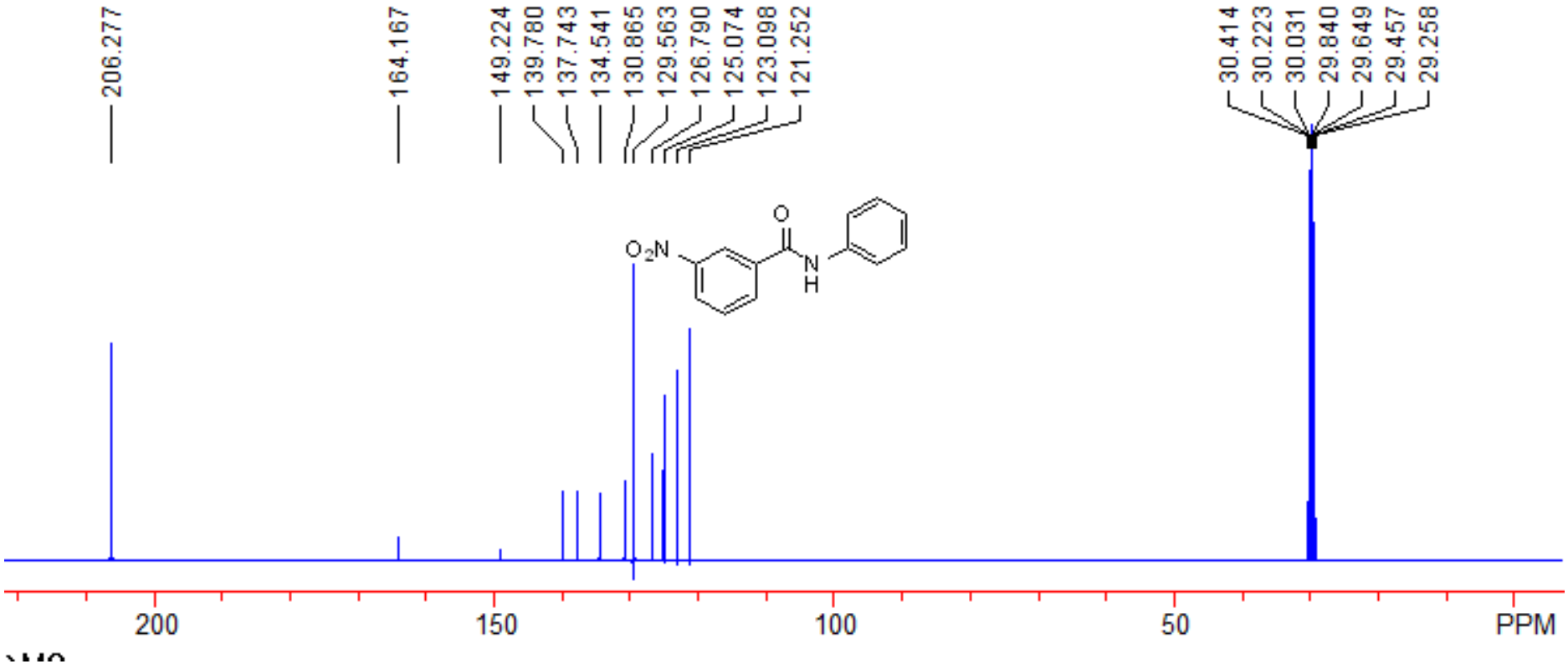
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3g**.



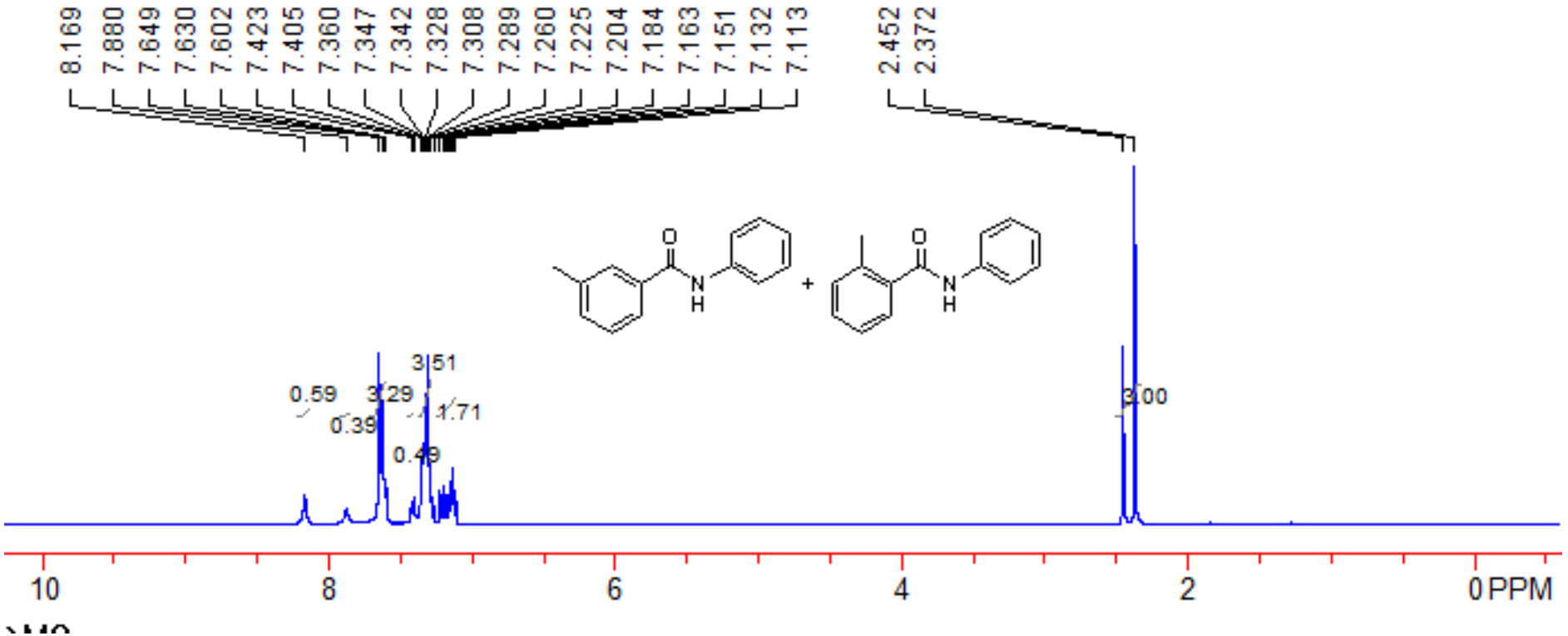
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **3g**.



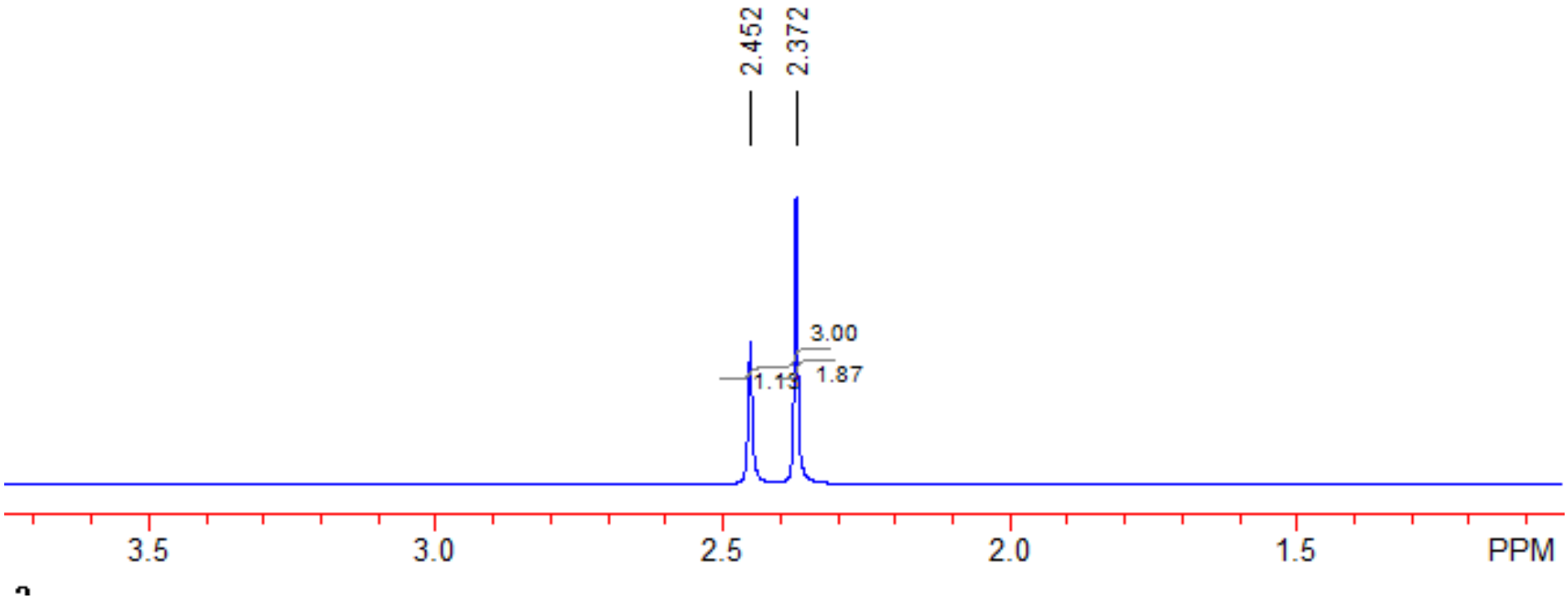
^{1}H NMR (400 MHz, acetone-d_6) spectrum of **3g**.



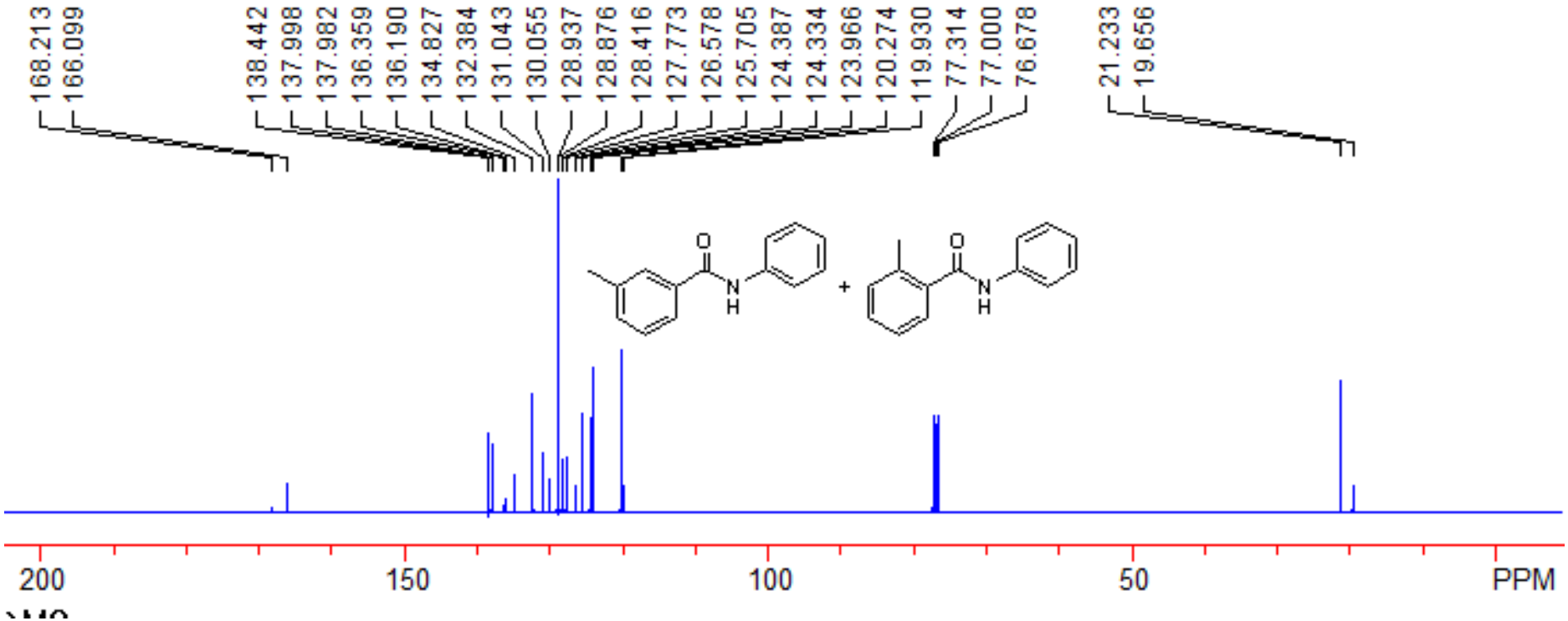
$^{13}C\{^{1}H\}$ NMR (100 MHz, acetone-d_6) spectrum of **3g**.



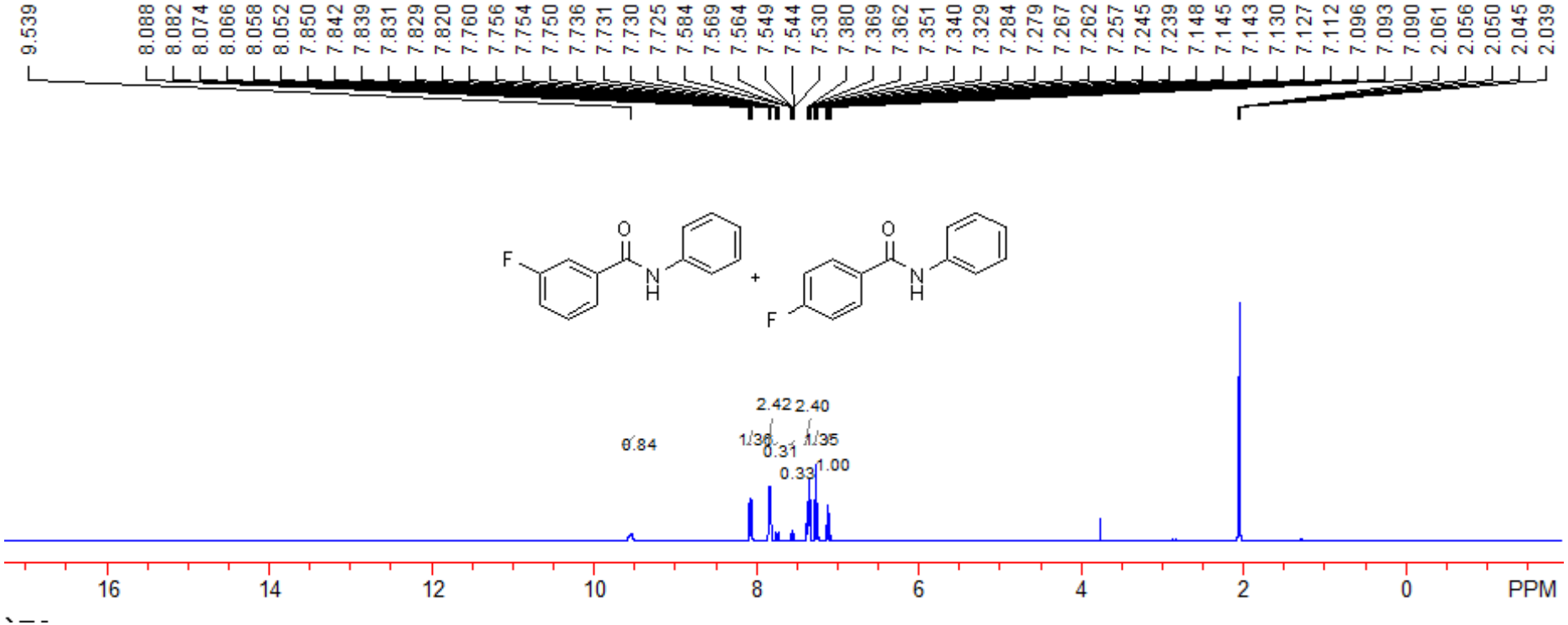
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3h** (mixture of ***o*-3h** and ***m*-3h**).



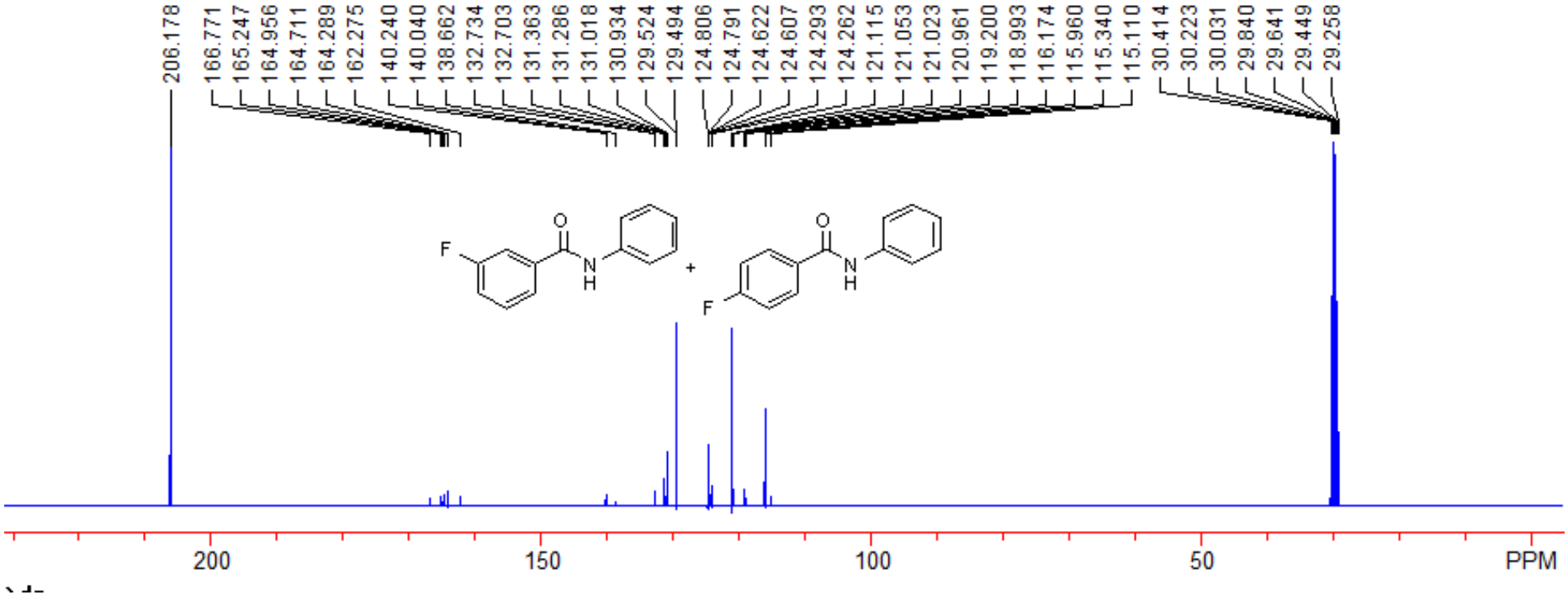
Zoom of the ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3h** to determine the ratio (38:62).



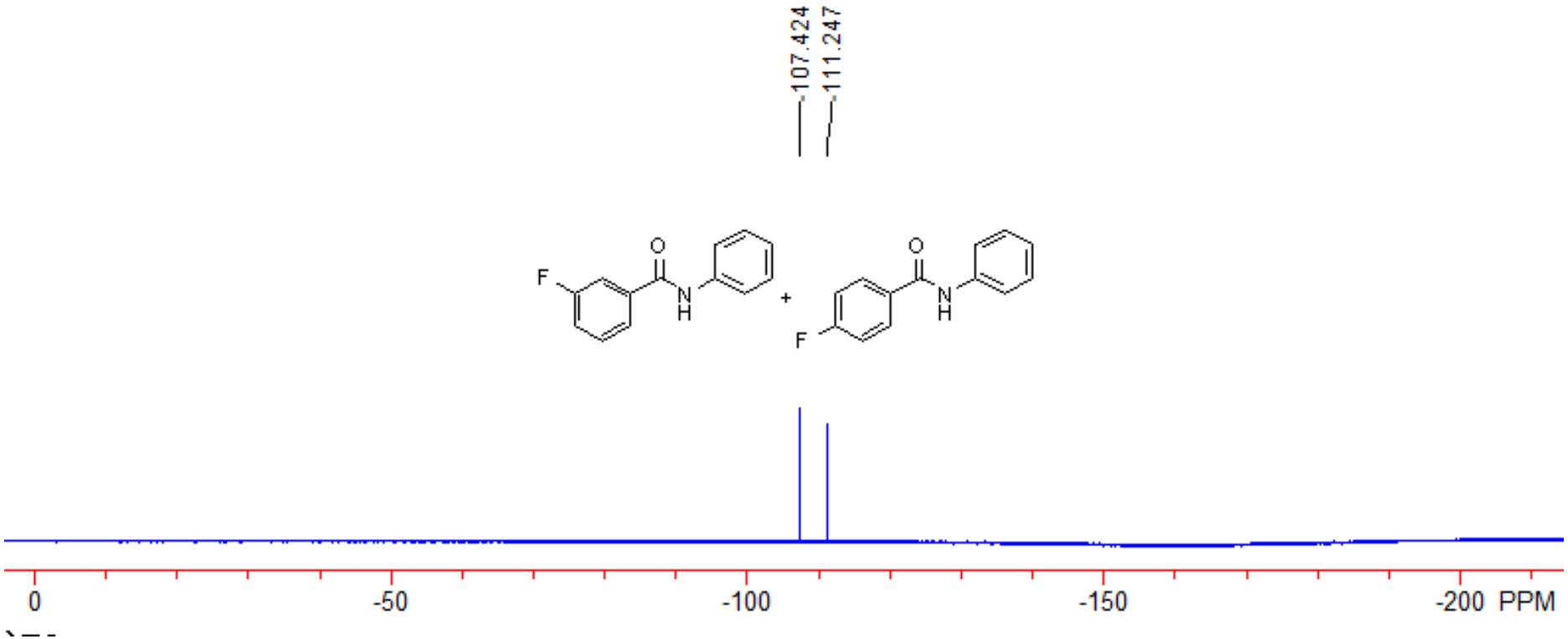
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **3h** (mixture of ***o*-3h** and ***m*-3h**).



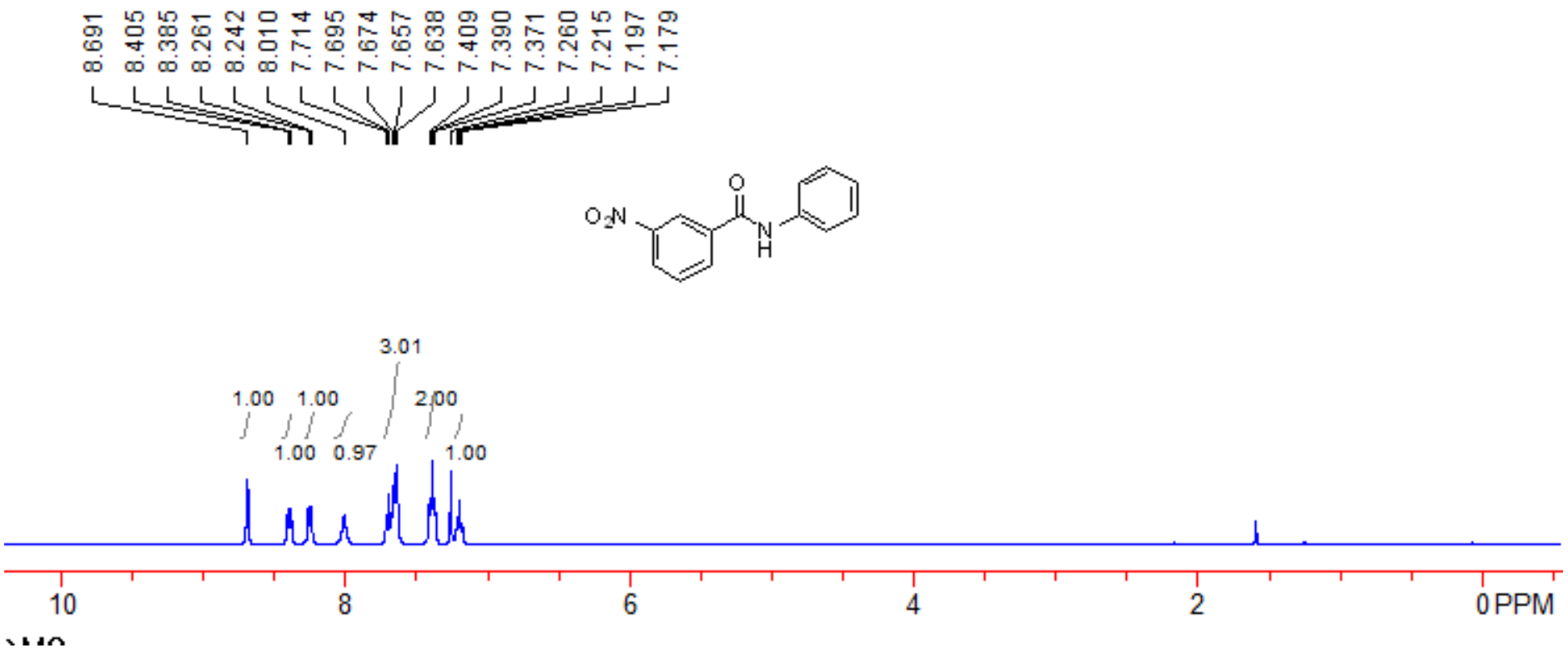
^{1}H NMR (400 MHz, acetone-d_6) spectrum of **3i** (mixture of ***m*-3i** and ***p*-3i**).



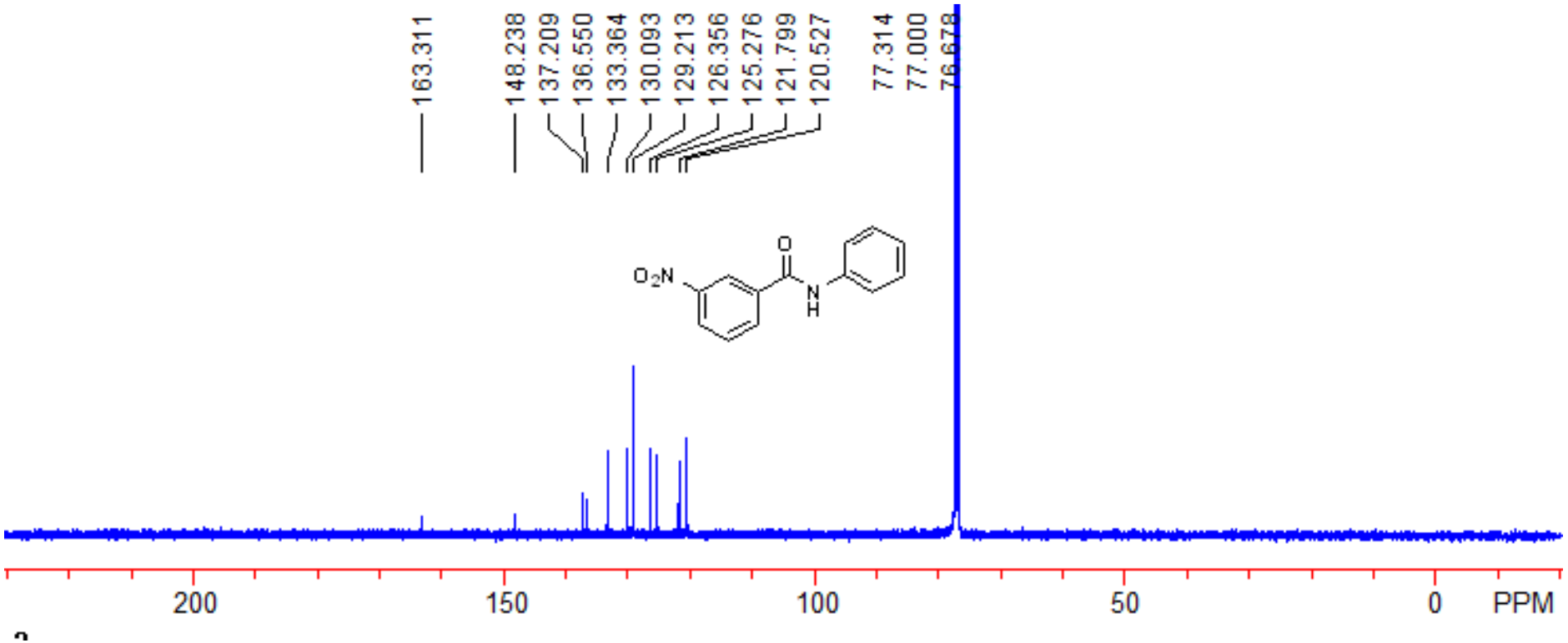
$^{13}C\{^{1}H\}$ NMR (100 MHz, acetone-d_6) spectrum of **3i** (mixture of ***m*-3i** and ***p*-3i**).



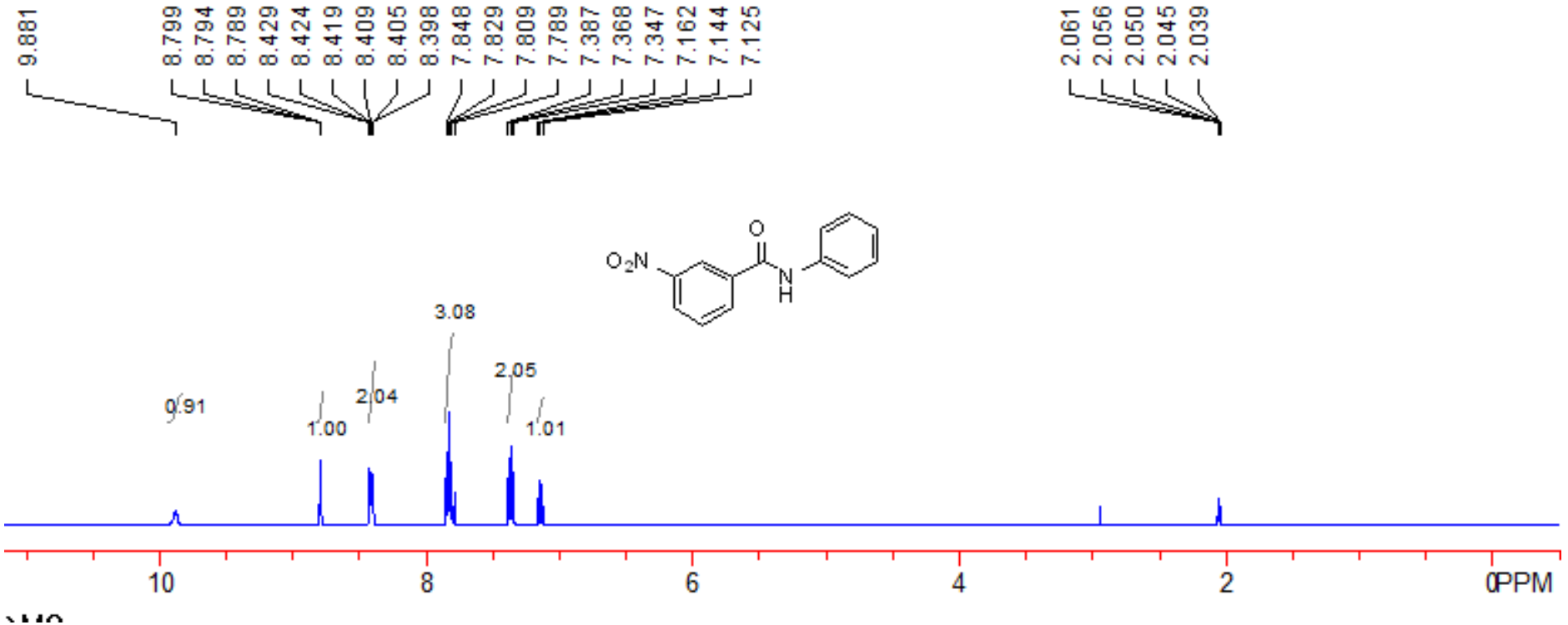
$^{19}F\{^{1}H\}$ NMR (376 MHz, acetone-d_6) spectrum of **3i** to determine the ratio (59:41).



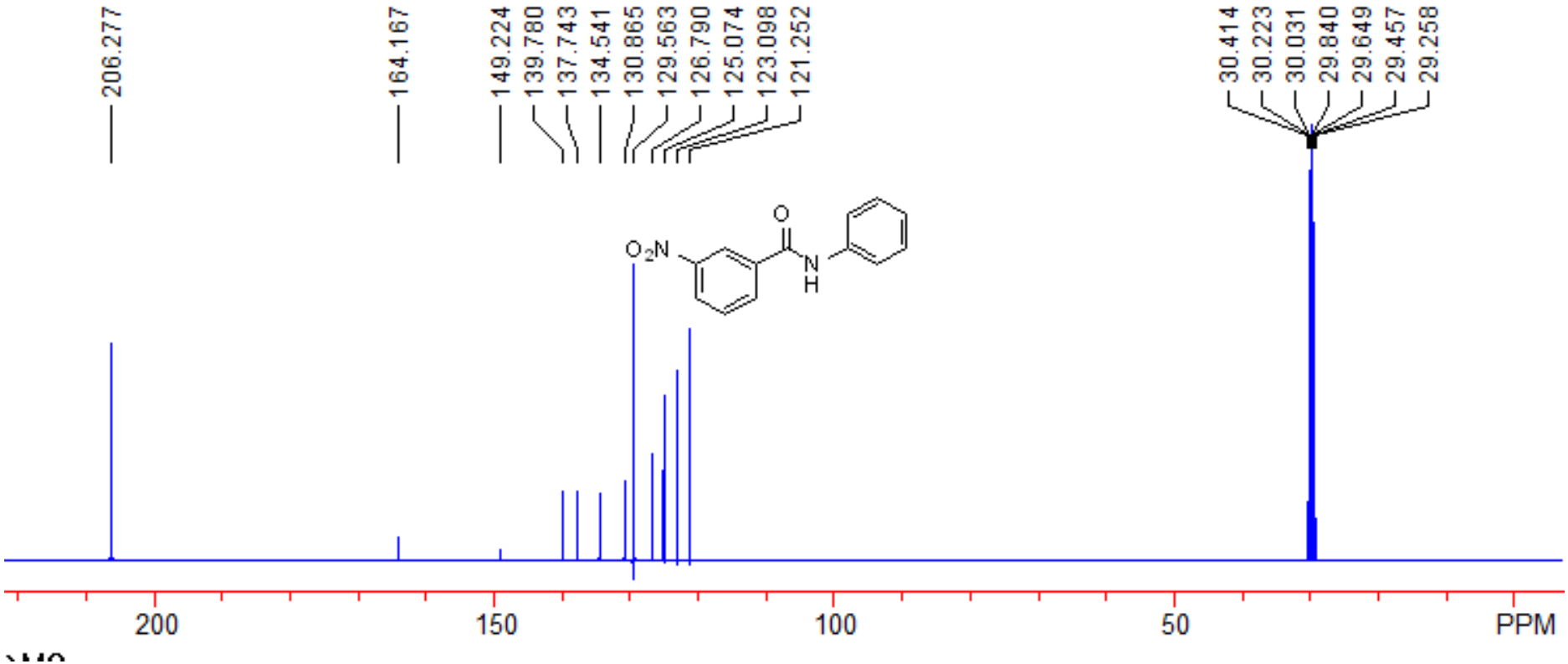
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of ***m*-3j**.



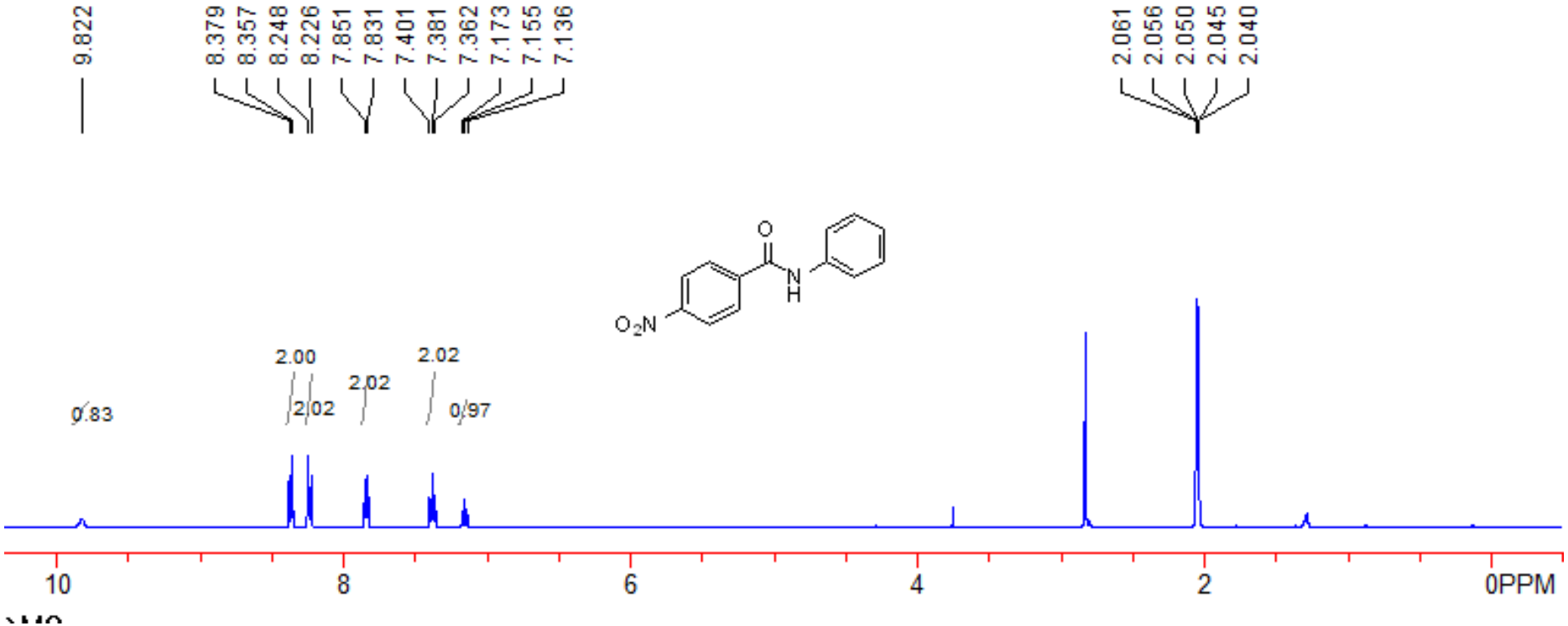
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of ***m*-3j**.



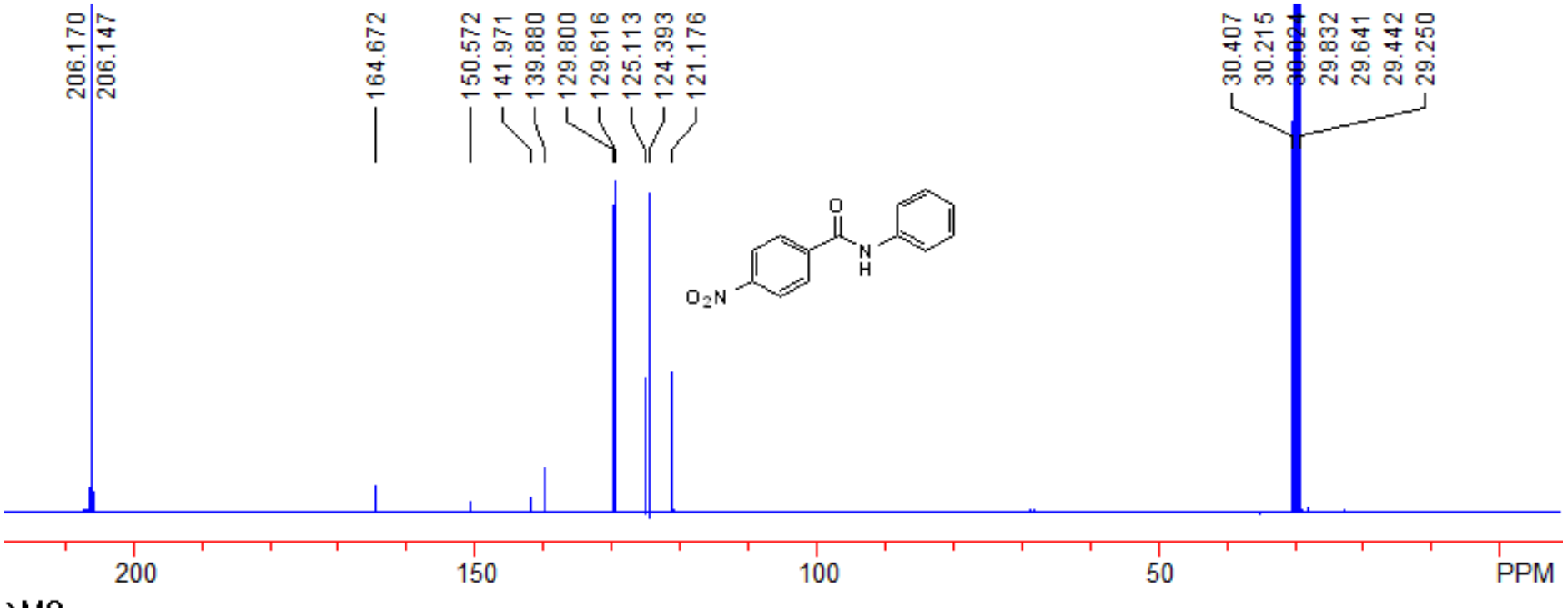
^{1}H NMR (400 MHz, acetone-d_6) spectrum of ***m*-3j**.



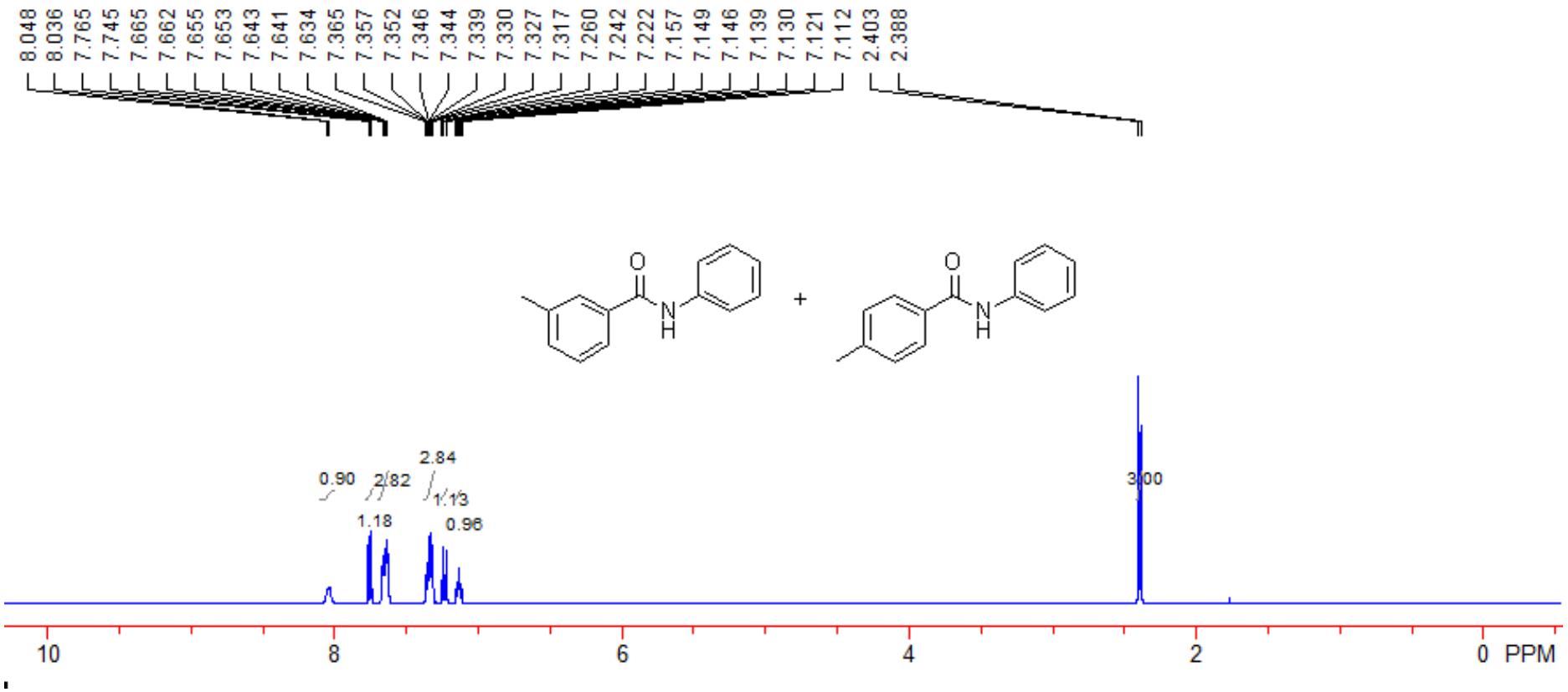
$^{13}C\{^{1}H\}$ NMR (100 MHz, acetone-d_6) spectrum of ***m*-3j**.



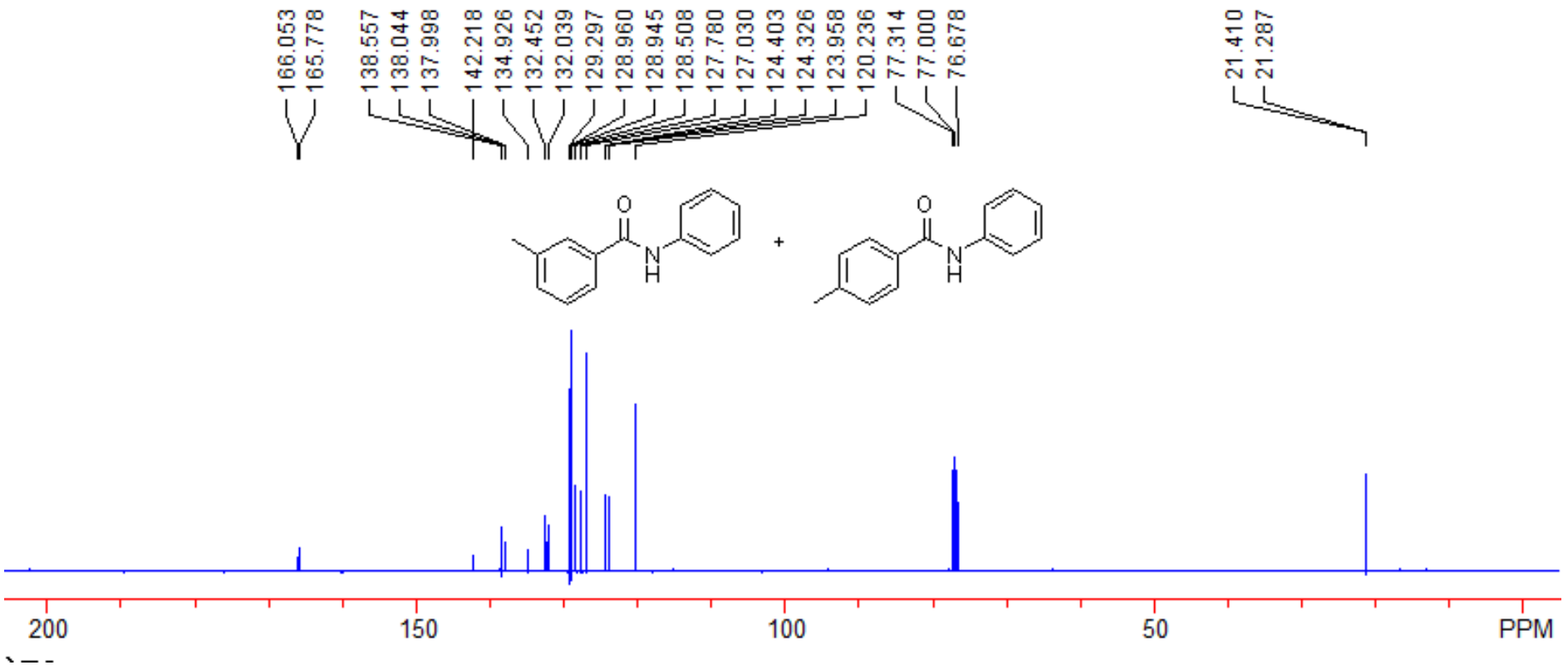
^{1}H NMR (400 MHz, acetone-d_6) spectrum of ***p*-3j**.



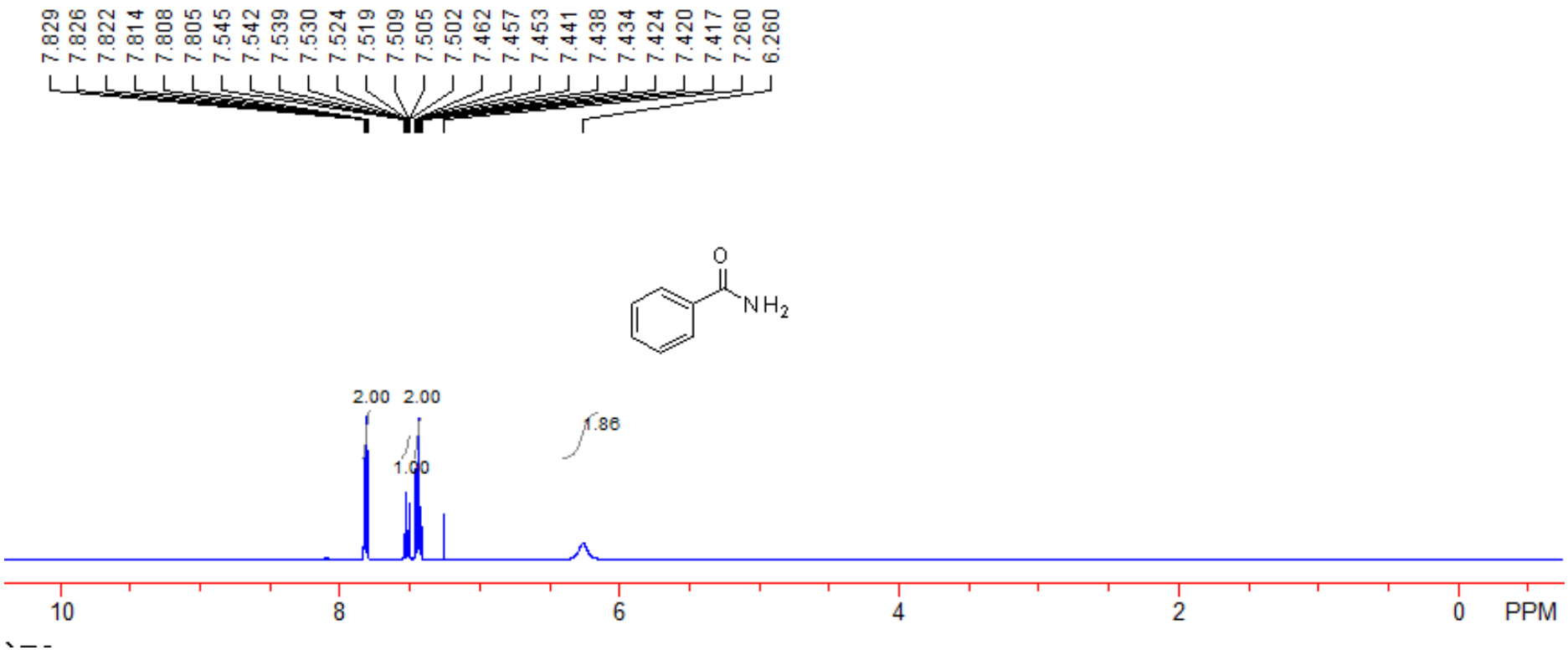
$^{13}C\{^{1}H\}$ NMR (100 MHz, acetone-d_6) spectrum of ***p*-3j**.



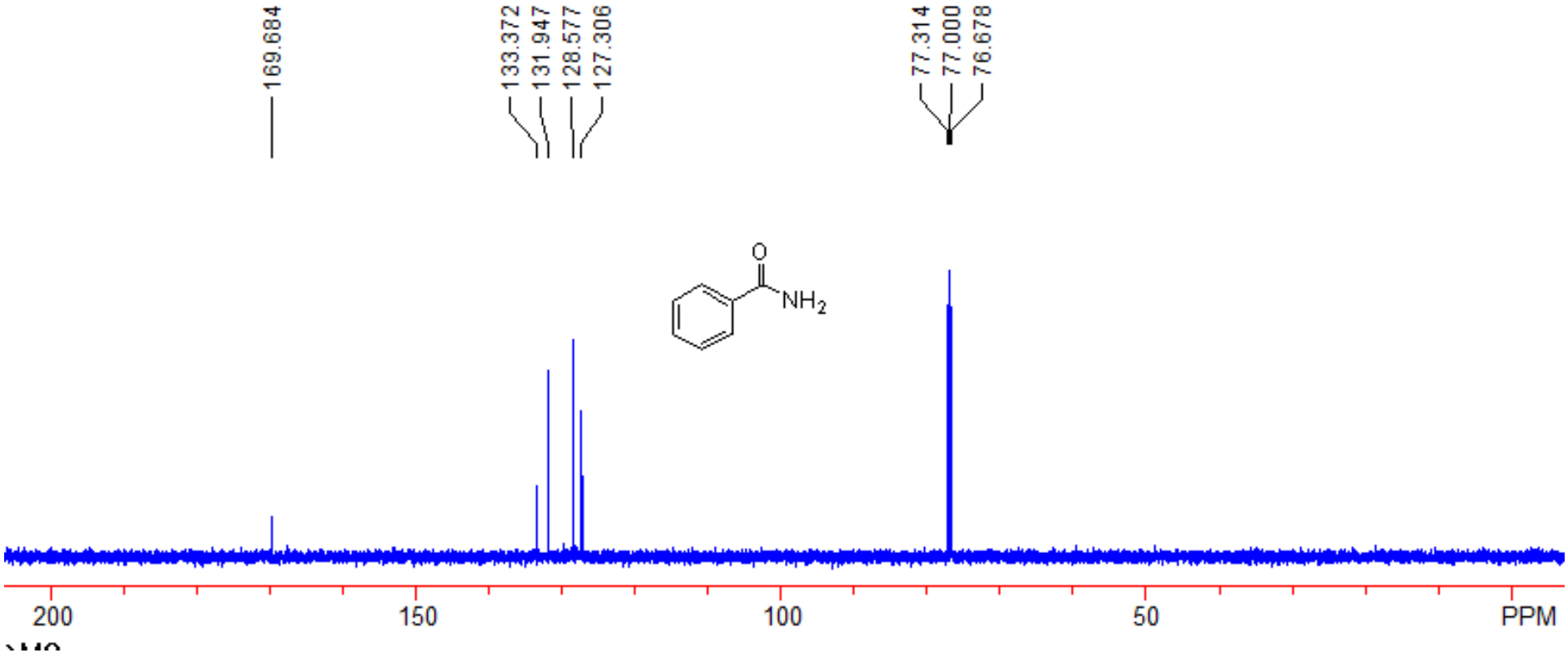
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **3k** (mixture of ***m*-3k** and ***p*-3k**).



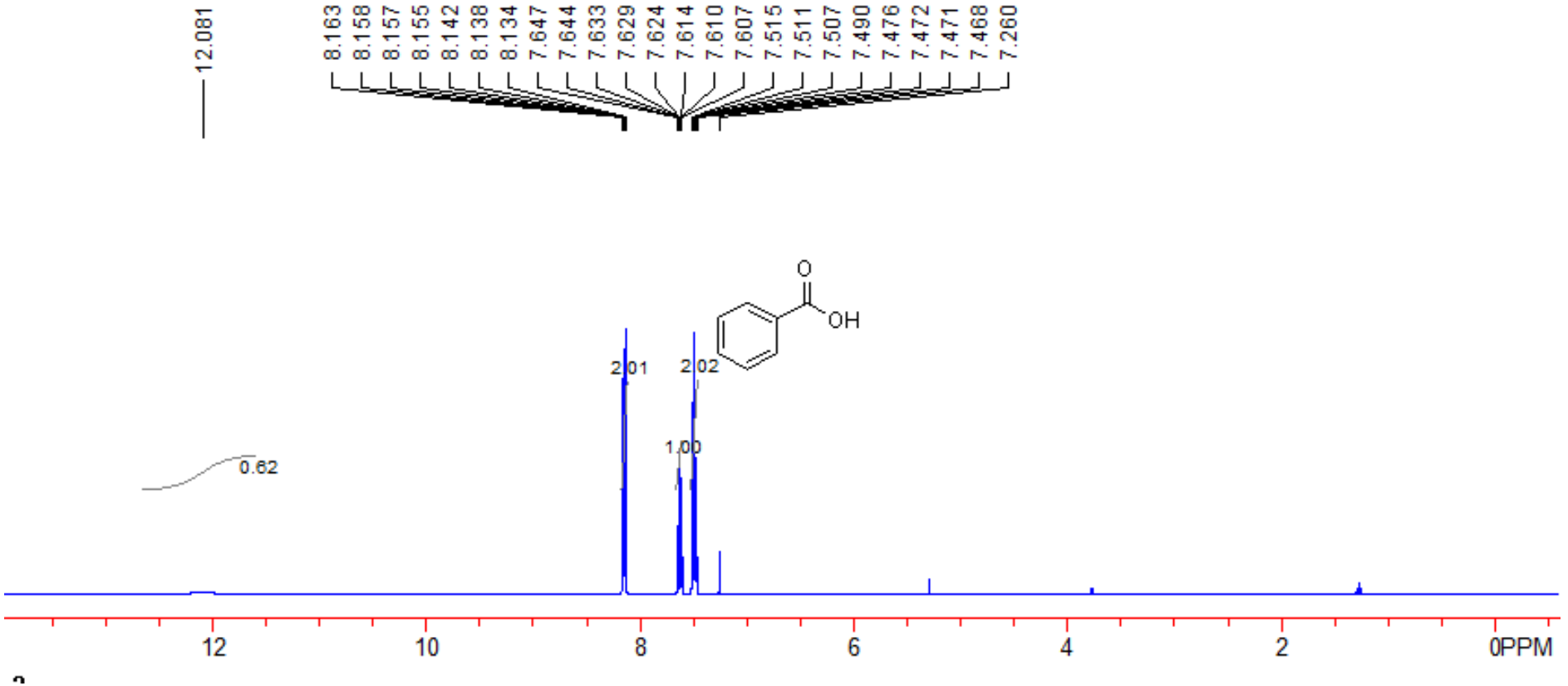
$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **3k** (mixture of ***m*-3k** and ***p*-3k**).



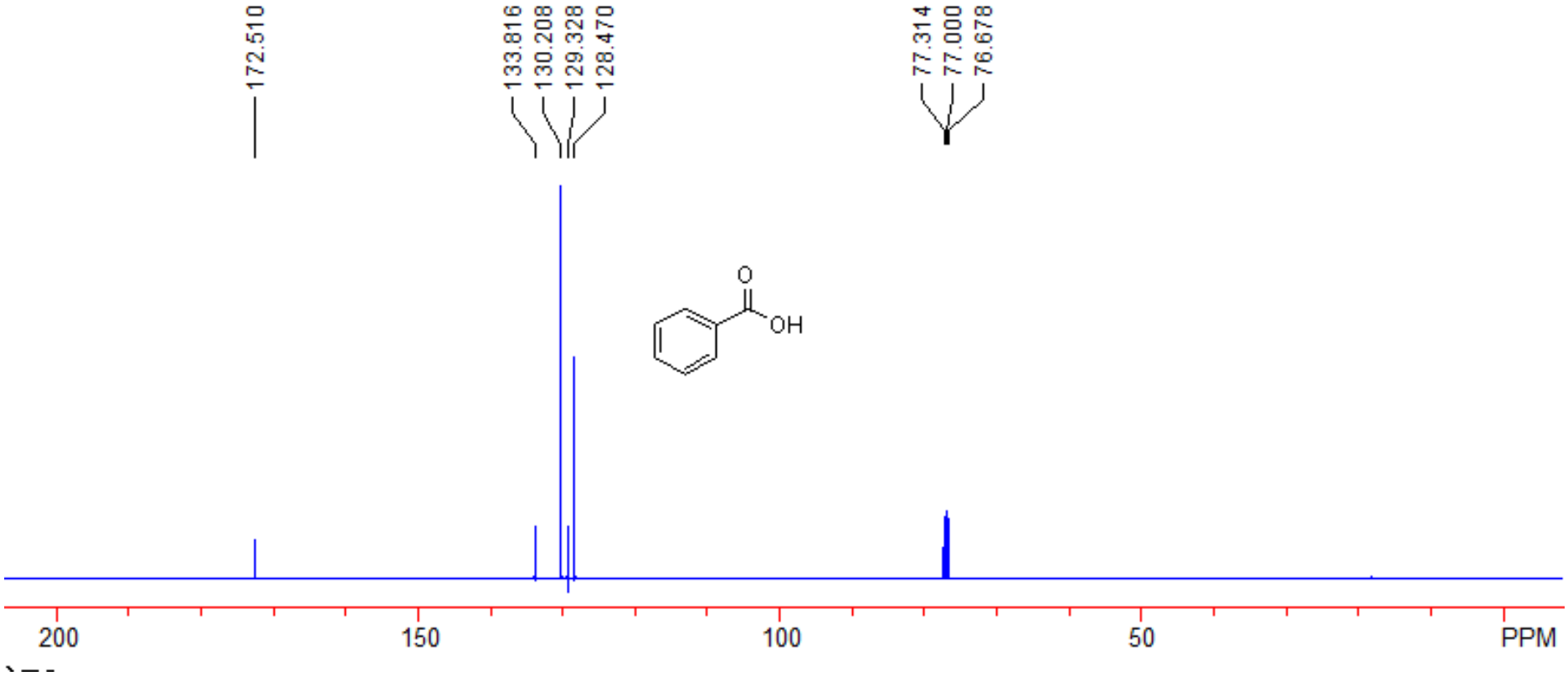
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **benzamide**.



$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **benzamide**.



^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **benzoic acid**.



$^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **benzoic acid**.