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Cross-coupling Reaction of Alcohols for Carbon–carbon Bond Formation Using Pincer-type-NHC/Palladium Catalysts

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Experimental Section

General: All reactions were carried out under argon atmosphere and in dried up glassware by means of standard Schlenk techniques unless otherwise noted. All reagents were purchased from Aldrich, Wako, TCI, Kanto and used without further purification except for benzyl alcohol which is simply distilled under reduced pressure. Column chromatography was performed using silica gel (silica gel 60, 230-400 mesh, Merck). TLC was performed using pre-coated silica gel plate (silica gel 60 F₂₅₄, Merck) and products were observed under UV light or with either phosphomolybdic acid reagent. ¹H NMR and ¹³C NMR spectra were recorded on a JEOL ECA-600 spectrometer, operating in CDCl₃ at 600 MHz. Chemical shifts and coupling constants are presented in ppm δ relative to tetramethylsilane and Hz respectively. High resolution mass spectra was obtained on JEOL JMS-700. Chiral high performance liquid chromatography analysis was conducted using Shimadzu LC-10AD coupled with photo diode array-detector SPD-M20A and chiral column of CHIRALCEL OD-H (Daicel chemical industries, LTD.).

Typical procedure for cross-coupling reaction of alcohols: The reaction of 1,2,3,4-tetrahydro -naphthol 1a with benzyl alcohol 2a (Table 1).

All reactions were carried out in argon-filled Schlenk tube. A degassed and subsequently argon-filled *p*-xylene (1 mL) suspension of 1,2,3,4-tetrahydronaphthol **1a** (2 mmol, 294 mg), benzyl alcohol **2a** (1 mmol, 108 mg), 2,6-bis(*N*-butyl-*N'*-methylenebenzimidazolium)-1-bromobenzene dibromide **5a** (0.04 mmol, 27.7 mg), Pd₂(dba)₃ (0.02 mmol, 18.3 mg), and cesium hydroxide (0.4 mmol, 60 mg) was prepared at room temperature. The mixture was stirred at 125 °C for 24 h, cooled down to room temperature and directly purified by column chromatography on silica gel (EtOAc/*n*-hexane = from 1/100 to 1/30) to give a mixture of **3aa**

and **4aa** (99% yield in the ratio of 13:1, which was determined by internal standard (1,1,2,2,-tetrachloro -ethane) method with ^1H NMR spectroscopy).

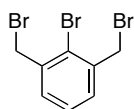
General procedure for cross-coupling reaction of various primary alcohols and secondary alcohols without H_2 (Table 2)

All reactions were carried out in Schlenk tube. A degassed and subsequently argon-filled *p*-xylene suspension of secondary alcohol (2 mmol), primary alcohol (1 mmol), 2,6-bis(*N*-butyl-*N'*-methylenebenzimidazolium)-1-bromobenzene dibromide (**5a**) (0.05 mmol, 34.6 mg), $\text{Pd}_2(\text{dba})_3$ (0.025 mmol, 22.9 mg), and cesium hydroxide (0.4 mmol, 60 mg) was prepared at room temperature. The mixture was stirred at 125 °C for 12–24 h, cooled down to room temperature, and directly purified by column chromatography on silica gel (EtOAc/*n*-hexane = from 1/100 to 1/30) to give alcohol **3**, along with ketone **4** in some cases. A ratio of **3** and **4** was determined by ^1H NMR spectroscopy.

Representative Procedure for the alcohol–alcohol coupling under H_2 (1 atm) pressure (Table 2)

To a 30 mL flask stoppered by a Young's stopcock was added $\text{Pd}_2(\text{dba})_3$ (22.5 mg, 0.025 mmol), **6** (35 mg, 0.05 mmol), **1b** (244 mg, 2 mmol), **2f** (74 mg, 1 mmol), NaOH (16 mg, 0.4 mmol) and anhydrous *p*-xylene (1.0 mL) at 25 °C. The flask was degassed and subsequently filled with H_2 , and was stoppered again by a Young's stopcock to make a closed system. The resulting suspension was immediately heated and stirred at 125 °C for 24 h. The mixture was cooled down to 25 °C, and was directly purified by column chromatography on silica gel (EtOAc/*n*-hexane = from 1/100 to 1/30) to give alcohol **3bf** and ketone **4bf** in a ratio of 9.2:1 (92% ^1H NMR yield, using 1,1,2,2-tetrachloroethane as an internal standard).

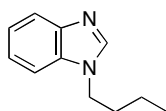
Preparation of 1-bromo-2,6-bis(bromomethyl)benzene¹



To a stirred solution of 1-bromo-2,6-dimethylbenzene (160 mmol, 29.7 g) in tetrachlorobenzene (200 mL) were added *N*-bromosuccinimide (330 mmol, 59.0 g) and a catalytic amount of benzoyl peroxide (5.8 mmol, 1.4 g). The mixture was heated gradually up to a reflux temperature. Then further catalytic amount of benzoyl peroxide (3.5 mmol, 0.85 g) was added

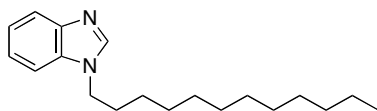
and kept stirring under reflux for 5 hours. The brown solution was cooled to room temperature and left to stand for overnight. The white solid was filtered off and discarded, and the brown filtrate was evaporated to dryness over a rotary evaporator. The residue was dissolved in a minimum volume of acetone and stored in a freezer for overnight. This gave a white crystalline solid, which was filtered off, washed with a minimum amount of cooled acetone and dried under reduced pressure (0.5–1.0 mmHg). The isolated yield of 1-bromo-2,6-bis(bromomethyl)benzene was 40% (21.9 g). IR (KBr): $\nu_{\max}/\text{cm}^{-1} = 3034, 2975, 1575, 1426, 1260, 1208, 1027, 863, 801, 724, 581$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.41 (d, $J = 7.5$ Hz, 2H), 7.29 (t, $J = 7.5$ Hz, 1 H), 4.64 (s, 4H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 138.8, 131.7, 128.4, 34.2.

Preparation of *N*-butylbenzimidazole²



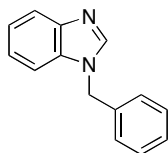
Benzimidazole (5 mmol, 0.34 g) was slowly added to a suspension of oil-free sodium hydride (10 mmol 0.24 g) in THF (10 mL) under Ar at room temperature. After evolution of hydrogen gas ceased, a solution of *n*-butyl bromide (5 mmol, 0.87 g) in THF (10 mL) was dropwise added, then the mixture was stirred at 60 °C for overnight, and a yellow solution was obtained. The solvent was removed with a rotary evaporator and H_2O was added to the residue. Then the solution was extracted with dichloromethane (50 mL \times 3), and the extracted solution was dried over sodium sulfate. After removal of dichloromethane, yellow liquid was obtained. This was simply purified by short column chromatography (EtOAc/*n*-hexane = 1/20) to give *N*-butyl benzimidazole as colorless liquid (0.78 g, 89% yield). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 2872, 1498, 1458, 1375, 1288, 1098, 948, 844, 741$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.88 (s, 1H), 7.81 (d, $J = 6.8$ Hz, 1H), 7.40 (dd, $J = 6.8$ Hz, 1.38 Hz, 1H), 7.30–7.26 (m, 2H), 4.16 (t, $J = 6.8$ Hz, 2H), 1.87–1.84 (m, 2H), 1.39–1.33 (m, 2H), 0.95 (t, $J = 7.2$ Hz, 3H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 144.2, 143.2, 134.1, 123.0, 122.3, 120.7, 109.9, 45.1, 32.1, 20.3, 13.9.

Preparation of *N*-dodecylbenzimidazole



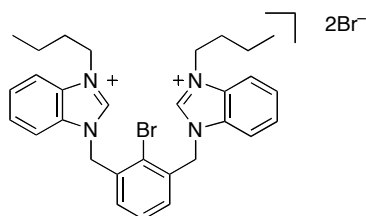
Benzimidazole (5 mmol, 0.34 g) was slowly added to a suspension of oil-free sodium hydride (10 mmol 0.24 g) in THF (10 mL) under Ar at room temperature. After evolution of hydrogen gas ceased, a solution of *n*-dodecyl bromide (5 mmol, 1.25 g) in THF (10 mL) was dropwise added, then the mixture was stirred at 60 °C for 36 h, and a yellow solution was obtained. The solvent was removed with a rotary evaporator and H₂O was added to the residue. Then the solution was extracted with dichloromethane (50 mL × 3), and the extracted solution was dried over sodium sulfate. After removal of dichloromethane, yellow liquid was obtained. This was simply purified by short column chromatography (EtOAc/*n*-hexane = 1/20) to give *N*-dodecyl benzimidazole as colorless liquid (1.29 g, 90% yield). IR (ATR): $\nu_{\max}/\text{cm}^{-1}$ = 2922, 2852, 1495, 1457, 1364, 1286, 1201, 1007, 889, 768. ¹H NMR (600 MHz, CDCl₃) δ (ppm): 7.88 (s, 1H), 7.81 (dd, *J* = 6.9 Hz, 1.4 Hz, 1H), 7.40 (dd, *J* = 6.9 Hz, 1.4 Hz, 1H), 7.31–7.24 (m, 2H), 4.16 (t, *J* = 7.2 Hz, 2H), 1.88 (m, 2H), 1.32–1.24 (m, 18H), 0.88 (t, *J* = 6.9 Hz, 3H). ¹³C NMR (600MHz, CDCl₃) δ (ppm): 144.3, 143.3, 134.2, 123.1, 122.3, 120.7, 110.0, 45.4, 32.2, 30.1, 29.9, 29.8, 29.6, 29.4, 27.1, 23.0, 14.5. HRMS (FAB) Calcd for C₁₉H₃₀N₂ : 286.2409. Found *m/z* = 286.2412.

Preparation of *N*-benzylbenzimidazole³



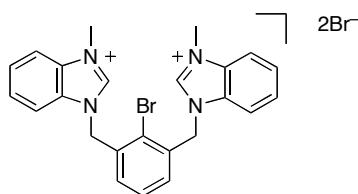
Benzimidazole (5 mmol, 340 mg) was slowly added to a suspension of oil-free sodium hydride (10 mmol 0.24 g) in THF (10 mL) under Ar at room temperature. After evolution of hydrogen gas ceased, a solution of benzyl bromide (5 mmol, 0.86 g) in THF (10 mL) was dropwise added, then the mixture was stirred at 60 °C for 12 h, and a yellow solution was obtained. The solvent was removed with a rotary evaporator and H₂O (50 mL) was added to the residue. Then the solution was extracted with dichloromethane (50 mL × 3), and the extracted solution was dried over sodium sulfate, after removal of dichloromethane, yellow liquid was obtained. This was simply purified by short column chromatography (EtOAc/*n*-hexane = 1/20) to give *N*-benzylbenzimidazole as colorless liquid (0.96 g, 92%). IR (KBr): $\nu_{\max}/\text{cm}^{-1}$ = 3080, 1613, 1494, 1443, 1366, 1286, 1266, 1202, 1187, 973, 887, 766, 731, 695. ¹H NMR (600 MHz, CDCl₃) δ (ppm): 7.93 (s, 1H), 7.83 (d, *J* = 8.9 Hz, 1H), 7.32–7.23 (m, 6H), 7.16 (d, *J* = 6.9 Hz, 2H), 5.33 (s, 2H). ¹³C NMR (600MHz, CDCl₃) δ (ppm): 144.3, 143.5, 135.8, 134.2, 129.3, 128.5, 127.4, 123.4, 122.5, 120.7, 110.3, 49.1.

Preparation of 2,6-bis(*N*-butyl-*N'*-methylenebenzimidazolium)-1-bromobenzene Dibromide (**5a**)⁴



A solution of *N*-butylbenzimidazole (4 mmol, 0.70 g) and 1-bromo-2,6-bis(bromomethyl)benzene (3.9 mmol, 1.33 g) in 1,4-dioxane (50 mL) was heated under reflux for overnight. White precipitate was filtered off, washed with ether (30 mL × 2) and dried under reduced pressure (0.5–1.0 mmHg). Without further purification, 2,6-bis(*N*-butyl-*N'*-methylenebenzimidazolium)-1-bromobenzene dibromide **5a** was obtained as a white solid (2.54 g, 92% yield). IR (KBr): $\nu_{\max}/\text{cm}^{-1} = 3449, 2961, 1561, 1460, 1430, 1346, 1203, 1016, 759$. ¹H NMR (600 MHz, DMSO-*d*₆) δ (ppm): 9.95 (s, 2H), 8.07 (d, *J* = 8.3 Hz, 2H), 7.83 (d, *J* = 8.3 Hz, 2H), 7.62–7.55 (m, 4H), 7.39–7.36 (m, 3H), 5.82 (s, 4H), 4.48 (t, *J* = 7.2 Hz, 2H), 2.39 (s, 2H), 1.80–1.76 (m, 4H), 1.25–1.20 (m, 4H), 0.80 (t, *J* = 7.2 Hz, 6H). ¹³C NMR (600MHz, DMSO-*d*₆) δ (ppm): 143.9, 135.1, 132.0, 129.7, 127.7, 125.6, 115.0, 114.8, 51.7, 47.5, 31.5, 20.0, 14.3.

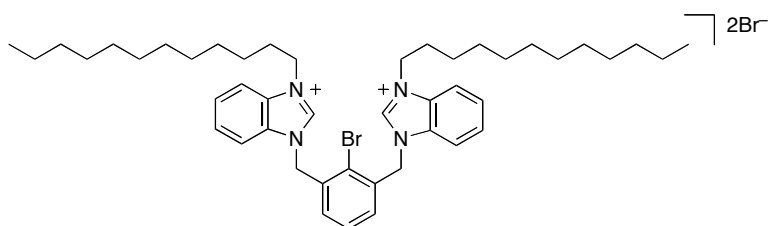
Preparation of 2,6-bis(*N*-methyl-*N'*-methylenebenzimidazolium)-1-bromobenzene Dibromide (**5b**)



A solution of *N*-methylbenzimidazole (5 mmol, 0.66 g) and 1-bromo-2,6-bis(bromomethyl)benzene (2.5 mmol, 0.85 g) in 1,4-dioxane (50 mL) was heated under reflux for overnight. White precipitate was filtered off, washed with ether (30 mL × 2) and dried under reduced pressure (0.5–1.0 mmHg). Without further purification, 2,6-bis(*N*-methyl-*N'*-methylenebenzimidazolium)-1-bromobenzene dibromide **5b** was obtained as a white solid (2.76 g, 91% yield). IR (KBr): $\nu_{\max}/\text{cm}^{-1} = 3406, 3018, 2963, 1567, 1432, 1352, 1199, 1029, 759$. ¹H NMR (600 MHz, DMSO-*d*₆) δ (ppm): 9.93 (s, 2H), 8.13 (d, *J* = 7.6 Hz,

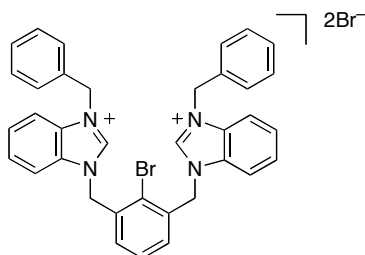
2H), 7.98 (d, $J = 8.2$ Hz, 2H), 7.77 (t, $J = 7.6$ Hz, 2H), 7.72 (t, $J = 7.6$ Hz, 2H), 7.53 (t, $J = 7.2$ Hz, 1H), 7.45 (d, $J = 7.6$ Hz, 2H), 5.97 (s, 4H), 4.19 (s, 6H). ^{13}C NMR (600MHz, DMSO- d_6) δ (ppm): 144.3, 135.1, 132.9, 131.8, 130.0, 129.6, 127.8, 127.7, 125.4, 114.8, 114.6, 51.5, 34.4. HRMS (FAB) Calcd for $\text{C}_{24}\text{H}_{23}\text{Br}_2\text{N}_4(\text{M}-\text{Br}^-)$: 527.0289. Found $m/z = 527.0314$.

Preparation of 2,6-bis(*N*-dodecyl-*N'*-methylenebenzimidazolium)-1-bromobenzene Dibromide (5c)



A solution of *N*-dodecylbenzimidazole (5 mmol, 1.43 g) and 1-bromo-2,6-bis(bromomethyl)benzene (2.5 mmol, 0.85 g) in 1,4-dioxane (50 mL) was heated under reflux for overnight. White precipitate was filtered off, washed with ether (30 mL \times 2) and dried under reduced pressure (0.5–1.0 mmHg). Without further purification, 2,6-bis(*N*-dodecyl-*N'*-methylenebenzimidazolium)-1-bromobenzene dibromide **5c** was obtained as a white solid (3.94 g, 86% yield). IR (KBr): $\nu_{\text{max}}/\text{cm}^{-1} = 3430, 2920, 2853, 1562, 1430, 1354, 1204, 1023, 761, 743$. ^1H NMR (600 MHz, DMSO- d_6) δ (ppm): 10.1 (s, 2H), 8.21 (d, $J = 8.3$ Hz, 2H), 7.98 (d, $J = 8.3$ Hz, 2H), 7.76–7.69 (m, 4H), 7.53 (s, 3H), 6.97 (s, 4H), 4.62 (t, $J = 7.2$ Hz, 4H), 1.96–1.93 (m, 4H), 1.33–1.24 (m, 36H), 0.87 (t, $J = 6.9$, 6H). ^{13}C NMR (600MHz, DMSO- d_6) δ (ppm): 143.9, 135.0, 132.1, 124.1, 127.8, 127.7, 125.5, 115.0, 114.8, 51.7, 47.8, 31.2, 29.6, 26.4, 23.0, 14.9. HRMS (FAB) Calcd for $\text{C}_{46}\text{H}_{67}\text{Br}_2\text{N}_4(\text{M}-\text{Br}^-)$: 833.3732. Found $m/z = 833.3730$.

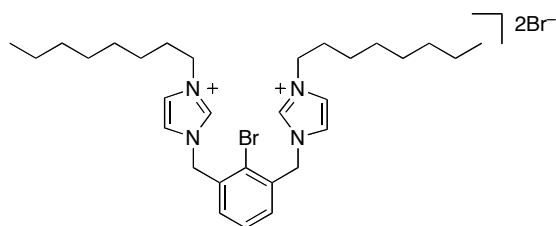
Preparation of 2,6-bis(*N*-benzyl-*N'*-methylenebenzimidazolium)-1-bromobenzene Dibromide (5d)



A solution of *N*-benzylbenzimidazole (5 mmol, 1.04 g) and

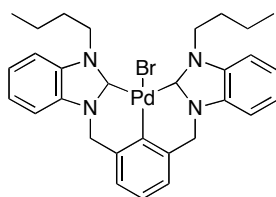
1-bromo-2,6-bis(bromomethyl)benzene (2.5 mmol, 0.85 g) in 1,4-dioxane (50 mL) was heated under reflux for overnight. White precipitate was filtered off, washed with ether (30 mL × 2) and dried under reduced pressure (0.5–1.0 mmHg). Without further purification, 2,6-bis(*N*-benzyl-*N*'-methylenebenzimidazolium)-1-bromobenzene dibromide **5d** was obtained as a white solid (3.27 g, 86% yield). IR (KBr): $\nu_{\max}/\text{cm}^{-1} = 3438, 3014, 1618, 1560, 1429, 1191, 1024, 758, 704$. ^1H NMR (600 MHz, DMSO- d_6) δ (ppm): 10.1 (s, 2H), 8.04 (d, $J = 7.6$ Hz, 2H), 8.02 (d, $J = 8.2$ Hz, 2H), 7.72–7.67 (m, 4H), 7.57–7.52 (m, 7H), 7.46–7.41 (m, 6H), 5.98 (s, 4H), 5.88 (s, 4H). ^{13}C NMR (600MHz, DMSO- d_6) δ (ppm): 144.2, 135.0, 134.8, 132.3, 132.2, 131.8, 129.9, 129.8, 129.6, 129.2, 127.9, 128.9, 125.8, 115.1, 115.0, 67.3, 51.9, 50.8. HRMS (FAB) Calcd for $\text{C}_{36}\text{H}_{31}\text{Br}_2\text{N}_4$ (M-Br $^-$): 677.0915. Found $m/z = 677.0898$.

Preparation of 2,6-bis(*N*-octyl-*N*'-methyleneimidazolium)-1-bromobenzene Dibromide (**6**)



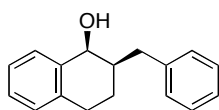
A solution of *N*-octylimidazole (5 mmol, 0.9 g) and 1-bromo-2,6-bis(bromomethyl)benzene (2.5 mmol, 0.85 g) in 1,4-dioxane (50 mL) was heated under reflux for overnight. White precipitate was filtered off, washed with ether (30 mL × 2) and dried under reduced pressure (0.5–1.0 mmHg). Without further purification, 2,6-bis(*N*-octyl-*N*'-methyleneimidazolium)-1-bromobenzene dibromide **6** was obtained as a white solid (3.27 g, 86% yield). IR (KBr): $\nu_{\max}/\text{cm}^{-1} = 3038, 2922, 2852, 1565, 1428, 1169, 1029, 743, 634$. ^1H NMR (600 MHz, DMSO- d_6) δ (ppm): 9.60 (s, 2H), 8.02 (s, 2H), 7.90 (s, 2H), 7.57–7.53 (m, 3H), 5.67 (s, 4H), 4.29(t, $J = 7.2$ Hz, 4H), 1.85–1.80 (m, 4H), 1.30–1.25 (m, 10H), 0.87 (t, $J = 6.9$ Hz, 6H). ^{13}C NMR (600MHz, DMSO- d_6) δ (ppm): 137.3, 135.5, 131.9, 129.4, 125.4, 123.4, 53.0, 49.5, 31.7, 30.0, 29.0, 28.8, 26.0, 22.6, 14.5. HRMS (FAB) Calcd for $\text{C}_{30}\text{H}_{47}\text{Br}_2\text{N}_4$ (M-Br $^-$): 621.2167. Found $m/z = 621.2198$.

Preparation of [2,6-bis(*N*-butyl-*N*-methylenebenzimidazolin-2-ylidene)phenylene]-bromopalladium (II) (**7**)⁴



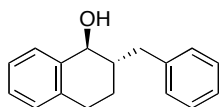
2,6-bis (*N*-butyl-*N'*-methylenebenzimidazolium)-1-bromobenzene dibromide **5a** (2 mmol, 1.38 g) was suspended in THF 200 mL) under Ar. To this was added dropwise at $-78\text{ }^{\circ}\text{C}$ a 1.59 M *n*-hexane solution of *n*-butyllithium (4.2 mmol, 2.67 mL). The reaction mixture was stirred for 30 min., and $\text{Pd}_2(\text{dba})_3$ (1.0 mmol, 0.92 g) was added. The reaction mixture was allowed to slowly warm to room temperature. Subsequently the obtained red solution was heated under reflux for 12 h. Solvent was removed with a rotary evaporator, and the residue was purified by recrystallization. This gave white crystals of [2,6-bis(*N*-butyl-*N*-methylenebenzimidazolin-2-ylidene)phenylene]bromopalladium (II) **7** (516 mg, 41% yield). (Found: C, 56.64; H, 5.25; N, 8.40. $\text{C}_{30}\text{H}_{35}\text{BrN}_4\text{Pd}$ requires C, 56.48; H, 5.53; N, 8.78%); IR (KBr): $\nu_{\text{max}}/\text{cm}^{-1} = 2951, 2868, 1478, 1413, 1341, 1203, 1030, 745$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.51 (d, $J = 7.6$ Hz, 2H), 7.40 (d, $J = 8.2$ Hz, 2H), 7.32–7.25 (m, 4H), 7.05 (d, $J = 6.8$ Hz, 2H), 6.85 (t, $J = 7.2$ Hz, 1H), 5.50 (d, $J = 13.8$ Hz, 2H), 5.46–5.41 (m, 2H), 5.14 (d, $J = 13.8$ Hz, 2H), 4.49–4.45 (m, 2H), 1.96–1.87 (m, 4H), 1.48–1.42 (m, 2H), 1.37–1.31 (m, 2H), 0.94 (t, $J = 7.2$ Hz, 6H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 189.3, 152.2, 141.2, 134.7, 134.3, 125.2, 123.2, 123.1, 122.9, 111.4, 110.3, 54.6, 48.1, 32.5, 20.3, 14.3.

2-benzyl-1,2,3,4-tetrahydronaphthalen-1-ol (**3aa**, *cis* isomer) ^{5,6}



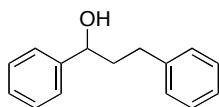
Yield (219 mg, 92 % (*cis* and *trans* isomers)). IR (ATR): $\nu_{\text{max}}/\text{cm}^{-1} = 3406, 3022, 2917, 1602, 1492, 1454, 1373, 1239, 1100, 1043, 942, 772, 736, 699$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.32–7.10 (m, 9H), 4.49 (bs, 1H), 2.94 (dd, $J = 7.9, 7.6$ Hz, 1H), 2.88–2.84 (m, 1H), 2.76–2.69 (m, 2H), 2.04–1.97 (m, 1H), 1.85–1.78 (m, 1H), 1.71–1.67 (m, 1H), 1.54 (s, 1H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 141.0, 138.8, 137.3, 130.4, 129.6 (2C), 129.4, 128.7 (2C), 128.3, 126.4, 126.2, 69.7, 42.1, 38.5, 29.5, 22.9.

2-benzyl-1,2,3,4-tetrahydronaphthalen-1-ol (**3aa**, *trans* isomer) ^{5,6}



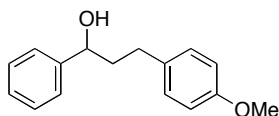
IR (KBr): $\nu_{\max}/\text{cm}^{-1}$ = 2931, 1602, 1490, 1454, 1322, 1039, 1000, 740, 700. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.50 (d, J = 7.6 Hz, 1H), 7.33–7.18 (m, 7H), 7.08 (d, J = 6.8 Hz, 1H), 4.49 (t, J = 7.2 Hz, 1H), 3.07 (dd, J = 13.8, 5.2 Hz, 1H), 2.79–2.71 (m, 2H), 2.51 (dd, J = 13.7, 8.9 Hz, 1H), 2.08–2.02 (m, 1H), 1.98–1.95 (m, 1H), 1.72 (d, J = 6.8 Hz, 1H), 1.52–1.47 (m, 1H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 140.7, 138.9, 137.1, 129.6 (2C), 129.1, 128.7 (2C), 128.6, 127.8, 126.6, 126.4, 73.3, 44.3, 38.7, 28.0, 24.9.

1,3-diphenylpropan-1-ol (3ba)^{7,8}



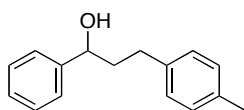
Yield (202 mg, 95%). IR (ATR): $\nu_{\max}/\text{cm}^{-1}$ = 3355, 3025, 2936, 1602, 1493, 1453, 1056, 910, 741, 695. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.37–7.20 (m 10H), 4.65 (t, J = 6.5 Hz, 1H), 2.78–2.64 (m, 2H), 2.40 (s, 1H), 2.16–2.00 (m, 2H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 144.8, 142.1, 128.6 (6C), 127.8, 126.2 (2C) 126.1, 74.0, 40.7, 32.3.

3-(4-methoxyphenyl)-1-phenylpropan-1-ol (3bb)⁷



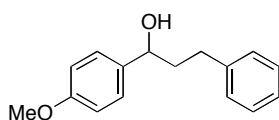
Yield (229 mg, 95%). IR (ATR): $\nu_{\max}/\text{cm}^{-1}$ = 3384, 2935, 1611, 1509, 1241, 1034, 826, 751, 699. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.32–7.30 (m, 4H), 7.26–7.24 (m, 1H), 7.07 (d, J = 8.3 Hz, 2H), 6.80 (d, J = 8.2 Hz, 2H), 4.62 (s, 1H), 3.75 (s, 3H), 2.67–2.63 (m, 1H), 2.60–2.55 (m, 1H), 2.17 (s, 1H), 2.09–2.03 (m, 1H), 1.98–1.93 (m, 1H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 158.0, 144.9, 134.1, 129.6 (2C), 128.7 (2C), 127.8, 126.2 (2C), 114.1 (2C), 74.0, 55.5, 41.0, 31.4.

3-(4-methyl)-1-phenylpropan-1-ol (3bc)⁷



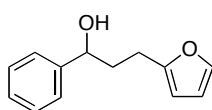
Yield (215 mg, 95%). IR (ATR): $\nu_{\max}/\text{cm}^{-1}$ = 3388, 3025, 2920, 2858, 1514, 1452, 1057, 913, 808, 753, 698. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.35–7.32 (m, 5H), 7.09–7.06 (m, 4H), 4.68–4.64 (m, 1H), 2.72–2.67 (m, 1H), 2.64–2.59 (m, 1H), 2.31 (s, 3H), 2.13–2.07 (m, 1H), 2.03–1.97 (m, 1H), 1.90 (s, 1H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 144.9, 139.0, 135.6, 129.4 (2C), 128.8 (2C), 128.6 (2C), 127.9, 126.3 (2C), 74.2, 40.9, 31.9, 21.3.

1-(4-methoxyphenyl)-3-phenylpropan-1-ol (3ca)⁵



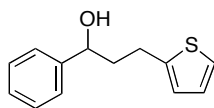
Yield (187 mg, 77%). IR (KBr): $\nu_{\max}/\text{cm}^{-1}$ = 3299, 2946, 2909, 1609, 1512, 1441, 1176, 1034, 829, 703. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.28–7.23 (m, 4H), 7.18–7.16 (m, 3H), 6.88–6.86 (m, 2H), 4.62–4.60 (m, 1H), 3.78 (s, 3H), 2.73–2.68 (m, 1H), 2.65–2.60 (m, 1H), 2.15–2.09 (m, 1H), 2.03–1.96 (m, 1H), 1.92 (m, 1H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 159.4, 142.2, 137.0, 128.8 (2C), 128.7 (2C), 127.5 (2C), 126.1, 114.2 (2C), 73.8, 55.6, 40.7, 32.4.

3-furyl-1-phenylpropan-1-ol (3bd)^{8,9}



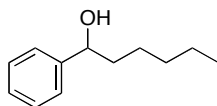
Yield (156 mg, 77%). IR (ATR): $\nu_{\max}/\text{cm}^{-1}$ = 3406, 2921, 1597, 1493, 1452, 1148, 1058, 1004, 931, 803, 729, 698. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.37–7.24 (m, 6H), 6.28–6.27 (m, 1H), 6.00 (d, J = 2.0 Hz, 1H), 4.70–4.68 (m, 1H), 2.76–2.67 (m, 2H), 2.15–2.02 (m, 2H), 1.98 (d, J = 2.8 Hz, 1H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 155.6, 144.7, 141.3, 128.9 (2C), 128.0, 126.2 (2C), 110.5, 105.4, 74.0, 37.5, 24.7.

1-phenyl-3-thiophenylpropan-1-ol (3be)



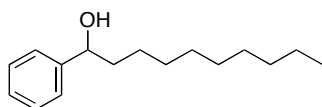
Yield (57 mg, 26%). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 3388, 2920, 1721, 1492, 1451, 1276, 1056, 914, 849, 752, 694$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.35–7.25 (m, 5H), 7.09 (d, $J = 4.1$ Hz, 1H), 6.90 (dd, $J = 5.2$ Hz, 3.4 Hz, 1H), 6.78 (t, $J = 1.7$ Hz, 1H), 4.68 (t, $J = 5.9$ Hz, 1H), 2.95–2.85 (m, 2H), 2.18–2.12 (m, 1H), 2.07–2.02 (m, 2H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 144.9, 144.7, 128.9 (2C), 128.0, 127.1, 126.2 (2C), 124.6, 123.4, 73.8, 41.0, 26.5. HRMS (EI) Calcd for $\text{C}_{13}\text{H}_{14}\text{OS}$: 218.0765. Found $m/z = 218.0756$.

1-phenylhexan-1-ol (3bf)⁷



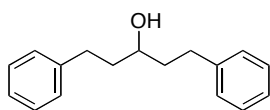
Yield (148 mg, 83%). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 3377, 2929, 2857, 1592, 1454, 1377, 1024, 914, 758, 698$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.35–7.24 (m, 5H), 4.65–4.60 (m, 1H), 1.94 (s, 1H), 1.76–1.79 (m, 1H), 1.71–1.67 (m, 1H), 1.43–1.38 (m, 1H), 1.30–1.24 (m, 5H), 0.91–0.86 (m, 3H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 145.3, 128.7 (2C), 127.8, 126.2 (2C), 75.0, 39.4, 32.1, 25.8, 22.9, 14.3.

1-phenyldecan-1-ol (3bg)⁷



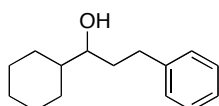
Yield (208 mg, 89%). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 3372, 2922, 2853, 1454, 1376, 1216, 1026, 757, 698$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.33–7.24 (m, 5H), 4.64–4.62 (m, 1H), 1.94 (s, 1H), 1.82–1.76 (m, 1H), 1.71–1.65 (m, 1H), 1.43–1.38 (m, 1H), 1.31–1.23 (m, 13H), 0.88–0.86 (m, 3H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 145.3, 128.7 (2C), 127.8, 126.2 (2C), 75.0, 39.4, 32.2, 29.9 (2C), 29.7, 29.6, 26.2, 23.0, 14.4.

1,5-diphenylpentan-3-ol (3da)



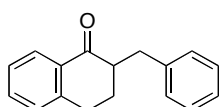
Yield (120 mg, 50%). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 3364, 3024, 2919, 1602, 1595, 1454, 1029, 911, 743, 696$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.29–7.26 (m, 4H), 7.19–7.17 (m, 6H), 3.68–3.65 (m, 1H), 2.81–2.76 (m, 2H), 2.69–2.64 (m, 2H), 1.85–1.74 (m, 4H), 1.44 (s, 1H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 142.4 (2C), 128.8 (8C), 126.2 (2C), 71.2, 39.5, 32.4. HRMS (EI) Calcd for $\text{C}_{17}\text{H}_{22}$ (M – H_2O): 222.1408. Found $m/z = 222.1406$.

1-cyclohexyl-3-phenylpropan-1-ol (3ea)¹⁰



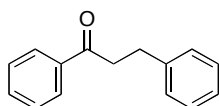
Yield (129 mg, 59%). IR (KBr): $\nu_{\max}/\text{cm}^{-1} = 3300, 2912, 2849, 1496, 1454, 1347, 1060, 1031, 745, 696$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.29–7.17 (m, 5H), 3.39 (bs, 1H), 2.86–2.81 (m, 1H), 2.67–2.62 (m, 1H), 1.84–1.65 (m, 7H), 1.37–0.97 (m, 7H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 142.7, 128.7 (4C), 126.1, 77.1, 44.1, 36.3, 32.7, 29.5, 28.1, 26.9, 26.8, 26.5.

2-benzyl-1,2,3,4-tetrahydronaphthalen-1-one (4aa)¹¹



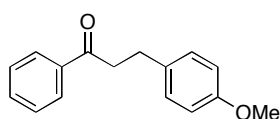
Yield (17 mg, 7%). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 3024, 2927, 1677, 1598, 1453, 1289, 1217, 932, 737, 698$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 8.07 (d, $J = 6.9$ Hz, 1H), 7.44 (t, $J = 7.9$ Hz, 1H), 7.31–7.28 (m, 3H), 7.23–7.20 (m, 4H), 3.49 (t, $J = 13.7, 3.8$ Hz, 1H), 2.98–2.88 (m, 2H), 2.76–2.70 (m, 1H), 2.66–2.60 (m, 1H), 2.12–2.07 (m, 1H), 1.82–1.74 (m, 1H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 199.6, 144.3, 140.3, 133.6, 132.8, 129.6(2C), 129.0, 128.7 (2C), 127.8, 126.9, 126.4, 49.7, 36.0, 28.9, 28.0.

1,3-diphenylpropan-1-one (4ba)¹²



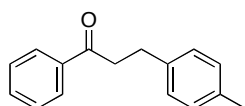
Yield (4.2 mg, 2%). IR (KBr): $\nu_{\max}/\text{cm}^{-1} = 3024, 2920, 1681, 1595, 1494, 1448, 1208, 744, 700$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.96 (d, $J = 6.8$ Hz, 2H), 7.55 (t, $J = 7.2$ Hz, 1H), 7.45 (t, $J = 7.6$ Hz, 2H), 7.31–7.19 (m, 5H), 3.31 (t, $J = 7.9$ Hz, 2H), 3.07 (t, $J = 7.9$ Hz, 2H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 199.5, 141.6, 137.2, 133.4, 128.9 (2C), 128.8 (2C), 128.7 (2C), 128.4 (2C), 126.5, 40.8, 30.4.

3-(4-methoxyphenyl)-1-phenylpropan-1-one (4bb)¹³



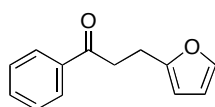
Yield (12.0 mg, 5%). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 2934, 1682, 1510, 1447, 1242, 1177, 1033, 974, 824, 742, 689$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.95 (d, $J = 6.8$ Hz, 2H), 7.55 (t, $J = 6.8$ Hz, 1H), 7.45 (t, $J = 7.5$ Hz, 2H), 7.17 (d, $J = 8.9$ Hz, 2H), 6.84 (d, $J = 6.2$ Hz, 2H), 3.78 (s, 3H), 3.27 (t, $J = 7.6$ Hz, 2H), 3.01 (t, $J = 7.9$ Hz, 2H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 199.7, 185.3, 137.2, 133.6, 133.4, 129.7 (2C), 128.9 (2C), 128.4 (2C), 114.3 (2C), 55.6, 41.1, 29.6.

3-(4-methylphenyl)-1-phenylpropan-1-one (4bc)



Yield (8.9 mg, 4%). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 2920$ (CH), 1684 (C=O), 1597, 1579, 1515, 1447, 1360, 1290, 1202, 973. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.97 (d, $J = 7.6$ Hz, 2H), 7.55 (t, $J = 7.6$ Hz, 1H), 7.45 (t, $J = 7.5$ Hz, 2H), 7.16 (d, $J = 7.6$ Hz, 2H), 7.12 (d, $J = 8.2$ Hz, 2H), 3.28 (t, $J = 7.9$ Hz, 2H), 3.04 (t, $J = 7.6$ Hz, 2H), 2.33 (s, 3H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 199.7, 138.7, 137.3, 136.1, 133.5, 129.7 (2C), 129.1 (2C), 128.8 (2C), 128.5 (2C), 41.0, 30.2, 21.5. HRMS (FAB) Calcd for $\text{C}_{16}\text{H}_{16}\text{O}$: 224.1201. Found $m/z = 224.1200$.

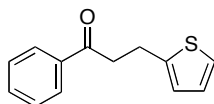
3-furyl-1-phenylpropan-1-one (4bd)¹³



Yield (10 mg, 5%). IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 2916, 1683, 1596, 1448, 1363, 1205, 1148, 1010, 974, 920, 727, 688$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.97 (dd, $J = 8.3, 1.4$ Hz, 2H), 7.56 (t,

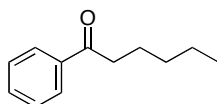
$J = 7.2$ Hz, 1H), 7.46 (t, $J = 7.9$ Hz, 2H), 7.31 (s, 1H), 6.3 (d, $J = 2.1$ Hz, 1H), 6.05 (d, $J = 4.1$ Hz, 1H), 3.34 (t, $J = 7.6$ Hz, 2H), 3.09 (t, $J = 7.6$ Hz, 2H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 199.0, 155.1, 141.5, 137.0, 133.4, 129.0 (2C), 128.4 (2C), 110.6, 105.7, 37.3, 22.8.

1-phenyl-3-thiophenylpropan-1-one (4be)



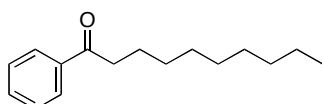
Yield (~1%). IR (ATR): $\nu_{\text{max}}/\text{cm}^{-1} = 3103, 2922, 1682, 1594, 1446, 1363, 1207, 971, 852, 748, 707, 692$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.97 (d, $J = 8.3$ Hz, 2H), 7.57 (t, $J = 7.6$, 1H), 7.47 (t, $J = 7.6$ Hz, 2H), 7.13–7.12 (m, 1H), 6.93–6.91 (m, 1H), 6.87 (t, $J = 1.4$ Hz, 1H), 3.38–3.35 (m, 2H), 3.32–3.29 (m, 2H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 198.9, 144.2, 137.1, 133.5, 129.0, 128.4 (2C), 127.2 (2C), 125.0, 123.7, 40.9, 24.5. HRMS (FAB) Calcd for $\text{C}_{13}\text{H}_{12}\text{OS}$: 216.0609. Found $m/z = 216.0607$.

1-phenylhexan-1-one (4bf)⁷



Yield (16 mg, 9%). IR (ATR): $\nu_{\text{max}}/\text{cm}^{-1} = 2930, 2859, 1683, 1596, 1448, 1362, 1232, 1203, 1179, 970, 746, 689$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.96 (d, $J = 7.6$ Hz, 2H), 7.55 (t, $J = 7.2$ Hz, 1H), 7.47–7.45 (m, 2H), 2.96 (t, $J = 7.2$ Hz, 2H), 1.77–1.72 (m, 2H), 1.38–1.36 (m, 4H), 0.93–0.90 (m, 3H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 201.0, 137.5, 133.2, 128.9 (2C), 128.4 (2C), 38.9, 31.9, 24.4, 22.9, 14.3.

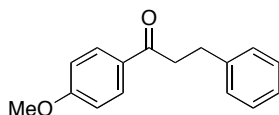
1-phenyldecan-1-one (4bg)



Yield (23 mg, 10%). IR (KBr): $\nu_{\text{max}}/\text{cm}^{-1} = 2918, 2846, 1686, 1596, 1473, 1447, 1375, 1256, 1220, 1193, 970, 733, 688$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.96 (d, $J = 7.6$ Hz, 2H), 7.55 (t, $J = 7.2$ Hz, 1H), 7.46 (t, $J = 7.6$ Hz, 2H), 2.96 (t, $J = 7.6$ Hz, 2H), 1.76–1.71 (m, 2H), 1.39–1.28 (m, 12H), 0.88 (t, $J = 7.2$ Hz, 3H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 201.0,

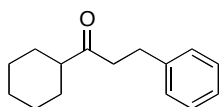
137.5, 133.2, 128.9 (2C), 128.4 (2C), 39.0, 32.2, 29.7 (4C), 24.7, 23.0, 14.4. HRMS (EI) Calcd for C₁₆H₂₄O: 232.1827. Found m/z = 232.1818.

1-(4-methoxyphenyl)-3-phenylpropan-1-one (4ca)¹⁴



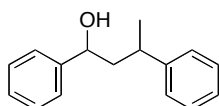
Yield (43 mg, 18%). IR (ATR): $\nu_{\max}/\text{cm}^{-1}$ = 2931, 1682, 1509, 1447, 1242, 1203, 1177, 1033, 974, 823, 724, 699. ¹H NMR (600 MHz, CDCl₃) δ (ppm): 7.93 (d, J = 8.9 Hz, 2H), 7.30–7.24 (m, 4H), 7.19 (t, J = 7.3 Hz, 1H), 6.91 (d, J = 8.9 Hz, 2H), 3.84 (s, 3H), 3.24 (t, J = 7.9 Hz, 2H), 3.05 (t, J = 7.9 Hz, 2H). ¹³C NMR (600MHz, CDCl₃) δ (ppm): 198.1, 163.7, 141.8, 130.6 (2C), 130.2, 128.8 (2C), 128.7 (2C), 126.4, 114.0 (2C), 55.7, 40.4, 30.6.

1-cyclohexyl-3-phenylpropan-1-one (4ea)¹⁵



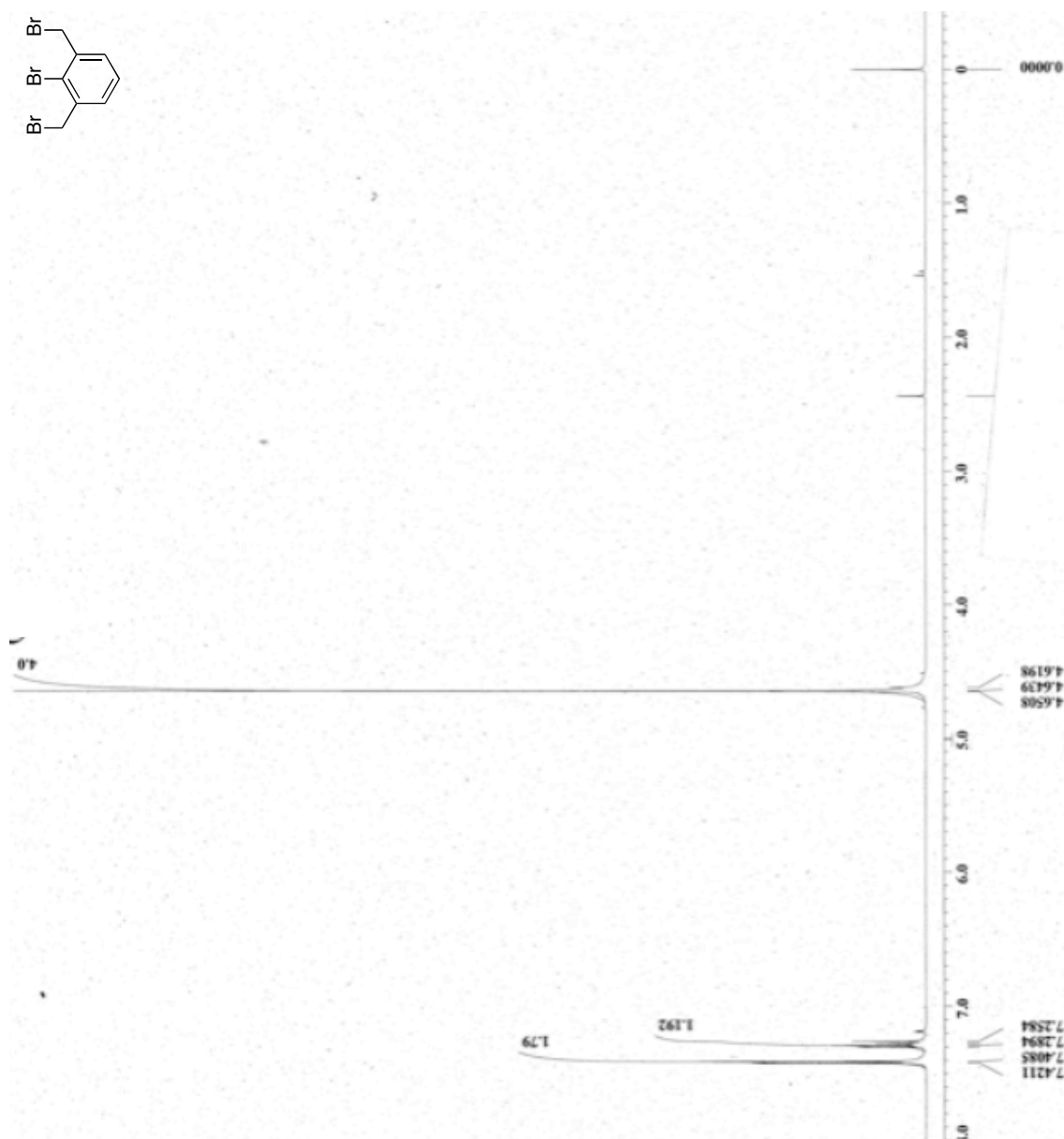
Yield (45 mg, 21%). IR (ATR): $\nu_{\max}/\text{cm}^{-1}$ = 2926, 2853, 1704, 1495, 1449, 1373, 1142, 990, 749, 697. ¹H NMR (600 MHz, CDCl₃) δ (ppm): 7.28–7.24 (m, 2H), 7.18–7.16 (m, 3H), 2.87 (t, J = 7.6 Hz, 2H), 2.75 (t, J = 7.9 Hz, 2H), 2.33–2.27 (m, 1H), 1.82–1.72 (m, 4H), 1.68–1.61 (m, 1H), 1.35–1.12 (m, 5H). ¹³C NMR (600MHz, CDCl₃) δ (ppm): 213.4, 141.7, 128.7 (2C), 128.6 (2C), 126.3, 51.2, 42.5, 30.0, 28.7 (2C), 26.1, 25.9 (2C).

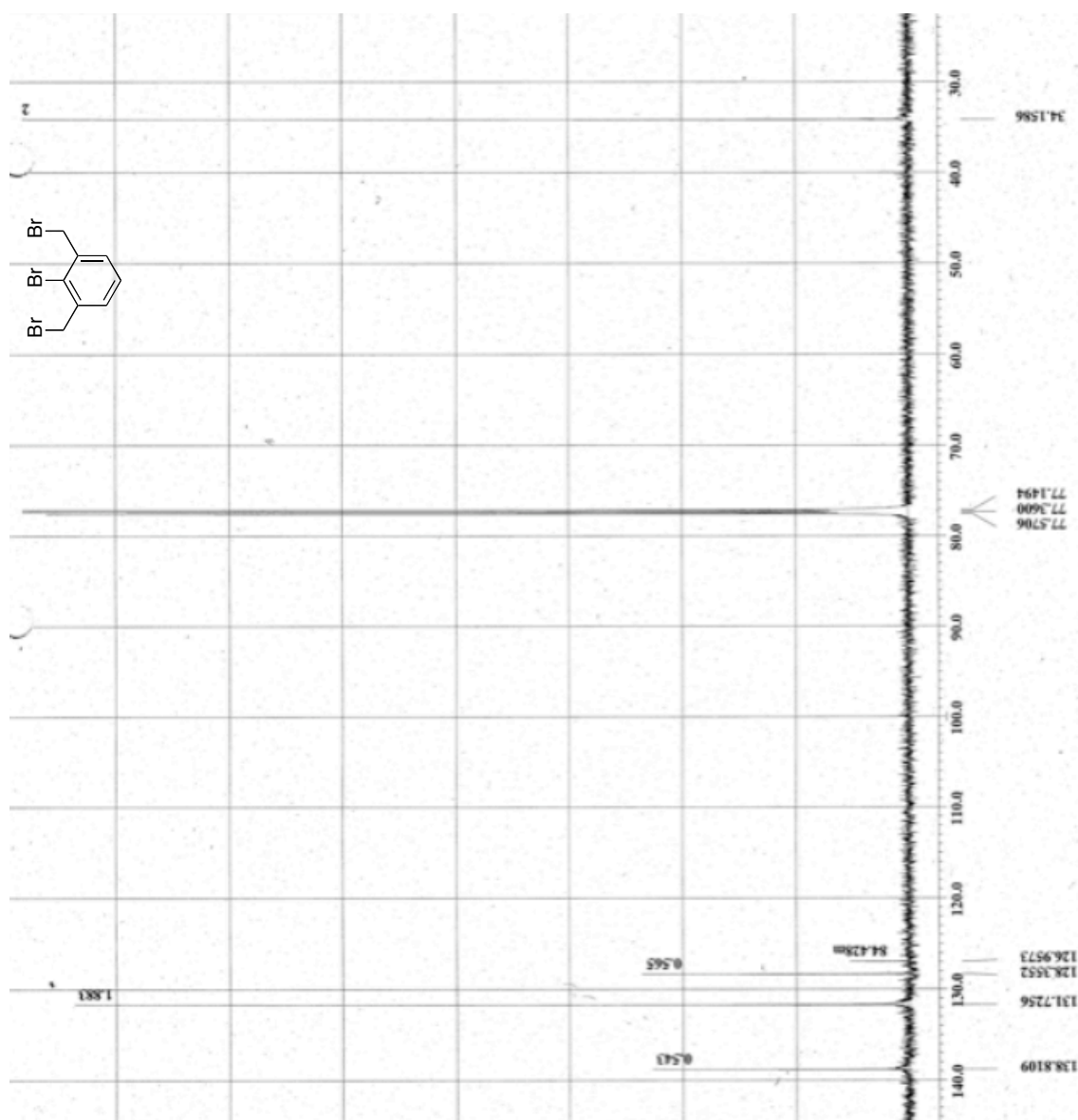
1,3-diphenyl-3-methylpropan-1-ol (8) (*cis* and *trans*)

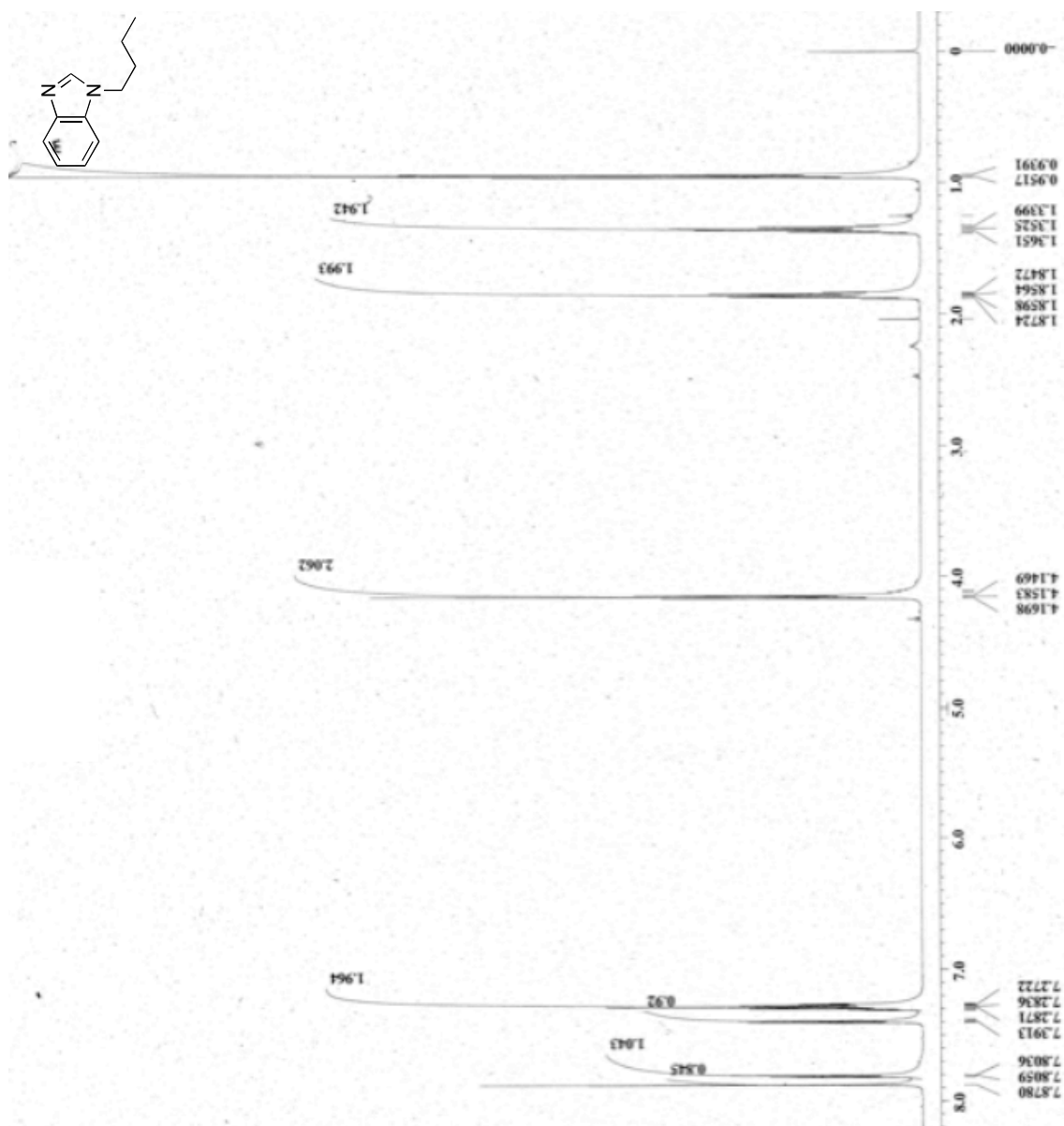


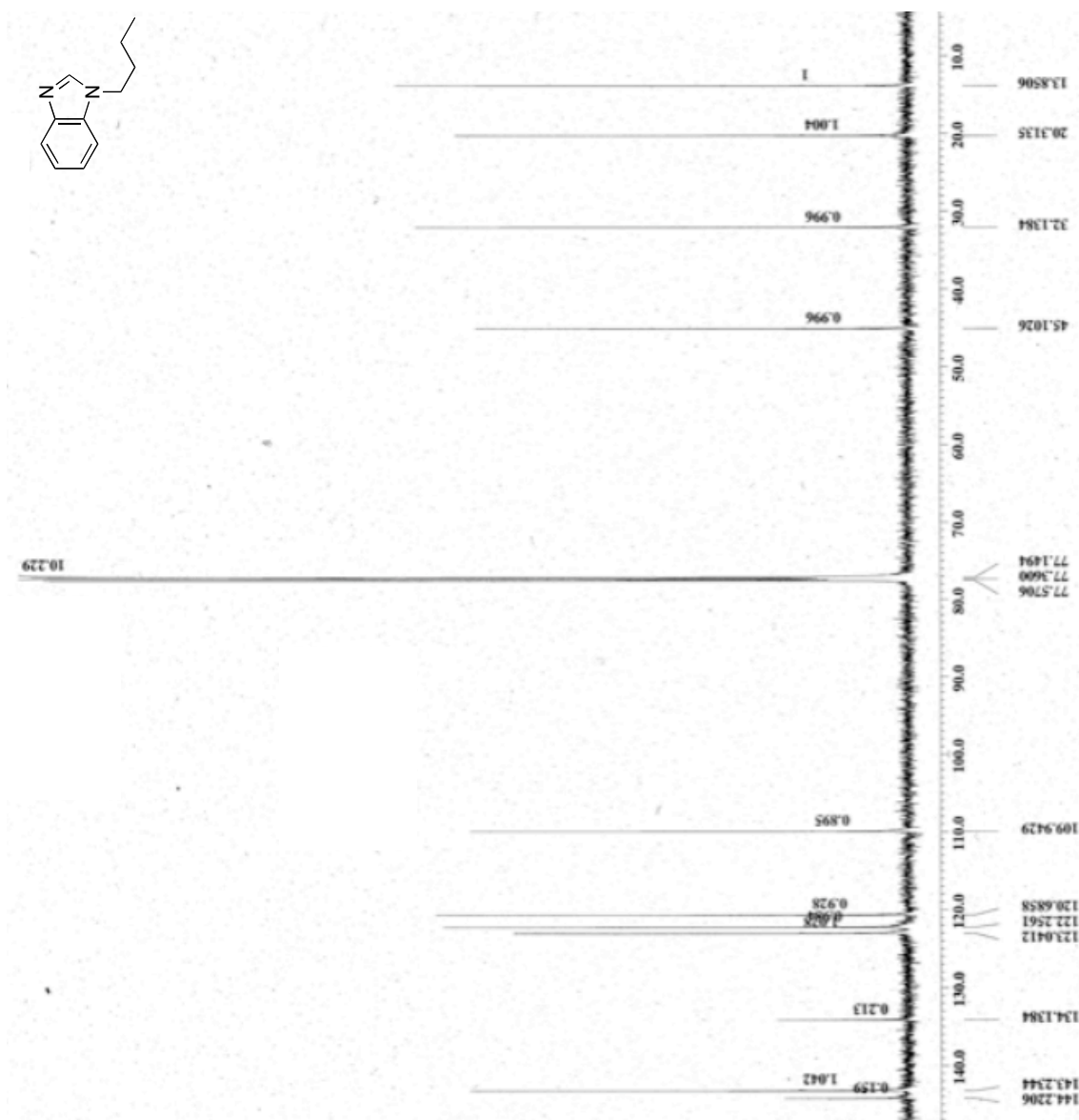
Diastereomer: Yield (158 mg, 70% (for two diastereomers)). IR (ATR): $\nu_{\max}/\text{cm}^{-1}$ = 2926, 2853, 1704, 1495, 1449, 1373, 1142, 990, 749, 697. ¹H NMR (600 MHz, CDCl₃) δ (ppm): 7.33–7.29 (m, 4H), 7.25–7.20 (m, 6H), 4.41–4.39 (m, 1H), 3.05–2.99 (m, 1H), 2.06–2.01 (m, 1H), 1.94–1.90 (m, 1H), 1.78 (d, J = 3.4 Hz, 1H), 1.28 (d, J = 6.8 Hz, 3H). ¹³C NMR (600MHz, CDCl₃) δ (ppm): 146.9, 145.5, 128.9 (2C), 128.8 (2C), 127.8, 127.5 (2C), 126.5, 126.0 (2C), 72.6, 48.0, 37.0, 23.4.

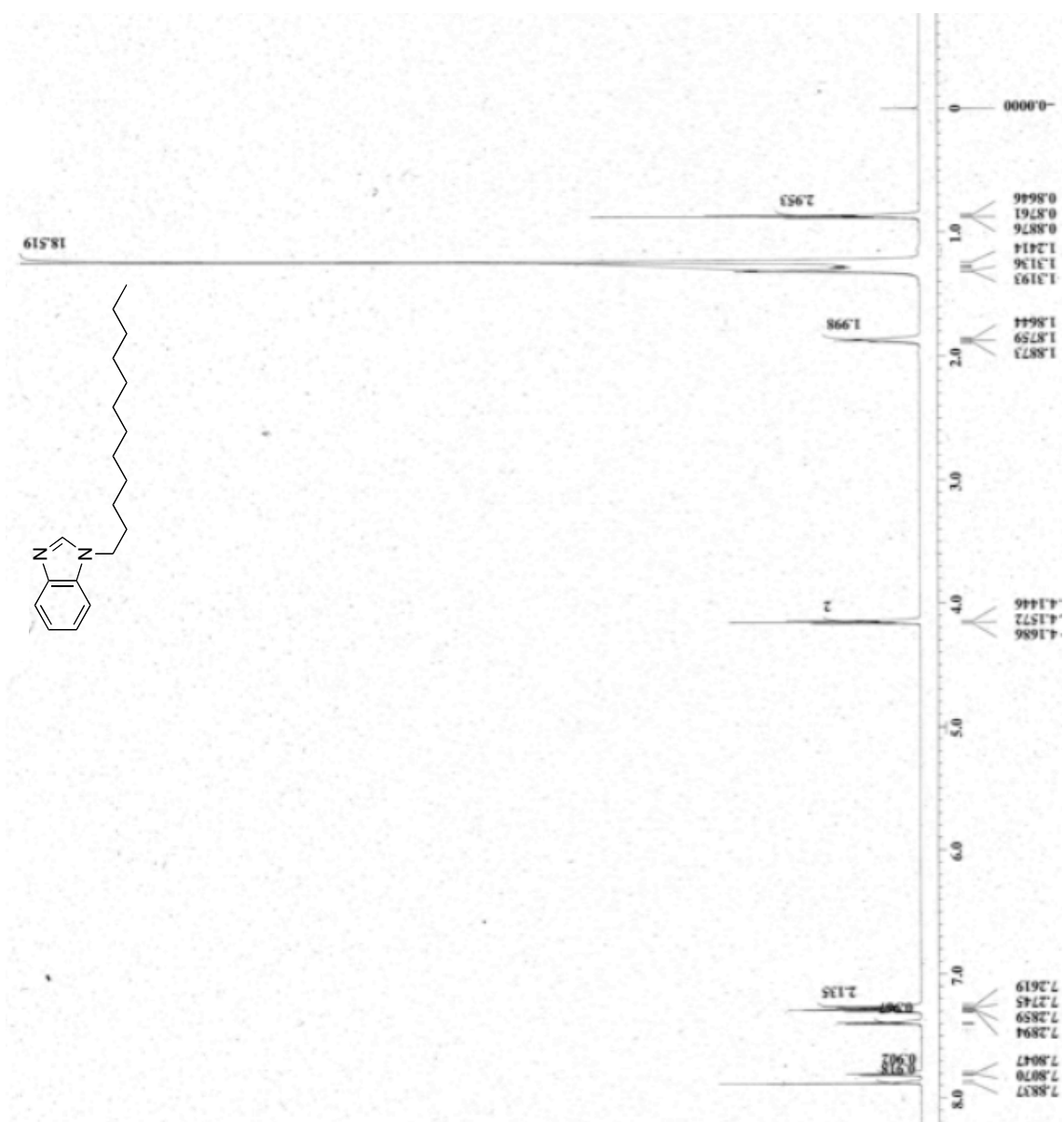
Another diastereomer: IR (ATR): $\nu_{\max}/\text{cm}^{-1} = 3357, 3026, 2925, 1602, 1492, 1452, 1051, 1015, 908, 760, 697$. ^1H NMR (600 MHz, CDCl_3) δ (ppm): 7.36–7.19 (m, 10H), 4.58–4.56 (m, 1H), 2.76–2.70 (m, 1H), 2.21–2.16 (m, 1H), 1.97–1.92 (m, 1H), 1.71 (d, $J = 2.8$ Hz, 1H), 1.27 (d, $J = 6.9$ Hz, 3H). ^{13}C NMR (600MHz, CDCl_3) δ (ppm): 147.3, 144.9, 128.7, 128.1, 127.4, 126.5, 73.3, 47.6, 37.0, 22.9. HRMS (EI) Calcd for $\text{C}_{16}\text{H}_{18}\text{O}$ (two diastereomers): 226.1358. Found $m/z = 226.1361$. The chiral HPLC analytical data for **8** were obtained using *i*-PrOH/hexane (2.5/97.5) as eluent at a flow rate of 1.0 mL/min. Column: OD-H, $t_R = 10.7$ min. and 11.9 min for the two enantiomers of either *cis* or *trans* isomer; $t_R = 14.8$ min and 15.5 min for the two enantiomers of either *cis* or *trans* isomer.

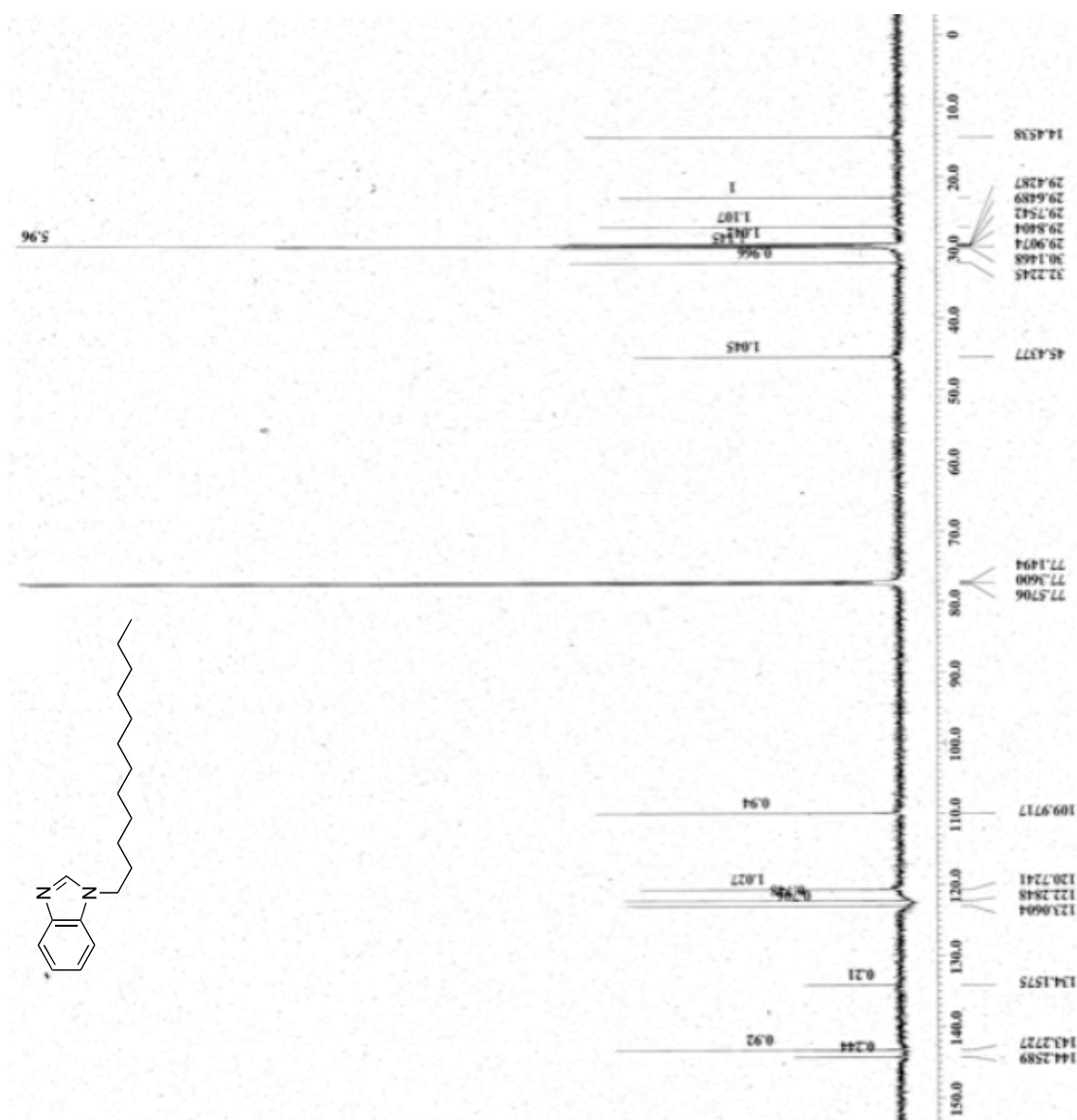


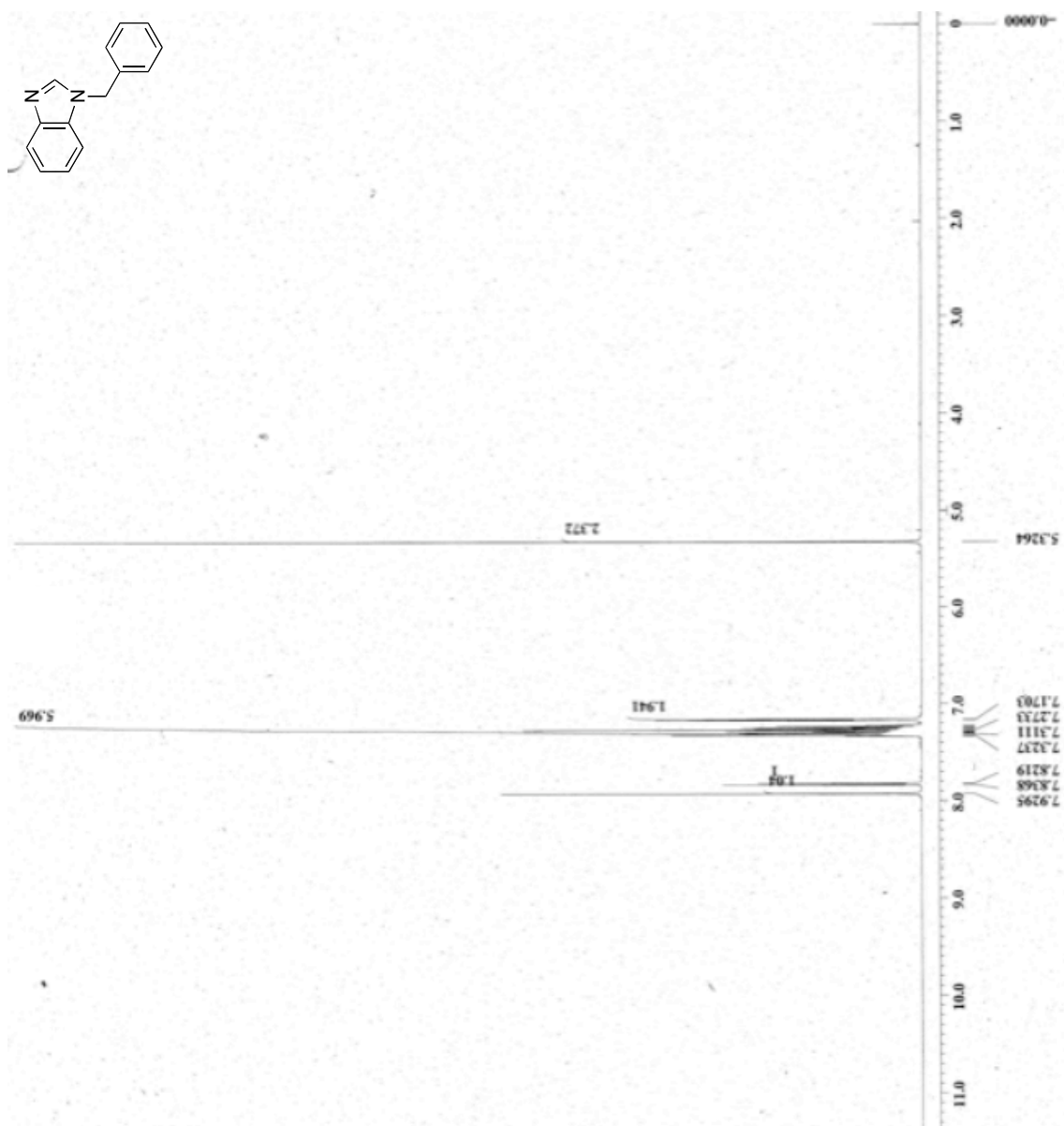


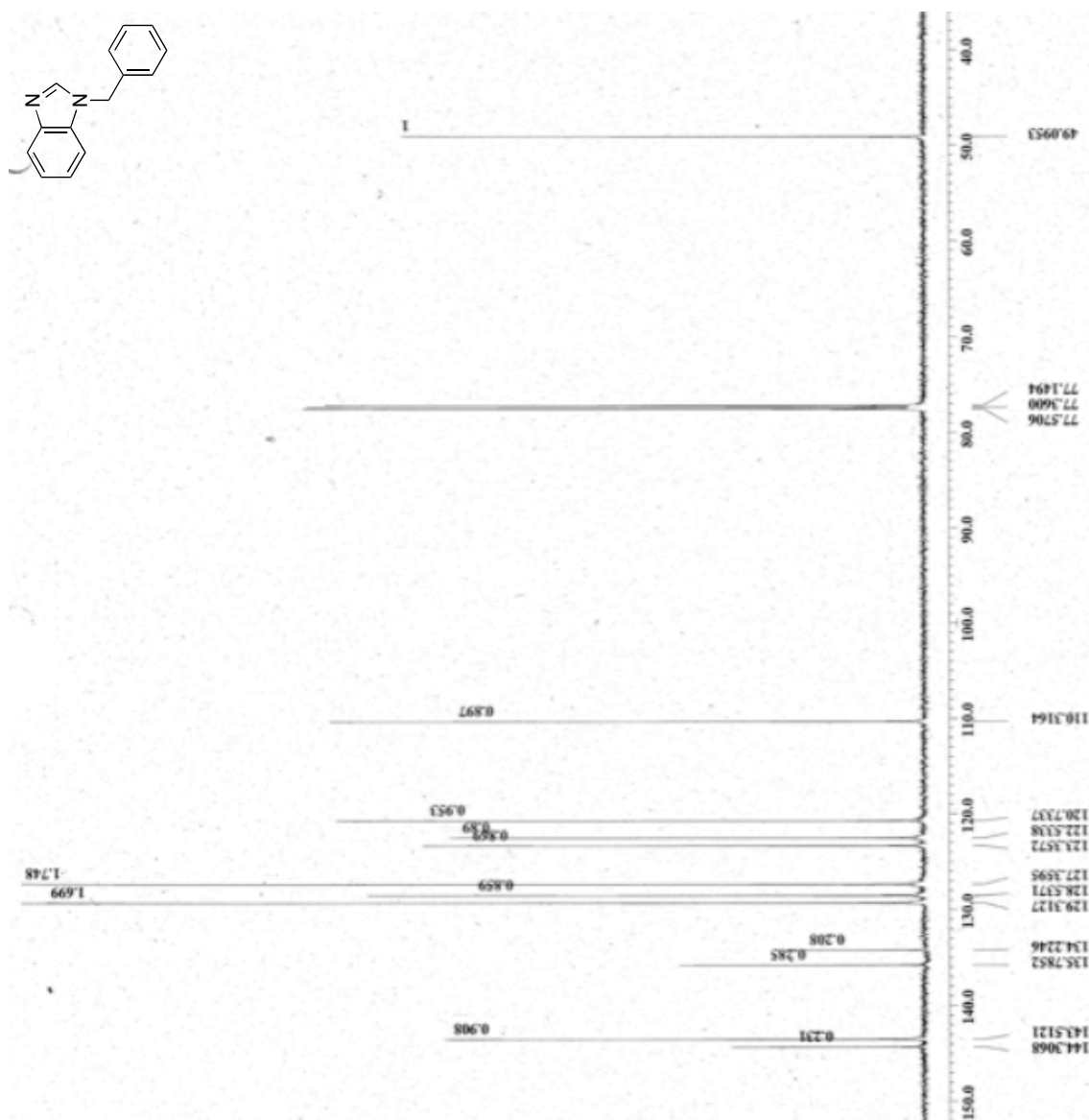


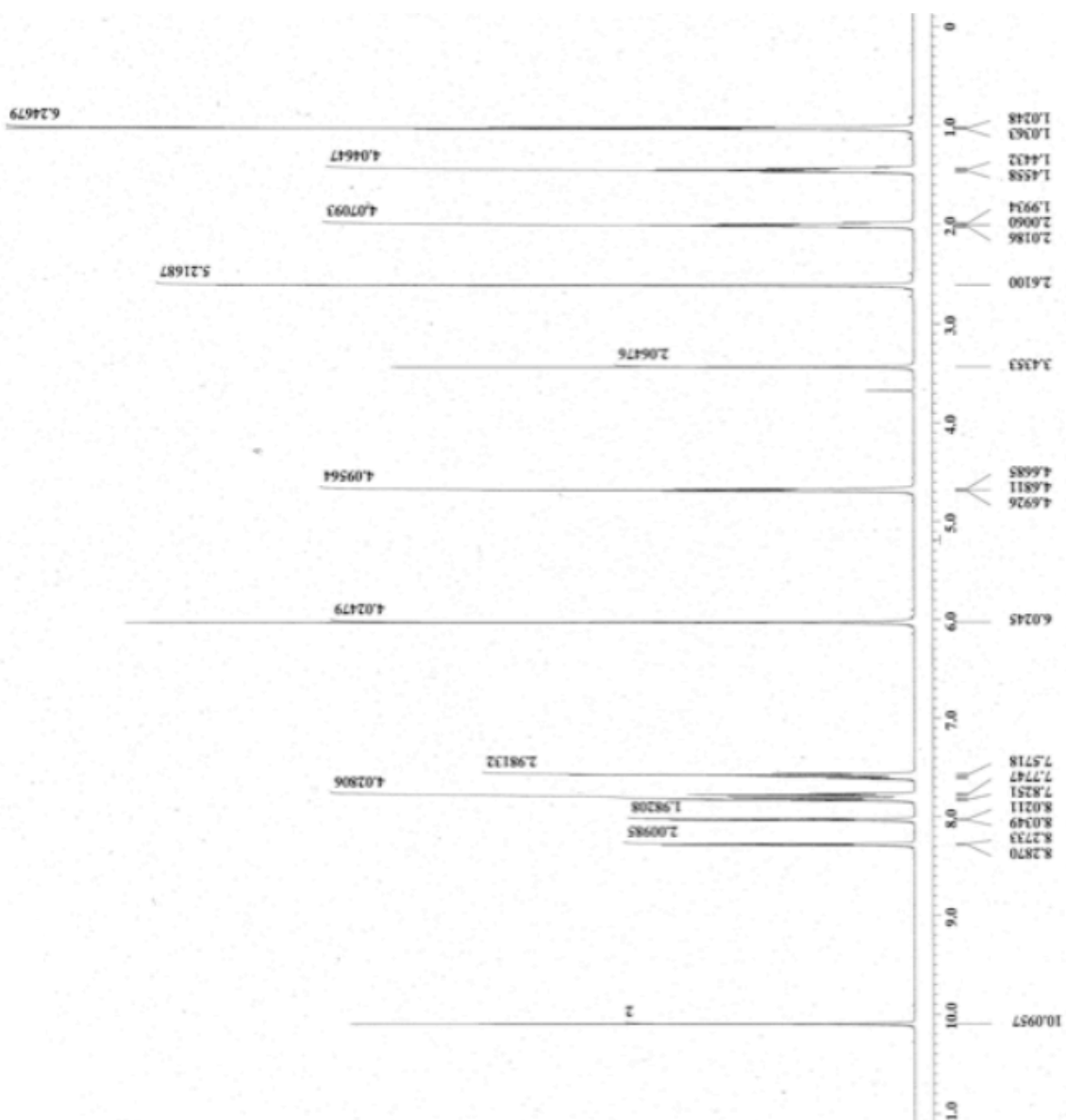


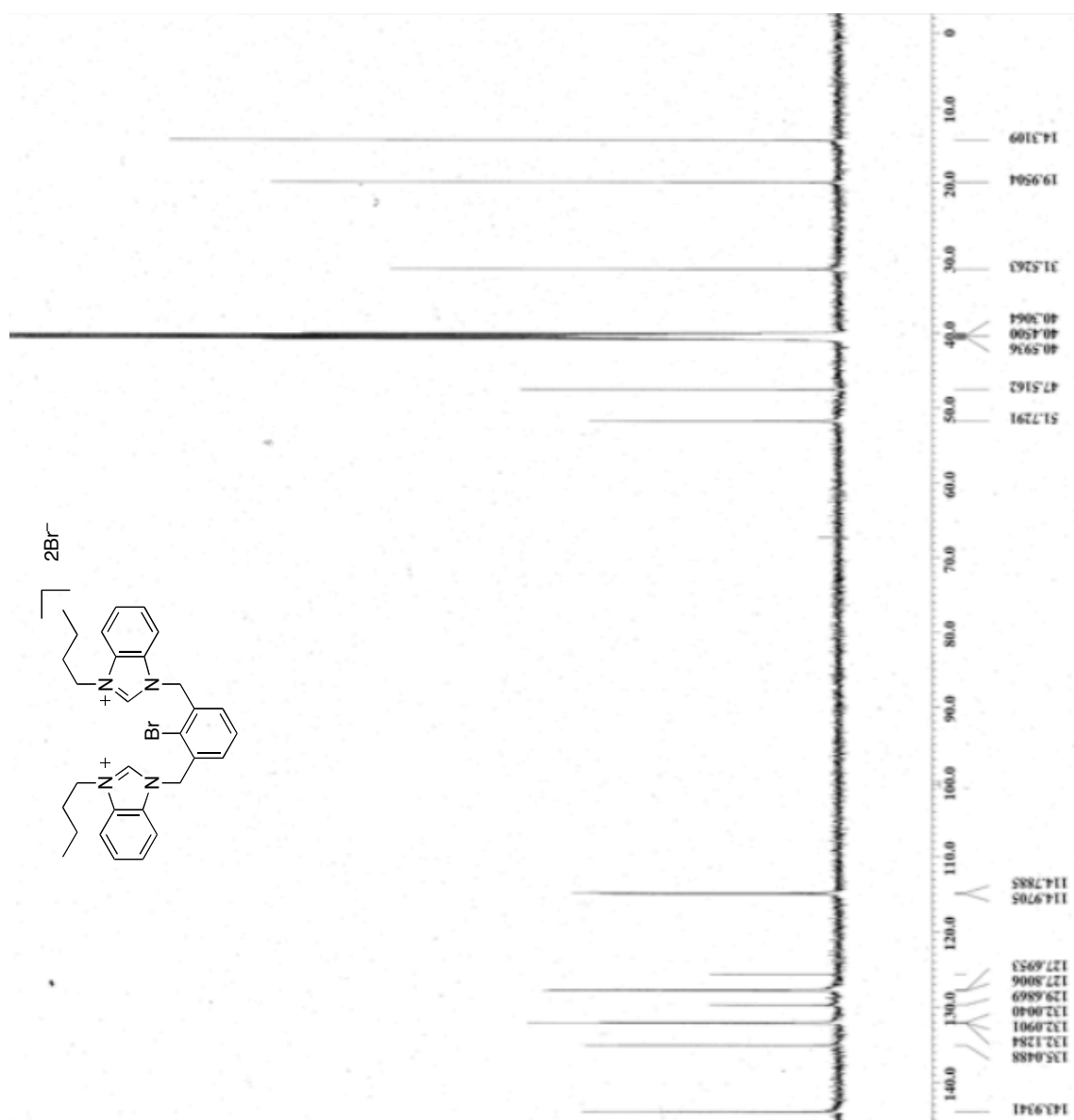


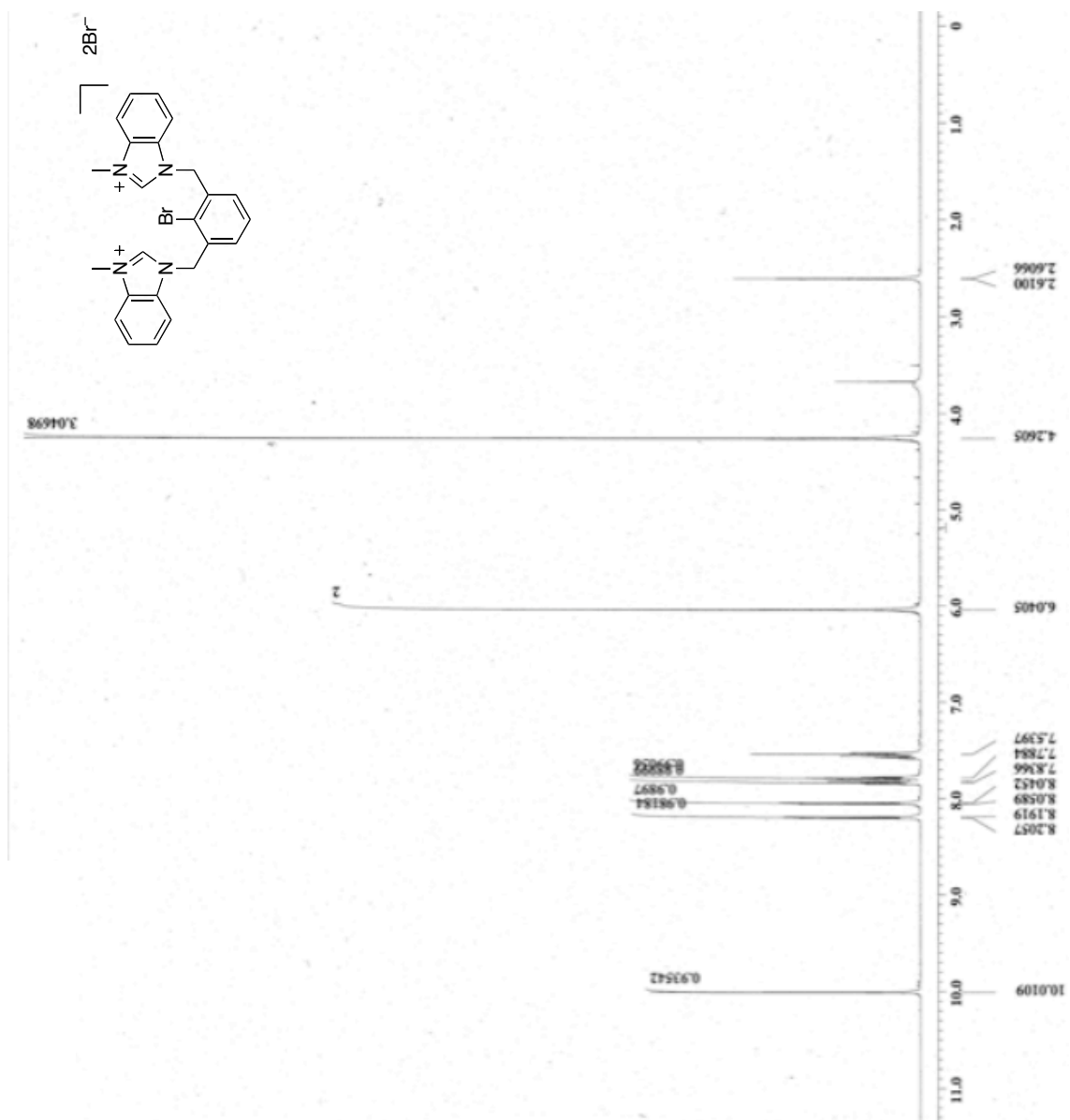


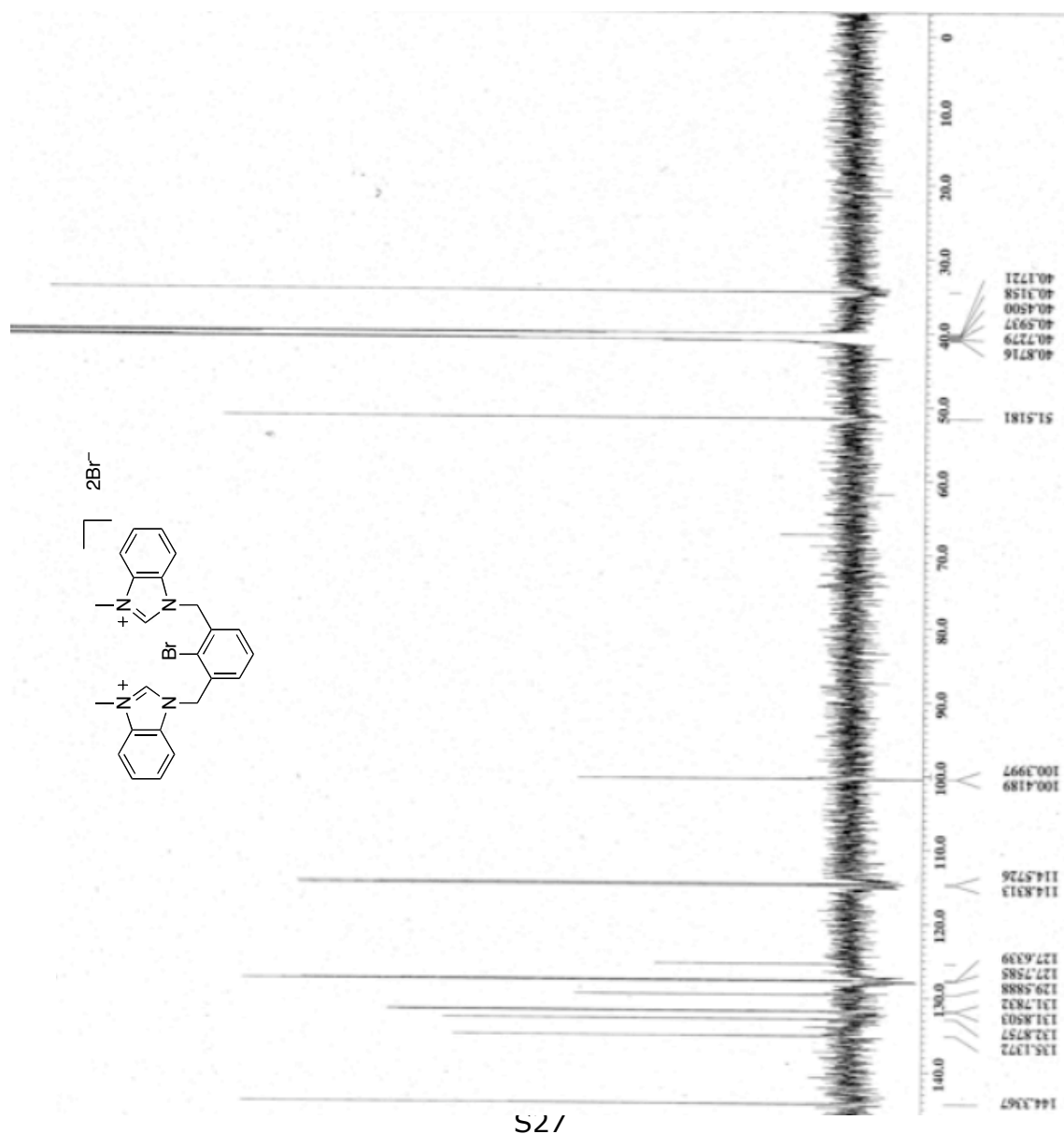


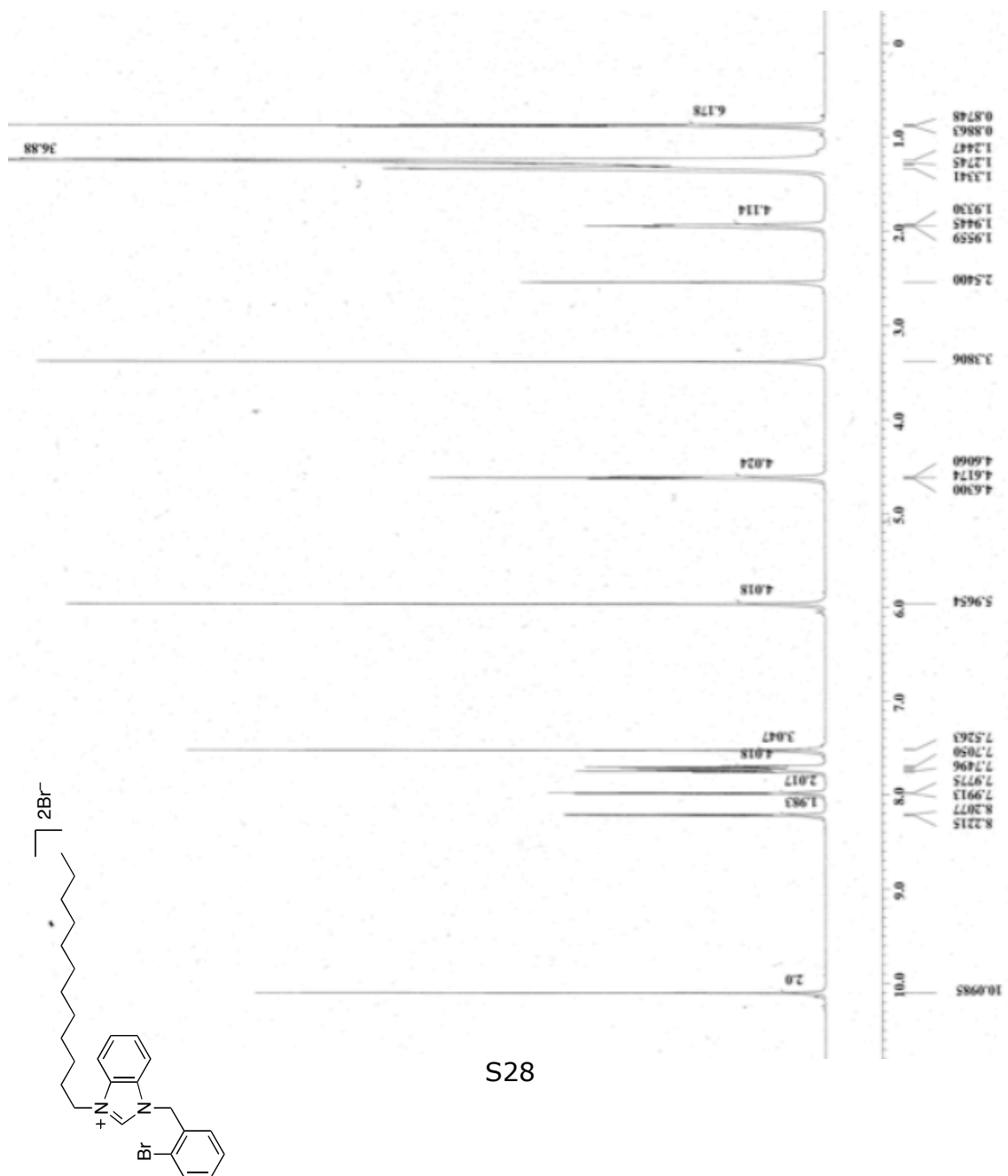


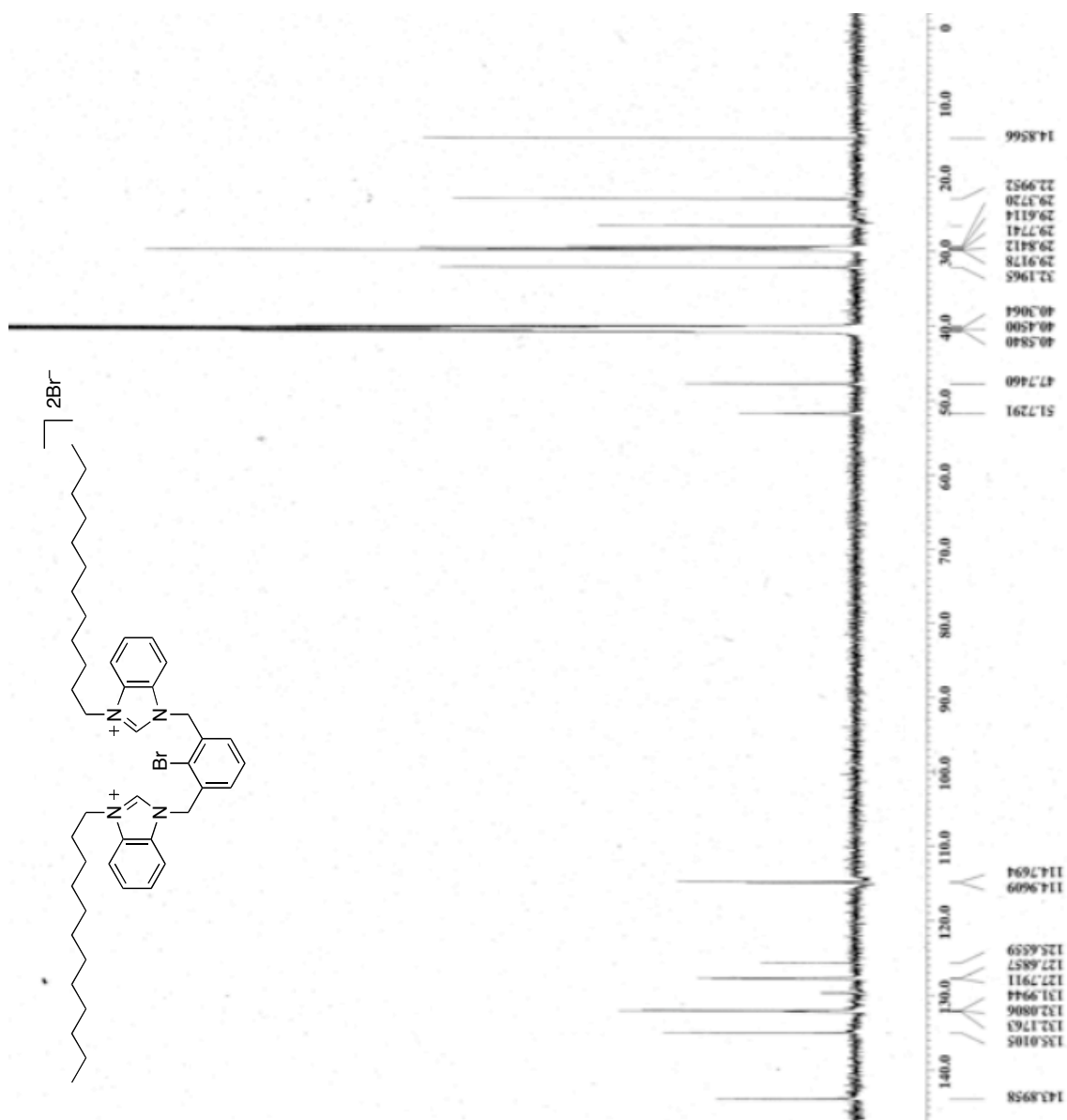


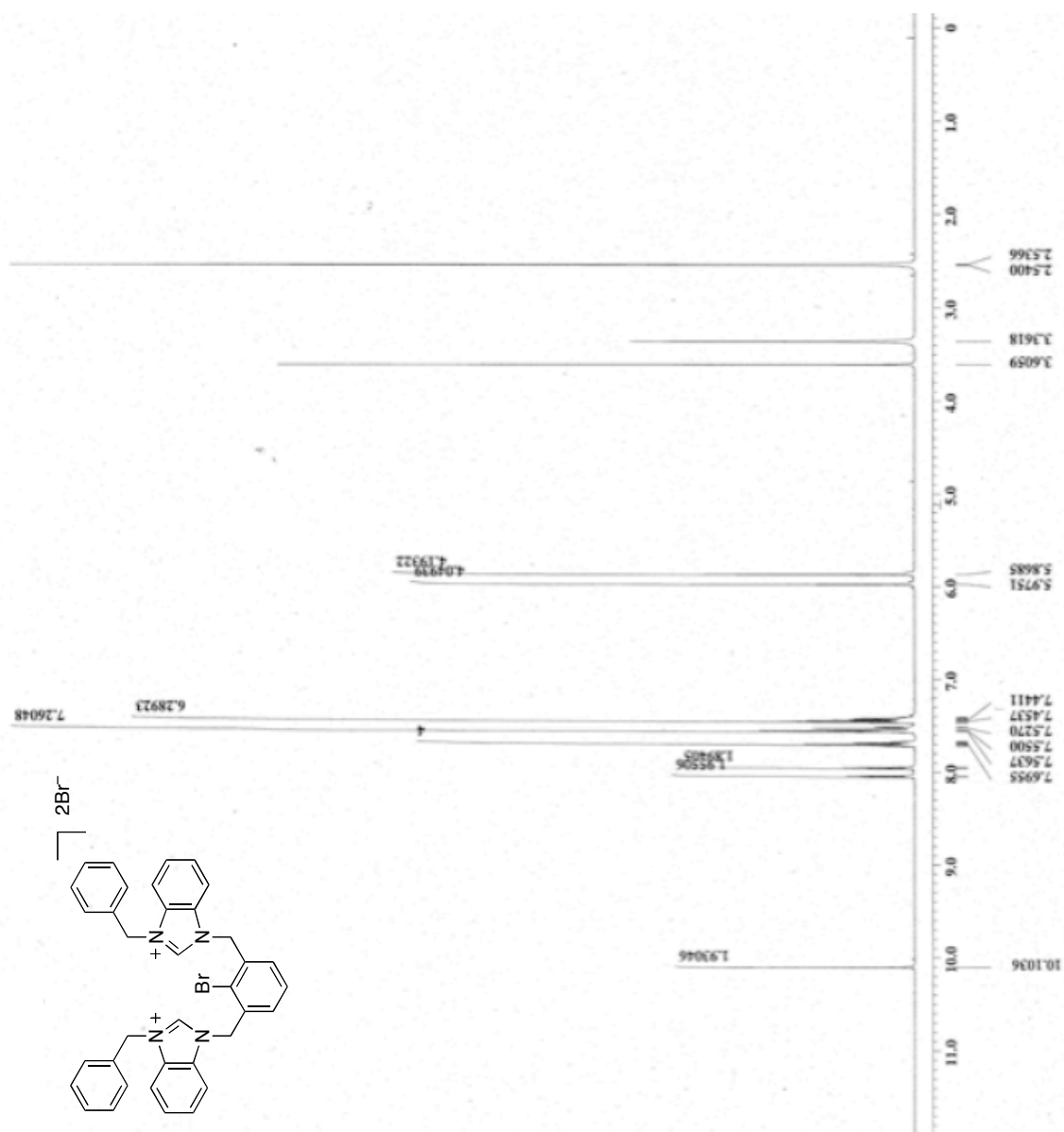


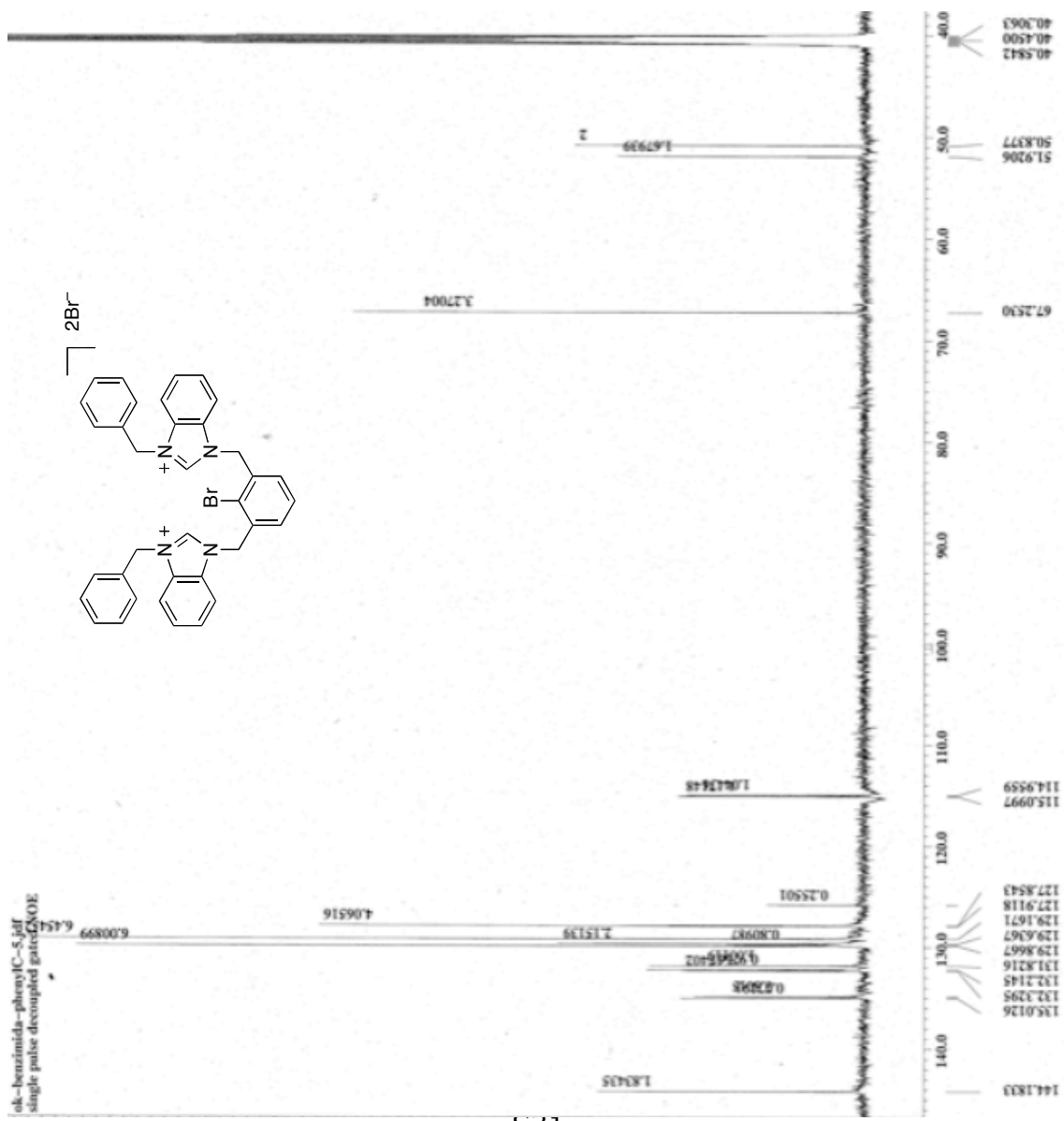


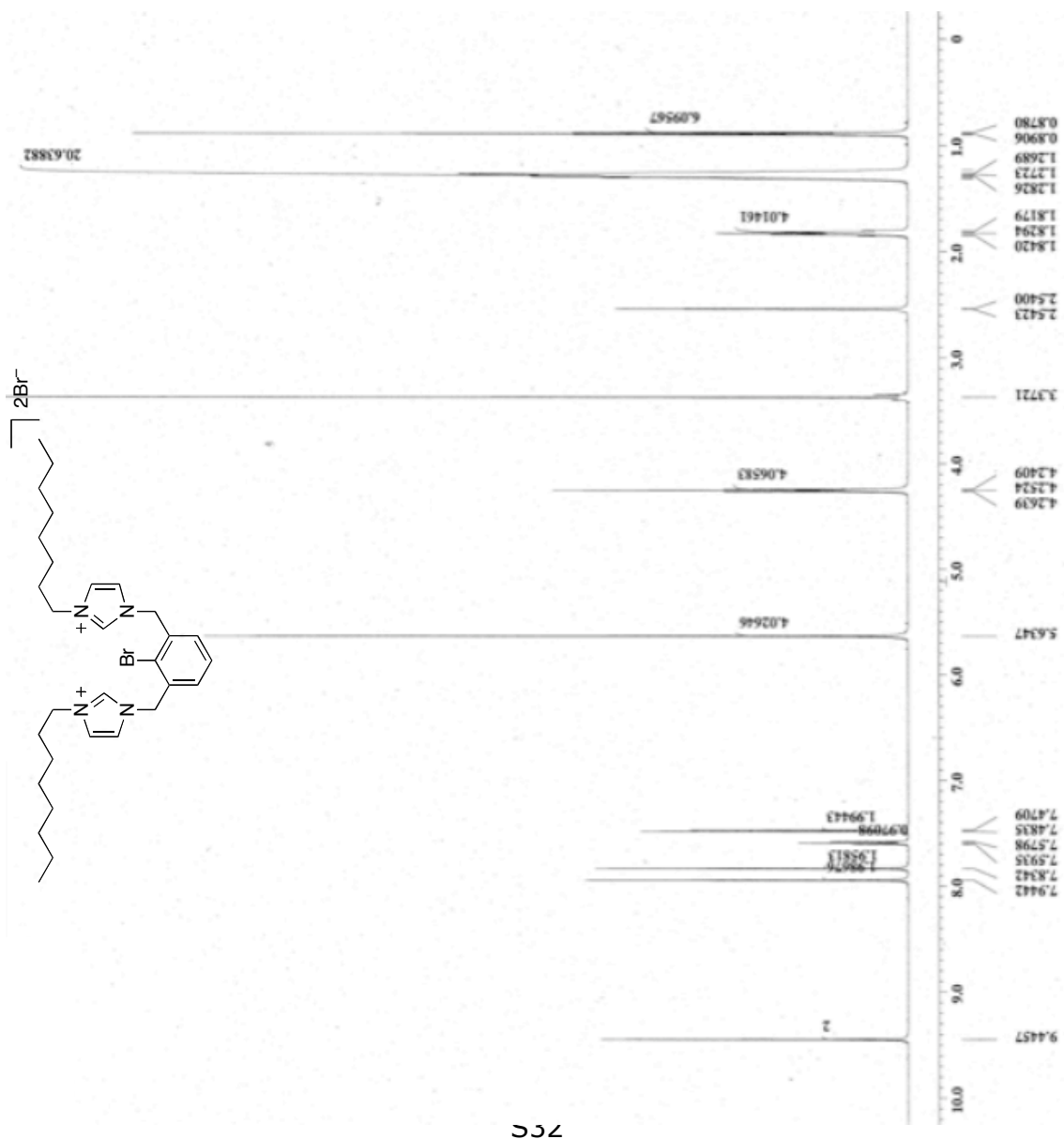


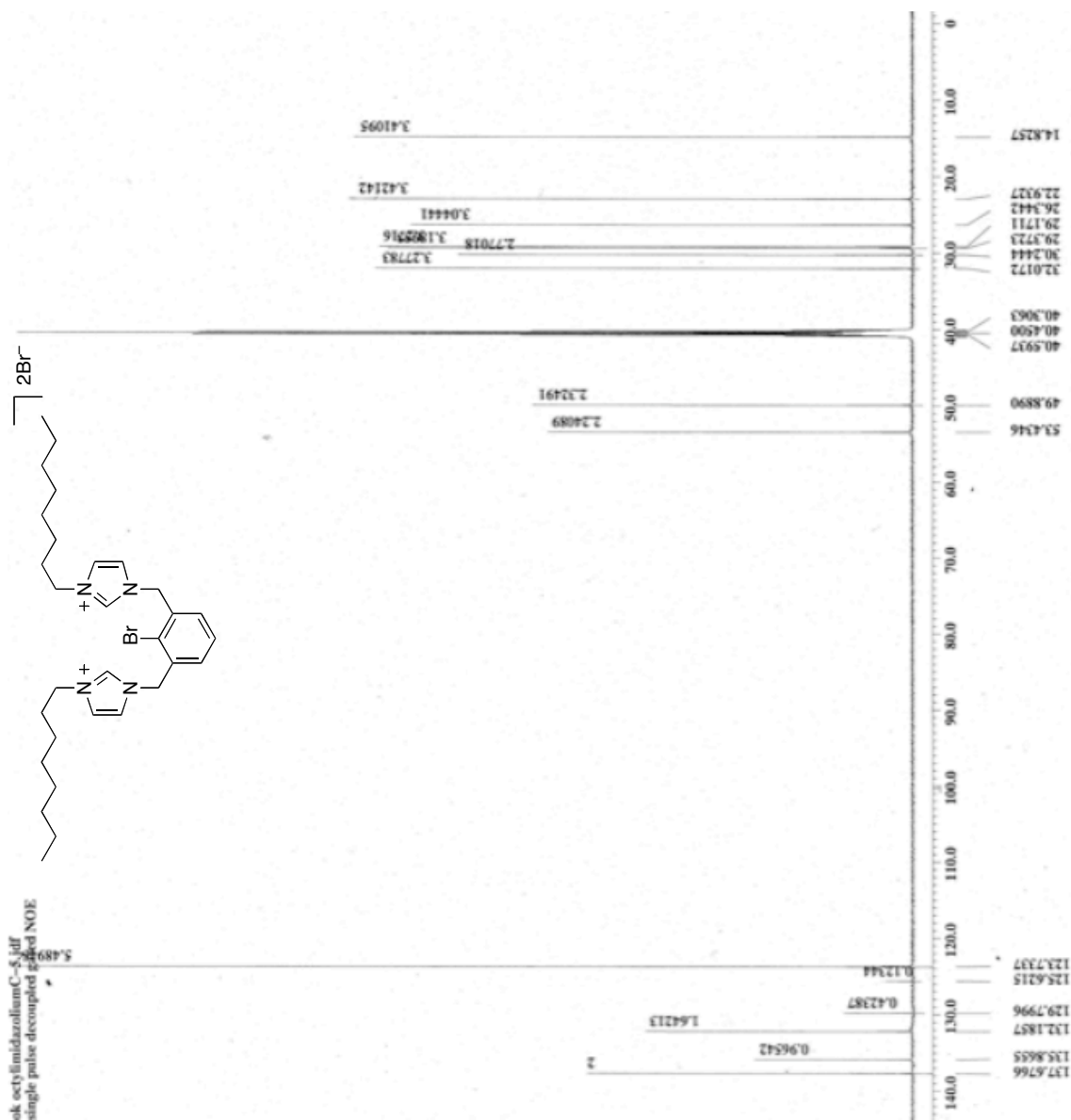


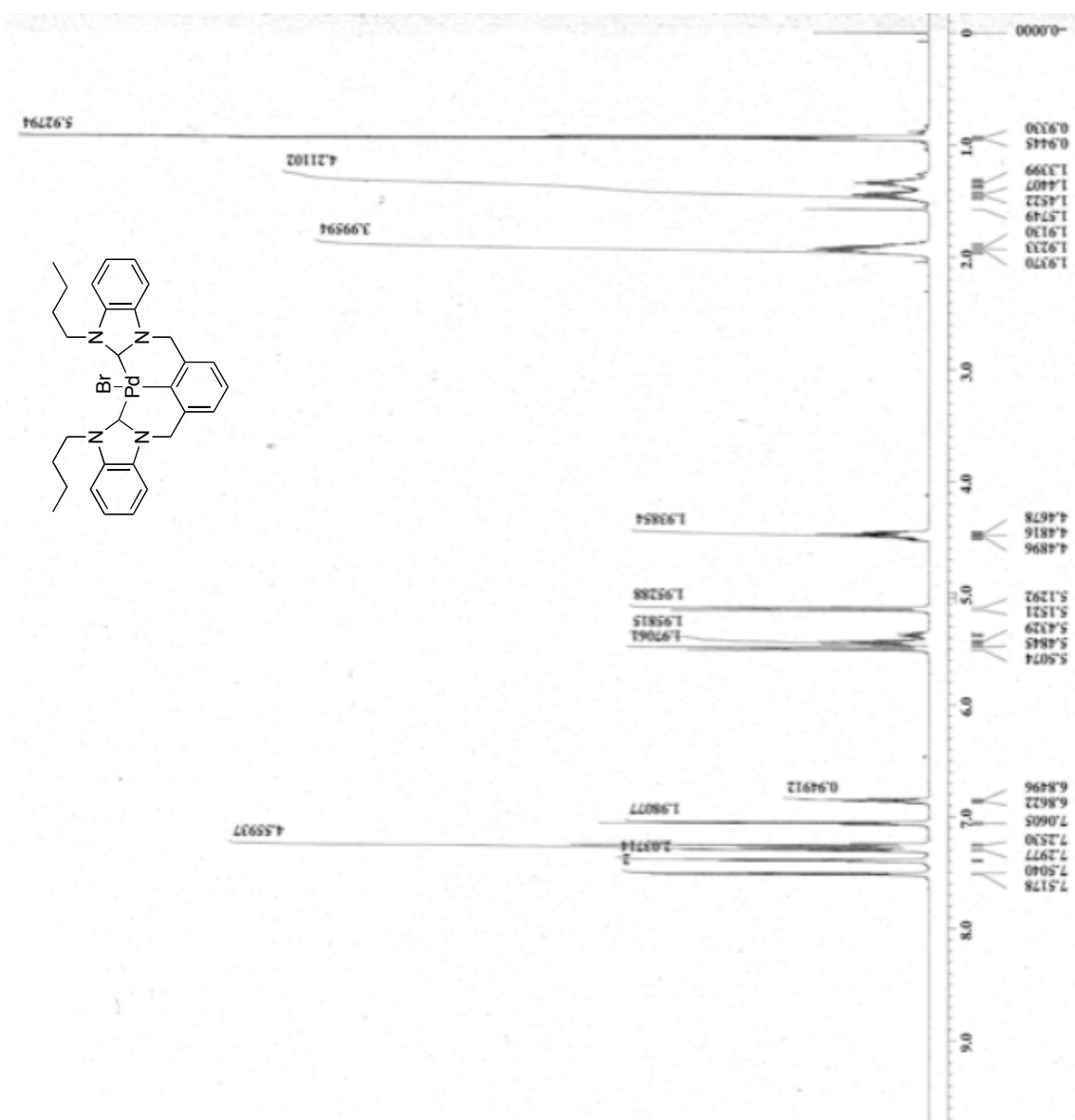


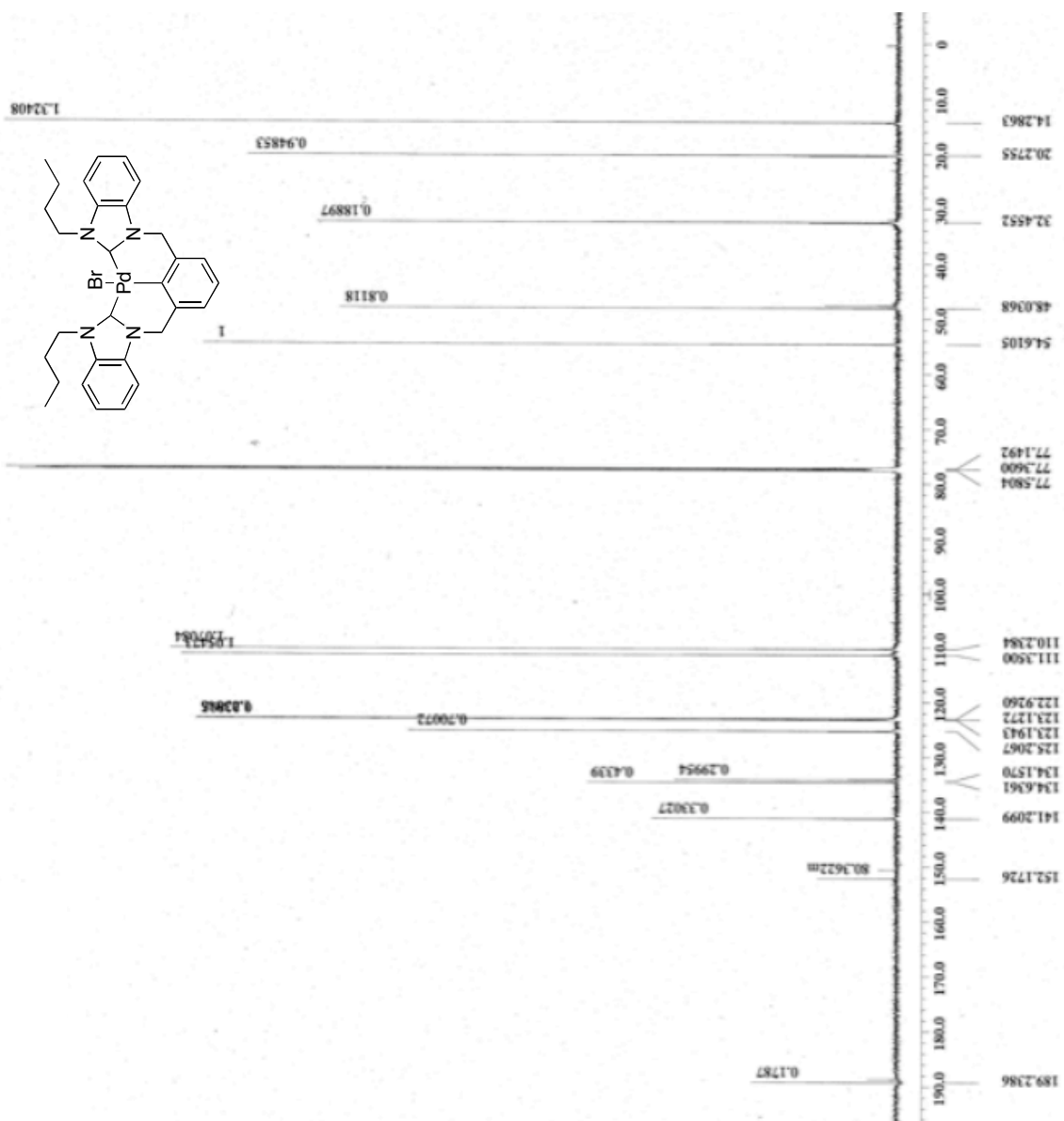


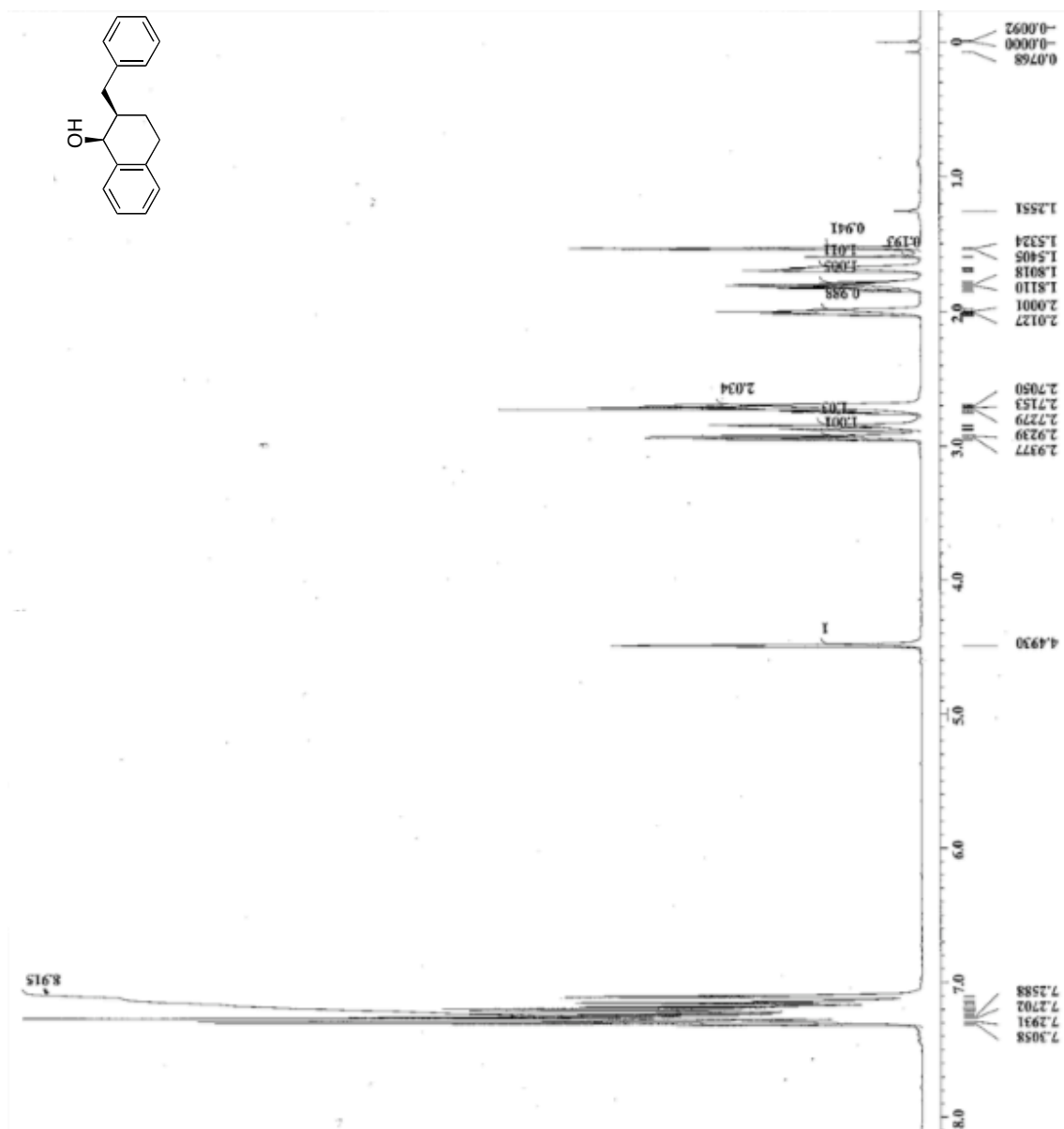


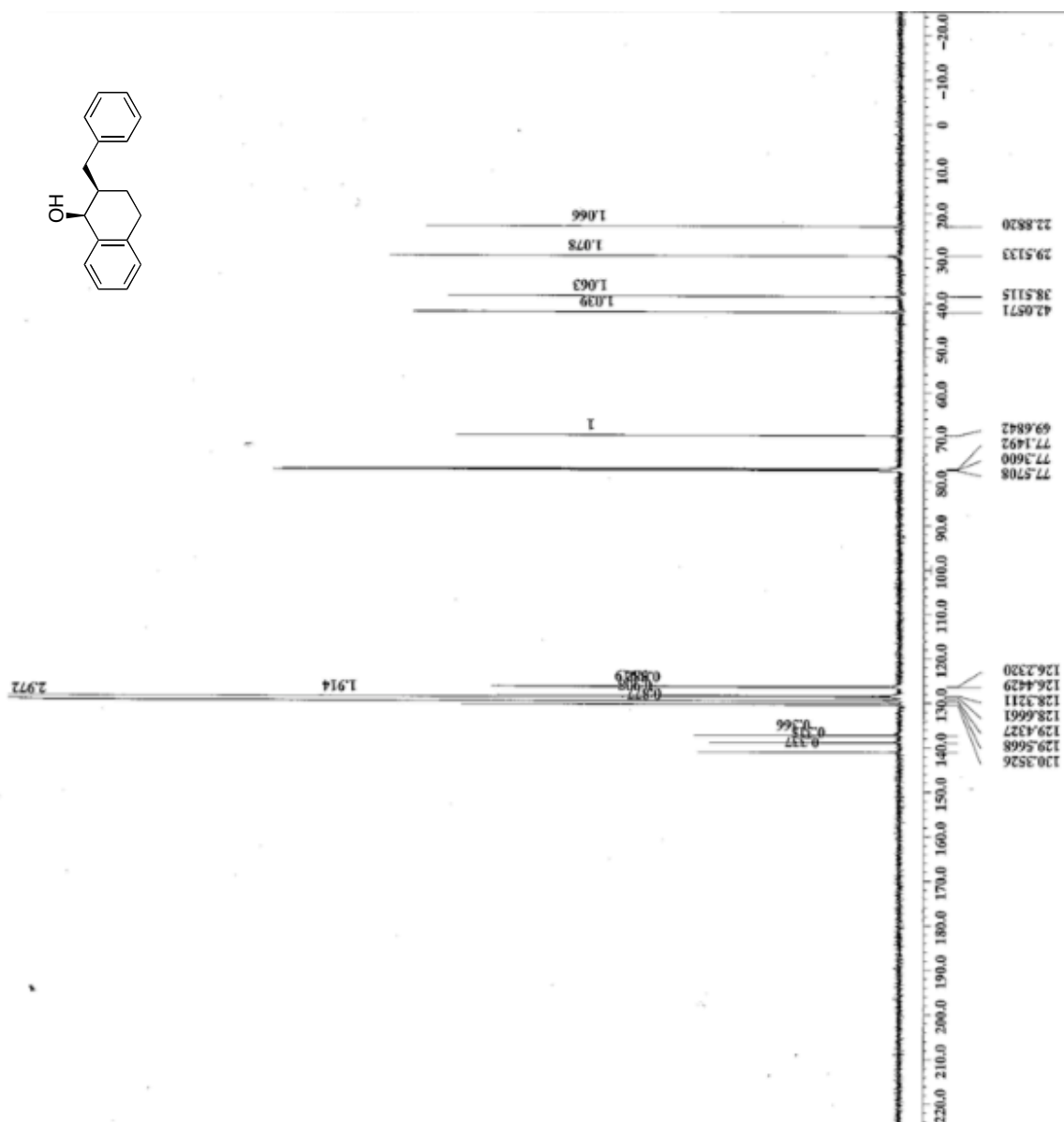


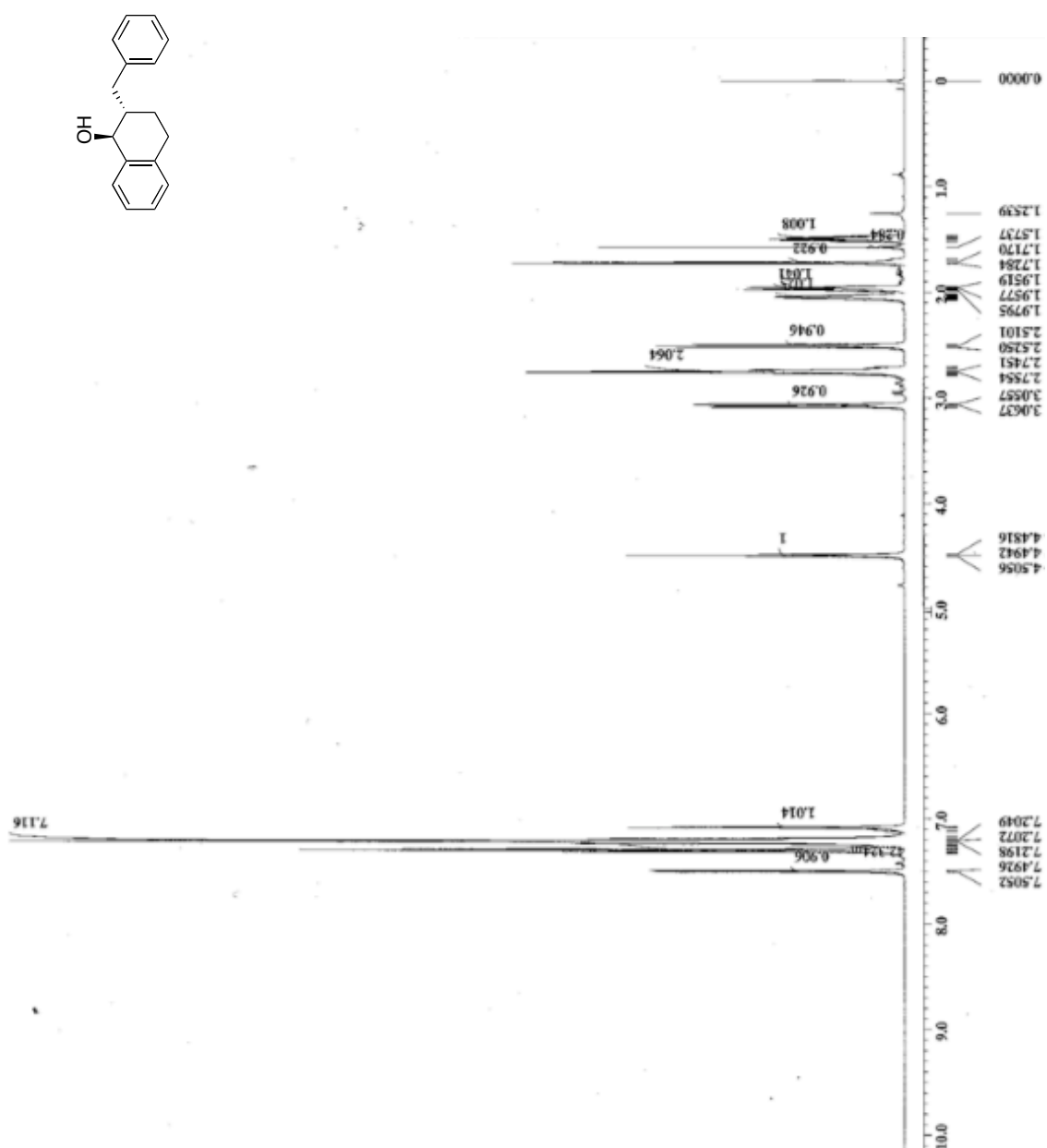


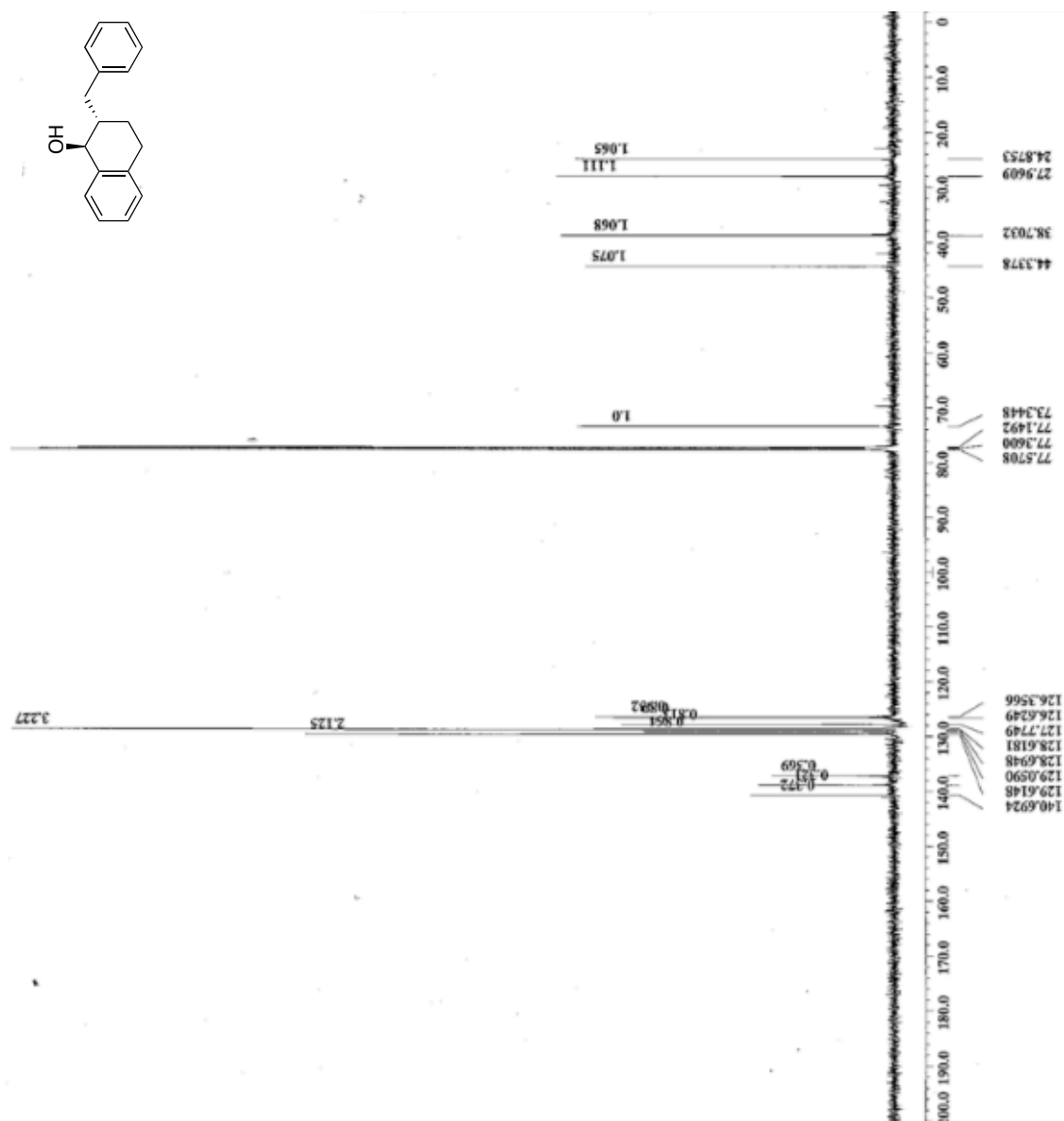


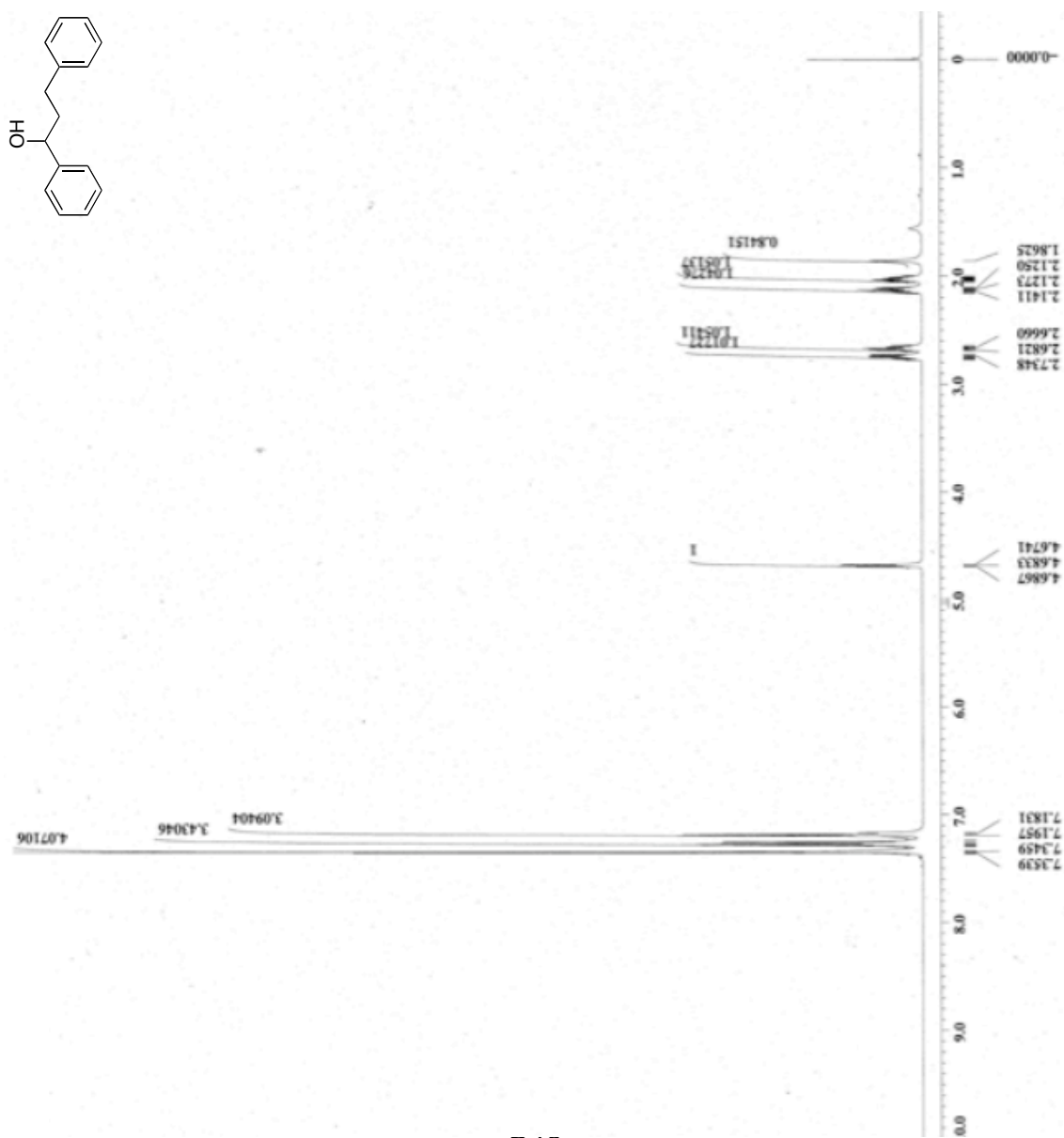


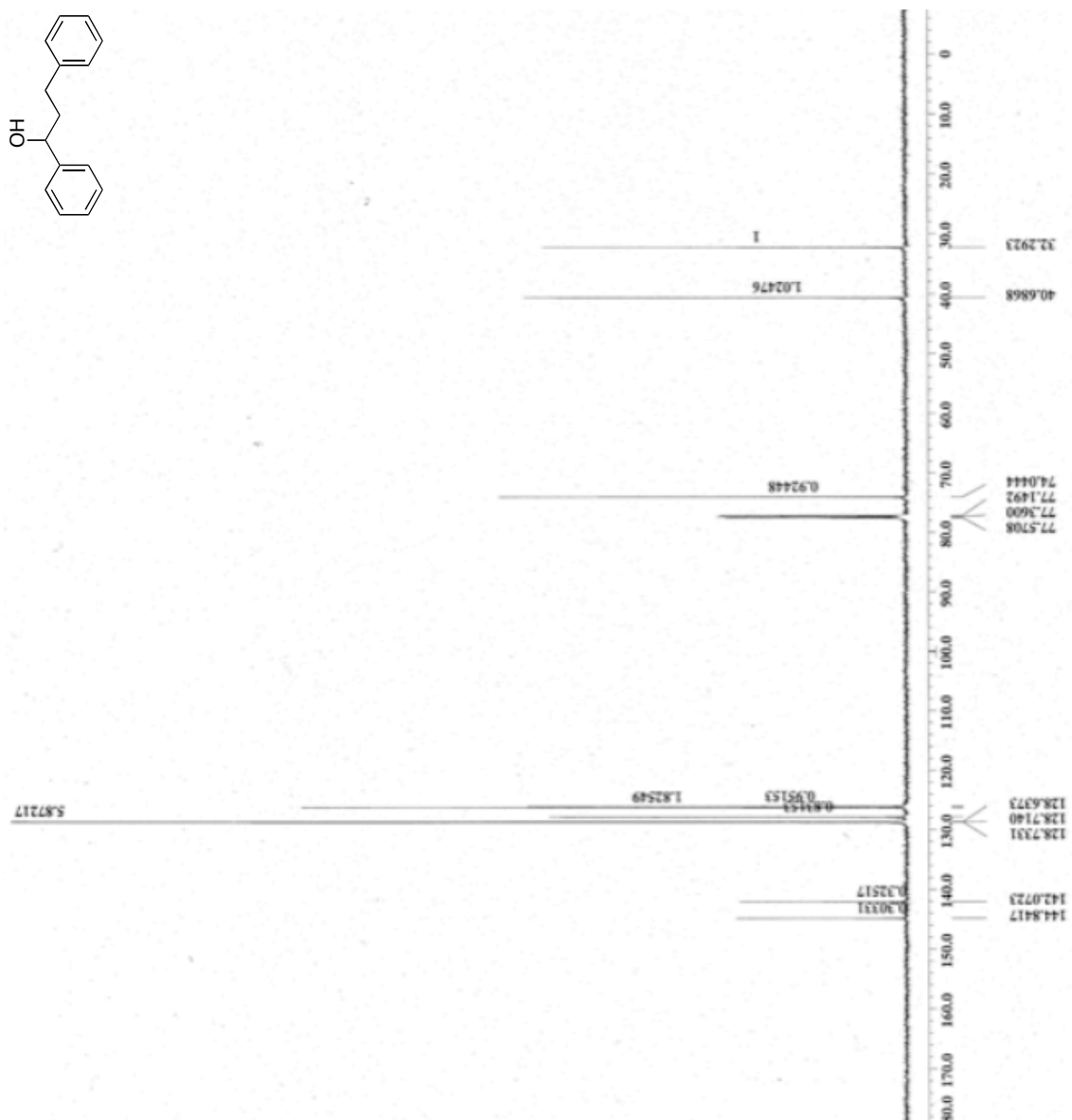


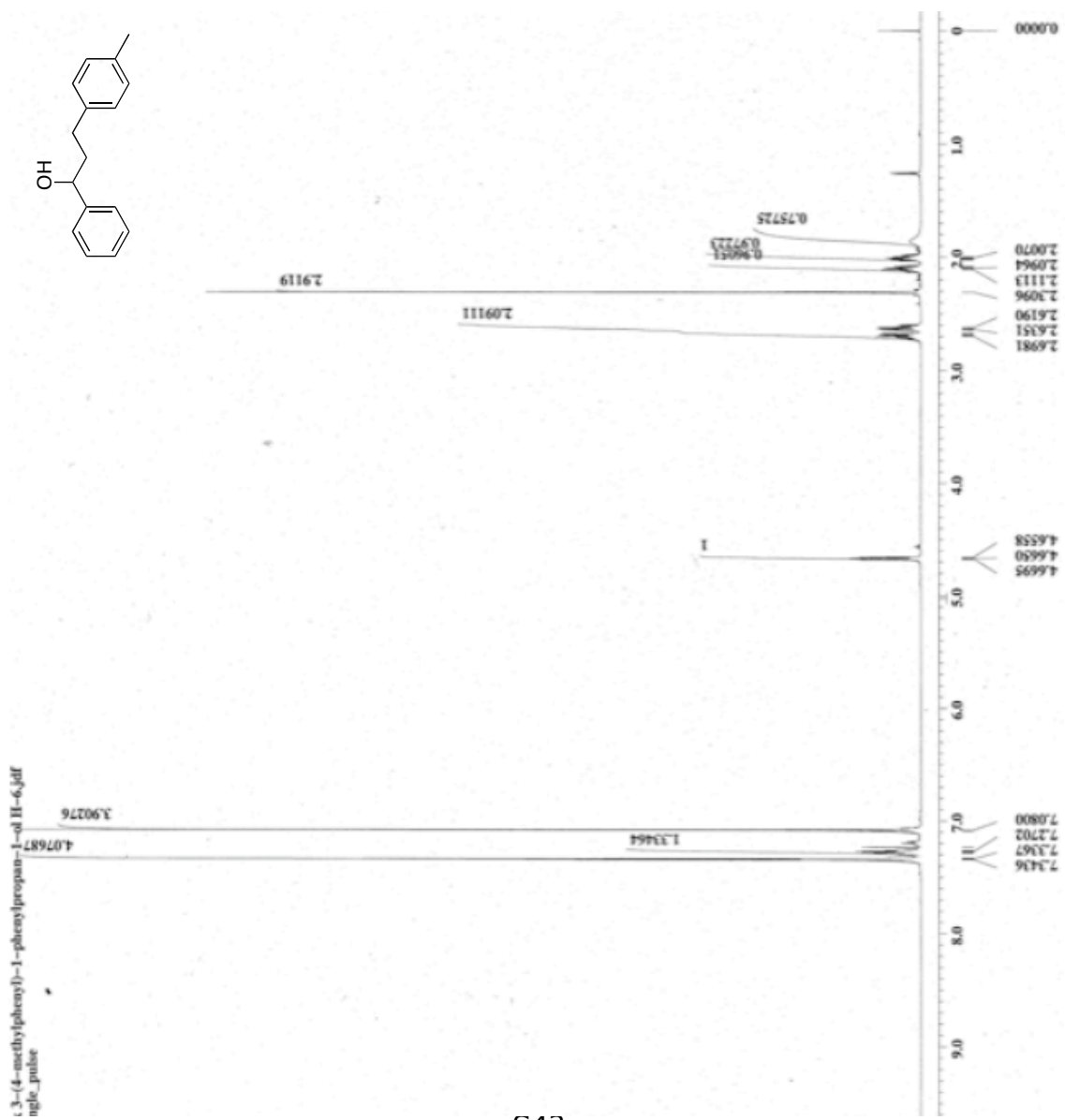


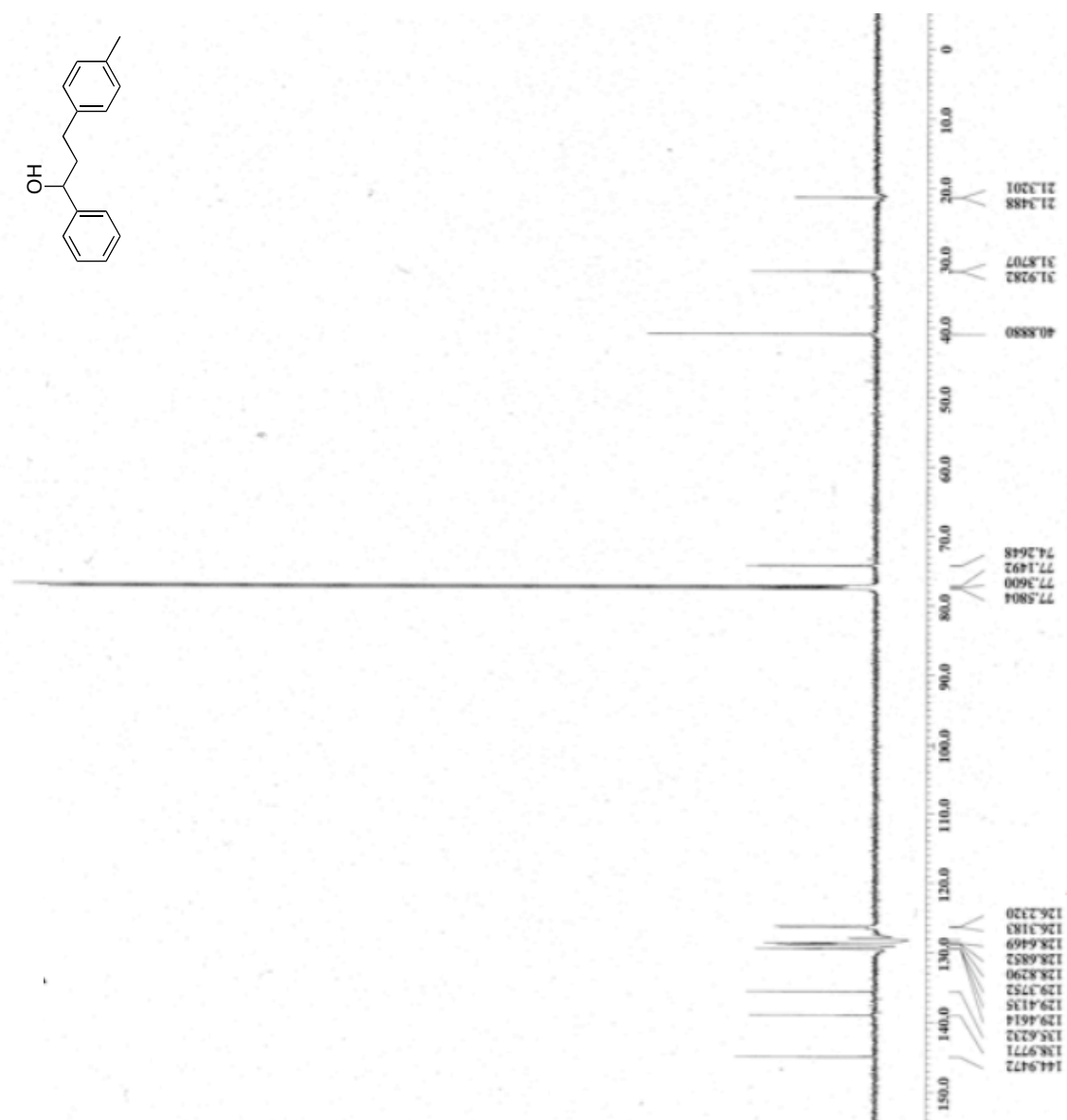
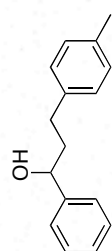


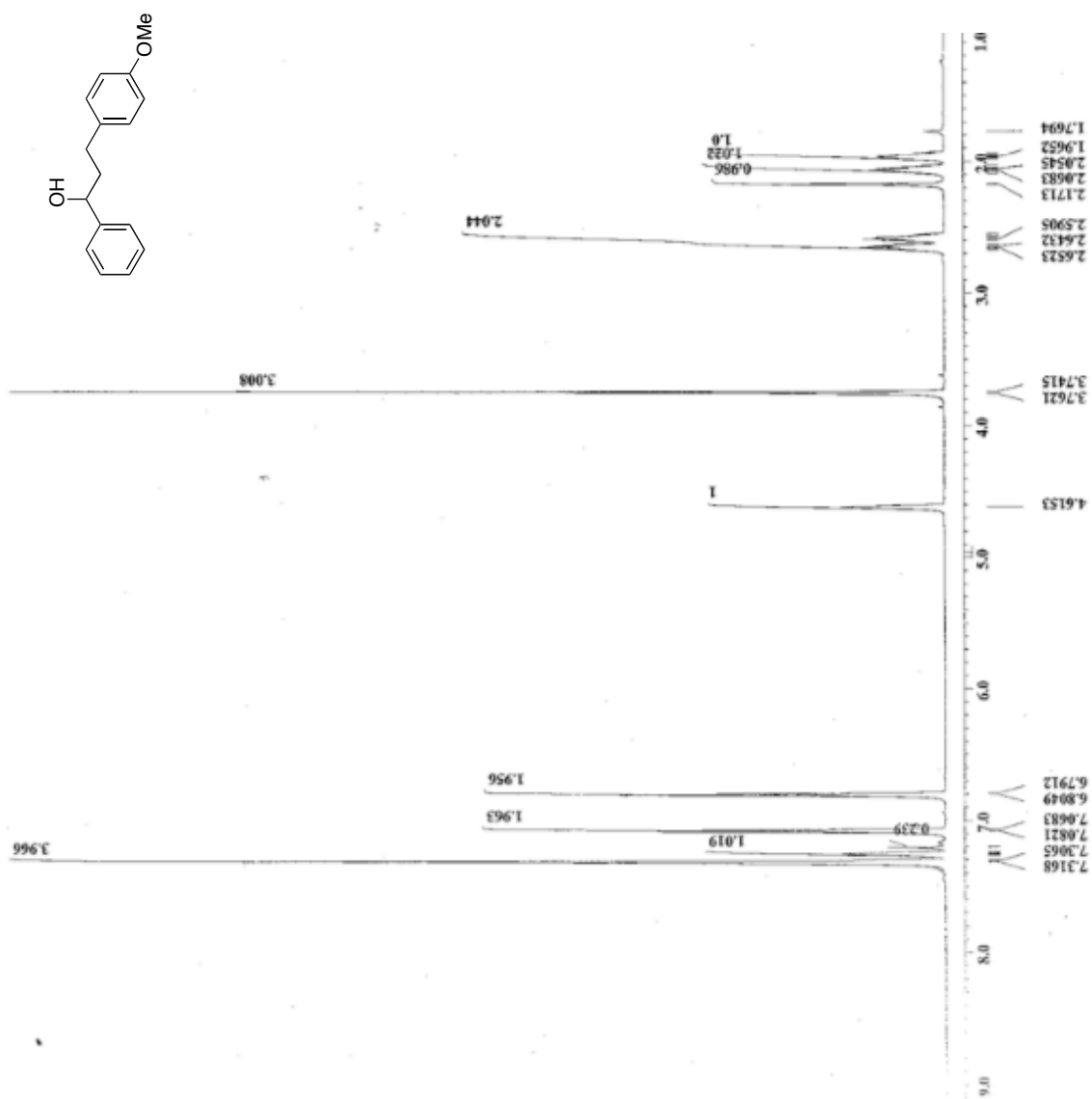


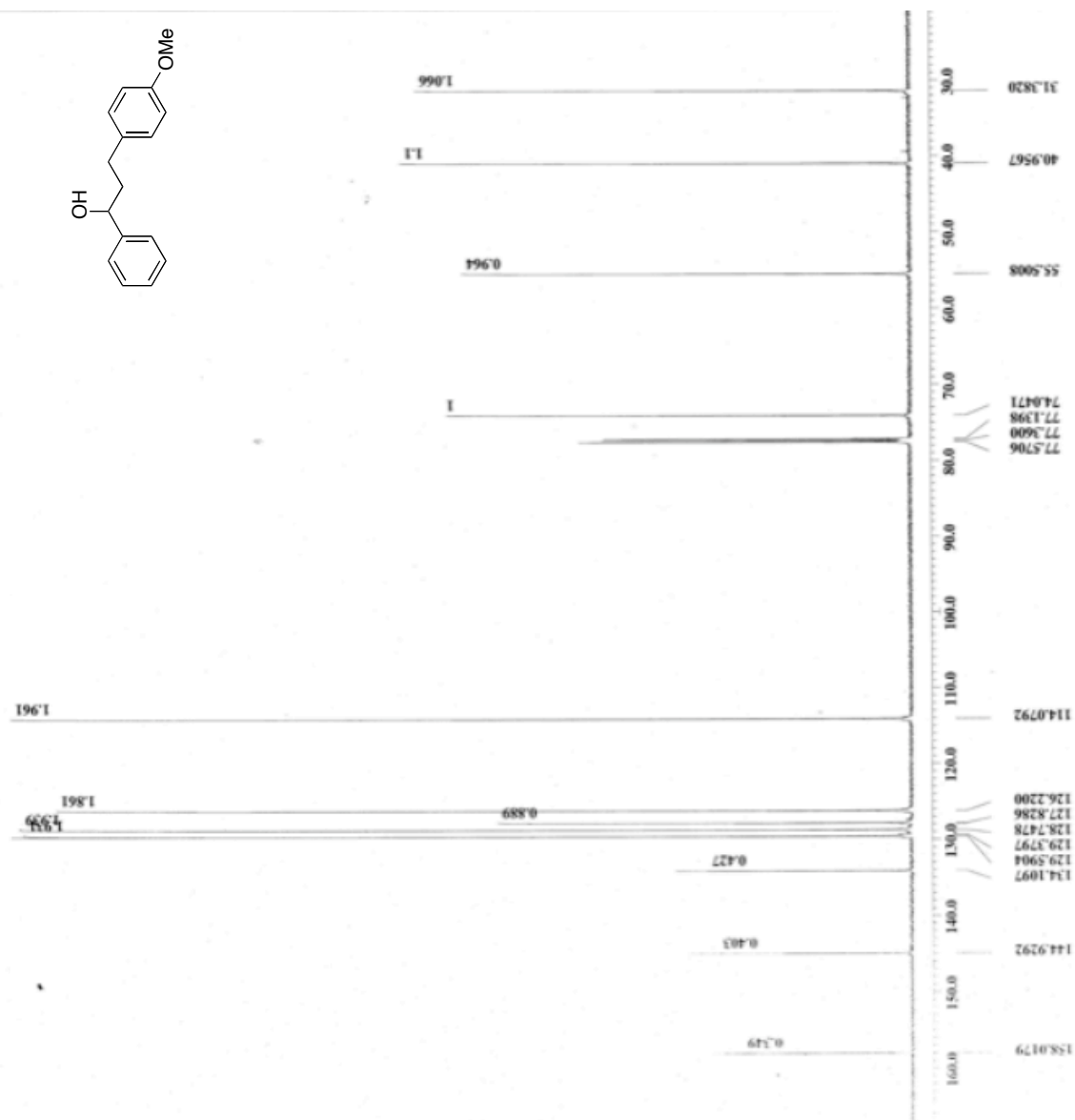


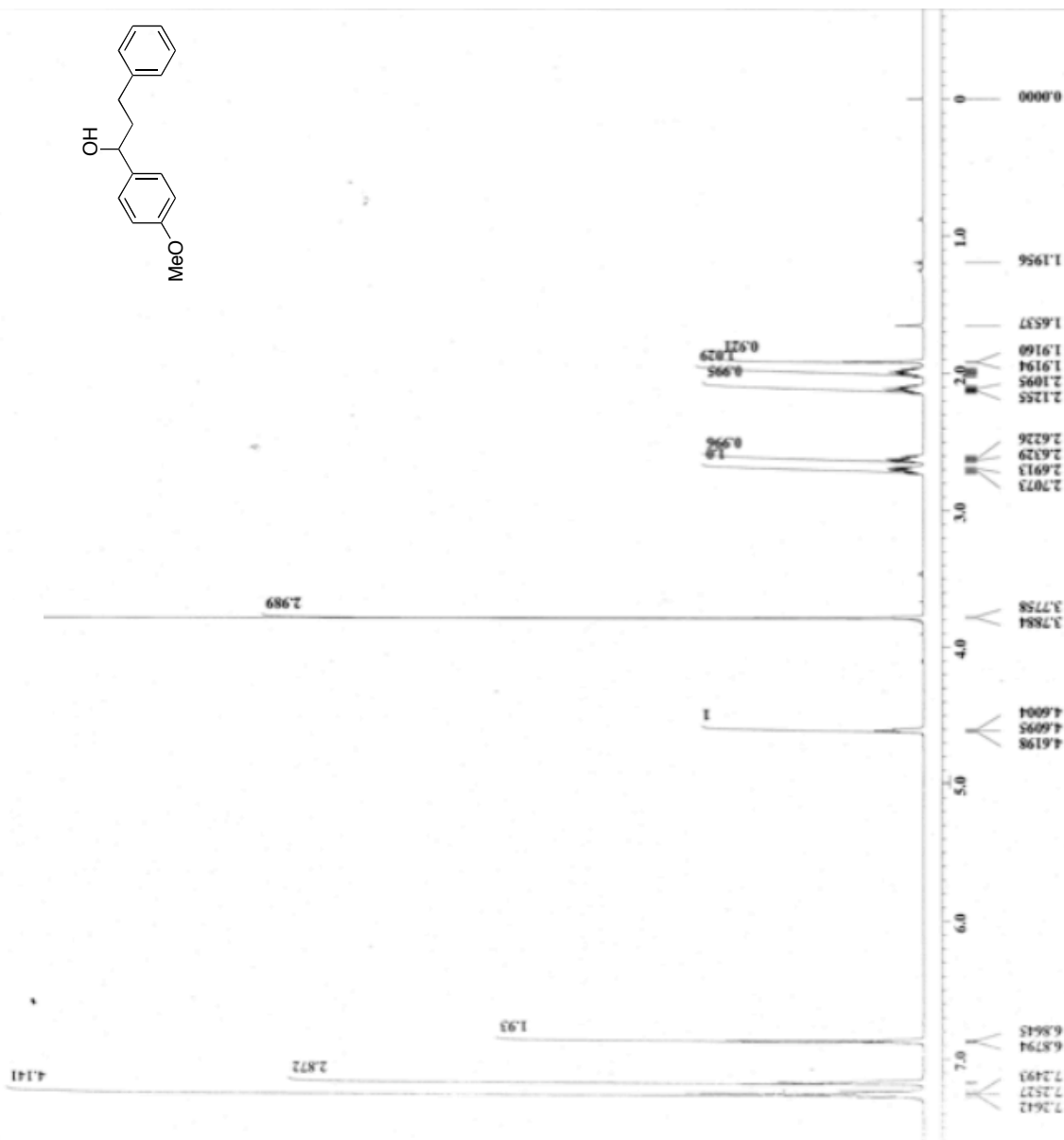


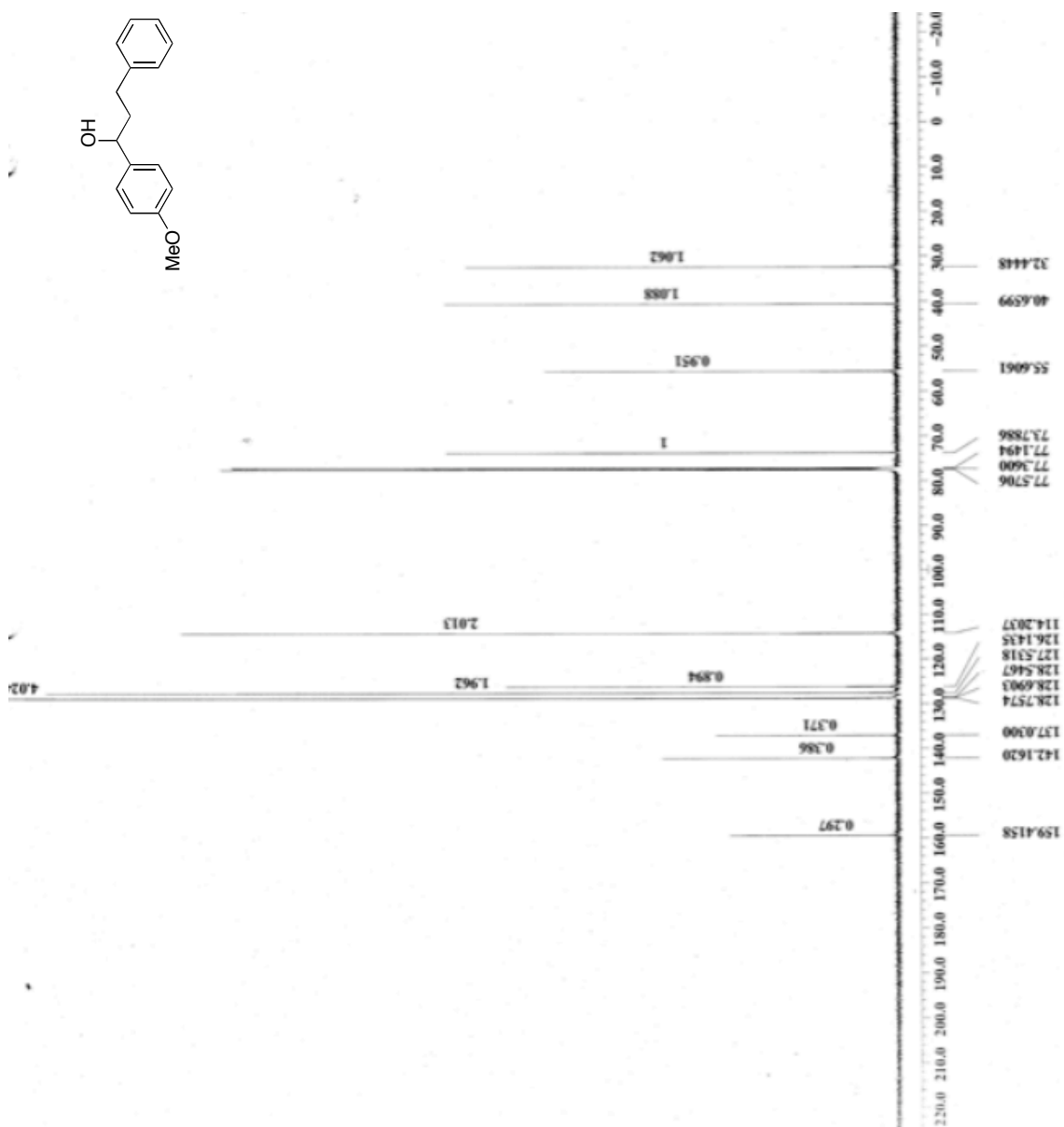


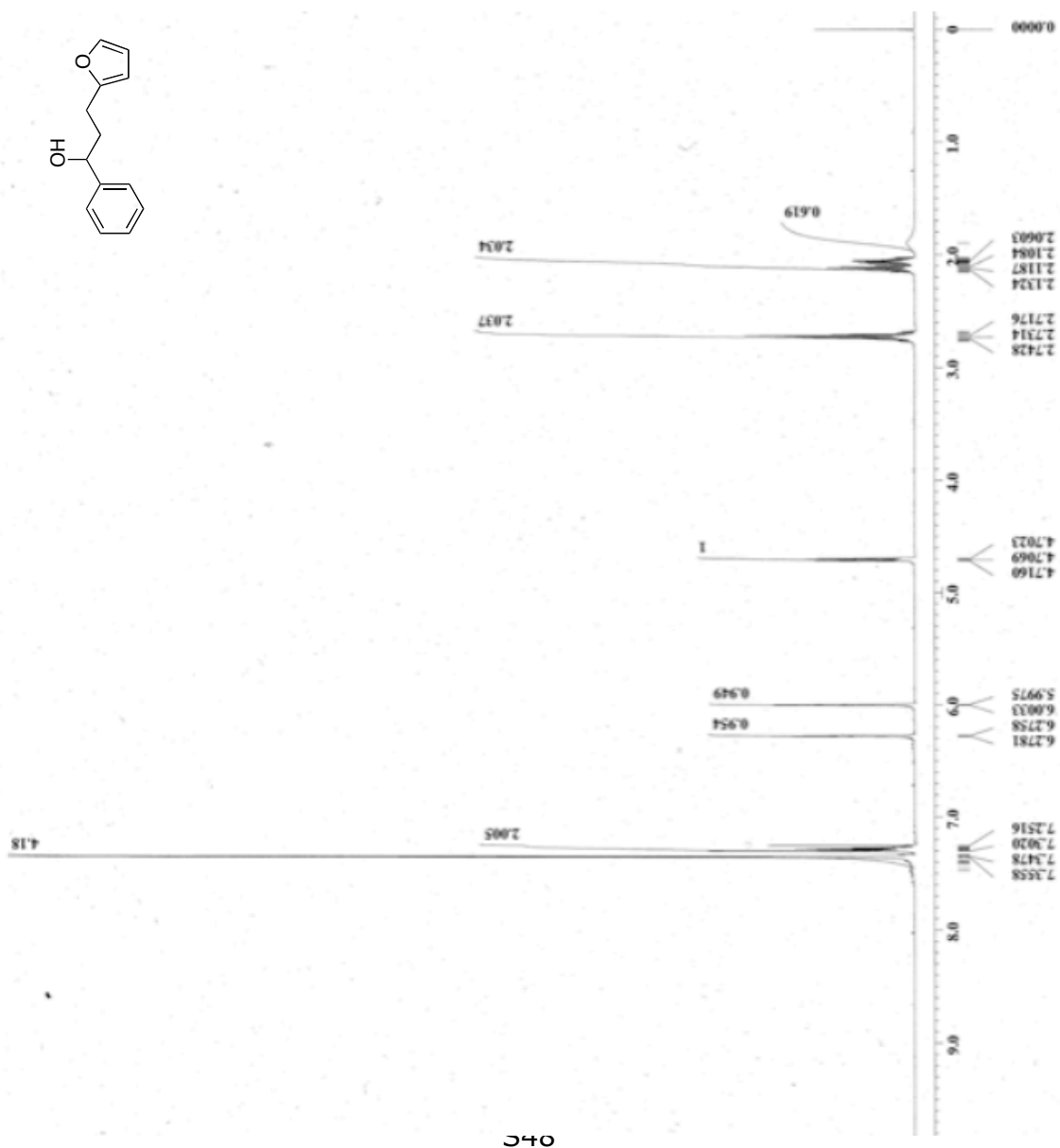


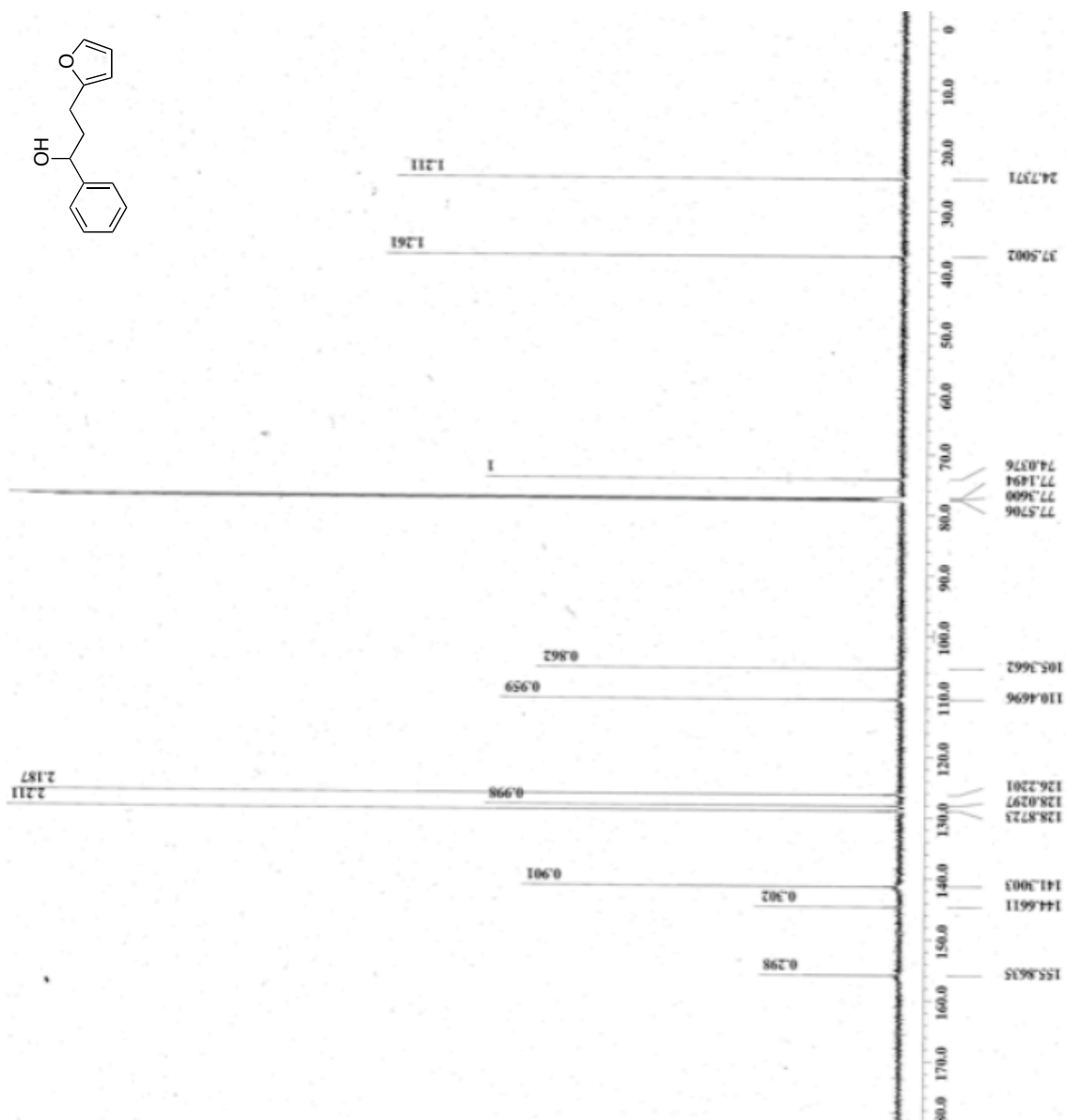


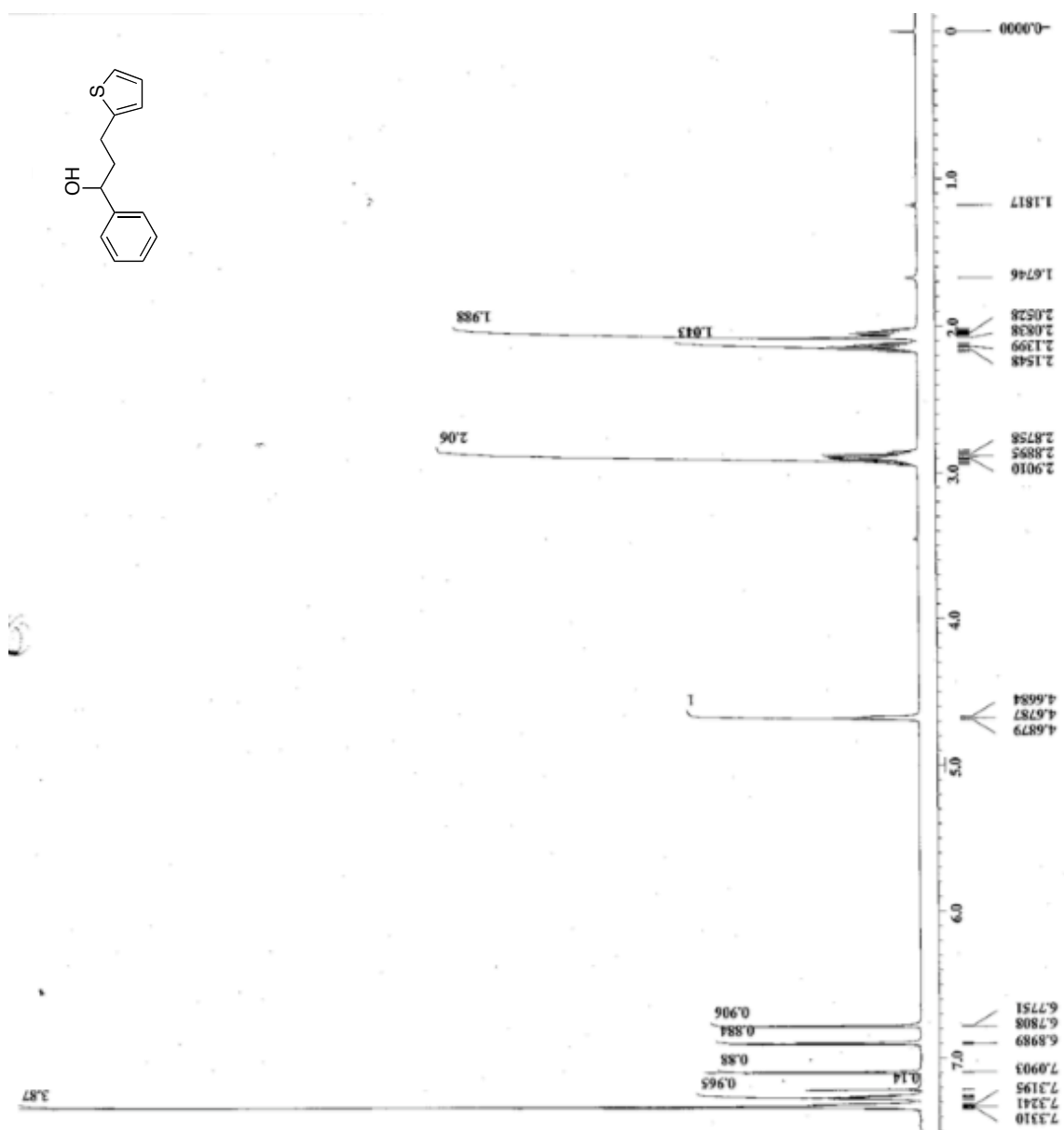


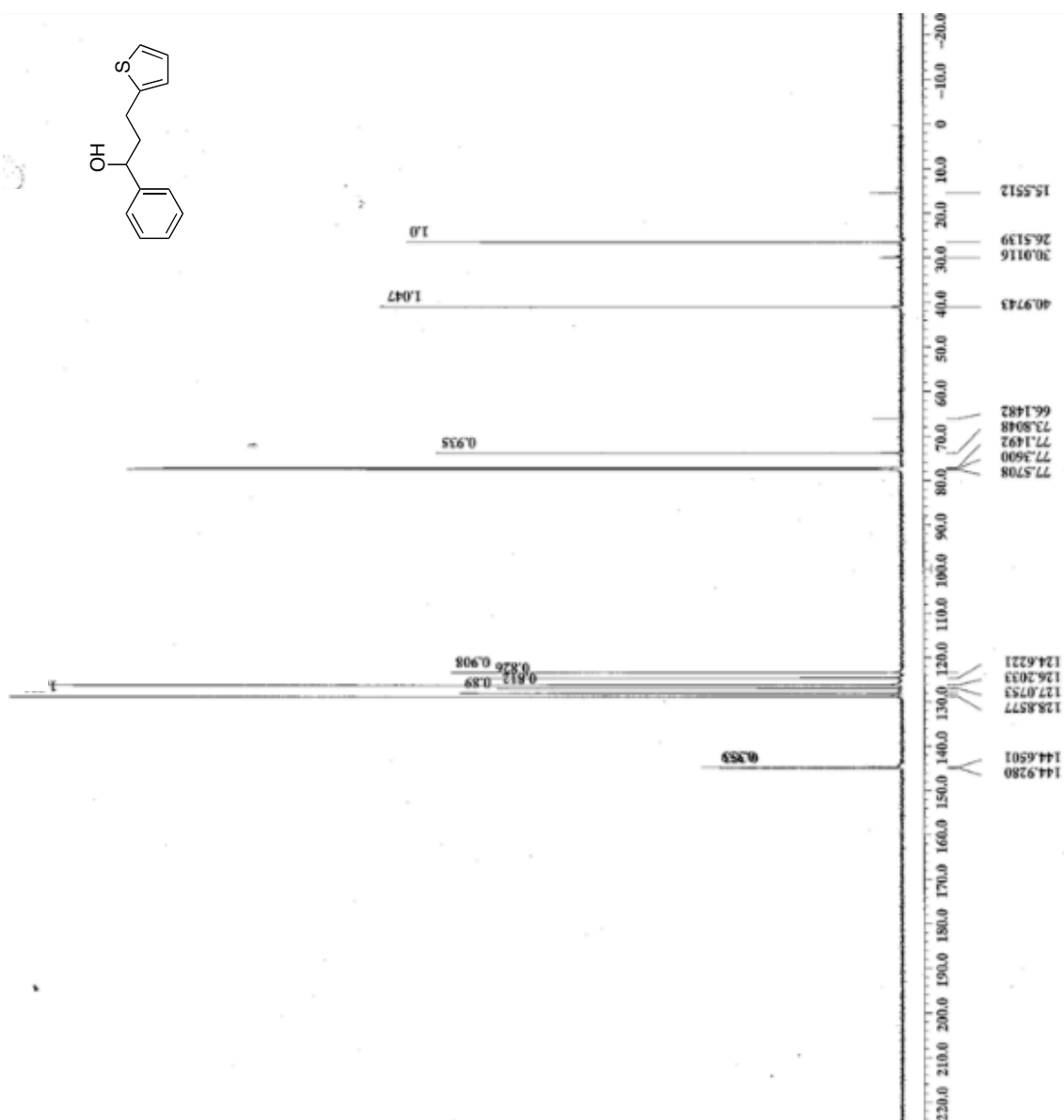


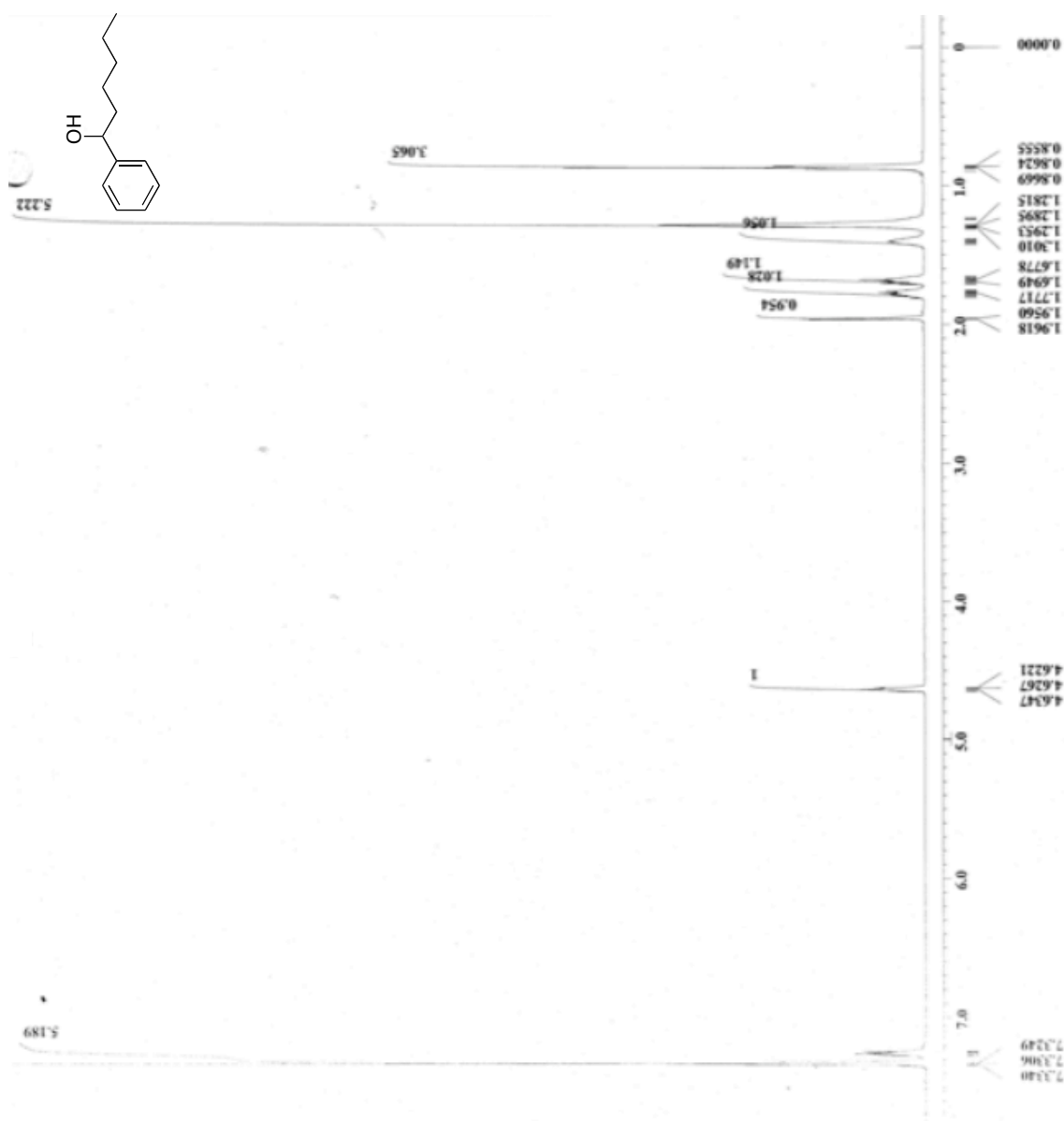


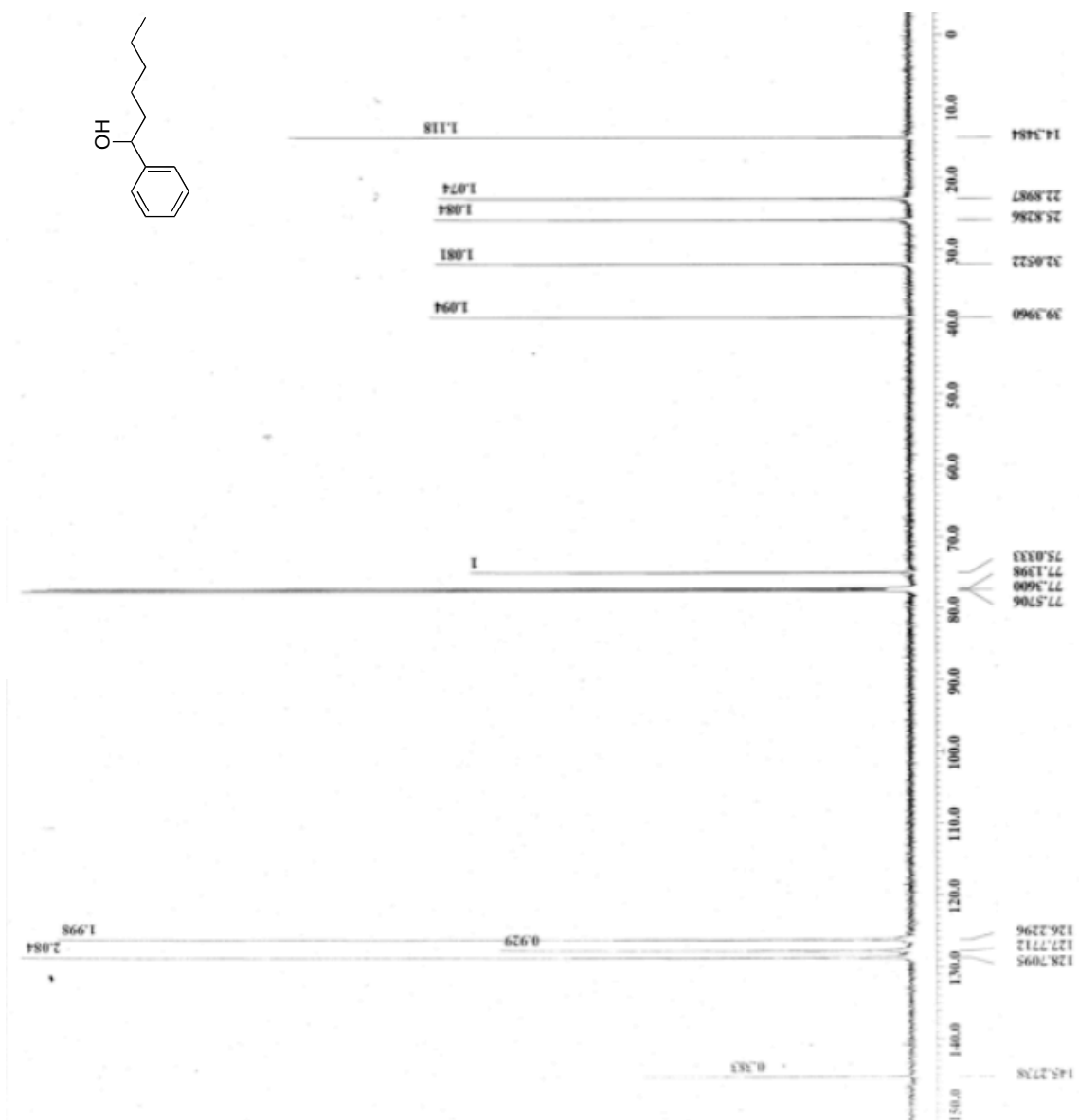


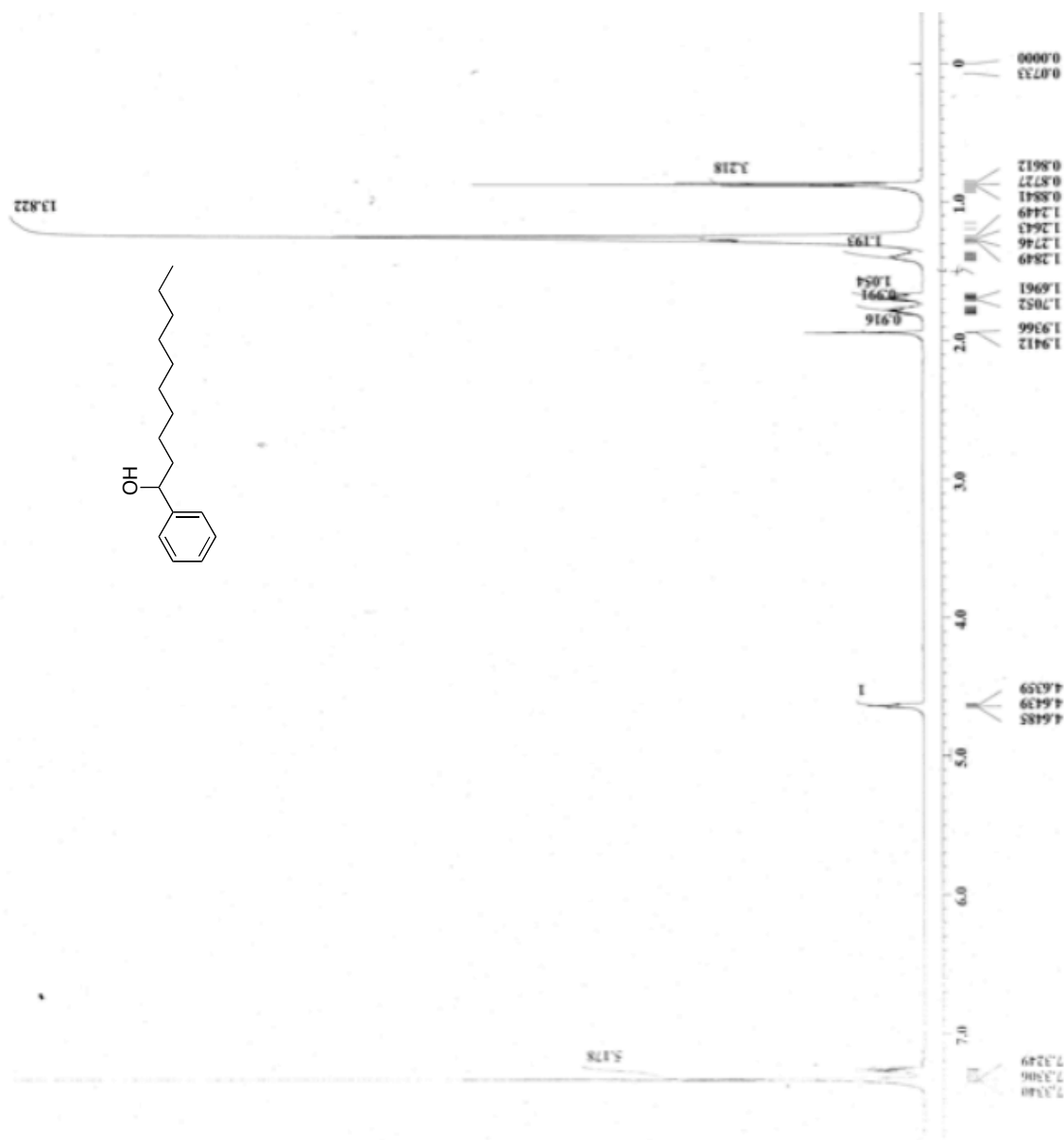


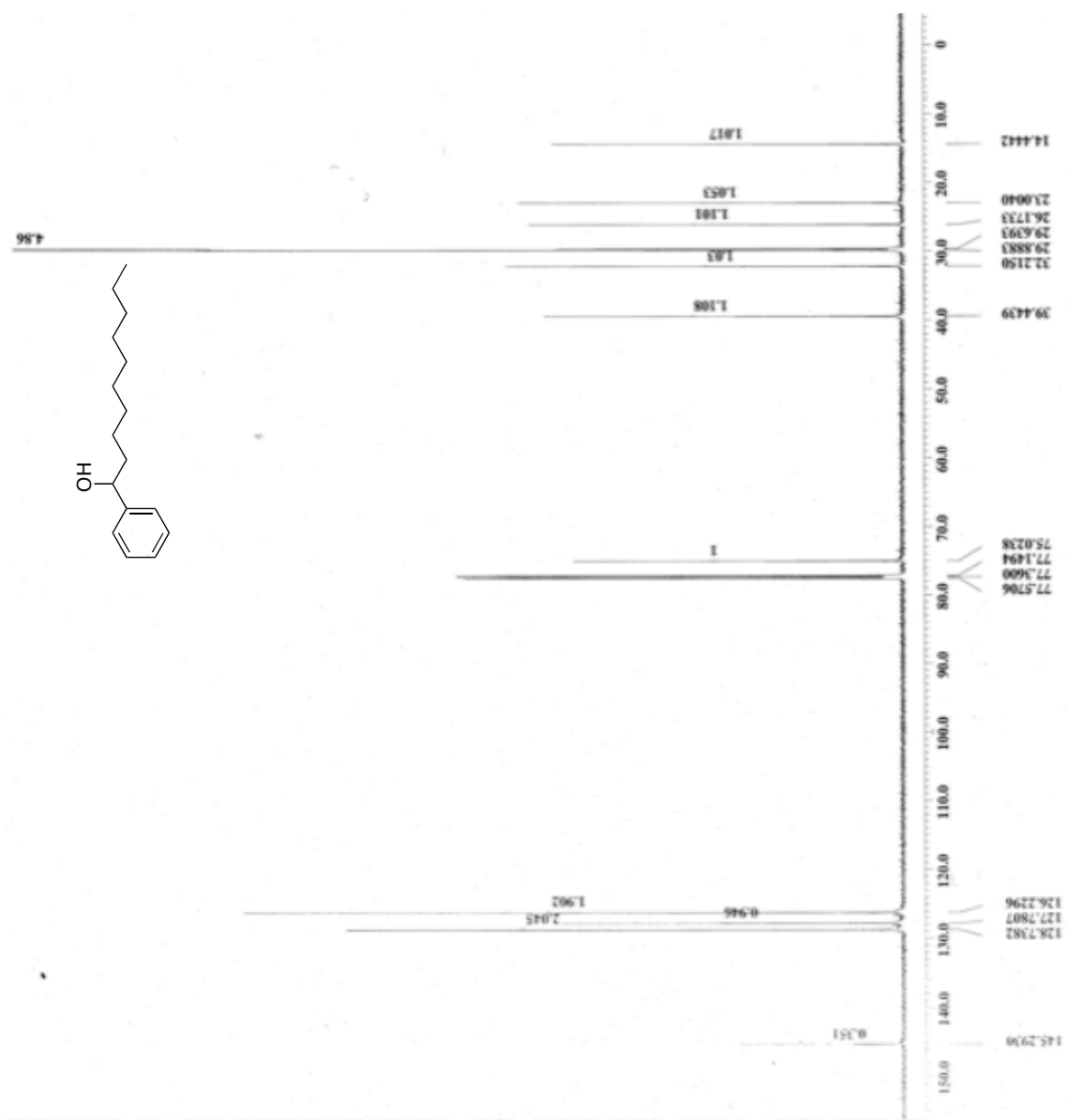


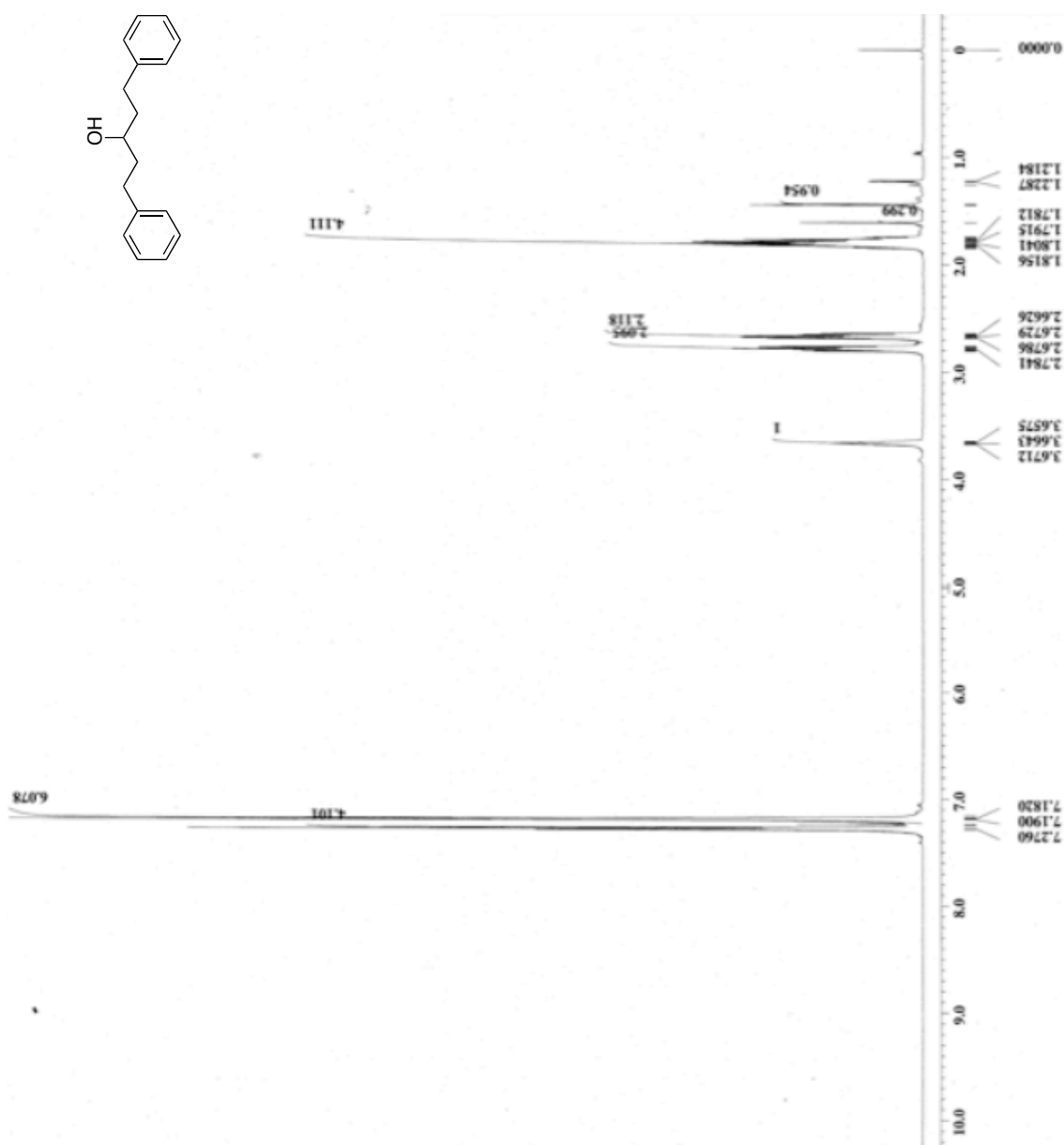


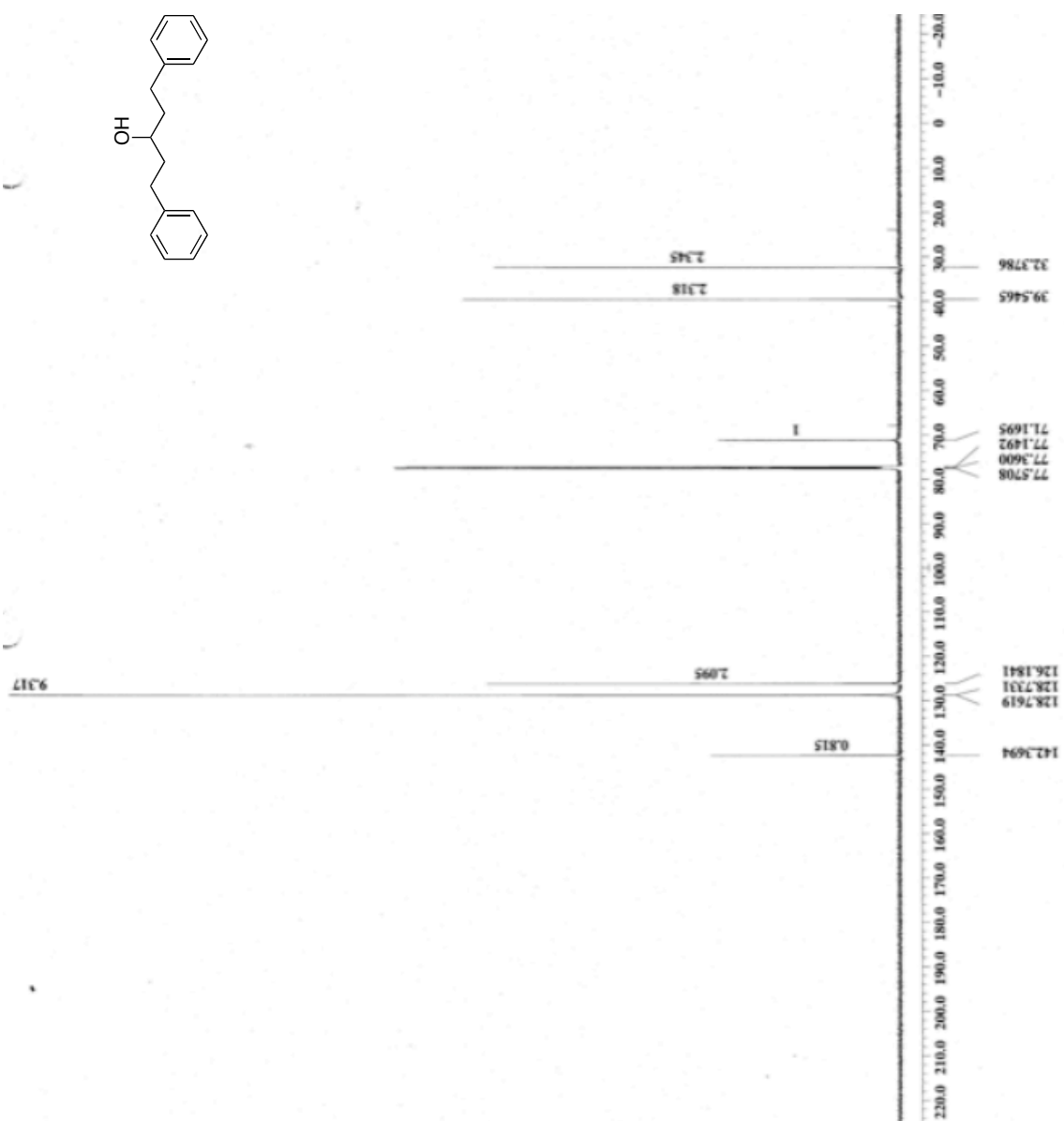
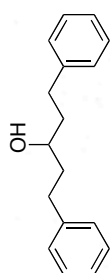


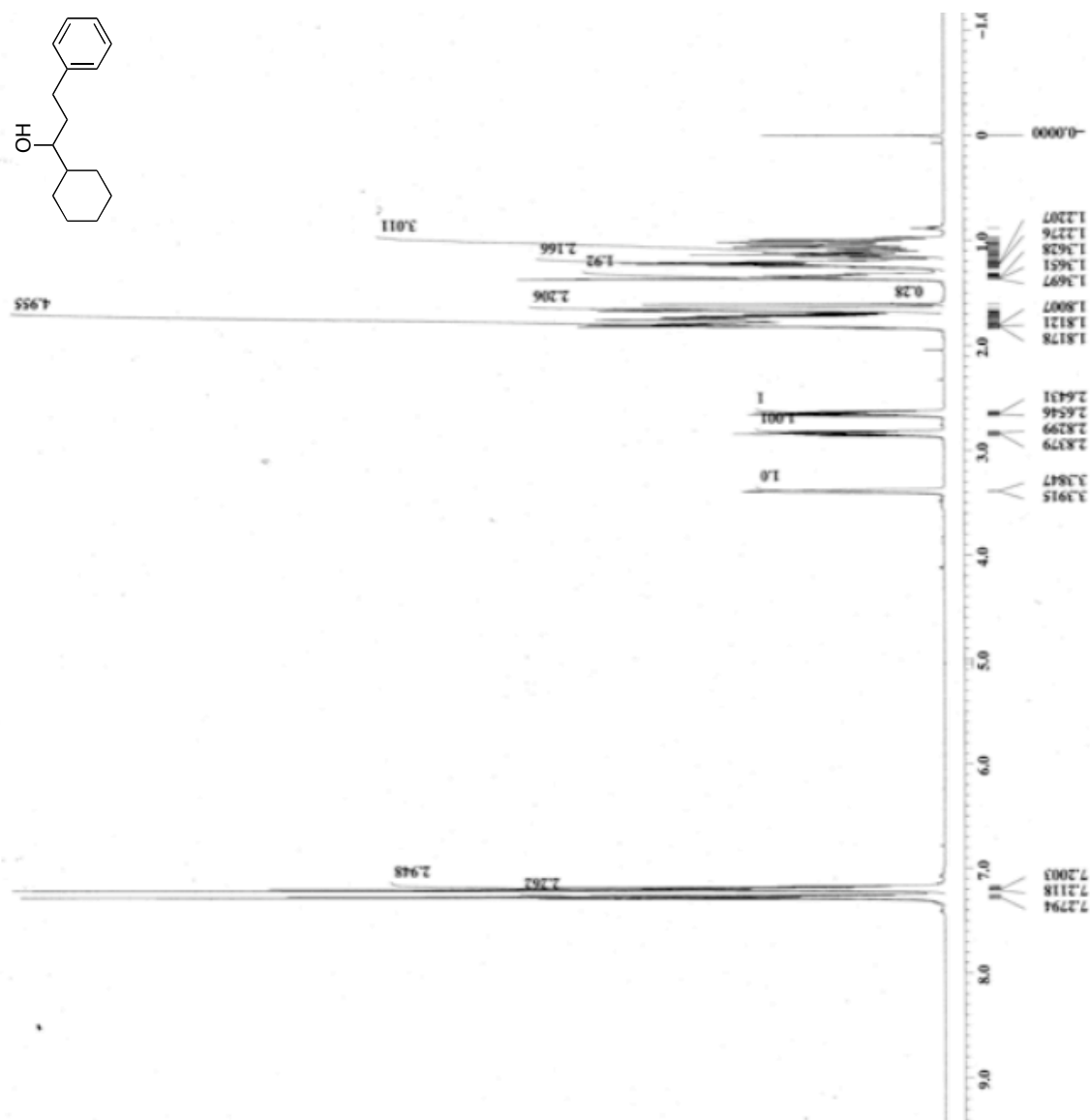


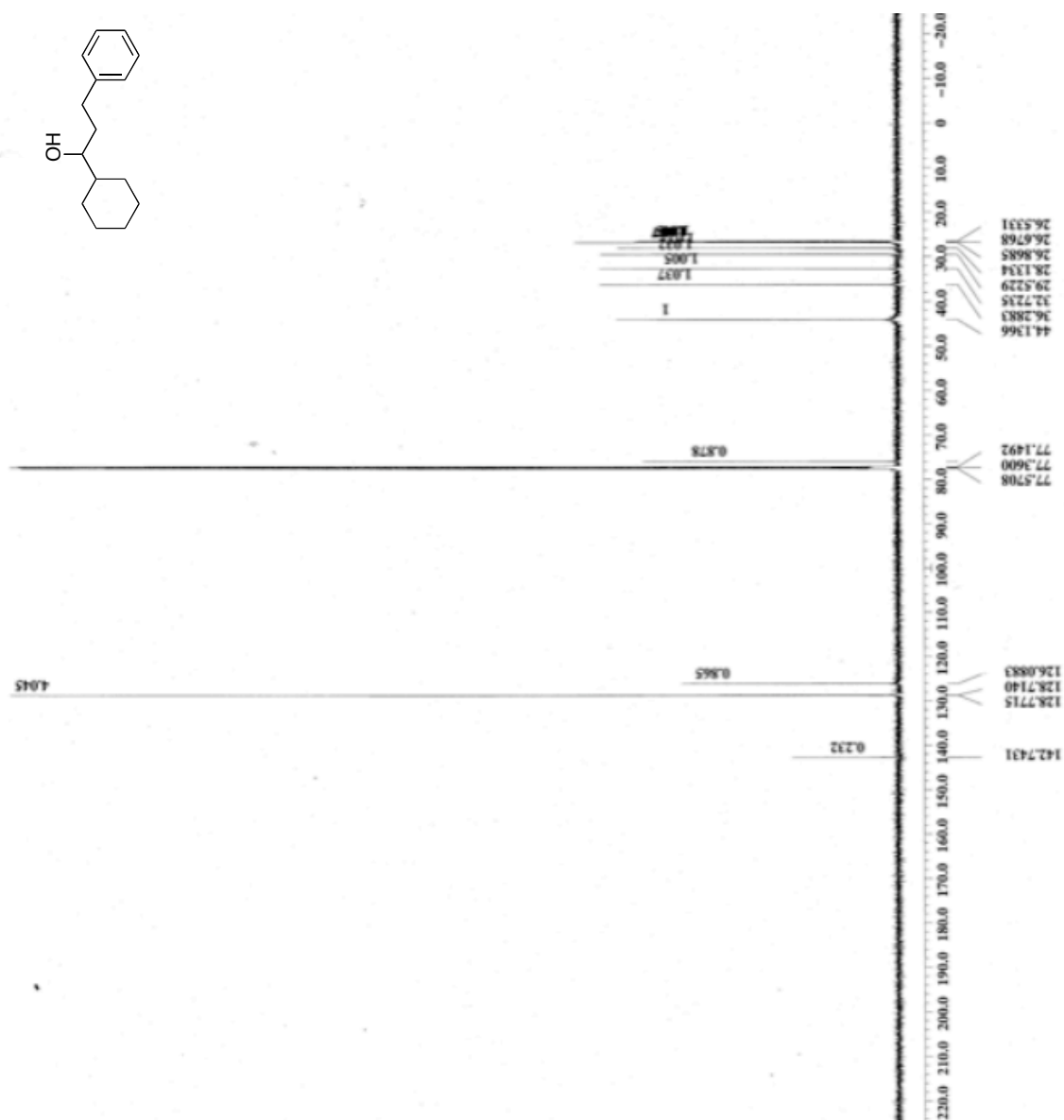


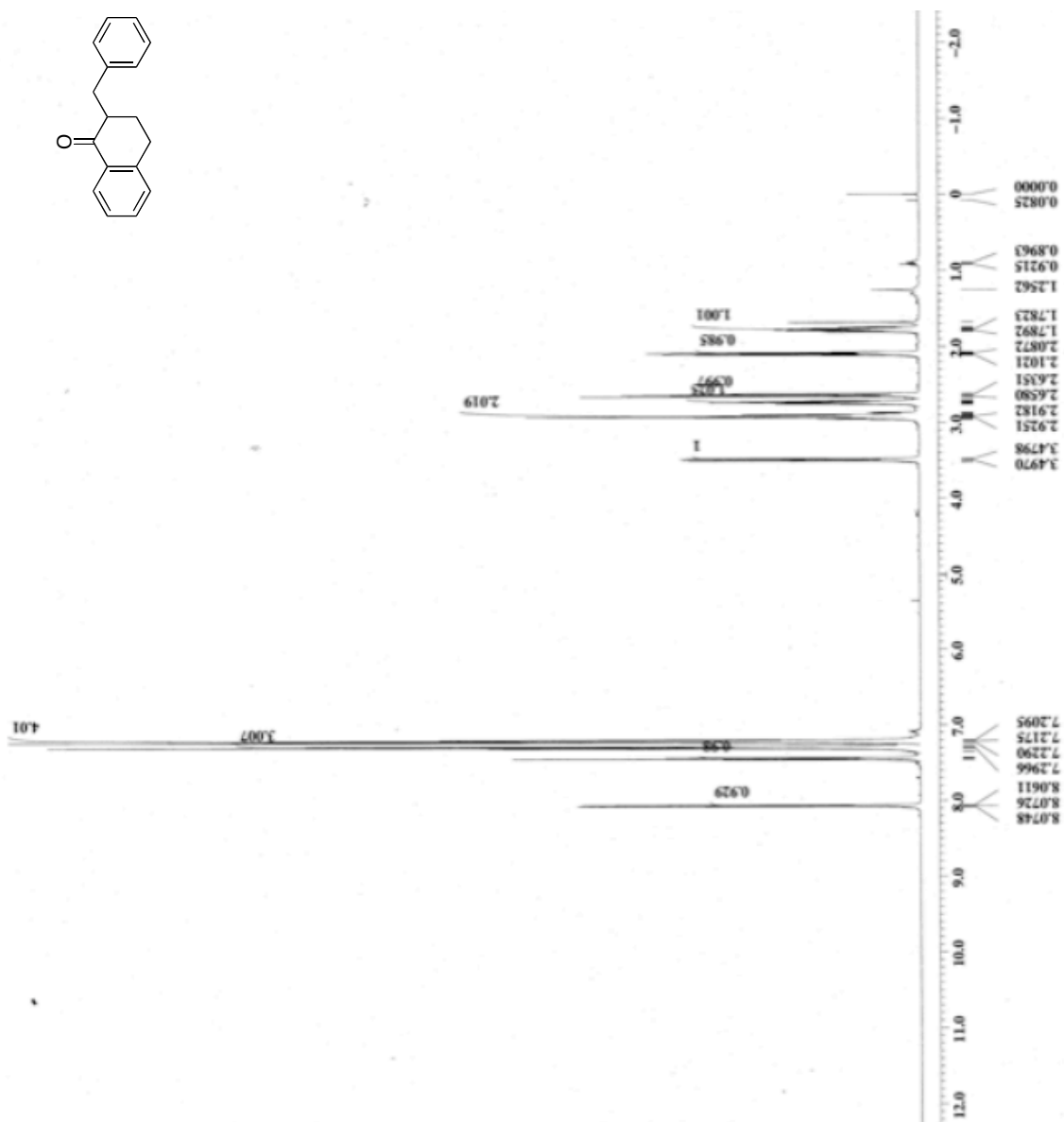


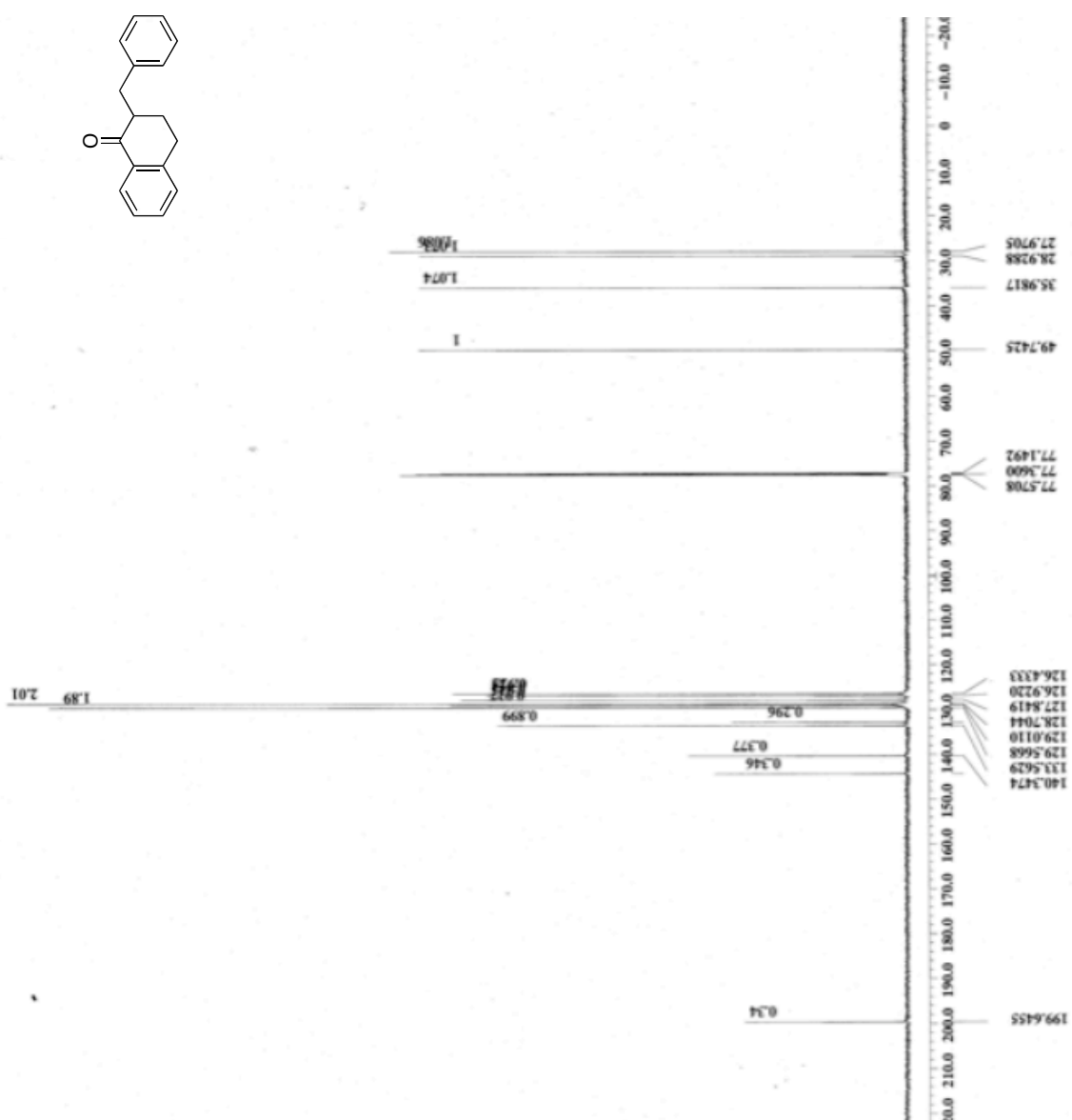


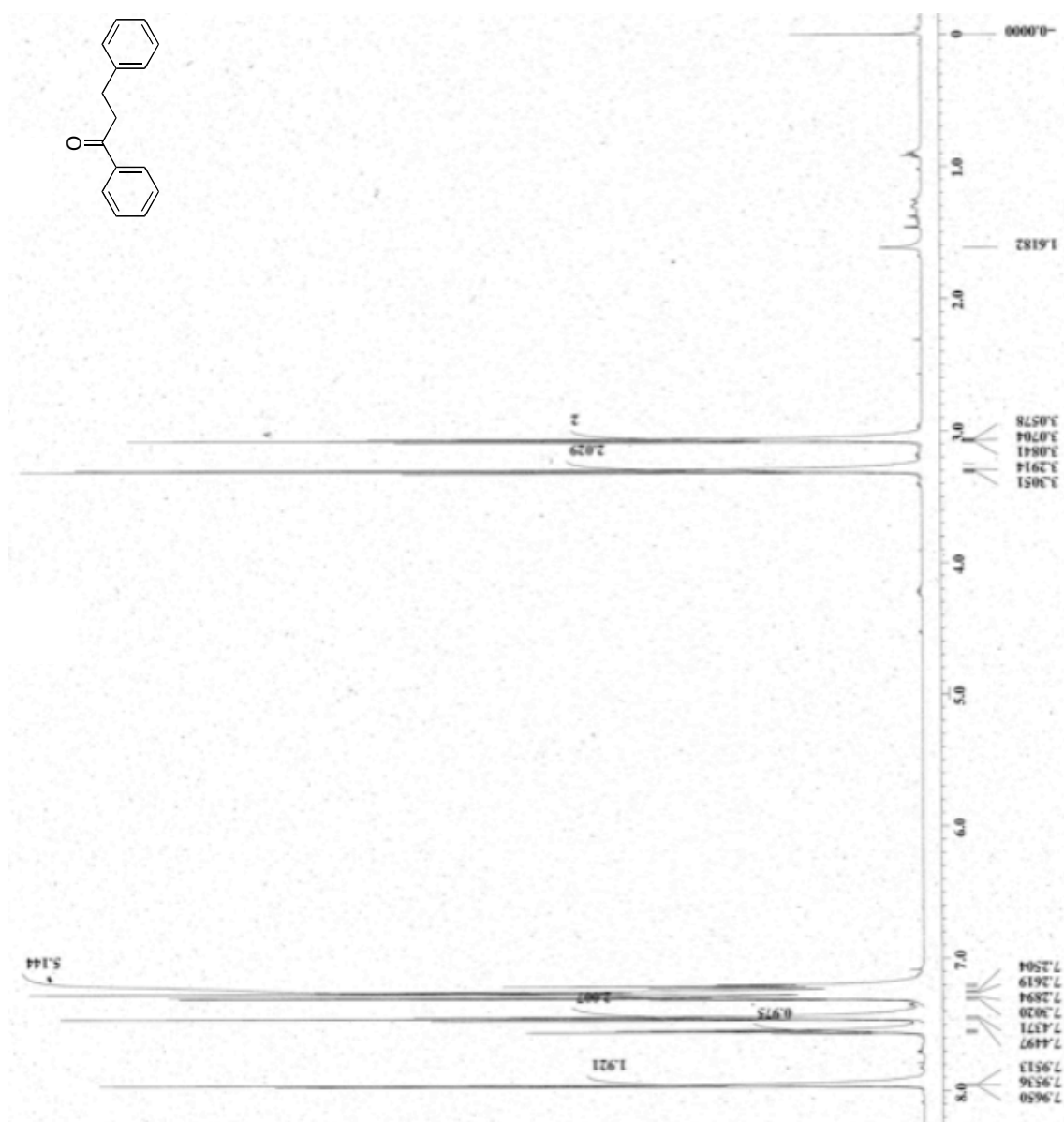


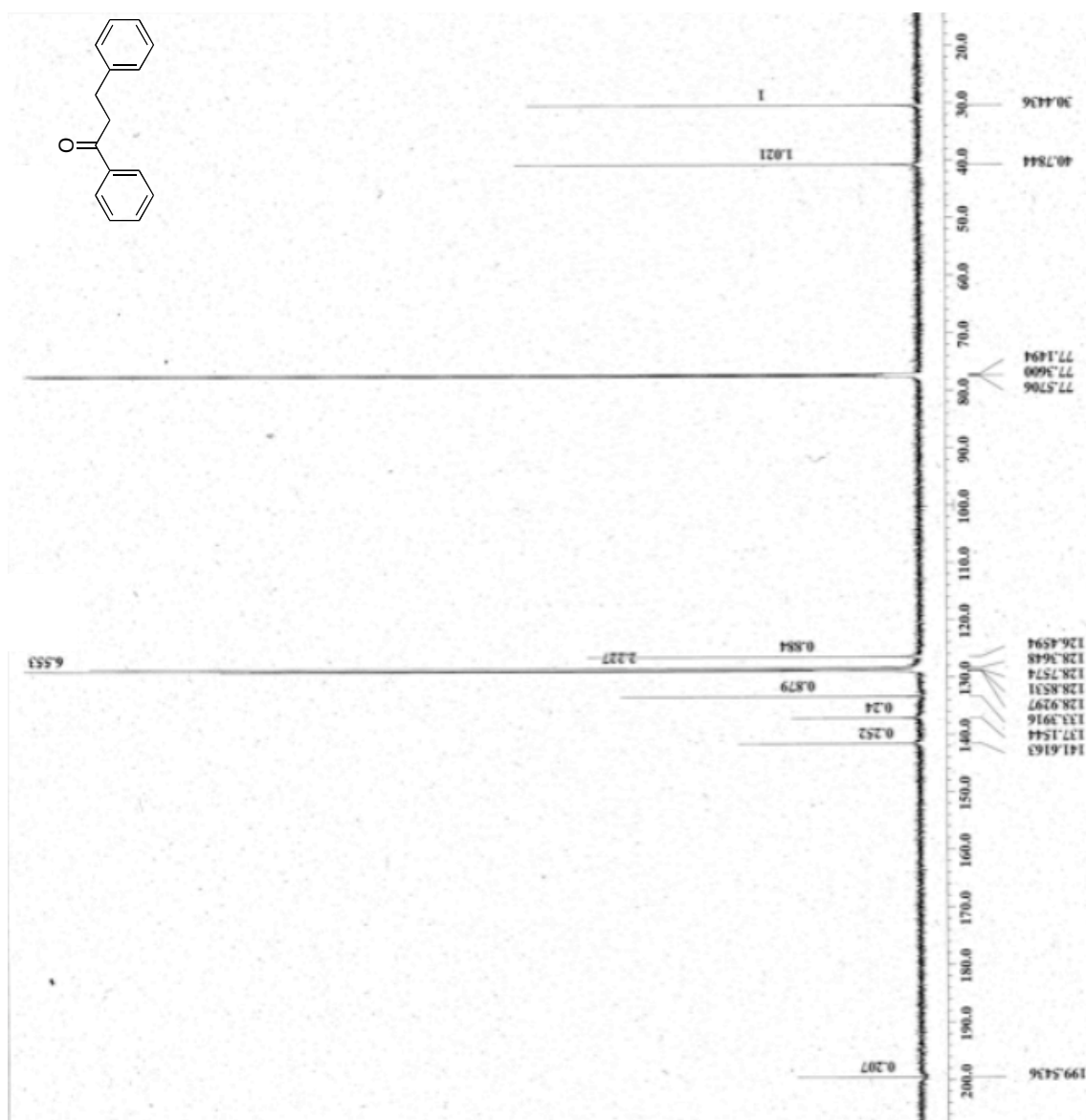


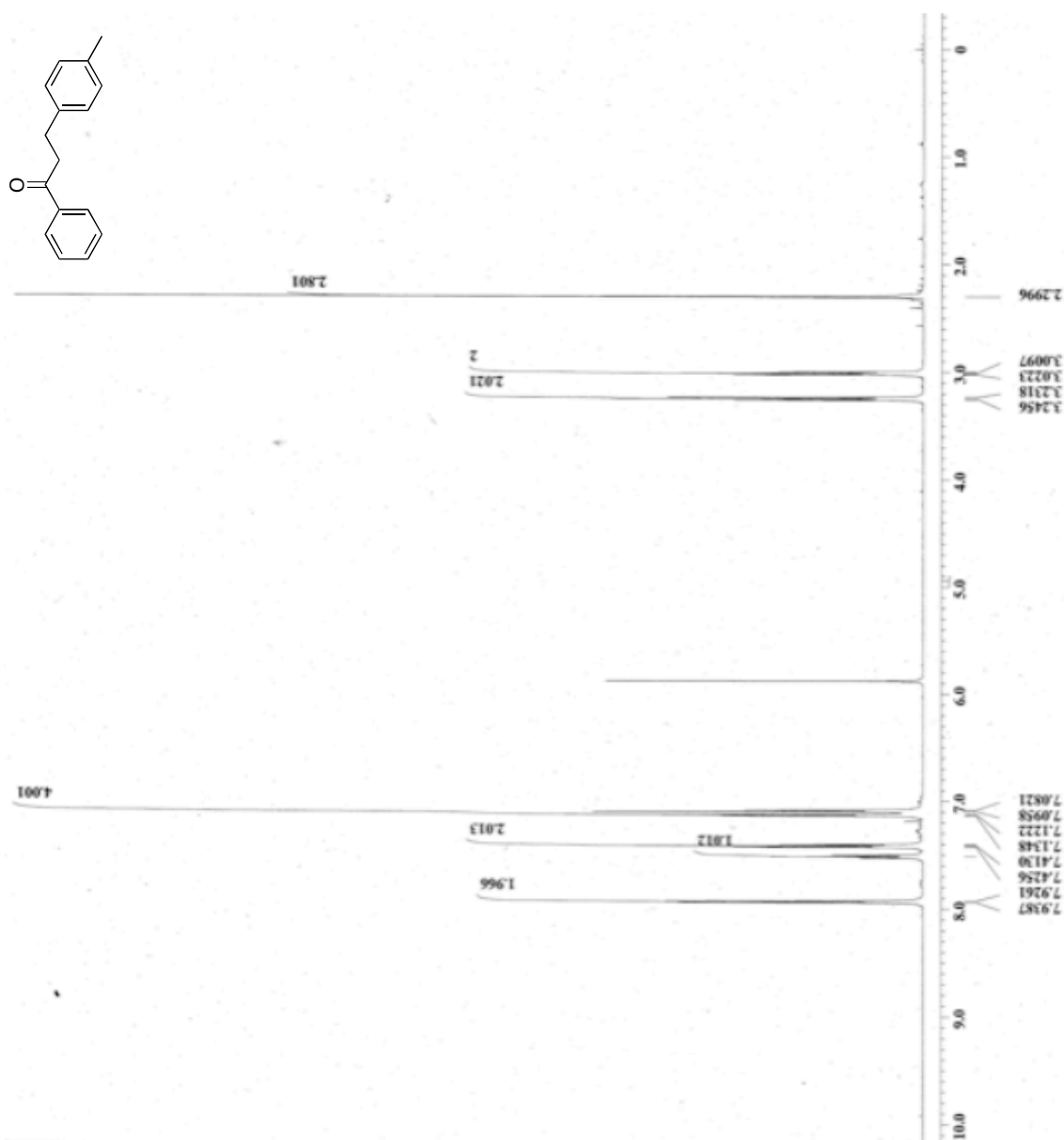


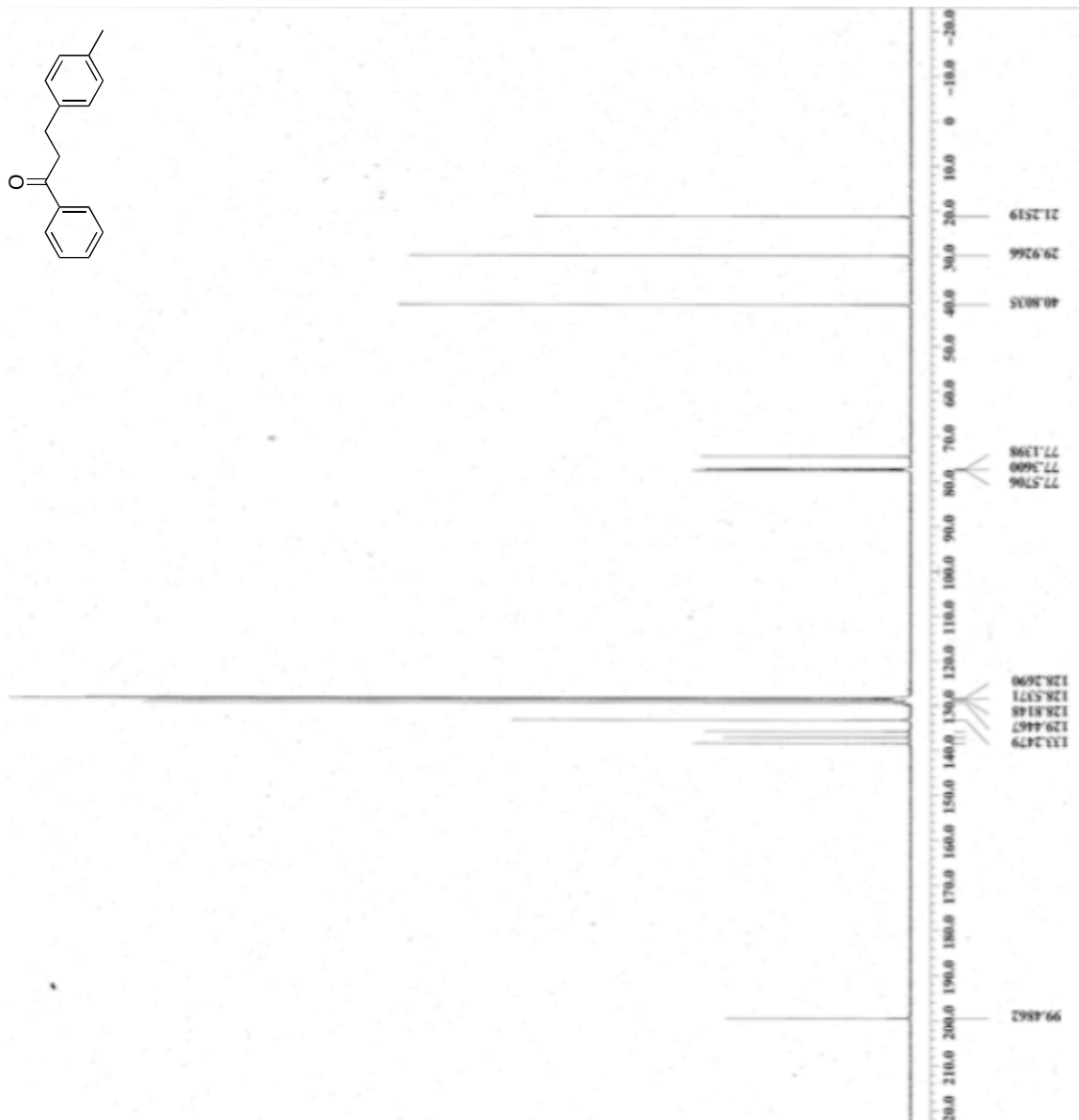


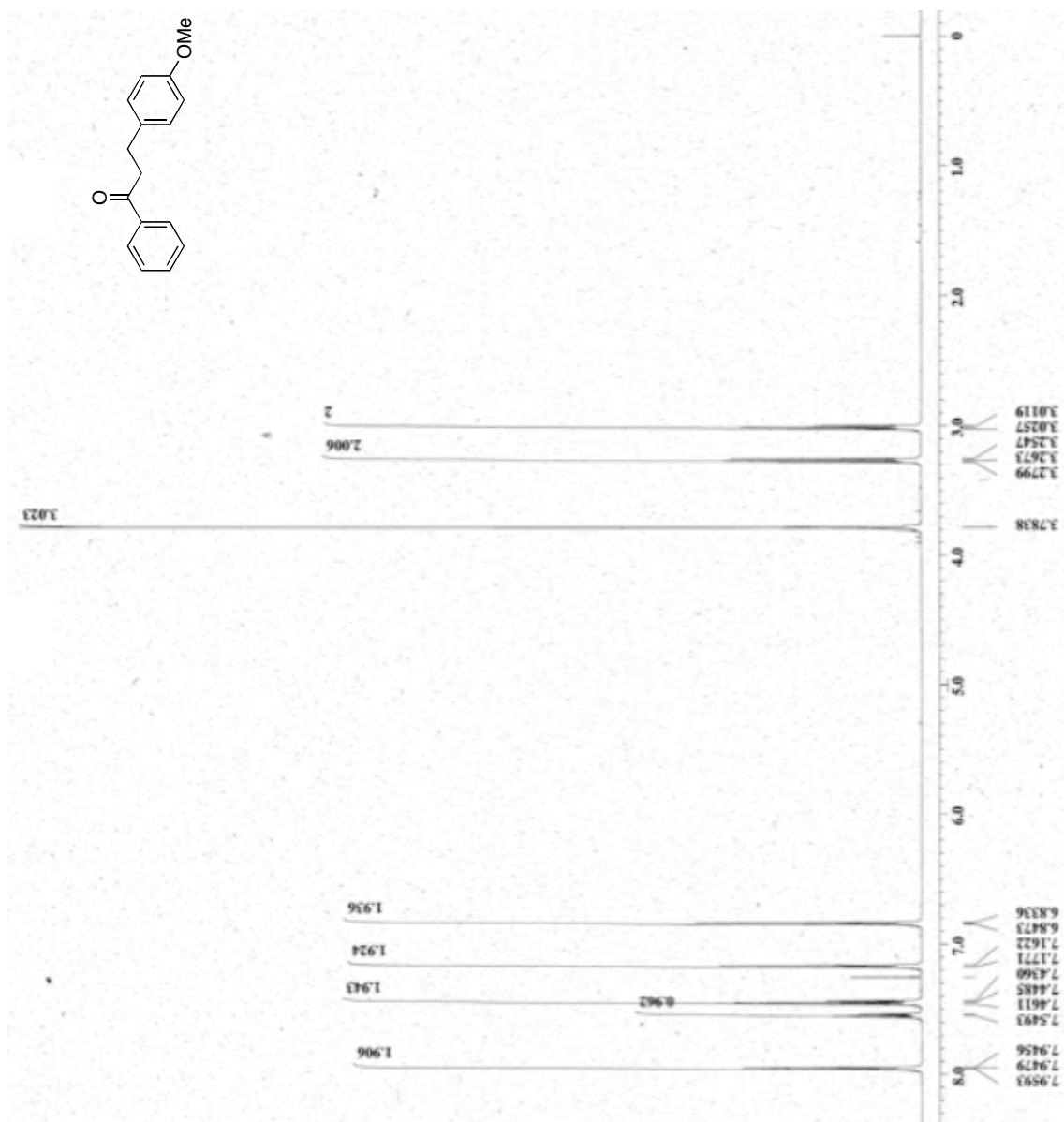


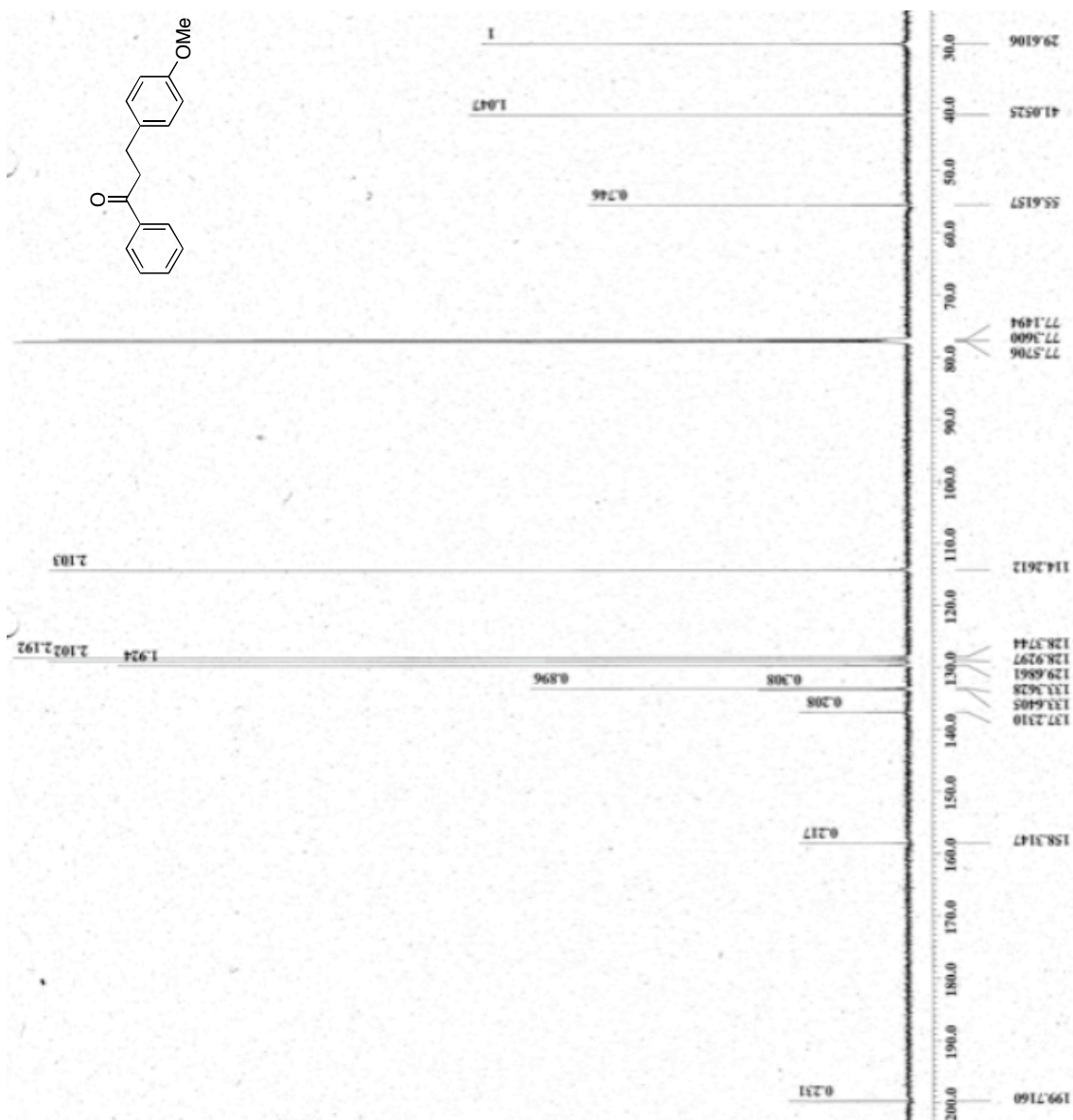


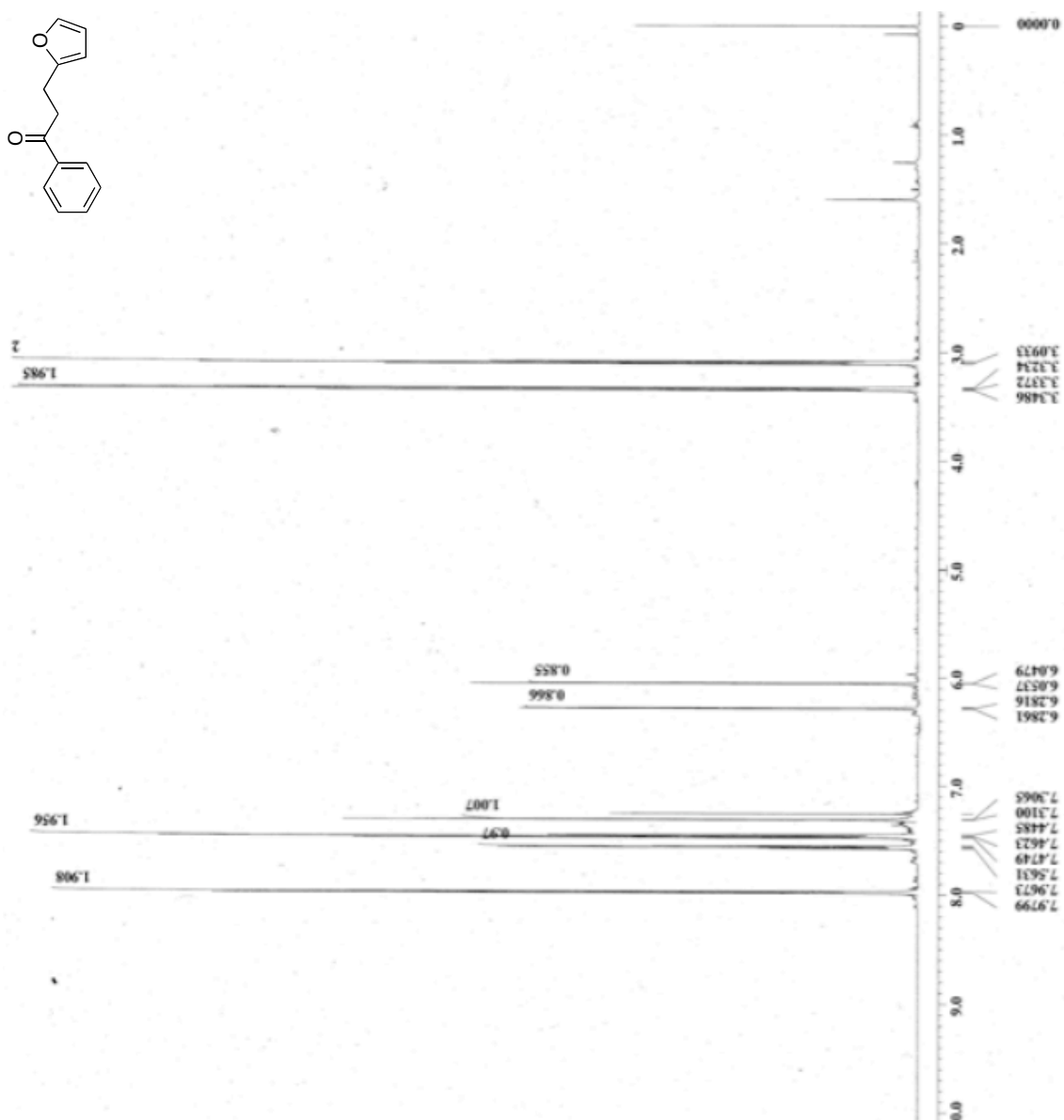


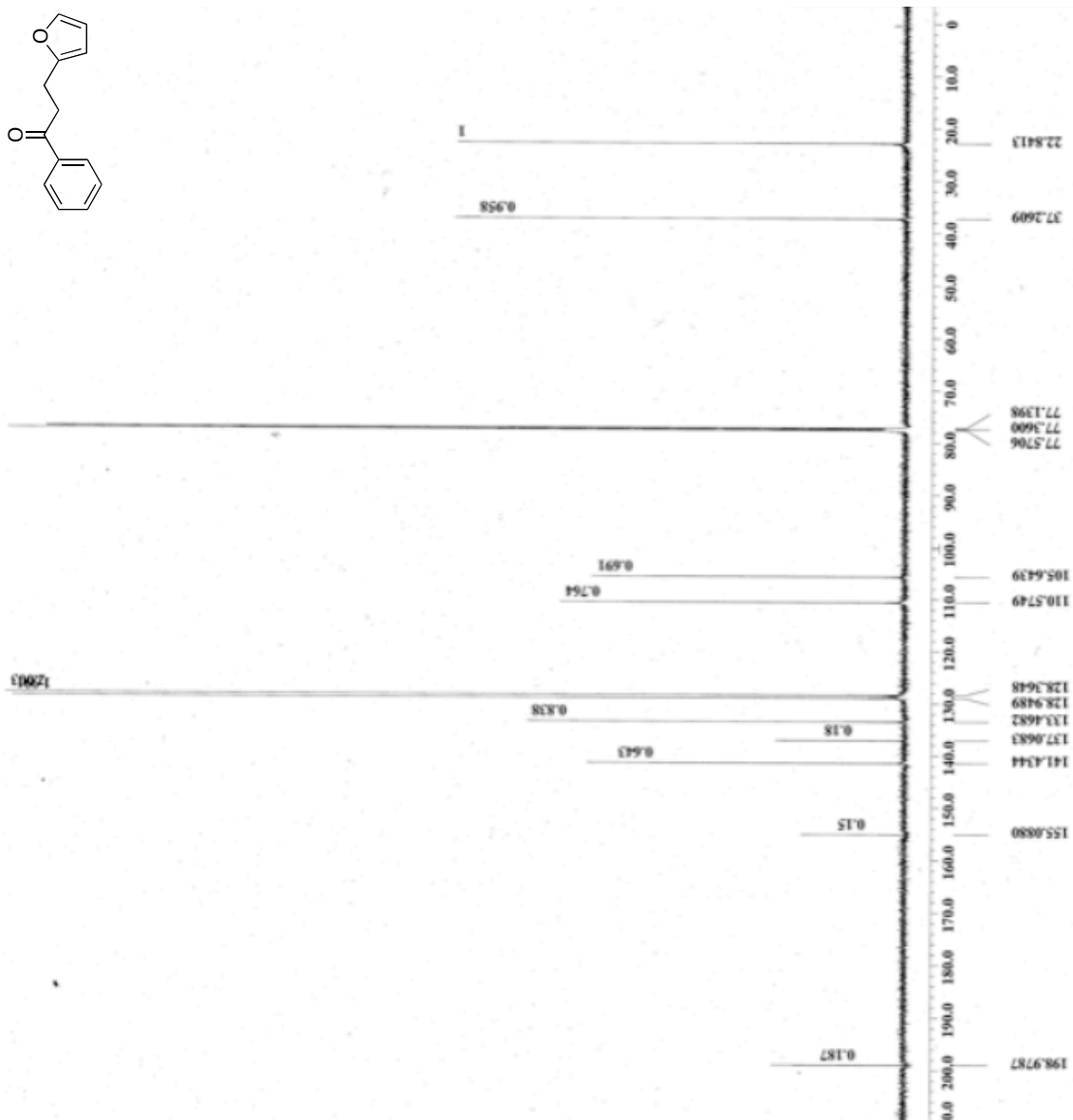


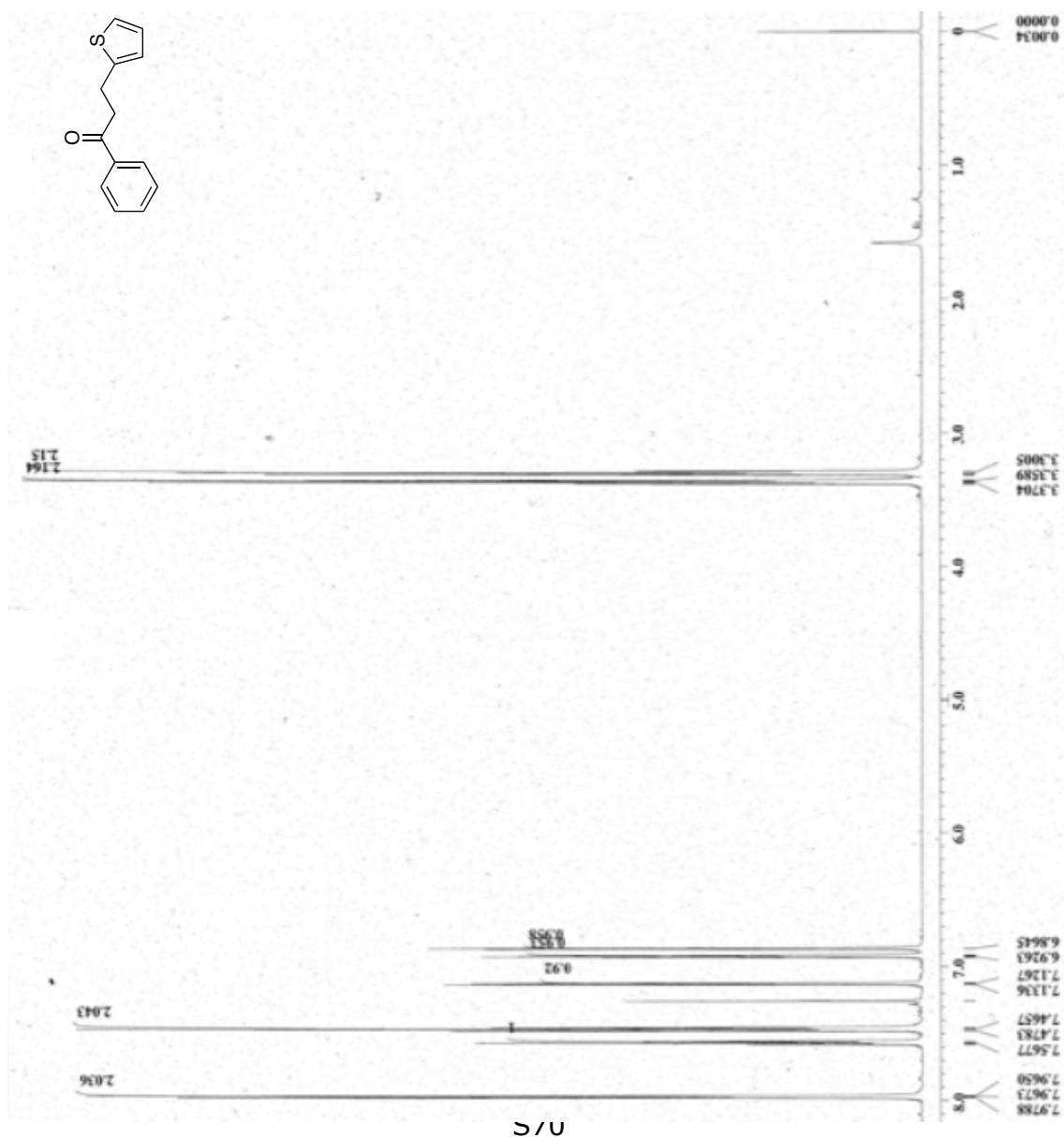


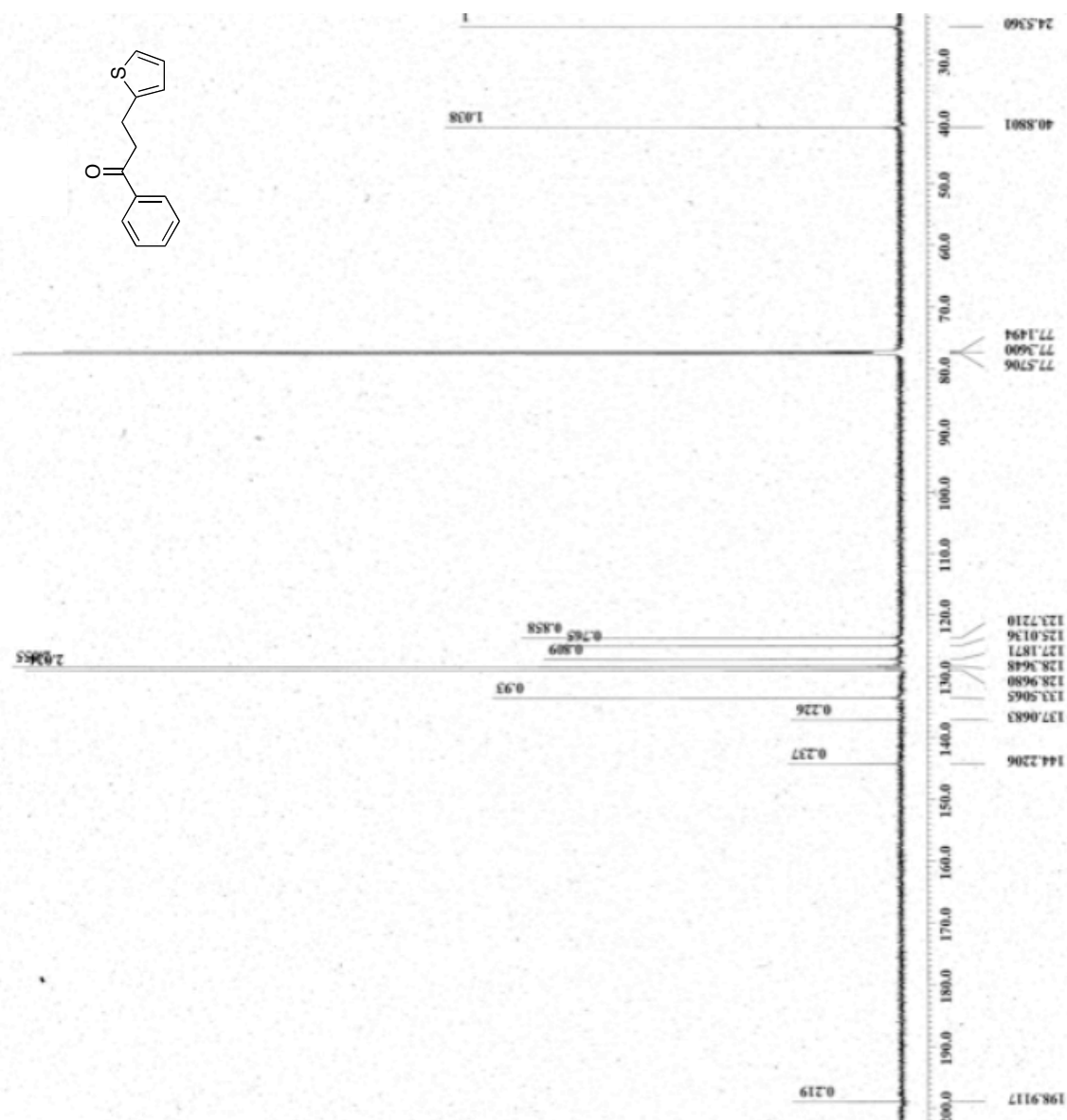


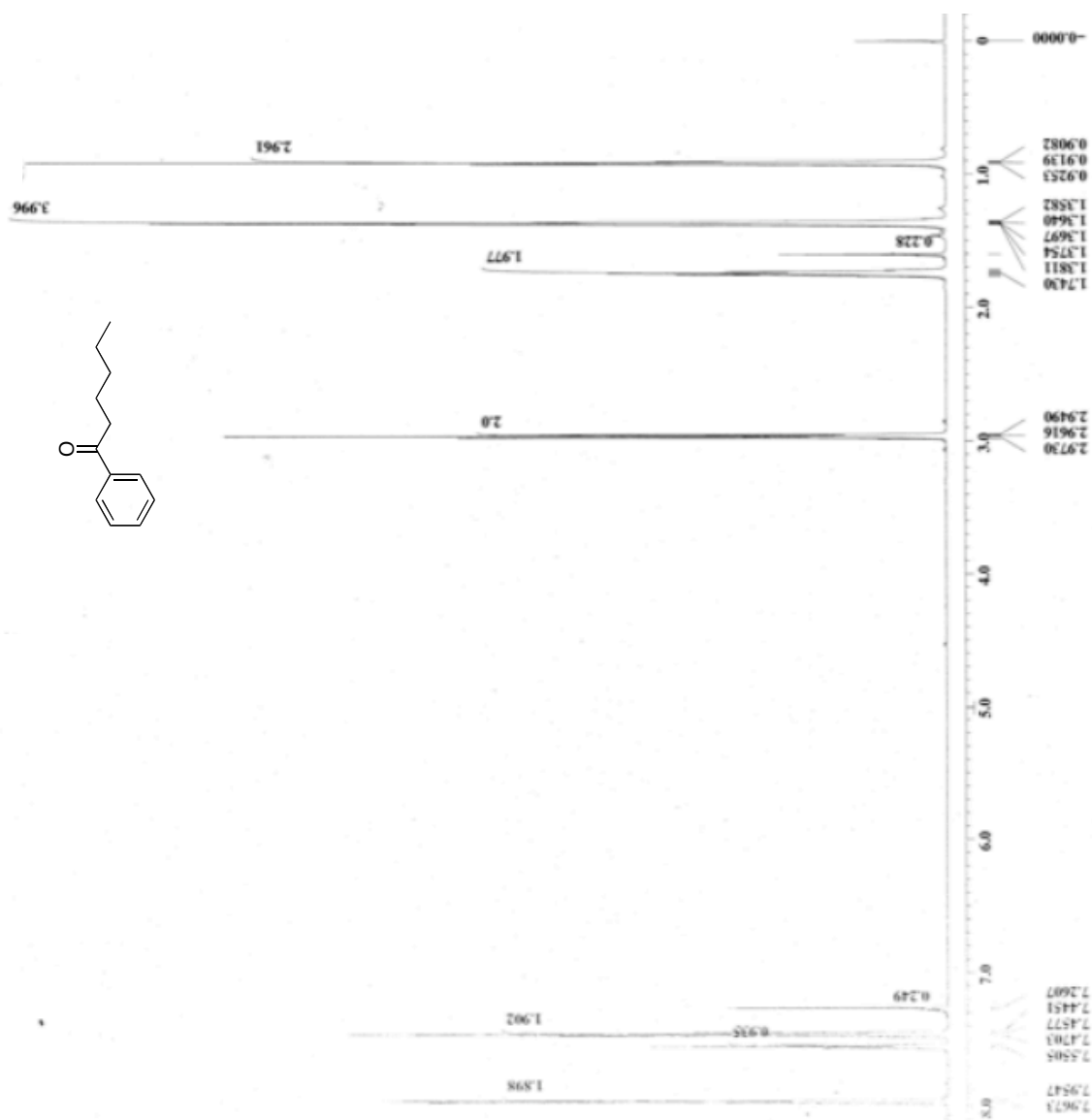


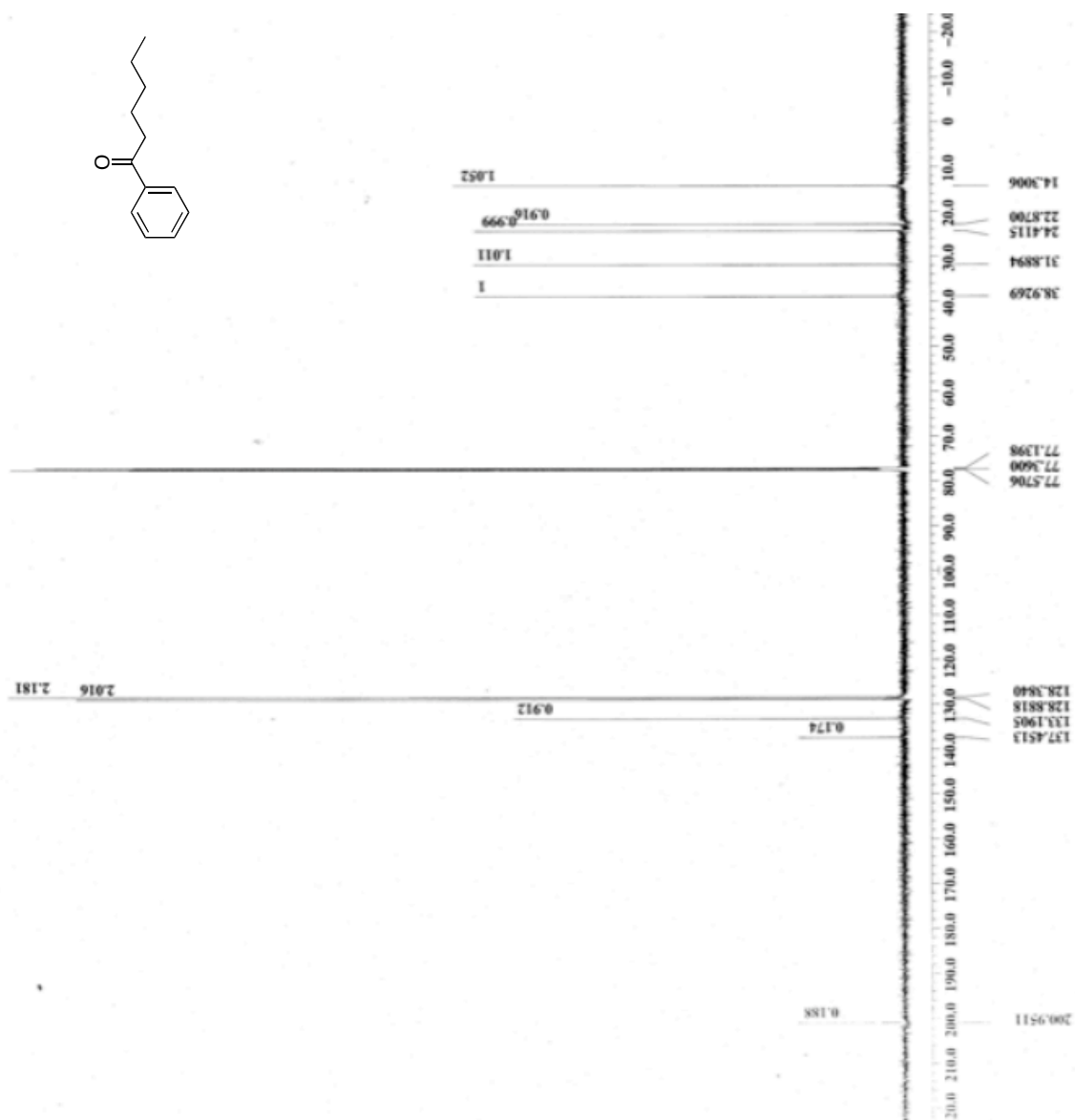


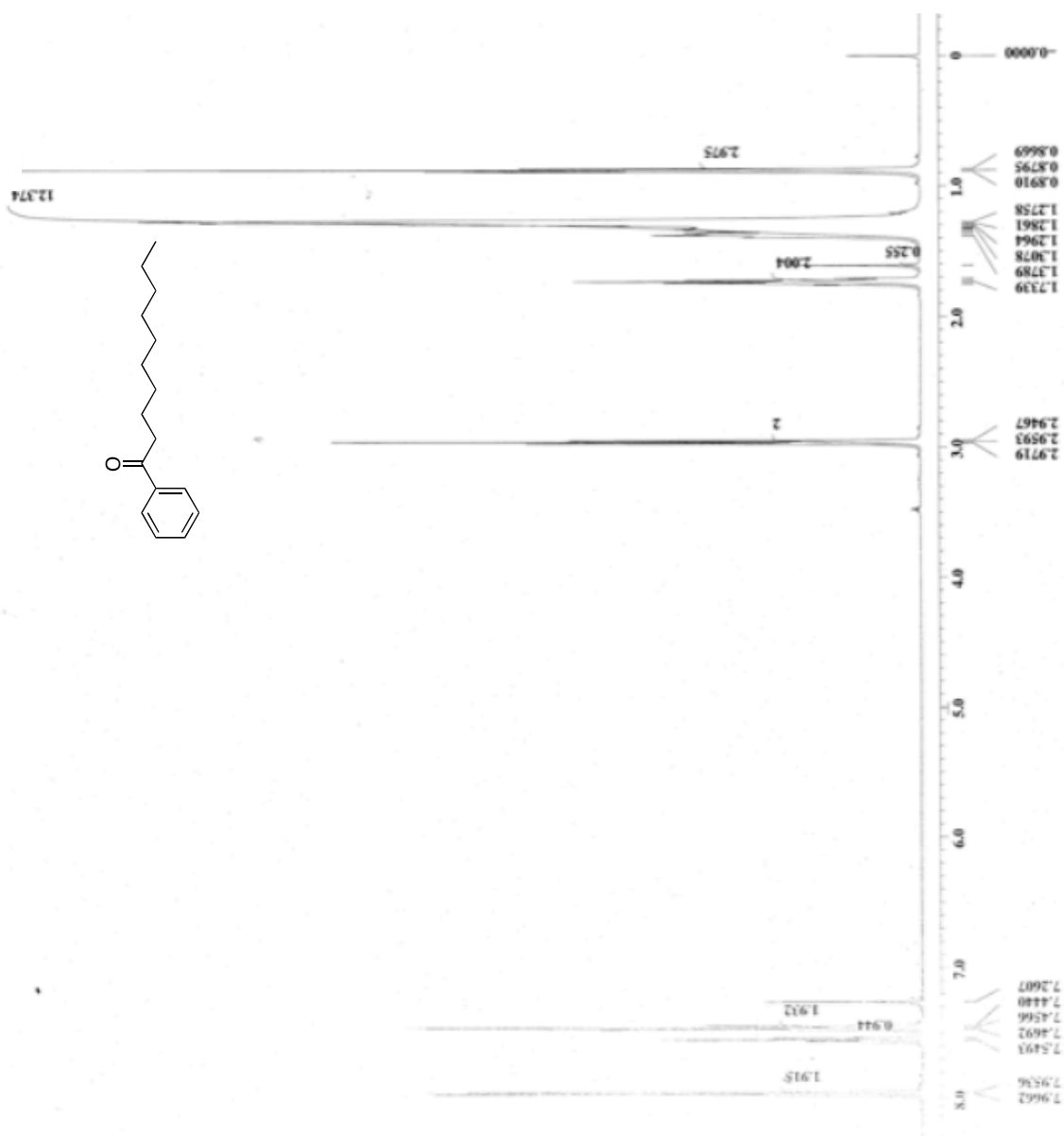


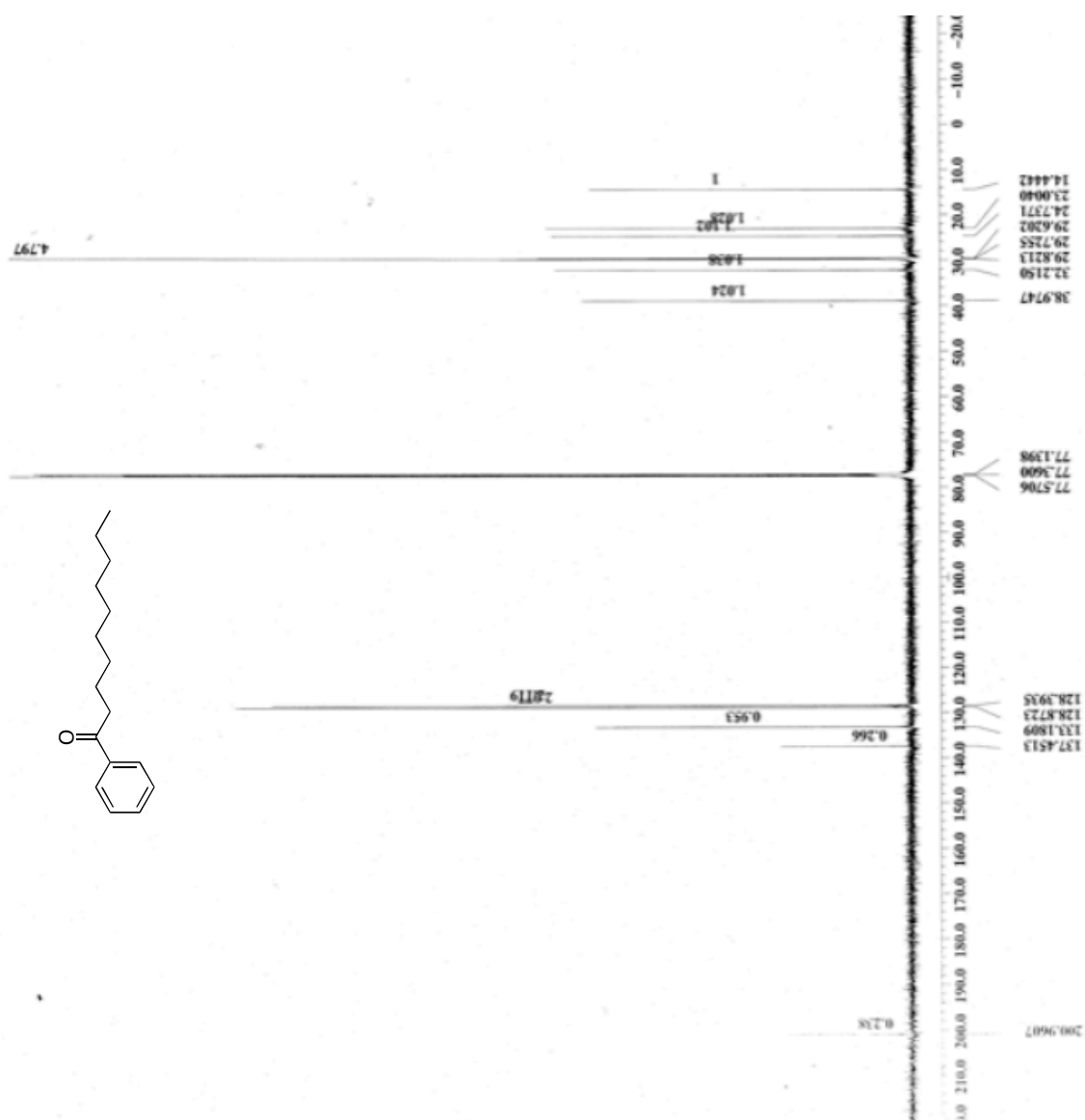


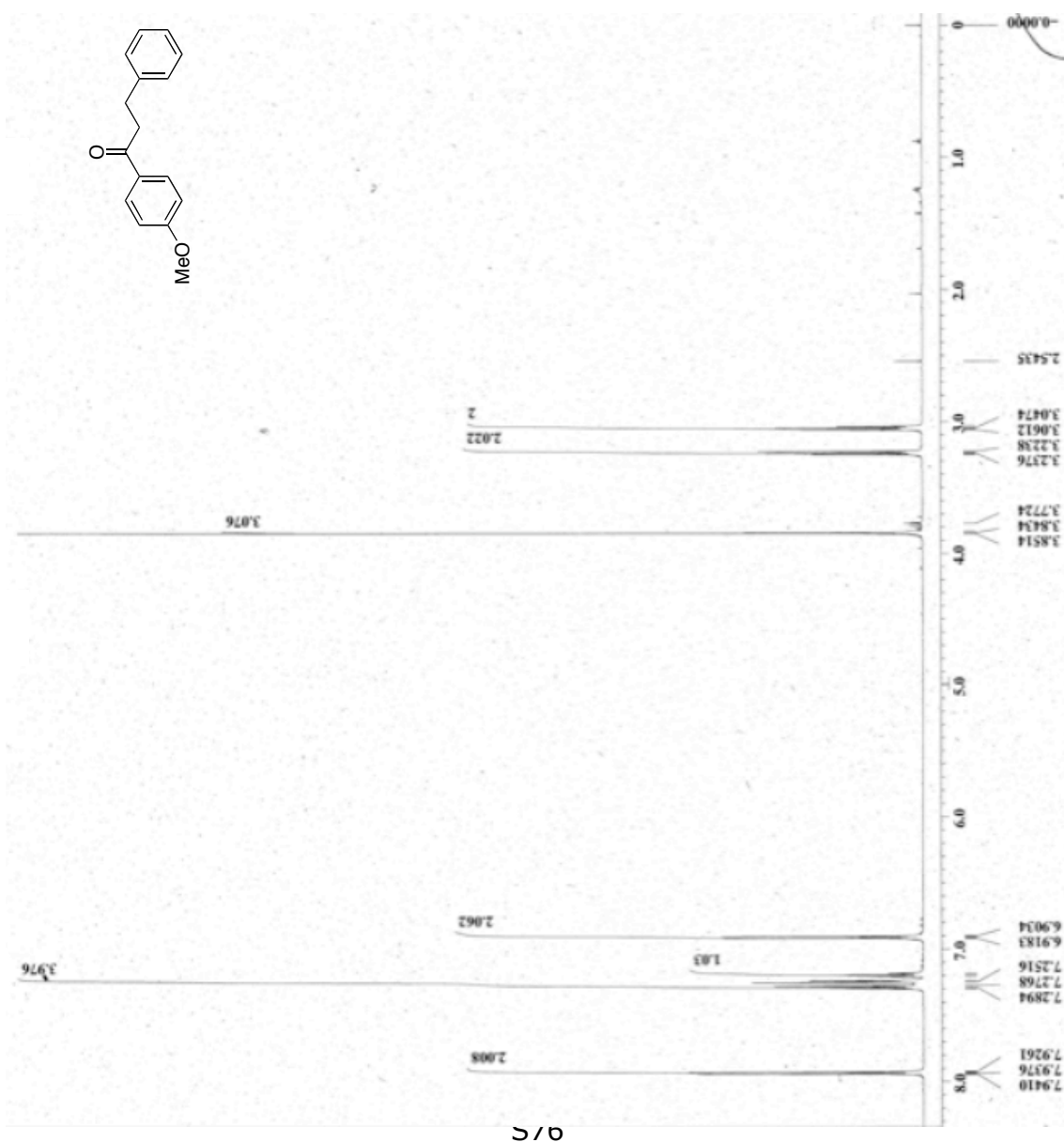


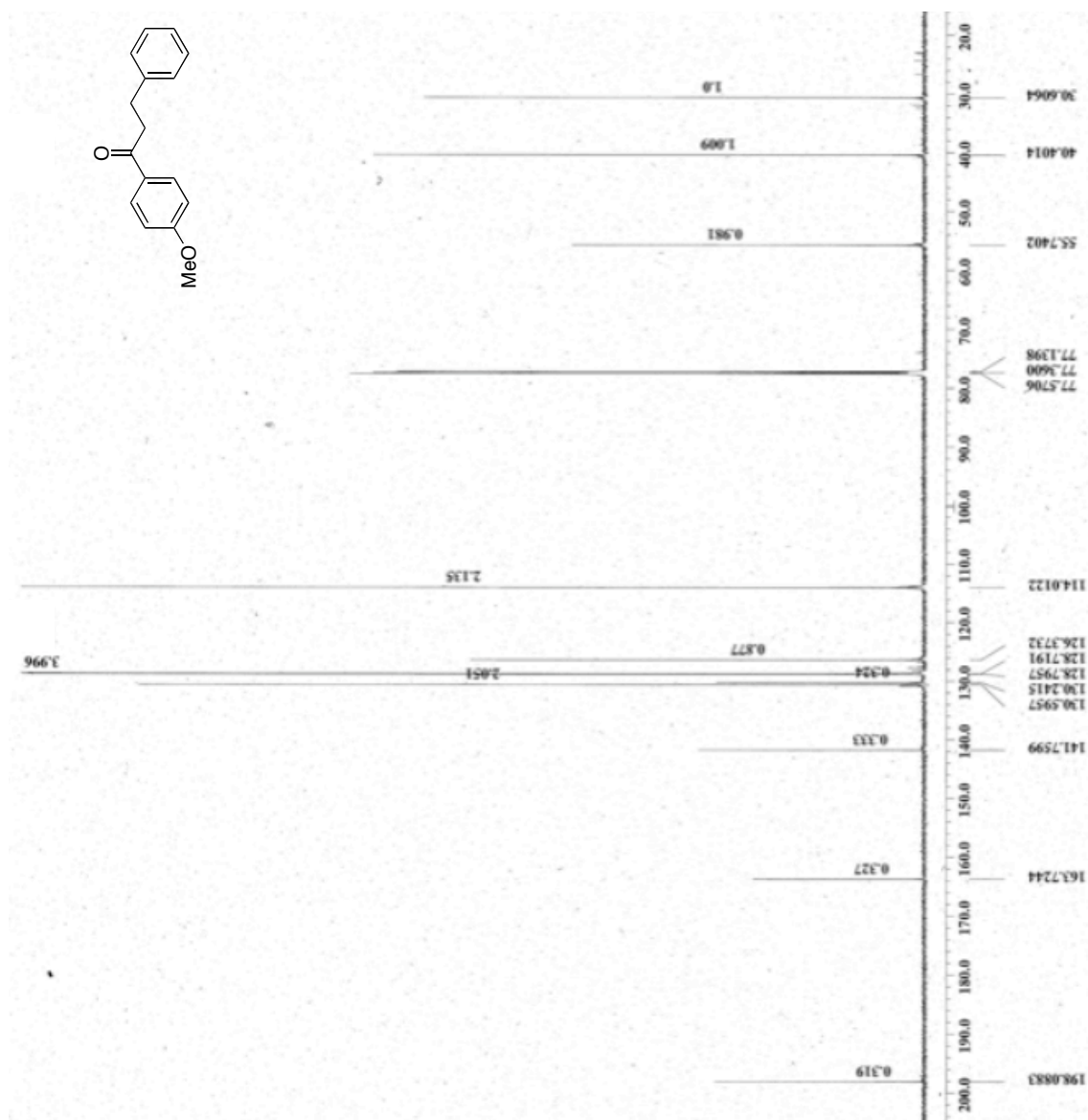


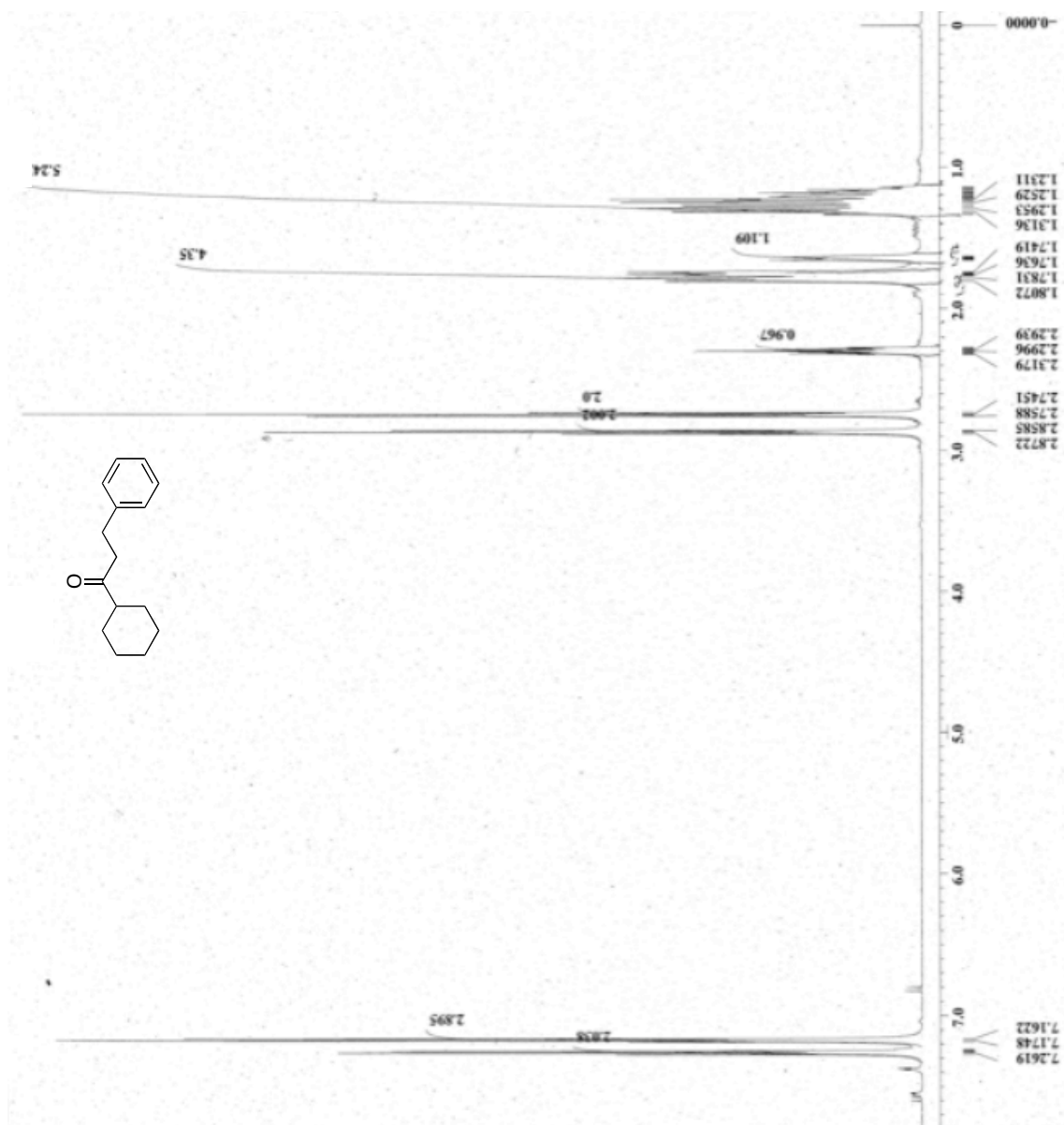


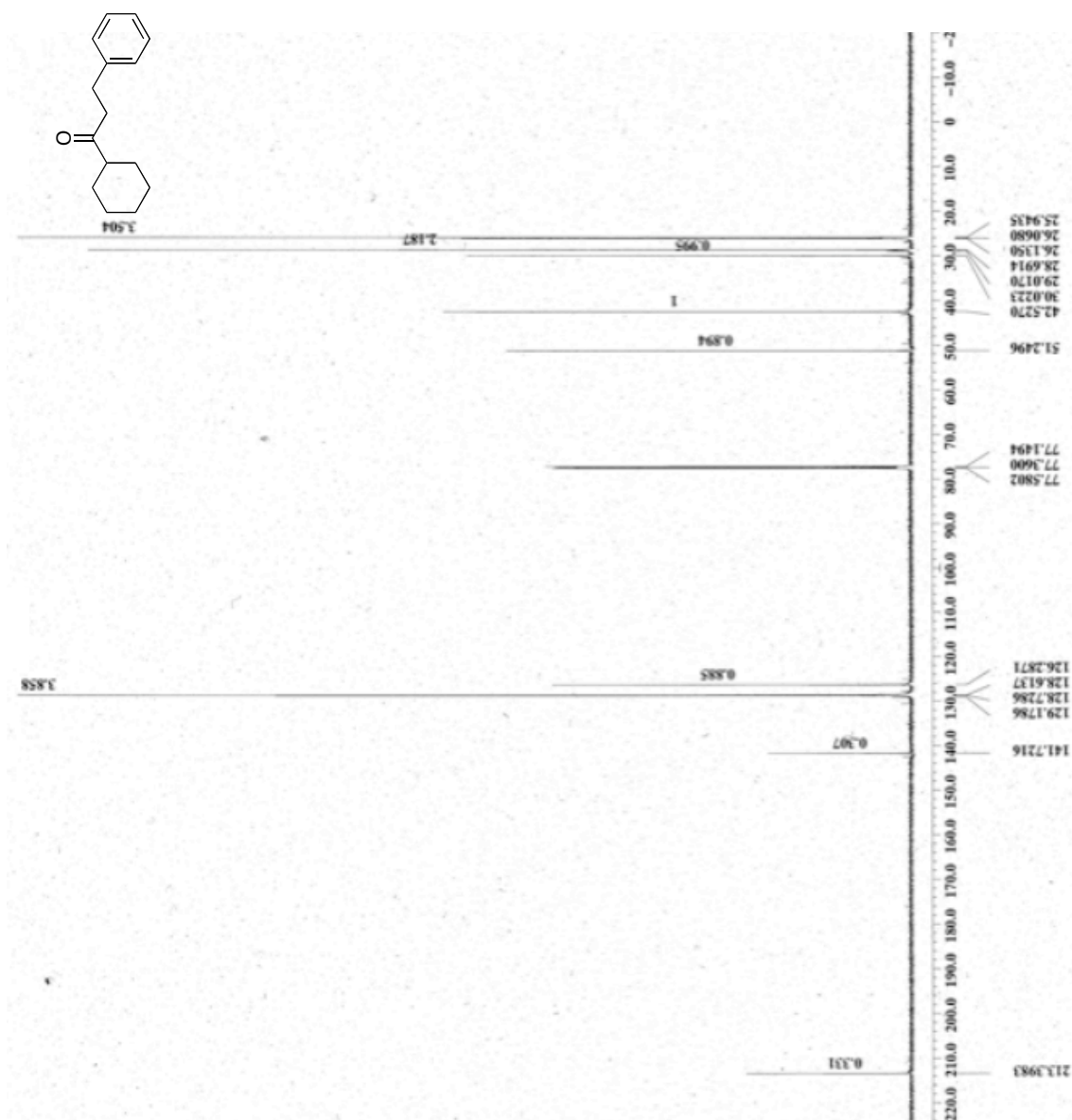


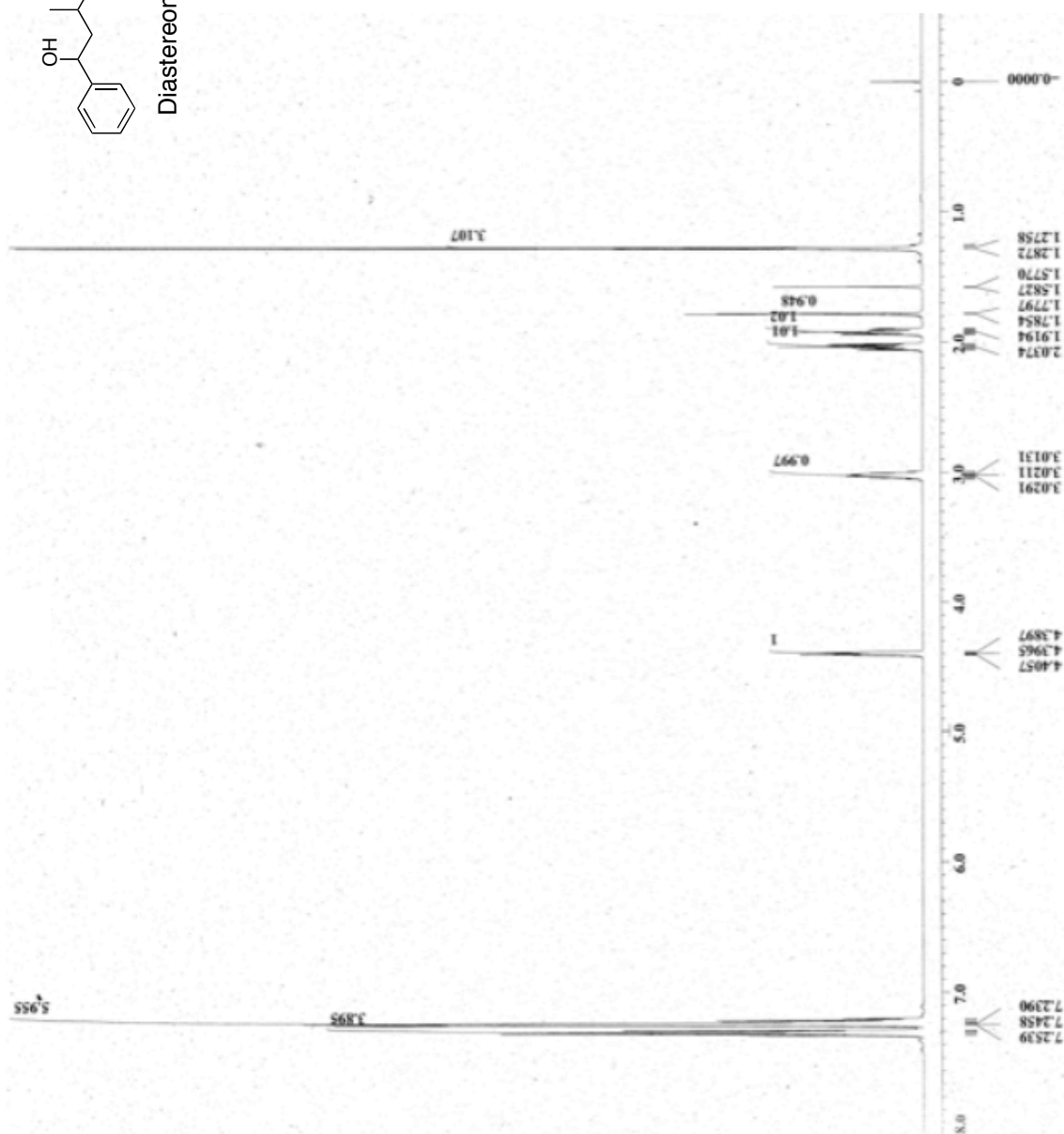
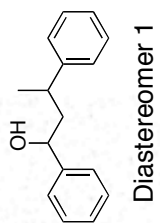


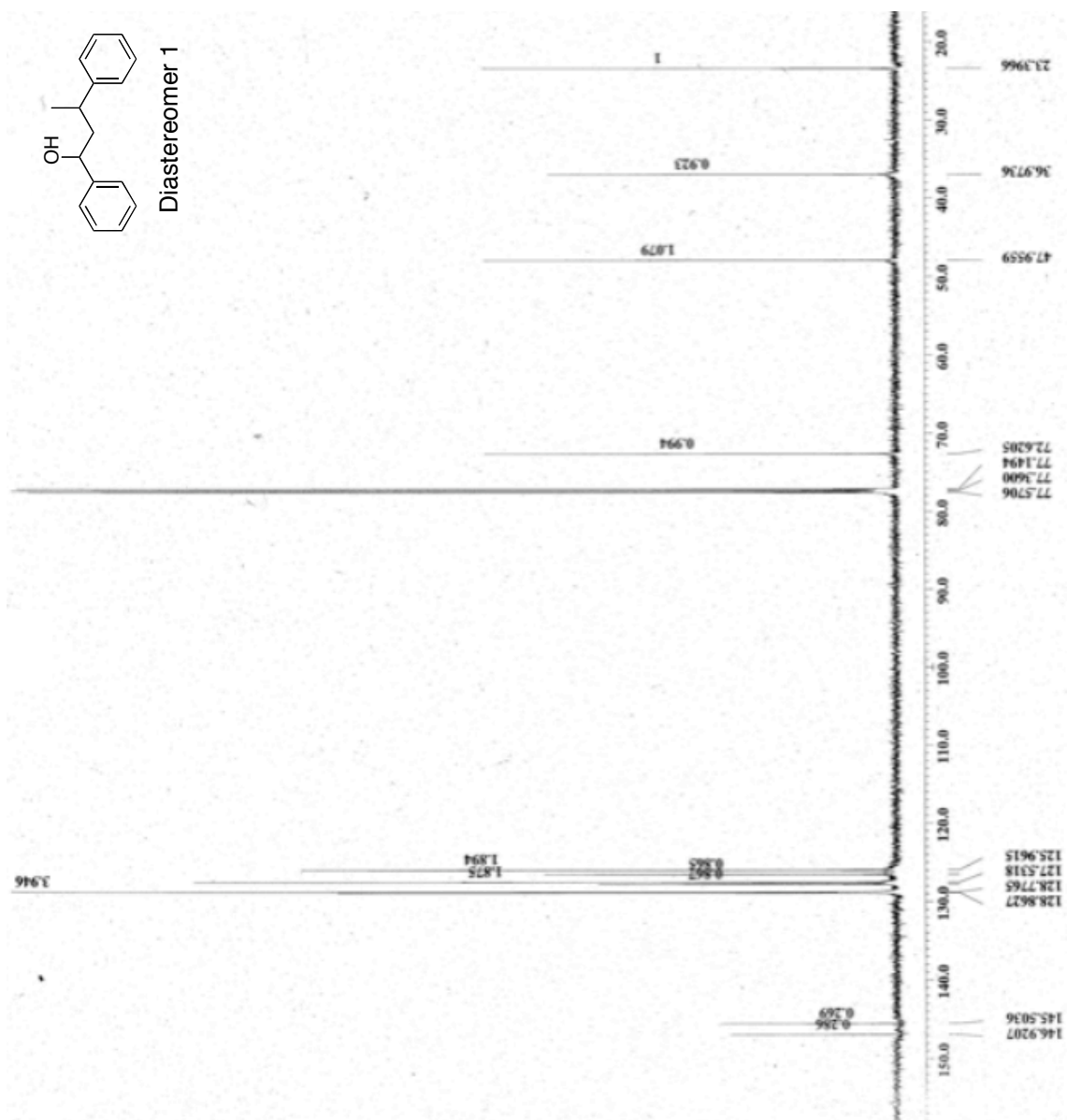


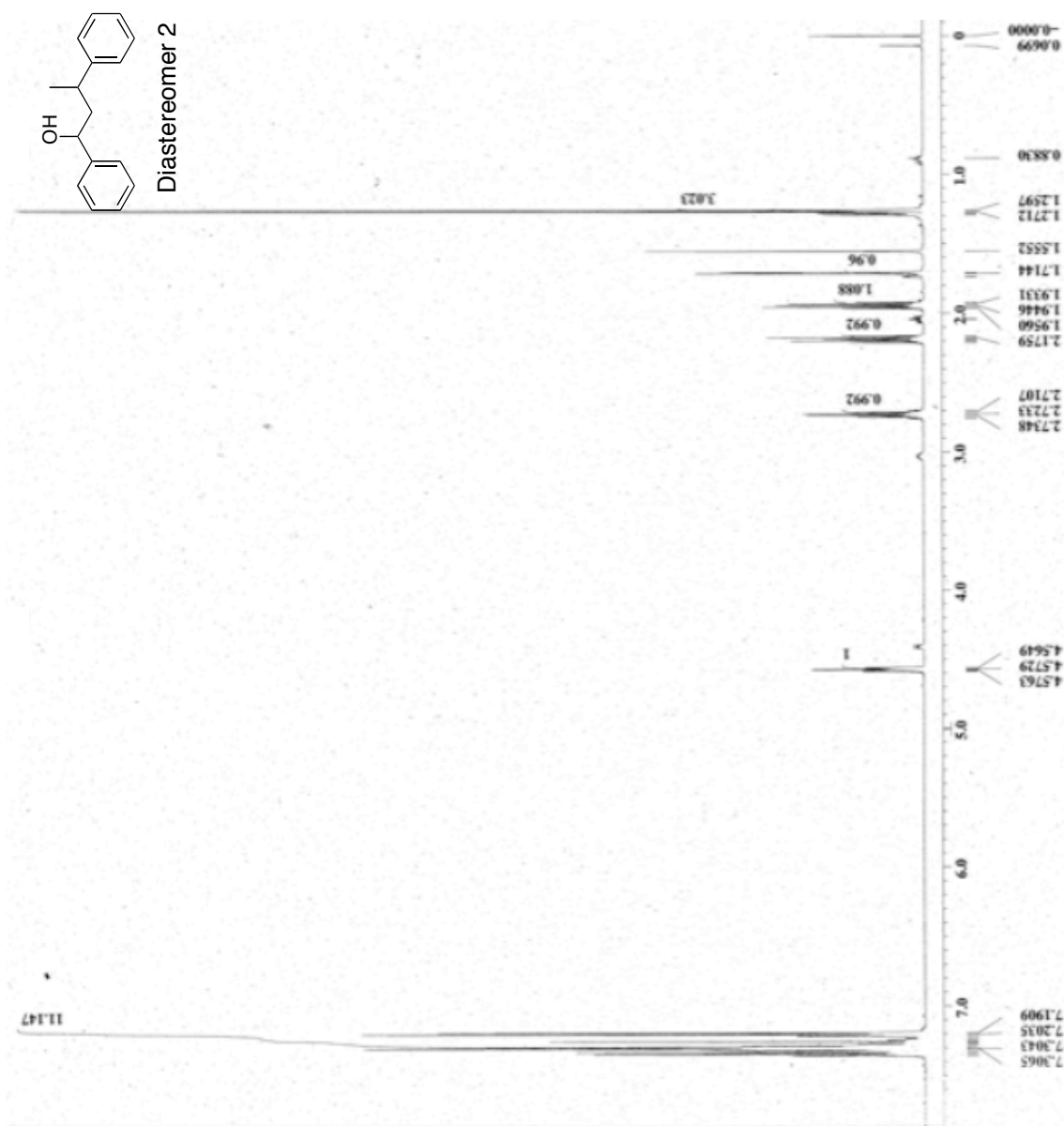


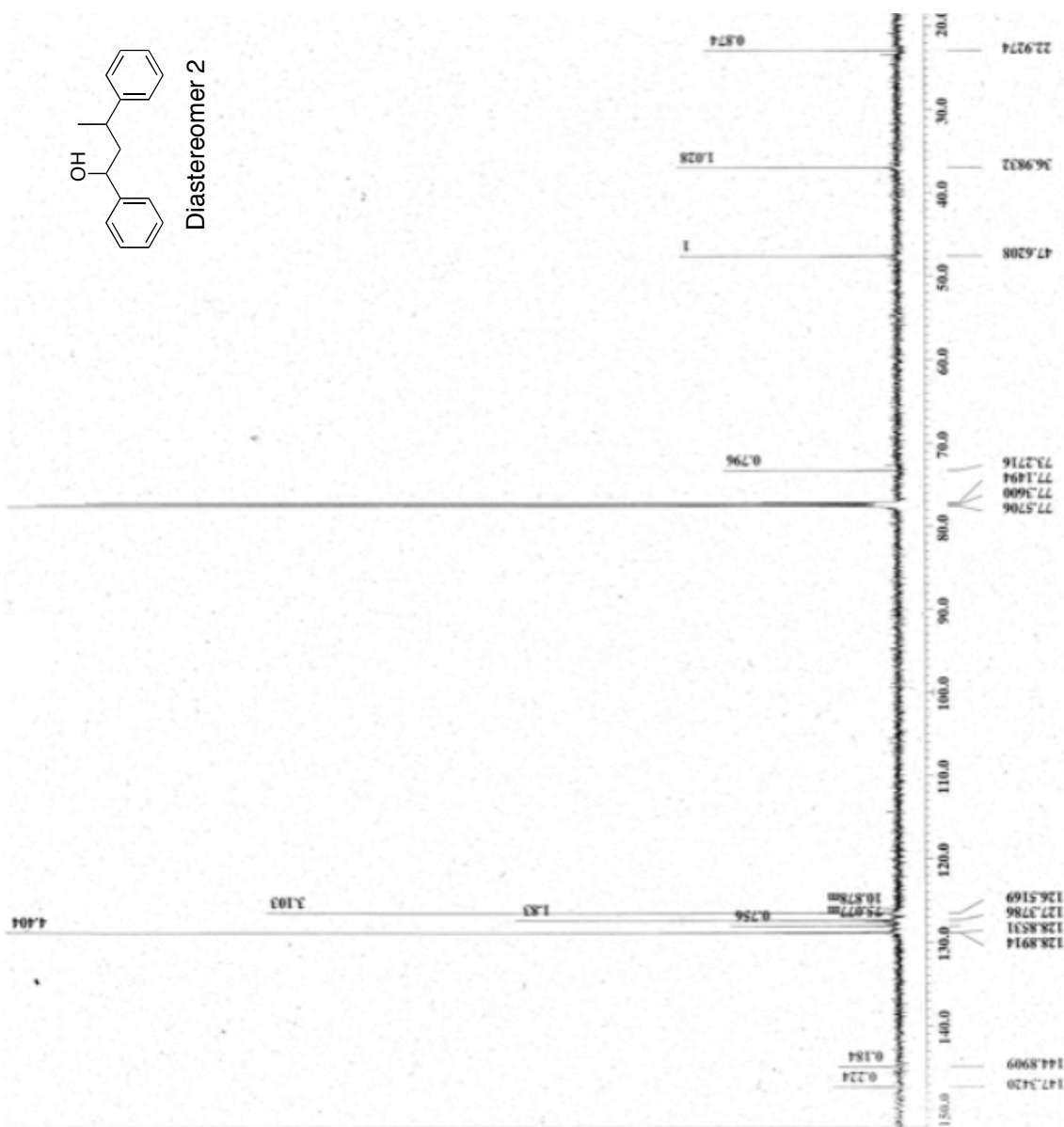
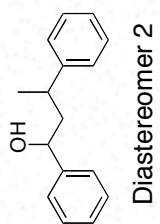












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