

Supporting Information

for

Magnesium bis(monoperoxyphthalate) hexahydrate as mild and efficient oxidant for the synthesis of selenones

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Dedicated to the memory of Professor Marcello Tiecco.

Experimental procedures, characterization data and copies of ¹H and ¹³C NMR spectra

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Materials and methods. Reactions were conducted in open glassware and were stirred with Teflon-coated magnetic stir bar. Solvents and reagents were used as received unless otherwise noted. Analytical thin layer chromatography (TLC) was performed on silica gel 60 F₂₅₄ pre-coated aluminium foil sheets and visualized by UV irradiation or by use of a KMnO₄ stain. Silica gel Kieselgel 60 (70–230 mesh) was used for column chromatography. NMR experiments were obtained at 25 °C on a Bruker DR 200 spectrometer operating at 200 MHz for ¹H and 50 MHz for ¹³C experiments. ¹H and ¹³C chemical shifts (δ) are reported in parts per million (ppm) and they are relative to the residual solvent peak of CDCl₃ at δ 7.26 and δ 77.00 in ¹H and ¹³C NMR, respectively. Data are reported as follows: chemical shift (multiplicity, number of hydrogens, coupling constants where applicable, and assignment where possible). Abbreviations are as follows: s (singlet), d (doublet), t (triplet), q (quartet), dd (doublet of doublet), dt (doublet of triplet), tt (triplet of triplet), m (multiplet), bs (broad signal). Coupling constant (J) quoted in Hertz (Hz) to the nearest 0.1 Hz. Infrared (IR) spectra were recorded with a Jasco model 410 spectrometer on a Diffuse Reflectance sampling cell. Only significant absorption maxima (ν_{max}) are reported in wavenumbers (cm⁻¹). HRMS analysis were obtained with a HP–LCMS ESI–qTOF 6200 with Phenomenex Aeris Peptide C18 column (1.7 μm, ID 2.1, 100 mm). Melting points were determined on a Kofler hotstage apparatus and are uncorrected. Compounds **1a** [1], **1b** [2], **1c** [3], **1d** [4], **1g** [4], **1h** [5], **1i** [5] and **1j** [6] were prepared according to the literature. Compound **1e** [7] was synthesized by opening of the corresponding epoxide with phenylselenolate anion [8] whereas compound **1f** [9] from azido selenenylation of styrene.

(Decylselenonyl)benzene (2a). To a solution of selenide **1a** (0.25 g, 0.85 mmol) in absolute ethanol (10 mL) at room temperature, magnesium monoperoxyphthalate, MMPP, (1.02 g, 2.04 mmol) was added. The reaction mixture was vigorously stirred until TLC analysis showed that the starting selenide was completely converted into the corresponding selenone (3 h). The mixture was then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 × 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum to afford **2a** as a colorless oil (0.24 g, 94%). FTIR: $\nu_{\max}/\text{cm}^{-1}$ 2924, 1723, 1445, 1286, 926, 879, 744. ^1H NMR (200 MHz, CDCl_3) δ/ppm 8.01-7.90 (m, 2H, H-Ar), 7.73-7.50 (m, 3H, H-Ar), 3.42 (t, 2H, $J = 7.3$ Hz, CH_2), 2.05-1.80 (m, 2H, CH_2), 1.48-1.03 (m, 14H, CH_2), 0.82 (t, 3H, $J = 6.0$ Hz, CH_3). ^{13}C NMR (50 MHz, CDCl_3) δ/ppm 139.8, 134.2, 130.2 (2C), 127.0 (2C), 60.0, 31.7, 29.3, 29.1 (2C), 28.8, 28.4, 22.5, 22.3, 14.0. HRMS (ESI-TOF): m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{16}\text{H}_{27}\text{O}_2\text{Se}$ 331.1176; found 331.1168.

At the end of the reaction the oxidant was present as phthalic acid which was extracted by aqueous base. Acidification (pH 2) of the aqueous extract with hydrochloric acid, extraction with dichloromethane (2 × 20 mL) and drying of the organic phase with sodium sulfate produced pure phthalic acid with a 85% yield.

[(*trans*-2-Methoxy-2-methylcyclohexyl)selenonyl]benzene (2b). Selenide **1b** (0.28 g, 1.00 mmol) was dissolved in THF (20 mL) and the resulting solution was stirred at room temperature. Powdered dipotassium hydrogen phosphate (0.55 g, 3.00 mmol) and magnesium monoperoxyphthalate (1.20 g, 2.40 mmol) were added and the resulting white suspension was vigorously stirred at room temperature. After 3 h 10% aqueous sodium carbonate (10 mL) was added to the stirred mixture and the solution extracted with ethyl acetate (2 × 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum to produce **2b** as a colorless oil (0.17 g, 54%). FTIR: $\nu_{\max}/\text{cm}^{-1}$ 2940, 1653, 1442, 1119, 965, 861, 744. ^1H NMR (200 MHz, CDCl_3) δ/ppm 7.75-7.60 (m, 2H, *H*-Ar), 7.58-7.40 (m, 3H, *H*-Ar), 3.31 (s, 3H, OCH_3), 2.81 (dd, 1H, $J = 12.0, 5.6$ Hz, *CH*), 2.12-2.01 (m, 1H, CH_2), 1.92-1.55 (m, 3H, CH_2), 1.48 (s, 3H, CH_3), 1.39-1.05 (m, 4H, CH_2). ^{13}C NMR (50 MHz,

CDCl₃) δ /ppm 140.6, 130.6, 129.3, 129.1, 126.0, 125.8, 76.5, 73.1, 48.7, 37.1, 25.7, 22.7, 20.7, 18.1. HRMS (ESI-TOF): m/z [M + H]⁺ calcd for C₁₄H₂₁O₃Se 317.0655; found 317.0654.

(Vinylselenonyl)benzene (2c). A mixture of selenide **1c** (0.06 g, 0.33 mmol), dipotassium hydrogen phosphate (0.17 g, 0.99 mmol) and magnesium monoperoxyphthalate (0.39 g, 0.70 mmol) in THF (10 mL) was stirred at room temperature for 2 h. The mixture was then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 \times 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum to afford **2c** as amorphous light yellow solid (0.06 g, 80%): mp 99-103 °C (from CH₂Cl₂) (lit. [9], 97-102 °C). ¹H and ¹³C NMR spectra are in agreement to that previously reported [9]. ¹H NMR (200 MHz, CDCl₃) δ /ppm 8.10-7.92 (m, 2H, *H*-Ar), 7.78-7.70 (m, 1H, *H*-Ar), 7.68-7.62 (m, 2H, *H*-Ar), 7.04 (dd, 1H, *J* = 16.6, 9.1 Hz, *CH*), 6.75 (dd, 1H, *J* = 16.6, 2.1 Hz, *CH*), 6.47 (dd, 1H, *J* = 9.1, 2.1 Hz, *CH*); ¹³C NMR (50 MHz, CDCl₃) δ /ppm 141.3, 138.9, 134.2, 131.2, 130.4 (2C), 126.9 (2C).

4-Methyl-N-[3-(phenylselenonyl)propyl]benzenesulfonamide (2d). To a solution of selenide **1d** (0.11 g, 0.30 mmol) in absolute ethanol (8 mL) at room temperature, magnesium monoperoxyphthalate (0.35 g, 0.71 mmol) was added. The reaction mixture was vigorously stirred until TLC analysis showed that the starting selenide was completely converted into the corresponding selenone (1 h). The mixture was then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 \times 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum. The solid residue was washed with ethyl acetate and pure **2d** was obtained as a light yellow solid (0.07 g, 63%); mp 137-139 °C (from EtOAc). FTIR: ν_{\max} /cm⁻¹ 3164, 1325, 1154, 927, 881, 751. ¹H NMR (200 MHz, CDCl₃) δ /ppm 8.03 (dd, 2H, *J* = 7.8, 1.4 Hz, *H*-Ar), 7.83-7.64 (m, 5H, *H*-Ar), 7.32 (d, 2H, *J* = 8.0 Hz, *H*-Ar), 5.52 (t, 1H, *J* = 6.5 Hz, *NH*), 3.67 (t, 2H, *J* = 7.0 Hz, *CH*), 3.21 (dt, 2H, *J* = 7.0, 6.5 Hz, *CH*), 2.45 (s, 3H, *CH*), 2.30 (quint, 2H, *J* = 7.0 Hz, *CH*). ¹³C NMR (50 MHz, CDCl₃) δ /ppm 143.6, 141.1, 136.6, 134.5 (2C), 130.4 (2C), 129.8 (2C), 127.0 (3C), 56.9,

41.2, 23.0, 21.5. HRMS (ESI-TOF): m/z $[M + H]^+$ calcd for $C_{16}H_{20}NO_4SSe$ 402.0278; found 402.0274.

Oxidative-Cyclization of **1d–j**

1-[(4-Methylphenyl)sulfonyl]azetidine (3d**)**. To a solution of phenylseleno amide **1d** (0.20 g, 0.54 mmol) in ethanol (20 mL) at room temperature magnesium monoperoxyphthalate (0.65 g, 1.30 mmol) was added. The reaction mixture was stirred until TLC analysis showed that the starting selenide was completely converted into the corresponding selenone (1 h). Powdered potassium hydroxide (0.24 g, 4.32 mmol) was then added and the consumption of the selenone was monitored by TLC (3 h). The mixture was poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 × 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum. Compound **3d** was obtained as amorphous solid (0.06 g, 55%) after chromatography (1: 1 diethyl ether/petroleum ether): mp 118-123 °C (from EtOH). The melting point range, 1H NMR and ^{13}C NMR spectra are in accordance with those obtained from a commercial sample. 1H NMR (200 MHz, $CDCl_3$) δ /ppm 7.78-7.65 (d, 2H, $J = 7.6$ Hz, $H-Ar$), 7.42-7.30 (d, 2H, $J = 7.6$ Hz, $H-Ar$), 3.75 (t, 4H, $J = 7.65$ Hz, CH_2), 2.46 (s, 3H, CH_3), 2.06 (quint, 2H, $J = 7.6$ Hz, CH_2). ^{13}C NMR (50 MHz, $CDCl_3$) δ /ppm 143.8, 131.0, 129.5 (2C), 128.10 (2C), 50.7, 21.3, 15.1.

At the end of the reaction the oxidant was present as phthalate and selenium as benzeneseleninate which were extracted by aqueous base. Acidification (pH 2) of the aqueous extract with hydrochloric acid and extraction with dichloromethane (2 × 20 mL) let us to recover the phthalic acid. The aqueous extract was evaporated and the residue was suspended in MeOH (10 mL). Hydrazine monohydrate (1.5 mmol, 0.07 mL) was added and the suspension stirred until diphenyl diselenide was formed, as indicated by the yellow color. The mixture was then concentrated in vacuo, poured into water and extracted with diethyl ether (3 × 5 mL). The organic layer was dried with sodium sulfate and concentrated in vacuum to afford diphenyl diselenide as a yellow solid (0.06 g, 67%).

cis-6-tert-Butyl-1-oxaspiro[2.5]octane (3e). To a solution of phenylseleno alcohol **1e** (0.13 g, 0.40 mmol) in THF (10 mL) at room temperature, powdered dipotassium hydrogen phosphate (0.21 g, 1.20 mmol) and magnesium monoperoxyphthalate (0.58 g, 0.96 mmol) were added. The reaction mixture was stirred until TLC analysis showed that the starting selenide was completely converted into the corresponding epoxide (16 h). The mixture was then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 × 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum to afford **3e** as a colorless oil (0.05 g, 77%): ¹H NMR spectra is identical to that previously reported [7]. ¹H NMR (200 MHz, CDCl₃) δ/ppm 2.67 (s, 2H, OCH₂), 2.02-1.72 (m, 4H, CH₂), 1.51-1.19 (m, 5H, CH and CH₂), 0.90 (s, 9H, CH₃). ¹³C NMR (50 MHz, CDCl₃) δ/ppm 58.2, 53.7, 47.1, 33.3 (2C), 32.4, 27.6 (2C), 27.5, 24.8 (2C).

2,4-Diphenyl-4,5-dihydro-1,3-oxazole (3f). To a solution of phenylseleno amide **1f** (0.30 g, 0.80 mmol) in methanol (10 mL) at room temperature, powdered magnesium monoperoxyphthalate (0.95 g, 1.92 mmol) was added. The reaction mixture was stirred for 4 h. The mixture was then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 × 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum to afford the 1,3-oxazoline **3f** as a colorless oil (0.15 g, 84%). FTIR: ν_{max}/cm⁻¹ 3062, 1723, 1647, 1275, 760, 697 cm⁻¹. ¹H NMR (200 MHz, CDCl₃) δ/ppm 8.20-7.90 (m, 2H, *H*-Ar), 7.65-7.12 (m, 8H, *H*-Ar), 5.41 (dd, 1H, *J* = 10.1, 8.1 Hz, OCH₂), 4.82 (dd, 1H, *J* = 10.1, 8.4 Hz, OCH₂), 4.31 (dd, 1H, *J* = 8.4, 8.1 Hz, NCH). ¹³C NMR (50 MHz, CDCl₃) δ/ppm 164.9, 142.3, 131.6, 129.1, 129.0, 128.9, 128.8, 128.5, 128.4, 127.8, 127.7, 126.8, 126.6, 74.9, 70.0. HRMS (ESI-TOF): *m/z* [M + H]⁺ calcd for C₁₅H₁₄NO ([M + H]⁺) 224.1075; found 224.1061.

2-Phenyl-5,6-dihydro-4*H*-1,3-oxazine (3g). To a solution of phenylseleno amide **1g** (0.25 g, 0.80 mmol) in methanol (15 mL) at room temperature, powdered magnesium monoperoxyphthalate (0.95 g, 1.92 mmol) were added. The reaction mixture was stirred for 7 h. The mixture was then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 × 20 mL).

The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum. The resulting oil was purified by chromatography on SiO₂ (4:6 ethyl ether/petroleum ether) to afford **3h** as a colorless oil (0.09 g, 67%): ¹H NMR and ¹³C NMR spectra are in agreement to that previously reported [10]. ¹H NMR (200 MHz, CDCl₃) δ/ppm 7.98-7.80 (m, 2H, *H*-Ar), 7.53-7.26 (m, 3H, *H*-Ar), 4.35 (t, 2H, *J* = 5.5 Hz, OCH₂), 3.63 (t, 2H, *J* = 5.7 Hz, NCH₂), 1.99 (tt, 2H, *J* = 5.7, 5.5 Hz, CH₂). ¹³C NMR (50 MHz, CDCl₃) δ/ppm 155.6, 134.0, 130.3, 128.0 (2C), 126.8 (2C), 65.2, 42.6, 21.9.

5-Decyl-3-[(4-methylphenyl)sulfonyl]-1,3-oxazolidin-2-one (3h). To a solution of phenylseleno carbamate **1h** (0.30 g, 0.55 mmol) in THF (20 mL) at room temperature, powdered dipotassium hydrogen phosphate (0.48 g, 1.65 mmol) and magnesium monoperoxyphthalate (0.66 g, 1.33 mmol) were added. The reaction mixture was stirred until TLC analysis showed that the selenone intermediate was completely converted into the cyclized product (10 h). The mixture was then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 × 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum to afford **3h** as a colourless oil (0.22 g, 94%). The ¹H NMR and ¹³C NMR spectra are in agreement to that previously reported [5]. ¹H NMR (200 MHz, CDCl₃) δ/ppm 7.92 (d, 2H, *J* = 7.6 Hz, *H*-Ar), 7.37 (d, 2H, *J* = 7.6 Hz, *H*-Ar), 4.60-4.40 (m, 1H, CH), 4.09 (dd, 1H, *J* = 9.0, 7.9 Hz, CH₂), 3.57 (dd, 1H, *J* = 9.0, 7.2 Hz, CH₂), 2.41 (s, 3H, CH₃), 1.80-1.10 (m, 18H, CH₂), 0.88 (t, 3H, *J* = 6.6 Hz, CH₃). ¹³C NMR (50 MHz, CDCl₃) δ/ppm 151.6, 145.5, 133.8, 129.7 (2 C), 128.0 (2 C), 74.6, 49.5, 34.0, 31.7, 29.3, 29.2, 29.1 (2 C), 28.9, 24.2, 22.5, 21.5, 13.9.

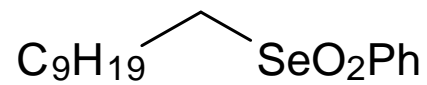
3-Benzoyl-5-[(benzyloxy)methyl]-1,3-oxazolidin-2-one (3i). To a solution of phenylseleno carbamate **1i** (0.469 g, 1.00 mmol) in THF (25 mL) at room temperature, powdered dipotassium hydrogen phosphate (0.522 g, 3.00 mmol) and magnesium monoperoxyphthalate (1.19 g, 2.40 mmol) were added. The reaction mixture was stirred until TLC analysis showed that the starting selenide was completely converted into the corresponding selenone (2 h) and then concentrated in vacuum. The residue was suspended in acetone (20 mL) and powdered potassium carbonate was

added (0.69 g, 5.00 mmol). The consumption of the selenone was monitored by TLC (4 h). The mixture was then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 × 20 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum to afford solid **3i** (0.25 g, 82%): mp 92-94 °C (from CH₂Cl₂) (lit. [8], 93-94 °C). ¹H NMR and ¹³C NMR spectra are identical to that previously reported [8]. ¹H NMR (200 MHz, CDCl₃) δ/ppm 7.60-7.43 (m, 3H, *H*-Ar), 7.42-7.28 (m, 7H, *H*-Ar), 4.75 (ddt, 1H, *J* = 8.5, 5.8, 3.5 Hz, OCH), 4.59 (m, 2H, CH₂), 4.16 (dd, 1H, *J* = 10.8, 8.5 Hz, CH₂), 4.04 (dd, 1H, *J* = 10.8, 5.8 Hz, CH₂), 3.77 (dd, 1H, *J* = 10.8, 3.5 Hz, CH₂), 3.65 (dd, 1H, *J* = 10.8, 3.5 Hz, CH₂). ¹³C NMR (50 MHz, CDCl₃) δ/ppm 169.4, 152.6, 137.1, 132.7, 131.9 (2 C), 128.6 (3 C), 128.5 (2 C), 127.8, 127.6 (2 C), 73.5, 72.4, 69.7, 45.3.

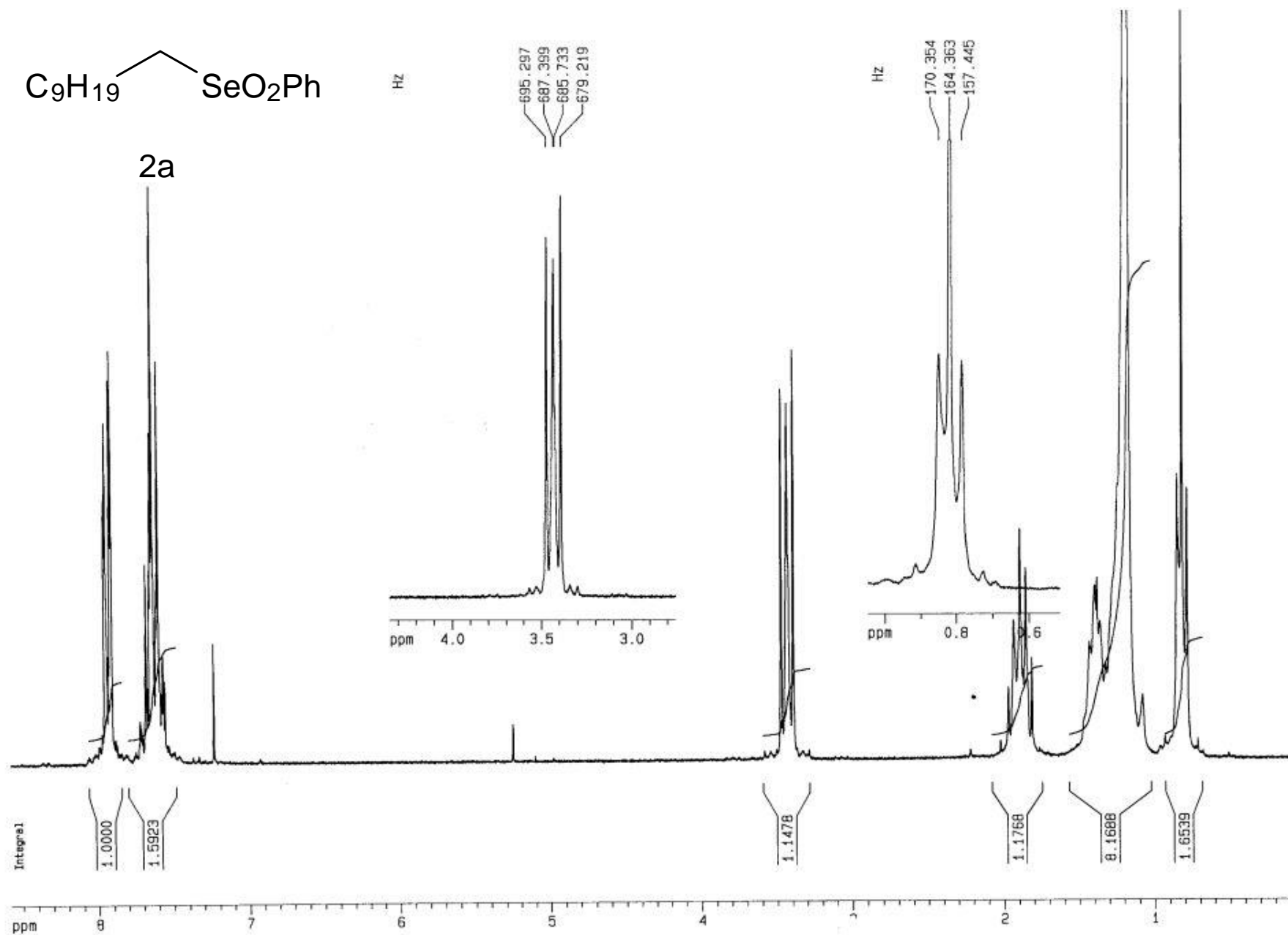
2-(3-Fluorophenyl)tetrahydrofuran (3j); To a solution of phenylseleno alcohol **1j** (0.06 g, 0.02 mmol) in methanol (8 mL) at room temperature, powdered magnesium monoperoxyphthalate (0.25 g, 0.50 mmol) was added. The reaction mixture was stirred for 7 h and then poured into 10% aqueous sodium carbonate (10 mL) and extracted with ethyl acetate (2 × 10 mL). The organic layer was washed with 10% aqueous sodium carbonate (10 mL), brine (15 mL), dried with sodium sulfate and concentrated in vacuum to afford **3j** as a colorless oil (0.02 g, 69%). The ¹H NMR and ¹³C NMR spectra are in agreement to that previously reported [6]. ¹H NMR (200 MHz, CDCl₃) δ/ppm . 7.30-7.05 (m, 1H, *H*-Ar), 7.05-6.73 (m, 3H, *H*-Ar), 4.90 (t, 1H, *J* = 7.1 Hz, OCH), 4.09 (ddd, 1H, *J* = 15.1, 8.2, 1.5 Hz, OCH₂), 3.91 (ddd, 1H, *J* = 15.1, 8.2, 1.5 Hz, OCH₂), 2.38-2.25 (m, 1H, CH₂), 2.05-1.70 (m, 3H, CH₂). ¹³C NMR (50 MHz, CDCl₃) δ/ppm 162.8 (d, *J*_{CF} = 243.0 Hz), 144.0 (d, *J*_{CF} = 5.5 Hz), 129.7 (d, *J*_{CF} = 8.2 Hz), 121.1 (d, *J*_{CF} = 2.7 Hz) 113.8 (d, *J*_{CF} = 21.2 Hz), 112.4 (d, *J*_{CF} = 22.0 Hz), 79.9, 68.7, 34.6, 25.8.

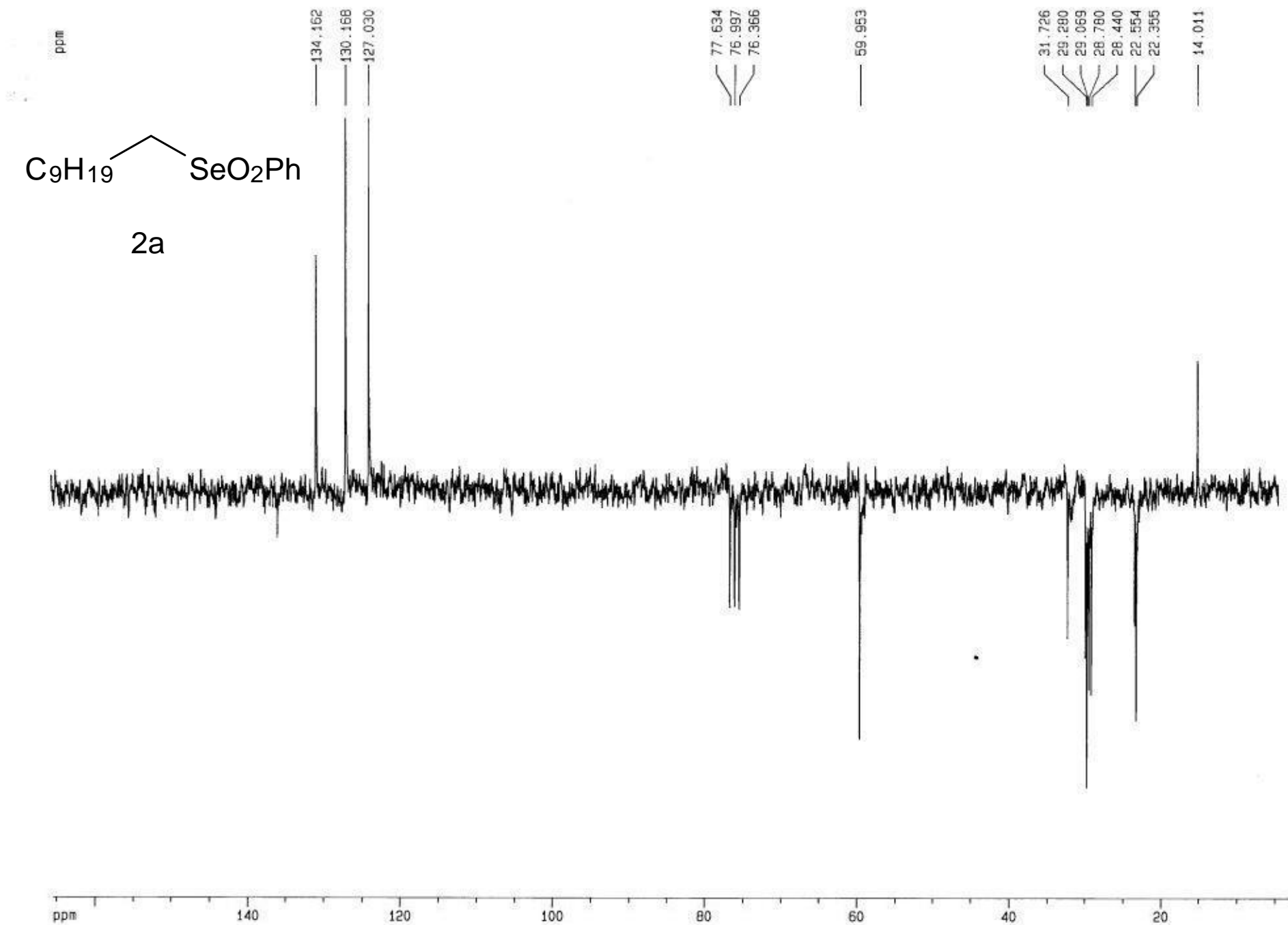
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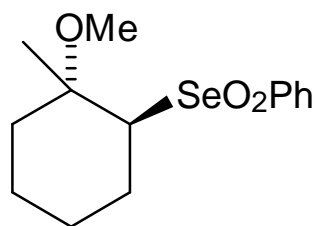
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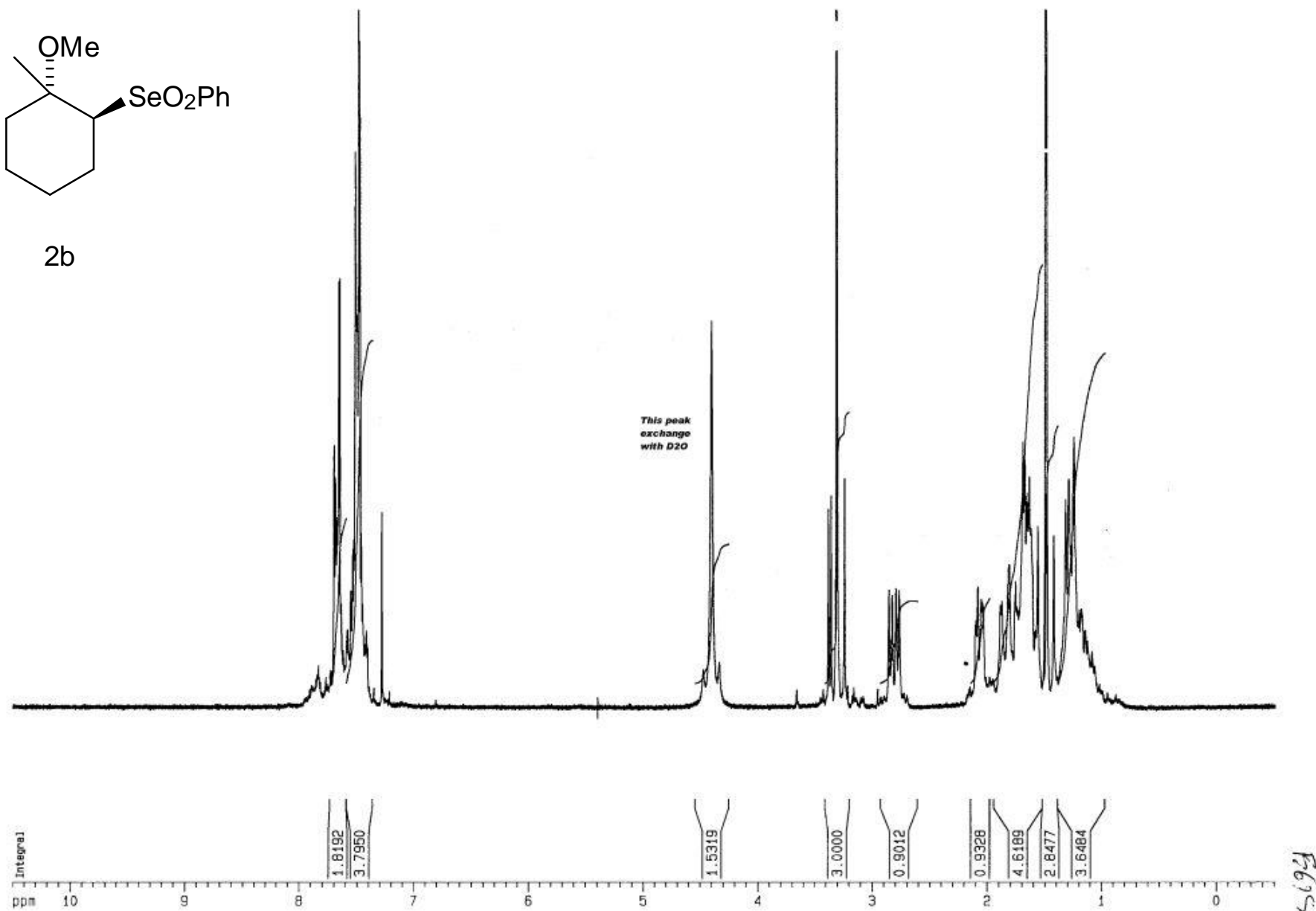
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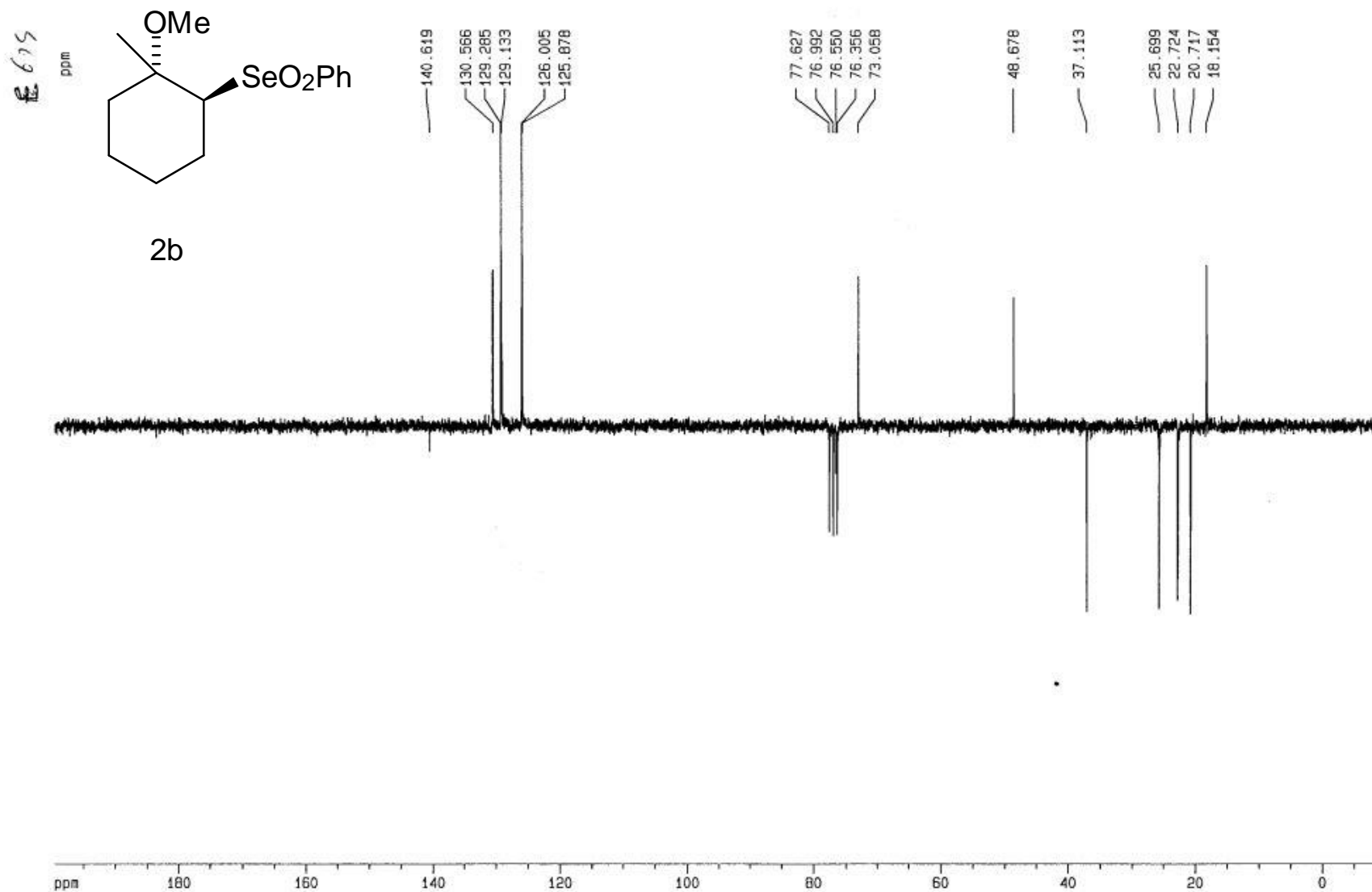




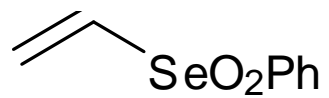


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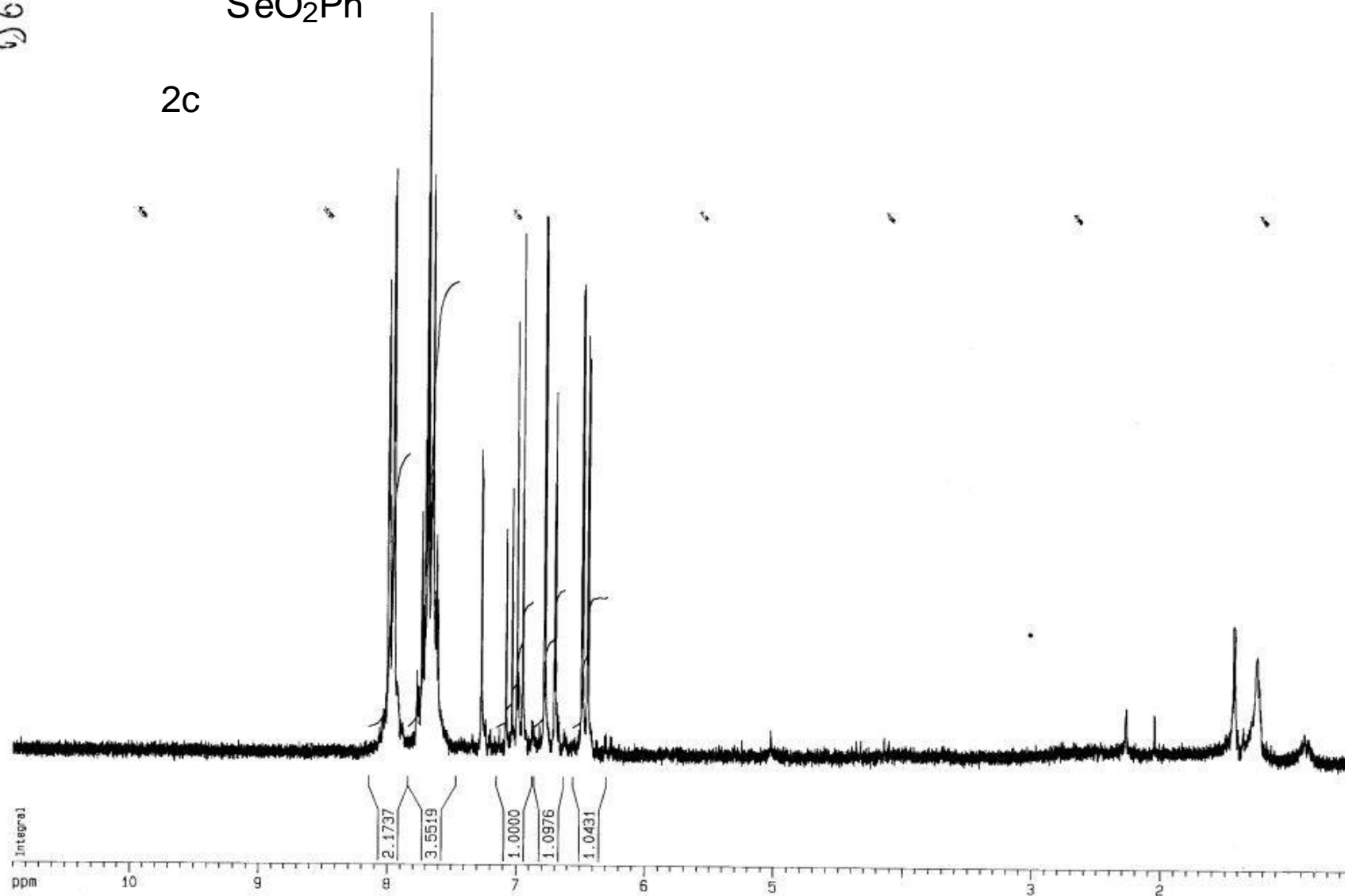




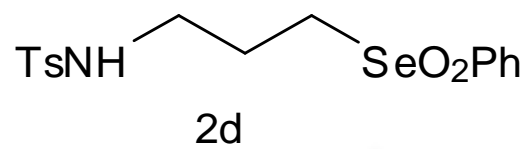
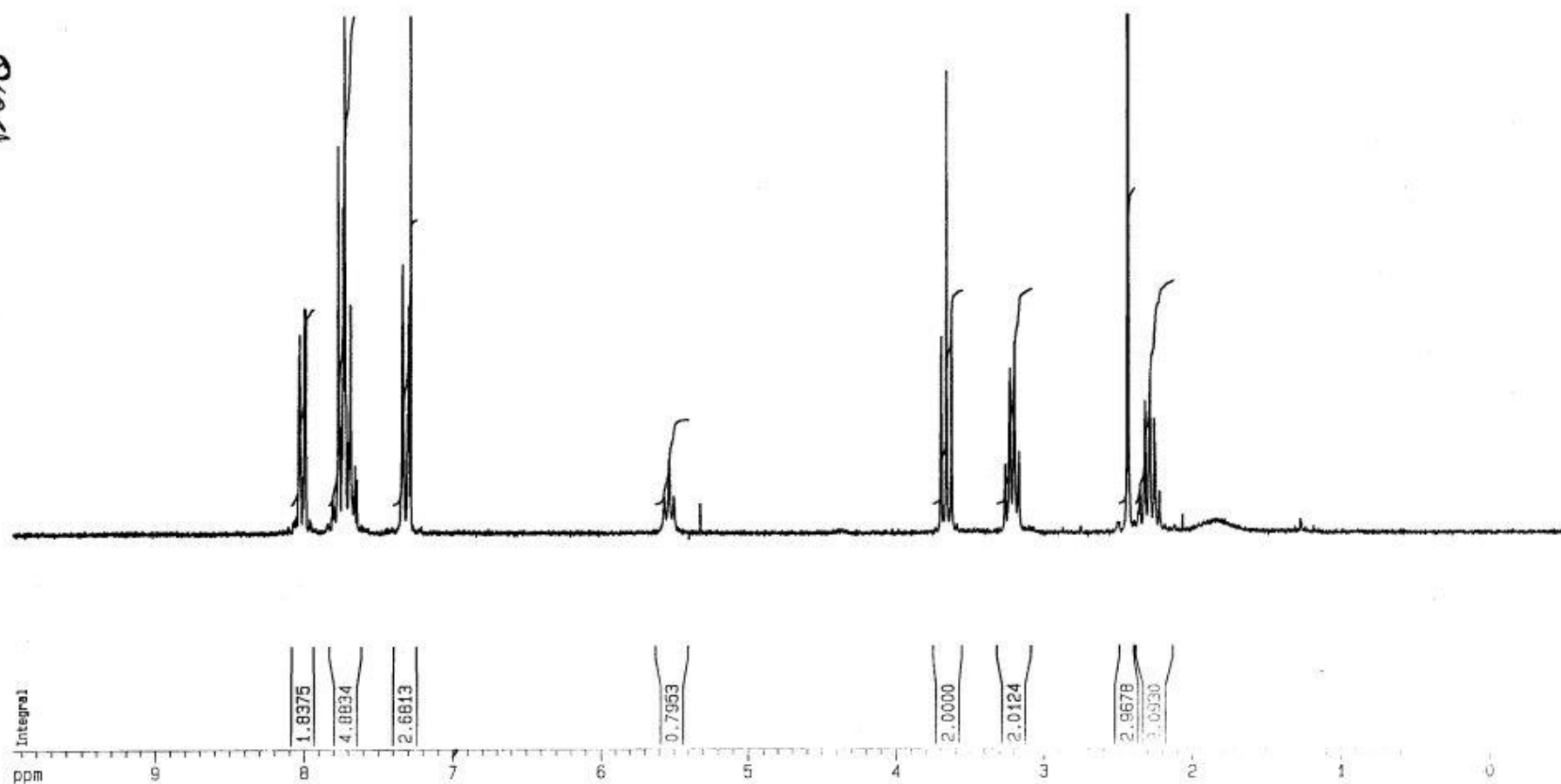
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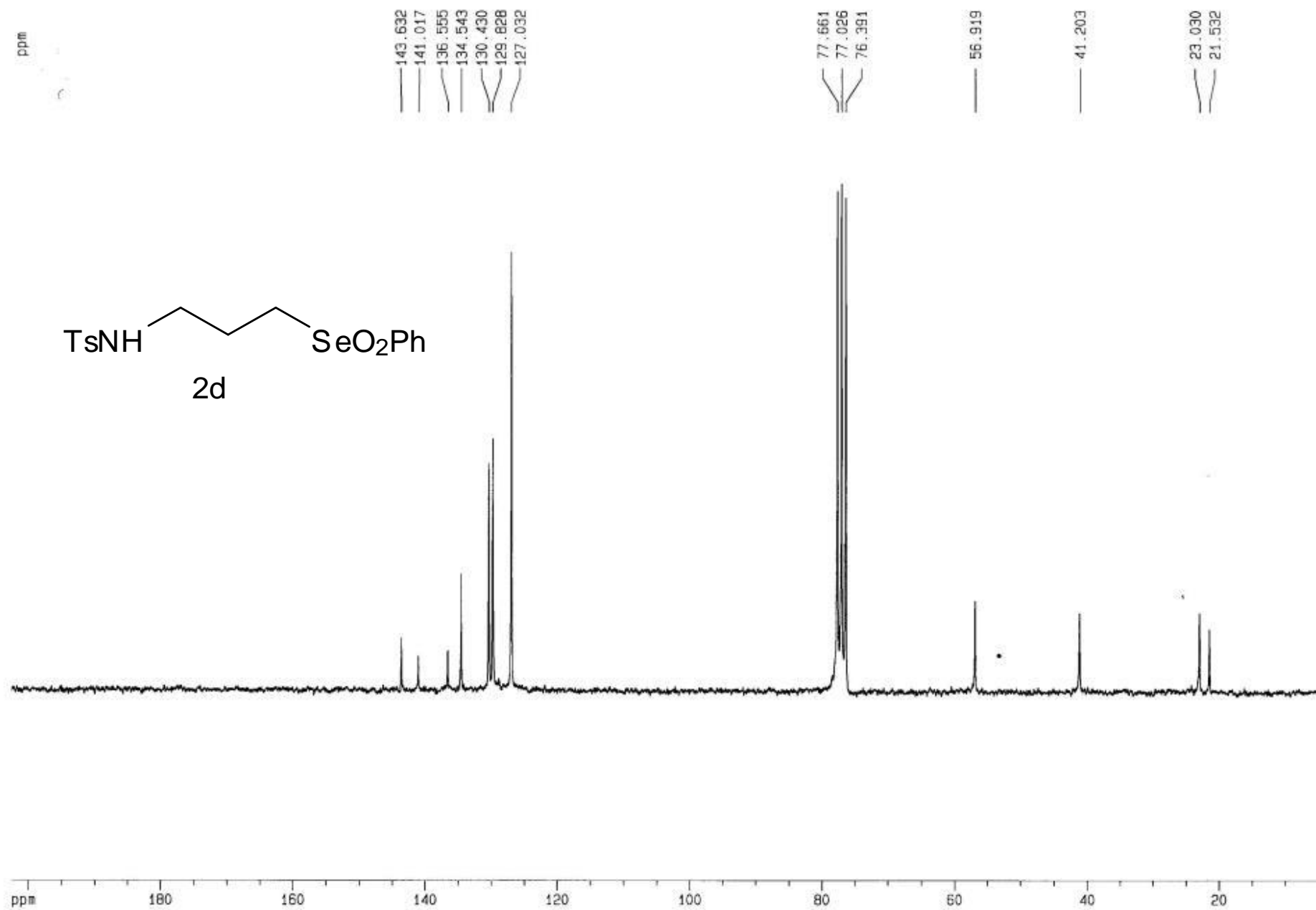


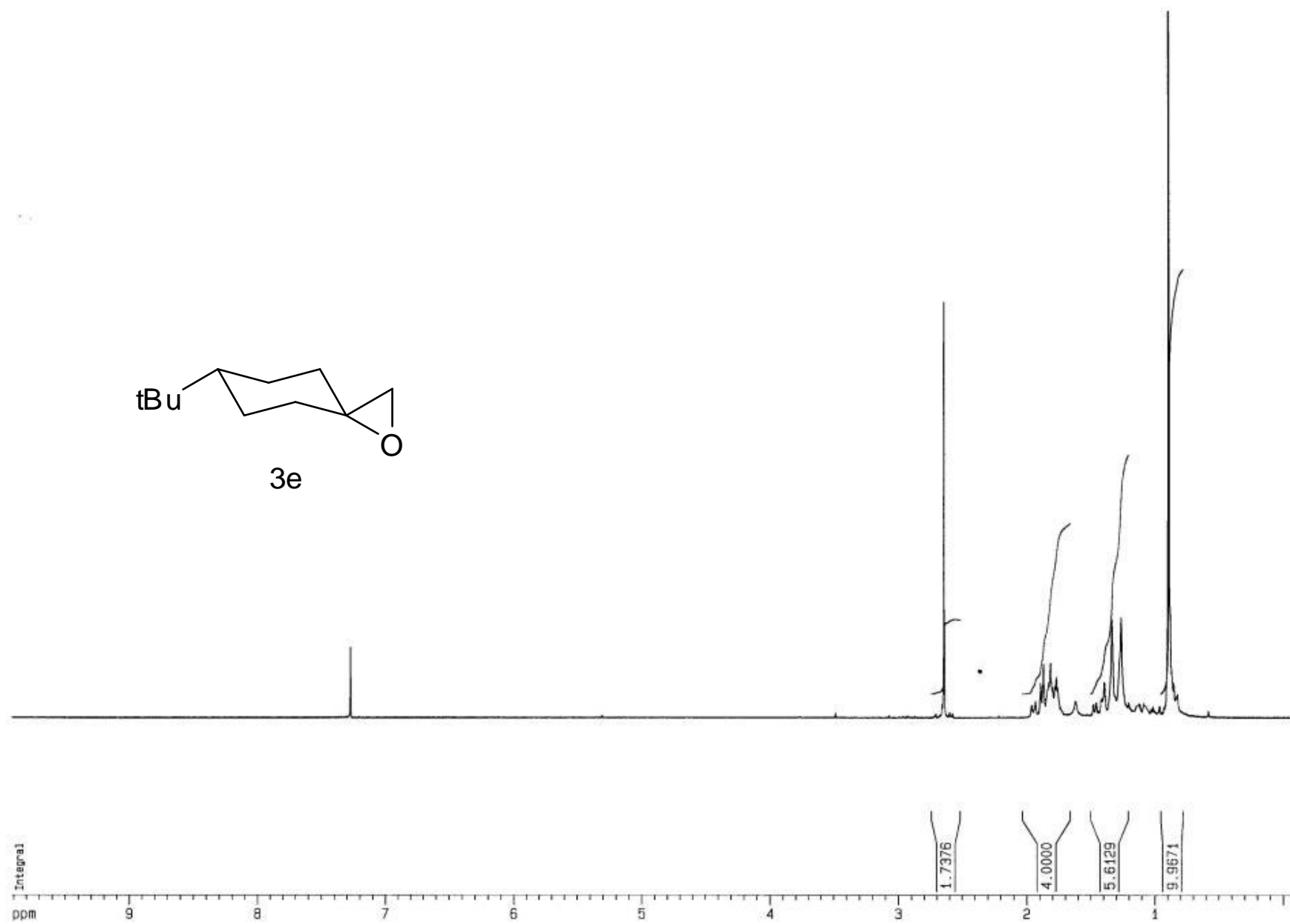
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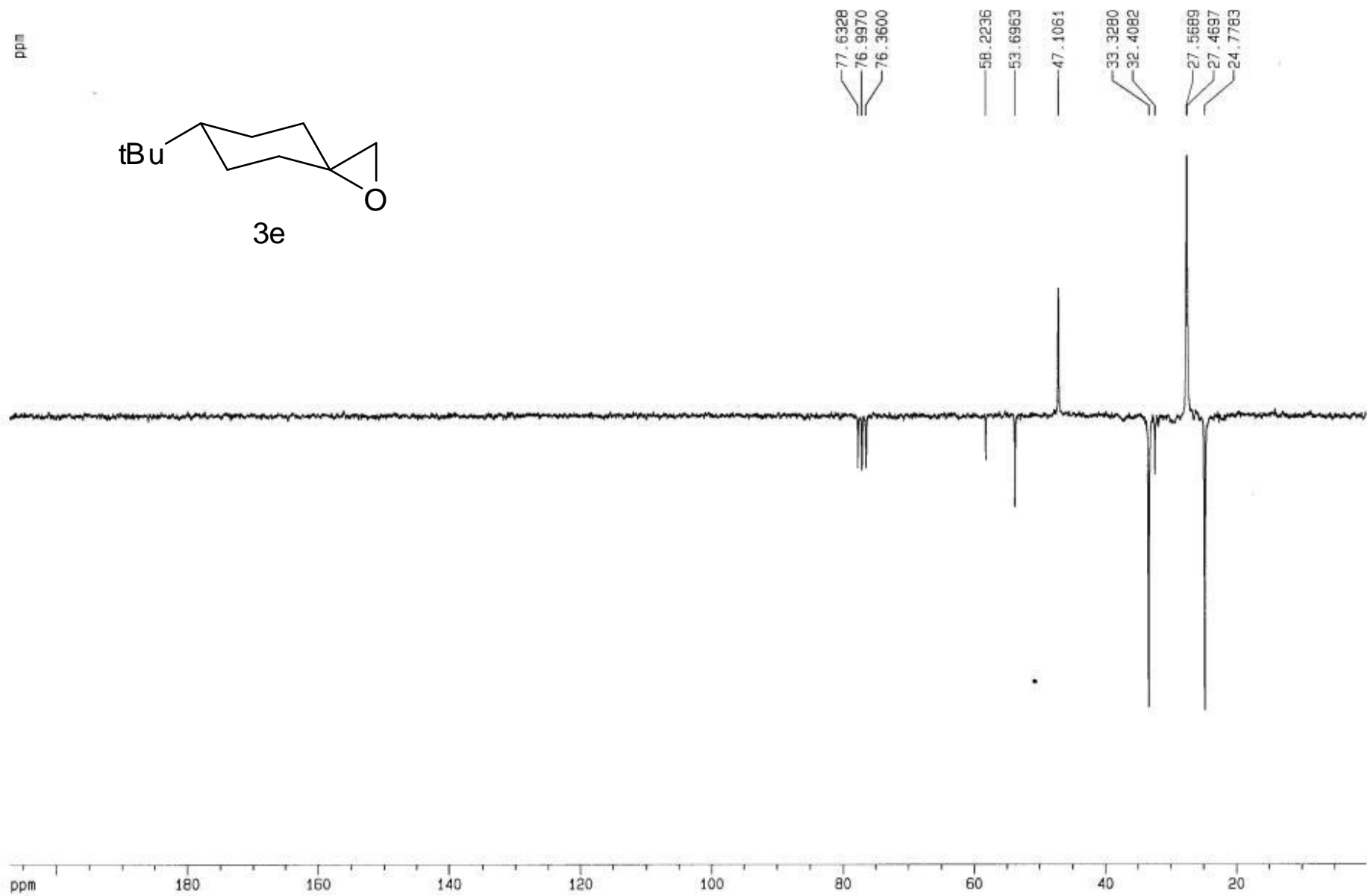


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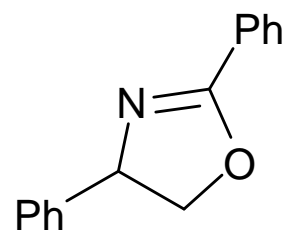




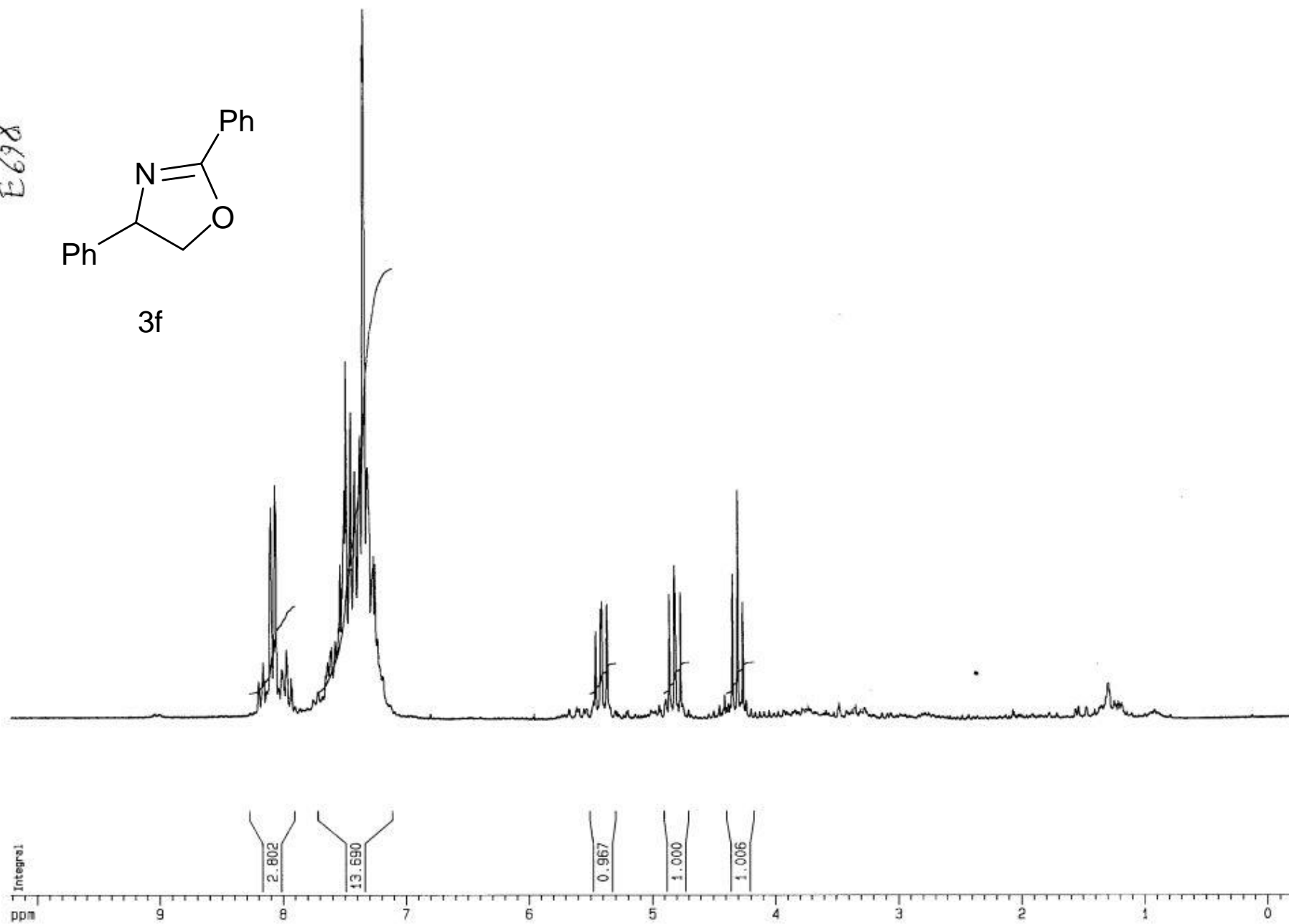


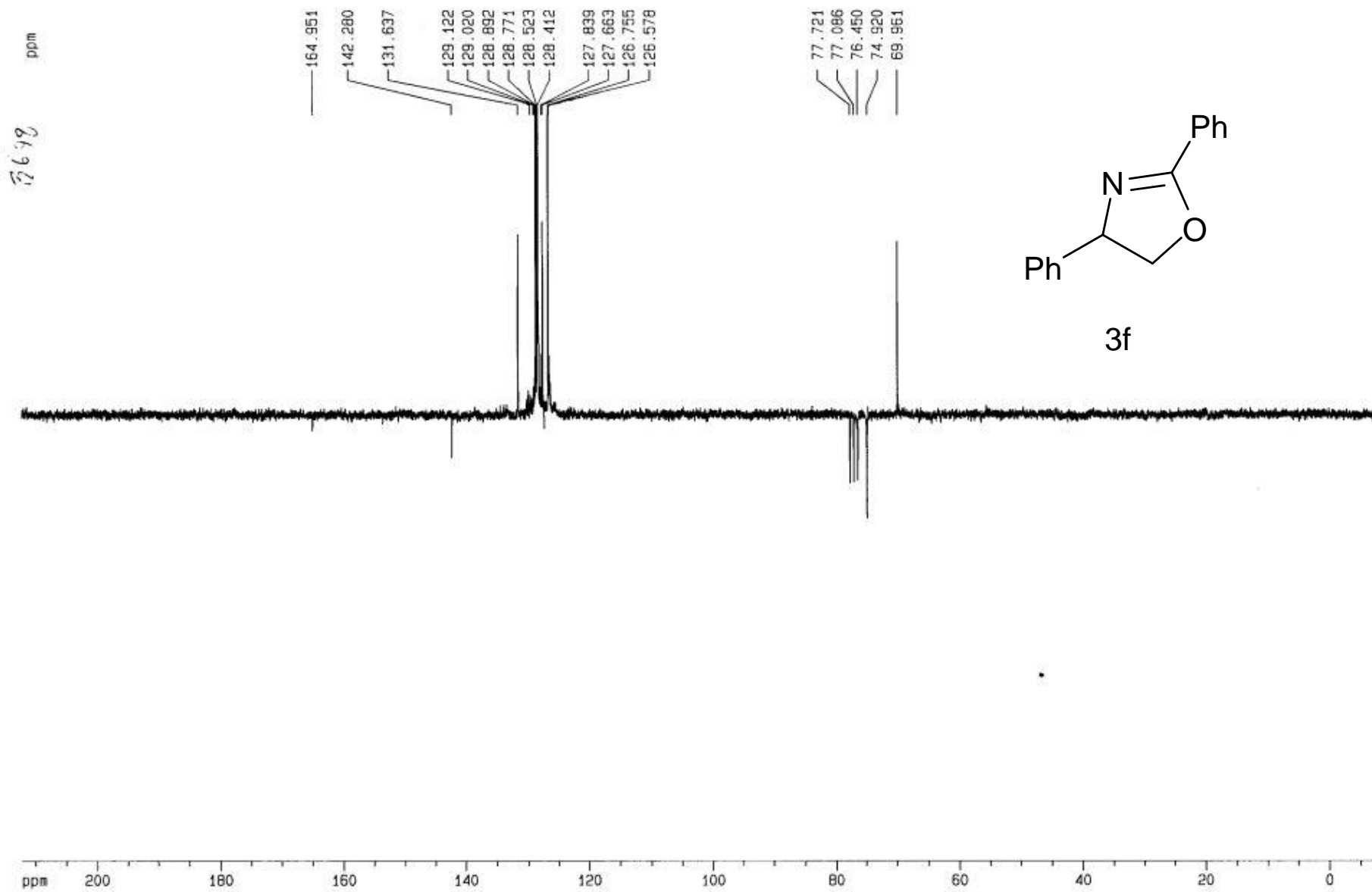


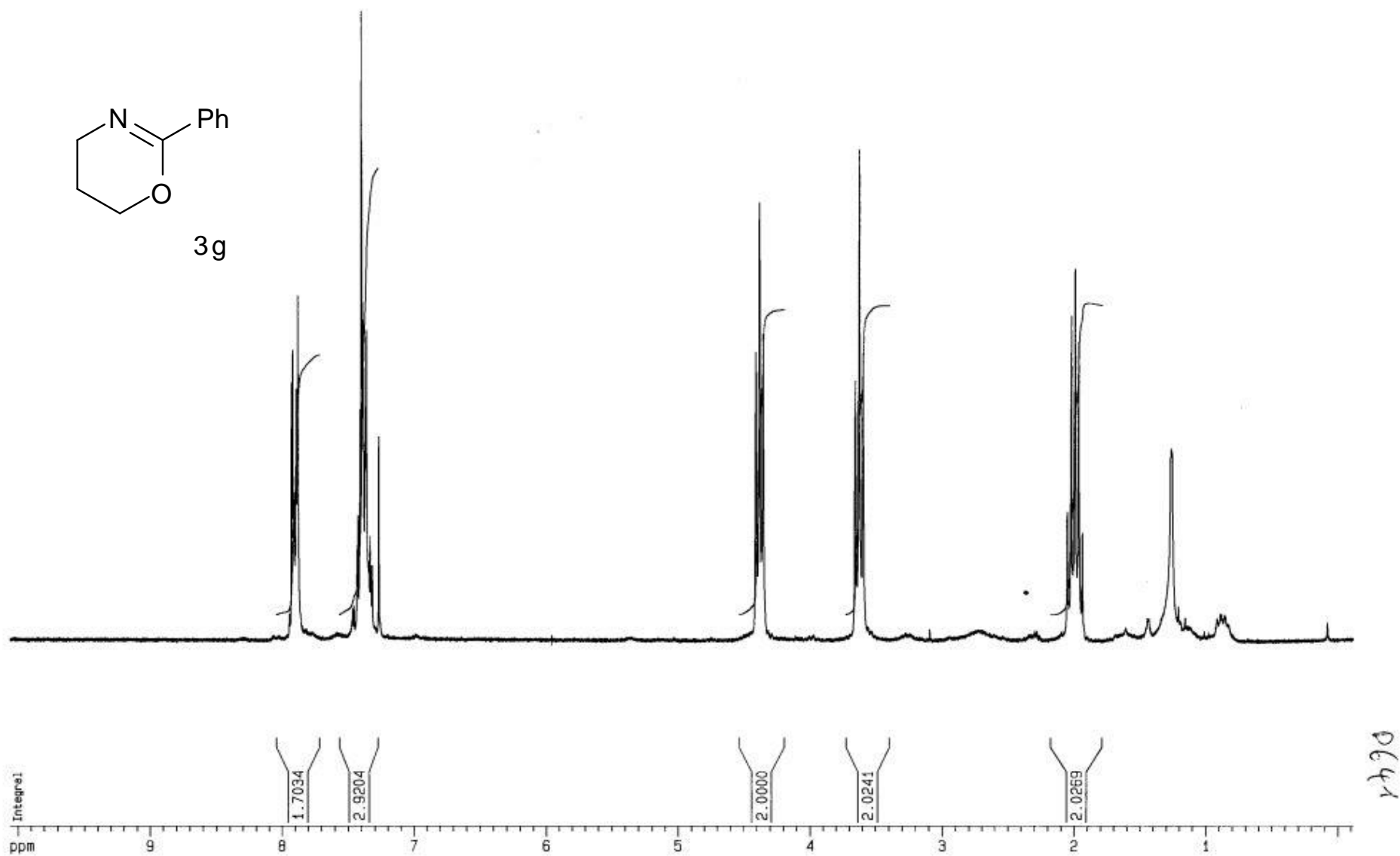
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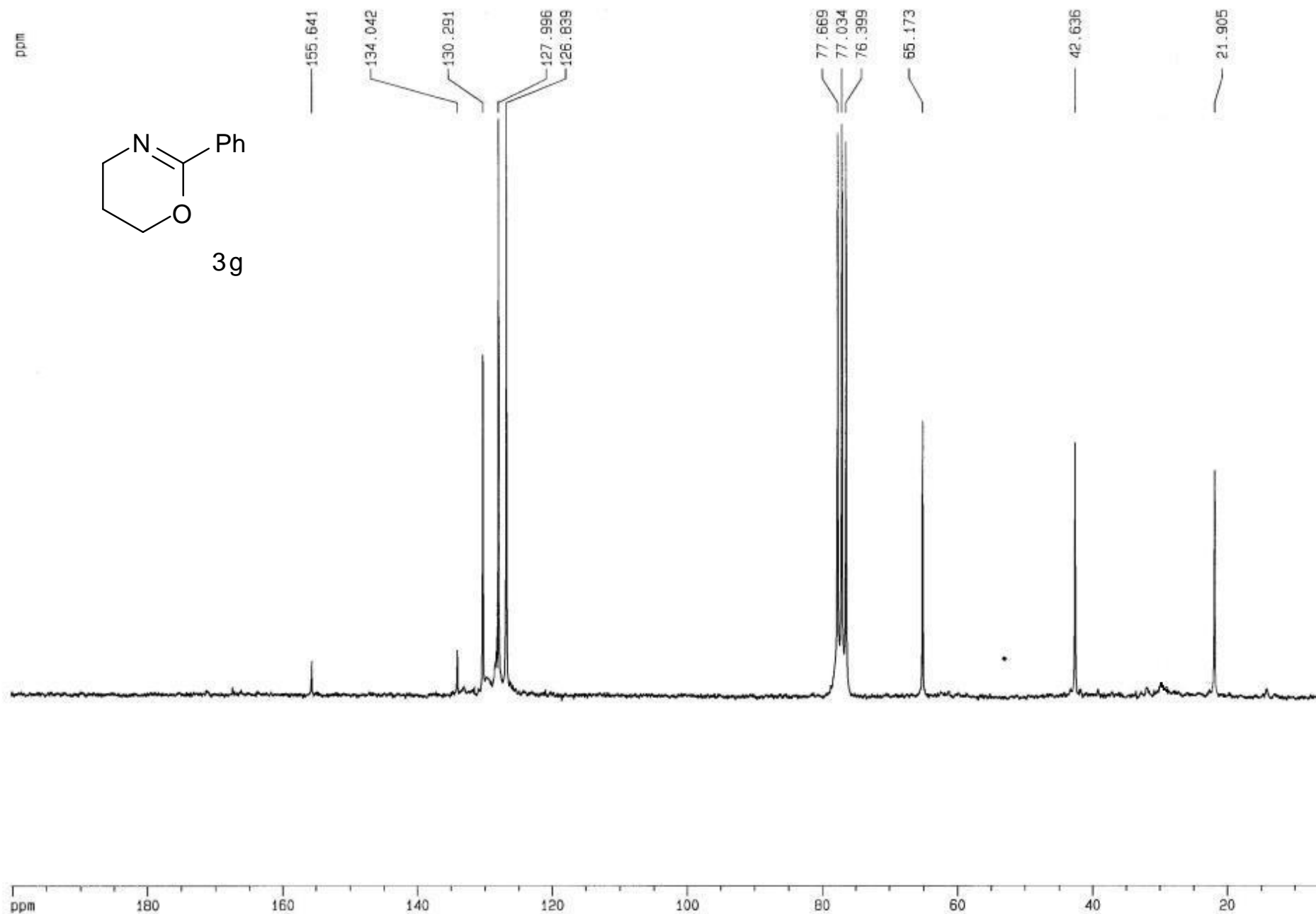


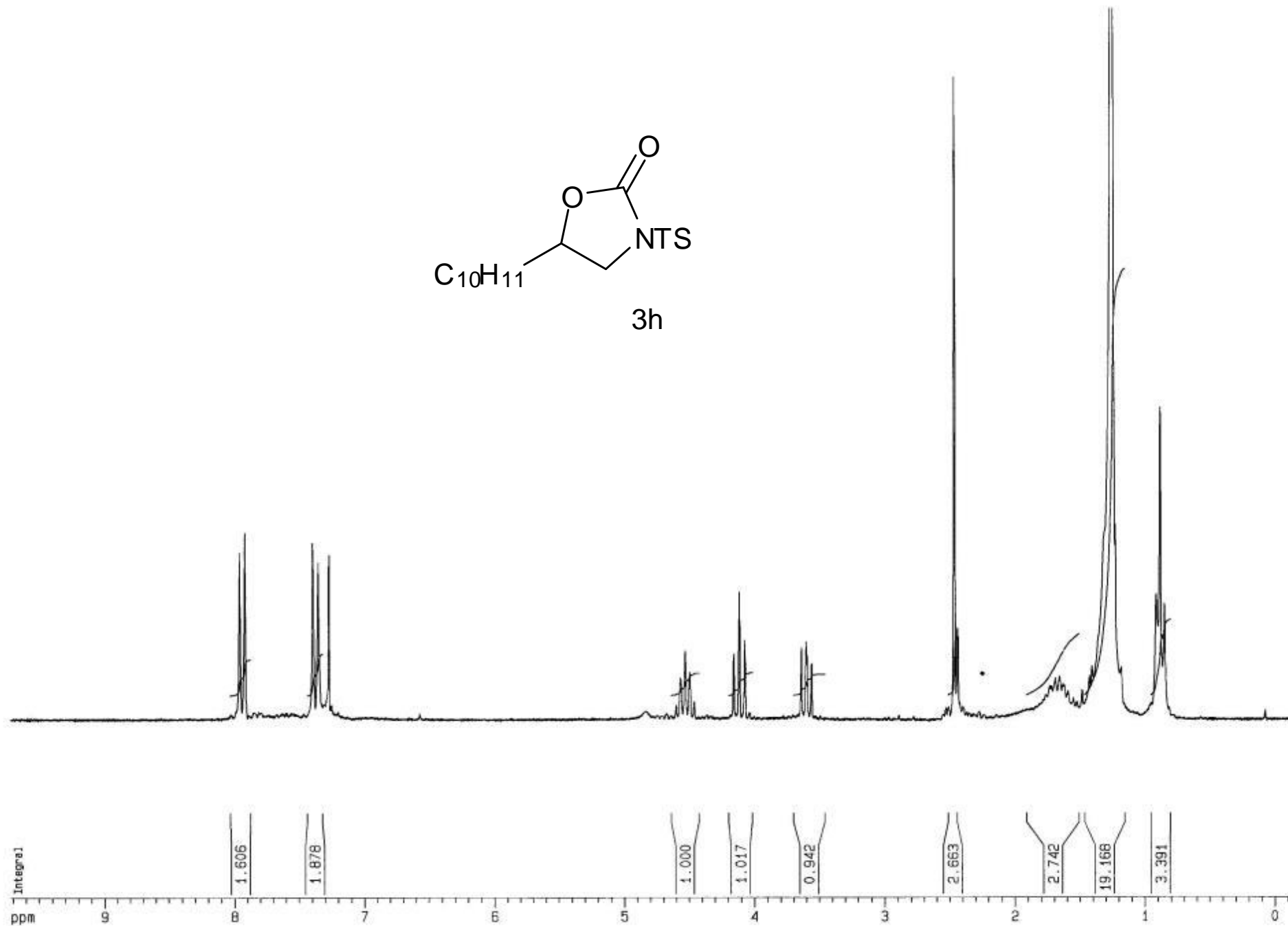
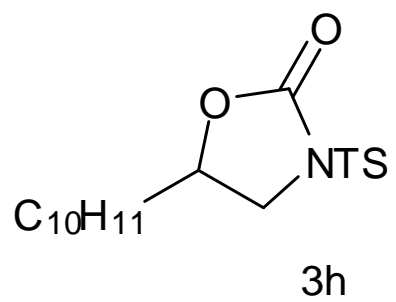
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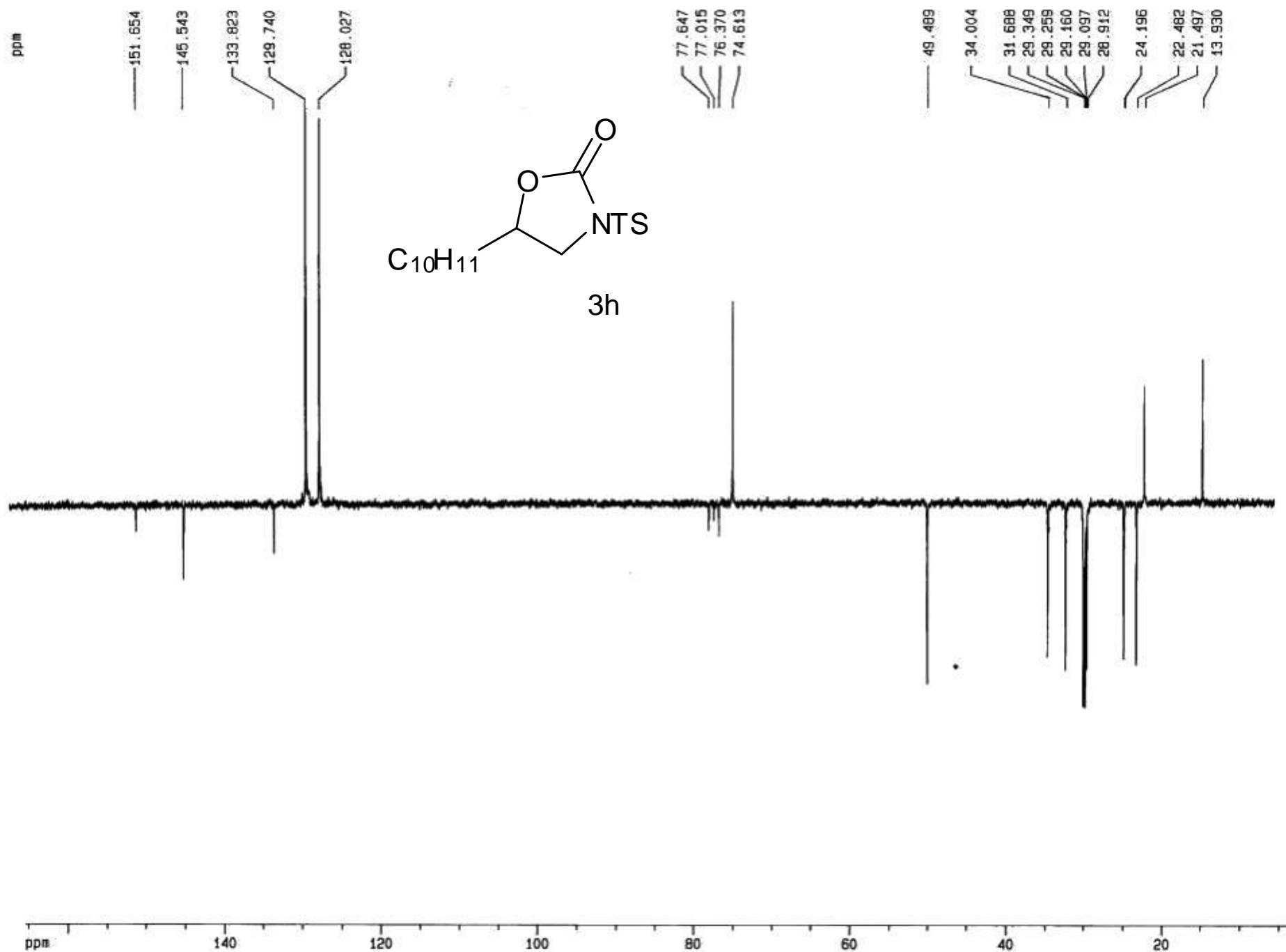


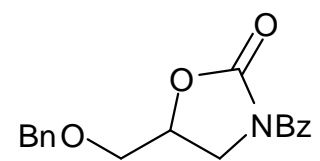




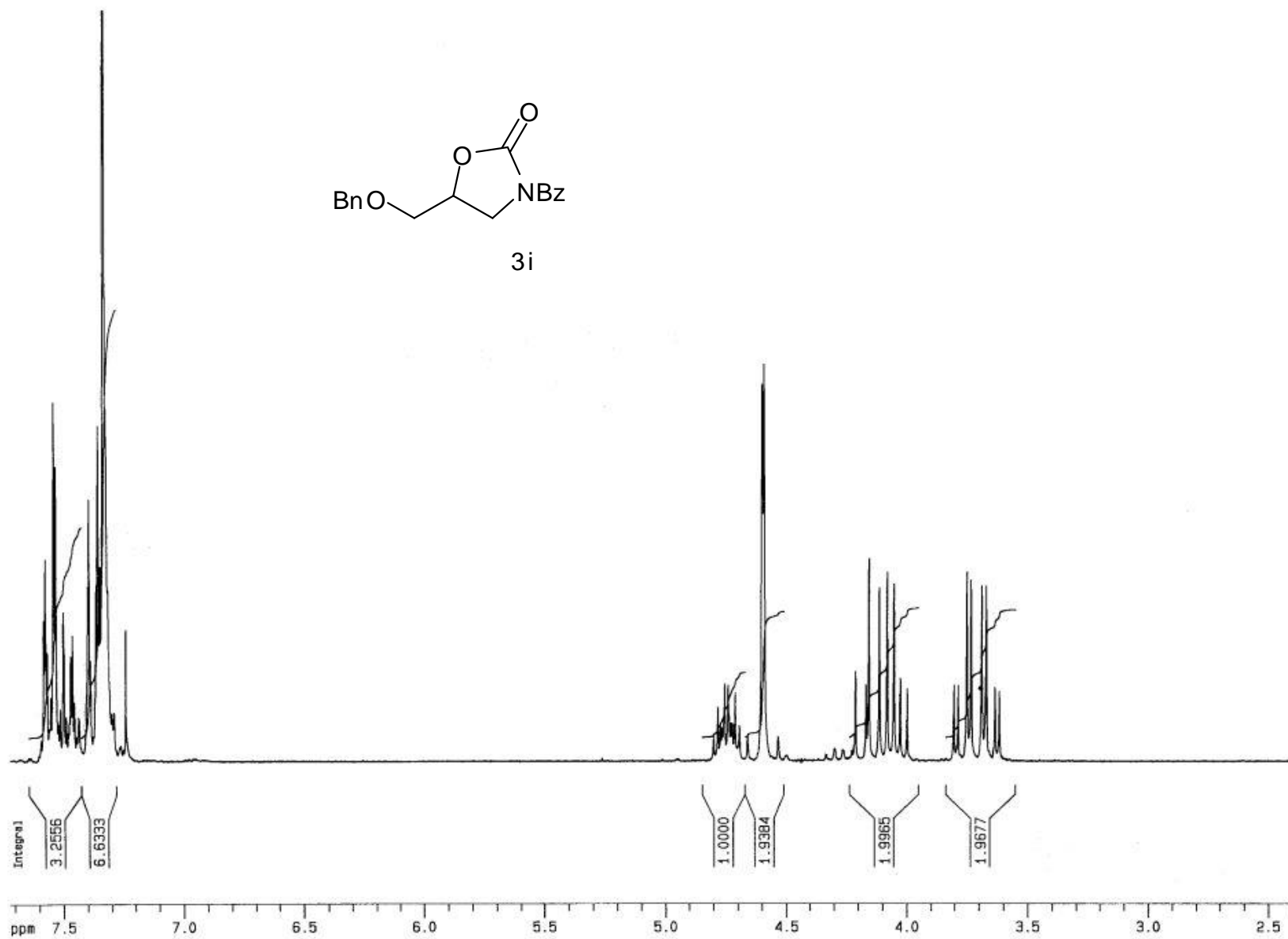


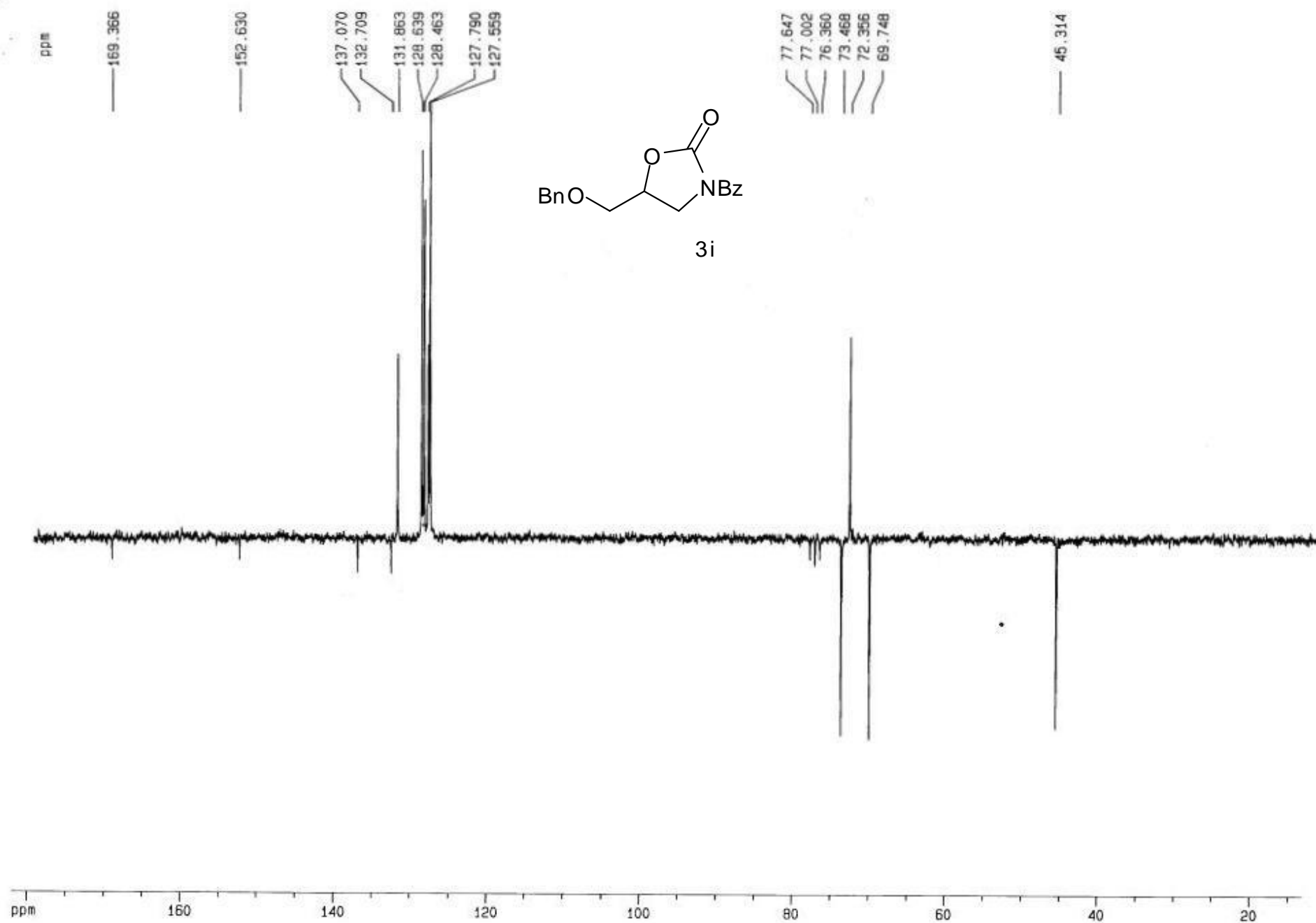
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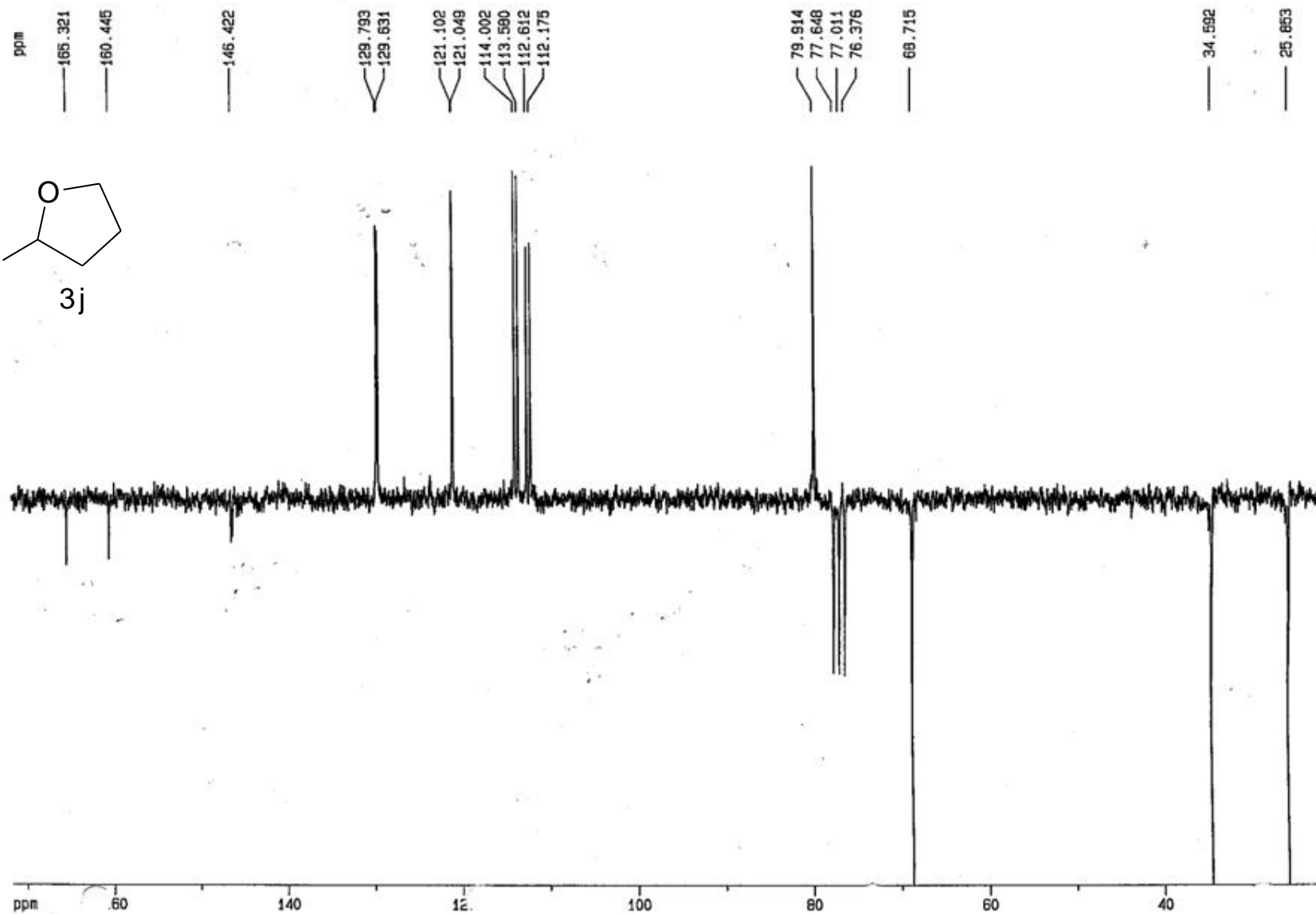
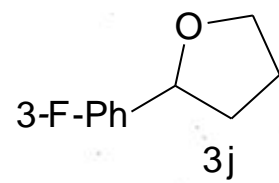




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