Electronic Supplementary Material (ESI) for ChemComm.

Supporting Online Material for

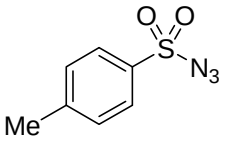
N-Sulfonyl acetylketeneimine as a highly reactive intermediate for the synthesis of *N*-sulfonyl amidines

Weiguang Yang, Dayun Huang, Xiaobao Zeng, Dongping Luo, Xinyan Wang,*

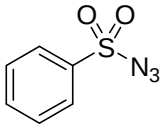
Yuefei Hu*

Key Laboratory of Bioorganic Phosphorus Chemistry and Chemical Biology (Ministry of Education), Department of Chemistry, Tsinghua University, Beijing 100084, P. R. China

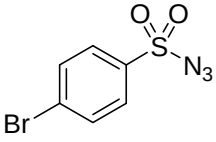
Structures of Starting Materials 2a-2h



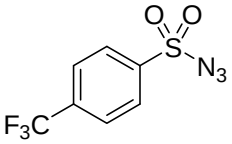
2a



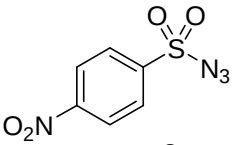
2b



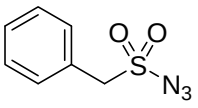
2c



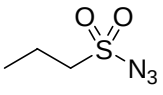
2d



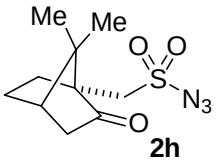
2e



2f

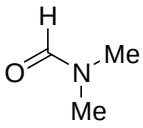


2g

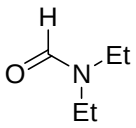


2h

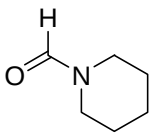
Structures of Starting Materials 10a-10h



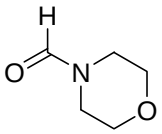
10a



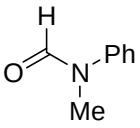
10b



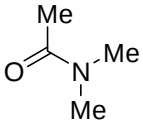
10c



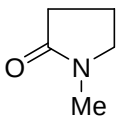
10d



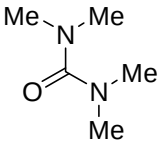
10e



10f



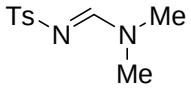
10g



10h

Experimental Section

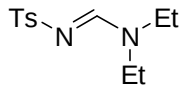
General Methods: All melting points were determined on a Yanaco melting point apparatus and are uncorrected. IR spectra were recorded as KBr pellets on a Nicolet FT-IR 5DX spectrometer. The ^{1}H NMR (300 or 400 MHz) and ^{13}C NMR (75 or 100 MHz) spectra were recorded on a JEOL JNM-ECA spectrometers 300 or 400 in $CDCl_3$. TMS was used as an internal reference and *J* values are given in Hz. HRMS were obtained on a Bruker micrOTOF-Q II spectrometer. PE is petroleum ether (60−90 ^{o}C). All starting materials **2a-2h** and **10a-10h** are known compounds. The compounds **2a-2c** and **10a-10h** were purchased directly and **2d-2h** were easily prepared according to the reported procedure (Pi, C; Cui, X.; Wu, Y. *J. Org. Chem.* **2015**, *80*, 7333.).



12a

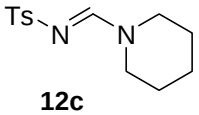
A typical procedure for preparation of *N*-[(dimethylamino)methylene]-4-methylbenzenesulfonamide (12a). To a stirred solution of 3-butyn-2-one (**9a**, 136 mg, 2 mmol) and CuI (19 mg, 0.1 mmol) in DMF (**10a**, 2 mL) was added TsN_3 (**2a**, 197 mg, 1 mmol). After the mixture was heated under 60 ^{o}C for 15 min, it was cooled to room temperature. The system was extracted with EtOAc and washed with brine. After the organic layer was dried over $MgSO_4$, the solvent was evaporated on rotavapor. The residue was purified by a flash chromatography (silica gel, petroleum ether/ethyl acetate = 1:1) to give the pure product **12a** (219 mg, 97%) as white solid, mp 135–136 ^{o}C (lit.[1] mp 132–133 °C). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 (s, 1H), 7.78 (d, *J* = 8.2 Hz, 2H), 7.26 (d, *J* = 8.2 Hz, 2H), 3.12 (s, 3H), 3.01 (s, 3H), 2,40 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 159.0, 142.4, 139.5, 129.3 (2C), 126.5 (2C), 41.4, 35.5, 21.5.

The products **12b-12t** were prepared by the similar procedure.



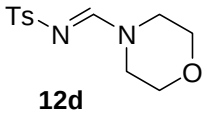
12b

***N*-[(Diethylamino)methylene]-4-methylsulfonamide (12b).** 234 mg (92%), white solid, mp 76–77 °C (lit.[1] mp 77–78 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 (s, 1H), 7.76 (d, J = 8.2 Hz, 2H), 7.25 (d, J = 8.2 Hz, 2H), 3.47 (q, J = 7.3 Hz, 2H), 3.38 (d, J = 7.3 Hz, 2H), 2.40 (s, 3H), 1.25 (t, J = 7.3 Hz, 3H), 1.14 (t, J = 7.3 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.0, 142.2, 139.7, 129.2 (2C), 126.3 (2C), 47.0, 40.8, 21.4, 14.5, 12.0.



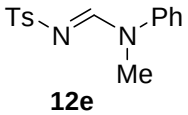
12c

***N*-(1-Piperidinylmethylene]-4-methylsulfonamide (12c).** 253 mg (95%), white solid, mp 150–151 °C (lit.[1] mp 147–149 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12 (s, 1H), 7.76 (d, J = 8.2 Hz, 2H), 7.25 (d, J = 8.2 Hz, 2H), 3.57 (t, J = 5.5 Hz, 2H), 3.40 (t, J = 5.5 Hz, 2H), 2.38 (s, 3H), 1.65–1.53 (m, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.0, 142.0, 139.5, 129.0 (2C), 126.1 (2C), 51.6, 44.3, 26.1, 24.6, 23.6, 21.2.



12d

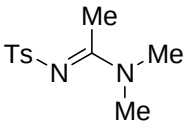
***N*-(4-Morpholinylmethylene)-4-methylsulfonamide (12d).** 188 mg (70%), white solid, mp 171–172 °C (lit.[1] mp 176–177 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.20 (s, 1H), 7.77 (d, J = 7.8 Hz, 2H), 7.27 (d, J = 7.8 Hz, 2H), 3.74 (t, J = 5.0 Hz, 2H), 3.67 (s, 4H), 3.49 (d, J = 5.0 Hz, 2H), 2.40 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.6, 142.7, 139.0, 129.4 (2C), 126.5 (2C), 66.8, 65.9, 50.3, 44.2, 21.5.



12e

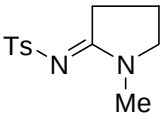
***N*-[(*N*-Methyl-*N*-phenylamino)methylene]-4-methylsulfonamide (12e).** 104

mg (36%), white solid, mp 117–118 °C (lit.[2] mp 108 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.56 (s, 1H), 7.82 (d, J = 8.3 Hz, 2H), 7.42 (t, J = 7.8 Hz, 2H), 7.33–7.28 (m, 3H), 7.19 (d, J = 8.2 Hz, 2H), 3.43 (s, 3H), 2.41 (s, 3H); ^{13}C NMR (100 MHz) δ 158.2, 143.0, 142.7, 138.8, 129.7 (2C), 129.3 (2C), 127.2, 126.6 (2C), 121.9 (2C), 35.9, 21.4.



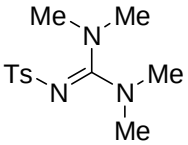
12f

***N*-[(Dimethylamino)ethylidene]-4-methylsulfonamide (12f).** 125 mg (52%), white solid, mp 123–124 °C, (lit.[3] mp 122–123 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.82 (d, J = 8.2 Hz, 2H), 7.25 (d, J = 8.2 Hz, 2H), 3.08 (s, 3H), 3.07 (s, 3H), 2.48 (s, 3H), 2.39 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 165.8, 141.7, 141.1, 129.0 (2C), 126.1 (2C), 38.8, 38.7, 21.3, 17.8.



12g

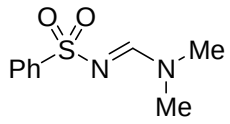
***N*-(1-Methylpyrrolidin-2-ylidene)-4-methylsulfonamide (12g).** 174 mg (69%), white solid, mp 127–128 °C (lit.[4] mp 145 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.81 (d, J = 8.3 Hz, 2H), 7.25 (d, J = 8.3 Hz, 2H), 3.45 (t, J = 7.3, Hz, 2H), 3.03 (t, J = 8.2, Hz, 2H), 2.97 (s, 3H), 2.40 (s, 3H), 2.08–2.01 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 169.8, 141.9, 140.5, 129.0 (2C), 126.4 (2C), 51.6, 31.9, 30.6, 21.3, 18.9.



12h

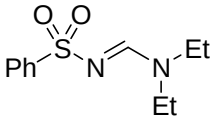
***N*-[Bis(dimethylamino)methylene]-4-methylsulfonamide (12h).** 204 mg (76%), yellow solid, mp 139–140 °C (lit.[5] mp 142–144 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.81 (d, J = 8.2 Hz, 2H), 7.22 (d, J = 7.8 Hz, 2H), 2.93 (s, 12H), 2.38 (s, 3H); ^{13}C

NMR (100 MHz, $CDCl_3$) δ 161.8, 143.1, 140.9, 128.9 (2C), 125.5 (2C), 40.4 (4C), 21.3.



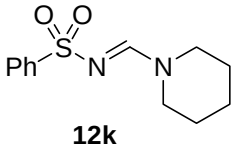
12i

***N*-[(Dimethylamino)methylene]benzenesulfonamide (12i).** 197 mg (93%), white solid, mp 130–131 °C (lit.[6] mp 128–129 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.15 (s, 1H), 7.89 (d, J = 7.6 Hz, 2H), 7.50-7.44 (m, 3H), 3.12 (s, 3H), 3.01 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 159.1, 142.3, 131.7, 128.6 (2C), 126.2 (2C), 41.4, 35.4.



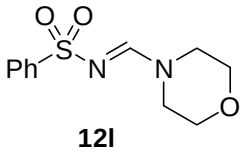
12j

***N*-[(Diethylamino)methylene]-benzenesulfonamide (12j).** 199 mg (83%), white solid, mp 75–76 °C (lit.[1] mp 73–75 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.16 (s, 1H), 7.88 (d, J = 7.2 Hz, 2H), 7.52–7.31 (m, 3H), 3.47 (q, J = 7.3 Hz, 2H), 3.38 (q, J = 6.9 Hz, 2H), 1.24 (t, J = 7.3 Hz, 3H), 1.13 (t, J = 6.9 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.0, 142.5, 131.5, 128.5 (2C), 126.1 (2C), 46.9, 40.8, 14.3, 11.9.



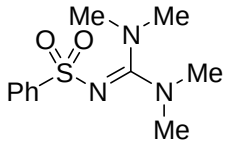
12k

***N*-(1-Piperidinylmethylene)-benzenesulfonamide (12k).** 229 mg (91%), white solid, mp 149–150 °C (lit.[7] no mp was reported); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 (s, 1H), 7.89 (d, J = 7.6 Hz, 2H), 7.51–7.44 (m, 3H), 3.60 (t, J = 5.5 Hz, 2H), 3.42 (d, J = 5.0 Hz, 2H), 1.67–1.66 (m, 4H), 1.59–1.57 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.3, 142.5, 131.6, 128.6 (2C), 126.3 (2C), 51.8, 44.6, 26.3, 24.7, 23.8.



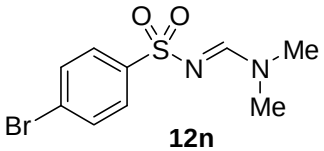
12l

***N*-(4-Morpholinylmethylene)-benzenesulfonamide (12l).** 191 mg (75%), white solid, mp 137–138 °C (lit.[7] no mp was reported); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.24 (s, 1H), 7.89 (d, J = 7.4 Hz, 2H), 7.55–7.46 (m, 3H), 3.74 (t, J = 4.6 Hz, 2H), 3.67 (s, 4H), 3.52 (t, J = 5.0 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.7, 141.9, 131.9, 128.7 (2C), 126.4 (2C), 66.7, 65.8, 50.2, 44.1.



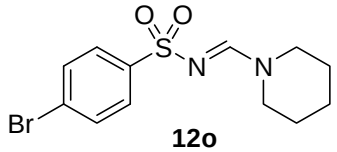
12m

***N*-[Bis(dimethylamino)methylene]benzenesulfonamide (12m).** 163 mg (64%), white solid, mp 130–131 °C (lit.[8] no mp); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.93–7.91 (m, 2H), 7.44–7.42 (m, 3H), 2.92 (s, 12H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 161.7, 145.7, 130.5, 128.2 (2C), 125.3 (2C), 40.3 (4C).



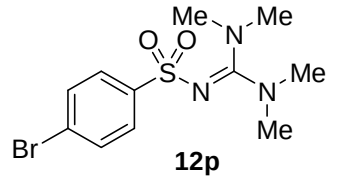
12n

***N*-[(Dimethylamino)methylene]-4-bromobenzenesulfonamide (12n).** 253 mg (87%), white solid, mp 139–141 °C (lit.[9] mp 139–140 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 (s, 1H), 7.75 (d, J = 8.7 Hz, 2H), 7.59 (d, J = 8.2 Hz, 2H), 3.15 (s, 3H), 3.02 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 159.1, 141.4, 131.7 (2C), 127.9 (2C), 126.4, 41.4, 35.4.



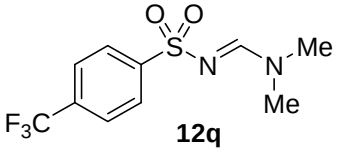
12o

***N*-(1-Piperidinylmethylene)-4-bromobenzenesulfonamide (12o).** 298 mg (90%), white solid, mp 132–133 °C; IR (KBr) ν 2983, 2868, 1624, 1311, 1140 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$) δ 8.12 (s, 1H), 7.75 (d, J = 8.2 Hz, 2H), 7.59 (d, J = 7.8 Hz, 2H), 3.59 (t, J = 6.0 Hz, 2H), 3.44 (t, J = 5.0 Hz, 2H), 1.68–1.67 (m, 4H), 1.60–1.55 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.2, 141.5, 131.7 (2C), 127.9 (2C), 126.2, 51.8, 44.5, 26.2, 24.7, 23.7. HRMS (ESI-TOF) (m/z): calcd for $C_{12}H_{15}BrN_2O_2S$, $[M+Na]^+$ 352.9930; found 352.9934.



12p

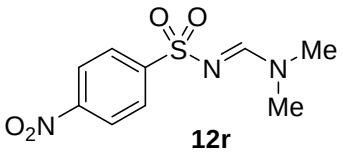
***N*-[Bis(dimethylamino)methylene]-4-bromo-benzenesulfonamide (12p).** 250 mg (75%), white solid, mp 153–154 °C (lit.[10] no mp); IR (KBr) ν 2952, 2864, 1611, 1463, 1158 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$) δ 7.80 (d, J = 8.7 Hz, 2H), 7.58-7.56 (m, 2H), 2.92 (s, 12H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 161.6, 144.8, 131.3 (2C), 127.1 (2C), 124.9, 40.3 (4C). HRMS (ESI-TOF) (m/z): calcd for $C_{11}H_{16}BrN_3O_2S$, $[M+Na]^+$ 356.0039; found 356.0036.



12q

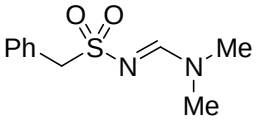
***N*,*N*-dimethyl-N'-((4-(trifluoromethyl)phenyl)sulfonyl)formimidamide (12q).** 207 mg (74%),White solid, mp 167–168 °C (lit. [11] no report); ^1H NMR (400 MHz, DMSO-d_6) δ 8.27 (s, 1H), 8.00 (d, J = 8.2 Hz, 2H), 7.89 (d, J = 8.7 Hz, 2H), 3.16 (s, 3H), 2.92 (s, 3H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 160.2, 146.9, 131.6 (q, J = 31.5

Hz, 1C), 126.9 (2C), 126.2 (q, J = 2.9 Hz, 2C), 122.6 (q, J = 271.7 Hz, 1C), 41.0, 35.2.



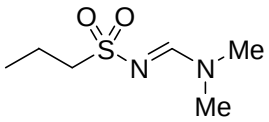
12r

***N*-[(Dimethylamino)methylene]-4-nitrobenzenesulfonamide (12r).** 141 mg (55%), white solid, mp 197–199 °C (lit.[11] mp 197–201 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.31 (d, J = 8.7 Hz, 2H), 8.17 (s, 1H), 8.08 (d, J = 8.7 Hz, 2H), 3.19 (s, 3H), 3.06 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 159.4, 149.5 148.1, 127.8 (2C), 124.0 (2C), 41.7, 35.7.



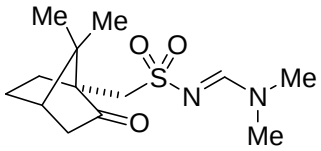
12s

***N*-[(Dimethylamino)methylene]-benzylsulfonamide (12s).** 99 mg (44%), white solid, mp 117–118 °C (lit.[12] 135–137 °C); ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.50 (s, 1H), 7.36–7.28 (m, 5H), 4.26 (s, 2H), 2.99 (s, 3H), 2.93 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 160.2, 130.9 (2C), 130.2, 128.3 (2C), 128.2, 59.6, 41.0, 35.2.



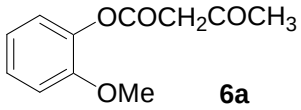
12t

***N*-[(Dimethylamino)methylene]propylsulfonamide (12t).** 75 mg (42%), colorless oil; IR (KBr) ν 2968, 2822, 1633, 1281, 1120 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.04 (s, 1H), 3.13 (s, 3H), 3.04 (s, 3H), 3.01–2.96 (m, 2H), 1.86–1.78 (m, 2H), 1.04 (t, J = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 159.3, 55.5, 41.2, 35.2, 17.3, 12.9. HRMS (ESI-TOF) (m/z): calcd for $C_6H_{14}N_2O_2S$, $[M+Na]^+$ 201.0668; found 201.0665.



12u

(1*R*)-*N*-[(Dimethylamino)methylene]camphorsulfonamide (12u). 212 mg (74%), white solid, mp 108–109 °C; IR (KBr) ν 3452, 2961, 2889, 1734,1627, 1297, 1137 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$) δ 8.04 (s, 1H), 3.46 (d, J = 1.4 Hz, 1H), 3.16 (s, 3H), 3.02 (s, 3H), 2.96 (d, J = 1.4 Hz, 1H), 2.67–2.68 (m, 1H), 2.35 (d, J = 1.9 Hz, 1H), 2.09–2.00 (m, 2H), 1.91 (d, J = 1.8 Hz, 1H), 1.78–1.71 (m, 1H), 1.45–1.38 (m, 1H), 1.12 (s, 3H), 0.86 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 215.1, 159.6, 58.3, 50.6, 47.8, 42.5, 42.4, 41.2, 35.2, 26.8, 24.6, 19.8, 19.6. HRMS (ESI-TOF) (m/z): calcd for $C_{13}H_{22}N_2O_3S$, $[M+Na]^+$ 309.1243; found 309.1246.

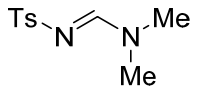


6a

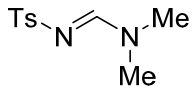
Preparation of 2-methoxyphenylacatoacetate (14). To a stirred solution of 3-butyn-2-one (68 mg, 1 mmol), 2-methoxyphenol (124 mg, 1 mmol) and CuI (19 mg, 0.1 mmol) in DMF (2 mL) was added methanesulfonyl azide (171 mg, 1.4 mmol). The mixture was heated under 50 °C for 1 h and then cooled to room temperature. The system was extracted with ethyl acetate and brine. After the organic layer was dried over $MgSO_4$, removal of the solvent under vacuum gave the crude product as a residue, which was purified by a flash chromatography (silica gel, PE : EtOAc = 2 : 1) to give 139 mg of the product **14** (67%) as a colorless oil. It was a mixture with enol structure (6:1) (lit.[13] mp 62–63 °C); ^1H NMR (300 MHz, $CDCl_3$) δ 11.83 (s, 0.18H), 7.26–7.19 (m, 1.6H), 7.08–7.05 (m, 1.16H), 6.98–6.95 (m, 2.41H), 5.29 (s, 0.19H), 3.83 (s, 0.5H), 3.82 (s, 3H), 3.68 (s, 2H), 2.38 (s, 3H), 2.03 (s, 0.5H) [peaks for the major product: 7.26–7.19 (m, 1H), 7.08–7.05 (m, 1H), 6.98–6.92 (m, 2H), 3.83 (s, 3H), 3.68 (s, 2H), 2.38 (s, 3H)]. ^{13}C NMR (75 MHz, $CDCl_3$) δ 199.9, 165.1, 150.8, 139.3, 127.2, 122.6, 120.7, 112.4, 55.7, 49.8, 29.9.

References:

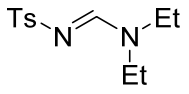
1. Xu, X.; Ma, L.;Ye, N.; Weng, B. *J. Am. Chem. Soc*. **2008**, *130*, 14048–14049.
2. Jakobsen, P.; Treppendahl, S. *Tetrahedron* **1977**, *33*, 3137–3140.
3. Sevast'yanov, V. N.; Abrazhanova, E. A.; Skripets, V. I. *Voprosy Khimii i Khimicheskoi Tekhnologii*. **1983**, *72*, 82–84.
4. Ahuja, P.; Singh, J.; Asthana, M. B.; Sardana, V.; Anand, N. *Indian J. Chem., Sect. B: Org. Chem. Incl. Med. Chem.* **1989**, *28*, 1034–1038.
5. Qi, Y.; Gao, H.; Yang, M.; Xia, C.-G.; Suo, J. *Synth. Commun*. **2003**, *33*, 1073–1079.
6. Derkach, N. Y.; Barashenkov, G. G.; Slyusarenko, E. I. *Zhurnal Organicheskoi Khimii*, **1982**, *18*, 70–78.
7. Krasnov, V. L.; Vasyanina, G. I.; Bodrikov, I. V. *Zhurnal Organicheskoi Khimii*, **1991**, *27*, 1552–1556.
8. Kessler. H.; Leibfritz. D.; Burk, C. *Tetrahedron* **1970**, 1805–1820.
9. Dudutiene, V.; Asta, Z.; Alexey, S.; Joana, G.; David, T. *Bioorg. Med. Chem*. **2013**, *21*, 2093–2106.
10. Munakata, T.; Hasegawa, H.; Furuta, T. *Jpn. Tokkyo Koho.* **1970**, JP 45017680 B4 19700618.
11. Chen, S.; Wan, X. *Org. Lett*. **2011**, *13*, 6152–6155.
12. Kresze, G.; Albrecht, R. *Angew. Chem.* **1962**, *74*, 781–782; *Angew. Chem. Int. Ed.* **1962**, *1*, 595-596.
13. Rall, K. B.; Perekalin, V. V. *Zhurnal Obshchei Khimii*. **1955**, *25*, 276–281.



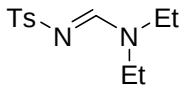
12a, ^{1}H NMR (400 MHz, $CDCl_3$)



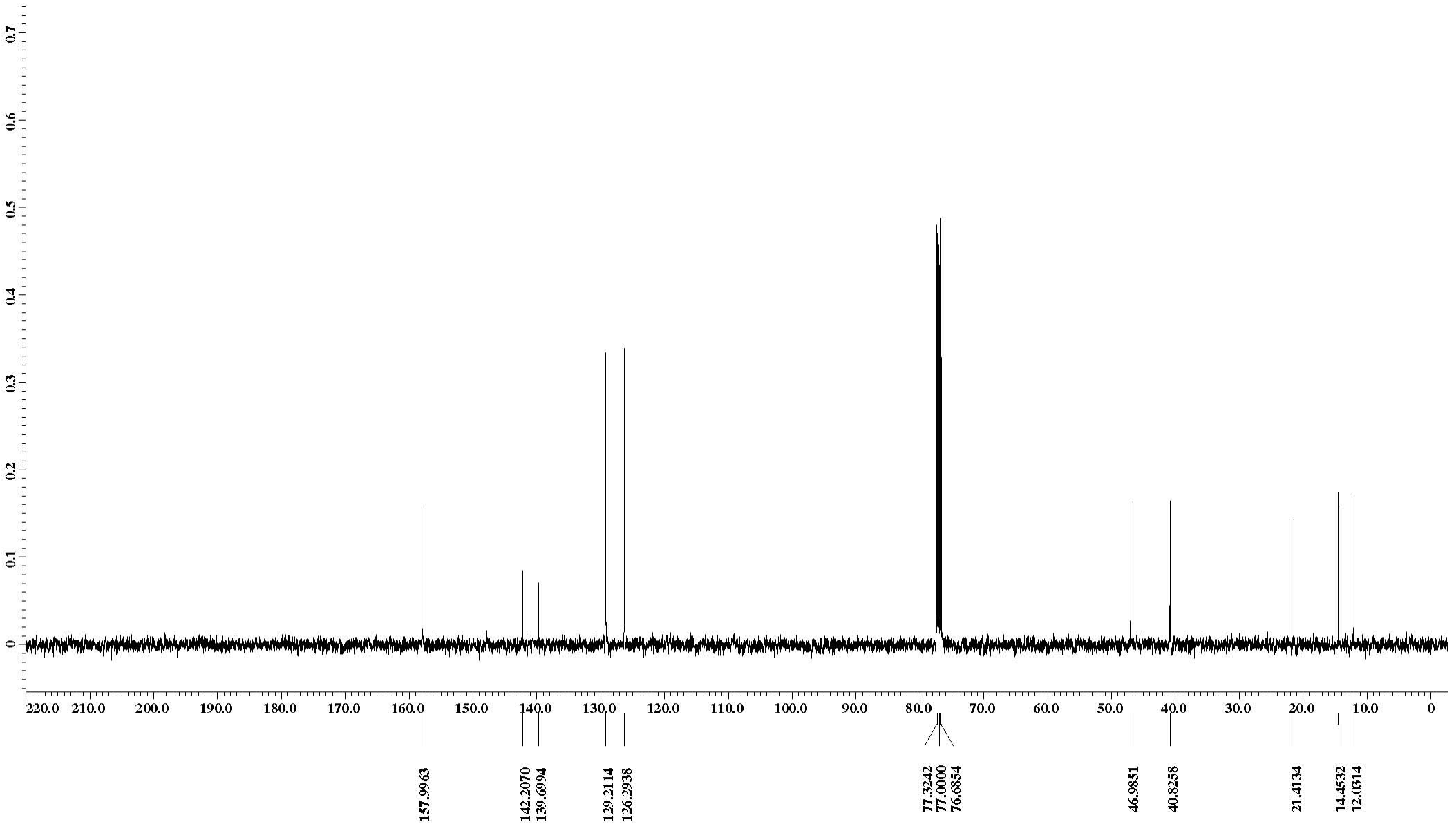
12a, ^{13}C NMR (100 MHz, $CDCl_3$)

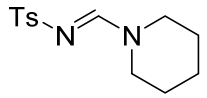


12b, ^{1}H NMR (400 MHz, $CDCl_3$)

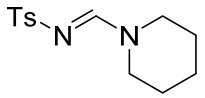


12b, ^{13}C NMR (100 MHz, $CDCl_3$)

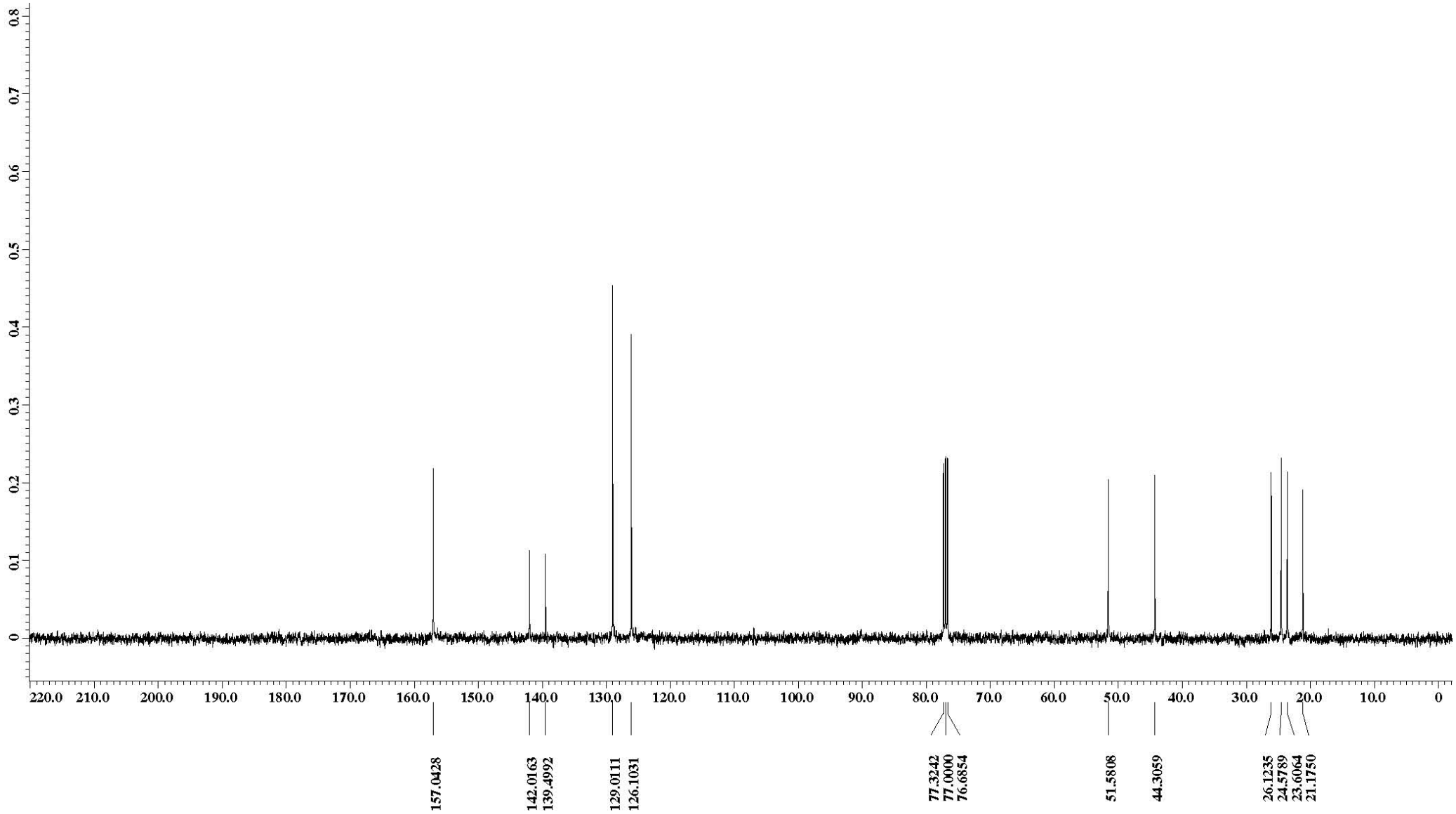


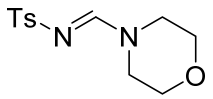


12c, ^{1}H NMR (400 MHz, $CDCl_3$)

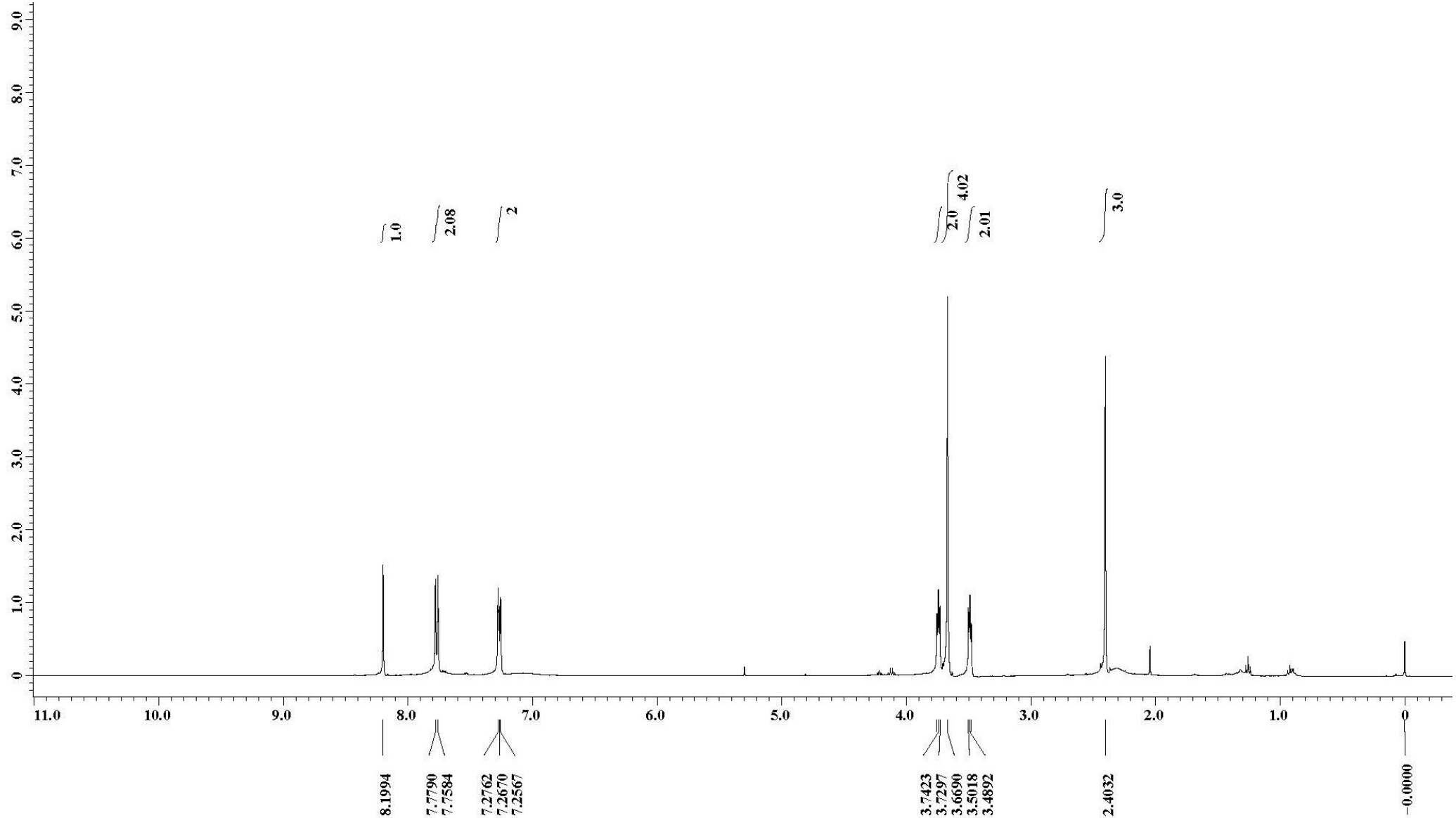


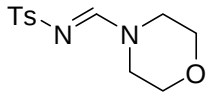
12c, ^{13}C NMR (100 MHz, $CDCl_3$)



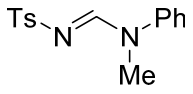


12d, ^{1}H NMR (400 MHz, $CDCl_3$)

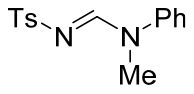




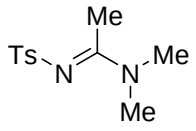
12d, ^{13}C NMR (100 MHz, $CDCl_3$)



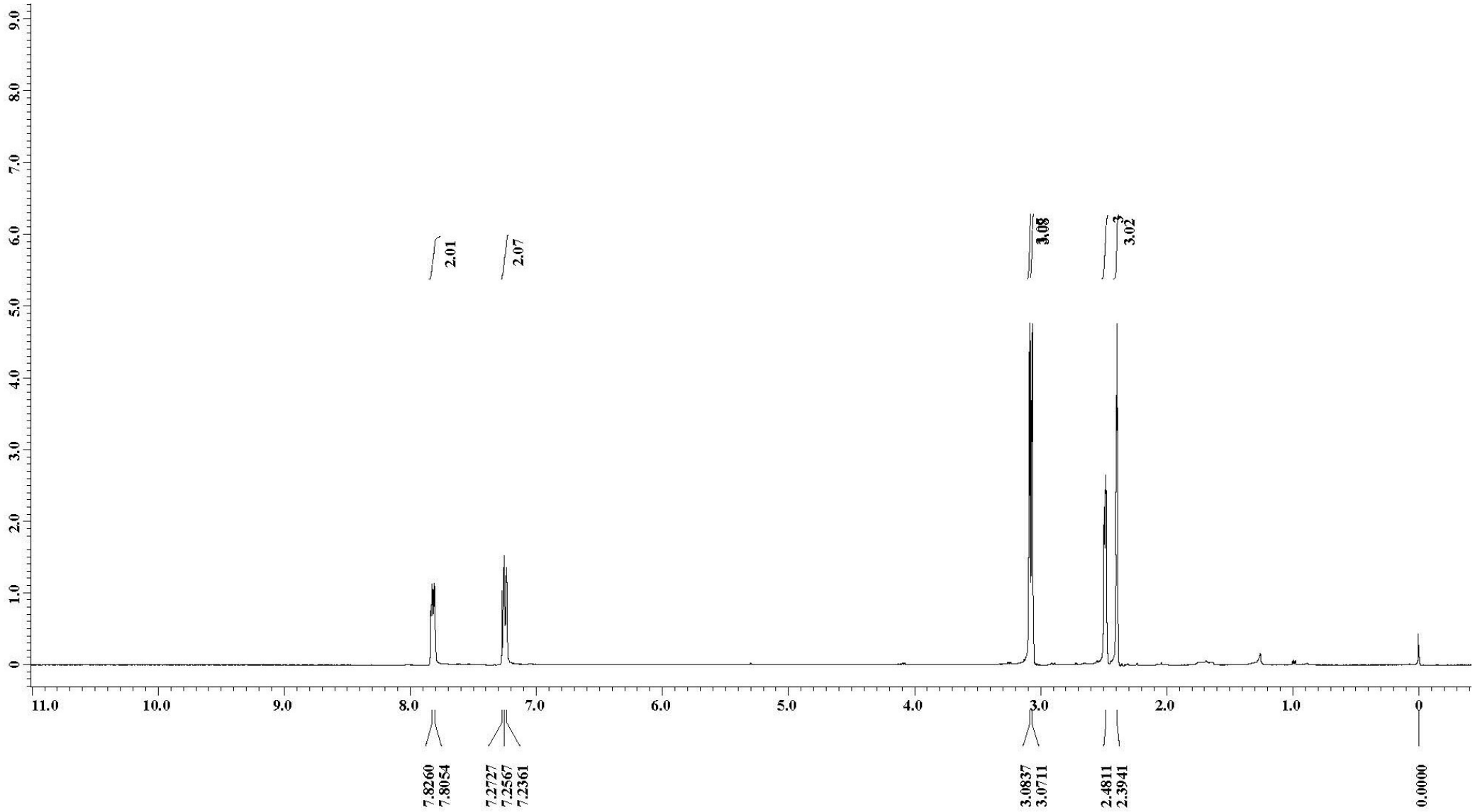
12e, ^{1}H NMR (400 MHz, $CDCl_3$)

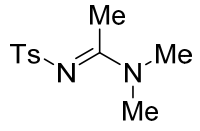


12e, ^{13}C NMR (100 MHz, $CDCl_3$)

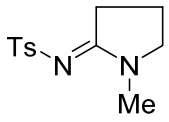


12f, ^{1}H NMR (400 MHz, $CDCl_3$)

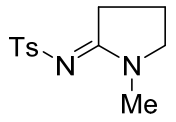




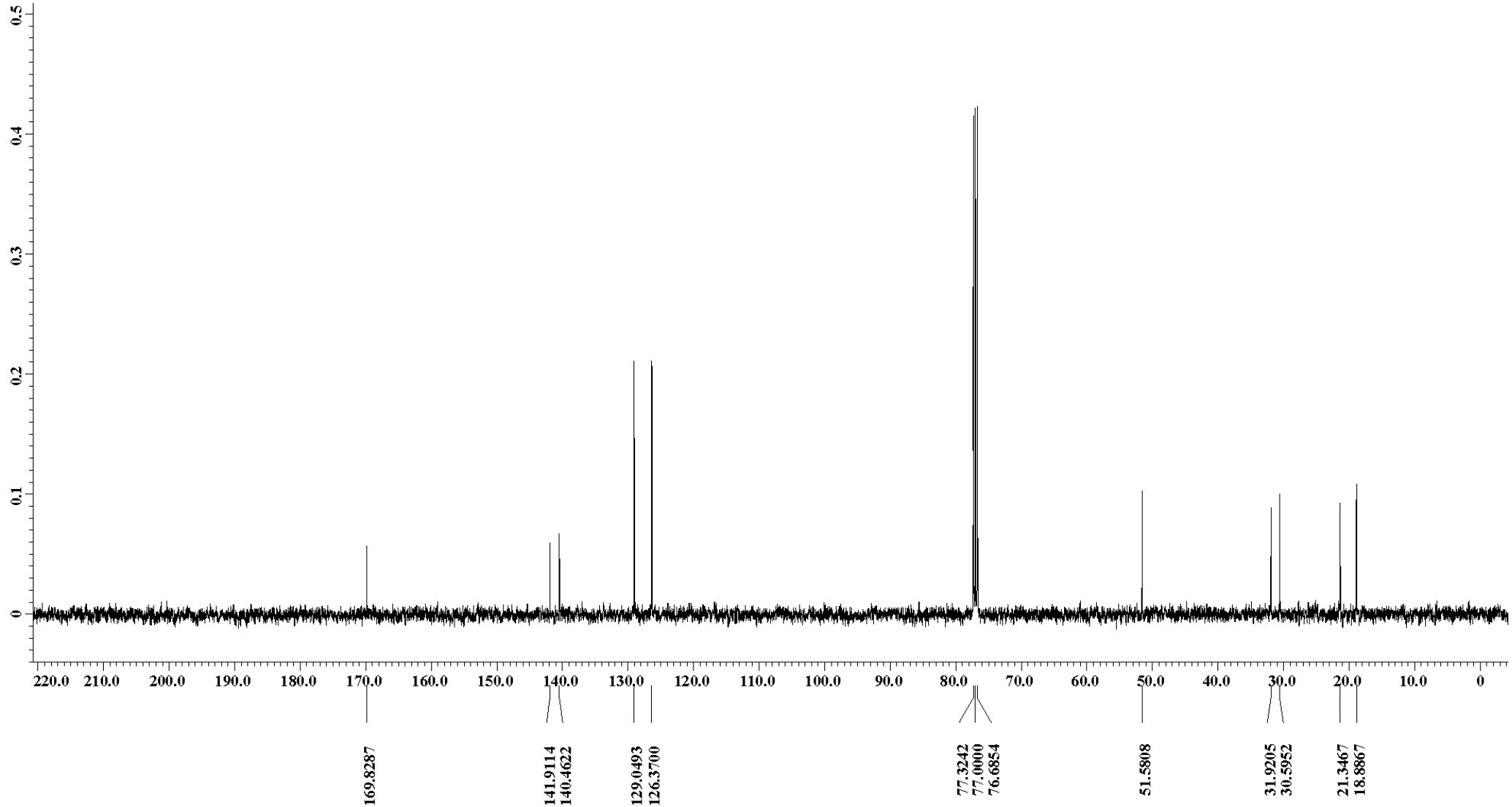
12f, ^{13}C NMR (100 MHz, $CDCl_3$)

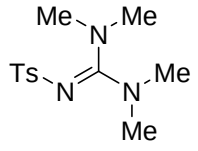


12g, ^{1}H NMR (400 MHz, $CDCl_3$)

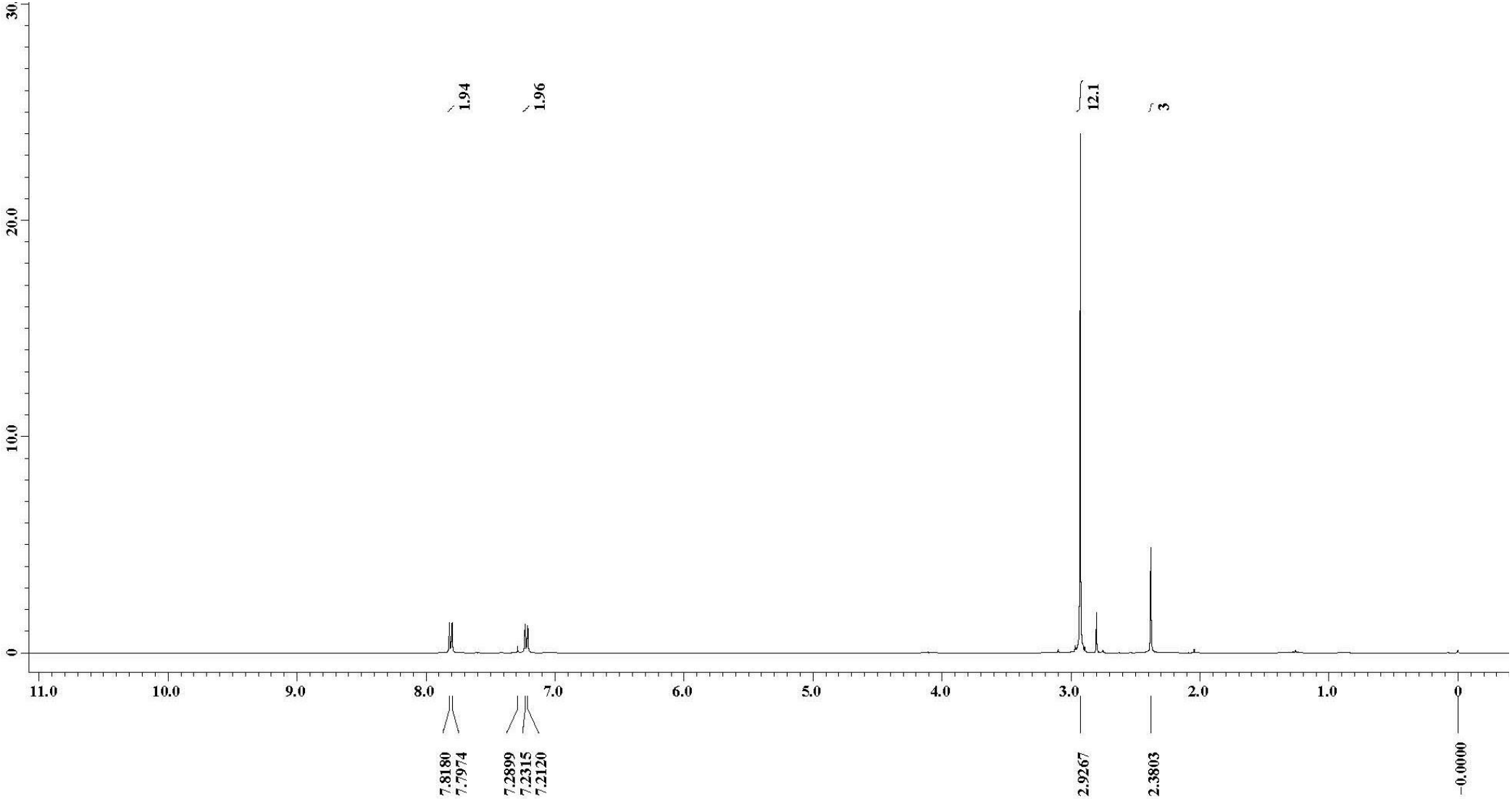


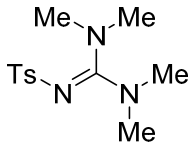
12g, ^{13}C NMR (100 MHz, $CDCl_3$)



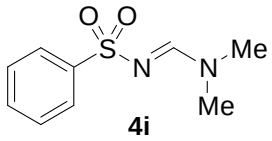


12h, ^{1}H NMR (400 MHz, $CDCl_3$)



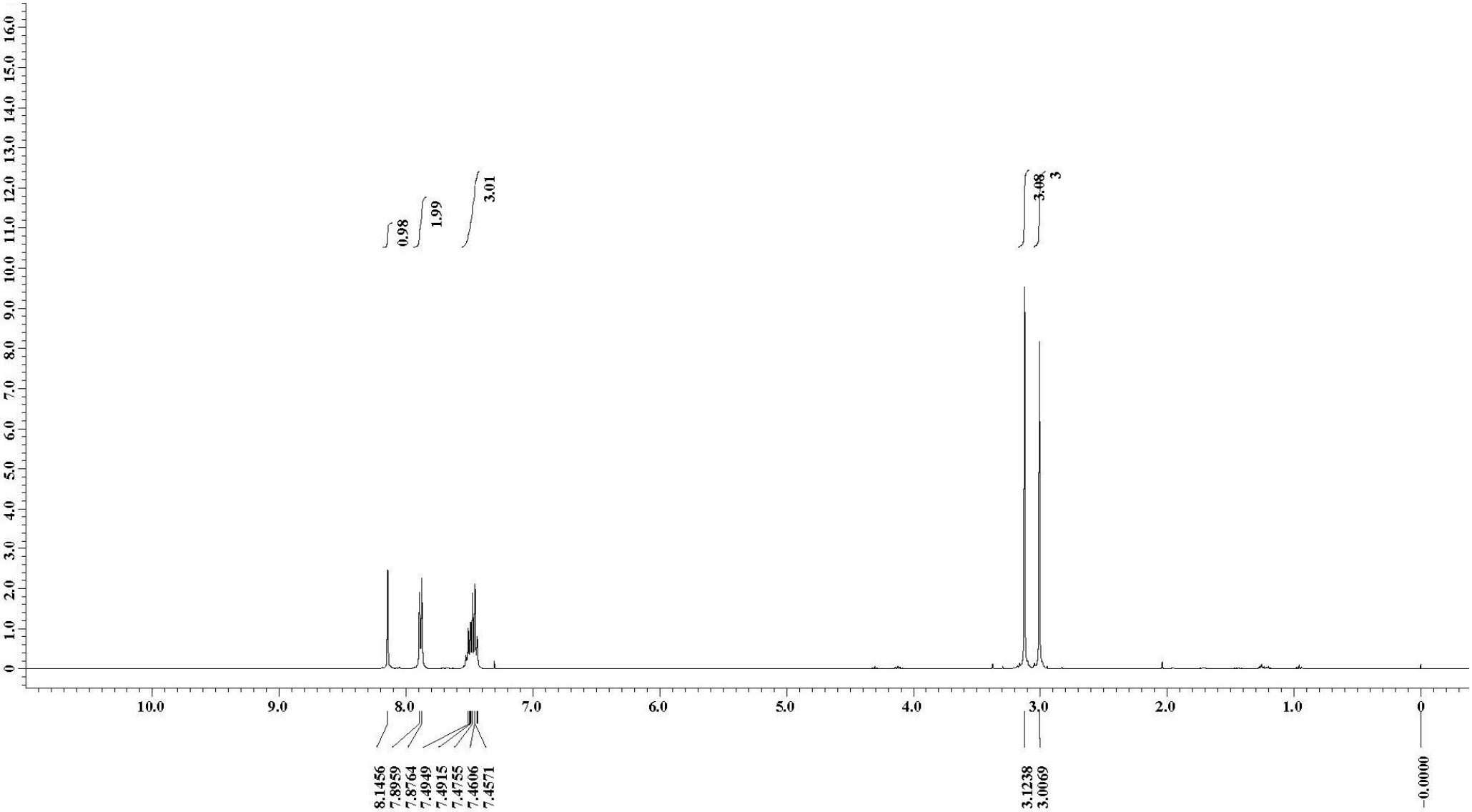


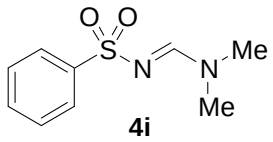
12h, ^{13}C NMR (100 MHz, $CDCl_3$)



4i

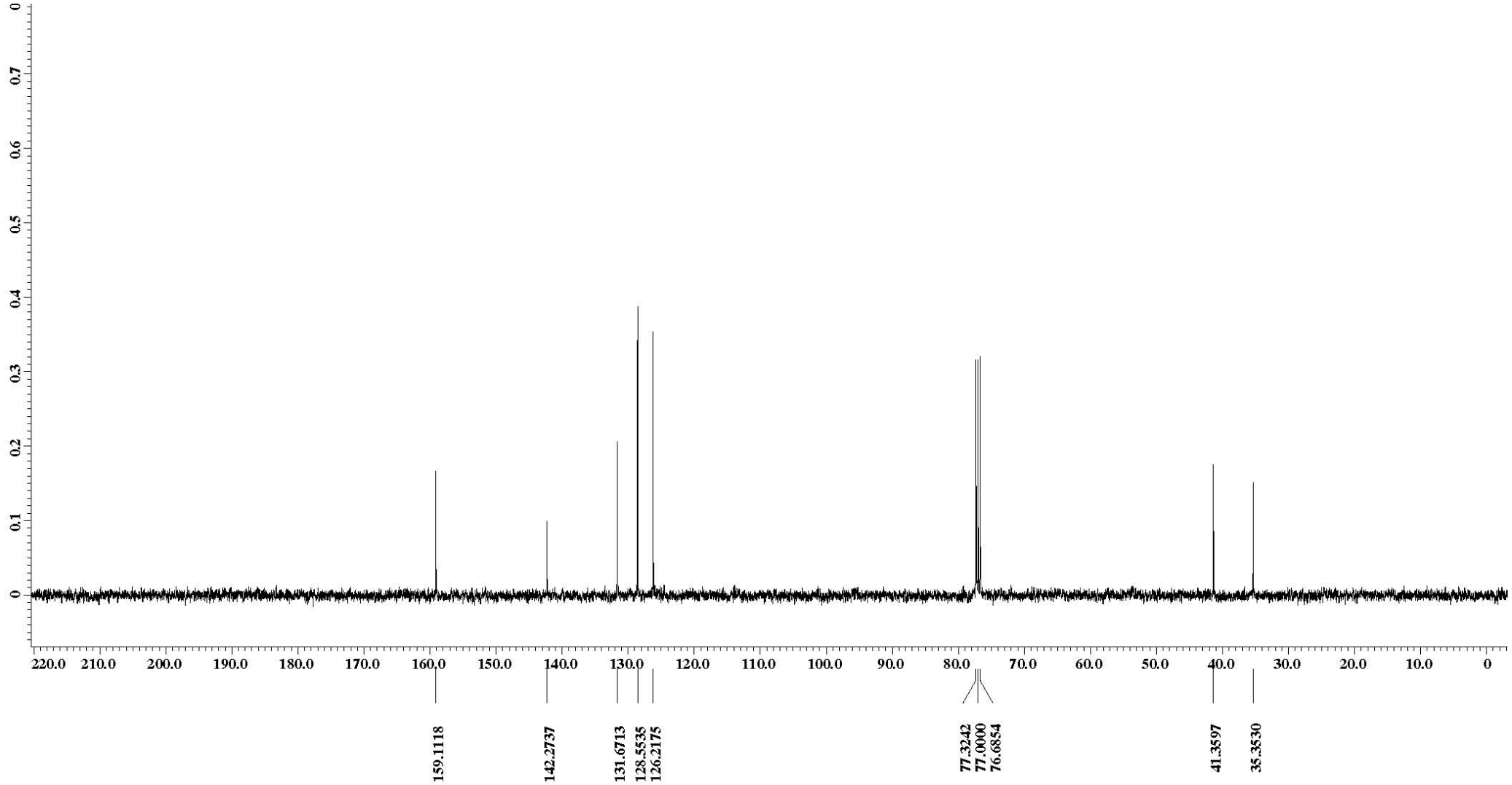
12i, ^{1}H NMR (400 MHz, $CDCl_3$)

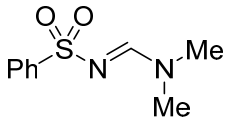




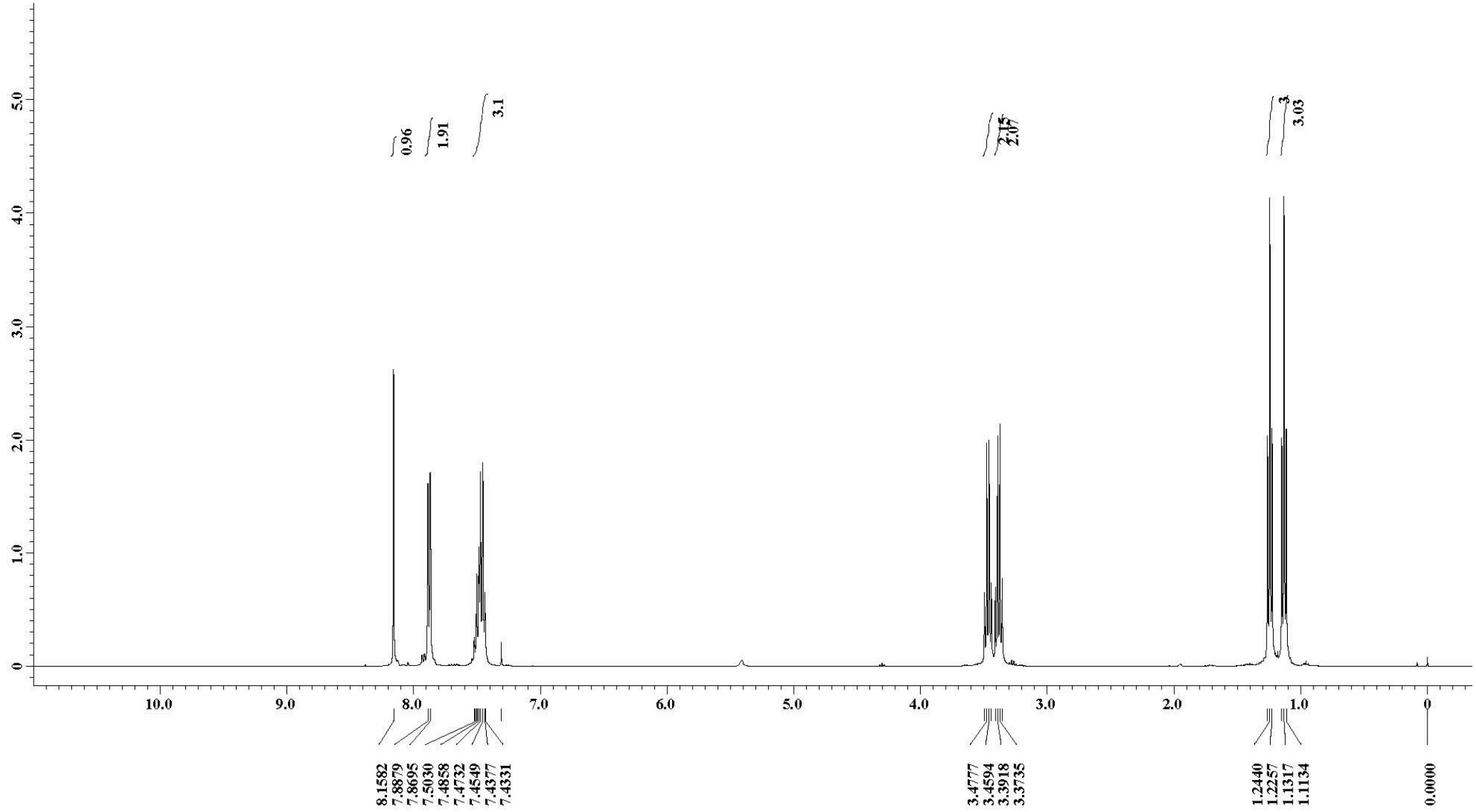
4i

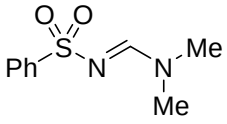
12i, ^{13}C NMR (100 MHz, $CDCl_3$)



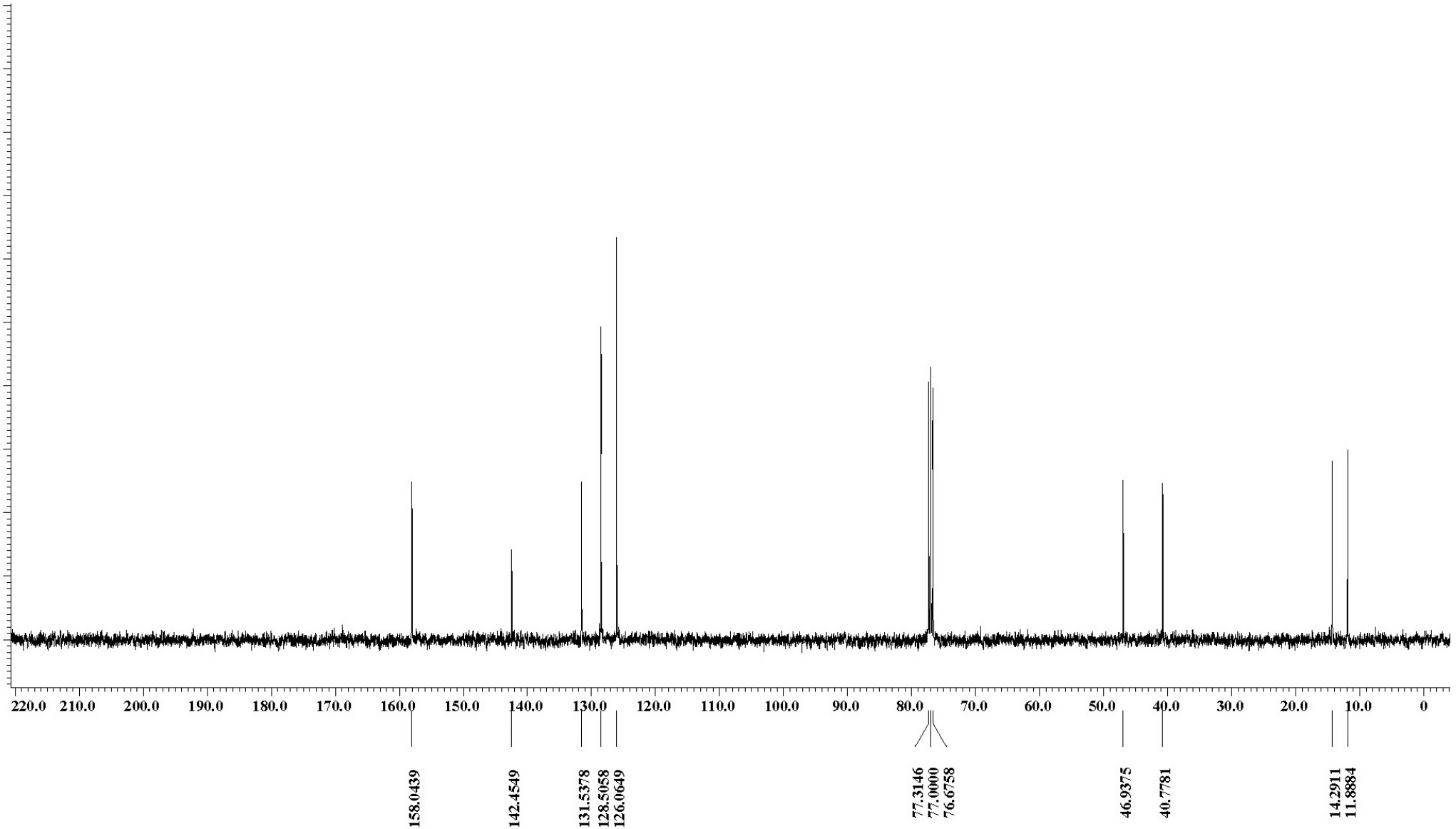


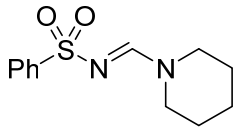
12j, ^{1}H NMR (400 MHz, $CDCl_3$)



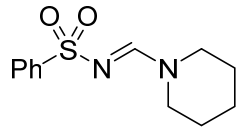


12j, ^{13}C NMR (100 MHz, $CDCl_3$)

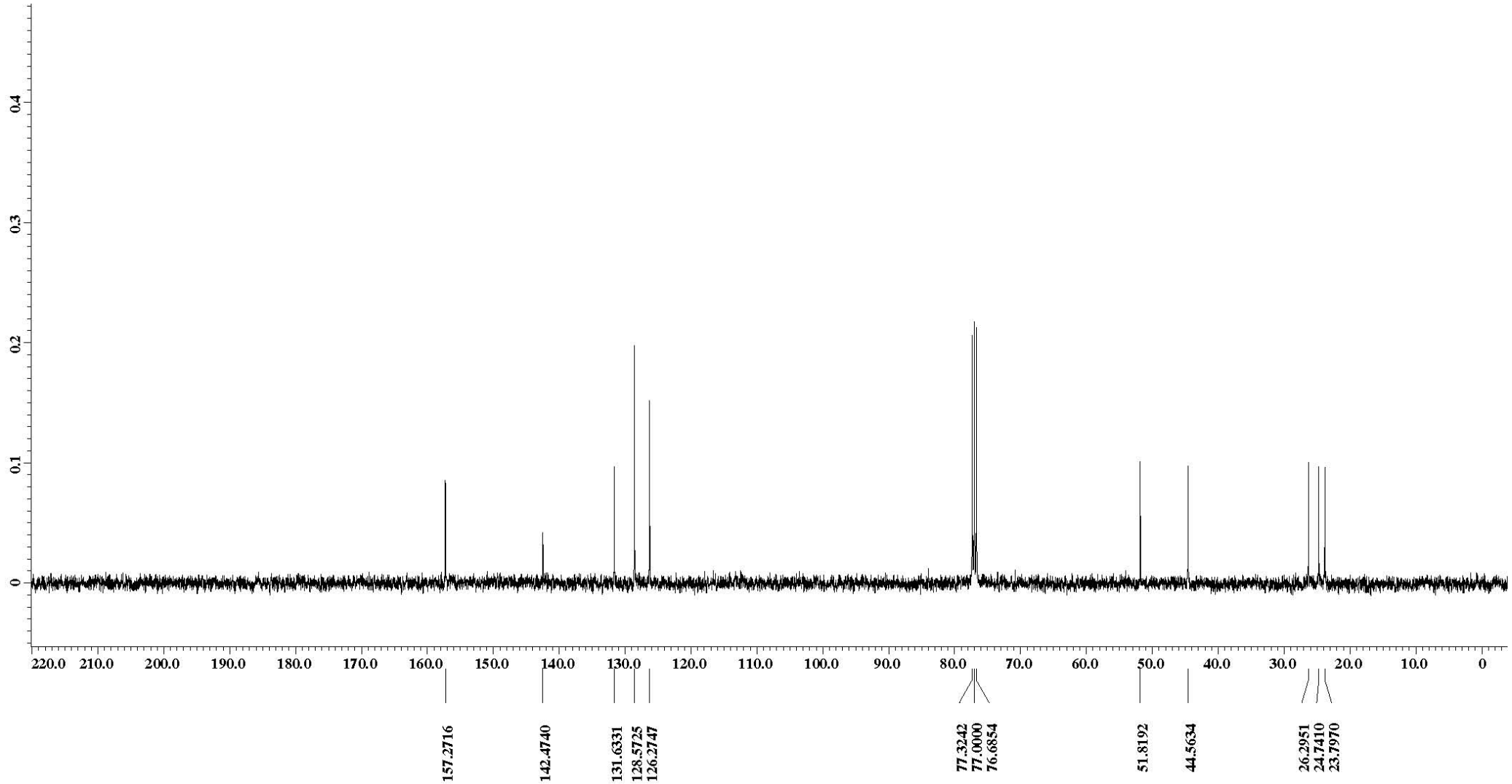


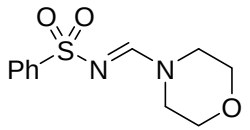


12k, ^{1}H NMR (400 MHz, $CDCl_3$)

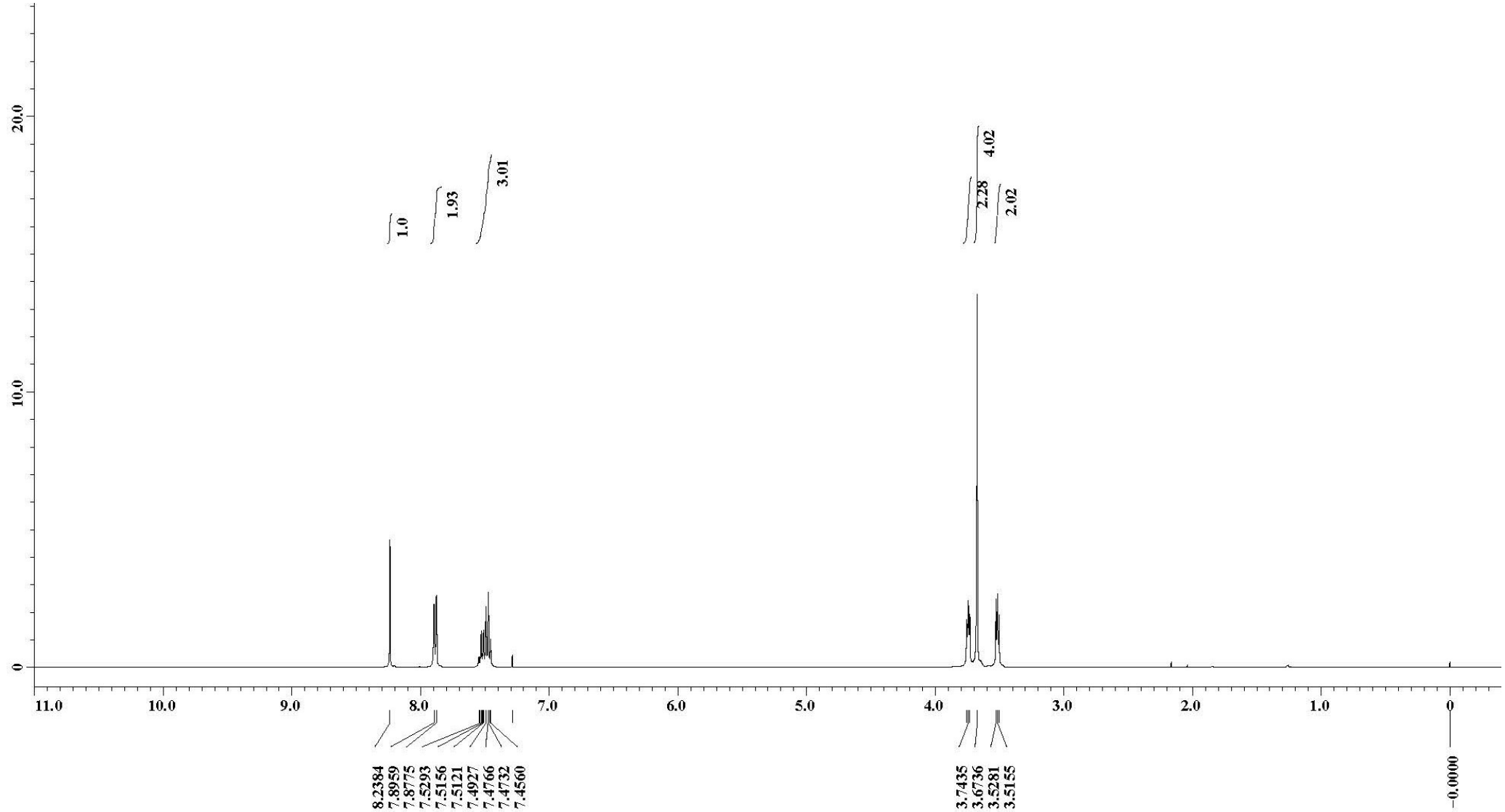


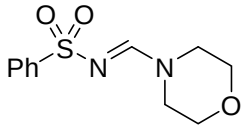
12k, ^{13}C NMR (100 MHz, $CDCl_3$)



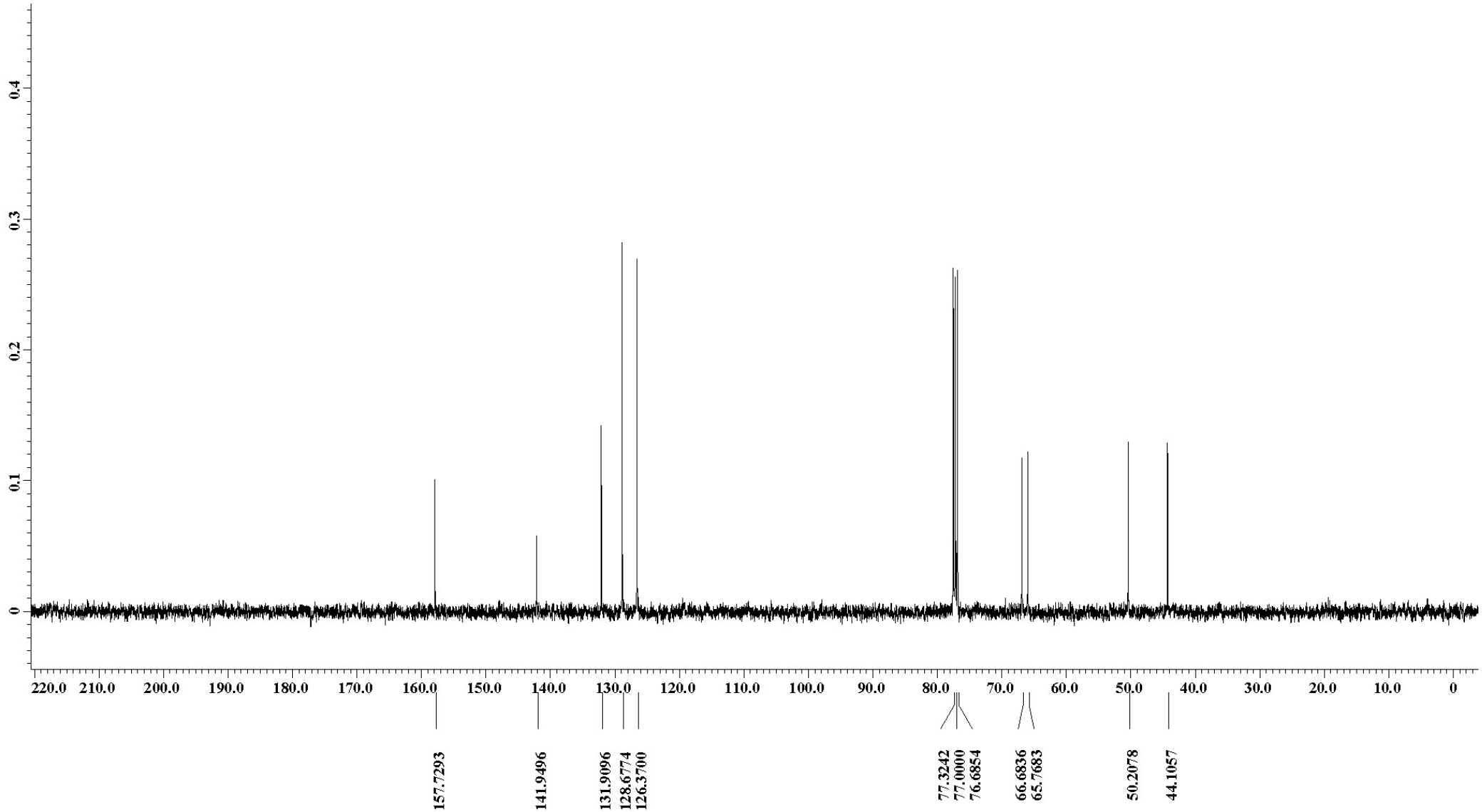


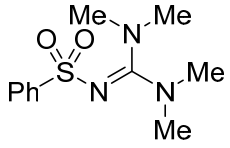
12l, ^{1}H NMR (400 MHz, $CDCl_3$)



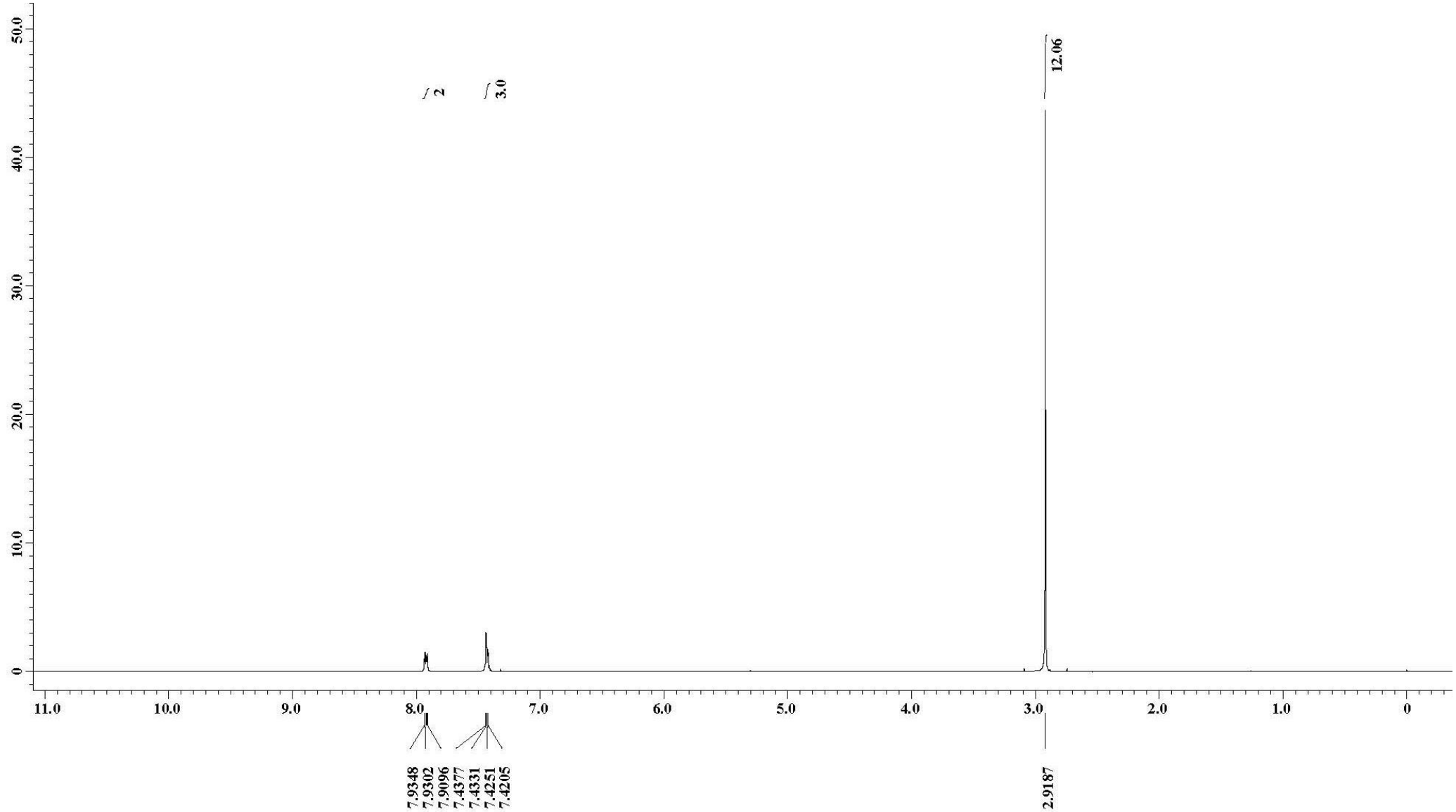


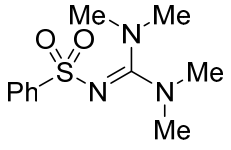
12l, ^{13}C NMR (100 MHz, $CDCl_3$)



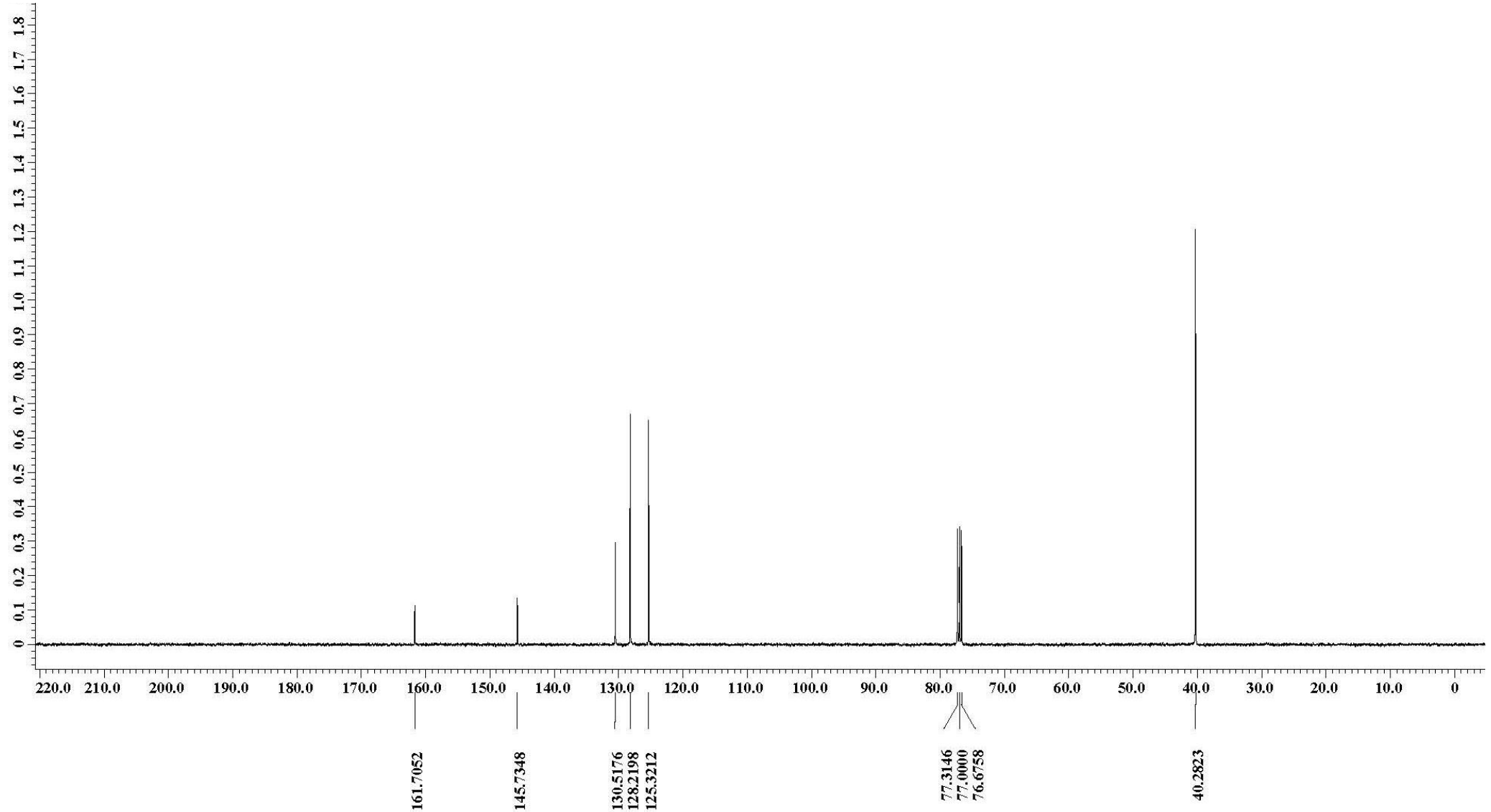


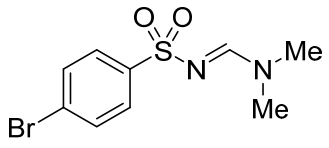
12m, ^{1}H NMR (400 MHz, $CDCl_3$)



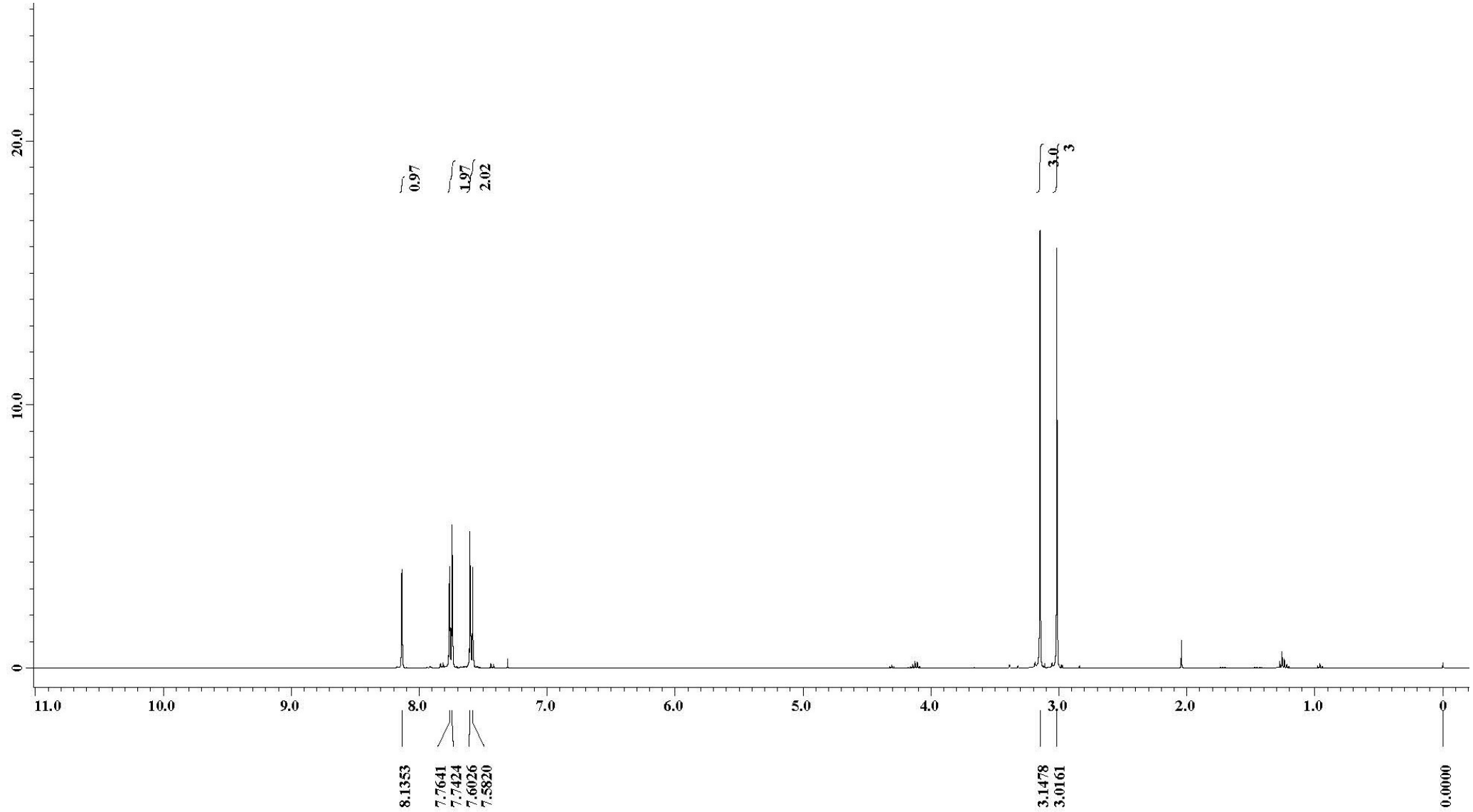


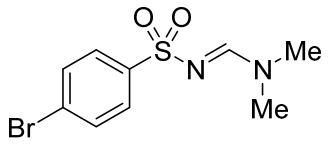
12m, ^{13}C NMR (100 MHz, $CDCl_3$)



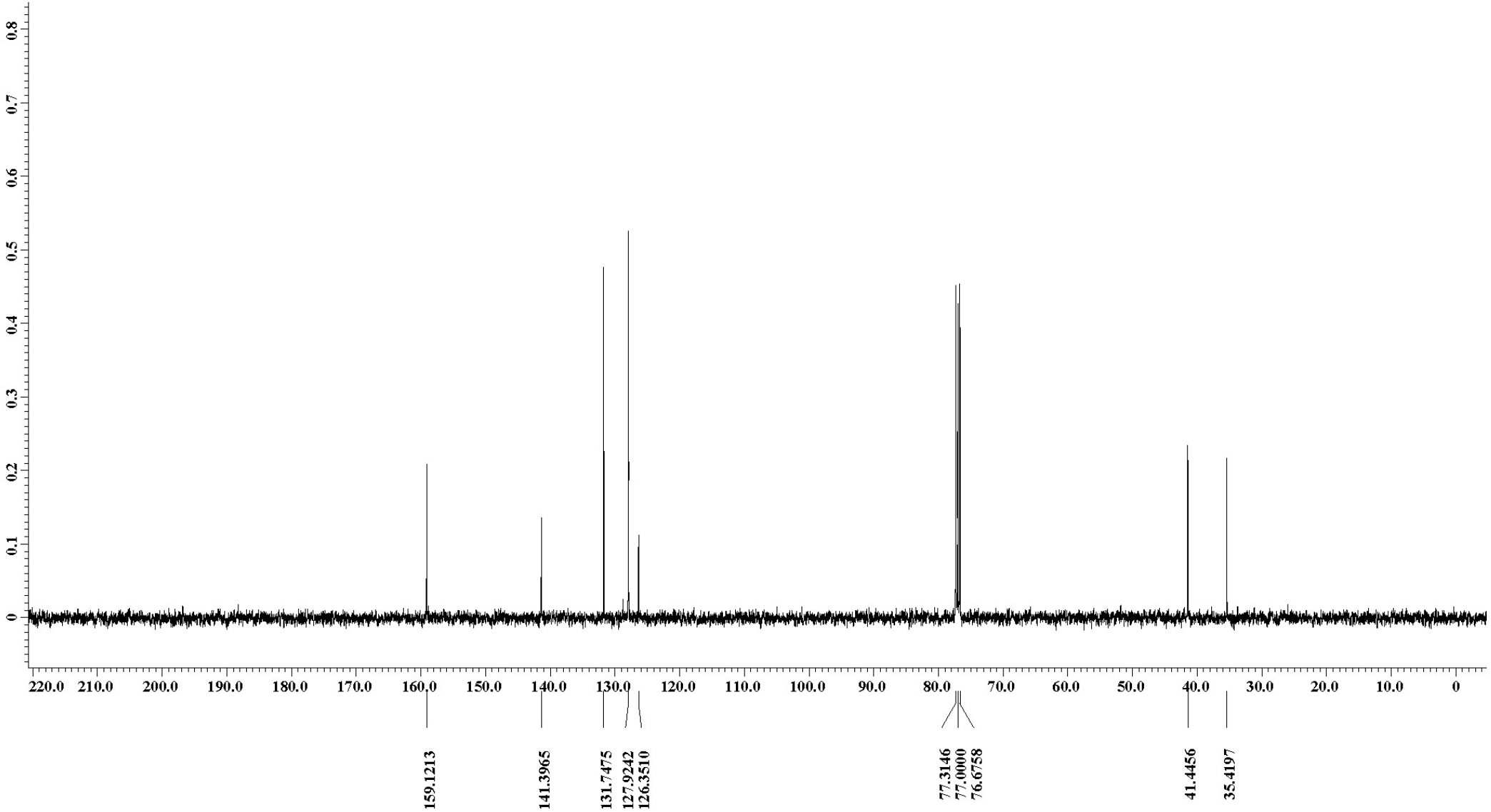


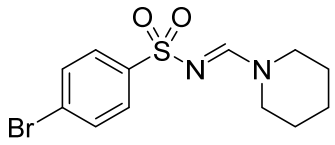
12n, ^{1}H NMR (400 MHz, $CDCl_3$)



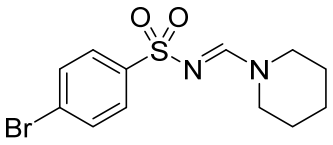


12n, ^{13}C NMR (100 MHz, $CDCl_3$)

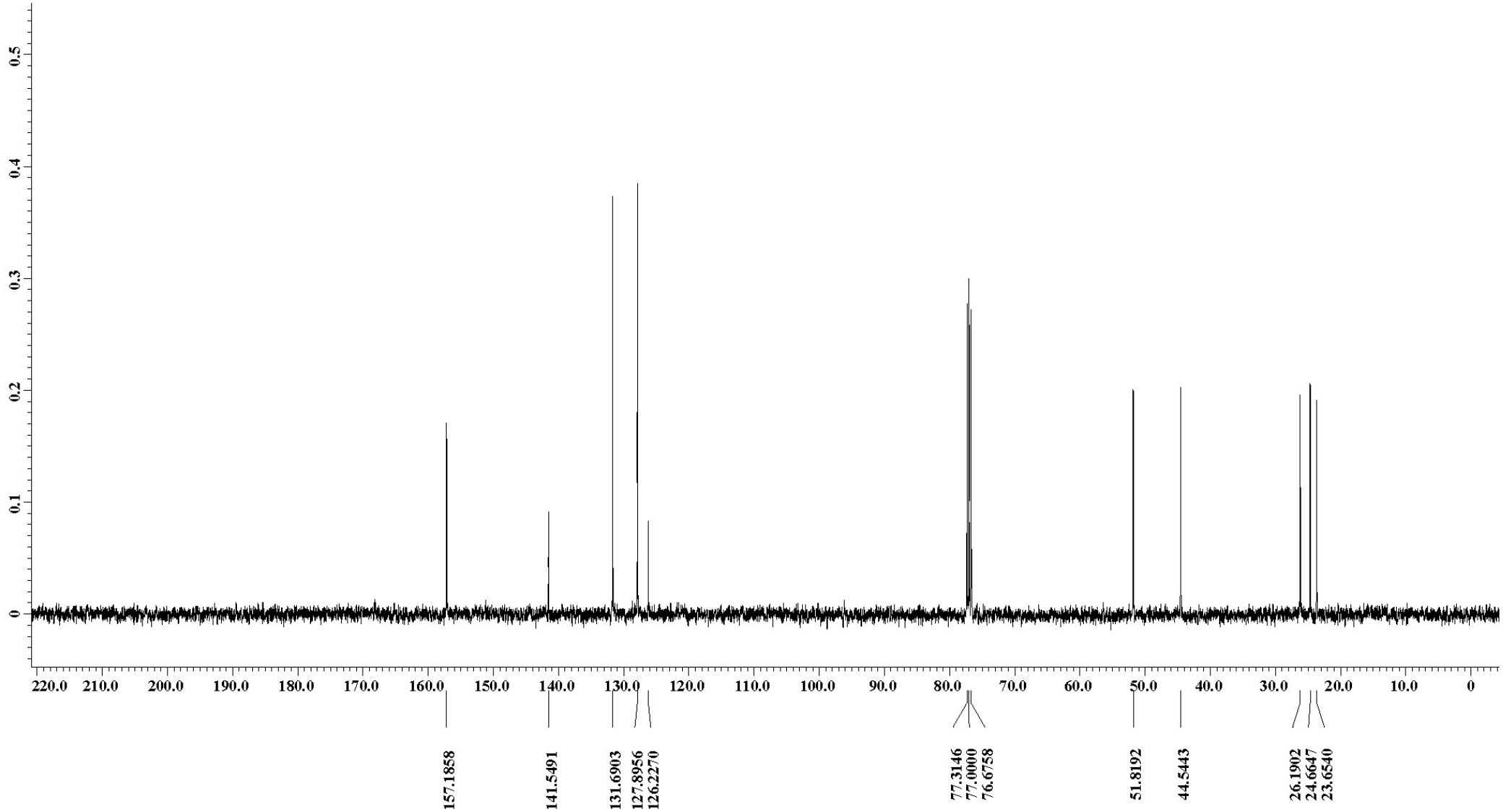


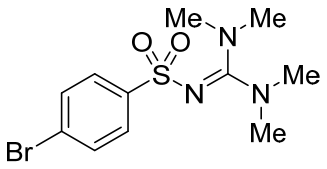


12o, ^{1}H NMR (400 MHz, $CDCl_3$)

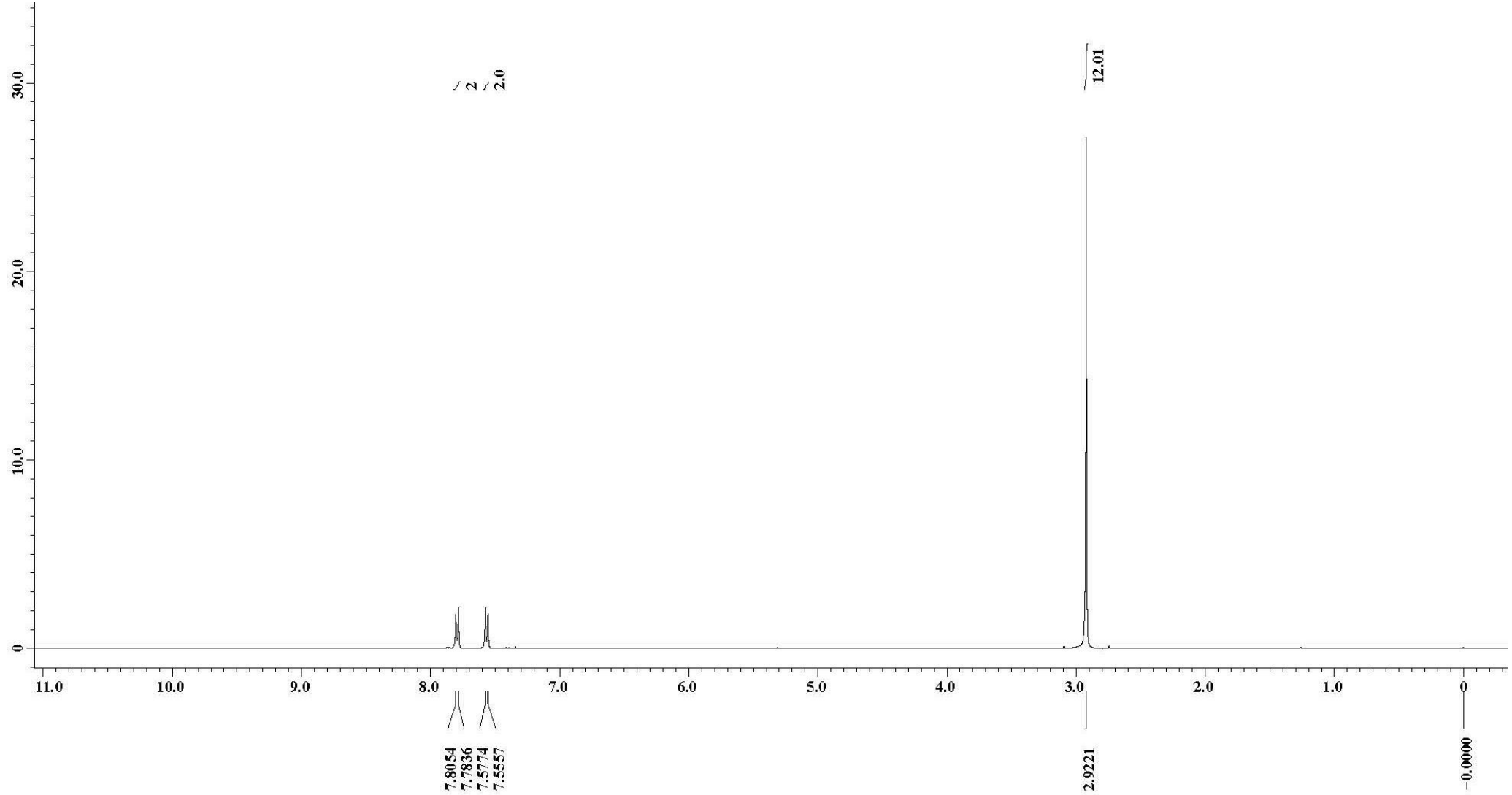


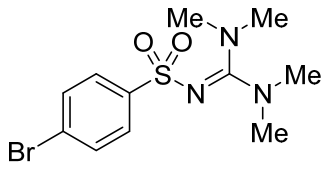
12o, ^{13}C NMR (100 MHz, $CDCl_3$)



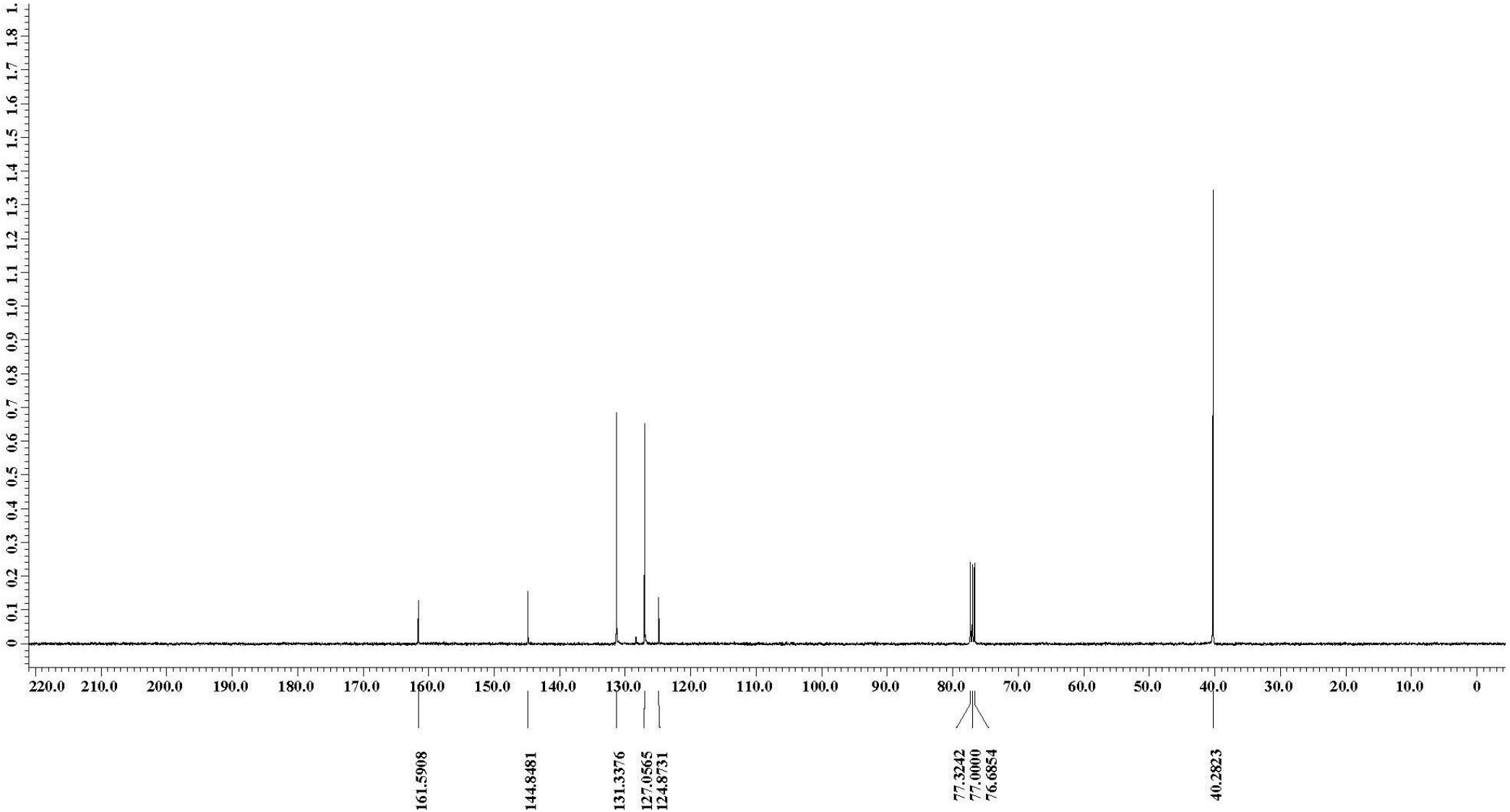


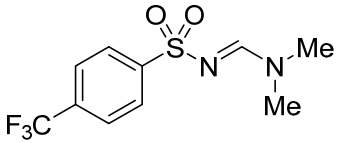
12p, ^{1}H NMR (400 MHz, $CDCl_3$)



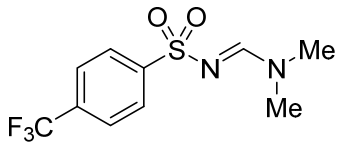


12p, ^{13}C NMR (100 MHz, $CDCl_3$)

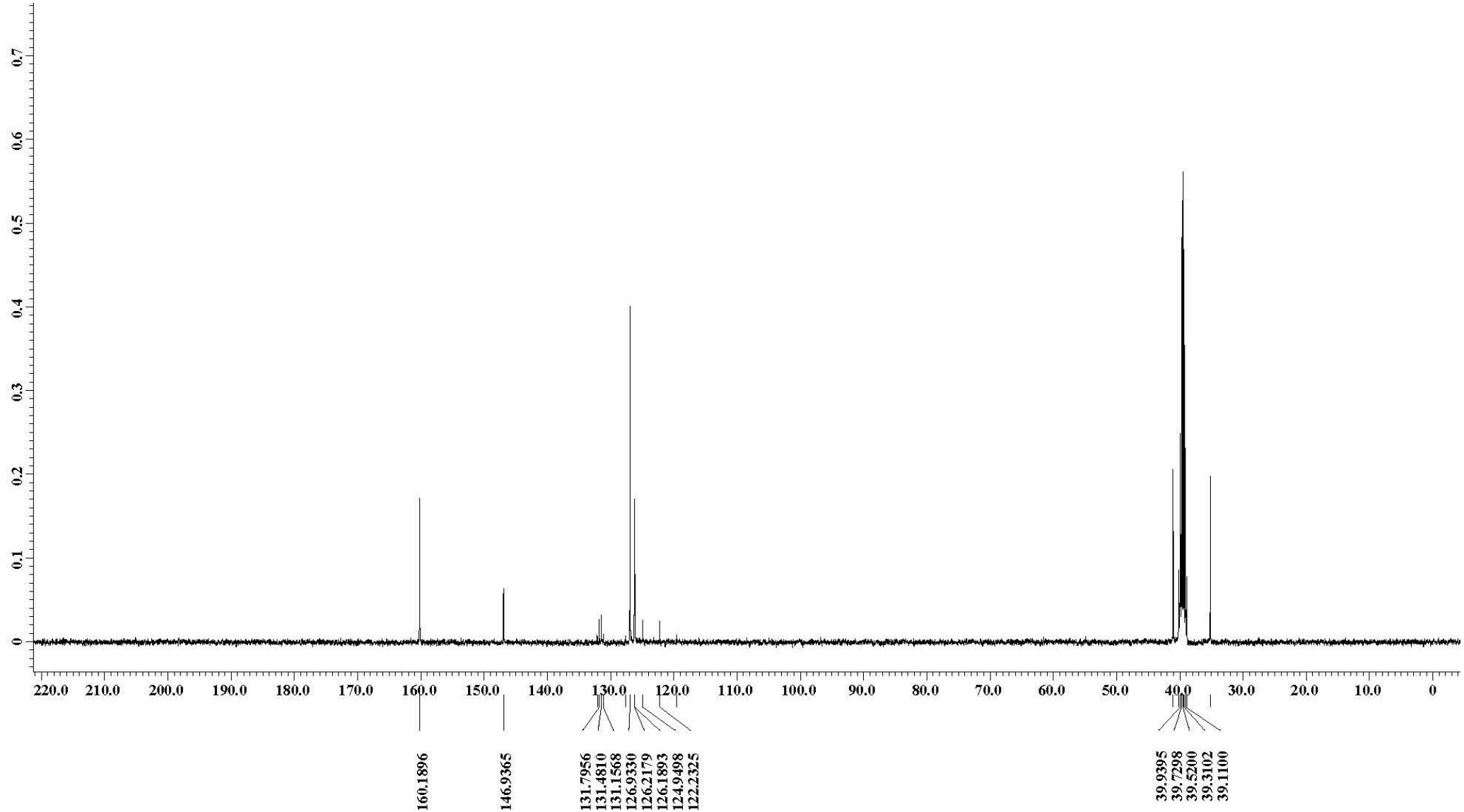


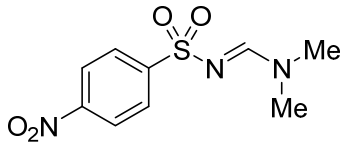


12q, ^{1}H NMR (400 MHz, DMSO-d_6)

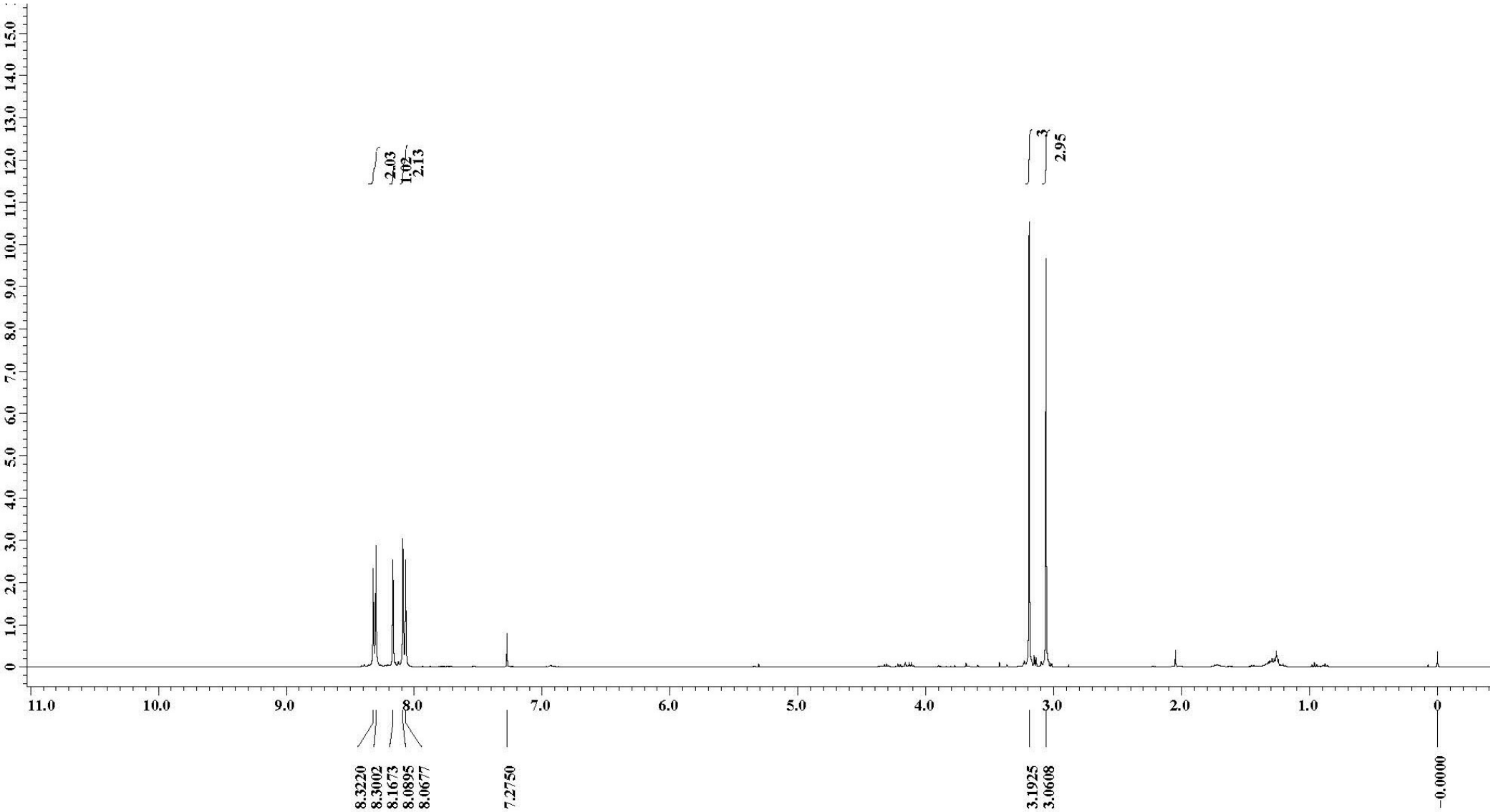


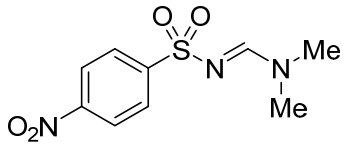
12q, ^{13}C NMR (100 MHz, DMSO-d_6)



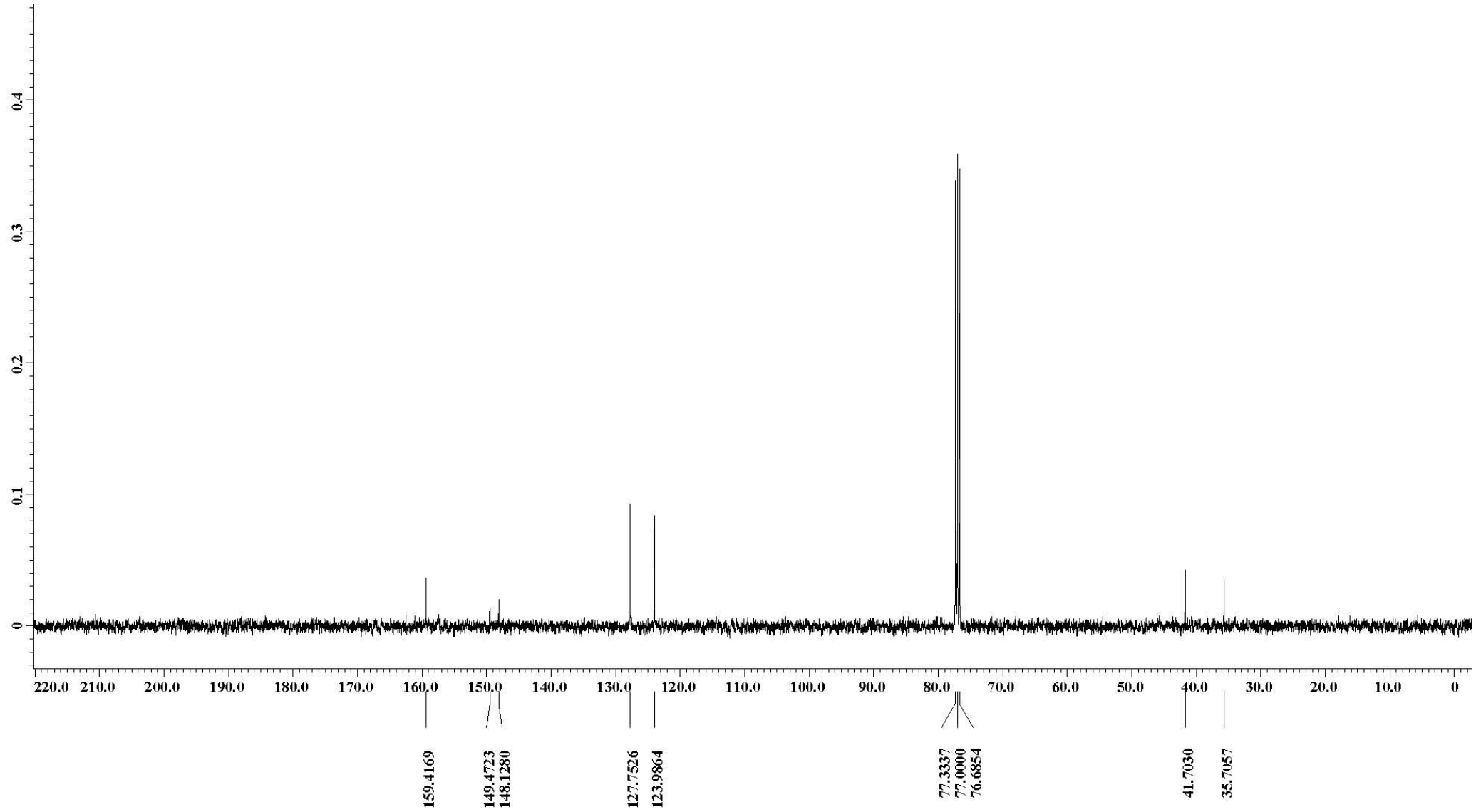


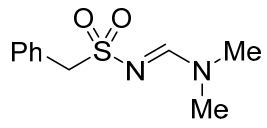
12r, ^{1}H NMR (400 MHz, $CDCl_3$)



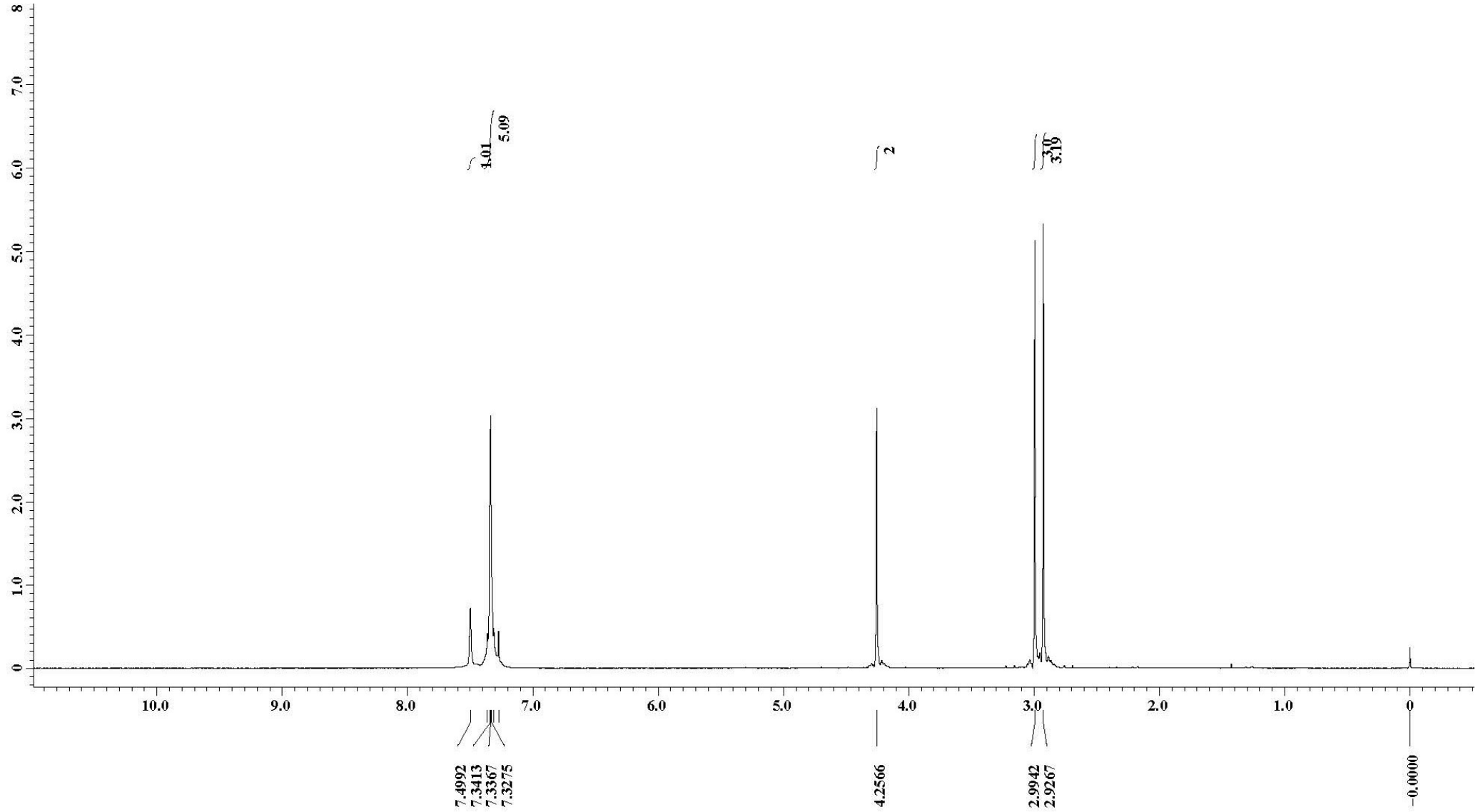


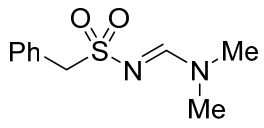
12r, ^{13}C NMR (100 MHz, $CDCl_3$)



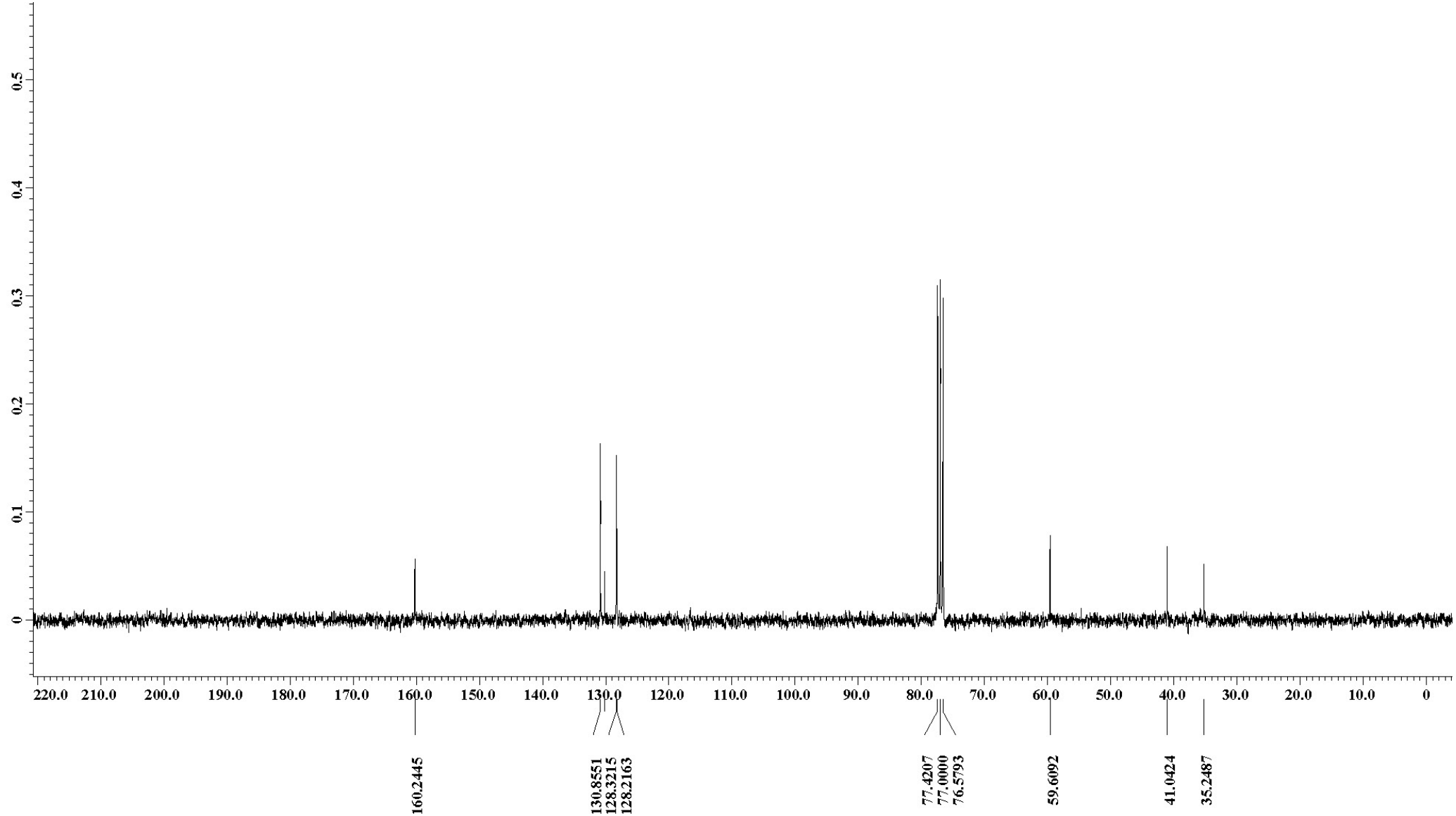


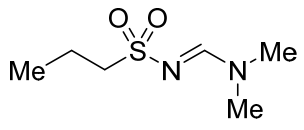
12s, ^{1}H NMR (300 MHz, $CDCl_3$)



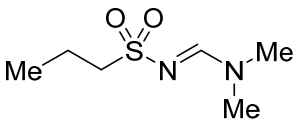


12s, ^{13}C NMR (75 MHz, $CDCl_3$)

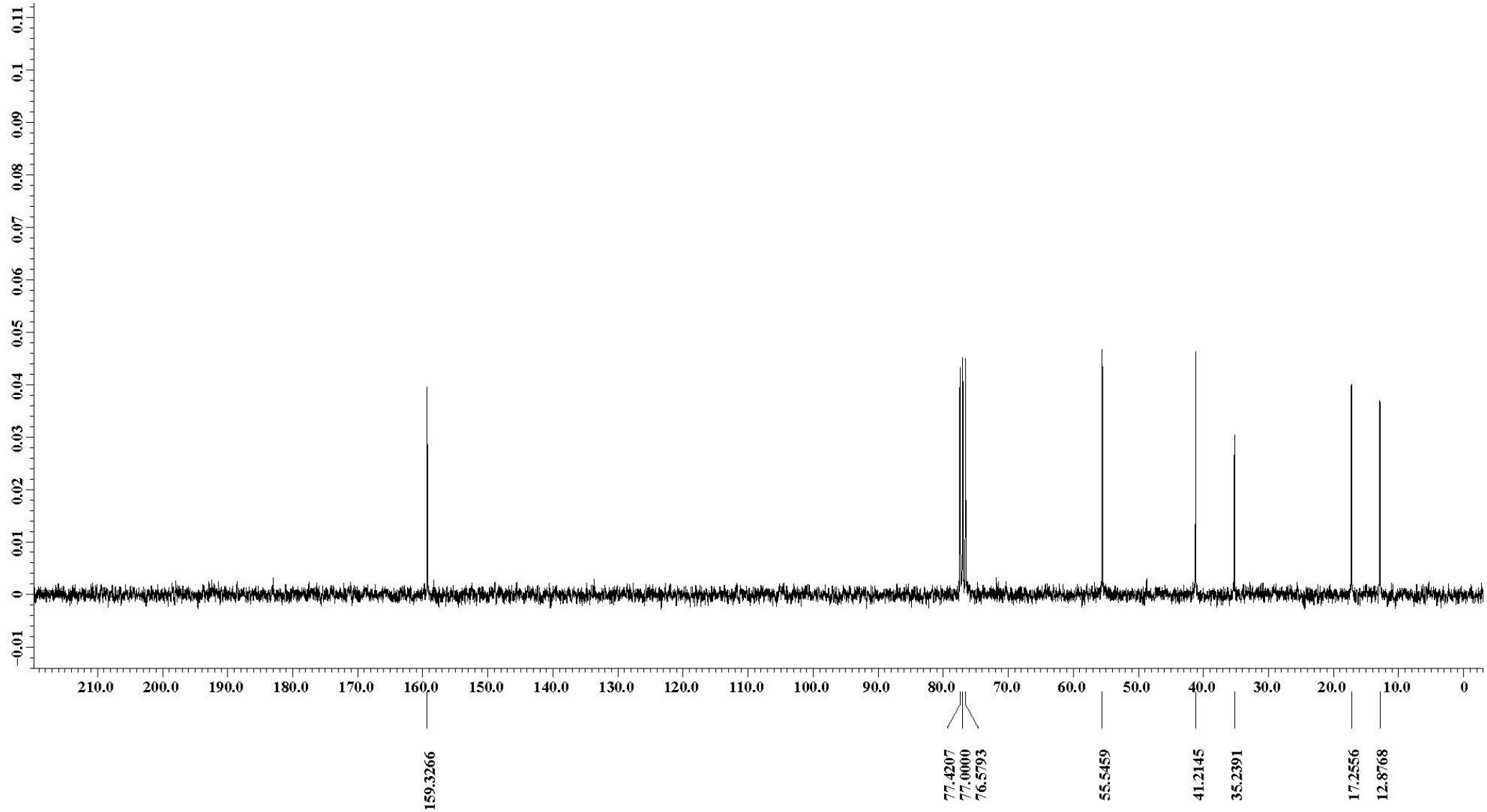


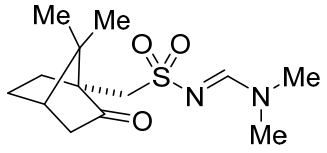


12t, ^{1}H NMR (300 MHz, $CDCl_3$)

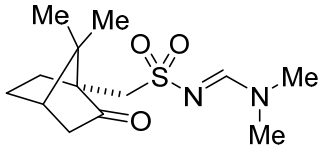


12t, ^{13}C NMR (75 MHz, $CDCl_3$)

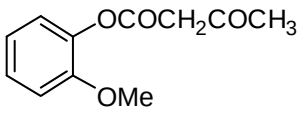




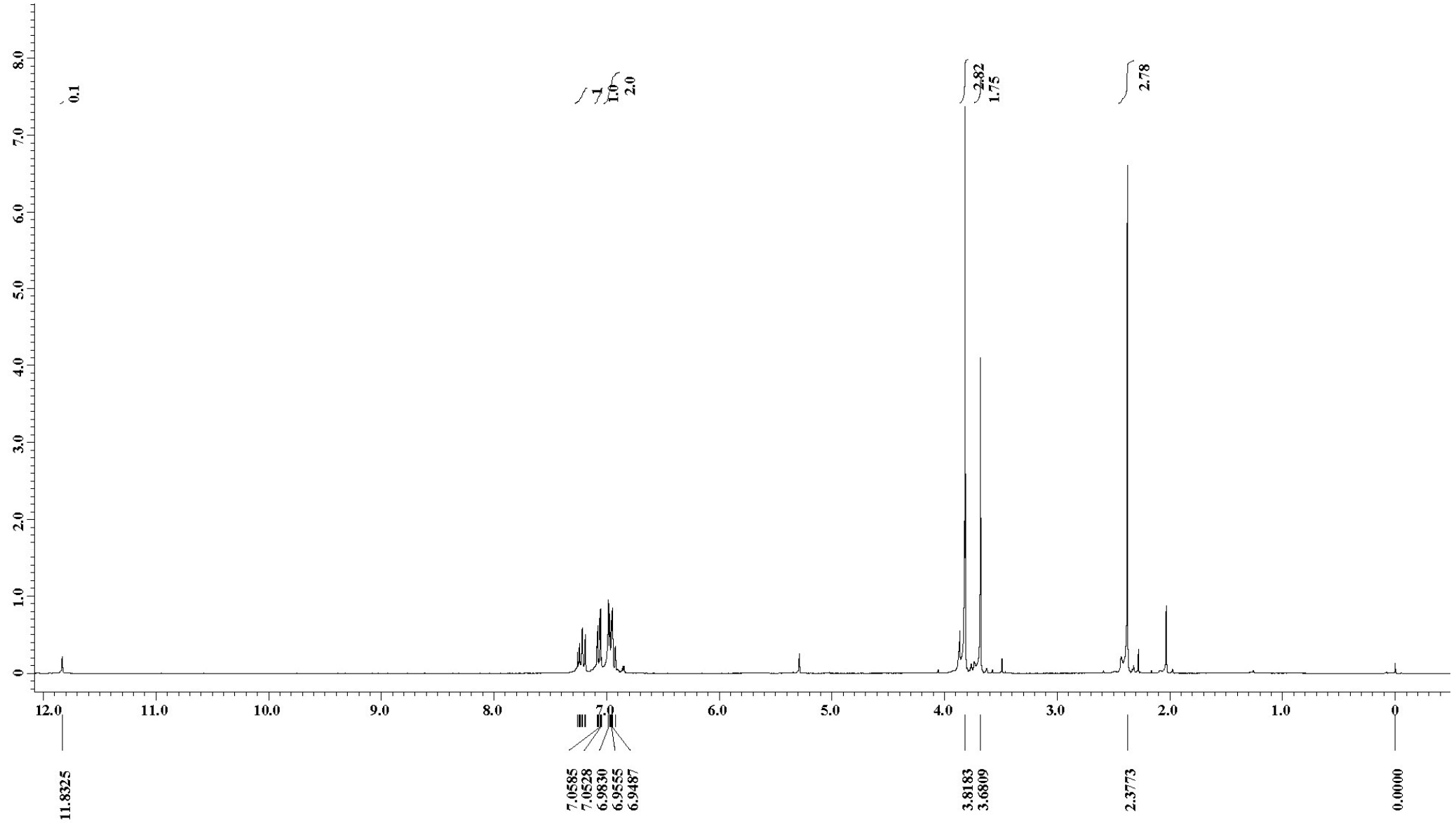
12u, ^{1}H NMR (300 MHz, $CDCl_3$)

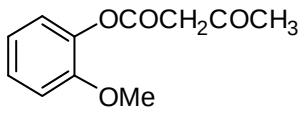


12u, ^{13}C NMR (75 MHz, $CDCl_3$)



14, ^{1}H NMR (300 MHz, $CDCl_3$)





14, ^{13}C NMR (75 MHz, $CDCl_3$)