SUPPORTING INFORMATION

Spirocyclization by Palladium-Catalyzed Domino Heck—Direct C-H Arylation Reactions: Synthesis of Spirodihydroquinolin-2-ones

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General information

Reagents and solvents were purchased from commercial sources (Aldrich, Acros, Merck, Fluka and VWR international). More sensitive compounds were stored in a desiccator or glove-box if required. Reagents were used without further purification unless otherwise noted.

All reactions were performed under argon (or nitrogen) and stirring unless otherwise noted. When needed, glassware was dried overnight in an oven (T > 100 °C) or under vacuum with a heat gun (T > 200 °C).

When solvents are indicated as dry they were either purchased as such, distilled prior to use or dried by a passage through a column of anhydrous alumina or copper using a Puresolv MD 5 from Innovative Technology Inc., based on the Grubbs' design [Pangborn, A. B. et al. *Organometallics* **1996**, *15*, 1518–1520].

Flash column chromatography was performed using Silicycle silica gel: 230-400 mesh (40-63 μm) silica. Reactions were monitored using Merck Kieselgel 60 F254 aluminium. TLC's were visualized by UV fluorescence (254 nm) then one of the following: KMnO4, ninhydrine, pancaldi, p-anisaldehyde or vanillin.

NMR spectra were recorded on a Brüker Avance III-400, Brüker Avance-400 or Brüker DPX-400 spectrometer at room temperature, ^{1}H frequency is at 400.13 MHz, ^{13}C frequency is at 100.62 MHz.

Chemical shifts (δ) were reported in parts per million (ppm) relative to residual solvent in $CDCl_3$ [^{1}H: 7.26, ^{13}C: 77.23]. Coupling constants (*J*) are reported in Hz to the nearest 0.1 Hz. Multiplicity is indicated as follows s (singlet), d (doublet), t (triplet), q (quartet), quint (quintet) m (multiplet) and br (broad). Attribution of peaks was done using the multiplicities and integrals of the peaks. When needed, COSY and/or HSQC experiments were used to confirm the attribution.

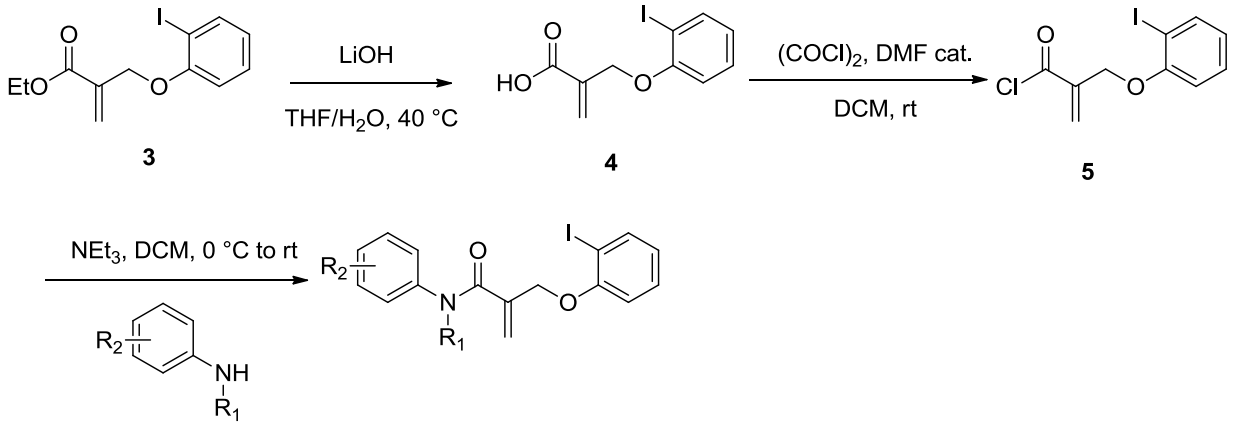
IR spectra were recorded in a Jasco FT/IR-4100 spectrometer outfitted with a PIKE technology

MIRacleTM ATR accessory as neat films compressed onto a Zinc Selenide window. The spectra are reported in cm^{-1}.

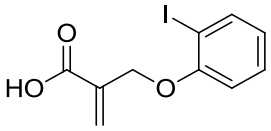
Mass spectra were determined with a Waters ACQUITY H-class UPLC/MS ACQ-SQD by electron ionisation (EI positive and negative) or a Finnigan TSQ7000 by electrospray ionization (ESI+). The accurate masses are done by the mass spectrometry service of the EPFL by ESI-TOF using a QTOF Ultima from Waters.

Melting points were determined using a Stuart SMP30 and were uncorrected.

Experimental procedure and spectroscopic data of compounds 3-4 and 1c-1l



*2-((2-Iodophenoxy)methyl)acrylic acid (**4**)*



To a solution of ethyl 2-((2-iodophenoxy)methyl)acrylate **3**[1] (4.2 g, 12.6 mmol, 1.0 equiv) in THF (12.0 mL), were added water (12.0 mL) and LiOH (900.0 mg, 37.8 mmol, 3.0 equiv). The mixture was stirred overnight at 40 °C. After completion of the reaction as shown by TLC, the THF was evaporated *in vacuo*, After cooling down to room temperature, the resulting mixture was extracted with ethyl ether. The aqueous layer was then acidified with 3 N aqueous HCl solutions (pH < 1.0), and extracted with ethyl acetate, dried over Na_2SO_4, evaporated under vacuum. The crude acrylic acid **4** were used directly for the subsequent reactions without further purification.

White solid. **M. p.** 110 °C. **R_f** (DCM/MeOH/AcOH 9:1:0.1) 0.65. **IR** (neat, cm^{-1}) ν 3673, 2900, 2871, 1690, 1435, 1056, 748. **^{1}H NMR** (400 MHz, CDCl3) δ 7.76 (dd, *J* = 1.6, 7.9 Hz, 1H), 7.31 (td, *J* = 1.1, 8.0 Hz, 1H), 6.93 (dd, *J* = 1.6, 8.0 Hz, 1H), 6.73 (td, *J* = 1.1, 7.9 Hz, 1H), 6.40 (d, *J* = 1.6 Hz, 1H), 6.20 (d, *J* = 1.6 Hz, 1H), 4.77 (s, 2H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.6, 158.4, 140.7, 137.8, 130.9, 127.0, 124.1, 113.7, 87.0, 68.3. **HRMS** m/z (ES^-) calcd for $[C_{10}H_8IO_3]^-$ 302.9524, found 302.9528.

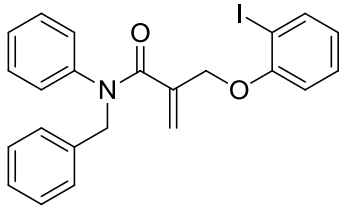
General Procedure A for the Synthesis of *N*-methyl-*N*-phenylacrylamide 1c-1l

Acrylic acid **4** (250 mg, 0.82 mmol, 1.0 equiv) was dissolved in dry dichloromethane (3.0 mL), followed by addition of oxalyl chloride (0.11 mL, 1.23 mmol, 1.5 equiv) and a drop of DMF. After 2 hours, the volatile were evacuated, the acid chloride **5** was obtained and used directly for the subsequent reactions without further purification.

[1] Szlosek-Pinauda, M.; Diazb, P.; Martineza, J.; Lamaty, F. *Tetrahedron* **2007**, *63*, 3340.

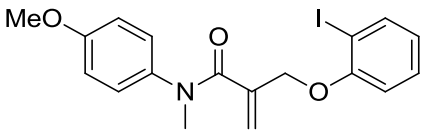
To a solution of *N*-substituted aniline (0.90 mmol, 1.0 equiv) and triethylamine (1.35 mmol, 1.5 equiv) in DCM (2.0 mL) was added dropwise at 0 °C a solution of freshly prepared acid chloride **5** (0.99 mmol, 1.1 equiv) in DCM (2.0 mL). The mixture was allowed to warm up at room temperature and stirred overnight. After completion of the reaction as shown by TLC, water was used to quench the reaction. The organic layer was extracted with ethyl acetate, washed successively with 1 N aqueous HCl solution, water, 1 N aqueous NaOH and brine. The crude product was purified by silica gel column chromatography to afford the corresponding product.

*N-benzyl-2-((2-iodophenoxy)methyl)-N-phenylacrylamide (***1c***)*



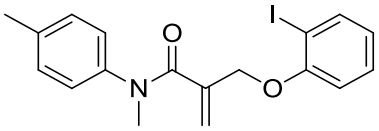
Yield = 58%. Yellow solid. **M. p.** 132 °C $\mathbf{R_f}$ (toluene/acetone 9:1) 0.23. **IR** (neat, cm^{-1}) ν 3675, 2987, 1617, 1497, 1397, 1051, 745, 700. **^{1}H NMR** (400 MHz, CDCl3) δ 7.78 (d, *J* = 7.5 Hz, 1H), 7.28-7.20 (m, 11H), 6.78 (d, *J* = 7.9 Hz, 1H). 6.71 (t, *J* = 7.5 Hz, 1H), 5.52 (s, 1H), 5.18 (s, 1H), 5.02 (s, 2H), 4.71 (s, 2H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.8, 157.1, 143.0, 139.7, 139.4, 137.2, 129.6, 129.3, 128.7, 128.1, 127.5, 127.4, 123.0, 122.3, 112.5, 86.3, 69.8, 53.7. **HRMS** m/z (ES+) calcd for $C_{23}H_{21}INO_2$ 470.0617, found 470.0615.

*2-((2-iodophenoxy)methyl)-N-(4-methoxyphenyl)-N-methylacrylamide (***1d***)*



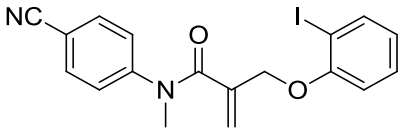
Yield = 62%. White solid. **M. p.** 119 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.13. **IR** (neat, cm^{-1}) ν 2969, 1623, 1512, 1248, 1017, 906, 727. **^{1}H NMR** (400 MHz, CDCl3) δ 7.77 (dd, *J* = 1.4, 7.5 Hz, 1H), 7.28-7.21 (m, 3H), 6.85 (d, *J* = 8.4 Hz, 2H), 6.76-6.26 (m, 2H), 5.51 (s, 1H), 5.16 (s, 1H), 4.64 (s, 2H), 3.80 (s, 3H), 3.39 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 158.6, 157.1, 139.7, 139.6, 137.3, 129.6, 128.4, 123.0, 121.7, 114.6, 112.6, 86.4, 69.7, 55.7, 38.3. **HRMS** m/z (ES+) calcd for $C_{18}H_{19}INO_3$ 424.0410, found 424.0413.

*2-((2-Iodophenoxy)methyl)-N-methyl-N-(p-tolyl)acrylamide (***1e***)*



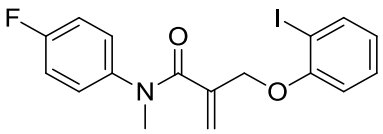
Yield = 53%. White solid. **M. p.** 119 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.25. **IR** (neat, cm^{-1}) ν 2929, 1654, 1626, 1513, 1380, 1240, 1017, 952, 760. **^{1}H NMR** (400 MHz, CDCl3) δ 7.76 (dd, *J* = 1.7, 7.7 Hz, 1H), 7.25 (td, *J* = 1.7, 7.7 Hz, 1H), 7.19 (d, *J* = 8.4 Hz, 2H), 7.13 (d, *J* = 8.4 Hz, 2H), 6.75 (dd, *J* = 1.3, 8.2 Hz, 1H), 6.70 (td, *J* = 1.3, 7.7 Hz, 1H), 5.51 (s, 1H), 5.16 (s, 1H), 4.66 (s, 2H), 3.39 (s, 3H), 2.34 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 157.1, 141.7, 139.6, 129.4, 137.41, 130.1, 129.6, 126.9, 122.9, 121.7, 112.5, 86.3, 69.6, 38.2, 21.2. **HRMS** m/z (ES+) calcd for $C_{18}H_{19}INO_2$ 408.0460, found 408.0461.

*N-(4-cyanophenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1f***)*



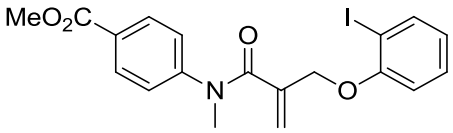
Yield = 15%. Yellow solid. **M. p.** 112 °C. $\mathbf{R_f}$ (toluene/acetone 9:1) 0.15. **IR** (neat, cm^{-1}) ν 2926, 2358, 2228, 1660, 1601, 1377, 1277, 1018, 751. **^{1}H NMR** (400 MHz, CDCl3) δ 7.79 (dd, *J* = 1.5, 8.0 Hz, 1H), 7.65 (d, *J* = 8.6 Hz, 2H), 7.54 (d, *J* = 8.6 Hz, 2H), 7.30 (td, *J* = 1.5, 8.1 Hz, 1H), 6.84 (dd, *J* = 1.3, 8.1 Hz, 1H), 6.75 (td, *J* = 1.3, 8.0 Hz, 1H), 5.58 (s, 1H), 5.10 (s, 1H), 4.75, (s, 2H), 3.47 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 157.0, 148.5, 139.5, 139.8, 139.1, 133.4, 129.8, 127.5, 123.4, 123.2, 118.4, 112.6, 86.3, 70.1, 37.9. **HRMS** m/z (ES+) calcd for $C_{18}H_{16}IN_2O_2$ 419.0256, found 419.0257.

*N-(4-fluorophenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1g***)*



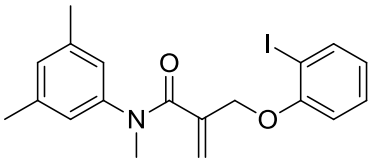
Yield = 18%. Colorless oil. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.15. **IR** (neat, cm^{-1}) ν 3675, 2920, 1661, 1510, 1240, 1017, 839, 751. **^{1}H NMR** (400 MHz, CDCl3) δ 7.78 (dd, *J* = 1.4, 7.5 Hz, 1H), 7.34-7.26 (m, 3H), 7.03 (t, *J* = 8.8 Hz, 2H), 6.79 (dd, *J* = 1.1, 8.2 Hz, 1H), 6.73 (td, *J* = 1.1, 7.5 Hz, 1H). 5.54 (s, 1H), 5.13 (s, 1H), 4.68 (s, 2H), 3.41 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 161.4 (d, *J* = 240.5 Hz), 157.1, 140.5, 139.7, 139.4, 129.7, 128.9 (d, *J* = 8.1Hz), 123.1, 122.1, 116.4 (d, *J* = 22.5 Hz), 112.6, 86.4, 70.0, 38.4. **HRMS** m/z (ES+) calcd for $C_{17}H_{16}FINO_2$ 412.0210, found 412.0206.

*Methyl 4-(2-((2-iodophenoxy)methyl)-N-methylacrylamido)benzoate (***1h***)*



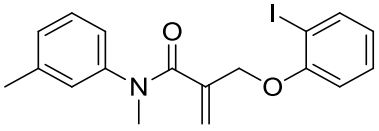
Yield = 49%. Yellow solid. **M. p.** 99 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.13. **IR** (neat, cm^{-1}) ν 2987, 1715, 1598, 1436, 1275, 1111, 1010, 753. **^{1}H NMR** (400 MHz, CDCl3) δ 8.02 (d, *J* = 8.5 Hz, 2H), 7.78 (d, *J* = 8.0 Hz, 1H), 7.45 (d, *J* = 8.5 Hz, 2H), 7.28 (t, *J* = 8.0 Hz, 1H). 6.82 (d, *J* = 8.0 Hz, 1H), 6.73 (t, *J* = 8.0 Hz, 1H), 5.54 (s, 1H), 5.11 (s, 1H), 4.72 (s, 2H), 3.92 (s, 3H), 3.46 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 166.5, 157.1, 148.7, 139.8, 139.2, 130.9, 129.7, 128.6, 126.8, 123.2, 122.8, 112.6, 86.4, 69.9, 52.5, 38.0. **HRMS** m/z (ES+) calcd for $C_{19}H_{19}INO_4$ 452.0359, found 452.0359.

*N-(3,5-dimethylphenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1i***)*



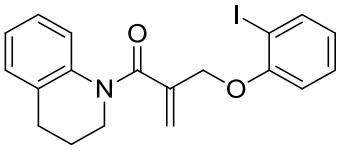
Yield = 93%.White solid. **M. p.** 98 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 9:1) 0.13. **IR** (neat, cm^{-1}) ν 2919, 1625, 1430, 1380, 1016, 930, 760. **^{1}H NMR** (400 MHz, CDCl3) δ 7.77 (dd, *J* = 1.7, 8.0 Hz, 1H), 7.25 (td, *J* = 1.7, 8.0 Hz, 1H), 6.89 (s, 1H), 6.88 (s, 2H), 6.73-6.69 (m, 2H), 5.51 (s, 1H), 5.24 (s, 1H), 4.63, (s, 2H), 3.39 (s, 3H), 2.27 (s, 6H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.7, 157.1, 144.3, 139.7, 139.7, 139.3, 129.6, 129.0, 124.7, 122.9, 121.5, 112.5, 86.4, 69.7, 38.4, 21.4. **HRMS** m/z (ES+) calcd for $C_{19}H_{21}INO_2$ 422.0617, found 422.0624

*2-((2-Iodophenoxy)methyl)-N-methyl-N-(m-tolyl)acrylamide (***1j***)*



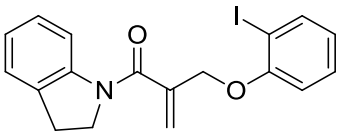
Yield = 66%. White solid. **M. p.** 80 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.19. **IR** (neat, cm^{-1}) ν 3674, 2972, 1619, 1384, 1227, 1020, 766. **^{1}H NMR** (400 MHz, CDCl3) δ 7.76 (dd, *J* = 1.7, 7.7 Hz, 1H), 7.27-7.20 (m, 2H), 7.10-7.05 (m, 3H), 6.75-6.69 (m, 2H), 5.51 (s, 1H), 5.19 (s, 1H), 4.64 (s, 2H), 3.40 (s, 3H), 2.31 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.8, 157.1, 144.4, 139.7, 139.6, 139.6, 129.6, 129.3, 128.1, 127.6, 124.3, 123.0, 121.7, 112.5, 86.4, 69.7, 38.3, 21.5. **HRMS** m/z (ES+) calcd for $C_{18}H_{19}INO_2$ 408.0460, found 408.0461.

*1-(3,4-dihydroquinolin-1(2H)-yl)-2-((2-iodophenoxy)methyl)prop-2-en-1-one (***1k***)*



Yield = 56%. White solid. **M. p.** 132 °C **R_f** (toluene/acetone 9:1) 0.13. **IR** (neat, cm^{-1}) ν 3673, 2926, 1624, 1492, 1388, 1244, 1053. **^{1}H NMR** (400 MHz, CDCl3) δ 7.77 (dd, *J* = 1.5, 7.8 Hz, 1H), 7.54 (d, *J* = 8.1 Hz, 1H), 7.29 (td, *J* = 2.0, 8.1 Hz, 1H), 7.15-7.12 (m, 2H), 7.08 (t, *J* = 8.1 Hz, 1H), 6.86 (d, *J* = 8.5 Hz, 1H). 6.73 (t, *J* = 7.7 Hz, 1H), 5.65 (s, 1H), 5.26 (s, 1H), 4.79 (s, 2H), 3.90 (t, *J* = 6.7 Hz, 2H), 2.77 (t, *J* = 6.7 Hz, 2H), 2.03 (quint, *J* = 6.7 Hz, 2H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.8, 157.2, 140.0, 139.7, 139.0, 131.8, 129.7, 128.5, 126.2, 125.7, 125.2, 123.1, 121.6, 112.6, 86.5, 69.7, 44.4, 27.0, 24.3. **HRMS** m/z (ES+) calcd for $C_{19}H_{19}INO_2$ 412.0210, found 412.0461.

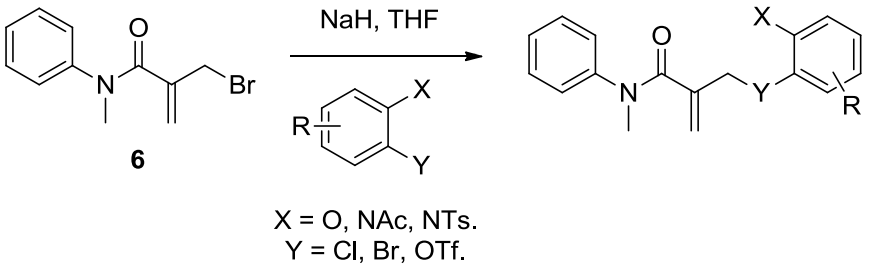
*1-(indolin-1-yl)-2-((2-iodophenoxy)methyl)prop-2-en-1-one (***1l***)*



Yield = 58%. White solid. **M. p.** 146 °C. **R_f** (petroleum ether/EtOAc 9:1) 0.22. **IR** (neat, cm^{-1}) ν 2924, 1657, 1612, 1459, 1356, 1133, 980, 841, 749. **^{1}H NMR** (400 MHz, CDCl3) δ 8.11 (s, 1H), 7.75 (d, *J* = 8.1 Hz, 1H), 7.29 (t, *J* = 8.1 Hz, 1H), 7.23-7.18 (m, 2H), 7.05 (t, *J* = 7.1 Hz, 1H), 6.88 (d, *J* = 8.0 Hz, 1H), 6.72 (t, *J* = 8.1 Hz, 1H), 5.83 (s, 1H), 5.57 (s, 1H), 4.86 (s, 2H), 4.28 (t, *J* = 8.3 Hz, 2H), 3.16 (t, *J* = 8.3 Hz, 2H). **^{13}C NMR** (75 MHz, CDCl3) δ 167.4, 156.9, 142.5, 141.0, 139.7, 132.7, 129.7, 127.6, 125.0, 124.5, 123.2, 118.8, 117.6, 112.5, 86.5, 69.6, 50.7, 28.4. **HRMS** m/z (ES+) calcd for $C_{18}H_{17}INO_2$ 406.0304, found 406.0298.

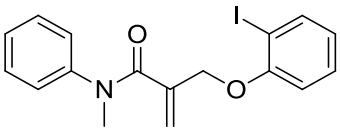
Experimental procedure and spectroscopic data of compounds 1a (X = I, Br, Cl, OTf), 1p-1r and 1t-1y

General Procedure B for the Synthesis of *N*-methyl-*N*-phenylacrylamide



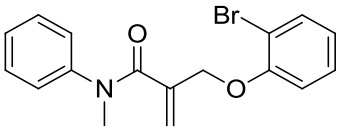
To a solution of aryl halide **5**[2] (1.0 equiv.) in THF (0.5 M) was added sodium hydride (60%, 1.1 equiv) and the mixture was stirred at room temperature for 30 min. Allyl halide **6** (1.2 equiv) was then added and the mixture was stirred until the reaction was judged to be completed by TLC analysis. Water was added and the mixture was diluted with ethyl acetate. The layers were separated and the organic layer was washed with water (x3) and brine, dried over anhydrous Na_2SO_4 and concentrated under reduced pressure. The crude product was purified by silica gel column chromatography to afford the corresponding product.

*2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a***)*



Yield = 84%. White solid. **M. p.** 89 °C. $\mathbf{R_f}$ (Petroleum ether/EtOAc 7:3) 0.39. **IR** (neat, cm^{-1}) ν 2928, 1657, 1626, 1473, 1239, 1010, 749. **^{1}H NMR** (400 MHz, CDCl3) δ 7.76 (dd, *J* = 1.7, 7.7 Hz, 1H), 7.37-7.24 (m, 6H), 6.76 (dd, *J* = 1.3, 8.4 Hz, 1H), 6.70 (td, *J* = 1.3, 7.7 Hz, 1H), 5.52 (s, 1H), 5.15 (s, 1H), 4.67 (s, 2H), 3.43 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 157.1, 144.5, 139.7, 139.4, 129.7, 129.5, 127.2, 127.2, 123.0, 122.0, 112.6, 86.4, 69.7, 38.3. **HRMS** m/z (ES+) calcd for $C_{17}H_{17}INO_2$ 394.0304, found 394.0298.

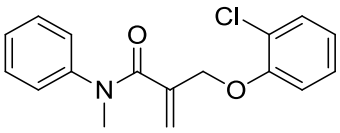
*2-((2-bromophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a, X = Br***)*



[2] Jaegli, S.; Vors, J.-P.; Neuville, L.; Zhu, J. *Tetrahedron* **2010**, *66*, 8911.

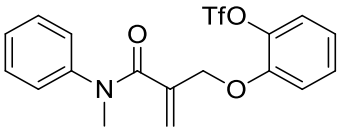
Yield = 74%. White solid. **M. p.** 98 °C. **R_f** (Petroleum ether/EtOAc 8:2) 0.27. **IR** (neat, cm^{-1}) ν 2928, 1625, 1594, 1479, 1382, 1243, 1031, 748, 699. **^{1}H NMR** (400 MHz, CDCl3) δ 7.55 (dd, *J* = 1.6, 8.3 Hz, 1H), 7.37-7.32 (m, 4H), 7.28-7.25 (m, 2H), 6.87-6.82 (m, 2H), 5.48 (s, 1H), 5.13 (s, 1H), 4.70 (s, 2H), 3.42 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.8, 154.9, 144.5, 139.4, 133.5, 129.5, 128.6, 127.2, 127.1, 122.3, 121.8, 113.4, 112.1, 69.6, 38.1. **HRMS** m/z (ES+) calcd for $C_{17}H_{17}BrNO_2$ 346.0443, found 346.0441.

*2-((2-chlorophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a, X = Cl***)*



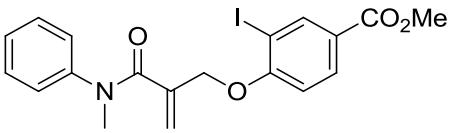
Yield = 59%. White solid. **M. p.** 94 °C. **R_f** (Petroleum ether/EtOAc 9:1) 0.14. **IR** (neat, cm^{-1}) ν 2940, 1656, 1627, 1480, 1382, 1242, 1010, 760, 701. **^{1}H NMR** (400 MHz, CDCl3) δ 7.37-7.32 (m, 5H), 7.28-7.25 (m, 1H), 7.18 (td, *J* = 1.2,8.6 Hz, 1H), 6.91-6.90 (m, 2H), 5.45 (s, 1H), 5.12 (s, 1H), 4.71 (s, 2H), 3.42 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 169.0, 154.0, 144.5, 139.4, 130.4, 129.5, 129.3, 127.9, 127.2, 127.1, 122.8, 121.8, 113.5, 69.6, 38.0. **HRMS** m/z (ES+) calcd for $C_{17}H_{17}ClNO_2$ 302.0948, found 302.0951.

*2-((2-(methyl(phenyl)carbamoyl)allyl)oxy)phenyl trifluoromethanesulfonate (***1a, X = OTf***)*



Yield = 69%. White solid. **M. p.** 215 °C. **R_f** (petroleum ether/EtOAc 8:2) 0.28. **IR** (neat, cm^{-1}) ν 2971, 1627, 1496, 1420, 1205, 1140, 1097, 890, 758. **^{1}H NMR** (400 MHz, CDCl3) δ 7.35 (t, *J* = 8.1 Hz, 2H), 7.29-7.21 (m, 5H), 7.02 (d, *J* = 8.4 Hz, 1H), 6.98 (t, *J* = 8.4 Hz, 1H), 5.51 (s, 1H), 5.16 (s, 1H), 4.73 (s, 2H), 3.40 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.6, 150.4, 144.4, 138.9, 138.9, 129.6, 129.4, 127.4, 127.0, 122.5, 122.1, 121.5, 118.9 (q, *J* = 321.2 Hz), 114.8, 69.7, 38.2. **HRMS** m/z (ES+) calcd for $[C_{18}H_{17}F_3NO_5S]^+$ 416.0774, found 416.0777.

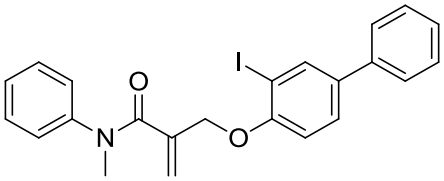
*methyl 3-iodo-4-((2-(methyl(phenyl)carbamoyl)allyl)oxy)benzoate (***1p***)*



Yield = 71%. Colorless oil. **R_f** (petroleum ether/EtOAc 8:2) 0.23. **IR** (neat, cm^{-1}) ν 2949, 1712, 1657, 1594, 1493, 1290, 1263, 991, 758, 700. **^{1}H NMR** (400 MHz, CDCl3) δ 8.45 (d, *J* = 1.8 Hz, 1H), 7.96 (dd, *J* = 1.8, 8.7 Hz, 1H), 7.37-7.25 (m, 5H), 6.75 (d, *J* = 8.7 Hz, 1H), 5.51 (s,

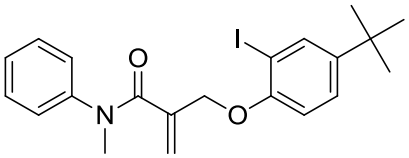
1H), 5.18 (s, 1H), 4.72 (s, 2H), 3.88 (s, 3H), 3.42 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.5, 165.6, 160.6, 144.4, 141.2, 138.8, 131.7, 129.5, 127.3, 127.1, 124.8, 122.5, 111.4, 85.6, 69.9, 52.3, 38.3. **HRMS** m/z (ES+) calcd for $C_{19}H_{19}INO_4$ 452.0359, found 452.0347.

*2-(((3-iodo-[1,1'-biphenyl]-4-yl)oxy)methyl)-N-methyl-N-phenylacrylamide (***1q***)*



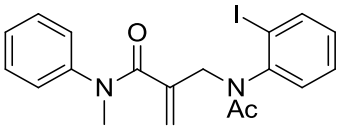
Yield = 71%. Colorless oil. **R_f** (petroleum ether/EtOAc 8:2) 0.34. **IR** (neat, cm^{-1}) ν 3058, 2932, 1624, 1478, 1382, 1274, 1014, 762. **^{1}H NMR** (400 MHz, CDCl3) δ 8.03 (d, *J* = 2.2 Hz, 1H), 7.53-7.48 (m, 3H), 7.42 (t, *J* = 7.6 Hz, 2H), 7.39-7.26 (m, 6H), 6.83 (d, *J* = 8.3 Hz, 1H), 5.54 (s, 1H), 5.18 (s, 1H), 4.72 (s, 2H), 3.45 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.8, 156.5, 144.4, 139.4, 139.4, 138.1, 136.2, 129.5, 129.0, 128.2, 127.3, 127.2, 127.1, 126.9, 122.0, 112.5, 86.8, 69.8, 38.2. **HRMS** m/z (ES+) calcd for $C_{23}H_{21}INO_2$ 470.0617, found 470.0622.

*2-((4-(tert-butyl)-2-iodophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1r***)*



Yield = 85%. Colorless oil. **R_f** (petroleum ether/EtOAc 8:2) 0.27. **IR** (neat, cm^{-1}) ν 2954, 1627, 1594, 1494, 1384, 1255, 1042, 933, 697. **^{1}H NMR** (400 MHz, CDCl3) δ 7.76 (d, *J* = 2.9 Hz, 1H), 7.36-7.26 (m, 6H), 6.71 (d, *J* = 9.0 Hz, 1H), 5.51 (s, 1H), 5.14 (s, 1H), 4.66 (s, 2H), 3.43 (s, 3H), 1.20 (s, 9H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 154.9, 146.1, 144.5, 139.6, 136.7, 129.4, 127.1, 126.5, 121.7, 112.0, 86.3, 69.8, 38.2, 34.2, 31.6. **HRMS** m/z (ES+) calcd for $C_{21}H_{25}INO_2$ 450.0930, found 450.0927.

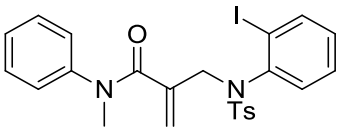
*2-((N-(2-iodophenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1t***)*



Yield = 65%. Yellow oil. **R_f** (petroleum ether/EtOAc 5:5) 0.26. **IR** (neat, cm^{-1}) ν 2922, 1648, 1632, 1389, 1283, 1016, 933, 769, 697. **^{1}H NMR** (400 MHz, CDCl3) δ 7.90 (d, *J* = 7.7 Hz, 1H), 7.60 (d, *J* = 7.7 Hz, 1H), 7.40-7.33 (m, 3H), 4.27-7.24 (m, 3H), 7.06 (t, *J* = 7.7 Hz, 1H), 5.20 (s, 1H), 5.00-4.96 (m, 2H), 3.31 (s, 3H), 3.12 (d, *J* = 13.1 Hz, 1H), 1.75 (s, 3H). **^{13}C NMR**

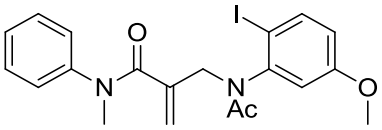
(75 MHz, CDCl3) δ 170.4, 169.9, 144.5, 143.9, 140.7, 140.0, 131.7, 130.1, 129.6, 129.4, 127.4, 121.4, 100.1, 51.4, 37.8, 22.9. **HRMS** m/z (ES+) calcd for $C_{19}H_{20}IN_2O_2$ 435.0569, found 435.0558.

*2-((N-(2-iodophenyl)-4-methylphenylsulfonamido)methyl)-N-methyl-N-phenylacrylamide (***1u***)*



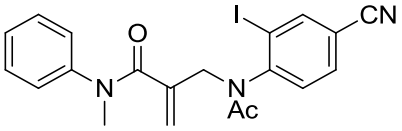
Yield = 86%. Orange oil. $\mathbf{R_f}$ (petroleum ether/EtOAc 7:3) 0.22. **IR** (neat, cm^{-1}) ν 3661, 2956, 1712, 1625, 1595, 1494, 1382, 1256, 1042, 770. **^{1}H NMR** (400 MHz, CDCl3) δ 7.85 (dd, *J* = 1.1, 8.0 Hz, 1H), 7.55 (d, *J* = 8.5 Hz, 2H), 7.29-7.20 (m, 6H), 7.13 (dd, *J* = 1.2, 8.1 Hz, 1H), 7.02-6.98 (m, 3H), 5.43 (s, 1H), 5.28 (s, 1H), 4.39 (d, *J* = 17.6 Hz, 1H), 4.00 (d, *J* = 17.6 Hz, 1H), 3.25 (s, 3H), 2.41 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 169.1, 144.0, 143.9, 141.5, 140.7, 139.8, 136.4, 132.6, 130.0, 129.6, 129.5, 128.7, 128.4, 127.2, 126.7, 124.4, 101.4, 53.6, 38.2, 21.8. **HRMS** m/z (ES+) calcd for $C_{24}H_{24}IN_2O_3S$ 547.0552, found 547.0553.

*2-((N-(2-iodo-5-methoxyphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1v***)*



Yield = 83%. Colorless oil. $\mathbf{R_f}$ (petroleum ether/EtOAc 6:4) 0.32. **IR** (neat, cm^{-1}) ν 2966, 2252, 1668, 1602, 1475, 1361, 1168, 907, 726. **^{1}H NMR** (400 MHz, CDCl3) δ 7.71 (d, *J* = 8.9 Hz, 1H), 7.37-7.24 (m, 6H), 6.67 (dd, *J* = 1.8, 8.9 Hz, 1H), 5.20 (s, 1H), 5.20-4.96 (m, 2H), 3.80 (s, 3H), 3.31 (s, 3H), 3.07 (d, *J* = 15.2 Hz, 1H), 1.79 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.3, 170.0, 160.8, 145.2, 143.8, 140.0, 139.8, 129.4, 127.5, 127.4, 121.3, 117.2, 116.9, 87.9, 55.9, 51.4, 37.8, 22.8. **HRMS** m/z (ES+) calcd for $[C_{20}H_{22}IN_2O_3]^+$ 465.0670, found 465.0674.

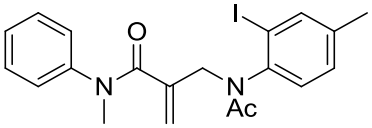
2-((N-(4-cyano-2-iodophenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide ***(1w)***



Yield = 87%. Colorless oil. $\mathbf{R_f}$ (petroleum ether/EtOAc 7:3) 0.11. **IR** (neat, cm^{-1}) ν 2923, 1650, 1593, 1495, 1376, 1291, 922, 729, 699. **^{1}H NMR** (400 MHz, CDCl3) δ 8.19 (d, *J* = 1.6 Hz, 1H), 7.94 (d, *J* = 8.4 Hz, 1H), 7.69 (dd, *J* = 1.6, 8.4 Hz, 1H), 7.39-7.27 (m, 5H), 5.15 (s, 1H), 5.05 (d, *J* = 12.6 Hz, 1H), 4.87 (s, 1H), 3.33 (s, 3H), 3.08 (d, *J* = 12.6 Hz, 1H), 1.76 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 169.5, 169.3, 148.3, 143.0, 140.1, 133.2, 132.4, 129.3, 127.5,

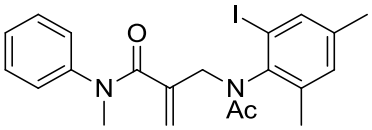
127.4, 121.4, 116.4, 114.0, 100.5, 51.2, 37.6, 22.8. **HRMS** m/z (ES+) calcd for $[C_{20}H_{19}IN_3O_2]^+$ 460.0516, found 460.0522.

*2-((N-(2-iodo-4-methylphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1x***)*



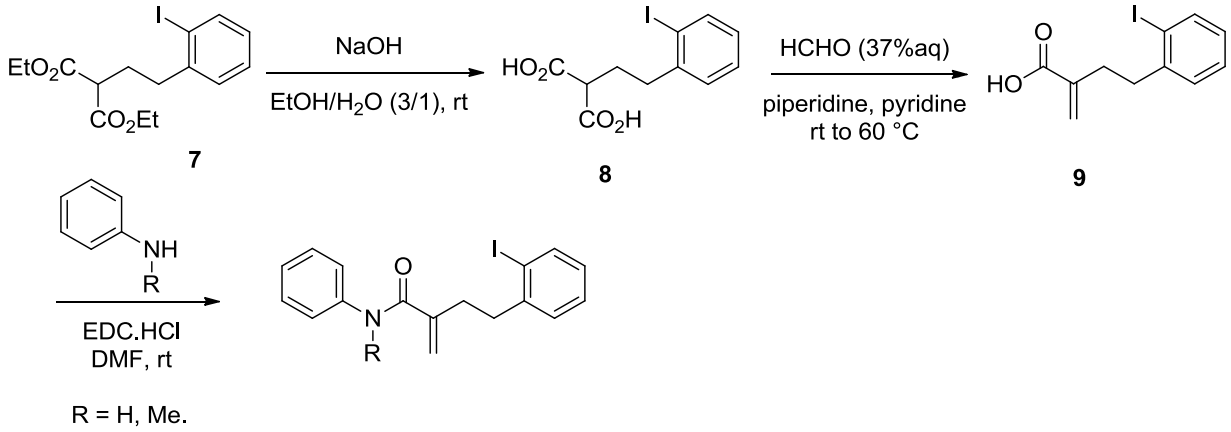
Yield = 77%. Yellow oil. $\mathbf{R_f}$ (petroleum ether/EtOAc 5:5) 0.17. **IR** (neat, cm^{-1}) ν 2930, 2359, 1659, 1586, 1381, 1303, 1232, 1029, 700. **^{1}H NMR** (400 MHz, CDCl3) δ 7.72 (d, *J* = 1.1 Hz, 1H), 7.46 (d, *J* = 7.9 Hz, 1H), 7.35 (t, *J* = 7.6 Hz, 2H), 7.28-7.24 (m, 3H), 7.19 (dd, *J* = 1.1, 7.9 Hz, 1H), 5.19 (s, 1H), 4.99-4.96 (m, 2H), 3.32 (s, 3H), 3.12 (d, *J* = 13.7 Hz, 1H), 2.33 (s, 3H), 1.76 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.7, 170.0, 143.9, 141.9, 140.8, 140.4, 140.3, 131.1, 130.4, 129.4, 127.4, 127.3, 121.2, 99.8, 51.5, 37.9, 22.9 20.8. **HRMS** m/z (ES+) calcd for $[C_{20}H_{22}IN_2O_2]^+$ 449.0720, found 449.0727.

*2-((N-(2-iodo-4,6-dimethylphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1y***)*

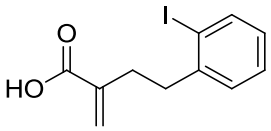


Yield = 56%. Colorless oil. $\mathbf{R_f}$ (petroleum ether/EtOAc 7:3) 0.14. **IR** (neat, cm^{-1}) ν 2973, 2233, 1668, 1627, 1380, 1293, 1076, 731. **^{1}H NMR** (400 MHz, CDCl3) δ 7.55 (s, 1H), 7.31 (t, *J* = 7.7 Hz, 2H), 7.21 (t, *J* = 7.7 Hz, 1H), 7.15 (d, *J* = 7.7 Hz, 2H), 7.02 (s, 1H), 5.34 (s, 1H), 5.26 (s,1H), 4.63 (d, *J* = 14.9 Hz, 1H), 3.30 (d, *J* = 14.9 Hz, 1H), 3.28 (s, 3H), 2.26 (s, 6H), 1.72 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 171.1, 169.9, 144.1, 141.8, 140.9, 140.0, 138.8, 138.5, 132.5, 129.4, 127.0, 127.0, 124.4, 100.6, 51.8, 38.2, 22.4, 20.6, 20.0. **HRMS** m/z (ES+) calcd for $[C_{21}H_{24}IN_2O_2]^+$ 463.0877, found 463.0884.

Experimental procedure and spectroscopic data of compounds 1b and 1s



4-(2-Iodophenyl)-2-methylenebutanoic acid (**9**)



To a solution of malonate **7**[3] (3.4 g, 8.7 mmol, 1.0 equiv) in a mixture of ethanol/water (3/1, 10.0 mL), was added sodium hydroxide (1.4 g, 34.9 mmol, 4.0 equiv). The mixture was stirred overnight at rt. After cooling down at rt, the ethanol was evaporated under vacuum, the resulting mixture was extracted with ethyl ether. The aqueous layer was then acidified with 3 N aqueous HCl solutions (pH < 1.0), extracted with ethyl acetate. the organic layer was dried over anhydrous Na_2SO_4, filtered, and concentrated under reduced pressure. The crude malonic acid **8** were used directly for the subsequent reactions without further purification.

To a solution of malonic acid **8** (2.2 g, 6.6 mmol, 1.0 equiv) in 22.0 mL of pyridine were added formaldehyde (37%aq) (0.7 mL, 8.6 mmol, 1.3 equiv) and piperidine (1.3 mL, 13.2 mmol, 2.0 equiv). The mixture was then stirred for 1 h at rt then 4 h at 60 °C. After completion of the reaction as shown by TLC, water was added and the pyridine was evaporated under vacuum. The mixture resulting mixture was extracted with ethyl acetate several times. The organic layer was washed with 1 N aqueous HCl, water and brine. The organic layer was dried over anhydrous Na_2SO_4, filtered, and concentrated under reduced pressure. The acrylic acid **9** was obtain as a white solid (1.8 g, 92%) and directly for the subsequent reactions without further purification.

M. p. 73 °C. $\mathbf{R_f}$ (DCM/MeOH/AcOH 9:1:0.1) 0.71. **IR** (neat, cm^{-1}) ν 3060, 2926, 2575, 1678, 1436, 1263, 951, 756. **^{1}H NMR** (400 MHz, CDCl3) δ 7.85 (d, *J* = 7.7 Hz, 1H), 7.30 (t, *J* = 7.2 Hz , 1H), 7.24 (dd, *J* = 1.5, 7.7 Hz, 1H), 6.92 (td, *J* = 1.5, 7.7 Hz, 1H), 6.38 (s, 1H), 5.71 (s,

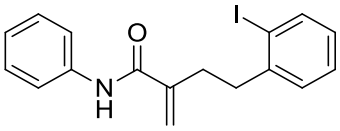
[3] Mukai, C.; Kuroda, N.; Ukon, R.; Itoh, I. *J. Org. Chem*. **2005**, *70*, 6282.

1H), 2.96 (t, *J* = 8.6 Hz, 2H), 2.63 (t, *J* = 8.6 Hz, 2H). **^{13}C NMR** (75 MHz, CDCl3) δ 172.7, 143.9, 139.7, 139.0, 129.9, 128.6, 128.5, 128.2, 100.7, 39.9, 32.3. **HRMS** m/z (ES+) calcd for $[C_{11}H_{12}IO_2]^+$ 302.9876, found 302.9530.

General procedure C for the synthesis of starting materials 1b and 1s

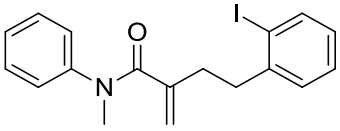
To a solution of acrylic acid **9** (1.0 equiv) and aniline (1.2 equiv) in DMF (*c* = 0.3 M), *N*-(3-dimethylaminopropyl)-*N*'-ethyl-carbodiimide hydrochloride (1.5 equiv) was added and the solution was stirred overnight at rt. The mixture was partitioned between water and ethyl acetate, the organic layer was washed with 1 N aqueuous HCl, water, 0.5 N aqueous NaOH, brine, dried over Na_2SO_4 and evaporated under vacuum. The crude material was chromatographed on silica using petroleum ether/AcOEt to give the desired compound.

*4-(2-Iodophenyl)-2-methylene-N-phenylbutanamide (***1b***)*



Yield = 96%. White solid. **M. p.** 98 °C. **R_f** (petroleum ether/EtOAc 8:2) 0.27. **IR** (neat, cm^{-1}) ν 3321, 2916, 1618, 1591, 1427, 1010, 754, 699. **^{1}H NMR** (400 MHz, CDCl3) δ 7.84 (dd, *J* = 0.8, 8.0 Hz, 1H), 7.59 (d, *J* = 7.6 Hz, 2H), 7.57 (brs, 1H), 7.37 (t, *J* = 7.6 Hz , 2H), 7.32-7.26 (m, 2H), 7.16 (t, *J* = 7.3 Hz , 1H), 6.91 (td, *J* = 0.8, 7.9 Hz, 1H), 5.80 (s, 1H), 5.47 (s, 1H), 2.99 (m, 2H), 2.73 (m, 2H).**^{13}C NMR** (75 MHz, CDCl3) δ 166.6, 145.1, 143.8, 139.7, 137.9, 130.0, 129.3, 128.7, 128.3, 124.7, 120.3, 119.4, 100.6, 39.9, 33.2. **HRMS** m/z (ES+) calcd for $C_{17}H_{17}INO$ 378.0355, found 378.0349.

*4-(2-Iodophenyl)-N-methyl-2-methylene-N-phenylbutanamide (***1s***)*

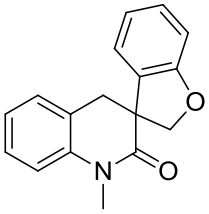


Yield = 65%. White solid. **M. p.** 72 °C. **R_f** (petroleum ether/EtOAc 8:2) 0.18. **IR** (neat, cm^{-1}) ν 3055, 16445, 1594, 1466, 1374, 1112, 1011, 920. **^{1}H NMR** (400 MHz, CDCl3) δ 7.79 (dd, *J* = 1.3, 7.9 Hz, 1H), 7.35 (t, *J* = 7.7 Hz, 2H), 7.28-7.23 (m, 2H), 7.18-7.15 (m, 3H), 6.87 (td, *J* = 1.8, 7.5 Hz, 1H), 5.13 (s, 1H), 5.10 (s, 1H), 3.37 (s, 3H), 2.82 (t, *J* = 8.3 Hz, 2H), 2.36 (t, *J* = 8.3 Hz, 2H). **^{13}C NMR** (75 MHz, CDCl3) δ 171.7, 144.6, 144.2, 143.9, 139.6, 129.6, 129.5, 128.6, 128.1, 127.2, 127.0, 119.0, 100.6, 39.2, 38.0, 34.2. **HRMS** m/z (ES+) calcd for $C_{18}H_{19}INO$ 392.0511, found 392.0506.

General Procedure for the Synthesis of Spirodihydroquinolin-2-ones

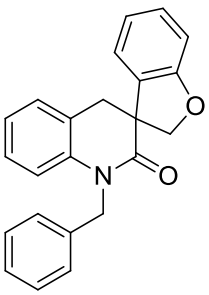
Anilide **2** (0.15 mmol, 1.0 equiv), $Pd(OAc)_2$ (0.0038 mmol, 0.025 equiv), Xphos (0.0075 mmol, 0.050 equiv) and K_2CO_3 (0.30 mmol, 2.0 equiv) were introduced in a screw cap vial equipped with a magnetic stir bar. The vial was purged with argon during 10 min. Degazed DMA (1.0 mL) was then added and the mixture was stirred at 100 °C until complete disappearance of the starting material as shown by TLC (usually 6-9 h). The reaction was next quenched with water and extracted with EtOAc. The combined organic layers were washed with brine, dried and concentrated under reduced pressure to give a residue which was purified by flash column chromatography.

*1'-Methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2a***)*



Yield = 85%. White solid. **M. p.** 140 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 9:1) 0.21. **IR** (neat, cm^{-1}) ν 2928, 1626, 1581, 1390, 1239, 1010, 749. **^{1}H NMR** (400 MHz, CDCl3) δ 7.37 (td, *J* = 1.7, 7.4 Hz, 1H), 7.16-7.06 (m, 4H), 6.82 (dd, *J* = 0.3, 8.2 Hz, 1H), 6.63 (td, *J* = 1.1, 7.4 Hz, 1H), 6.46 (dd, *J* = 1.1, 7.4 Hz, 1H), 5.20 (d, *J* = 8.9 Hz, 1H), 4.33 (d, *J* = 8.9 Hz, 1H), 3.46 (s, 3H), 3.23 (d, *J* = 15.4 Hz, 1H), 3.14 (d, *J* = 15.4 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.8, 159.9, 140.5, 129.8, 129.2, 129.0, 128.4, 124.0, 123.7, 123.6, 120.7, 114.8, 110.4, 79.2, 51.6, 37.8, 30.7. **HRMS** m/z (ES+) calcd for $C_{17}H_{16}NO_2$ 266.1181, found 266.1181.

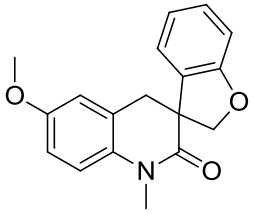
*1'-benzyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2c***)*



Yield = 90%. Yellow oil. $\mathbf{R_f}$ (petroleum ether/EtOAc 9:1) 0.38. **IR** (neat, cm^{-1}) ν 2933, 1667, 1609, 1462, 1376, 1188, 978, 751, 730. **^{1}H NMR** (400 MHz, CDCl3) δ 7.31-7.15 (m, 8H), 7.06-7.00 (m, 2H), 6.87 (dd, *J* = 1.4, 7.9 Hz, 1H), 6.67 (td, *J* = 1.2, 7.6 Hz, 1H), 6.55 (dd, *J* = 1.2, 7.7 Hz, 1H), 5.48 (d, *J* = 16.6 Hz, 1H), 5.35 (d, *J* = 9.5 Hz, 1H), 5.01 (d, *J* = 16.6 Hz, 1H), 4.42 (d, *J* = 9.5 Hz, 1H), 3.33 (d, *J* = 14.7 Hz, 1H), 3.22 (d, *J* = 14.7 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.9, 160.0, 139.9, 137.1, 129.9, 129.3, 129.0, 128.8, 128.3, 127.4, 126.6,

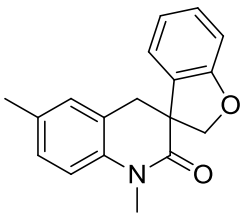
124.1, 123.7, 120.7, 115.6, 110.6, 79.1, 60.6, 51.6, 47.4, 38.0. **HRMS** m/z (ES+) calcd for $C_{23}H_{20}NO_2$ 342.1494, found 342.1416.

*6'-methoxy-1'-methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2d***)*



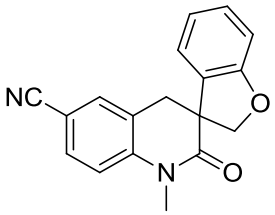
Yield = 97%. Yellow solid. **M. p.** 104 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.22. **IR** (neat, cm^{-1}) ν 3676, 2924, 1711, 1671, 1478, 1334, 1241, 978, 753. **^{1}H NMR** (400 MHz, CDCl3) δ 7.11 (t, *J* = 8.1 Hz, 1H), 7.00 (d, *J* = 9.2 Hz, 1H), 6.88 (dd, *J* = 2.3, 9.2 Hz, 1H), 6.81 (d, *J* = 8.1 Hz, 1H), 6.71 (d, *J* = 2.3 Hz, 1H), 6.64 (t, *J* = 7.6 Hz, 1H), 6.48 (d, *J* = 7.6 Hz, 1H), 5.20 (d, *J* = 8.6 Hz, 1H), 4.32 (d, *J* = 8.6 Hz, 1H), 3.80 (s, 3H), 3.43 (s, 3H), 3.20 (d, *J* = 15.6 Hz, 1H), 3.08 (d, *J* = 15.6 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.2, 159.9, 155.8, 134.0, 129.7, 129.1, 125.1, 124.1, 120.7, 115.8, 115.1, 112.9, 110.4, 79.2, 55.7, 51.5, 38.1, 30.8. **HRMS** m/z (ES+) calcd for $C_{18}H_{18}NO_2$ 296.1287, found 296.1281.

*1',6'-Dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2e***)*



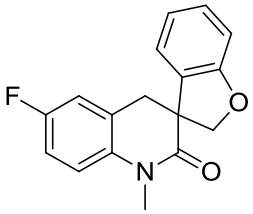
Yield = 92%. White solid. **M. p.** 174 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 9:1) 0.32. **IR** (neat, cm^{-1}) ν 2972, 1656, 1479, 1360, 1074, 977, 747. **^{1}H NMR** (400 MHz, CDCl3) δ 7.16 (d, *J* = 8.5 Hz, 1H), 7.12 (td, *J* = 0.3, 8.2 Hz, 1H), 6.97 (d, *J* = 8.2 Hz, 1H), 6.96 (s, 1H), 6.82 (dd, *J* = 0.3, 8.2 Hz, 1H), 6.64 (td, *J* = 1.1, 7.5 Hz, 1H), 6.48 (dd, *J* = 1.1, 7.5 Hz, 1H), 5.20 (d, *J* = 9.7 Hz, 1H), 4.33 (d, *J* = 9.7 Hz, 1H), 3.43 (s, 3H), 3.19 (d, *J* = 15.3 Hz, 1H), 3.09 (d, *J* = 15.3 Hz, 1H), 2.32 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.6, 159.9, 138.0, 133.2, 129.9, 129.7, 129.2, 128.7, 124.1, 123.4, 120.7, 114.7, 110.4, 79.2, 51.7, 37.8, 30.7, 20.8. **HRMS** m/z (ES+) calcd for $C_{18}H_{18}NO_2$ 280.1338, found 280.1335.

*1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-6'-carbonitrile (***2f***)*



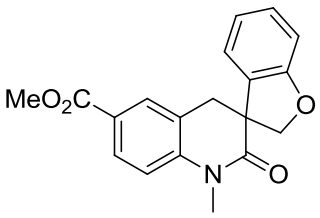
Yield = 76%. White solid. **M. p.** 103 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.25. **IR** (neat, cm^{-1}) ν 2927, 2224, 1667, 1605, 1473, 1353, 1130, 981, 833, 765. **^{1}H NMR** (400 MHz, CDCl3) δ 7.68 (dd, *J* = 1.7, 8.3 Hz, 1H), 7.44 (d, *J* = 1.7 Hz, 1H), 7.18-7.13 (m, 2H), 6.84 (d, *J* = 8.2 Hz, 1H), 6.65 (t, *J* = 7.3 Hz, 1H), 6.40 (d, *J* = 7.3 Hz, 1H), 5.19 (d, *J* = 8.8 Hz, 1H), 4.34 (d, *J* = 8.8 Hz, 1H), 3.47 (s, 3H), 3.25 (d, *J* = 16.2 Hz, 1H), 3.19 (d, *J* = 16.2 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.5, 159.8, 144.1, 132.9, 132.5, 130.3, 128.1, 124.7, 123.5, 121.0, 118.7, 115.2, 110.8, 106.9, 78.9, 51.2, 37.3, 30.9. **HRMS** m/z (ES+) calcd for $C_{18}H_{15}N_2O_2$ 291.1134, found 291.1133.

*6'-fluoro-1'-methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2g***)*



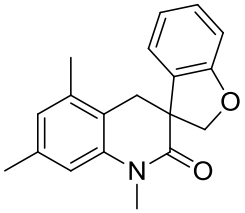
Yield = 44%. Yellow solid. **M. p.** 161 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 9:1) 0.21. **IR** (neat, cm^{-1}) ν 3661, 2920, 1656, 1505, 1479, 1242, 978, 748. **^{1}H NMR** (400 MHz, CDCl3) δ 7.13 (td, *J* = 1.4, 8.1 Hz, 1H), 7.09-7.01 (m, 2H), 6.89 (dd, *J* = 2.4, 8.1 Hz, 1H), 6.83 (d, *J* = 8.1 Hz, 1H), 6.65 (t, *J* = 7.6 Hz, 1H), 6.45 (d, *J* = 7.6 Hz, 1H), 5.20 (d, *J* = 9.0 Hz, 1H), 4.33 (d, *J* = 9.0 Hz, 1H), 3.44 (s, 3H), 3.22 (d, *J* = 14.3 Hz, 1H), 3.11 (d, *J* = 14.3 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.3, 159.8, 158.9 (d, *J* = 245.2 Hz), 136.7 (d, *J* = 2.1 Hz), 130.0, 128.7, 125.7, 123.8, 120.8, 116.3 (d, *J* = 22.1 Hz), 116.0 (d, *J* = 8.0 Hz), 114.7 (d, *J* = 22.1 Hz), 110.6, 79.1, 51.3, 37.7, 31.0. **HRMS** m/z (ES+) calcd for $C_{17}H_{15}FNO_2$ 284.1087, found 283.1009.

*Methyl 1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-6'-carboxylate (***2h***)*



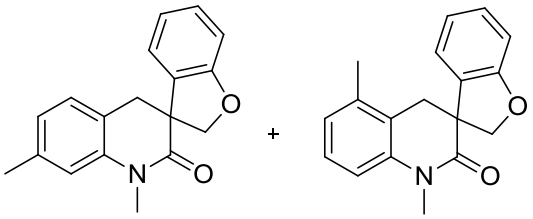
Yield = 91%. Yellow solid. **M. p.** 110 °C. **R_f** (petroleum ether/EtOAc 8:2) 0.33. **IR** (neat, cm^{-1}) ν 3660, 2925, 1711, 1672, 1264, 1110, 754. **^{1}H NMR** (400 MHz, CDCl3) δ 8.05 (dd, *J* = 1.9, 8.5 Hz, 1H), 7.83 (d, *J* = 1.9 Hz, 1H), 7.14-7.11 (m, 2H), 6.82 (d, *J* = 8.5 Hz, 1H), 6.62 (t, *J* = 7.7 Hz, 1H), 6.42 (d, *J* = 7.7 Hz, 1H), 5.20 (d, *J* = 9.3 Hz, 1H), 4.34 (d, *J* = 9.3 Hz, 1H), 3.90 (s, 3H), 3.48 (s, 3H), 3.26 (d, *J* = 15.8 Hz, 1H), 3.20 (d, *J* = 15.6 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.8, 166.6, 159.9, 144.2, 130.4, 130.3, 130.0, 128.5, 125.1, 123.8, 123.5, 120.8, 114.5, 110.6, 79.0, 52.3, 51.4, 37.5, 30.9. **HRMS** m/z (ES+) calcd for $C_{19}H_{18}NO_4$ 324.1236, found 324.1241.

*1',5',7'-trimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2i***)*



Yield = 93%. White solid. **M. p.** 126 °C. **R_f** (petroleum ether/EtOAc 9:1) 0.20. **IR** (neat, cm^{-1}) ν 2921, 1630, 1596, 1481, 1399, 1246, 1017, 751. **^{1}H NMR** (400 MHz, CDCl3) δ 7.12 (t, *J* = 8.3 Hz, 1H), 6.82 (d, *J* = 8.3 Hz, 1H), 6.79 (s, 1H), 6.77 (s, 1H), 6.65 (t, *J* = 7.2 Hz, 1H), 6.53 (d, *J* = 8.1 Hz, 1H), 5.16 (d, *J* = 8.1 Hz, 1H), 4.31 (d, *J* = 8.1 Hz, 1H), 3.41 (s, 3H), 3.16 (d, *J* = 16.6 Hz, 1H), 3.01 (d, *J* = 16.6 Hz, 1H), 2.38 (s, 3H), 2.18 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 167.4, 156.9, 142.5, 141.0, 139.7, 132.7, 129.8, 127.6, 125.0, 124.5, 123.2, 118.8, 117.6, 112.5, 86.5, 69.6, 50.7, 28.4. **HRMS** m/z (ES+) calcd for $C_{19}H_{20}NO_2$ 294.1494, found 294.1497.

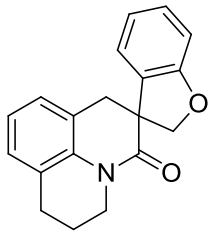
*1',7'-Dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one and 1',5'-dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2j** and **2j'***)*



Yield = 96%. Yellow solid. **M. p.** 128 °C. **R_f** (petroleum ether/EtOAc 9:1) 0.25. **IR** (neat, cm^{-1}) ν 2972, 1656, 1479, 1360, 1074, 977, 747. **^{1}H NMR** (400 MHz, CDCl3) δ 7.26 (t, *J* = 7.6 Hz, 1H-min), 7.13 (td, *J* = 1.0, 8.2 Hz, 1H-maj+ 1H-min), 7.03 (d, *J* = 7.4 Hz, 1H-maj), 6.98 (d, , *J* = 7.4 Hz, 1H-min), 6.97 (d, *J* = 8.2 Hz, 1H-min), 6.91 (s, 1H-maj), 6.90 (d, *J* = 8.2 Hz, 1H-maj), 6.83 (d, *J* = 8.1 Hz, 1H-min), 6.82 (d, *J* = 8.1 Hz, 1H-maj), 6.66 (td, *J* = 1.0, 7.2 Hz, 1H-min), 6.65 (td, *J* = 1.0, 7.2 Hz, 1H-maj), 6.52 (dd, *J* = 1.0, 7.2 Hz, 1H-min), 6.49 (dd, *J* = 1.0,

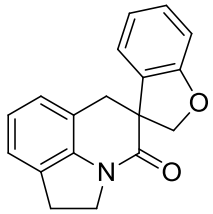
7.2 Hz, 1H-maj), 5.20 (d, *J* = 9.2 Hz, 1H-maj), 5.19 (d, *J* = 9.2 Hz, 1H-min), 4.34 (d, *J* = 9.2 Hz, 1H-min), 4.33 (d, *J* = 9.2 Hz, 1H-maj), 3.45 (s, 3H-maj+3H-min), 3.22 (d, *J* = 15.4 Hz, 1H-maj), 3.19 (d, *J* = 15.4 Hz, 1H-min), 3.10 (d, *J* = 15.4 Hz, 1H-maj), 3.07 (d, *J* = 15.4 Hz, 1H-min), 2.44 (s, 3H-maj), 2.24 (s, 3H-min). **^{13}C NMR** (75 MHz, CDCl3) δ 170.7 (maj), 170.4 (min), 159.7 (maj), 159.6 (min), 140.4 (min), 140.1 (maj), 138.0 (maj), 136.7 (min), 129.5 (maj+min), 129.1 (min), 129.0 (maj), 127.6 (min), 125.4 (min), 124.0 (maj), 123.9 (maj+min), 123.8 (maj), 122.1 (min), 120.9 (min), 120.5 (maj+min), 120.4 (min), 115.5 (maj), 122.8 (min), 110.2 (min), 110.2 (maj), 79.1 (min) , 79.0 (maj), 51.5 (maj), 51.2 (min) , 37.3 (maj), 34.1 (min), 30.8 (min), 30.5 (min), 21.7 (maj), 19.6 (min). **HRMS** m/z (ES+) calcd for C18H18NO2 280.1338, found 280.1338.

*6',7'-dihydro-1'H,2H-spiro[benzofuran-3,2'-pyrido[3,2,1-ij]quinolin]-3'(5'H)-one (***2k***)*



Yield = 48%. Colorless oil. **R_f** (petroleum ether/EtOAc 8:2) 0.50. **IR** (neat, cm^{-1}) ν 2936, 1665, 1593, 1477, 1375, 1243, 969. **^{1}H NMR** (400 MHz, CDCl3) δ 7.14-7.10 (m, 2H), 6.97-6.96 (m, 2H), 6.82 (d, *J* = 8.2 Hz, 1H), 6.65 (t, *J* = 7.4 Hz, 1H), 6.53 (dd, *J* = 1.0, 7.4 Hz, 1H), 5.23 (d, *J* = 8.7 Hz, 1H), 4.34 (d, *J* = 8.7 Hz, 1H), 4.19 (ddd, *J* = 5.1, 6.3, 13.1 Hz, 1H), 3.70 (ddd, *J* = 4.5, 7.0, 13.1 Hz, 1H), 3.19 (d, *J* = 15.1, 1H), 3.11 (d, *J* = 15.1, 1H), 2.87 (t, *J* = 5.6 Hz, 2H), 2.02-1.97 (m, 2H). **^{13}C NMR** (75 MHz, CDCl3) δ 169.9, 159.9, 136.0, 129.7, 129.3, 128.6, 127.2, 125.3, 123.9, 123.2, 123.0, 120.8, 110.4, 79.3, 51.3, 42.0, 37.8, 27.5, 21.8. **HRMS** m/z (ES+) calcd for $[C_{19}H_{18}INO_2]^+$ 292.1332, found 292.1336.

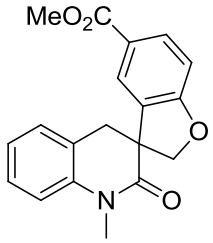
*1',2'-dihydro-2H-spiro[benzofuran-3,5'-pyrrolo[3,2,1-ij]quinolin]-4'(6'H)-one (***2l***)*



Yield = 38%. Colorless oil. **R_f** (petroleum ether/EtOAc 8:2) 0.37. **IR** (neat, cm^{-1}) ν 2938, 1666, 1596, 1480, 1392, 1247, 981, 751. **^{1}H NMR** (400 MHz, CDCl3) δ 7.18-7.12 (m, 2H), 6.99-6.98 (m, 2H), 6.84 (d, *J* = 8.0 Hz, 1H), 6.70-6.65 (m, 2H), 5.25 (d, *J* = 8.7 Hz, 1H), 4.37 (d, *J* = 8.7 Hz, 1H), 4.19 (ddd, *J* = 5.6, 10.5, 12.3 Hz, 1H), 4.10 (ddd, *J* = 7.6, 10.9, 12.3 Hz, 1H), 3.37-3.14 (m, 4H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.1, 159.5, 141.4, 130.4, 129.9, 128.9, 126.3,

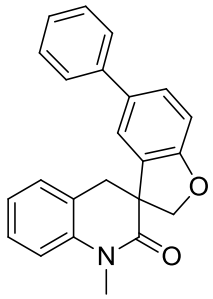
124.1, 124.1, 123.4, 120.9, 118.5, 110.5, 79.3, 53.2, 46.2, 38.3, 28.3. **HRMS** m/z (ES+) calcd for $C_{18}H_{16}NO_2$ 278.1181, found 278.1179.

*methyl 1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-5-carboxylate (***2p***)*



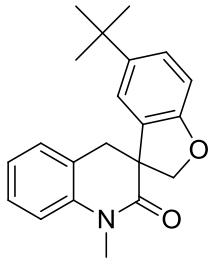
Yield = 97%. White solid. **M. p.** 156°C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.31. **IR** (neat, cm^{-1}) ν 2960, 1708, 1667, 1603, 1364, 1283, 1114, 986, 755. **^{1}H NMR** (400 MHz, CDCl3) δ 7.88 (dd, *J* = 2.0, 8.5, 1H), 7.39 (t, *J* = 7.7 Hz, 1H), 7.23 (d, *J* = 2.0 Hz, 1H), 7.17-7.08 (m, 3H), 6.83 (d, *J* = 8.5 Hz, 1H), 5.18 (d, *J* = 9.1 Hz, 1H), 4.41 (d, *J* = 9.1 Hz, 1H), 3.76 (s, 3H), 3.46 (s, 3H), 3.22 (d, *J* = 14.9 Hz, 1H), 3.16 (d, *J* = 14.9 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.3, 166.6, 163.9, 140.2, 132.5, 129.6, 129.2, 128.6, 126.2, 123.8, 123.1, 123.1, 115.1, 110.1, 80.1, 52.0, 51.4, 37.8, 30.8. **HRMS** m/z (ES+) calcd for $[C_{19}H_{18}NO_4]^+$ 342.1230, found 342.1234.

*1'-methyl-5-phenyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2q***)*



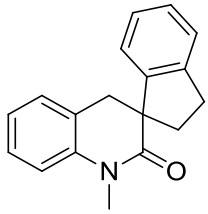
Yield = 83%. White solid. **M. p.** 167°C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.40. **IR** (neat, cm^{-1}) ν 2960, 1671, 1604, 1489, 1363, 1281, 1126, 977, 732. **^{1}H NMR** (400 MHz, CDCl3) δ 7.41-7.36 (m, 2H), 7.33-7.29 (m, 2H), 7.26-7.19 (m, 4H), 7.12 (t, *J* = 7.7 Hz, 2H), 6.89 (d, *J* = 8.5 Hz, 1H), 6.68 (d, *J* = 1.5 Hz, 1H), 5.25 (d, *J* = 8.7 Hz, 1H), 4.41 (d, *J* = 8.7 Hz, 1H), 3.47 (s, 3H), 3.28 (d, *J* = 15.2 Hz, 1H), 3.19 (d, *J* = 15.2 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.7, 159.6, 141.1, 140.5, 134.1, 129.7, 129.3, 128.9, 128.8, 128.4, 126.8, 126.7, 123.7, 123.7, 122.8, 114.9, 110.5, 79.5, 51.7, 37.9, 30.8. **HRMS** m/z (ES+) calcd for $C_{23}H_{20}NO_2$ 342.1494, found 342.1501.

*5-(tert-butyl)-1'-methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2r***)*



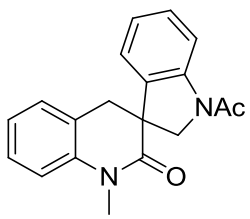
Yield = 76%. White solid. **M. p.** 167 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.43. **IR** (neat, cm^{-1}) ν 2925, 2364, 1669, 1604, 1476, 1365, 1126, 976, 909, 758. **^{1}H NMR** (400 MHz, CDCl3) δ 7.38 (t, *J* = 7.5 Hz, 1H), 7.16-7.06 (m, 4H), 6.73 (d, *J* = 8.4 Hz, 1H), 6.40 (d, *J* = 1.2 Hz, 1H), 5.23 (d, *J* = 8.2 Hz, 1H), 4.33 (d, *J* = 8.2 Hz, 1H), 3.47 (s, 3H), 3.24 (d, *J* = 16.0 Hz, 1H), 3.11 (d, *J* = 16.0 Hz, 1H), 1.04 (s, 9H). **^{13}C NMR** (75 MHz, CDCl3) δ 171.0, 157.6, 143.5, 140.7, 129.2, 128.3, 128.3, 126.6, 124.1, 123.5, 120.9, 114.7, 109.4, 79.2, 51.6, 37.8, 34.3, 31.6, 30.8. **HRMS** m/z (ES+) calcd for $C_{21}H_{24}NO_2$ 322.1807, found 322.1795.

*1'-Methyl-2,3-dihydro-1'H-spiro[indene-1,3'-quinolin]-2'(4'H)-one (***2s***)*



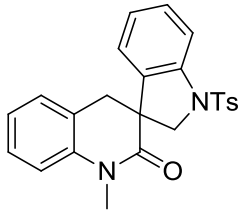
Yield = 53%. Light yellow solid. **M. p.** 130 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 8:2) 0.32. **IR** (neat, cm^{-1}) ν 2916, 2849, 1622, 1463, 1367, 1115, 752. **^{1}H NMR** (400 MHz, CDCl3) δ 7.33 (t, *J* = 7.5 Hz, 1H), 7.24 (d, *J* = 7.3 Hz, 1H), 7.19 (t, *J* = 7.3 Hz, 1H), 7.14 (d, *J* = 7.5 Hz, 1H) 7.06-7.03 (m, 3H), 6.81 (d, *J* = 7.6 Hz, 1H), 3.43 (s, 3H), 3.17 (d, *J* = 15.9 Hz, 1H), 3.10 (ddd, , *J* = 6.1, 8.1, 16.1 Hz, 1H), 3.05 (d, *J* = 15.9 Hz, 1H), 2.98 (ddd, *J* = 6.8, 8.6, 16.1 Hz, 1H), 2.59 (ddd, , *J* = 6.1, 8.6, 13.1 Hz, 1H), 1.99 (ddd, , *J* = 6.8, 8.1, 13.1 Hz, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 173.5, 145.1, 144.2, 140.7, 128.7, 127.9, 127.9, 126.7, 125.1, 125.0, 123.9, 123.0, 114.5, 54.0, 38.4, 36.2, 30.9, 30.4. **HRMS** m/z (ES+) calcd for $C_{18}H_{18}NO$ 264.1388, found 264.1386.

*1-Acetyl-1'-methyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2t***)*



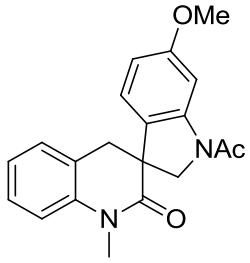
Yield = 92%. White solid. **M. p.** 183 °C. **R_f** (DCM/EtOAc 8:2) 0.28. **IR** (neat, cm^{-1}) ν 2987, 2359, 1666, 1480, 1403, 1338, 1127, 754. **^{1}H NMR** (400 MHz, CDCl3) δ 8.23 (d, *J* = 8.0 Hz, 1H), 7.38 (t, *J* = 7.5 Hz, 1H), 7.20 (t, *J* = 7.5 Hz, 1H), 7.14-7.07 (m, 3H), 6.78 (t, *J* = 7.6 Hz, 1H), 6.50 (d, *J* = 7.6 Hz, 1H), 4.80 (d, *J* = 11.0 Hz, 1H), 3.80 (d, *J* = 11.0 Hz, 1H), 3.47 (s, 3H), 3.25 (d, *J* = 15.6 Hz, 1H), 3.15 (d, *J* = 15.6 Hz, 1H), 2.26 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.6, 168.7, 142.5, 140.4, 132.5, 129.5, 129.2, 128.5, 123.8, 123.5, 123.3, 117.5, 114.9, 58.5, 49.8, 38.9, 30.8, 24.5. **HRMS** m/z (ES+) calcd for $C_{19}H_{19}N_2O_2$ 307.1447, found 307.1438.

*1'-methyl-1-tosyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2u***)*



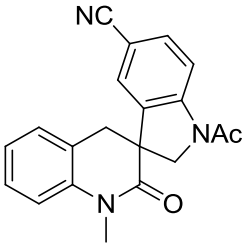
Yield = 80%. Orange solid. **M. p.** 165°C. **R_f** (petroleum ether/EtOAc 7:3) 0.54. **IR** (neat, cm^{-1}) ν 2966, 2252, 1668, 1475, 1361, 1168, 907, 727. **^{1}H NMR** (400 MHz, CDCl3) δ 7.72 (d, *J* = 8.2, 2H), 7.63 (d, *J* = 8.2 Hz, 1H), 7.34 (td, *J* = 1.0, 7.5 Hz, 1H), 7.28 (d, *J* = 8.2 Hz, 2H), 7.20 (td, *J* = 1.3, 8.2, Hz, 1H), 7.06-7.02 (m, 2H), 6.94 (d, *J* = 7.9 Hz, 1H), 6.79 (td, *J* = 1.0, 7.4 Hz, 1H), 6.53 (dd, *J* =1.1, 7.5 Hz, 1H), 4.41 (d, *J* = 10.5 Hz, 1H), 3.85 (d, *J* = 10.5 Hz, 1H), 3.41 (s, 3H), 2.90 (d, *J* = 16.1 Hz, 1H), 2.82 (d, *J* = 16.1 Hz, 1H), 2.40 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.1, 144.6, 141.4, 140.2, 133.9, 132.9, 130.0, 129.4, 129.2, 128.5, 127.7, 124.4, 123.8, 123.7, 123.4, 114.8, 114.8, 58.4, 50.0, 38.6, 30.8, 21.8. **HRMS** m/z (ES+) calcd for $[C_{24}H_{23}N_2O_3S]^+$ 419.1424, found 419.1429.

*1-Acetyl-6-methoxy-1'-methyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2v***)*



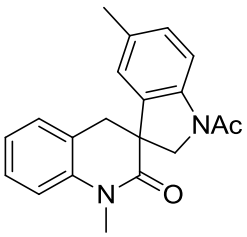
Yield = 95%. White solid. **M. p.** 171 °C. **R_f** (DCM/EtOAc 9:1) 0.21. **IR** (neat, cm^{-1}) ν 2970, 1662, 1603, 1489, 1402, 1127, 1034, 759, 732. **^{1}H NMR** (400 MHz, CDCl3) δ 7.90 (d, *J* = 1.3 Hz, 1H), 7.37 (t, *J* = 7.4 Hz, 1H), 7.14-7.06 (m, 3H), 6.34-6.29 (m, 2H), 4.84 (d, *J* = 10.4 Hz, 1H), 3.81 (d, *J* = 10.4 Hz, 1H), 3.45 (s, 3H), 3.24 (d, *J* = 15.0 Hz, 1H), 3.74 (s, 3H), 3.11 (d, *J* = 15.0 Hz, 1H), 2.26 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.8, 168.8, 160.8, 143.8, 140.4, 129.2, 128.5, 124.6, 123.7, 123.7, 133.5, 114.8, 110.2, 103.0, 59.2, 55.7, 49.0, 38.9, 30.8, 24.6. **HRMS** m/z (ES+) calcd for $[C_{20}H_{21}N_2O_3]^+$ 337.1547, found 337.1549.

*1-Acetyl-1'-methyl-2'-oxo-2',4'-dihydro-1'H-spiro[indoline-3,3'-quinoline]-5-carbonitrile (***2w***)*



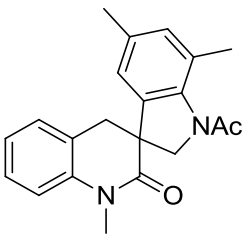
Yield = 60%. White solid. **M. p.** 216 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 5:5) 0.18. **IR** (neat, cm^{-1}) ν 2970, 2224, 1667, 1601, 1482, 1390, 1329, 908, 727. **^{1}H NMR** (400 MHz, CDCl3) δ 8.31 (d, *J* = 8.3 Hz, 1H), 7.50 (dd, *J* = 1.5, 8.3 Hz, 1H), 7.41 (td, *J* = 2.3, 7.7 Hz, 1H), 7.20-7.15 (m, 3H), 6.75 (s, 1H), 4.81 (d, *J* = 10.1 Hz, 1H), 3.86 (d, *J* = 10.1 Hz, 1H), 3.47 (s, 3H), 3.25 (d, *J* = 14.8 Hz, 1H), 3.15 (d, *J* = 14.8 Hz, 1H), 2.27 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 169.5, 169.3, 146.1, 139.9, 134.4, 133.6, 129.2, 129.1, 127.4, 124.3, 122.3, 119.1, 117.6, 115.2, 106.6, 58.6, 49.6, 38.6, 31.0, 24.5. **HRMS** m/z (ES+) calcd for $[C_{20}H_{18}N_3O_2]^+$ 332.1394, found 332.1393.

*1-Acetyl-1',5-dimethyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2w***)*



Yield = 94%. White solid. **M. p.** 205 °C. $\mathbf{R_f}$ (petroleum ether/EtOAc 5:5) 0.18. **IR** (neat, cm^{-1}) ν 2923, 1666, 1603, 1472, 1369, 1127, 733. **^{1}H NMR** (400 MHz, CDCl3) δ 8.09 (d, *J* = 8.7 Hz, 1H), 7.40 (t, *J* = 7.2 Hz, 1H), 7.15-7.00 (m, 4H), 6.34 (s, 1H), 4.69 (d, *J* = 11.0 Hz, 1H), 3.78 (d, *J* = 11.0 Hz, 1H), 3.47 (s, 3H), 3.20 (d, *J* = 14.5 Hz, 1H), 3.15 (d, *J* = 14.5 Hz, 1H), 2.22 (s, 3H), 2.10 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.8, 168.4, 140.3, 140.2, 133.4, 132.7, 130.0, 129.2, 128.5, 124.1, 123.7, 123.4, 117.1, 114.8, 58.6, 49.9, 38.9, 30.8, 24.4, 21.2. **HRMS** m/z (ES+) calcd for $[C_{20}H_{21}N_2O_2]^+$ 321.1598, found 321.1599.

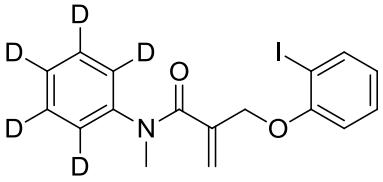
*1-Acetyl-1',5,7-trimethyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2y***)*



Yield = 57%. White solid. **R_f** (DCM/EtOAc 9:1) 0.33. **IR** (neat, cm^{-1}) ν 2970, 2224, 1667, 1482, 1329, 1127, 908, 727. **^{1}H NMR** (400 MHz, CDCl3) δ 7.36 (t, *J* = 6.6 Hz, 1H), 7.13 (d, *J* = 6.6 Hz, 1H), 7.09-7.06 (m, 3H), 6.86 (s, 1H), 4.62 (d, *J* = 10.4 Hz, 1H), 3.84 (d, *J* = 10.4 Hz, 1H), 3.42 (s, 3H), 3.14 (d, *J* = 15.0 Hz, 1H), 3.03 (d, *J* = 15.0 Hz, 1H), 2.29 (s, 3H), 2.27 (s, 3H), 2.09 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 170.7, 140.3, 135.3, 132.1, 130.9, 128.9, 128.4, 123.7, 123.6, 121.3, 114.9, 60.1, 50.6, 36.3, 30.5, 24.0, 21.2, 20.4. **HRMS** m/z (ES+) calcd for $[C_{21}H_{23}N_2O_2]^+$ 335.1754, found 335.1755.

Synthesis of the Deuterated Starting Materials

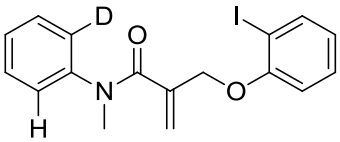
2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide-d_5 (**1a-D_5**)



N-methyl-*N*-phenylacrylamide-D_5 **1a-D_5** was prepared following general procedure A.

Yield = 90%. White solid. **M. p.** 91°C. **R_f** (petroleum ether/EtOAc 7:3) 0.39. **IR** (neat, cm^{-1}) ν 2922, 2361, 1622, 1563, 1472, 1241, 1017, 1006. **^{1}H NMR** (400 MHz, CDCl3) δ 7.76 (dd, *J* = 1.7, 7.7 Hz, 1H), 7.26 (td, *J* = 1.1, 7.7 Hz, 1H), 6.76 (dd, *J* = 1.3, 8.4 Hz, 1H), 6.70 (td, *J* = 1.3, 7.7 Hz, 1H), 5.52 (s, 1H), 5.15 (s, 1H), 4.67 (s, 2H), 3.43 (s, 3H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 157.1, 144.4, 139.7, 139.4, 129.7, 123.0, 122.0, 112.6, 86.4, 69.7, 38.3. **HRMS** m/z (ES+) calcd for $C_{17}H_{12}D_5INO_2$ 399.0618, found 399.0617.

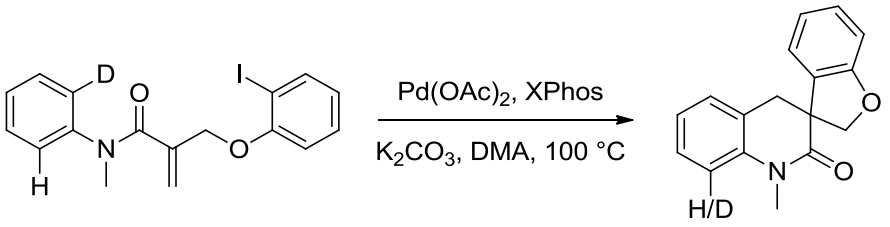
2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide-d_1 (**1a-D_1**)



N-methyl-*N*-phenylacrylamide-D_1 was prepared following general procedure A, starting from *N*-methyl-*N*-aniline-*D_1* (90% deuterium incorporation according to ^{1}H NMR)[4] and acrylic acid **4**. Yield = 92%. White solid. **M. p.** 90 °C. **R_f** (petroleum ether/EtOAc 7:3) 0.39. **IR** (neat, cm^{-1}) ν 2922, 2361, 1622, 1563, 1472, 1241, 1017, 1006. **^{1}H NMR** (400 MHz, CDCl3) δ 7.76 (dd, *J* = 1.7, 7.7 Hz, 1H), 7.26 (m, 5H), 6.76 (dd, *J* = 1.3, 8.4 Hz, 1H), 6.70 (td, *J* = 1.3, 7.7 Hz, 1H), 5.52 (s, 1H), 5.15 (s, 1H), 4.67 (s, 2H), 3.43 (s, 1H). **^{13}C NMR** (75 MHz, CDCl3) δ 168.9, 157.1, 144.4, 139.7, 139.4, 129.7, 129.4 (d, *J* = 10.0 Hz), 127.1 (d, *J* = 6.9 Hz), 123.0, 122.0, 112.6, 86.4, 69.7, 38.3. **HRMS** m/z (ES+) calcd for $C_{17}H_{16}DINO_2$ 395.0367, found 395.0363.

[4] Pinto, A.; Neuville, L.; Retailleau, P.; Zhu, J. *Org. Lett.* **2006**, 8, 4927.

Determination of Intramolecular Isotope Effect



1a-D$_1$ **2a/2a-D$_1$**

Anilide **1a-D$_1$** (30 mg, 0.076 mmol, 1.0 equiv), $Pd(OAc)_2$ (0.43 mg, 0.0019 mmol, 0.025 equiv), Xphos (2.4 mg, 0.0038 mmol, 0.050 equiv) and K_2CO_3 (21 mg, 0.15 mmol, 2.0 equiv) were introduced in a screw cap vial equipped with a magnetic stir bar. The vial was purged with argon during 10 min. Degazed DMA (0.5 mL) was then added and the mixture was stirred at 100 °C during 6 h until complete disappearance of the starting material as shown by TLC. The reaction was next quenched with water and extracted with EtOAc. The combined organic layers were washed with brine, dried and concentrated under reduced pressure. The residue was purified by flash column chromatography (petroleum ether/EtOAc 8/2) to give the expected spirodihydroquinolinone **2a/2a-D$_1$**(18 mg, 90%) as a white solid.

Inspection of the aromatic region of the ^{1}H NMR spectrum (7.16-7.06 for proton H2, H4, H5, H13 and 6.82 for proton H14) indicated that the position 2 was 30.5% protonated (after correction), indicating an intramolecular isotope effect of k_H/k_D = 2.53 (by taking account of the 90% incorporation of the starting material **1a-D$_1$** (Figure 1 and 2).

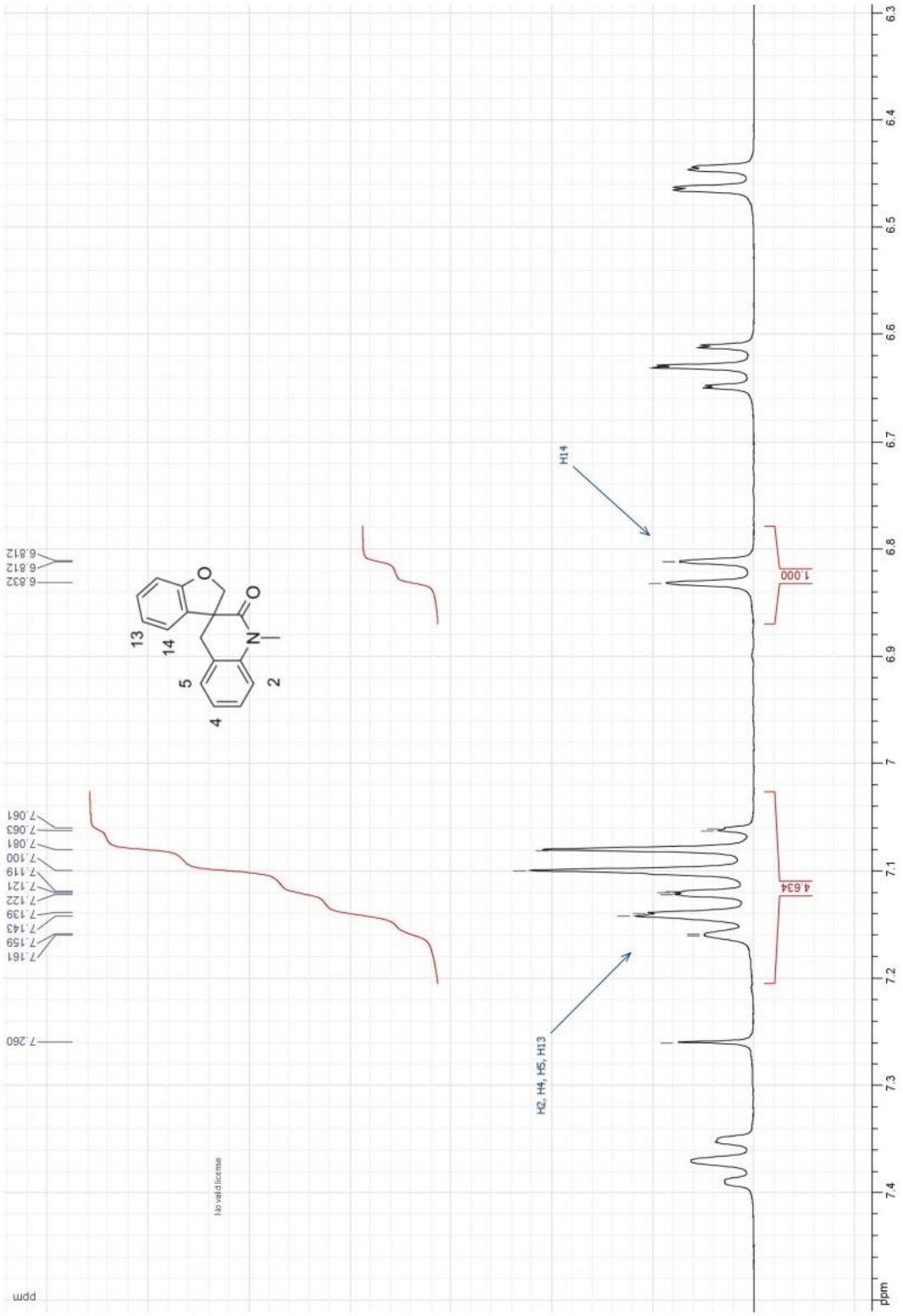


Figure 1. Aromatic region of spirodihydroquinolinone **2a**

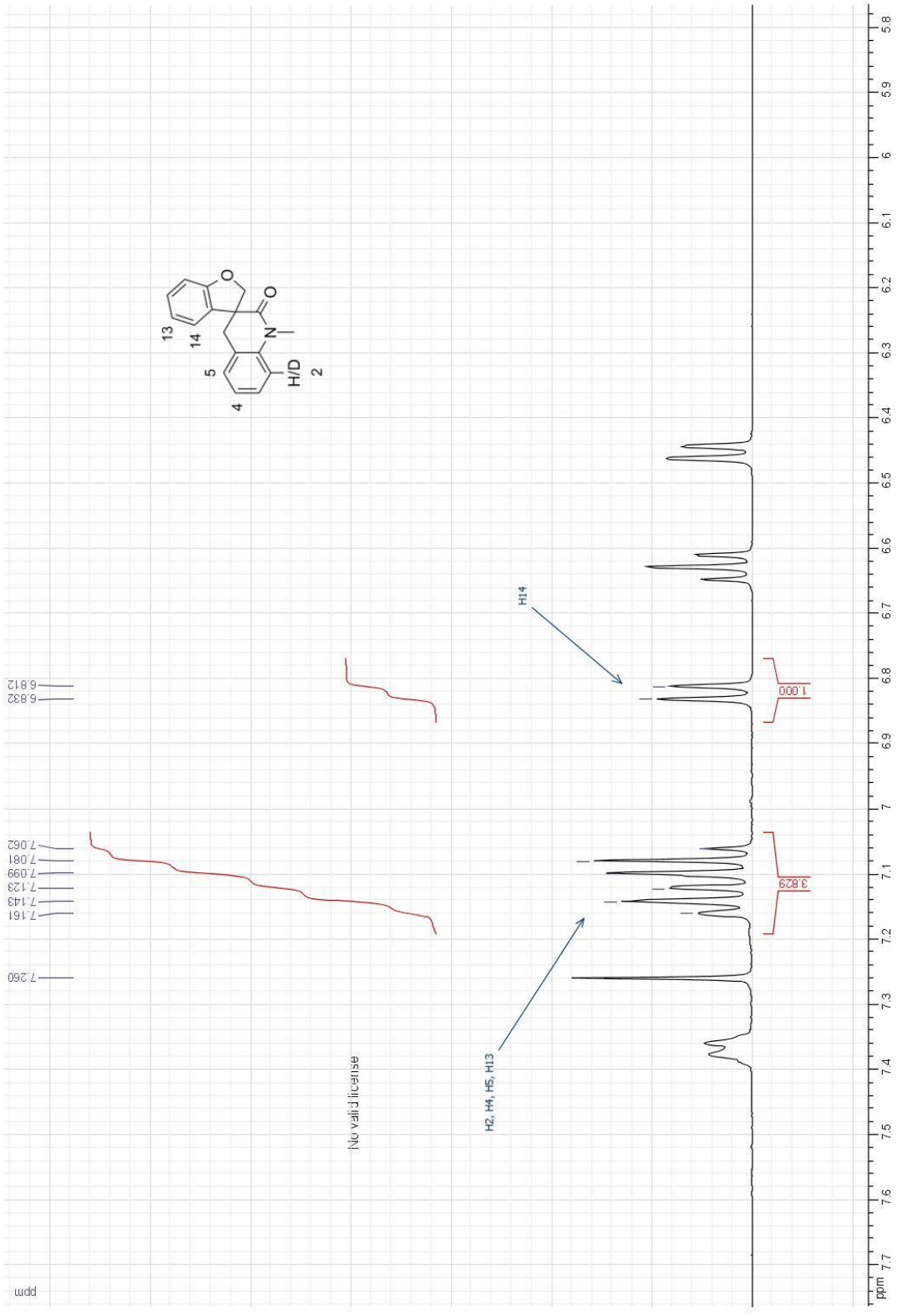
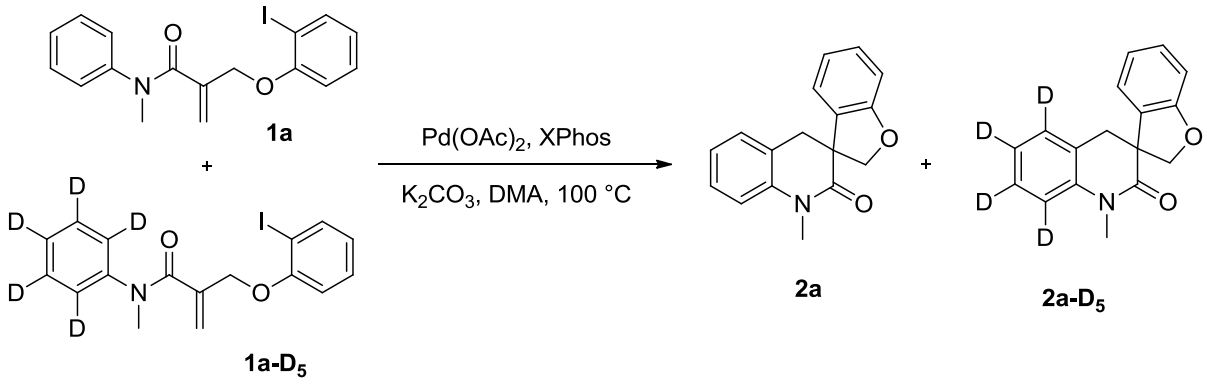


Figure 2. Aromatic region of isolated material from cyclization of monodeuterioanilide **2a-D$_1$**

Determination of Intermolecular Isotope Effect



Anilide **1a** (30 mg, 0.051 mmol, 0.5 equiv), anilide **1a-D_5** (30 mg, 0.076 mmol, 0.5 equiv), $Pd(OAc)_2$ (0.85 mg, 0.0038 mmol, 0.025 equiv), Xphos (3.6 mg, 0.0076 mmol, 0.050 equiv) and K_2CO_3 (42 mg, 0.30 mmol, 2.0 equiv) were introduced in a screw cap vial equipped with a magnetic stir bar. The vial was purged with argon during 10 min. Degazed DMA (0.7 mL) was then added and the mixture was stirred at 100 °C during 3 h. After cooling the reaction mixture, water was added. the mixture was extracted with EtOAc. The combined organic layers were washed with brine, dried and concentrated under reduced pressure.

A conversion of 30% was obtained by comparison of proton H6 of **1a**+**1a-D_5** (5.52 ppm) and H7 of **2a**+**2a-D_5** (4.33 ppm) (Figure 3). The residue was purified by flash column chromatography (petroleum ether/EtOAc 8/2) to give a mixture of **2a** and **2a-D_5**. The ratio between **2a** and **2a-D_5** was determinate by integration of the proton H3 (7.37 ppm) and H12 (6.63 ppm) (Figure 4 and 5), indicating an intramolecular isotope effect of $k_H/k_D = 1.4$.

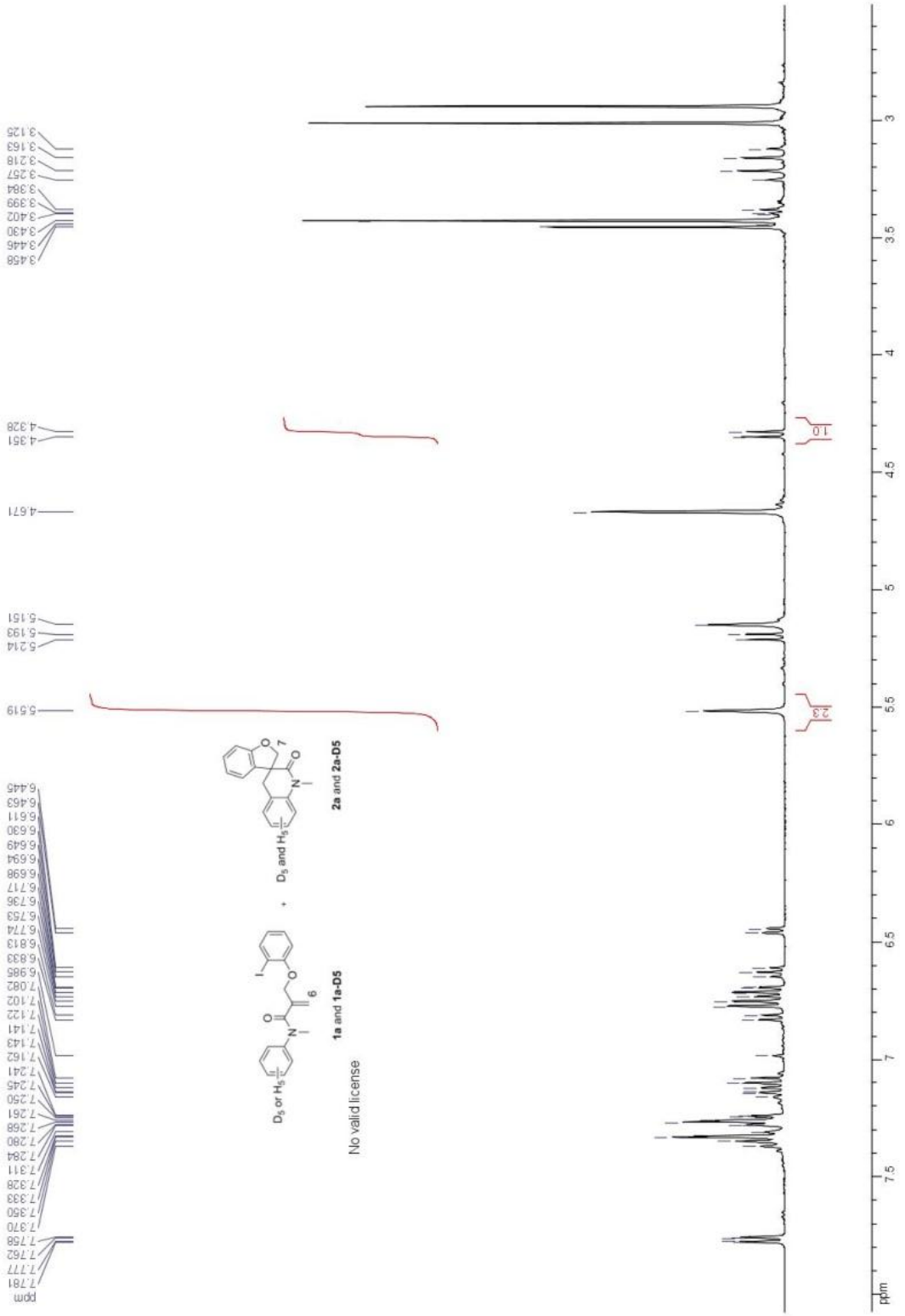


Figure 3. Reaction mixture of the intermolecular competition

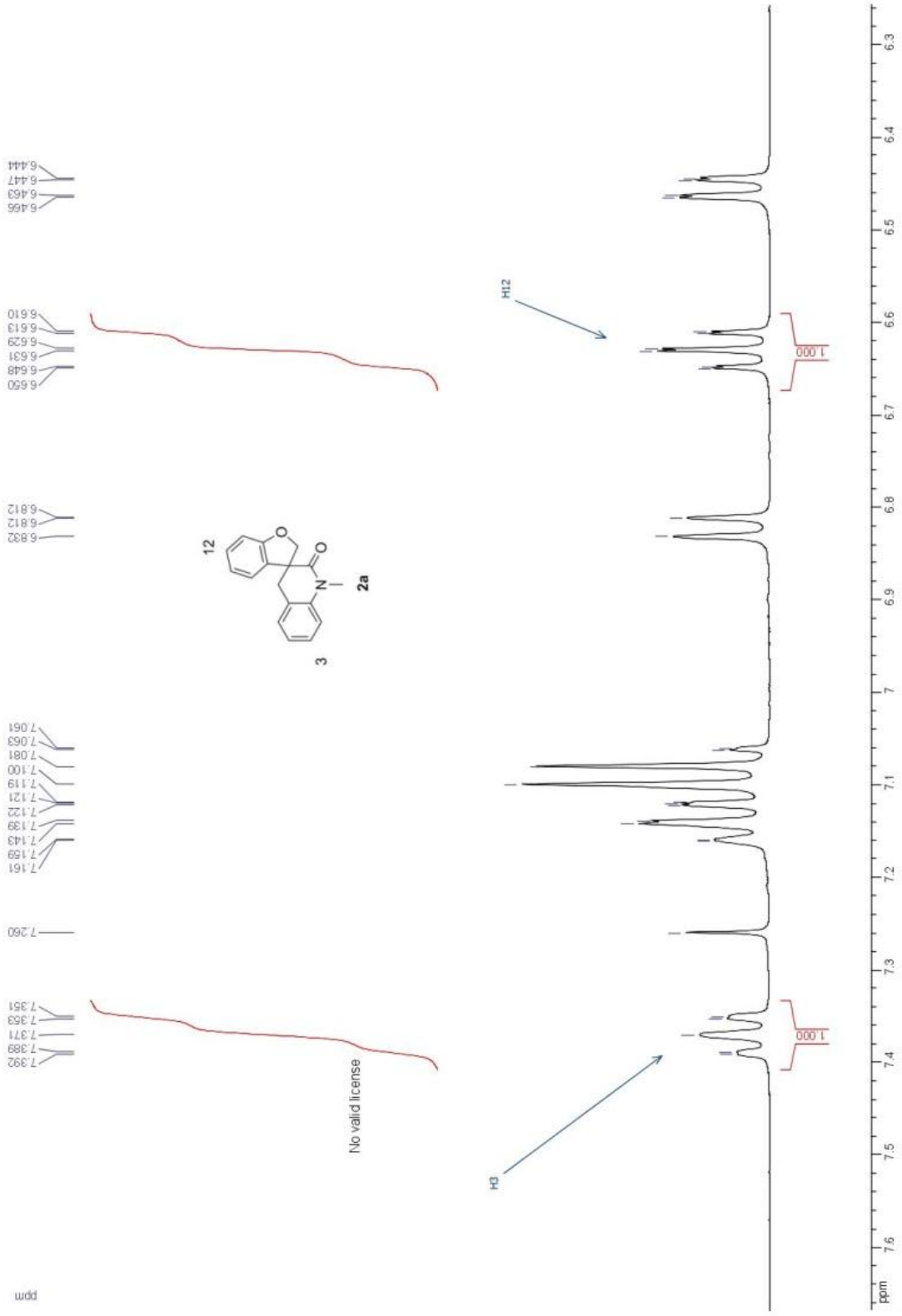


Figure 4. Aromatic region of spirodihydroquinolinone **2a**

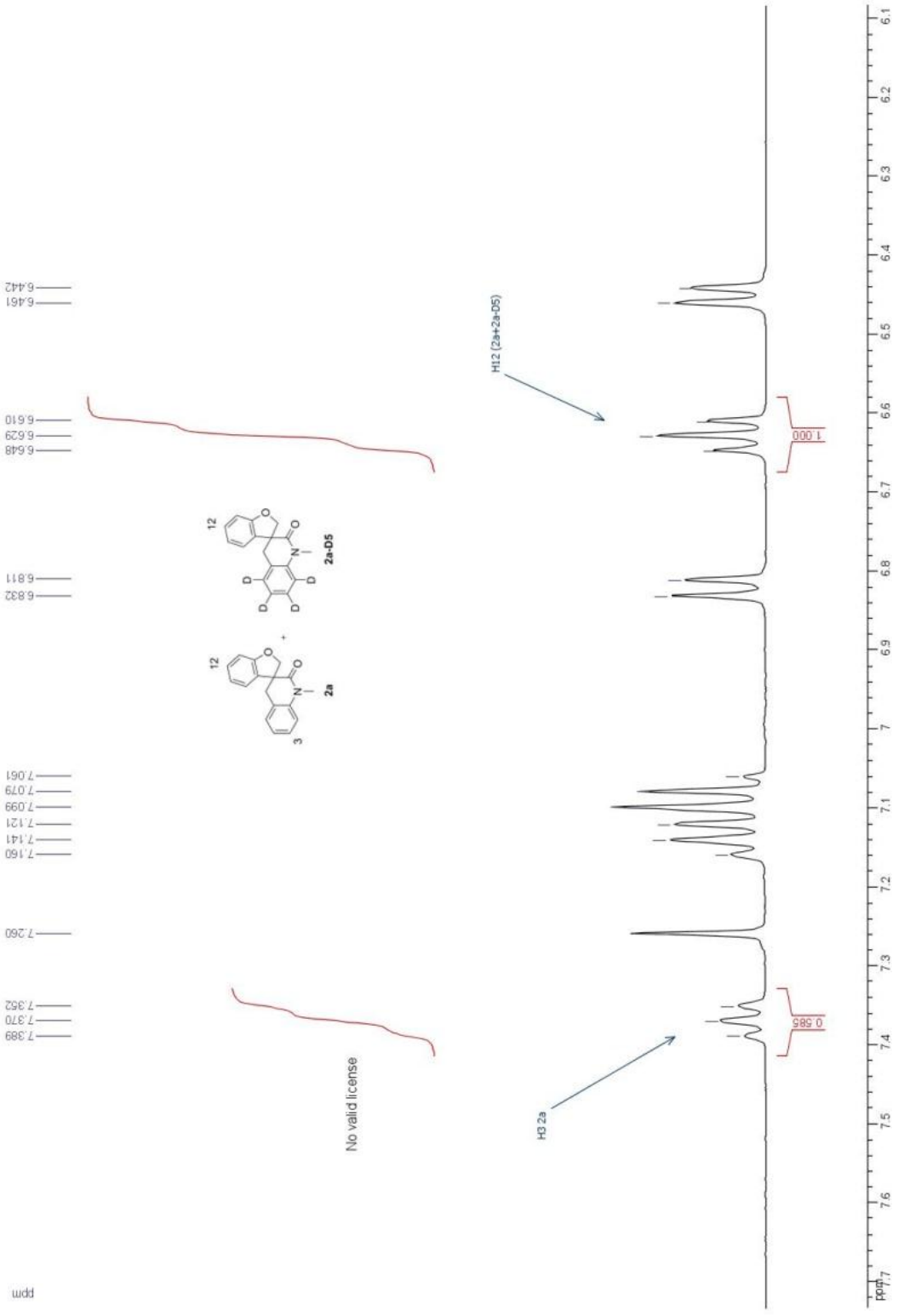
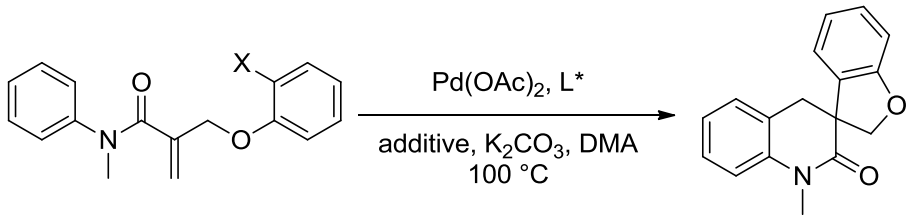


Figure 5. Aromatic region of isolated material from cyclization of compounds **2a** and **2a-D$_5$**

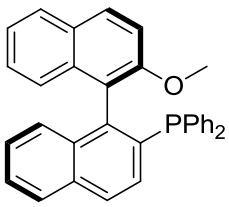
Chiral Ligands Screening



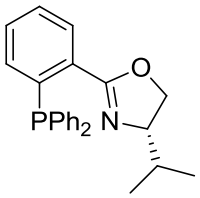
X = **1a**, I
= **1o**, OTf

2a

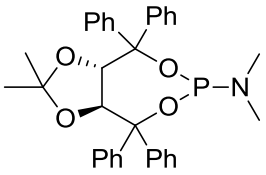
entry	starting material	additive	ligand	ee (%)
1	**1a**	Ag_3PO_4	L1	0
2	**1a**	Ag_3PO_4	L2	0
3	**1a**	Ag_3PO_4	L3	0
4	**1a**	Ag_3PO_4	L4	0
5	**1a**	Ag_3PO_4	L5	0
6	**1a**	Ag_3PO_4	L6	0
7	**1a**	Ag_3PO_4	L7	0
8	**2a**	-	L6	0
9	**2a**	-	L7	0
10	**2a**	-	L5	0
11	**2a**	-	L1	0
12	**2a**	-	L2	0



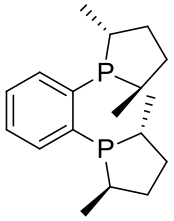
L1 (R)-MOP



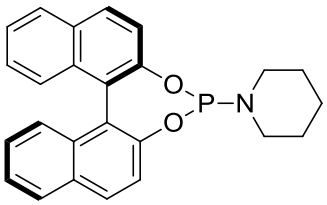
L2



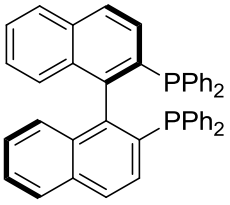
L3



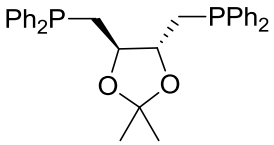
L4



L5

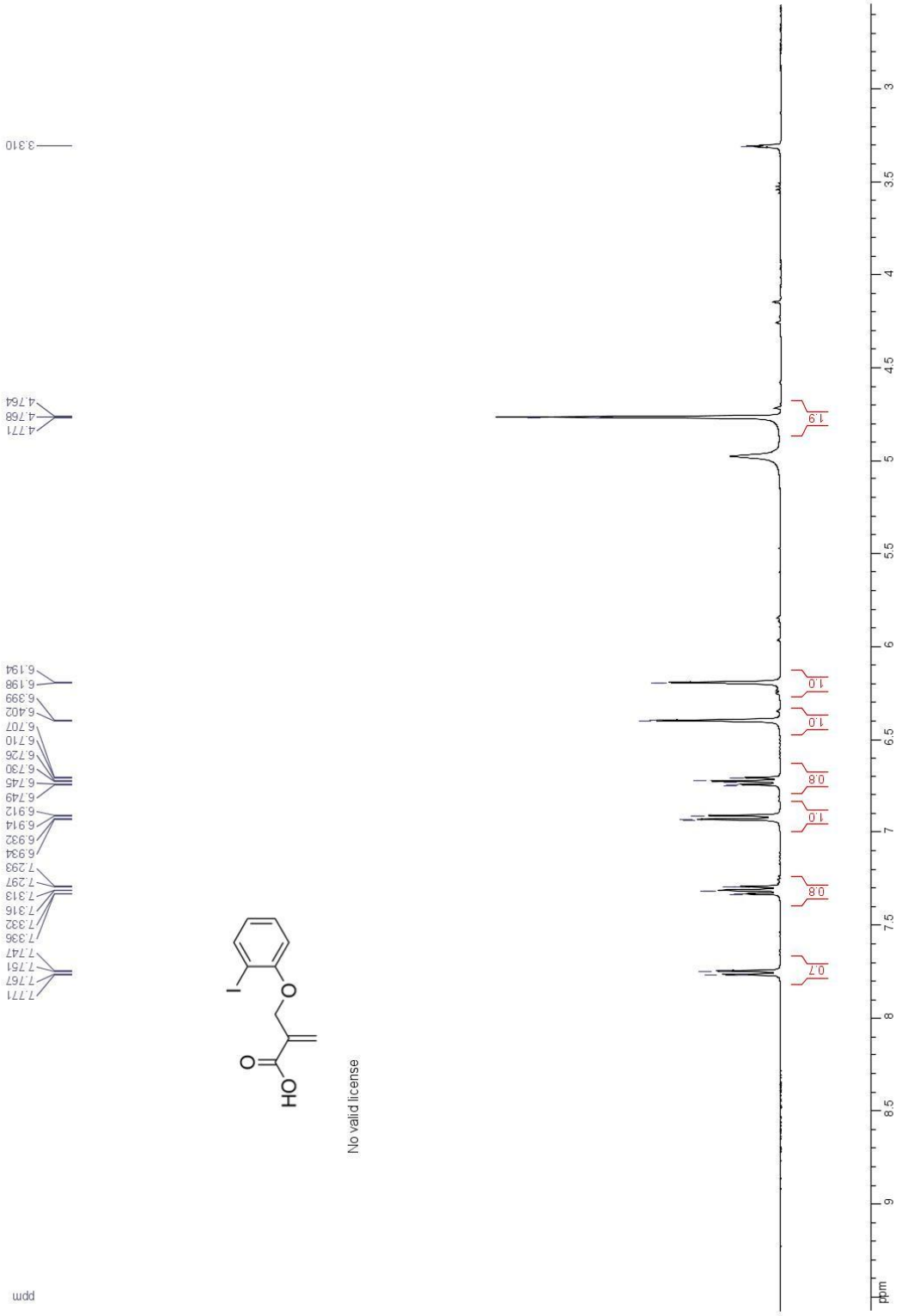


L6 (S)-BINAP

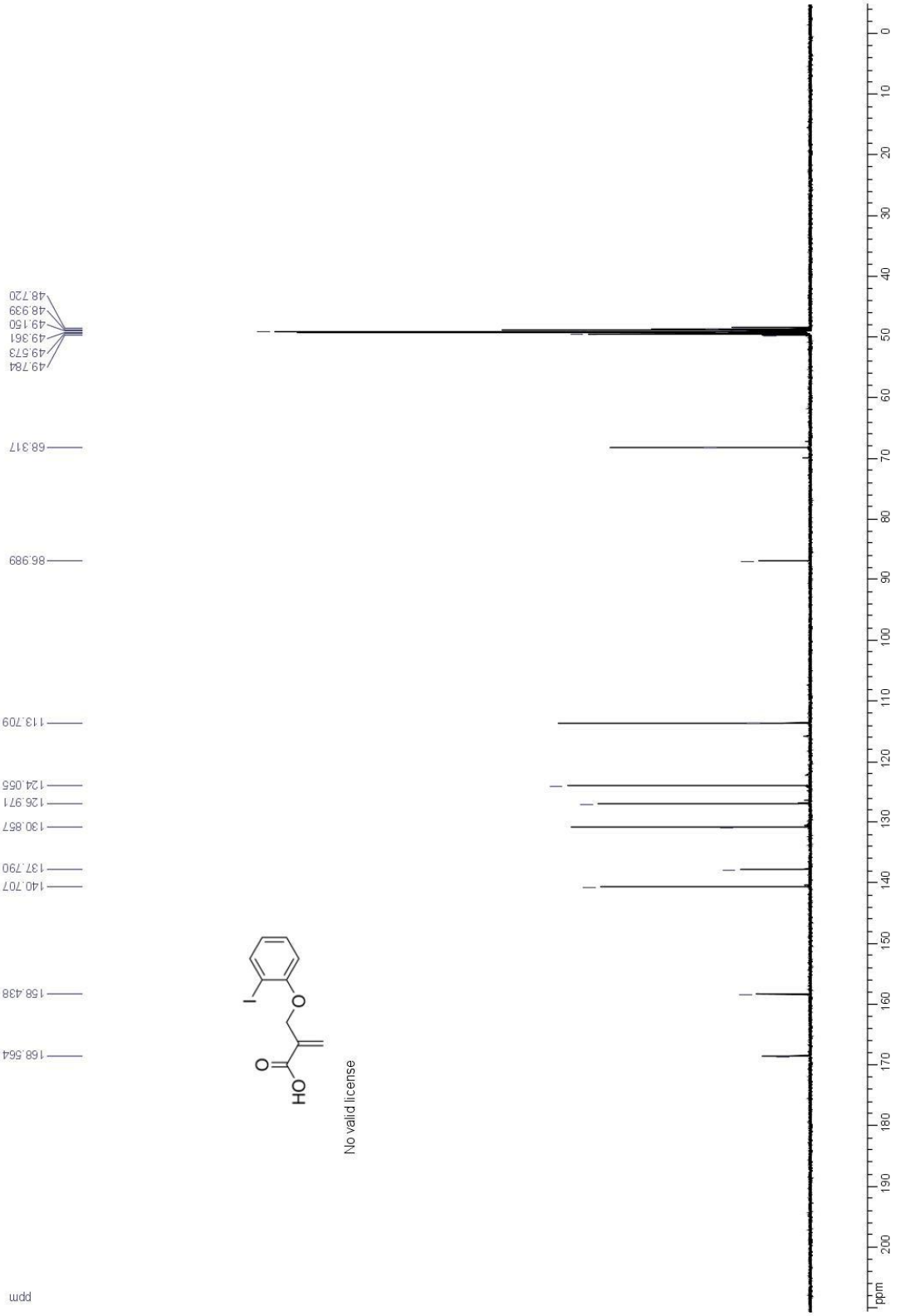


L7 (R,R)-DIOP

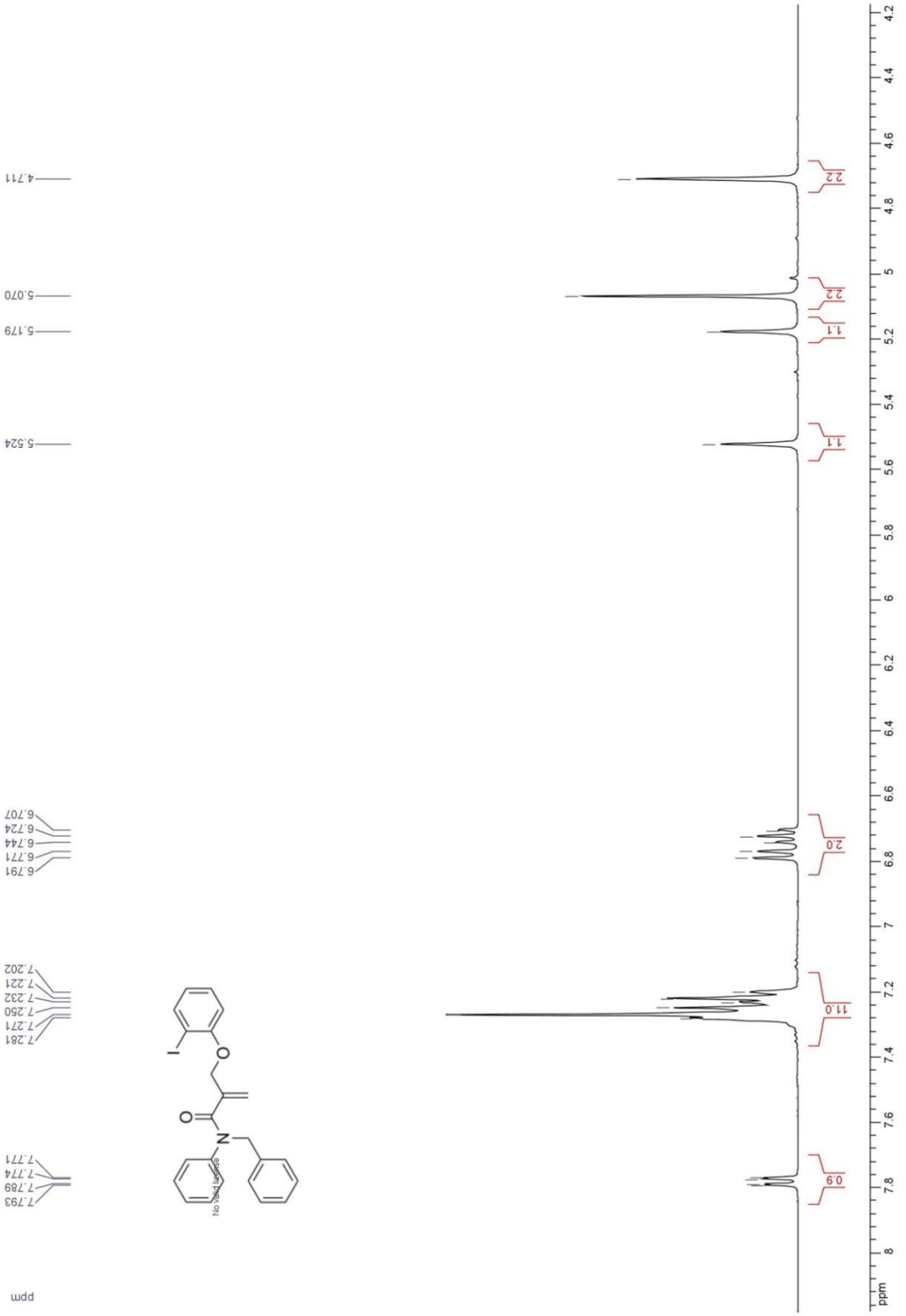
*2-((2-Iodophenoxy)methyl)acrylic acid (**4**)*



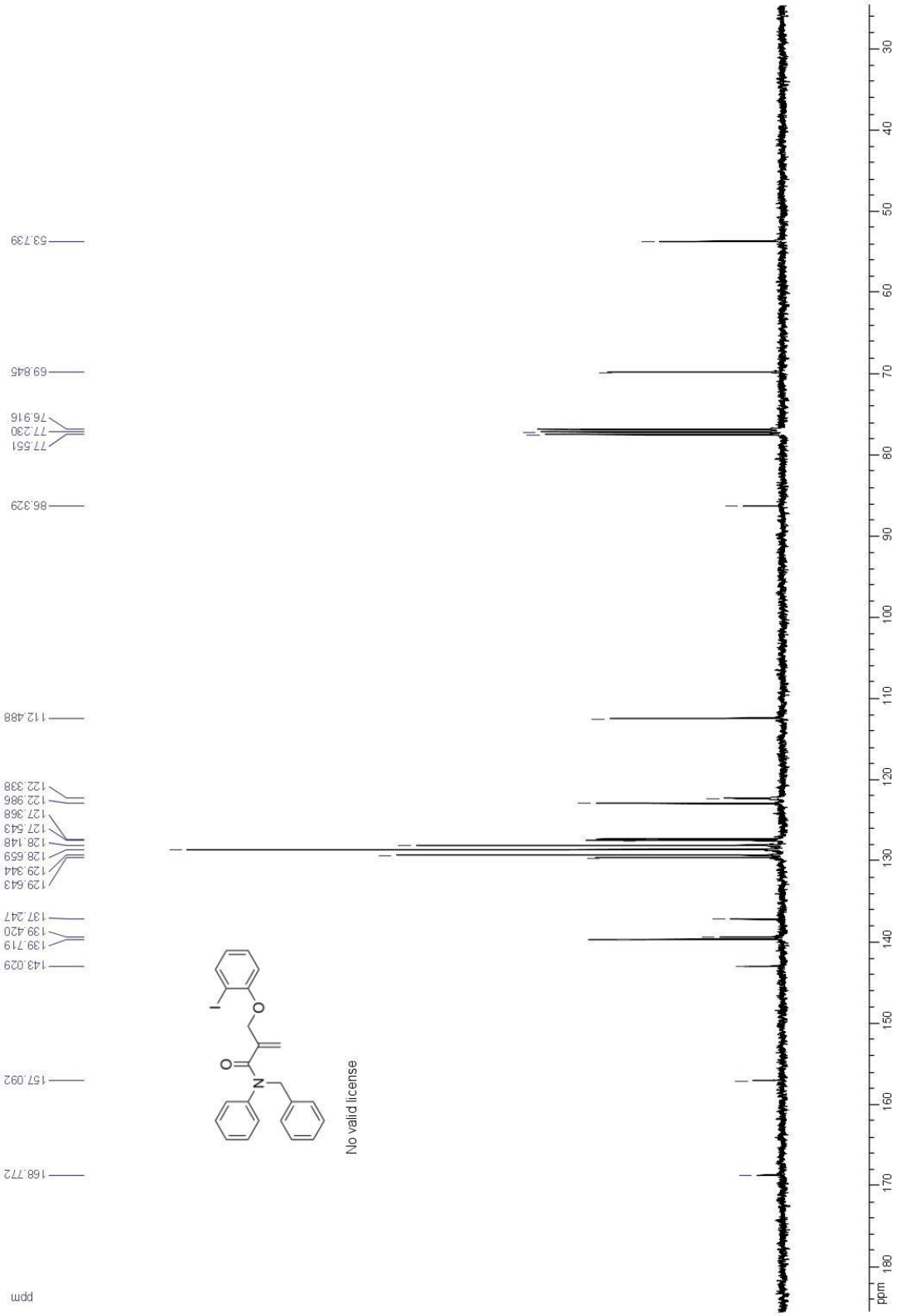
*2-((2-Iodophenoxy)methyl)acrylic acid (**4**)*



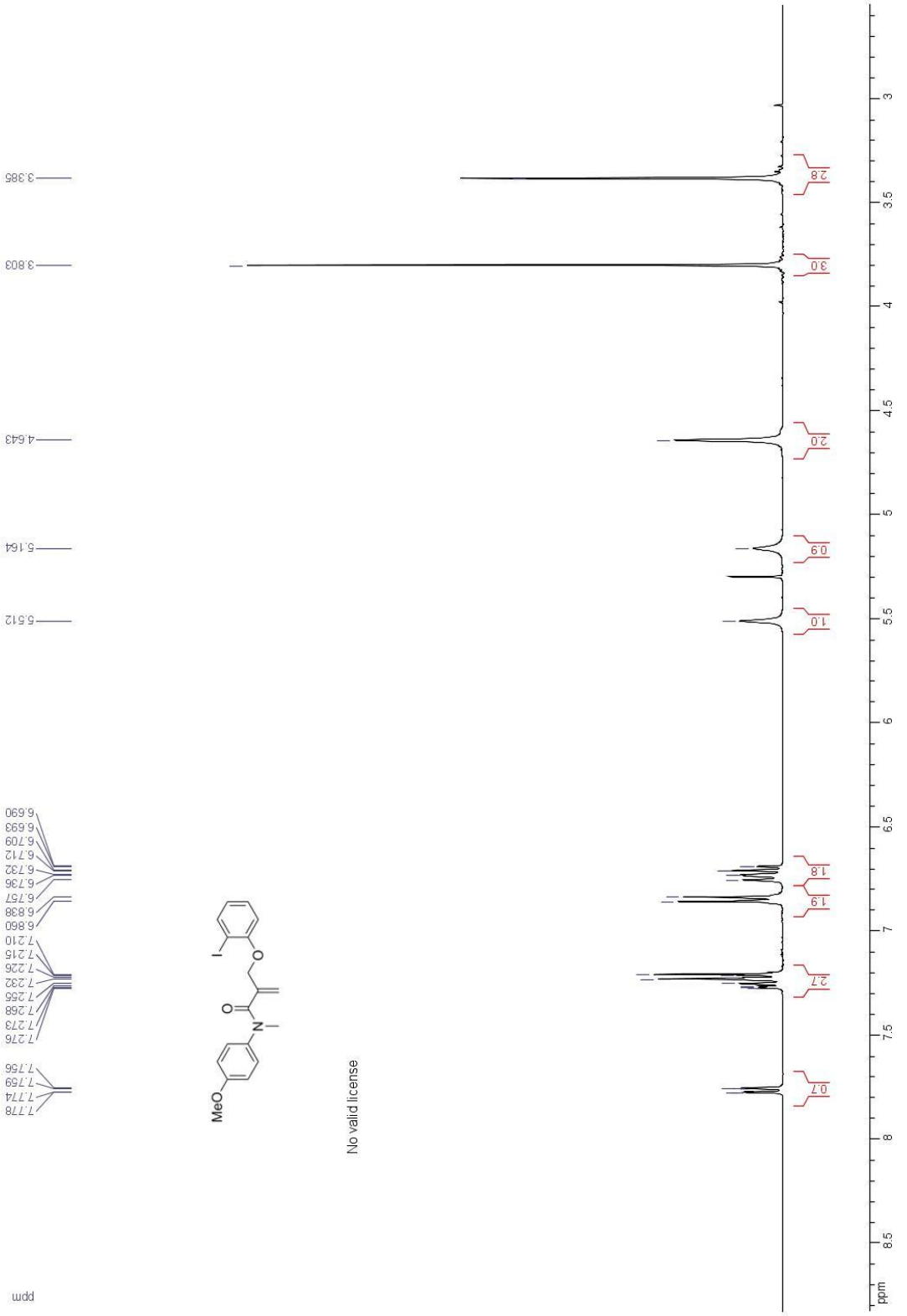
*N-benzyl-2-((2-iodophenoxy)methyl)-N-phenylacrylamide (***1c***)*



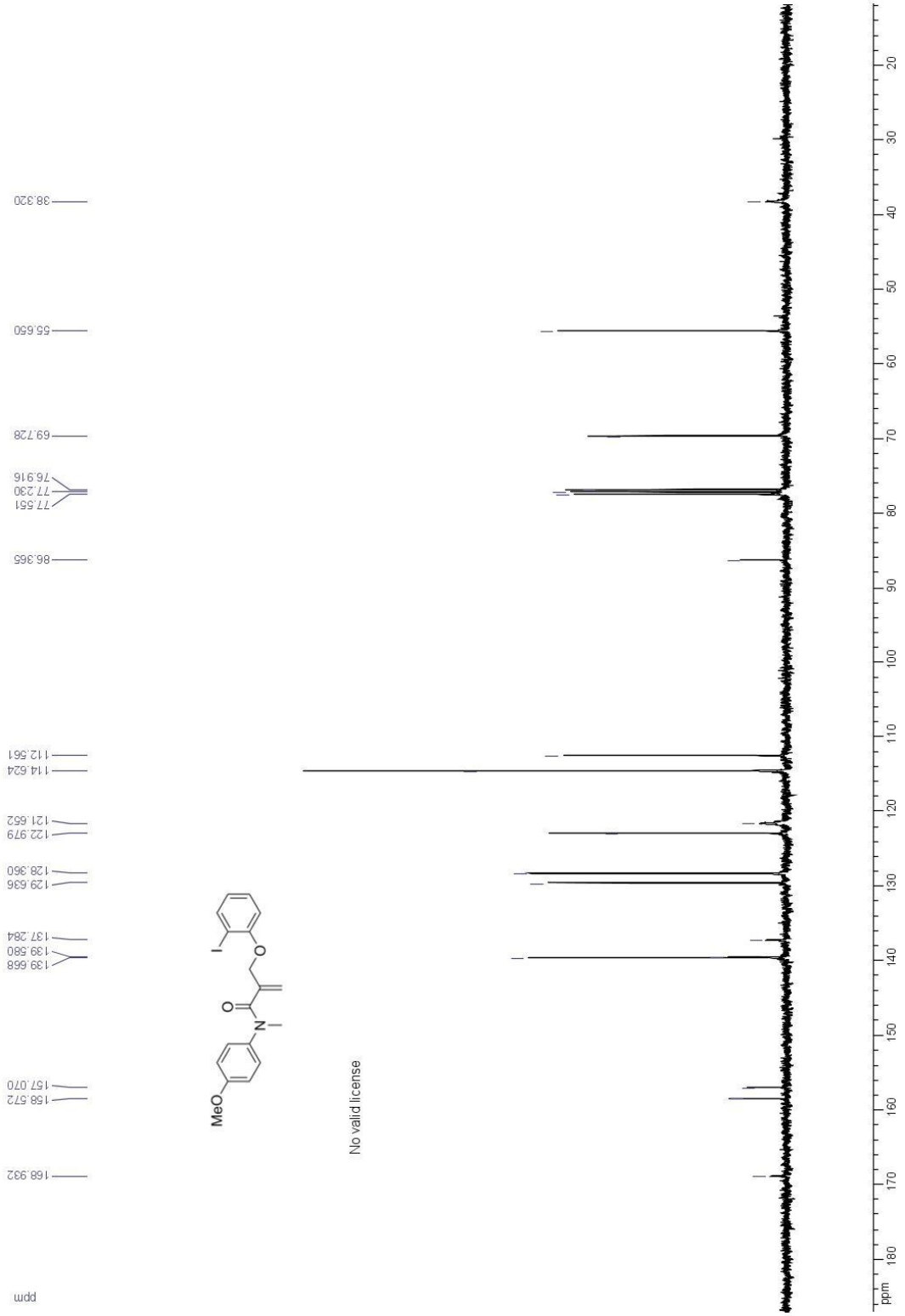
*N-benzyl-2-((2-iodophenoxy)methyl)-N-phenylacrylamide (***1c***)*



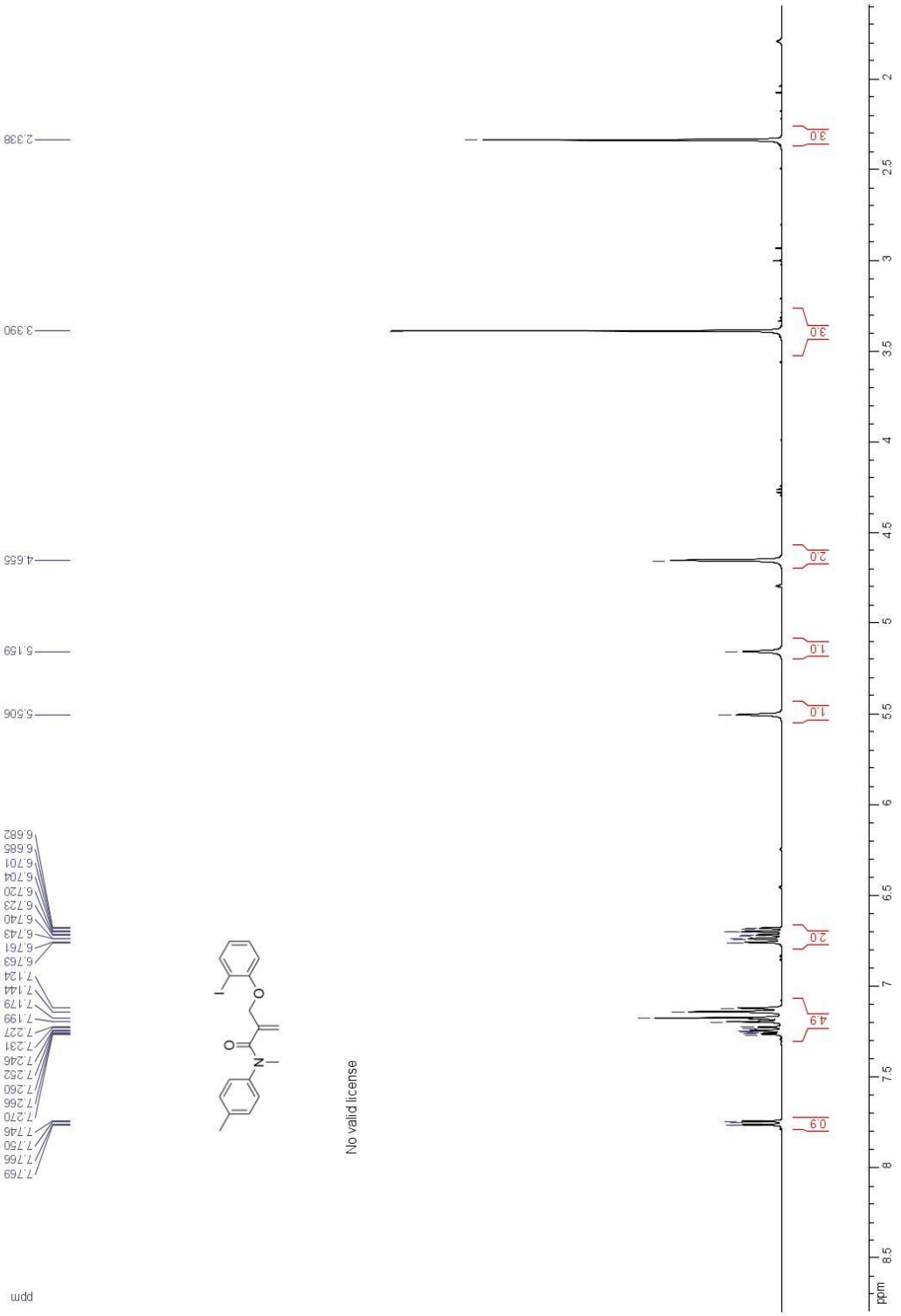
*2-((2-iodophenoxy)methyl)-N-(4-methoxyphenyl)-N-methylacrylamide (***1d***)*



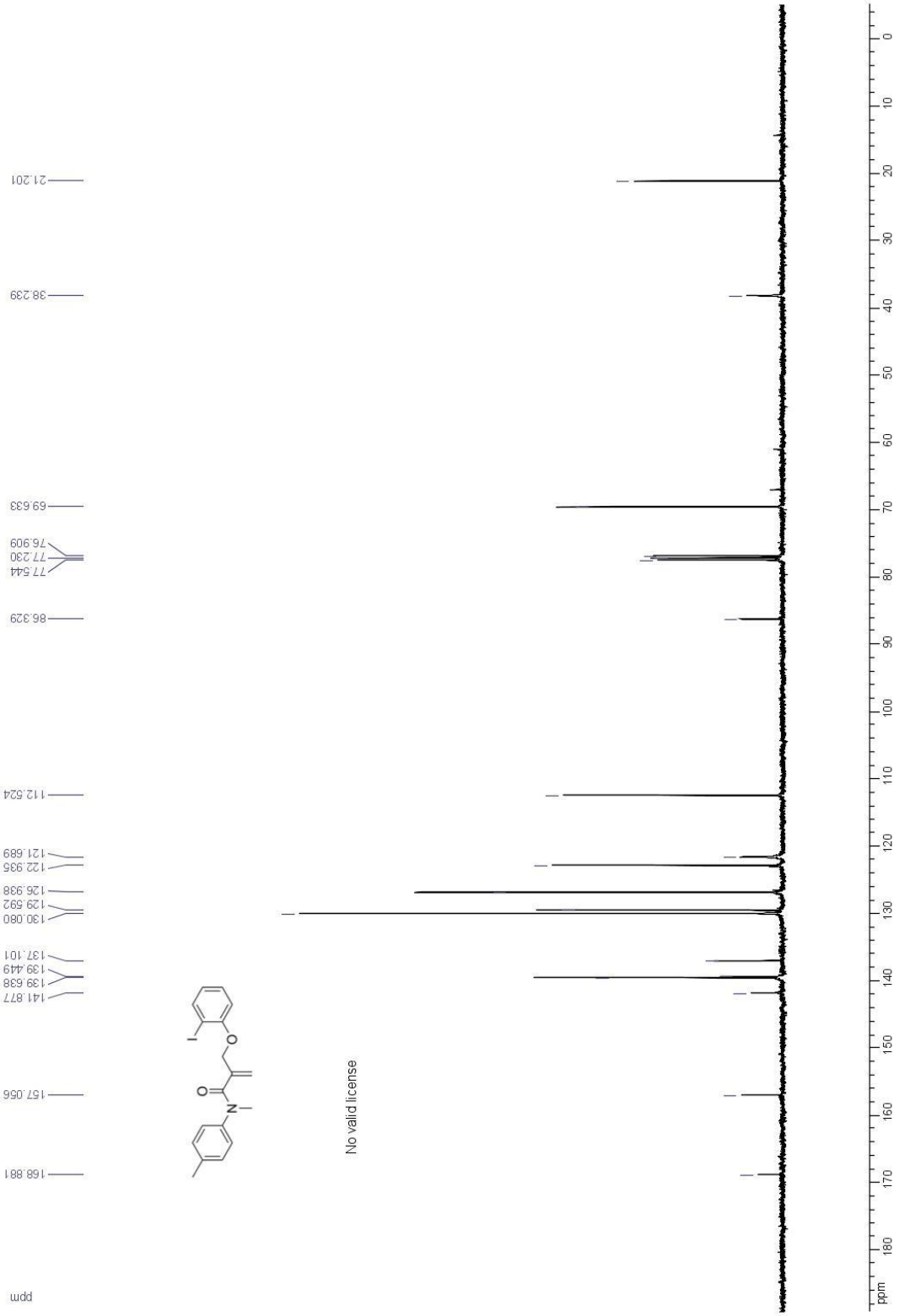
*2-((2-iodophenoxy)methyl)-N-(4-methoxyphenyl)-N-methylacrylamide (***1d***)*



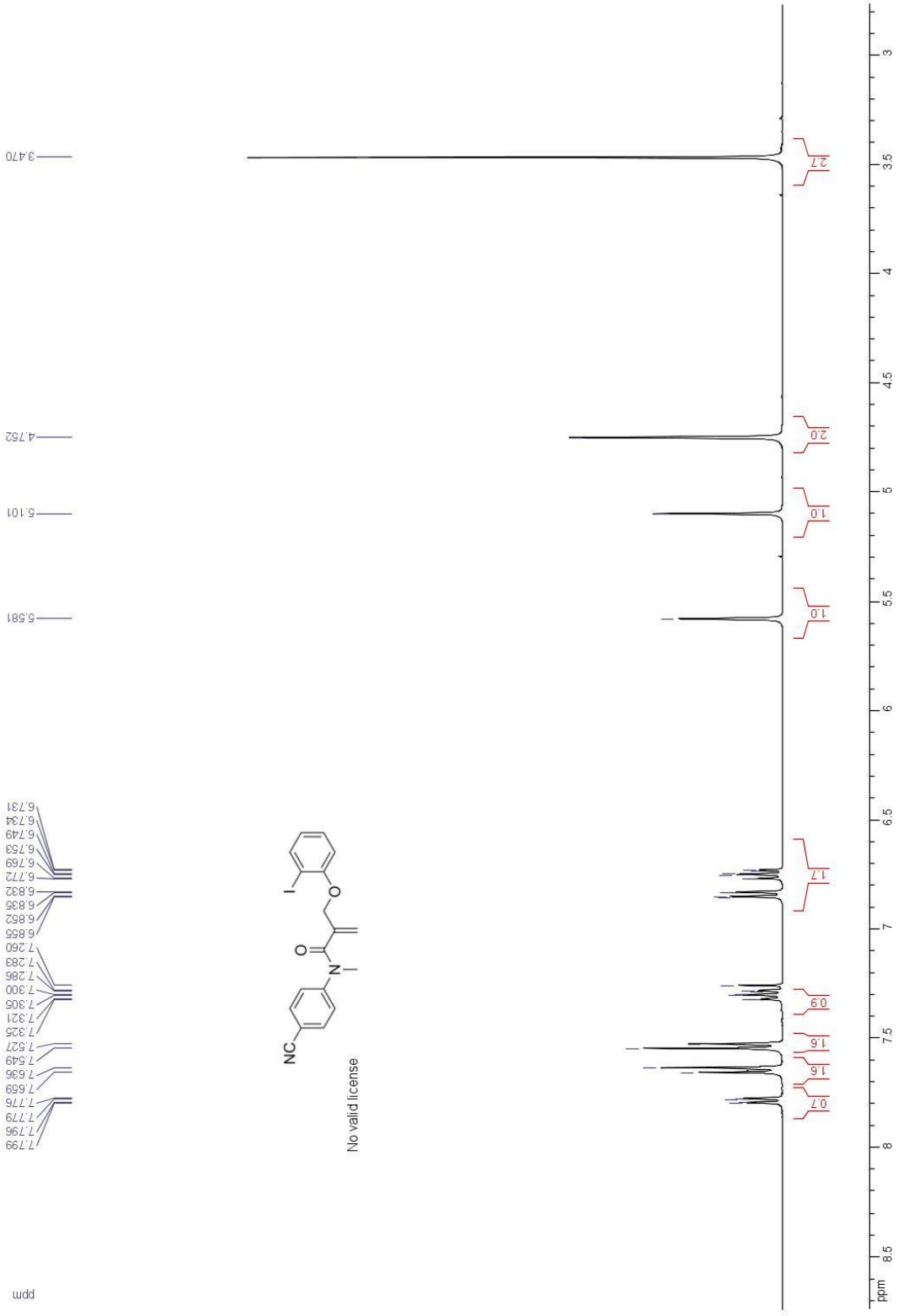
*2-((2-Iodophenoxy)methyl)-N-methyl-N-(p-tolyl)acrylamide (***1e***)*



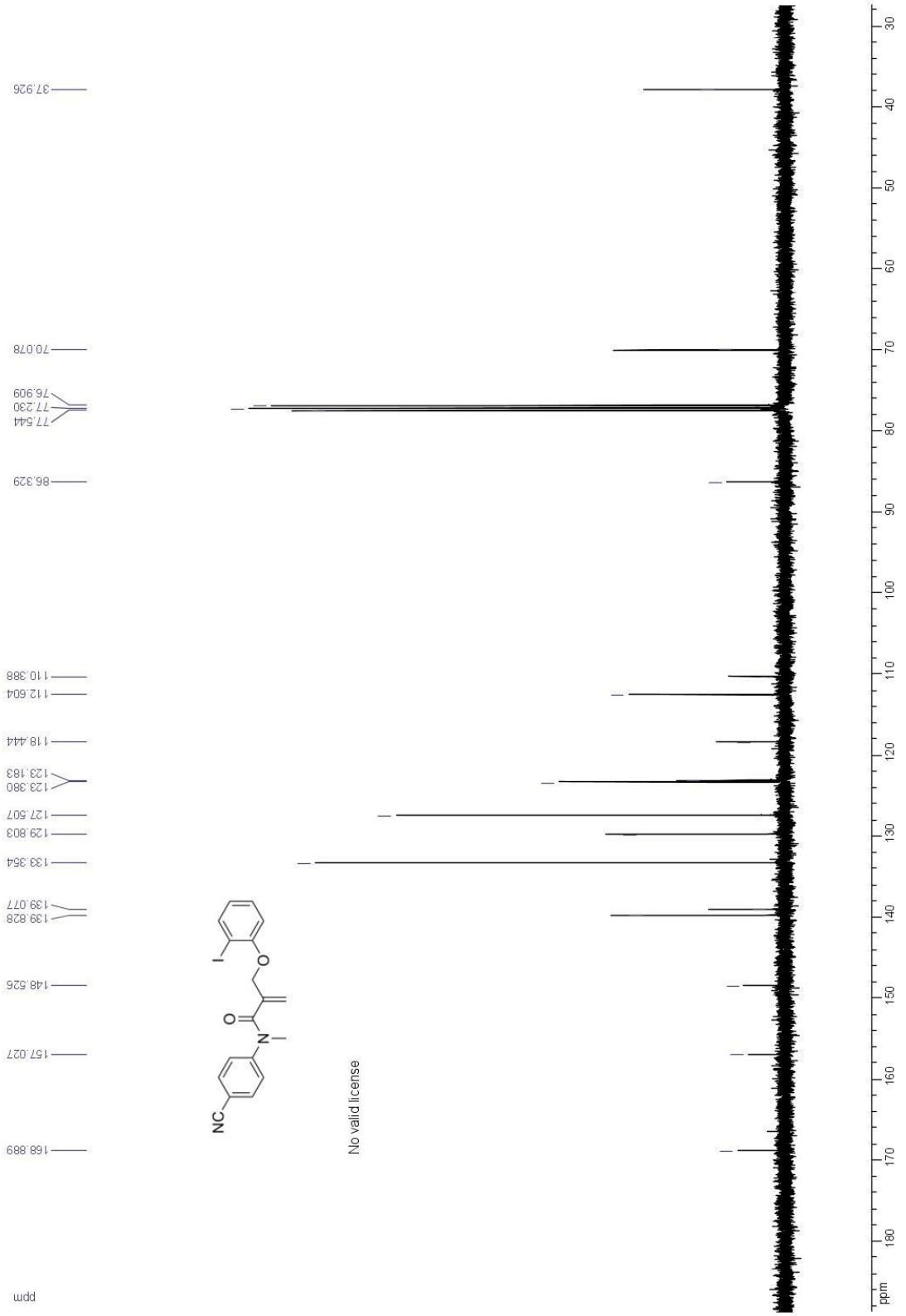
*2-((2-Iodophenoxy)methyl)-N-methyl-N-(p-tolyl)acrylamide (***1e***)*



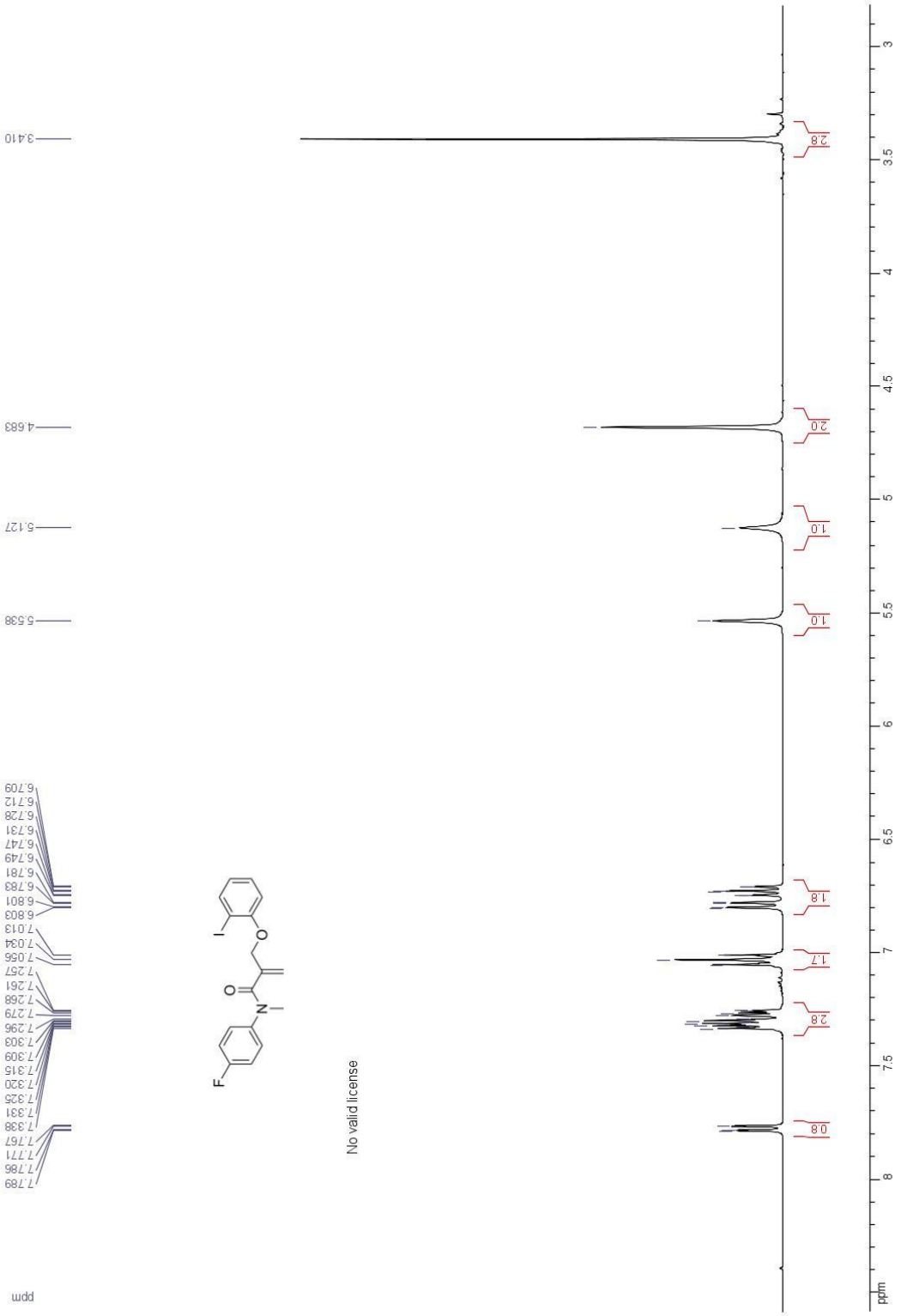
*N-(4-cyanophenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1f***)*



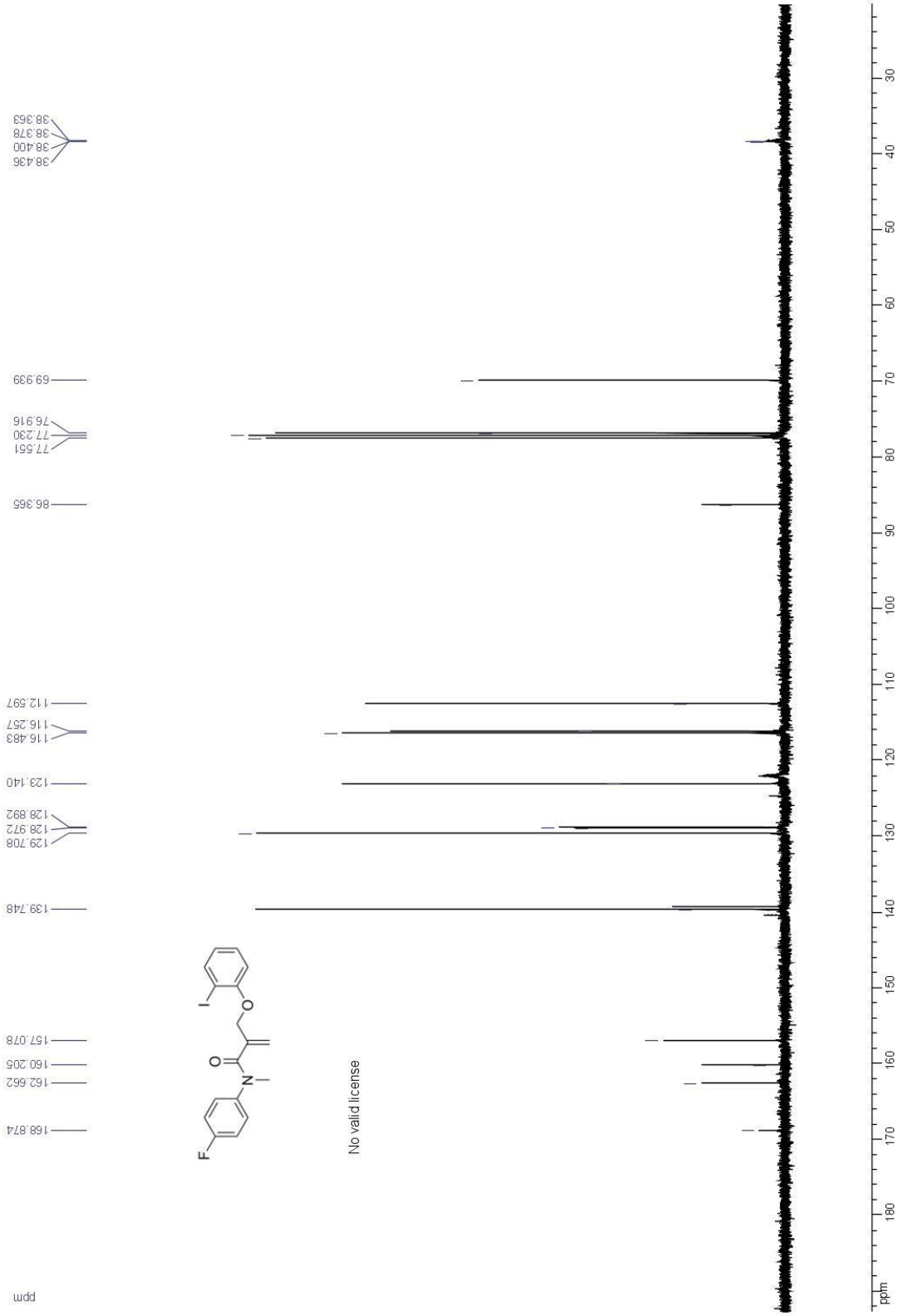
*N-(4-cyanophenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1f***)*



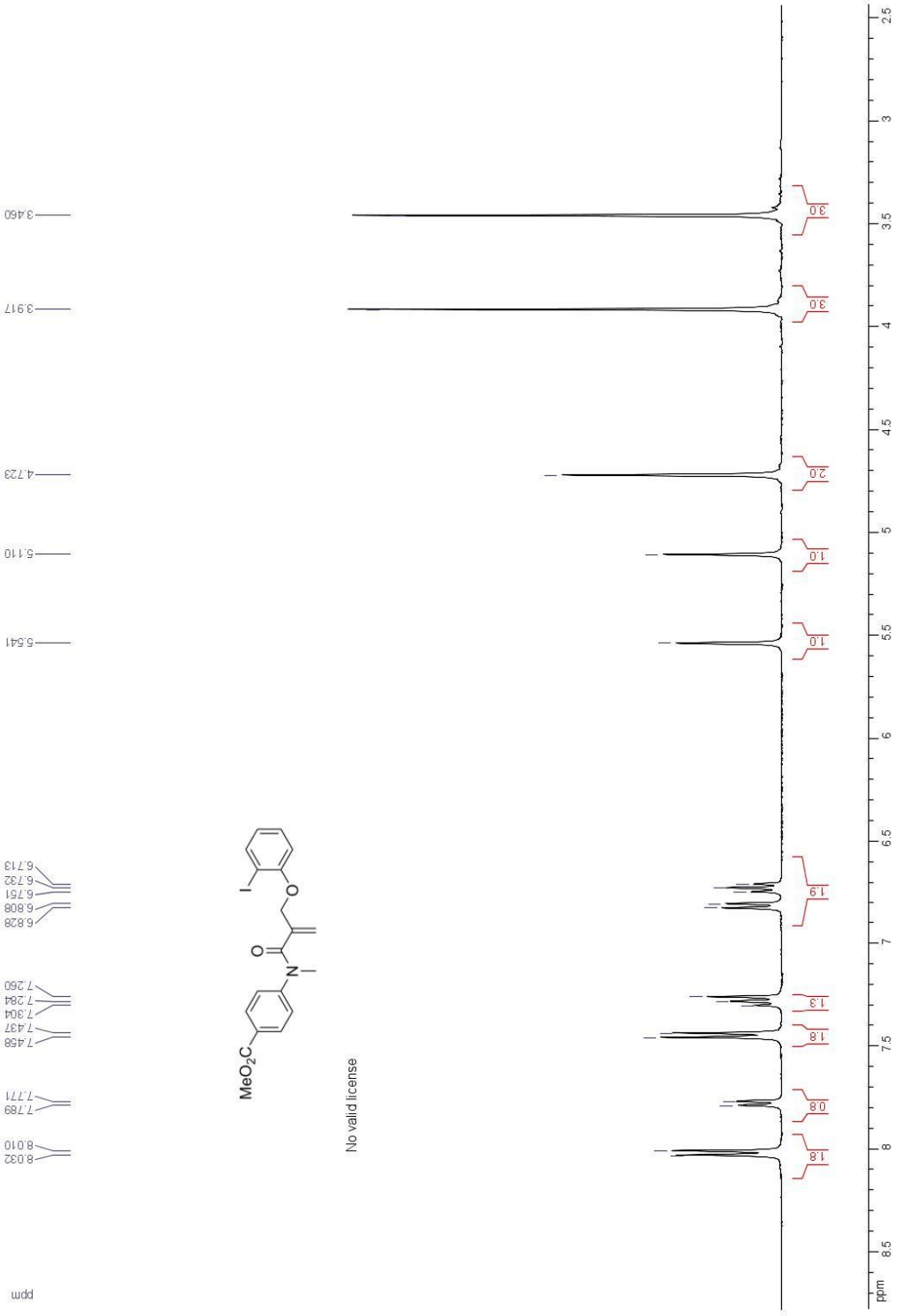
*N-(4-fluorophenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1g**)



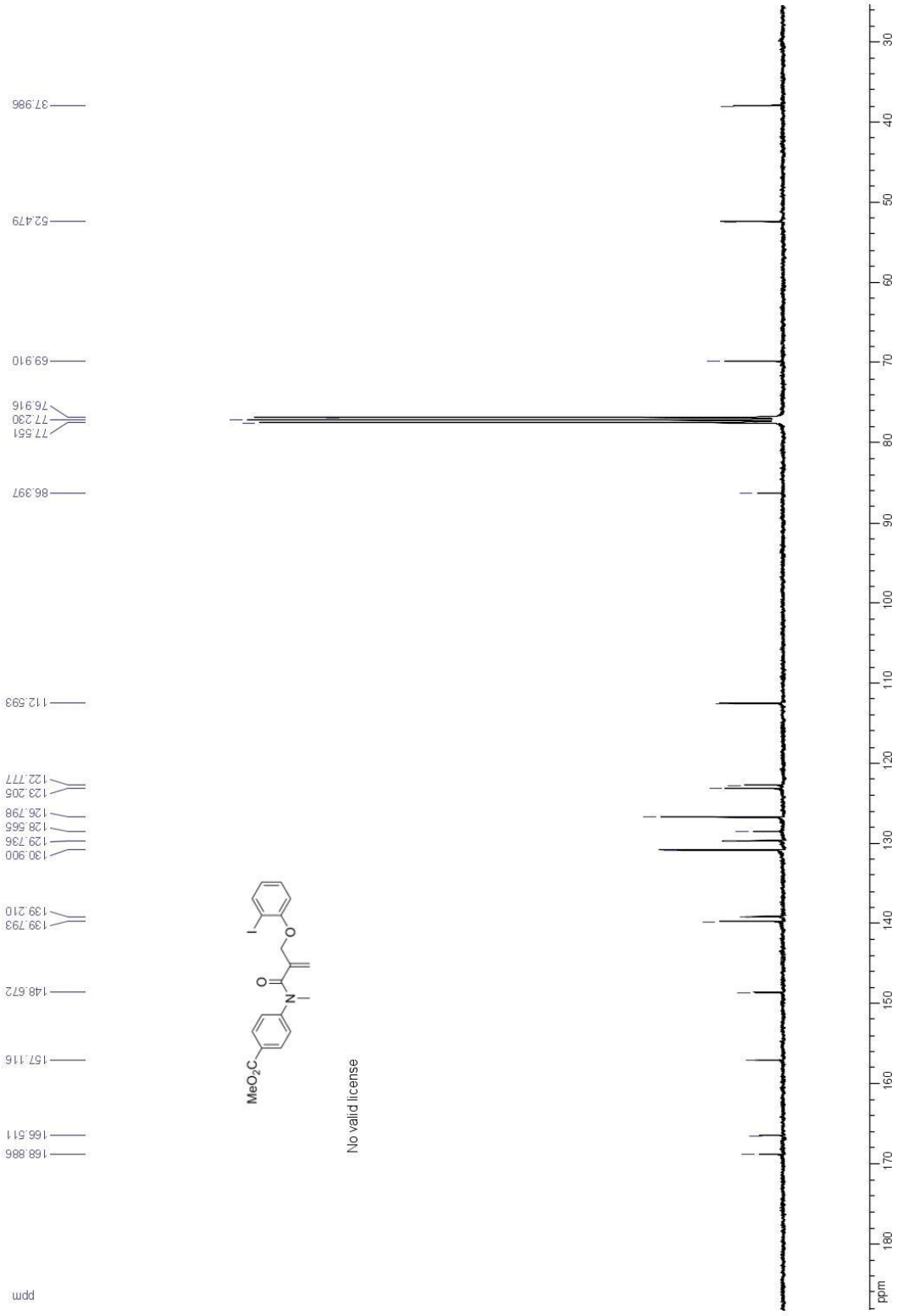
*N-(4-fluorophenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1g**)



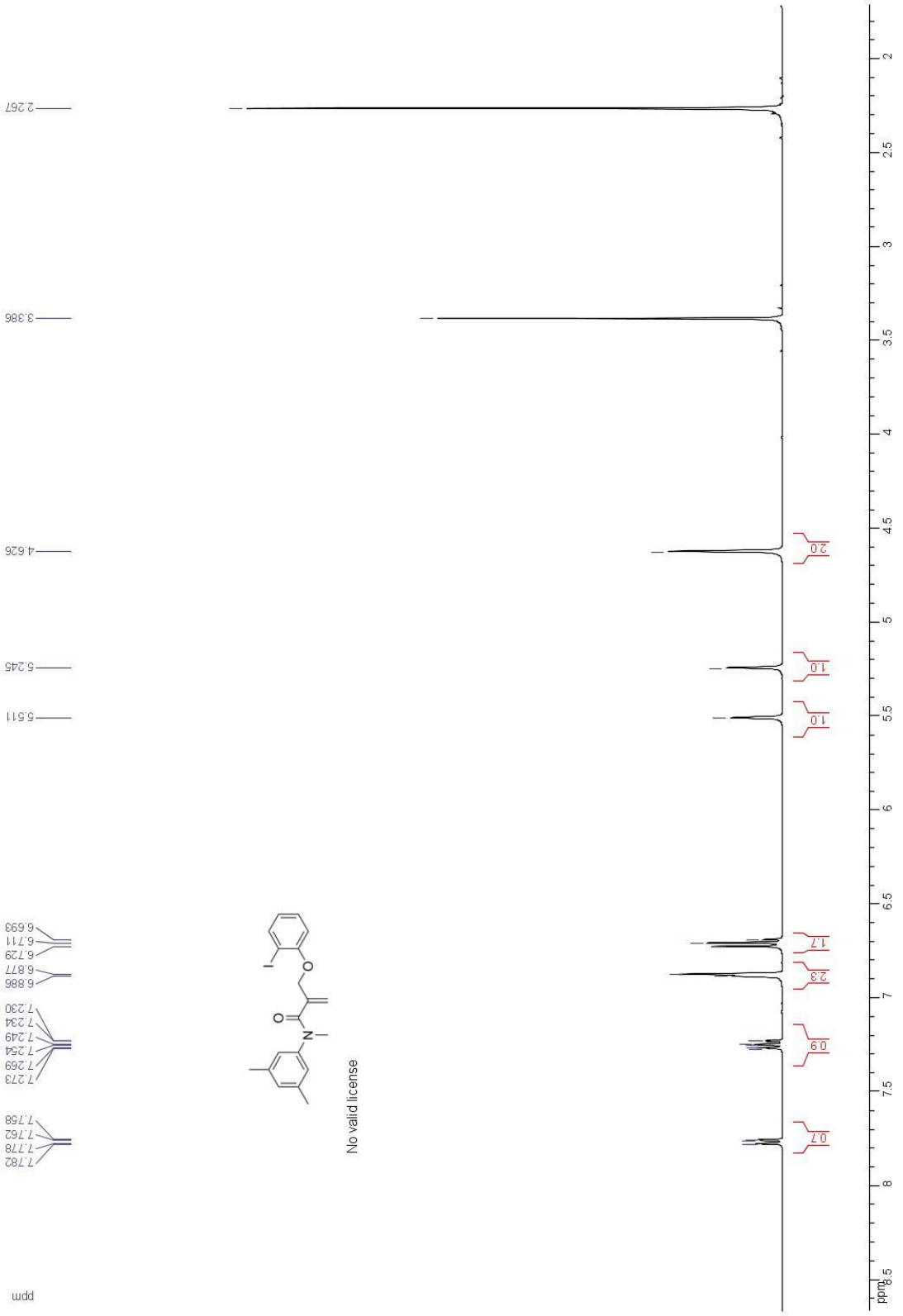
*Methyl 4-(2-((2-iodophenoxy)methyl)-N-methylacrylamido)benzoate (***1h***)*



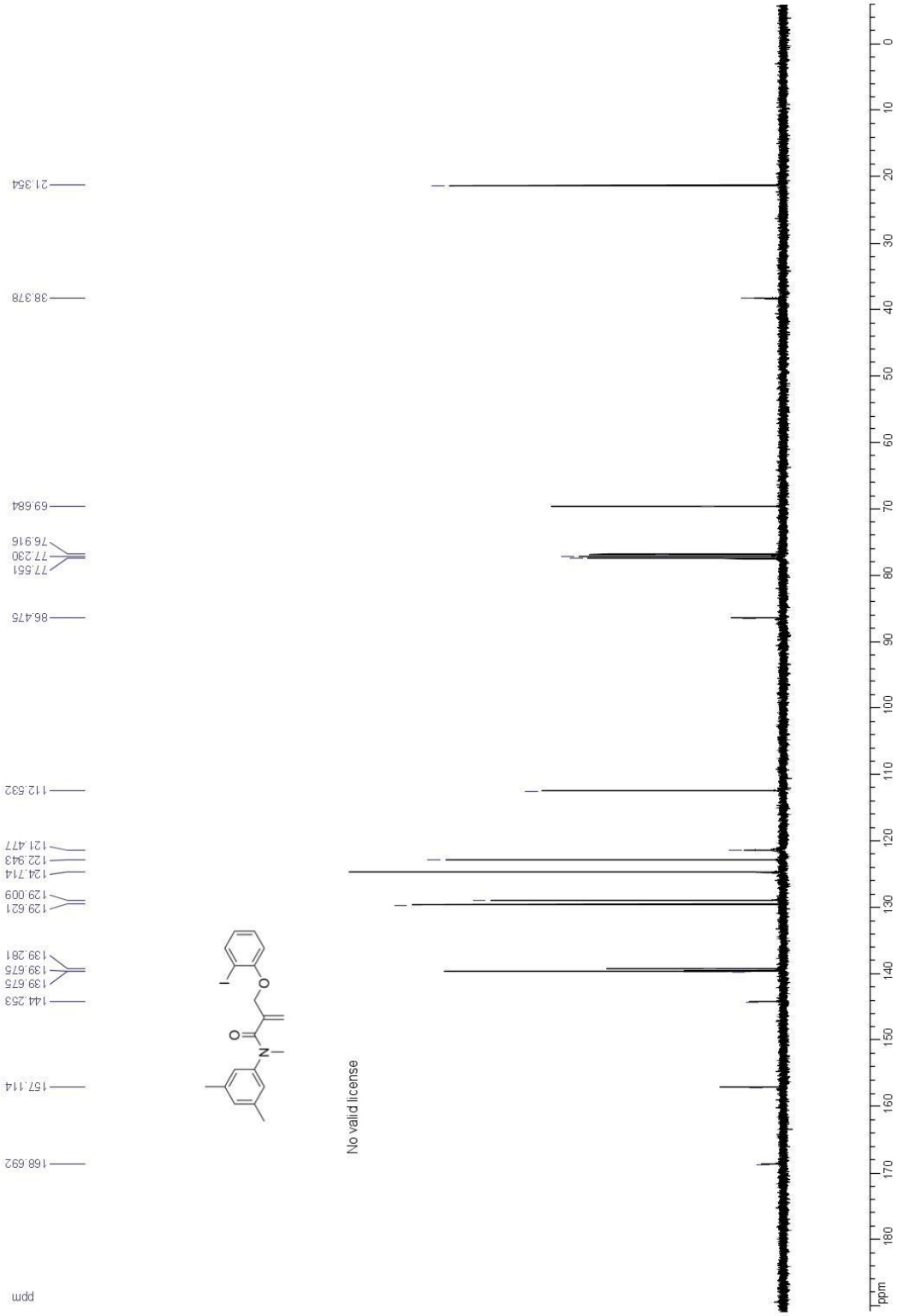
*Methyl 4-(2-((2-iodophenoxy)methyl)-N-methylacrylamido)benzoate (**1h**)*



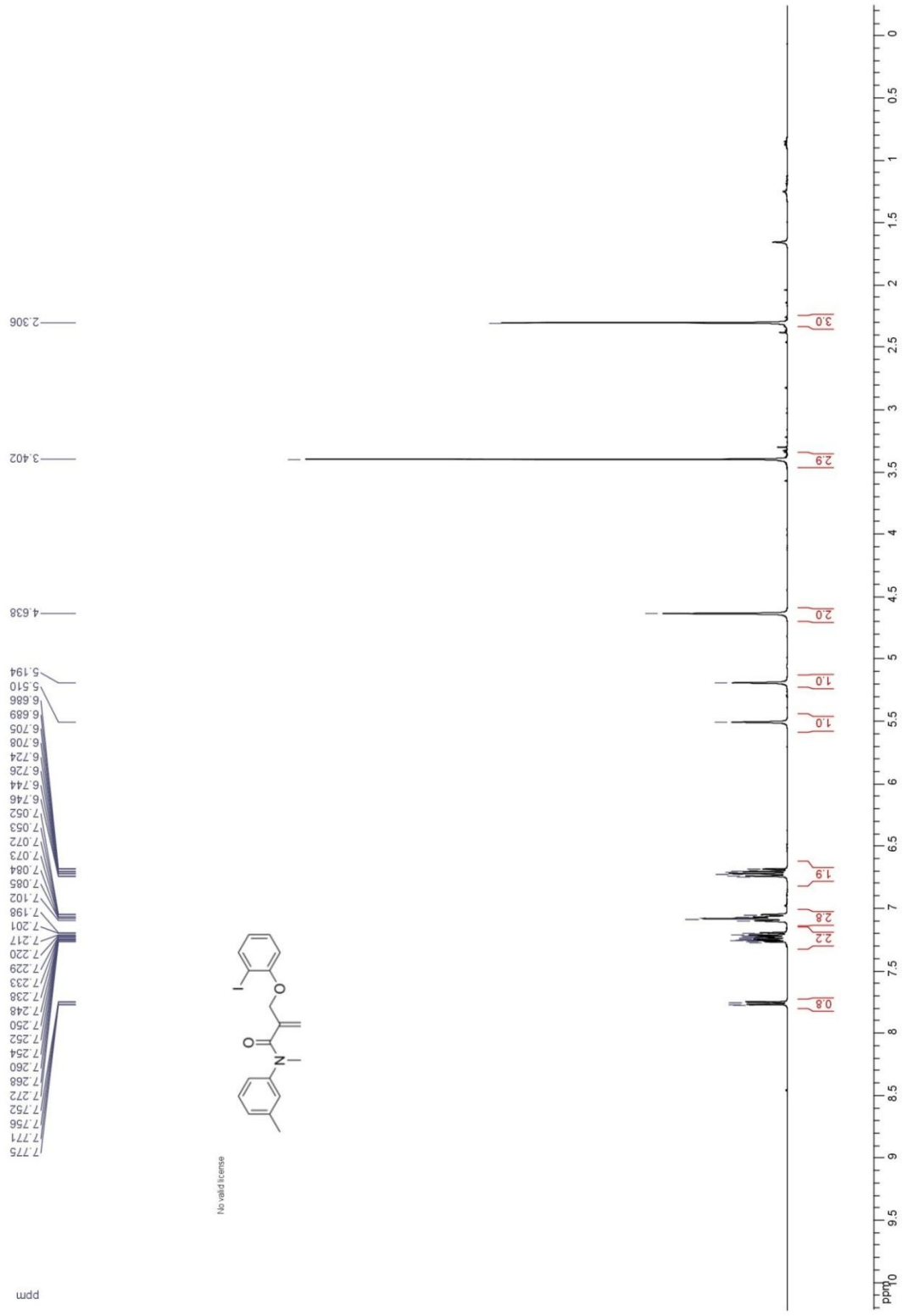
*N-(3,5-dimethylphenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1i***)*



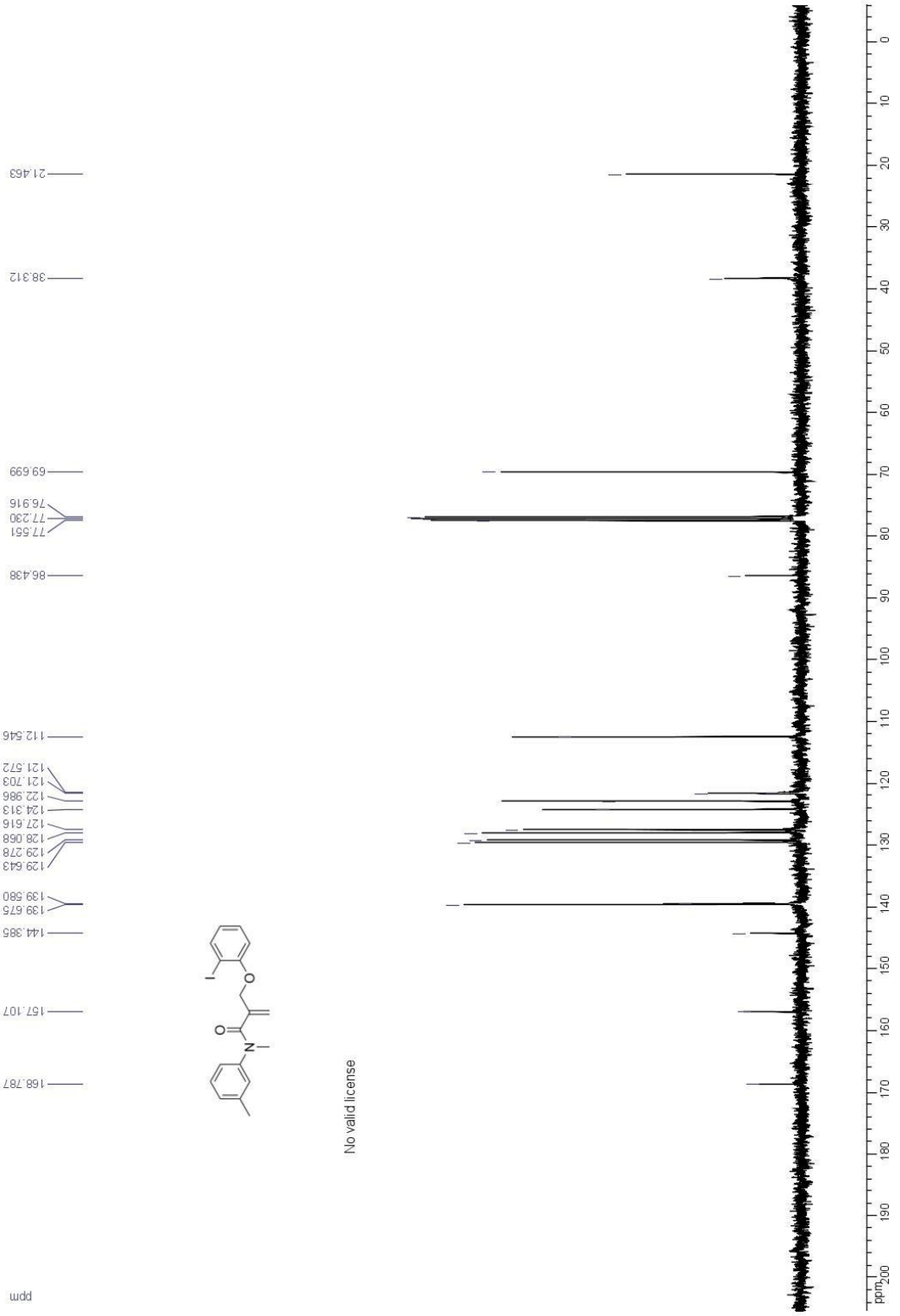
*N-(3,5-dimethylphenyl)-2-((2-iodophenoxy)methyl)-N-methylacrylamide (***1i***)*



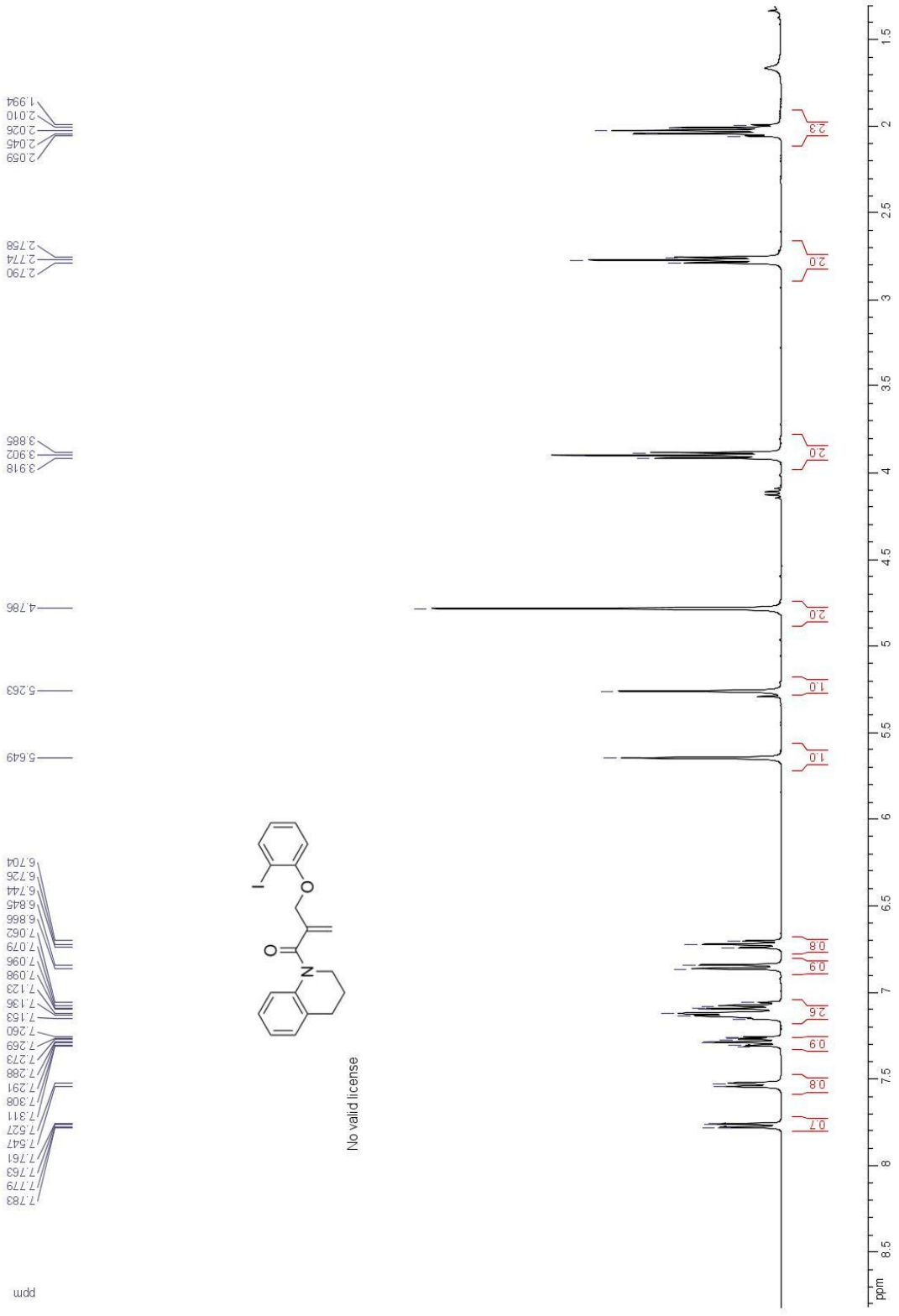
*2-((2-Iodophenoxy)methyl)-N-methyl-N-(m-tolyl)acrylamide (***1j***)*



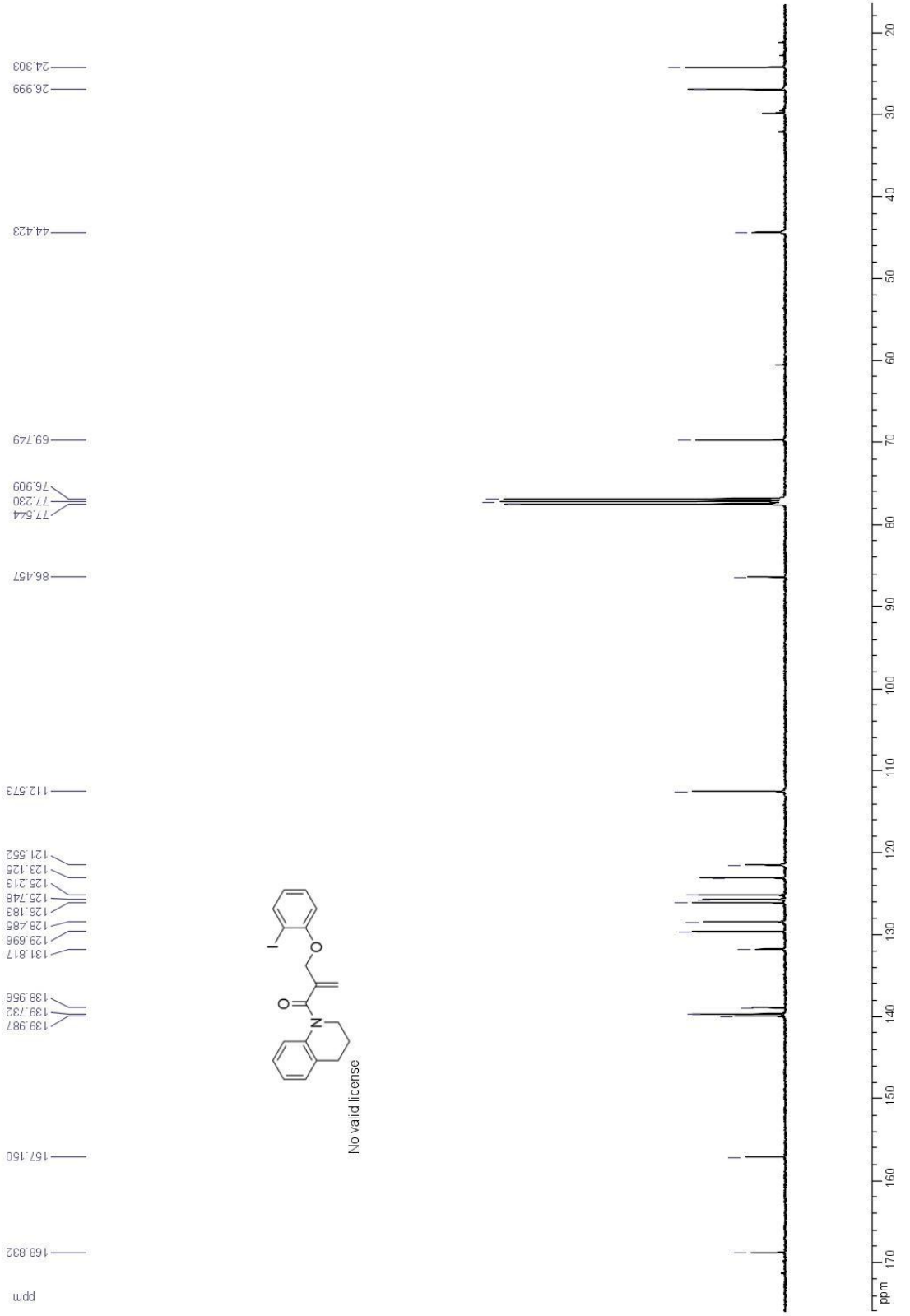
*2-((2-Iodophenoxy)methyl)-N-methyl-N-(m-tolyl)acrylamide (***1j***)*



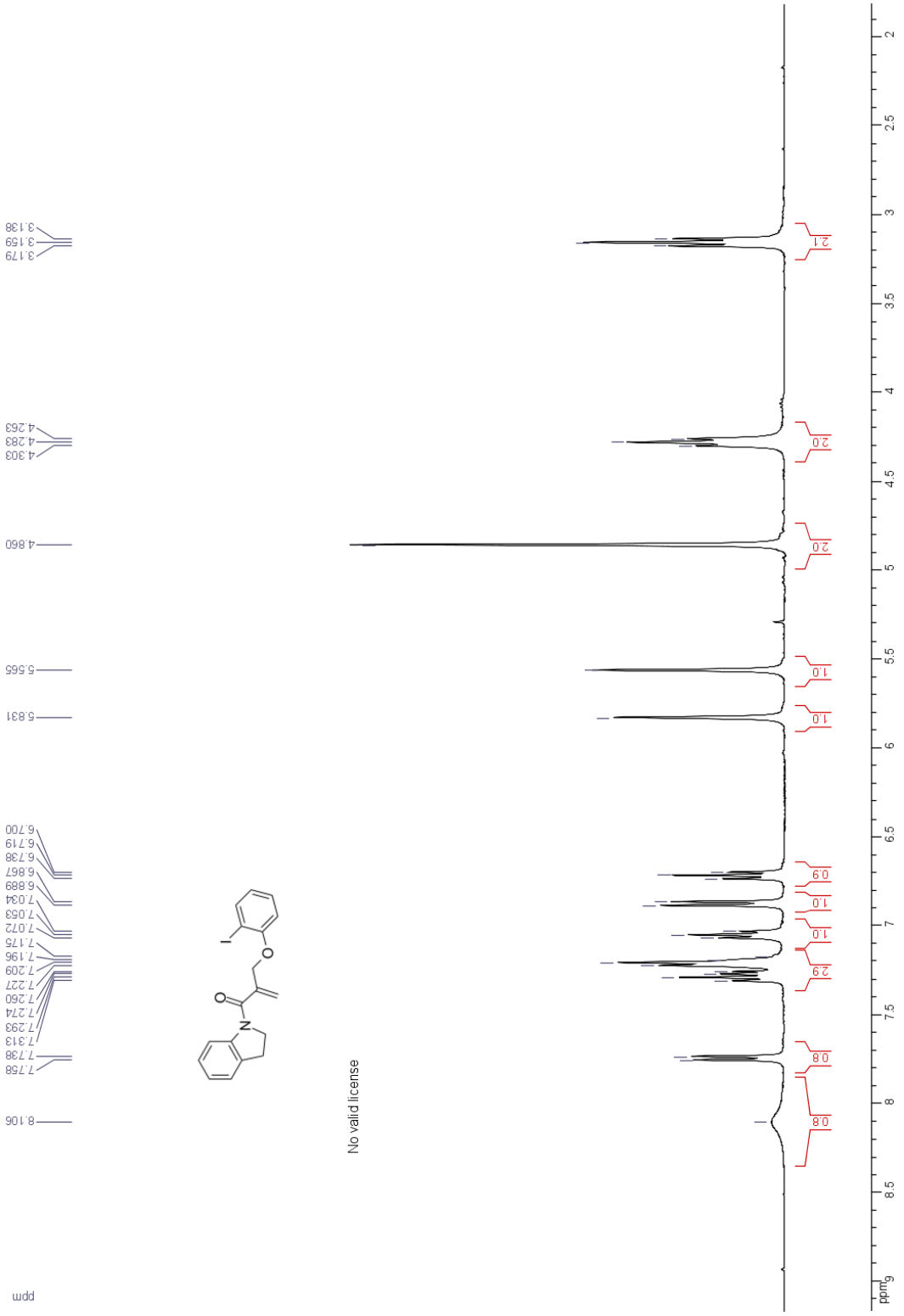
*1-(3,4-dihydroquinolin-1(2H)-yl)-2-((2-iodophenoxy)methyl)prop-2-en-1-one (***1k***)*



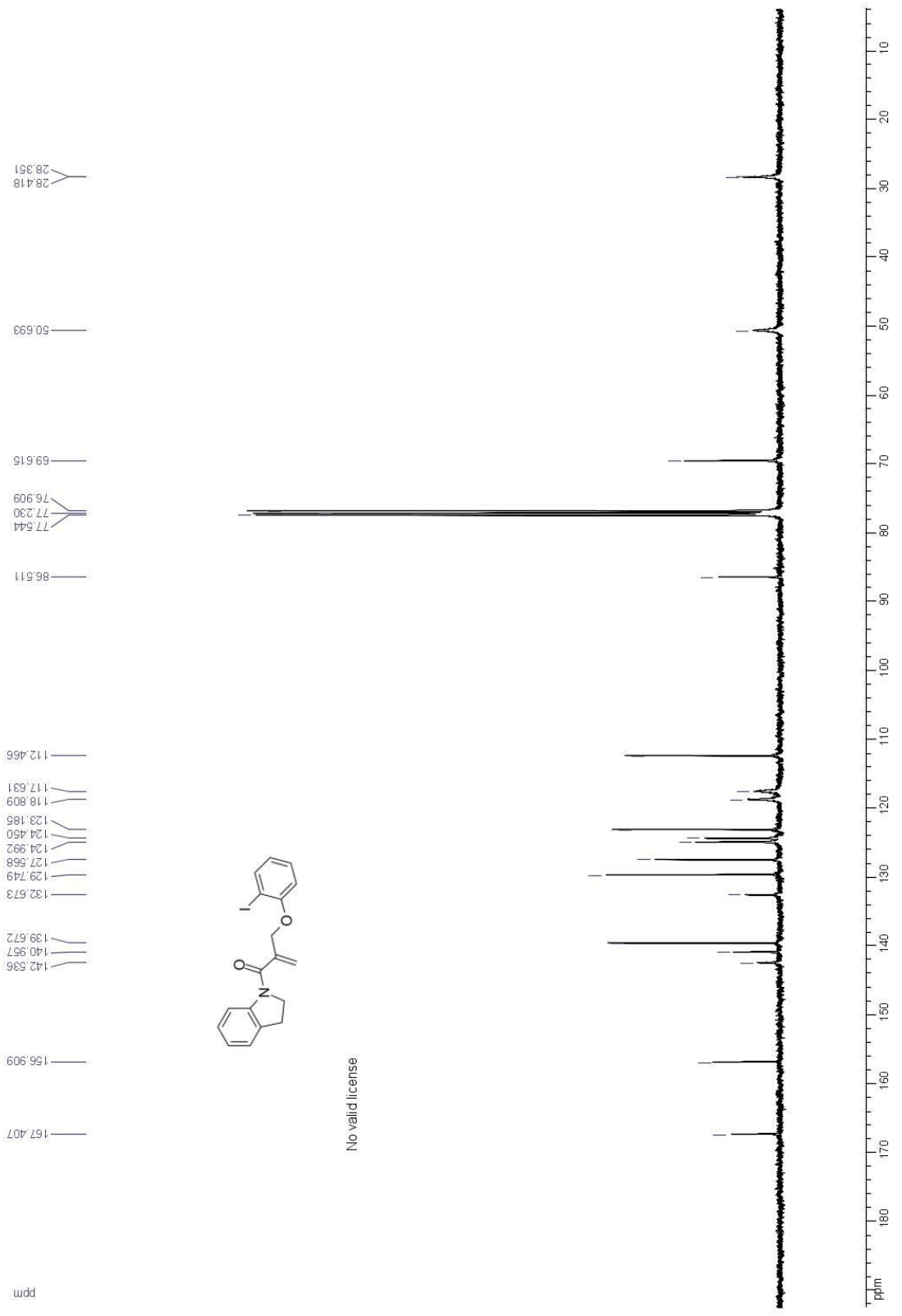
*1-(3,4-dihydroquinolin-1(2H)-yl)-2-((2-iodophenoxy)methyl)prop-2-en-1-one (***1k***)*



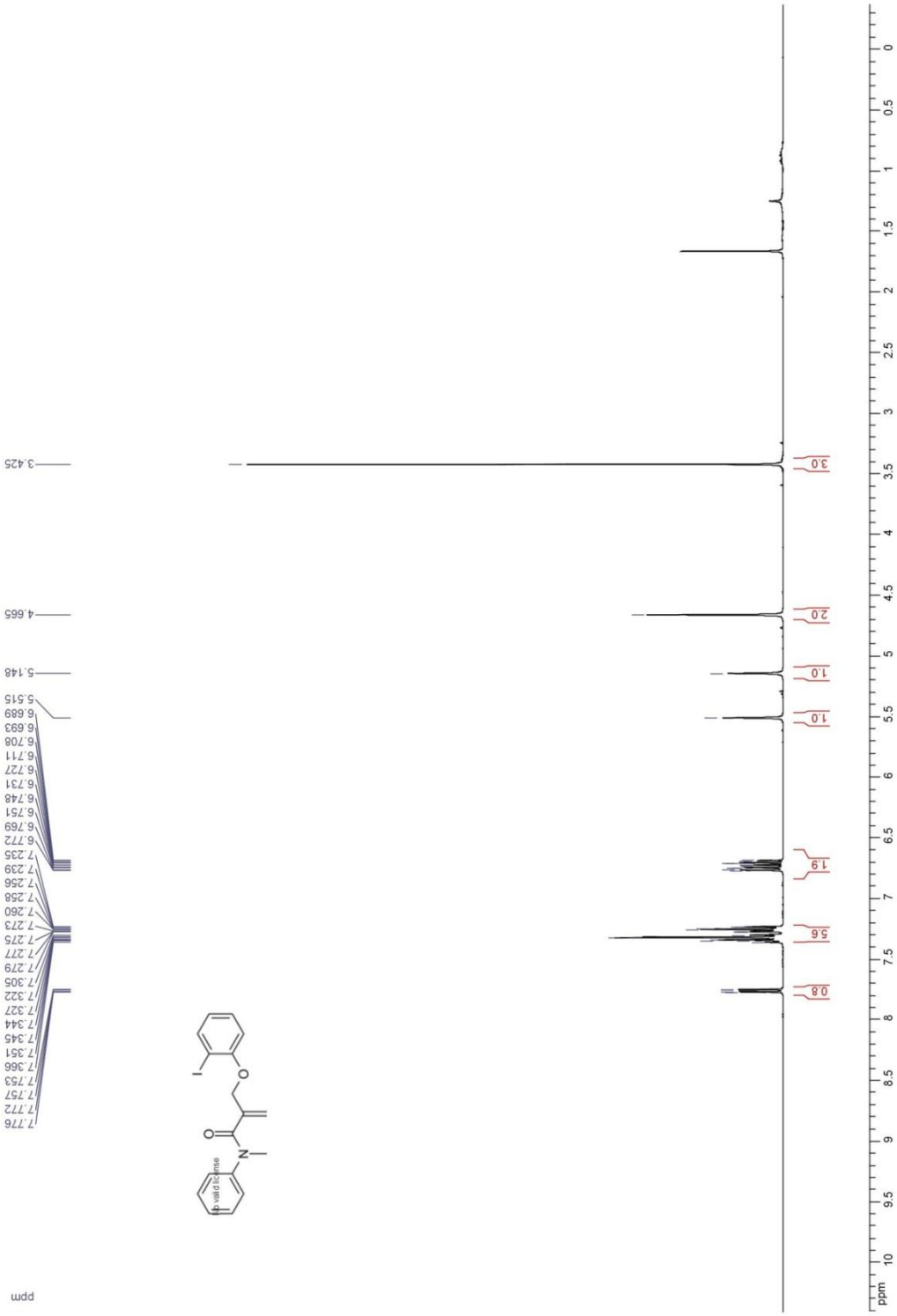
*1-(indolin-1-yl)-2-((2-iodo henoxy)methyl)prop-2-en-1-one (**1l**)*



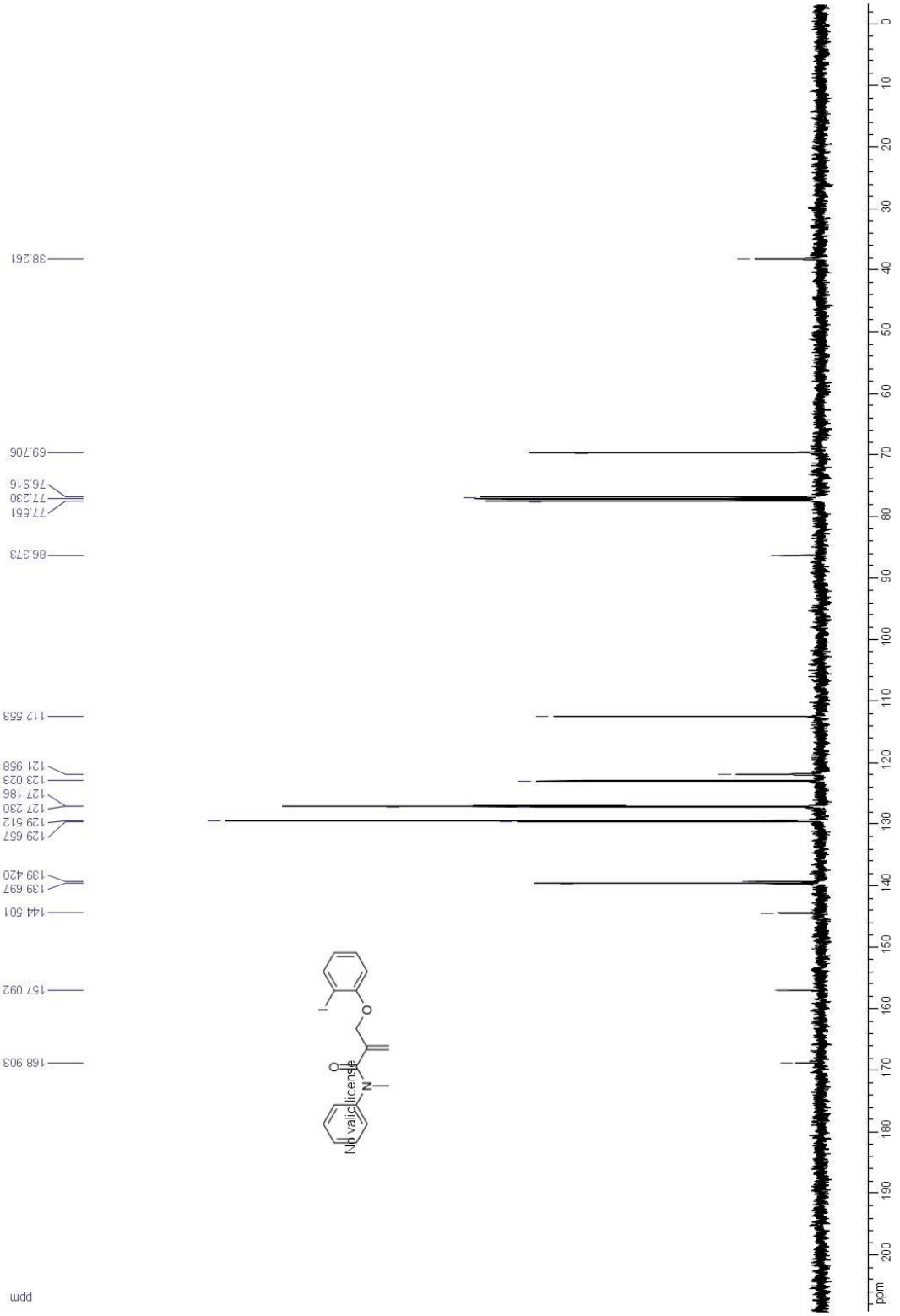
*1-(indolin-1-yl)-2-((2-iodophenoxy)methyl)prop-2-en-1-one (**1l**)*



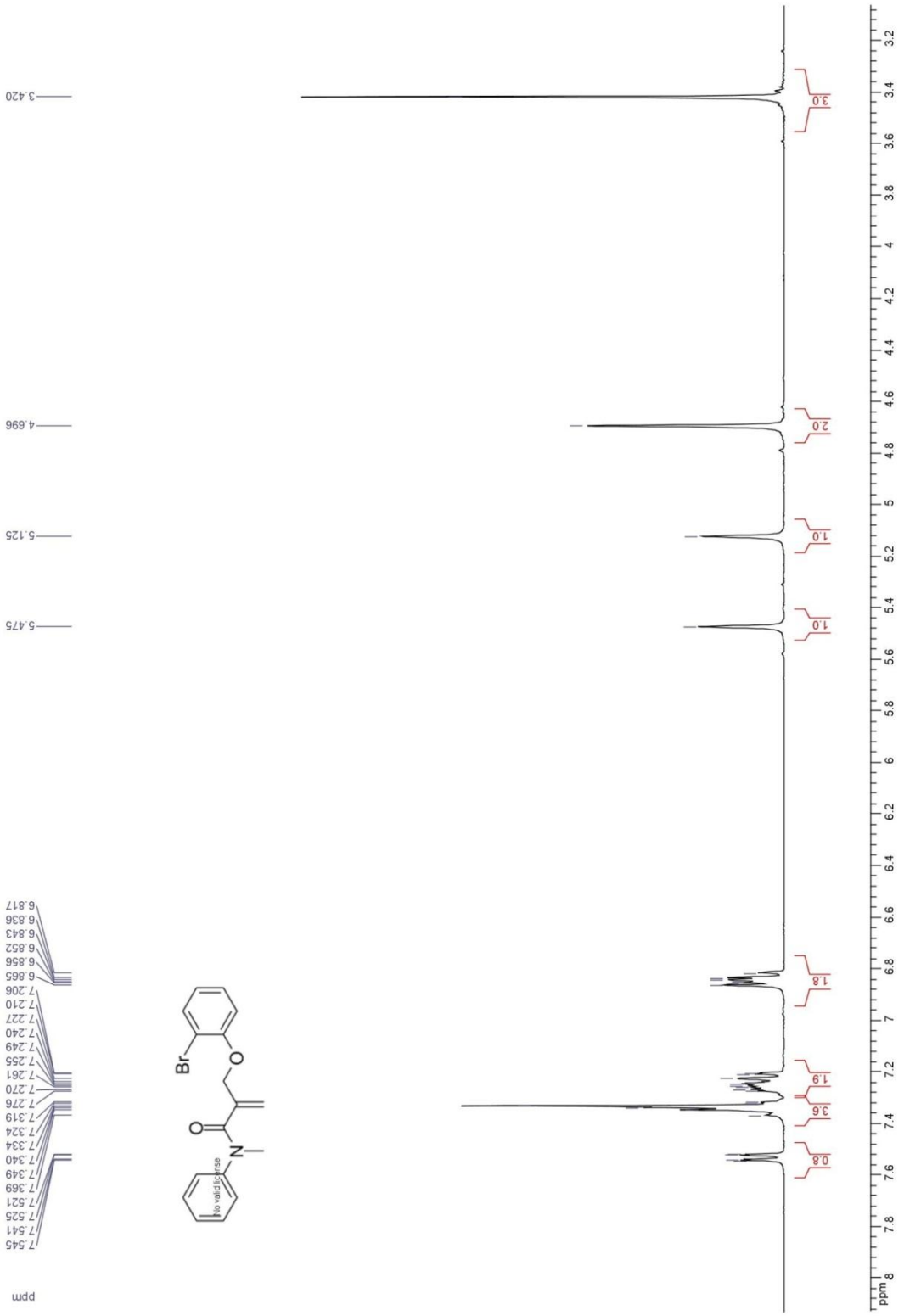
*2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a***)*



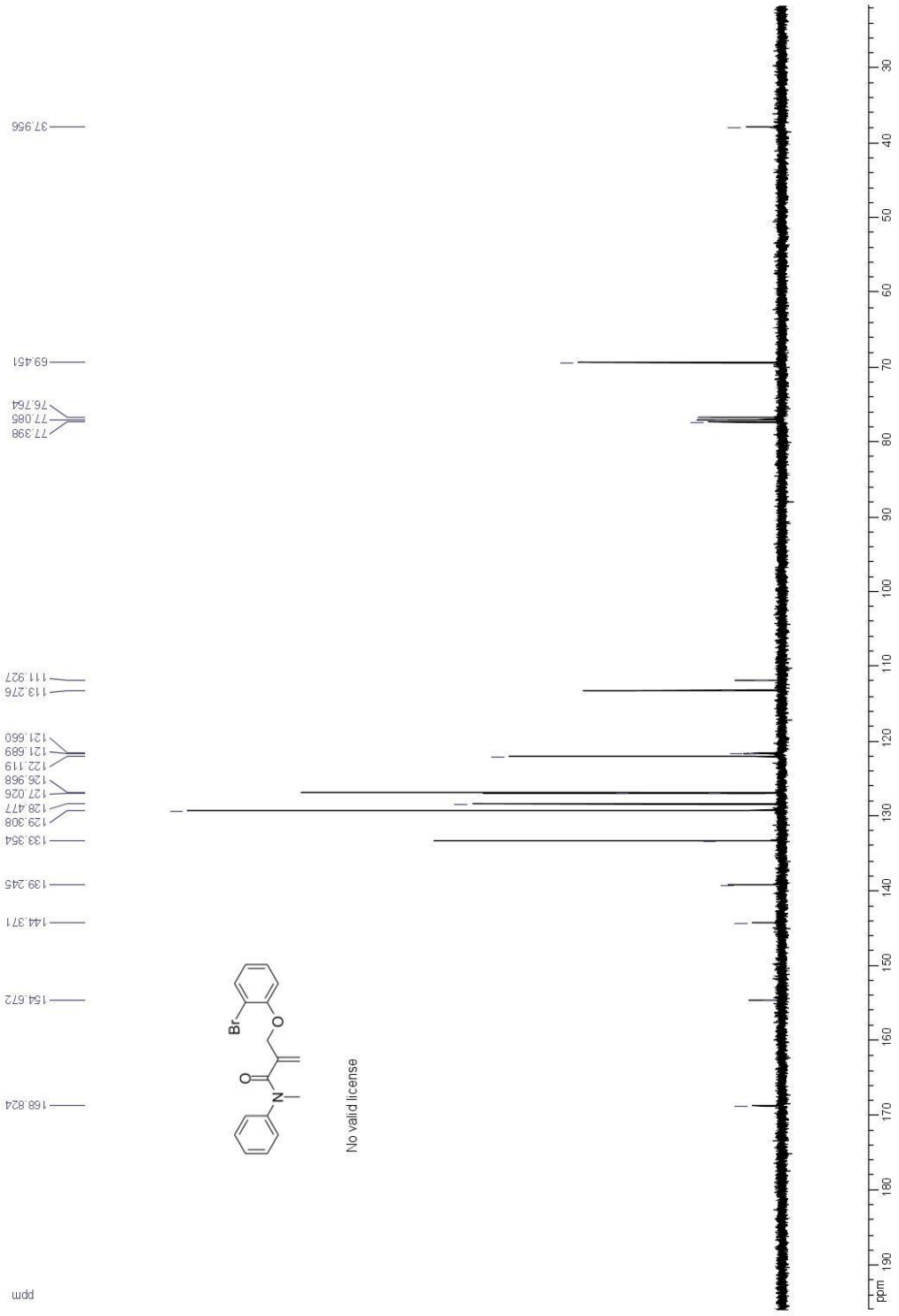
*2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a***)*



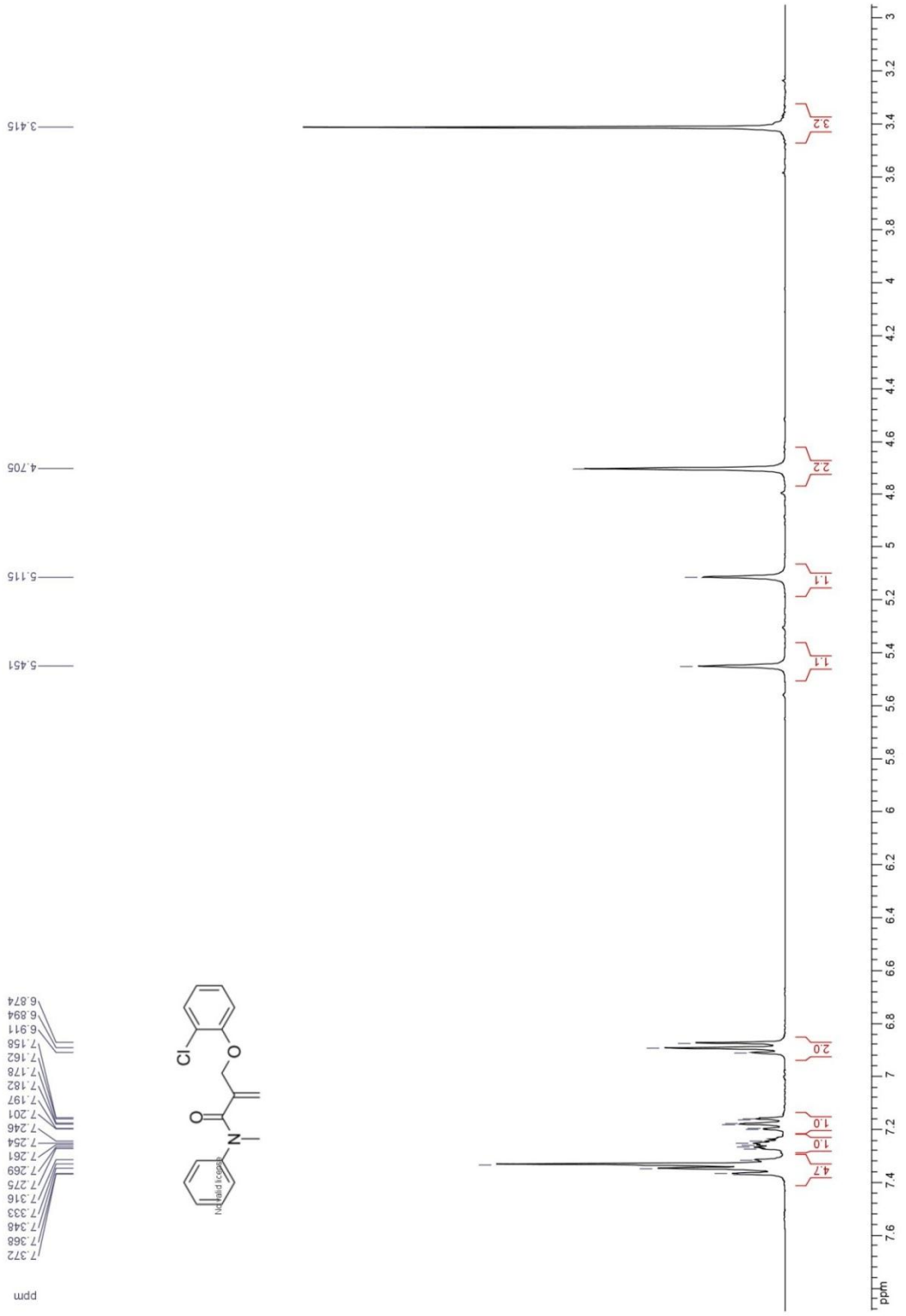
*2-((2-bromophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a, X = Br***)*



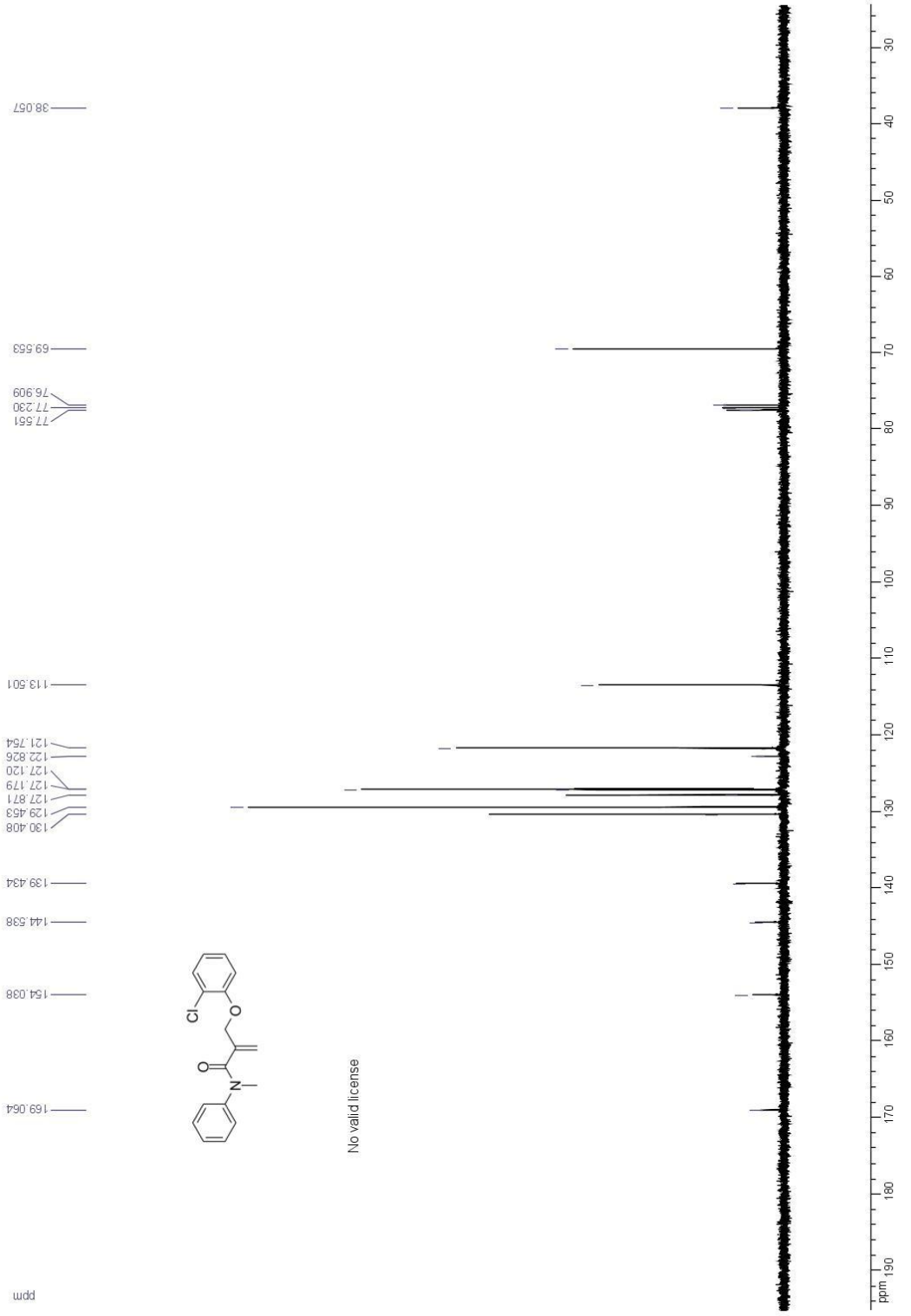
*2-((2-bromophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a, X = Br***)*



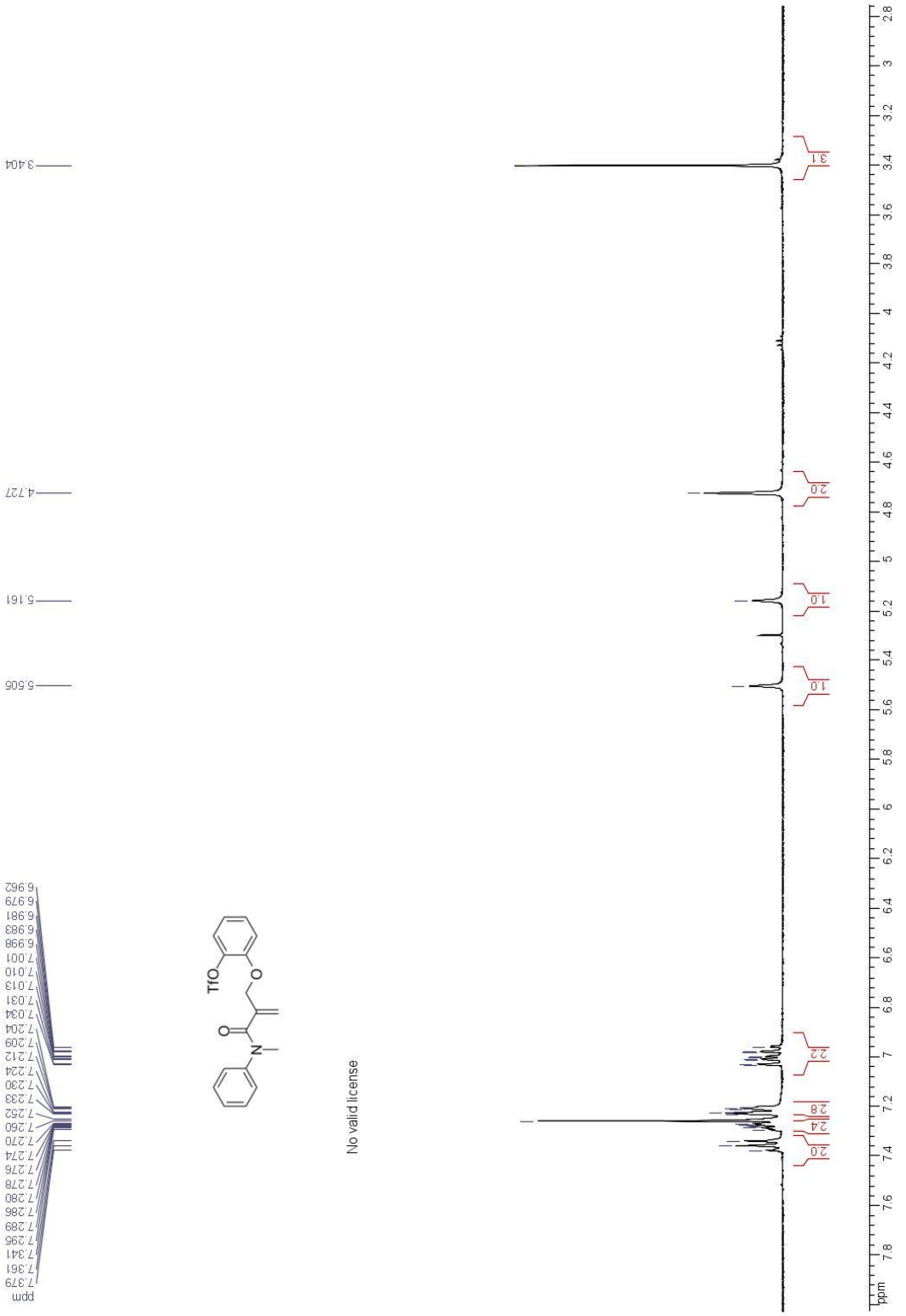
*2-((2-chlorophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a, X = Cl***)*



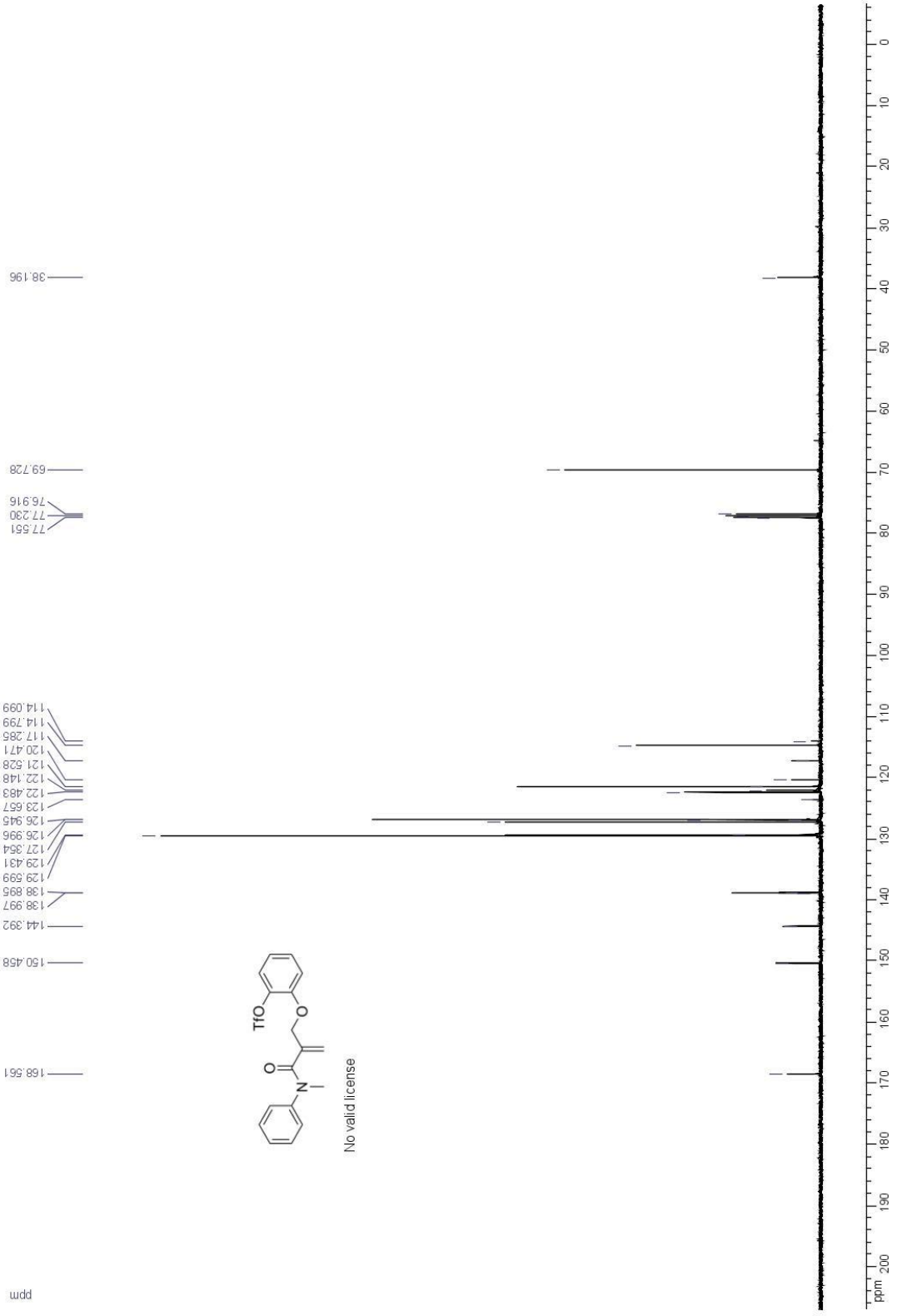
*2-((2-chlorophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1a, X = Cl***)*



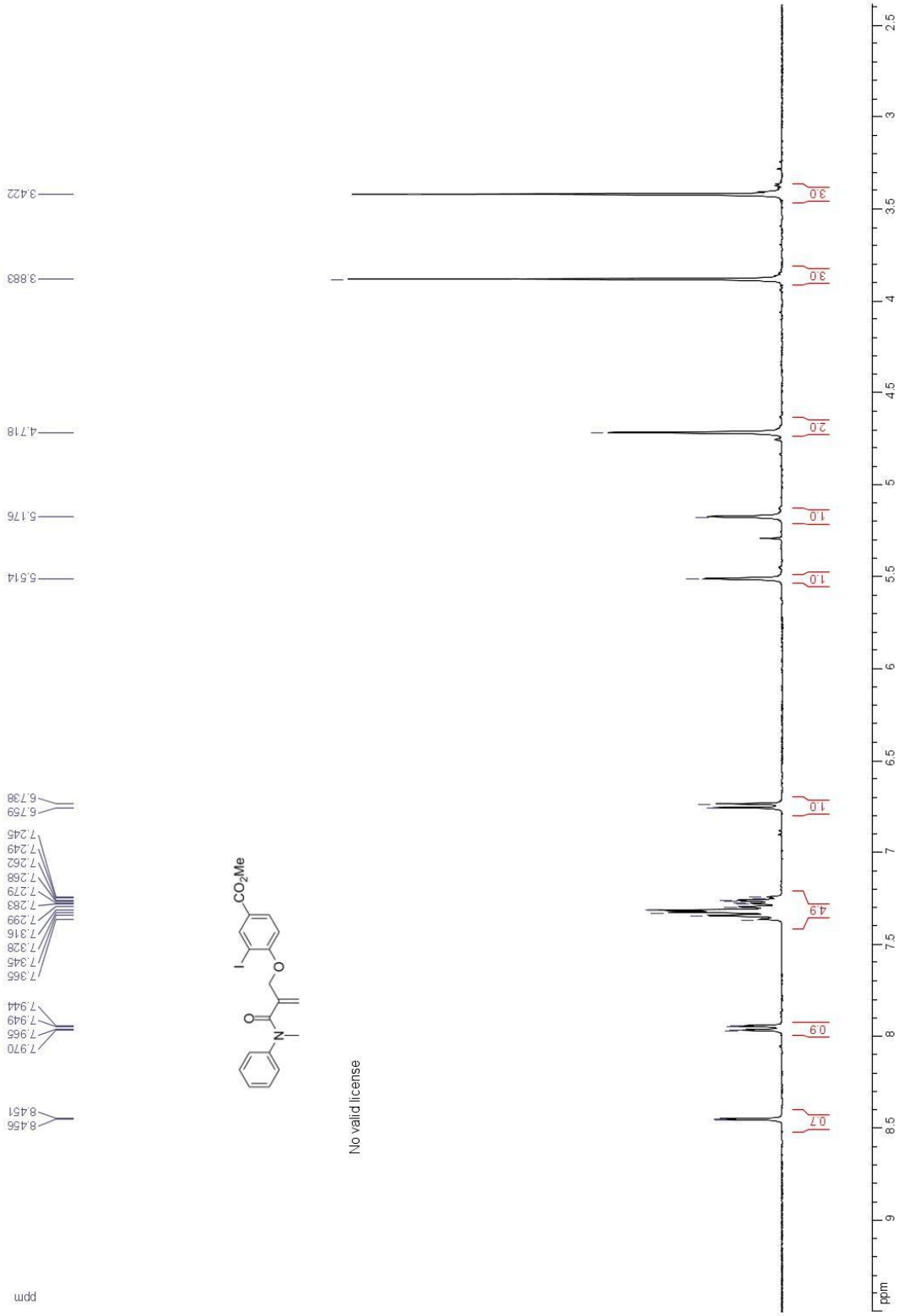
*2-((2-(methyl(phenyl)carbamoyl)allyl)oxy)phenyl trifluoromethanesulfonate (***1a, X = OTf***)*



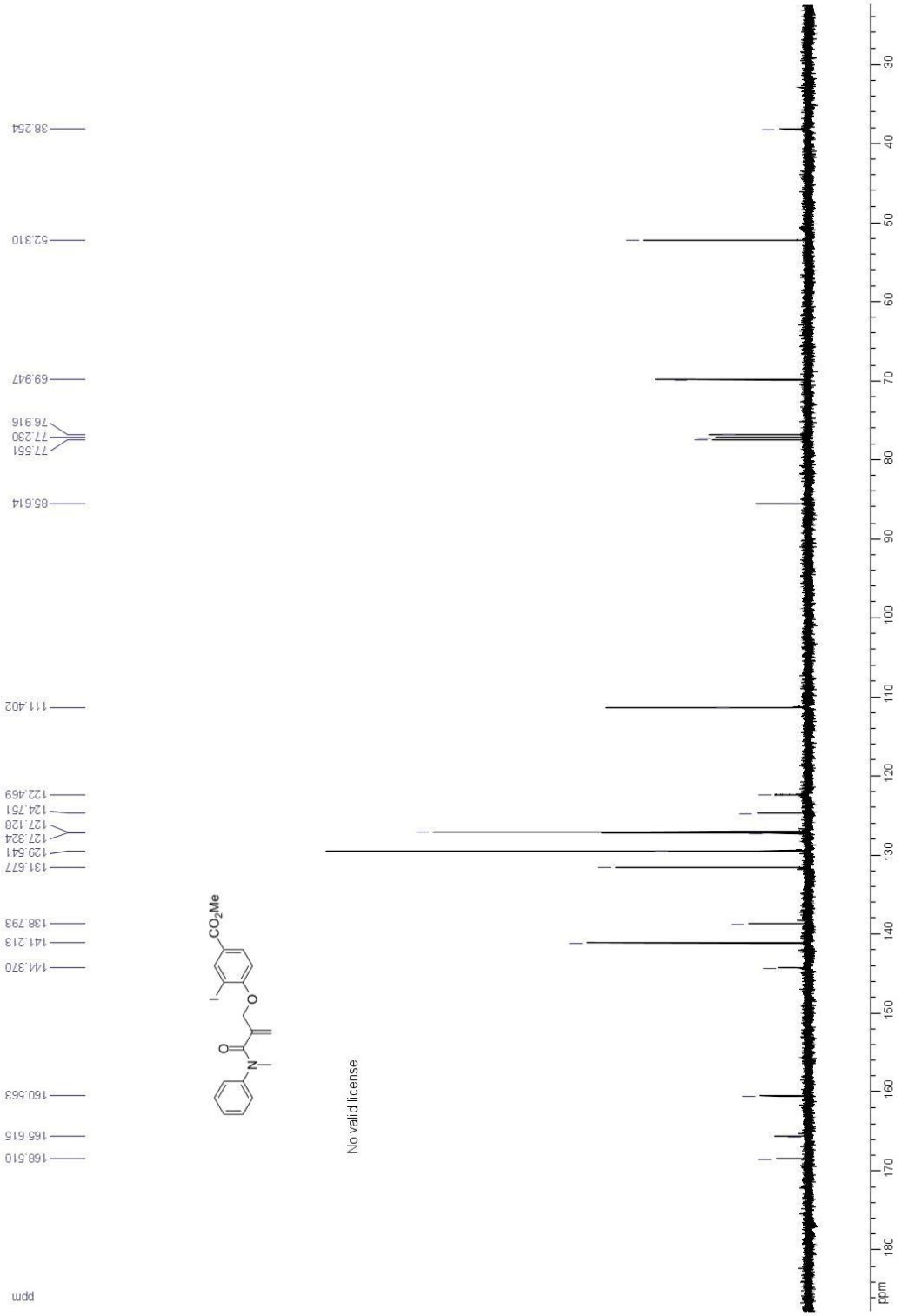
*2-((2-(methyl(phenyl)carbamoyl)allyl)oxy)phenyl trifluoromethanesulfonate (***1a, X = OTf***)*



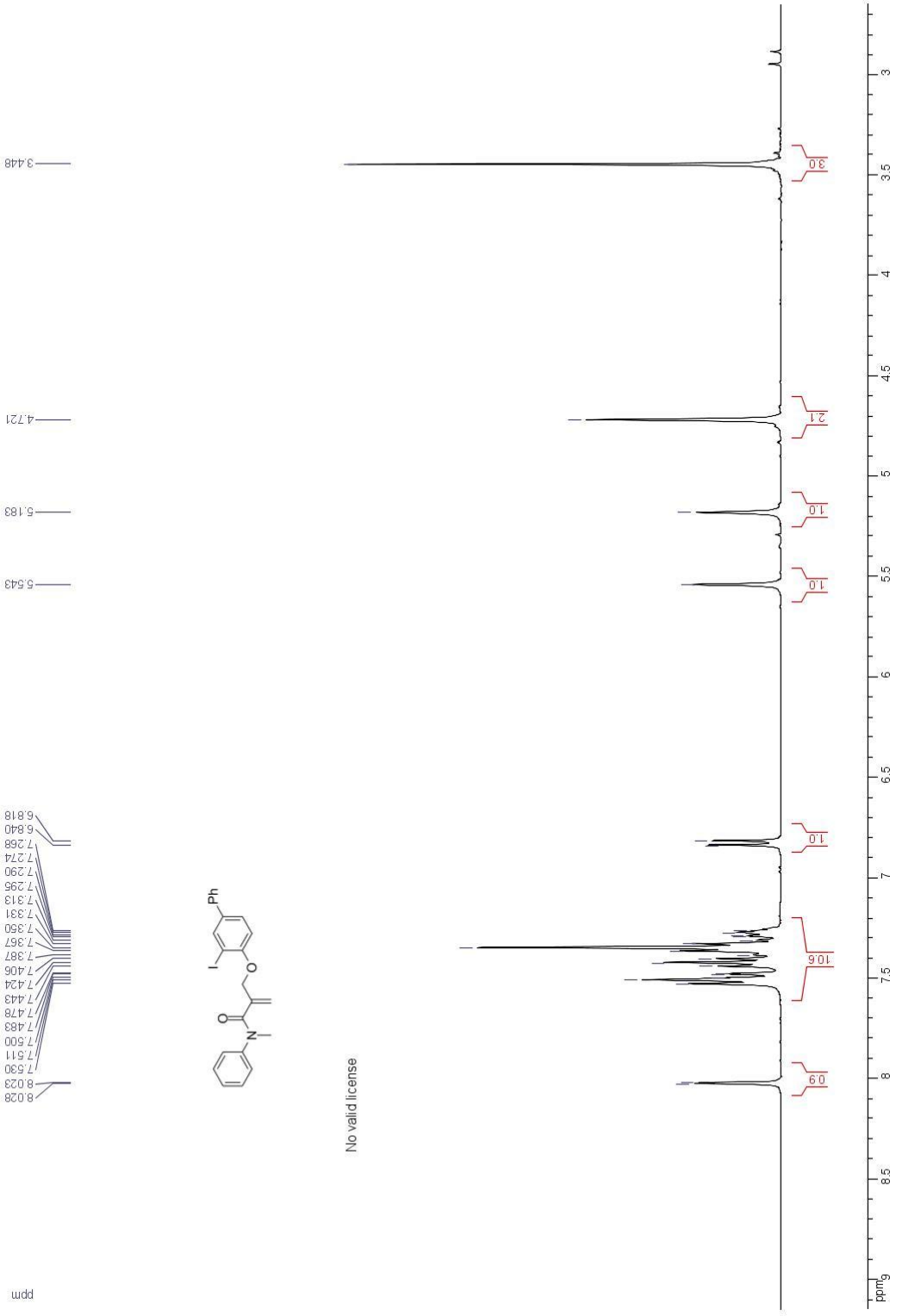
*methyl 3-iodo-4-((2-(methyl(phenyl)carbamoyl)allyl)oxy)benzoate (***1p***)*



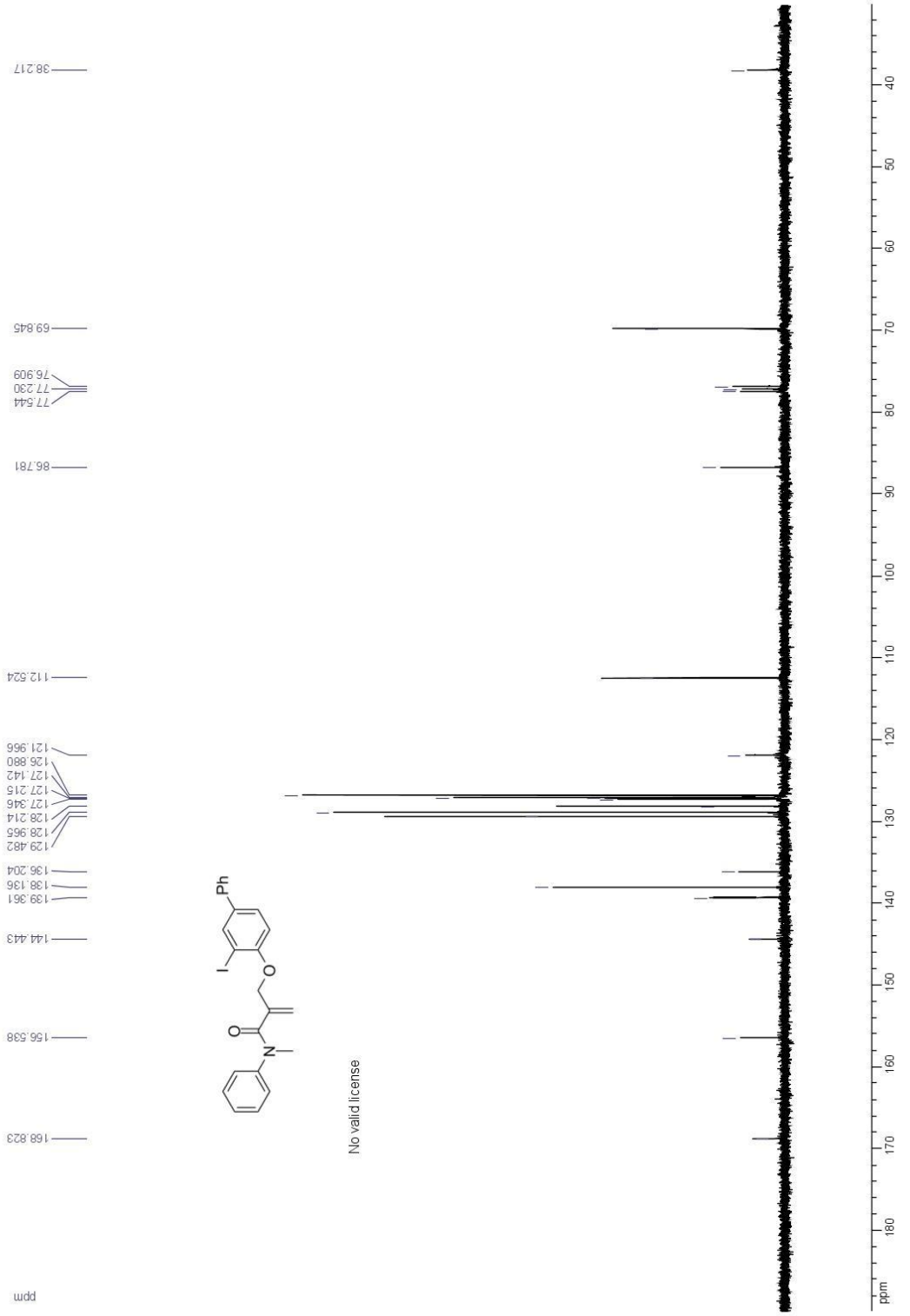
*methyl 3-iodo-4-((2-(methyl(phenyl)carbamoyl)allyl)oxy)benzoate (***1p***)*



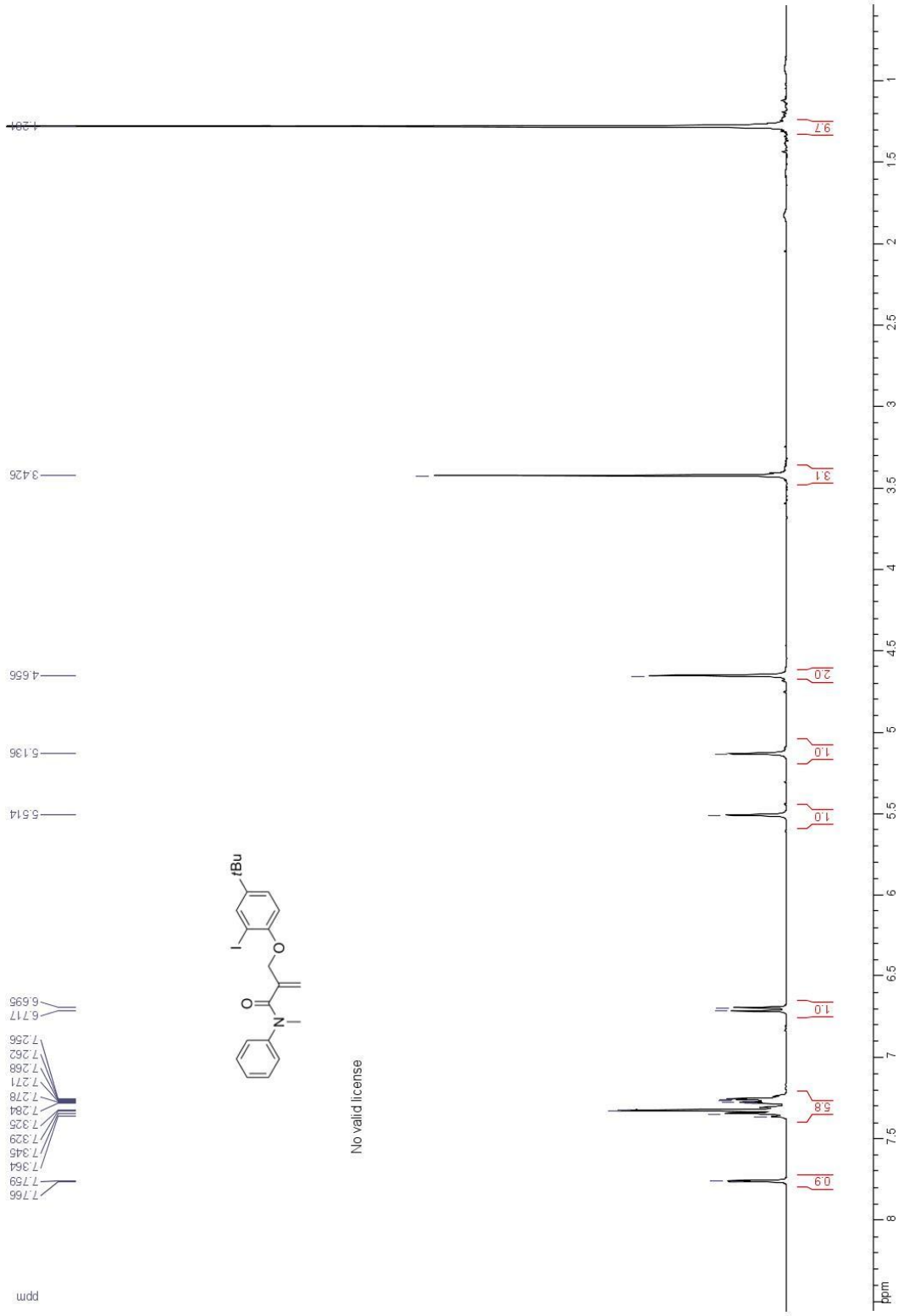
*2-(((3-iodo-[1,1'-biphenyl]-4-yl)oxy)methyl)-N-methyl-N-phenylacrylamide (***1q***)*



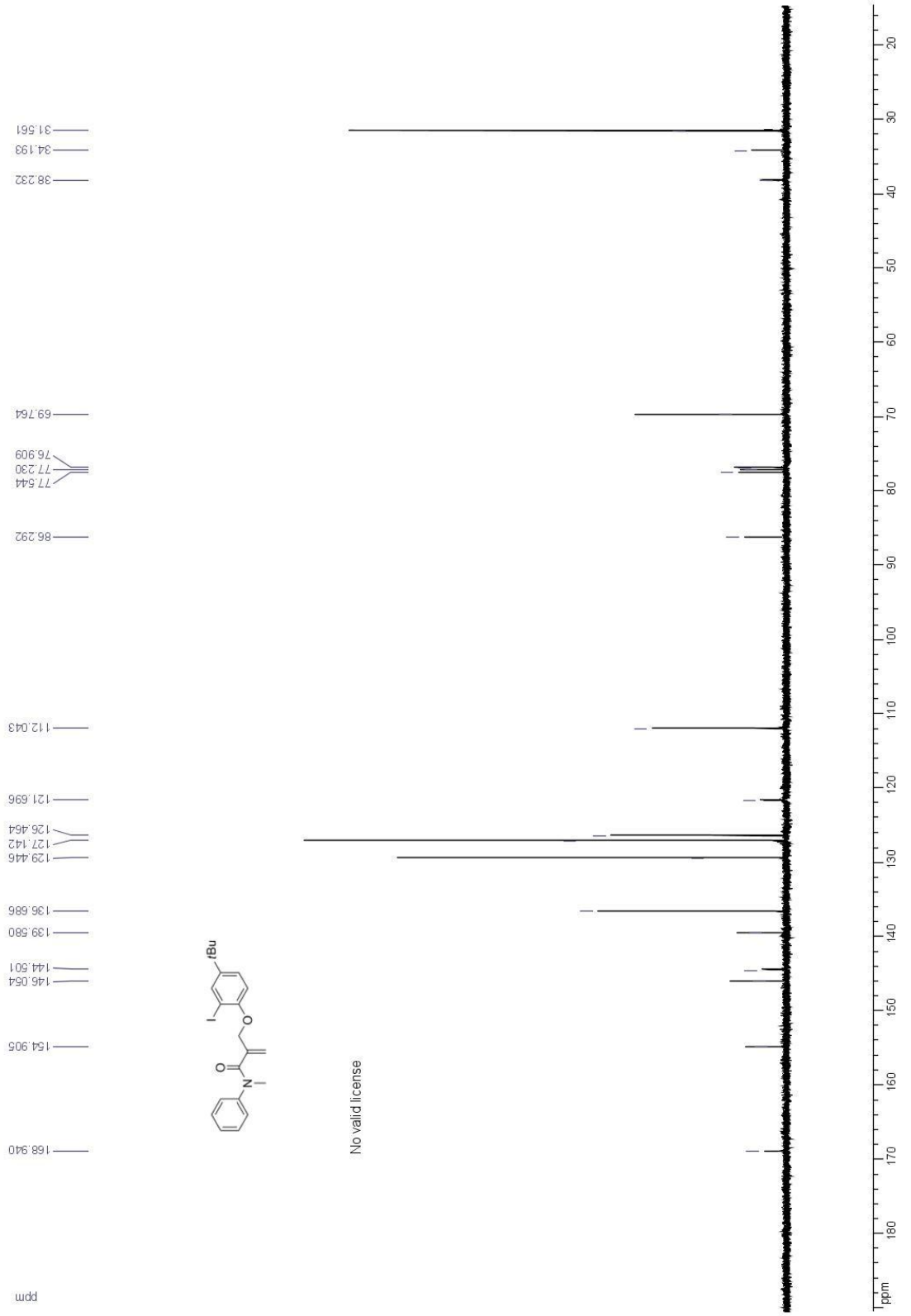
*2-(((3-iodo-[1,1'-biphenyl]-4-yl)oxy)methyl)-N-methyl-N-phenylacrylamide (***1q***)*



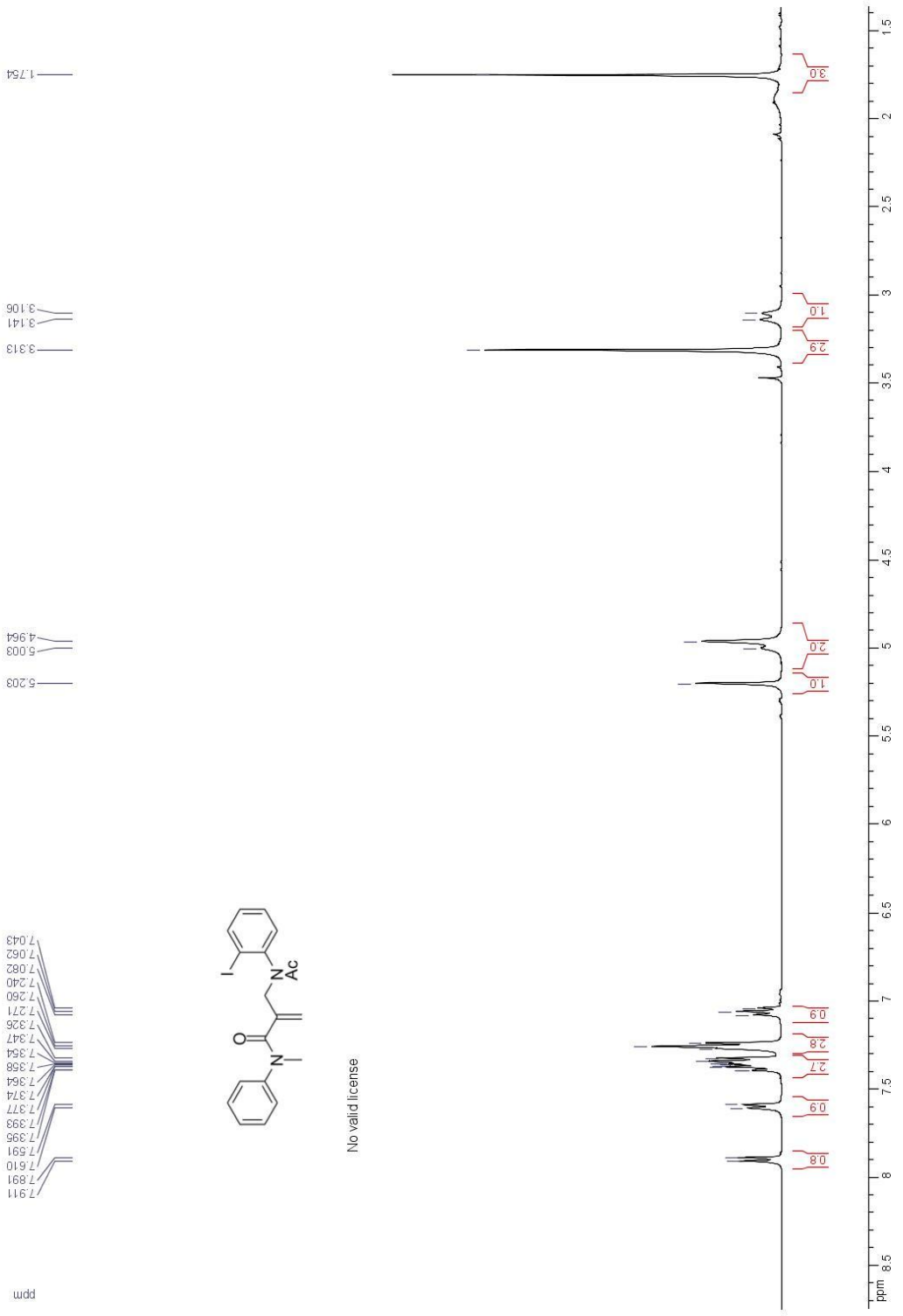
*2-((4-(tert-butyl)-2-iodophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1r***)*



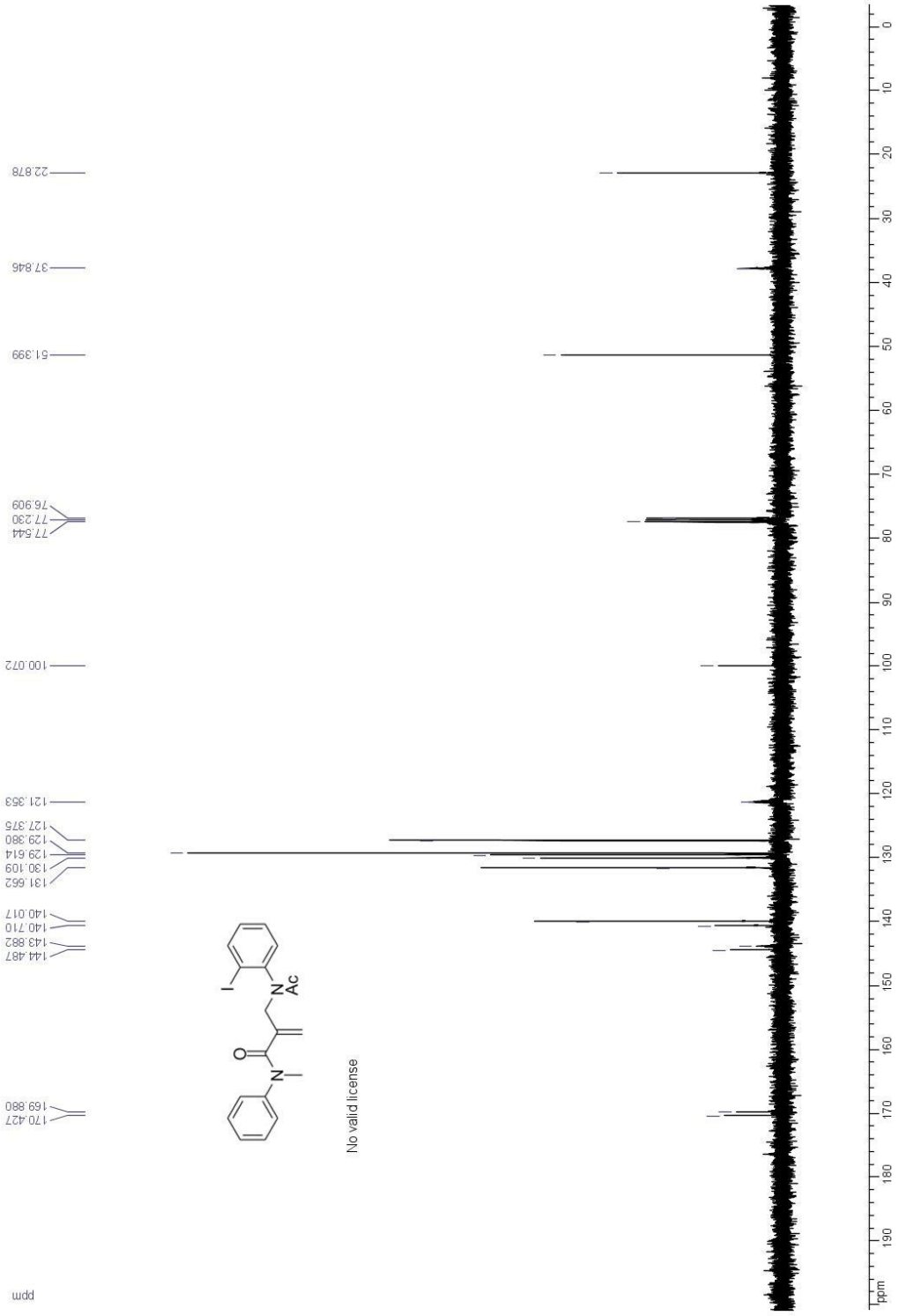
*2-((4-(tert-butyl)-2-iodophenoxy)methyl)-N-methyl-N-phenylacrylamide (***1r***)*



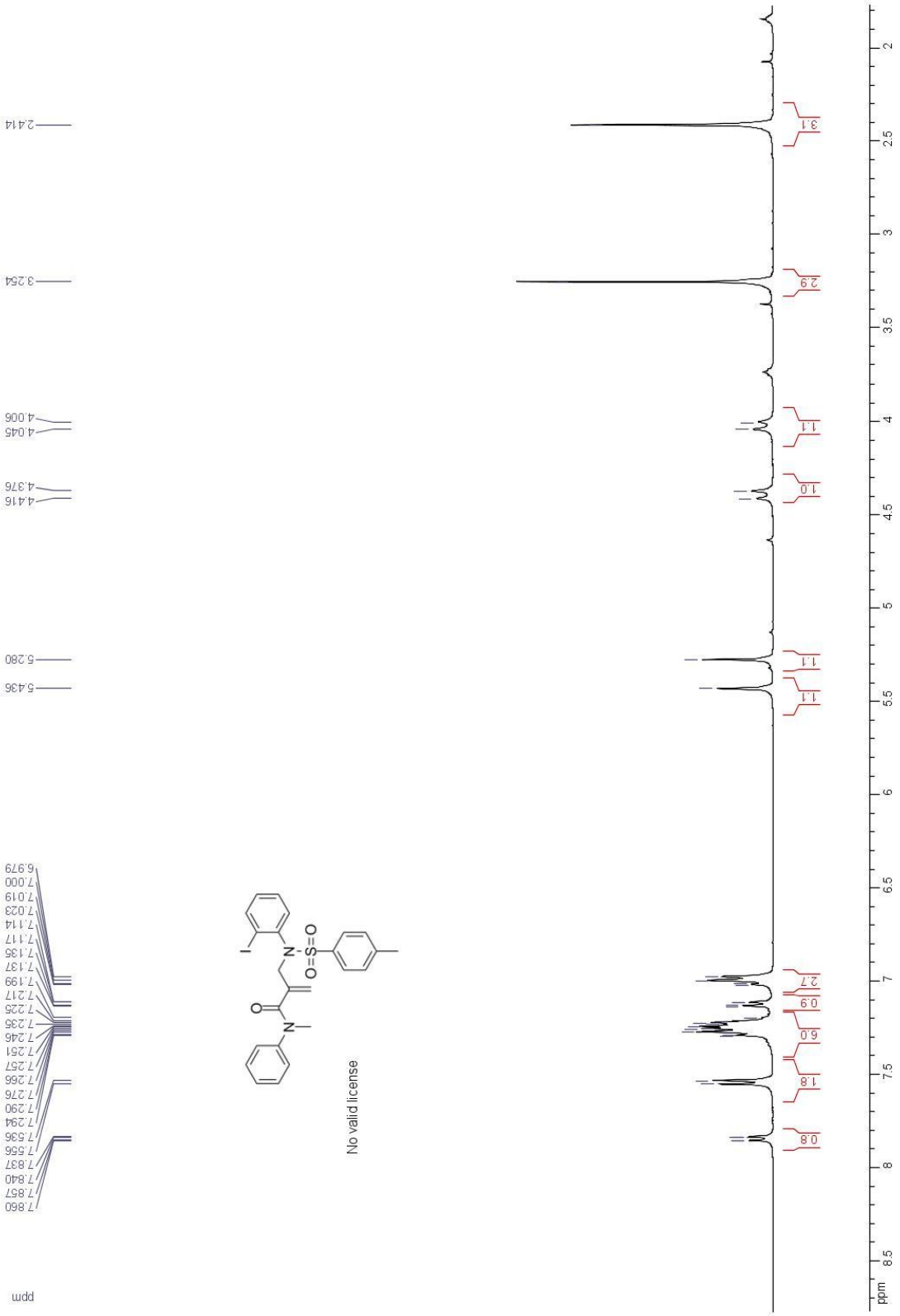
*2-((N-(2-iodophenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1t***)*



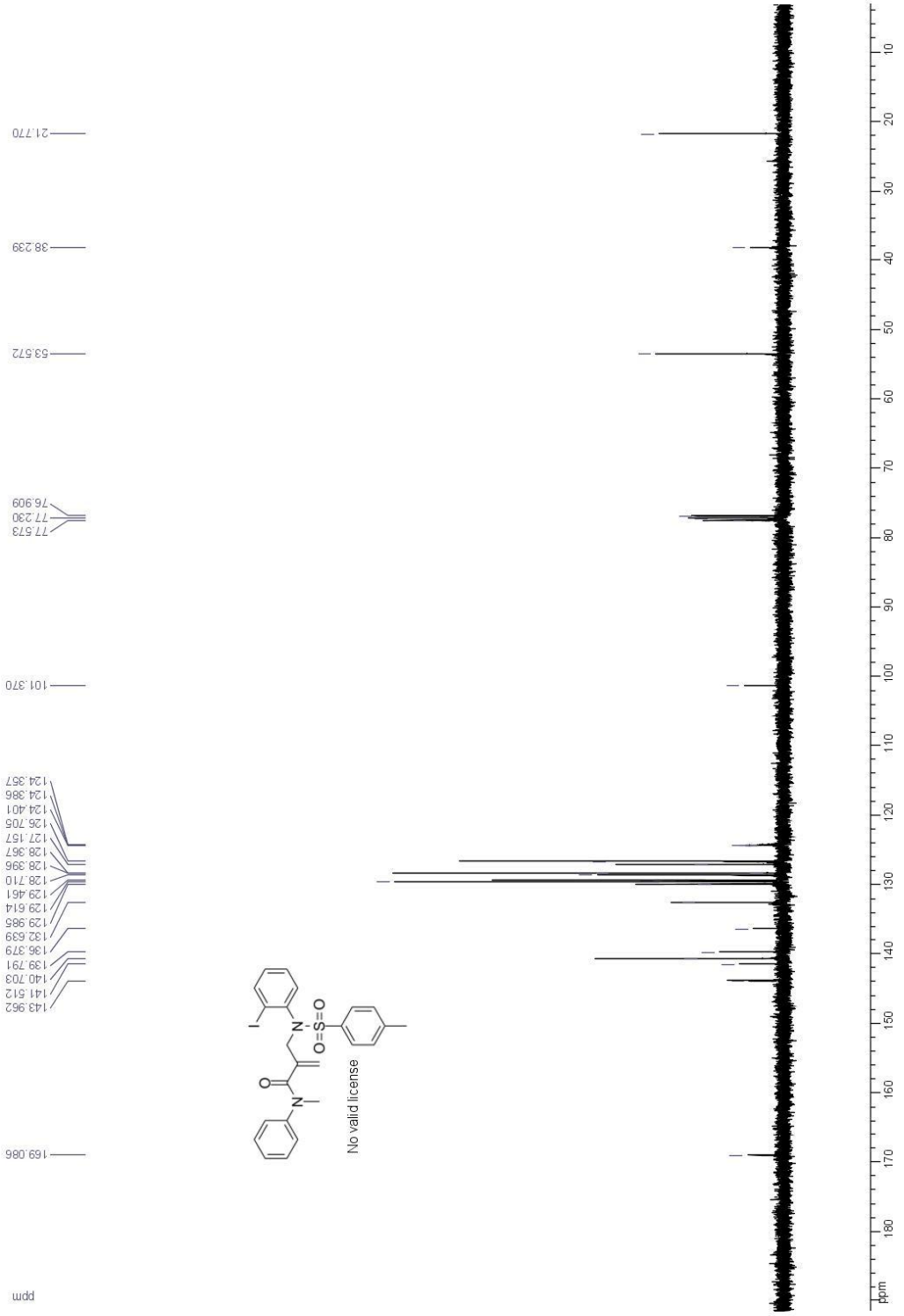
*2-((N-(2-iodophenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1t***)*



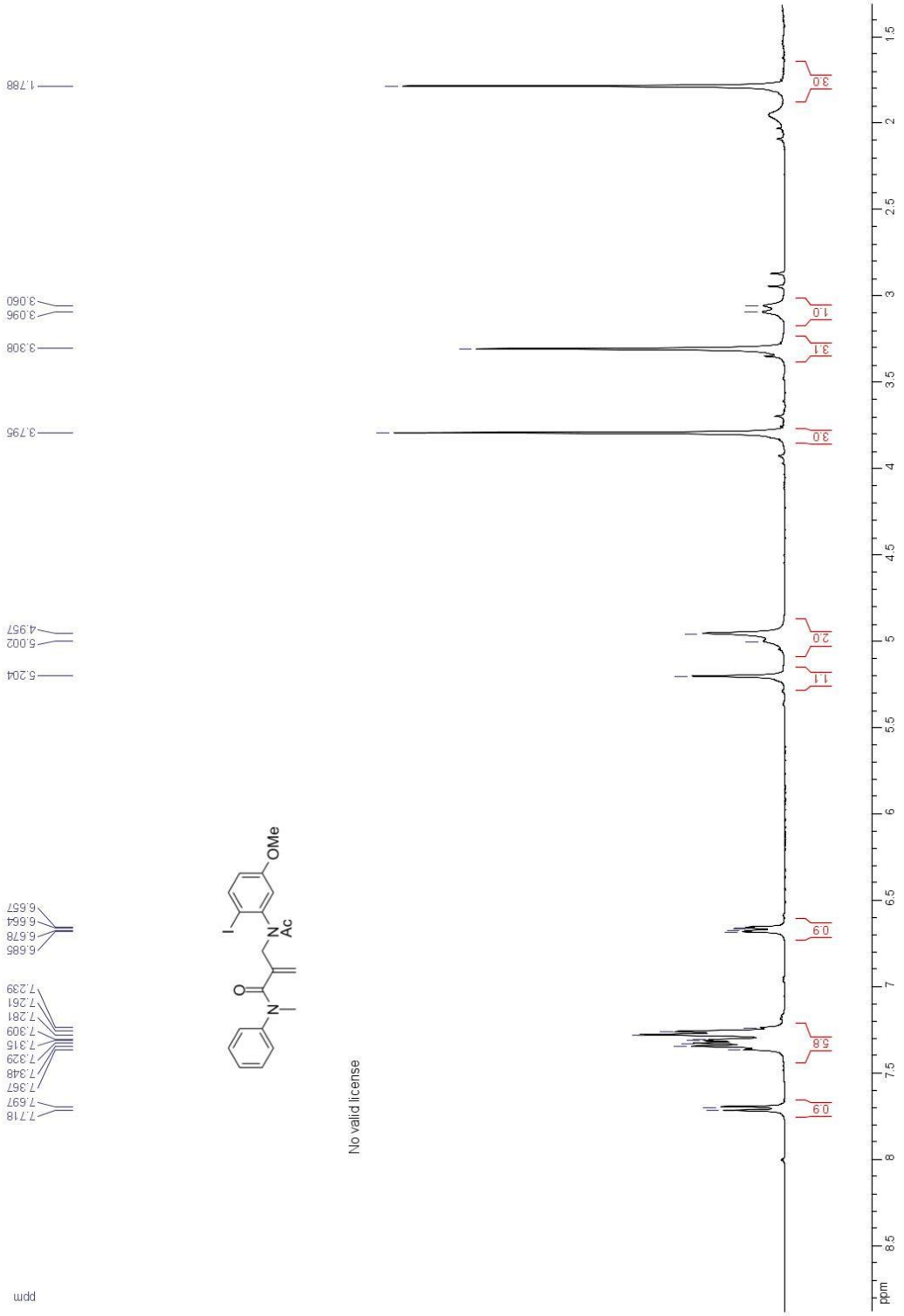
*2-((N-(2-iodophenyl)-4-methylphenylsulfonamido)methyl)-N-methyl-N-phenylacrylamide (***1u***)*



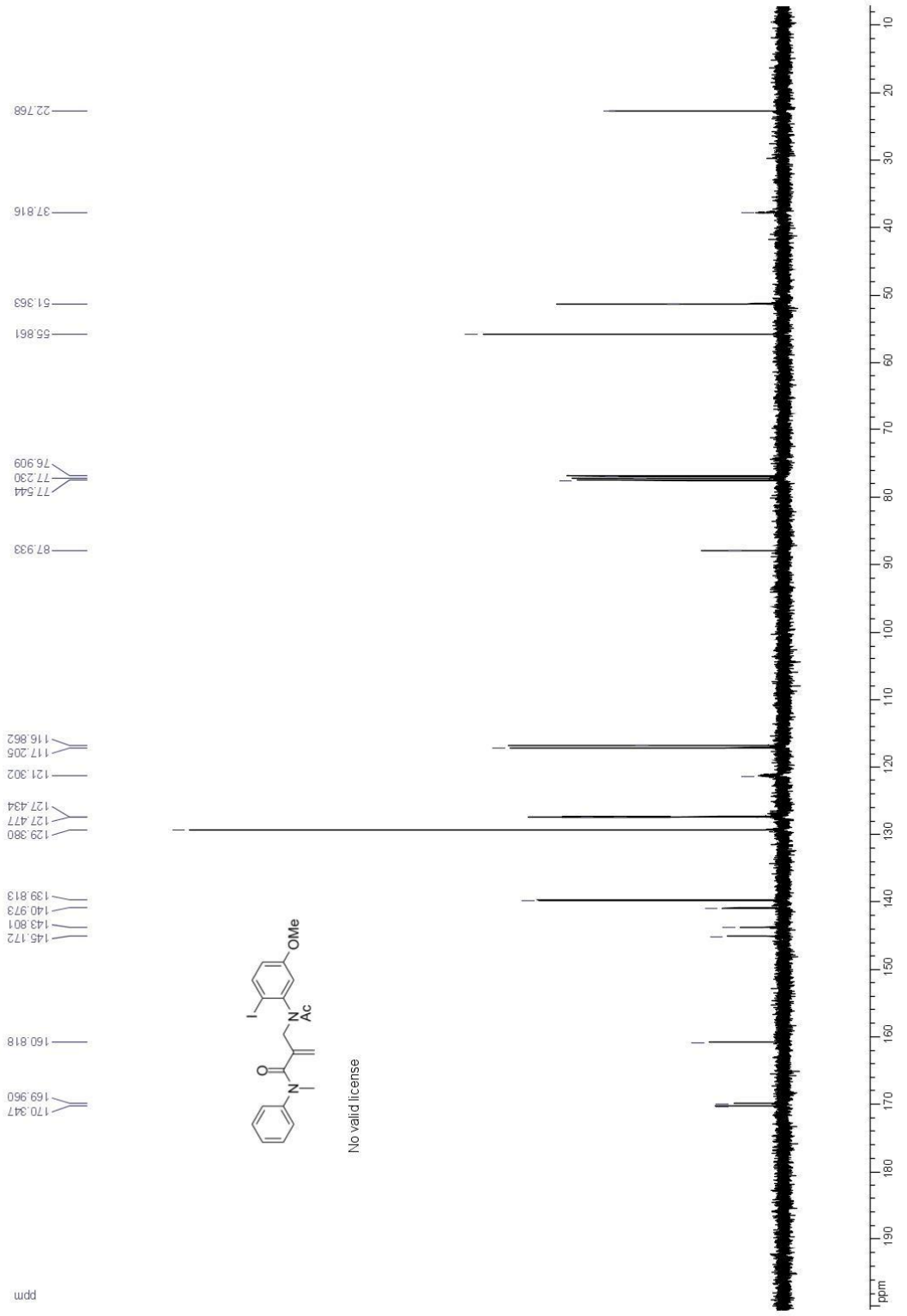
*2-((N-(2-iodophenyl)-4-methylphenylsulfonamido)methyl)-N-methyl-N-phenylacrylamide (***1u***)*



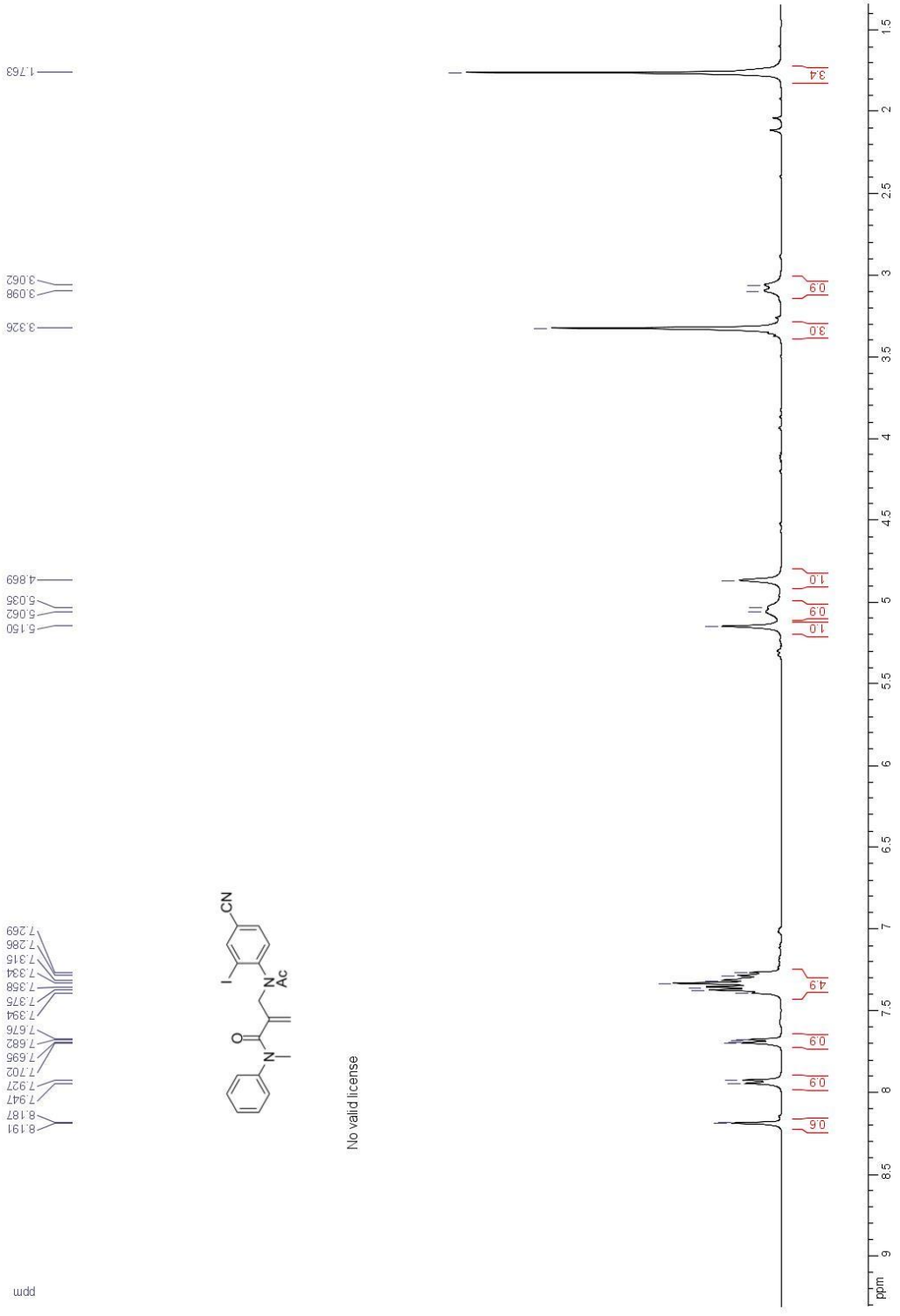
*2-((N-(2-iodo-5-methoxyphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1v***)*



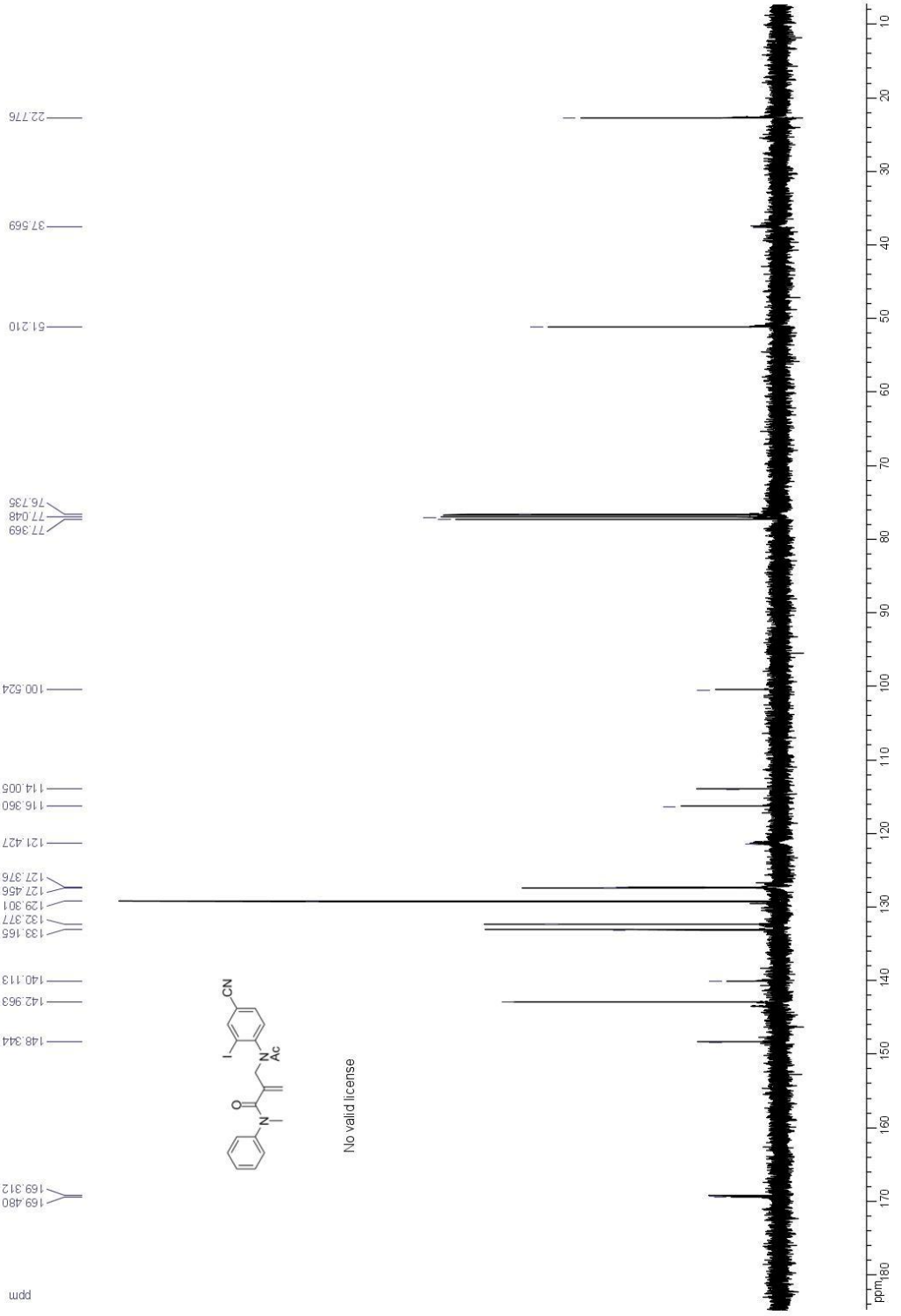
*2-((N-(2-iodo-5-methoxyphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1v***)*



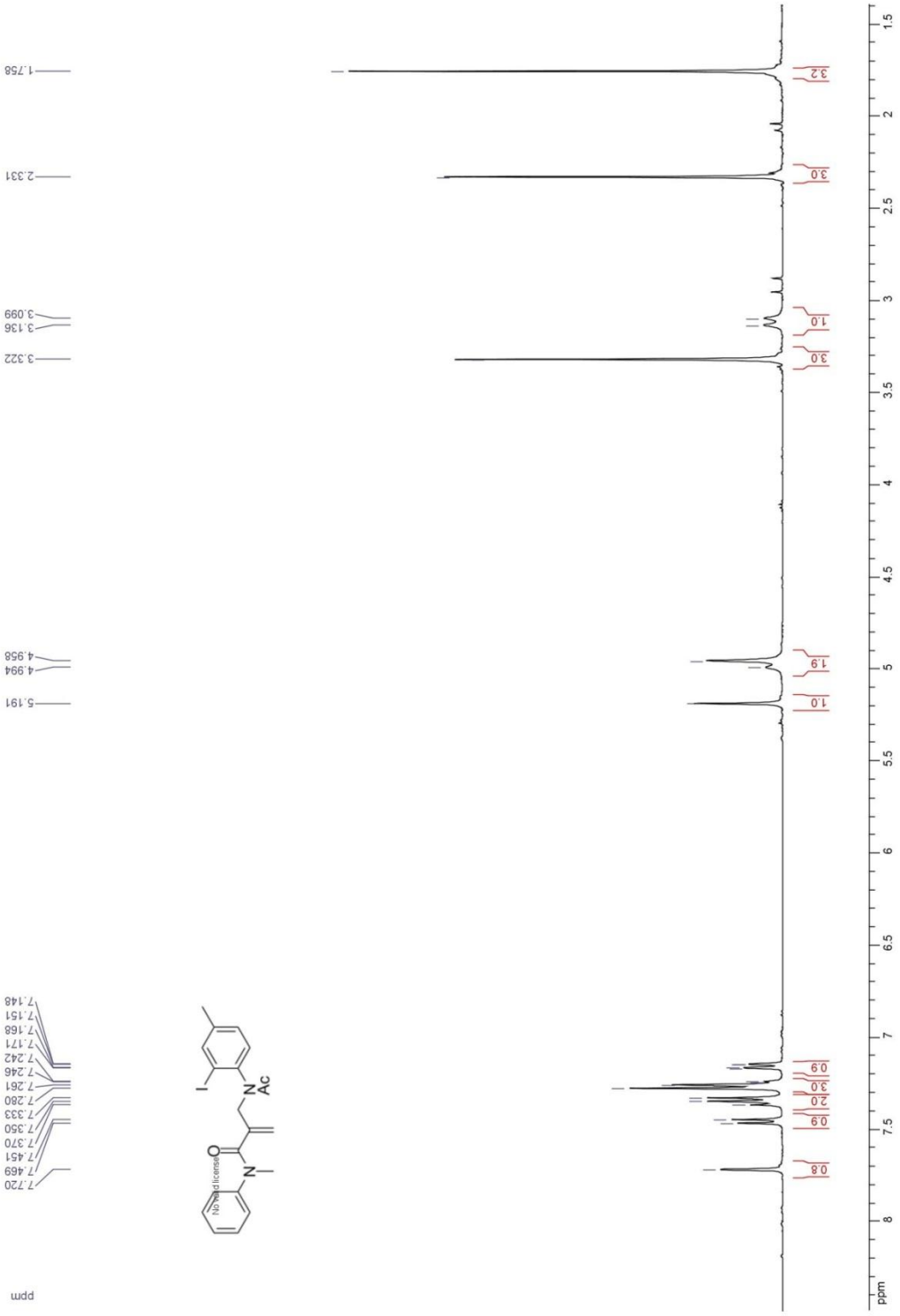
*2-((N-(4-cyano-2-iodophenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1w***)*



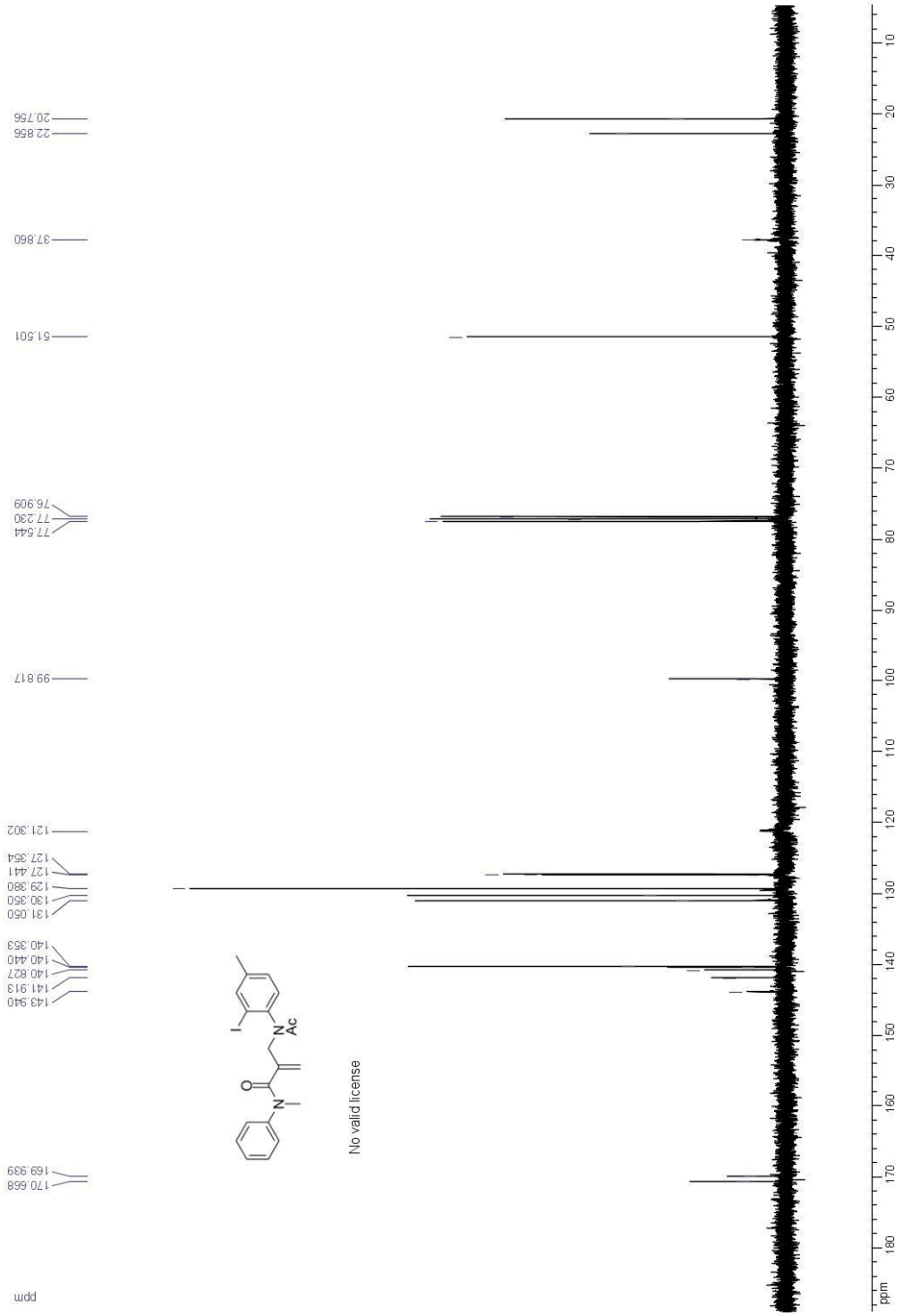
*2-((N-(4-cyano-2-iodophenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1w***)*



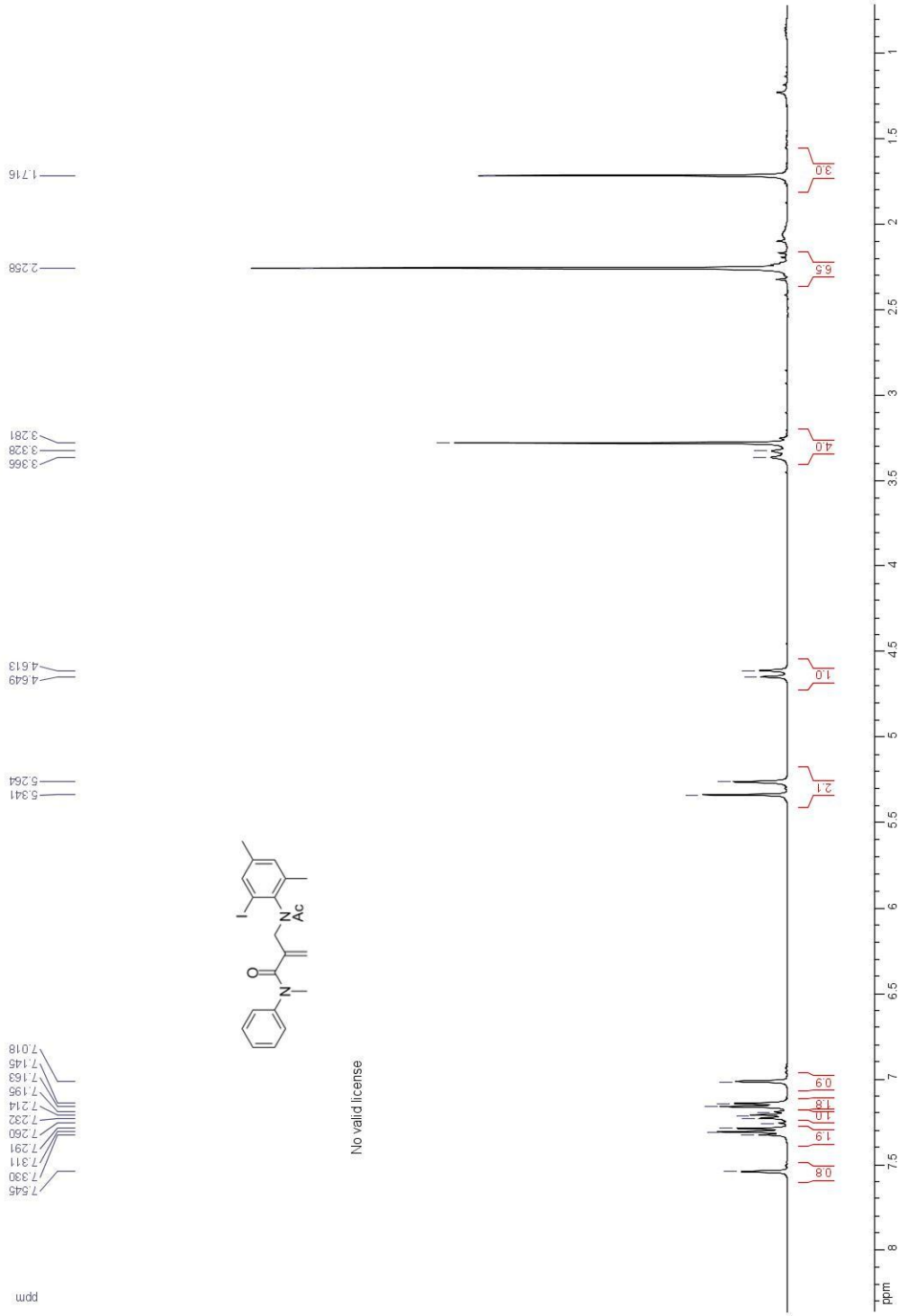
*2-((N-(2-iodo-4-methylphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1x***)*



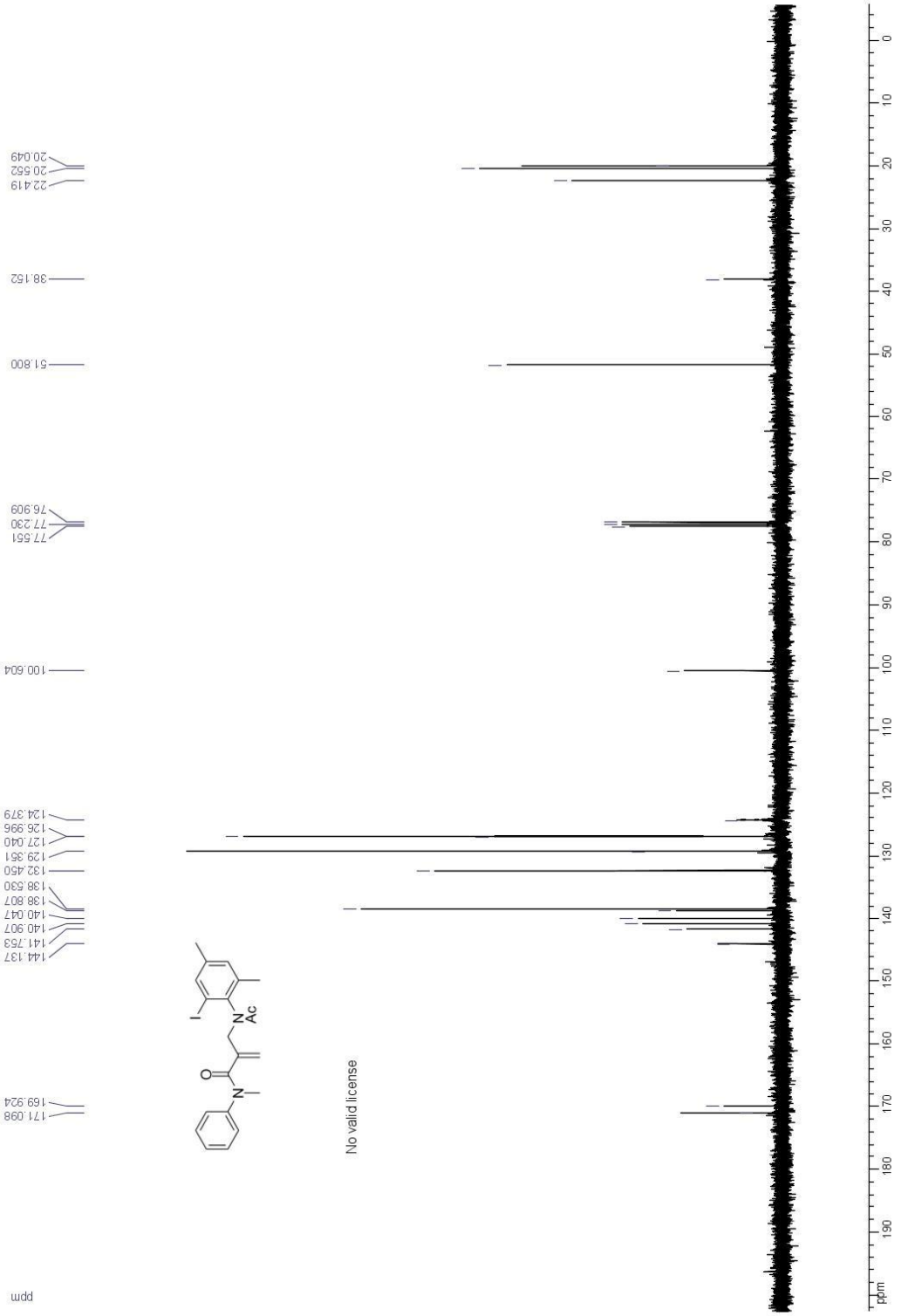
*2-((N-(2-iodo-4-methylphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1x***)*



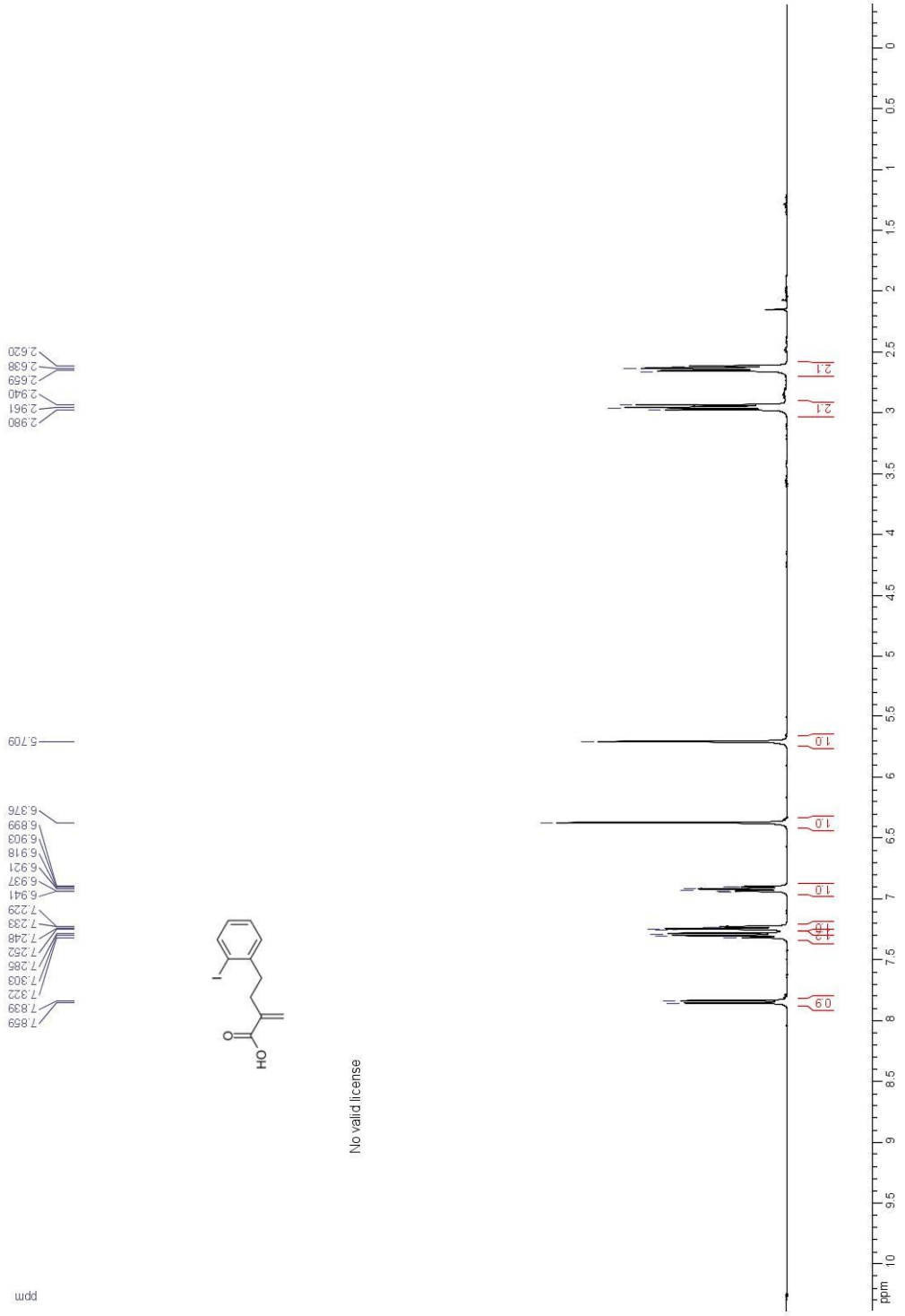
*2-((N-(2-iodo-4,6-dimethylphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1y***)*



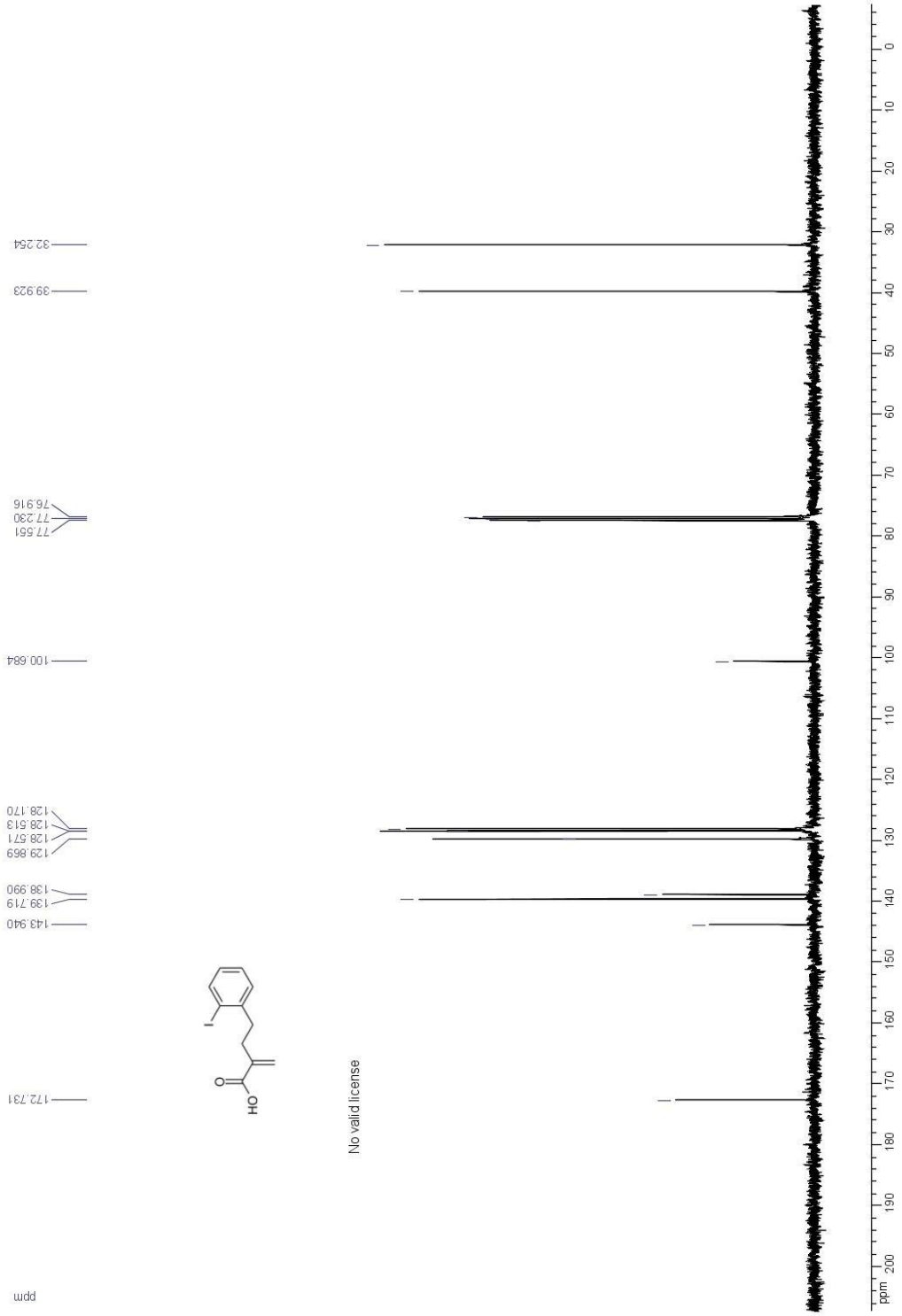
*2-((N-(2-iodo-4,6-dimethylphenyl)acetamido)methyl)-N-methyl-N-phenylacrylamide (***1y***)*



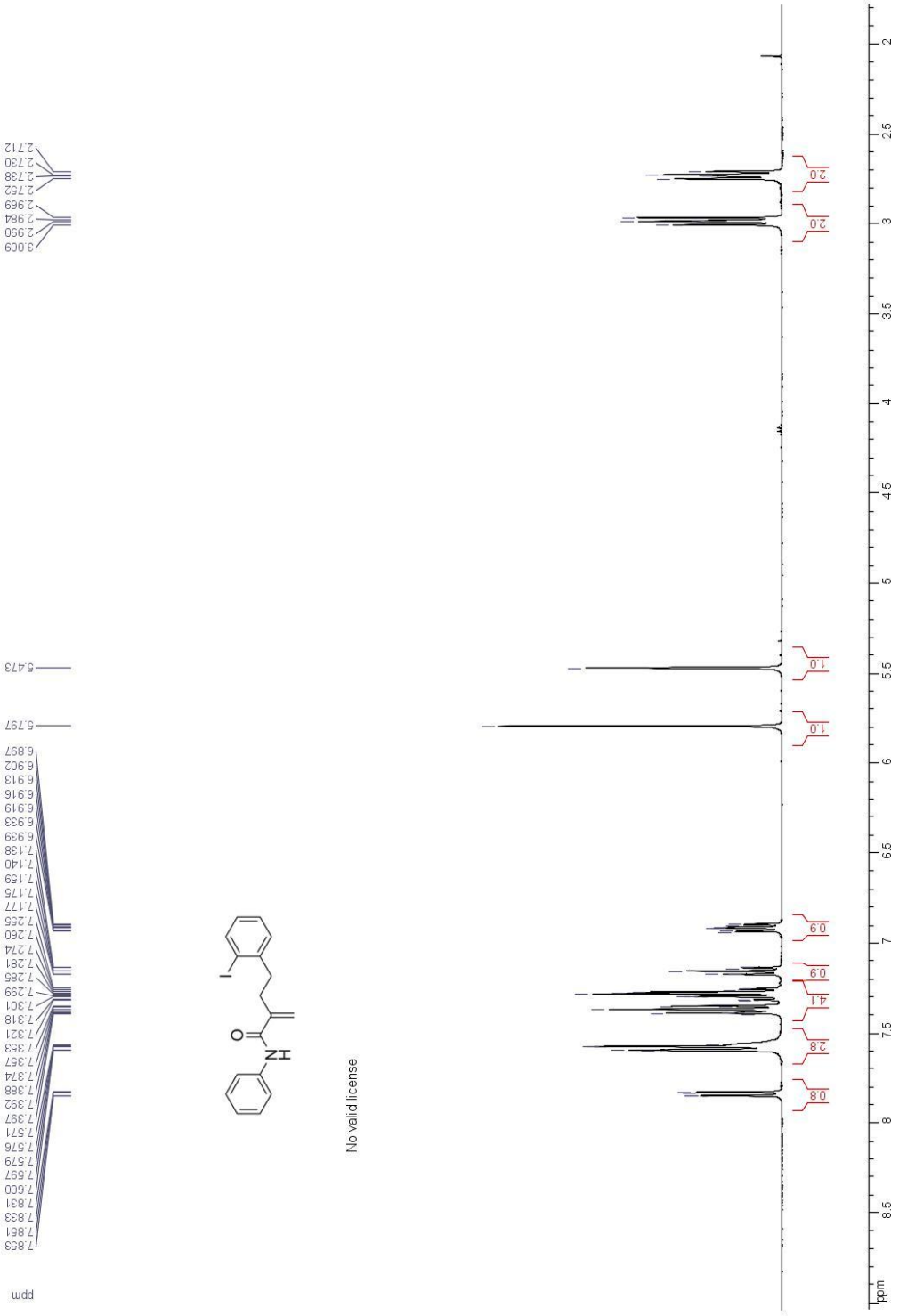
4-(2-Iodophenyl)-2-methylenebutanoic acid (**9**)



4-(2-Iodophenyl)-2-methylenebutanoic acid (**9**)

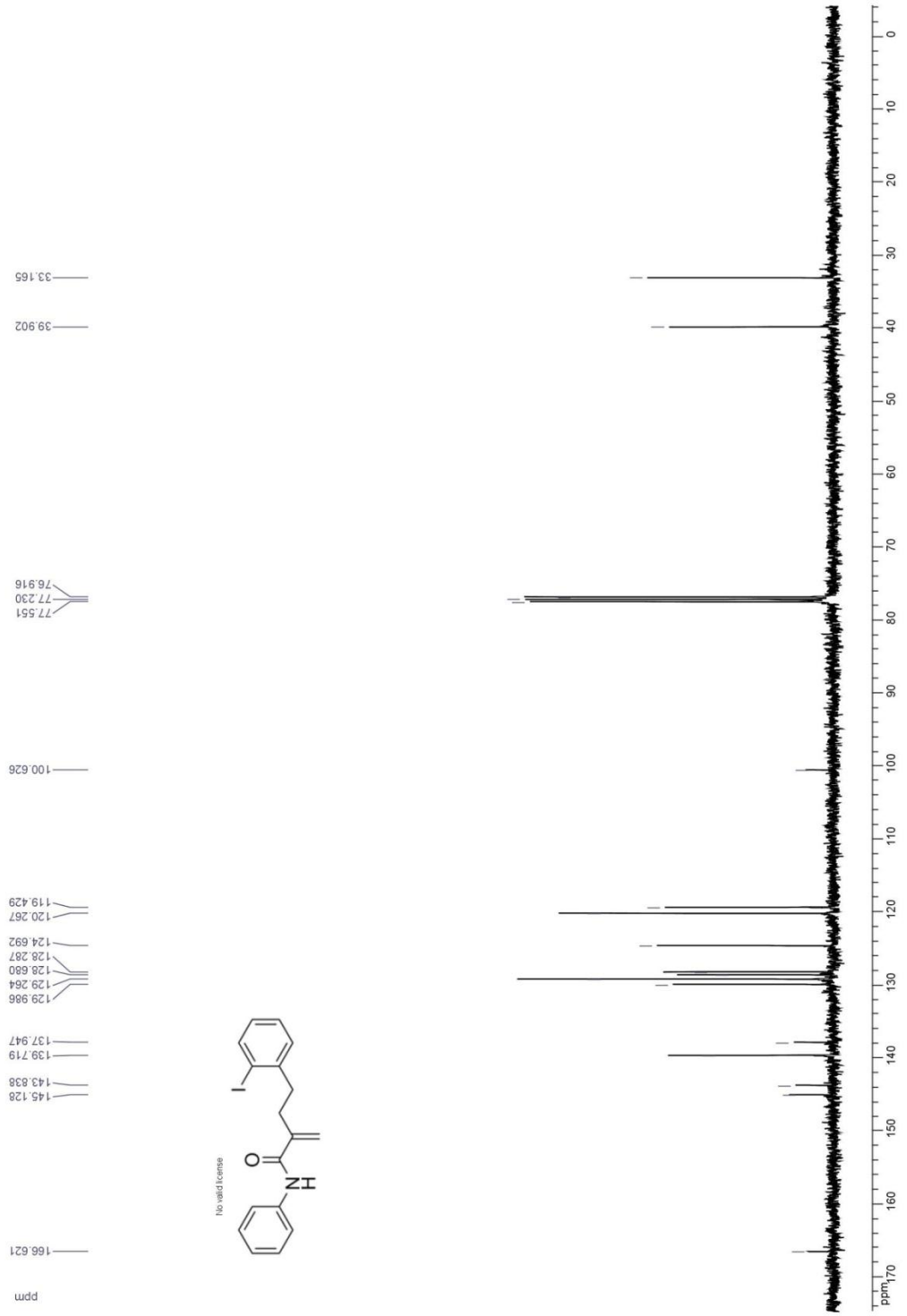


*4-(2-Iodophenyl)-2-methylene-N-phenylbutanamide (***1b***)*

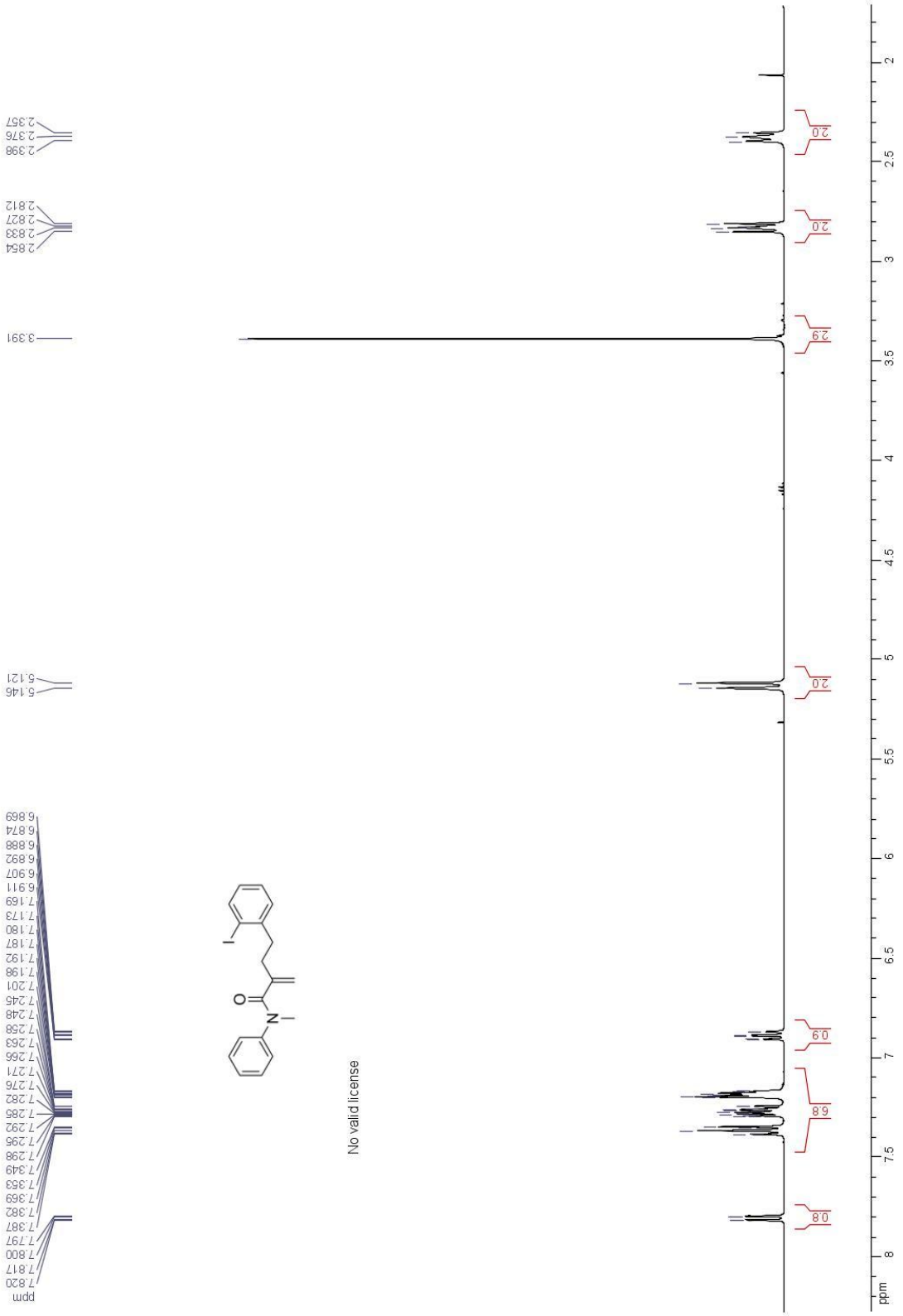


*4-(2-Iodophenyl)-2-methylene-N-phenylbutanamide (***1b***)*

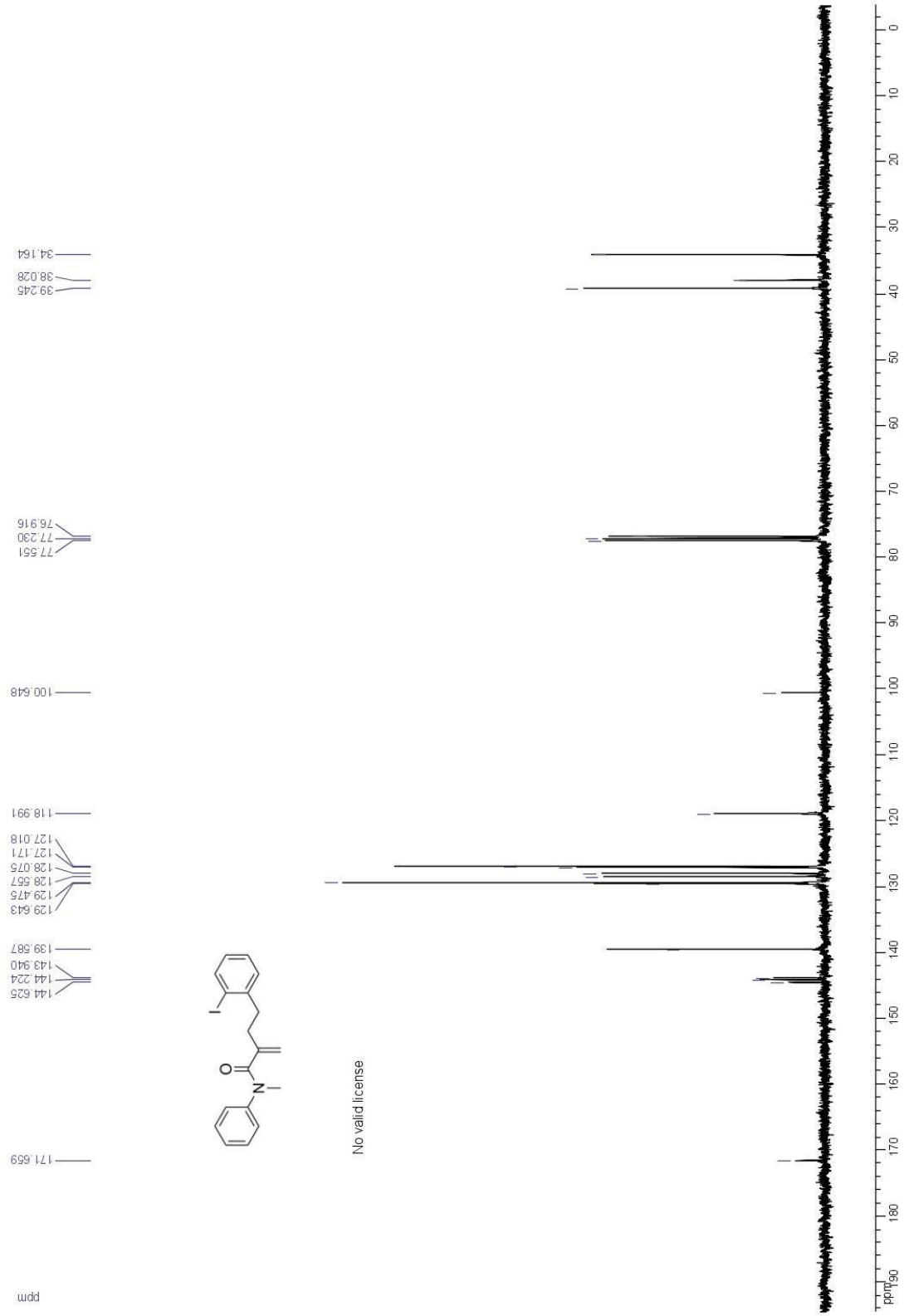
*4-(2-Iodophenyl)-2-methylene-N-phenylbutanamide (***1b***)*



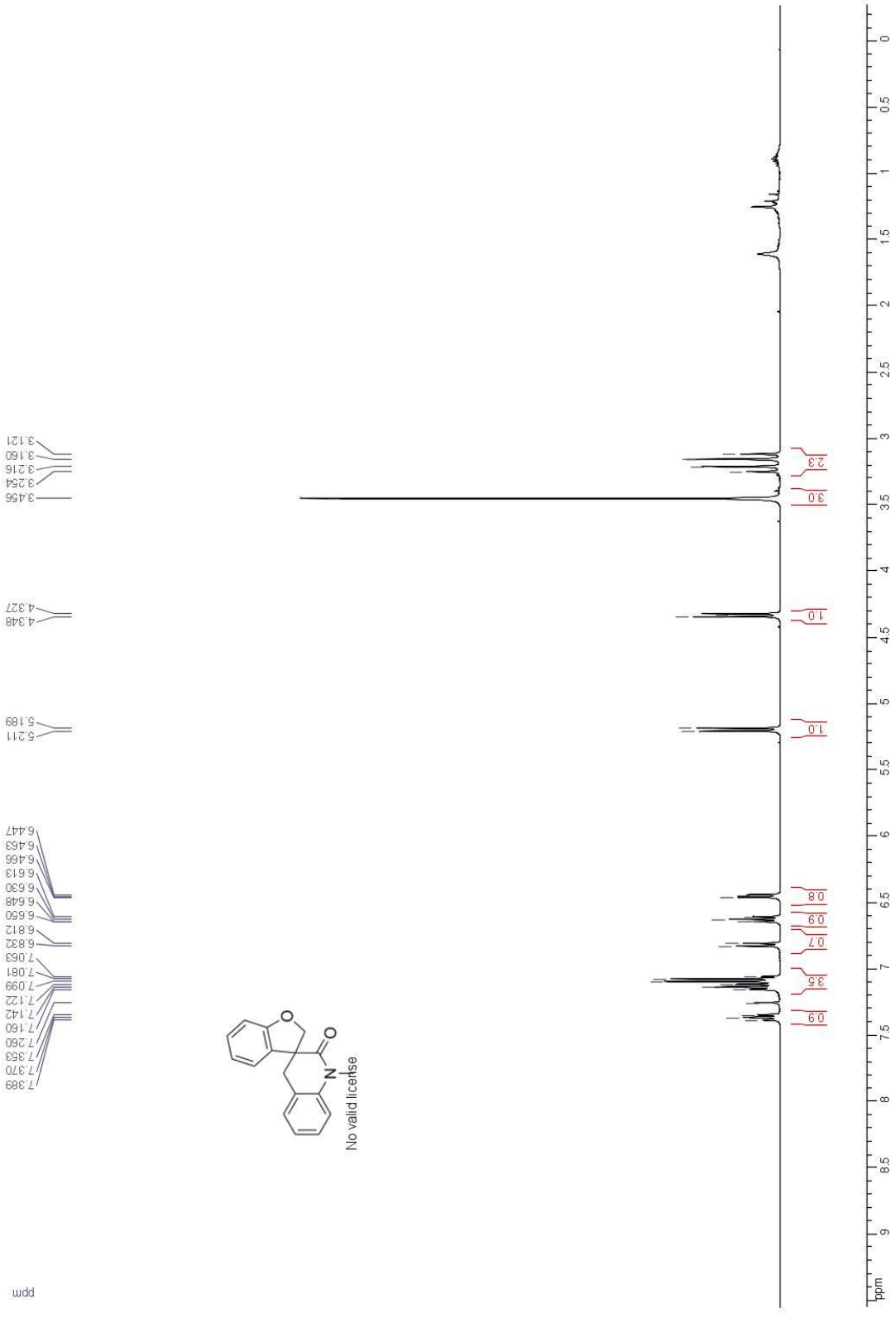
*4-(2-Iodophenyl)-N-methyl-2-methylene-N-phenylbutanamide (***1s***)*



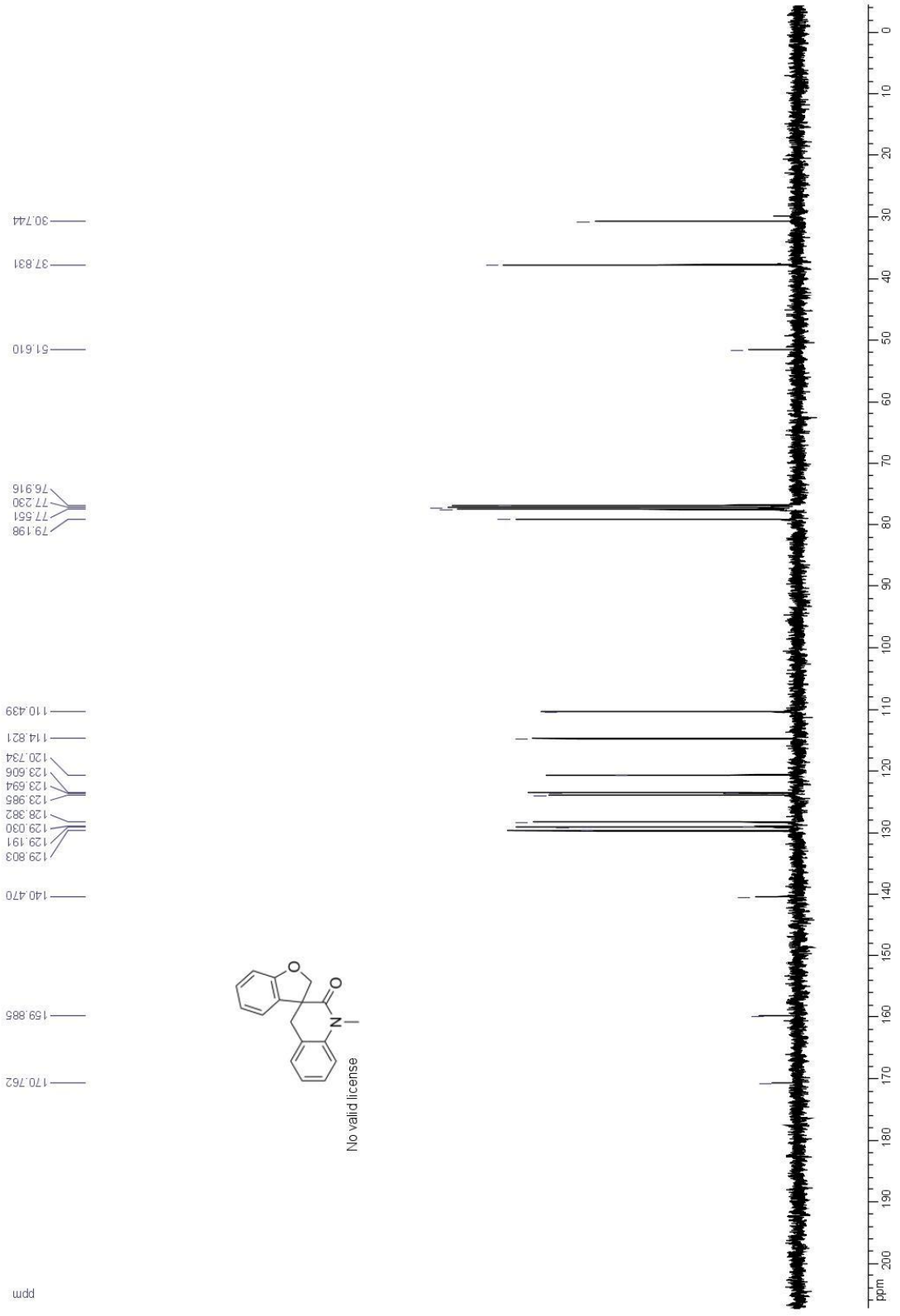
*4-(2-Iodophenyl)-N-methyl-2-methylene-N-phenylbutanamide (***1s***)*



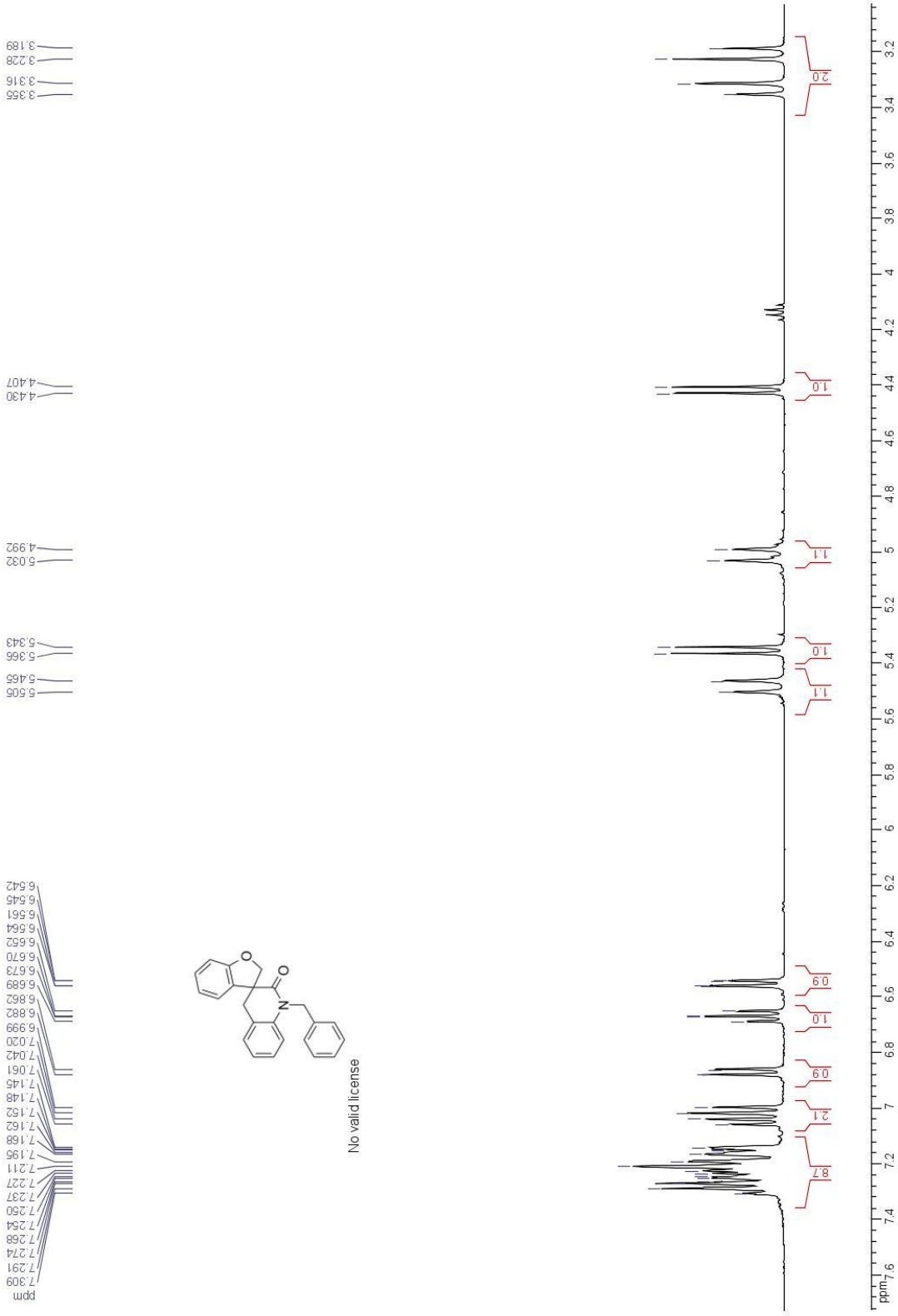
*1'-Methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2a***)*



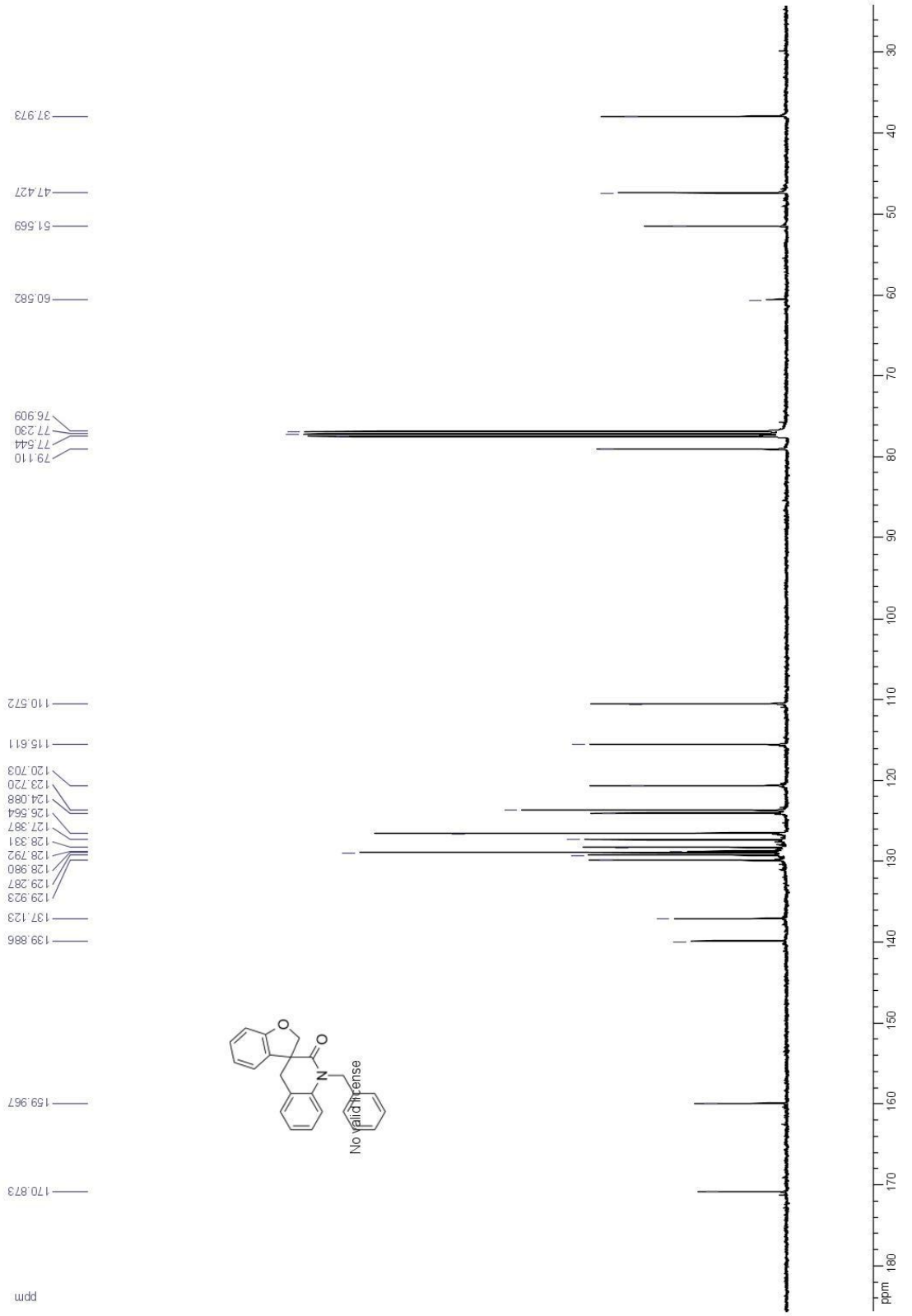
*1'-Methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2a***)*



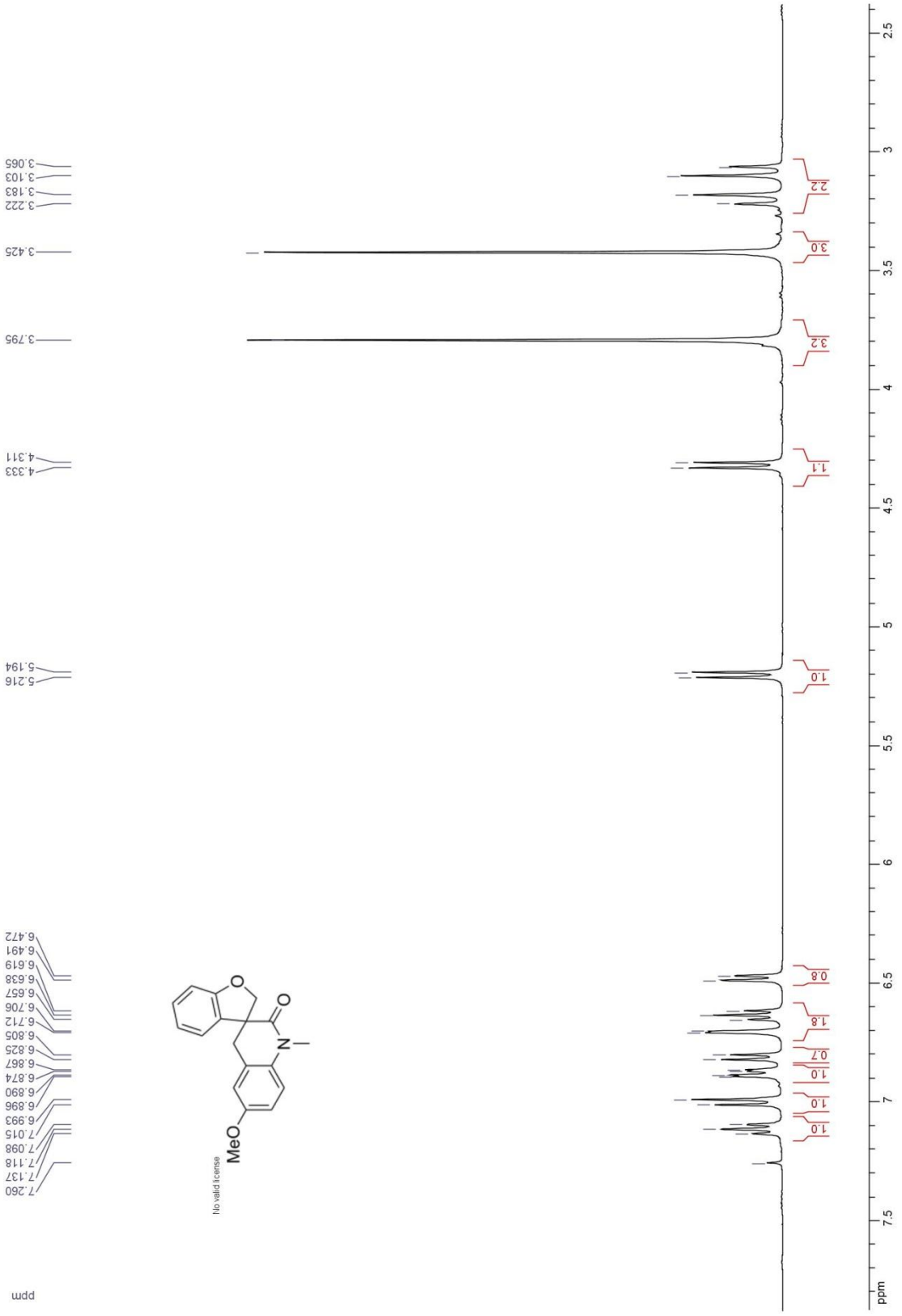
*1'-benzyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2c***)*



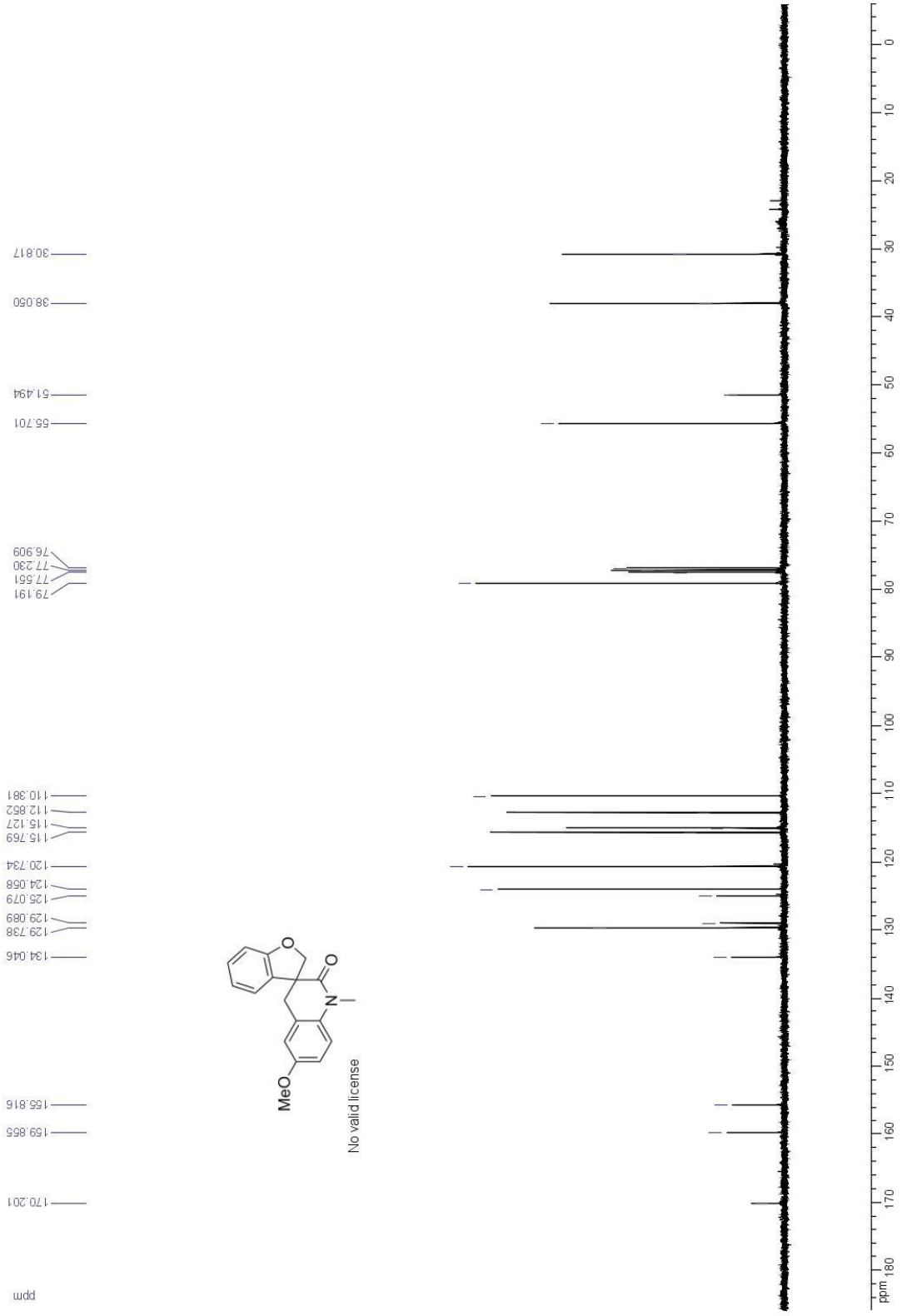
*1'-benzyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2c***)*



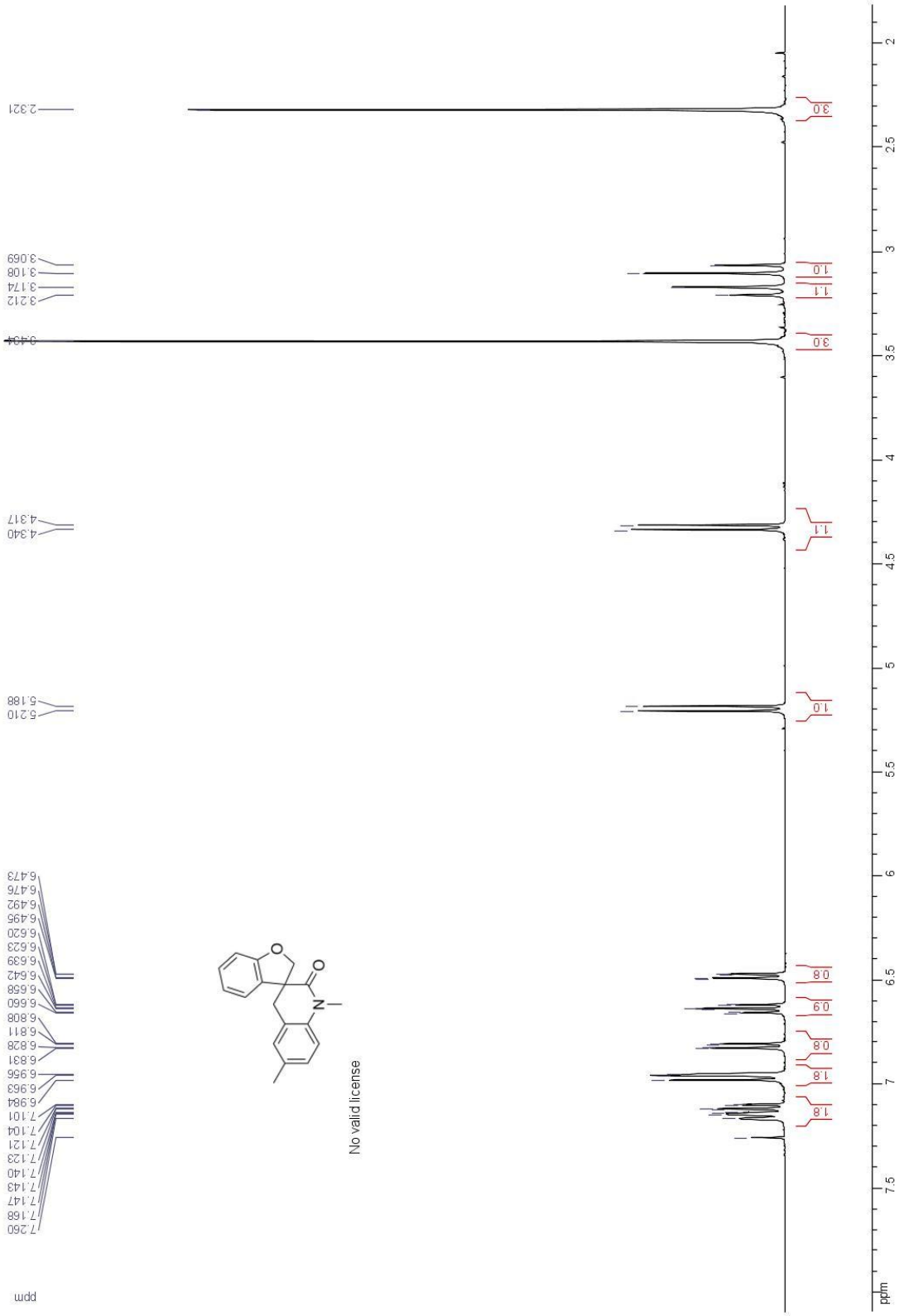
*6'-methoxy-1'-methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2d***)*



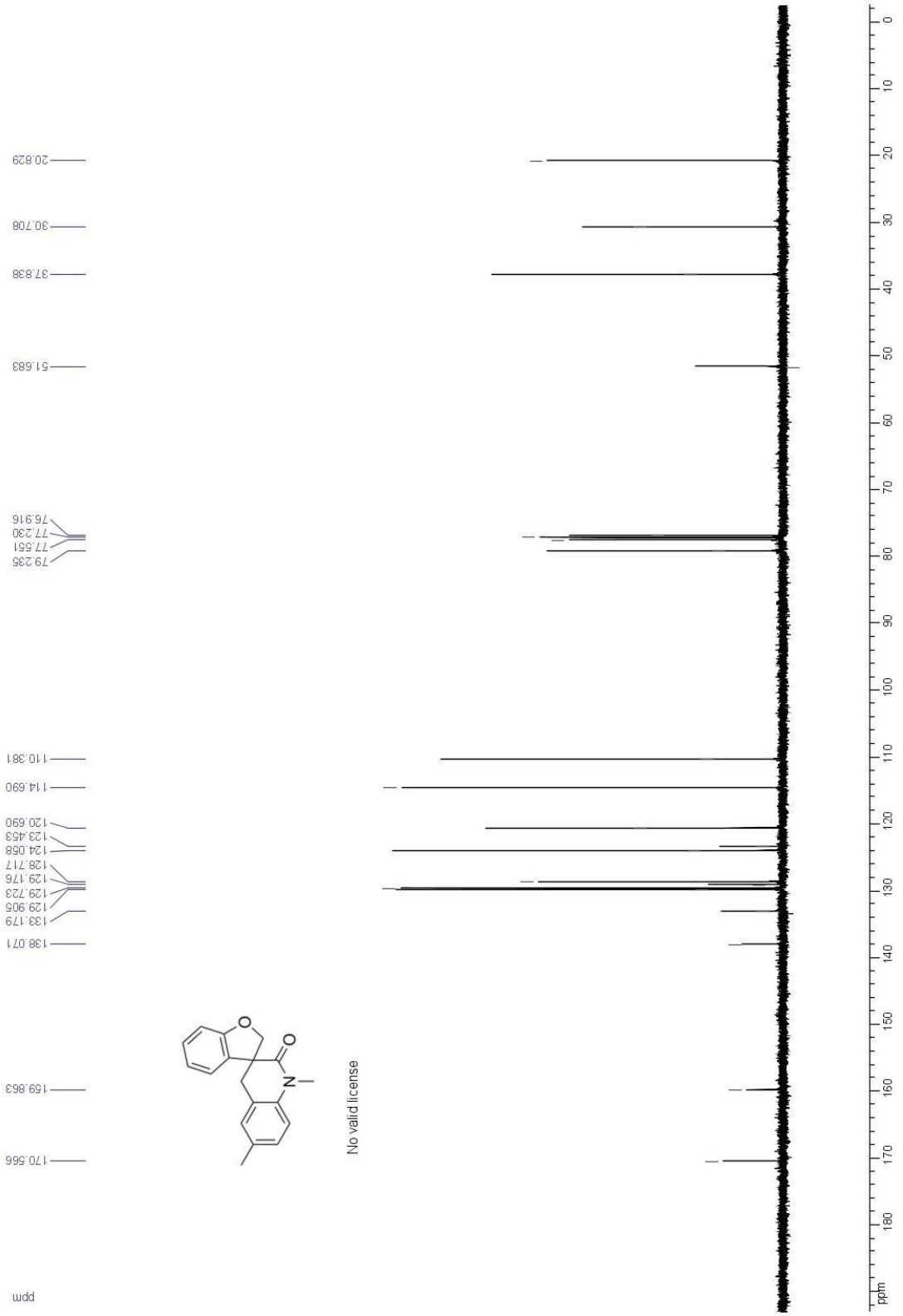
*6'-methoxy-1'-methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2d***)*



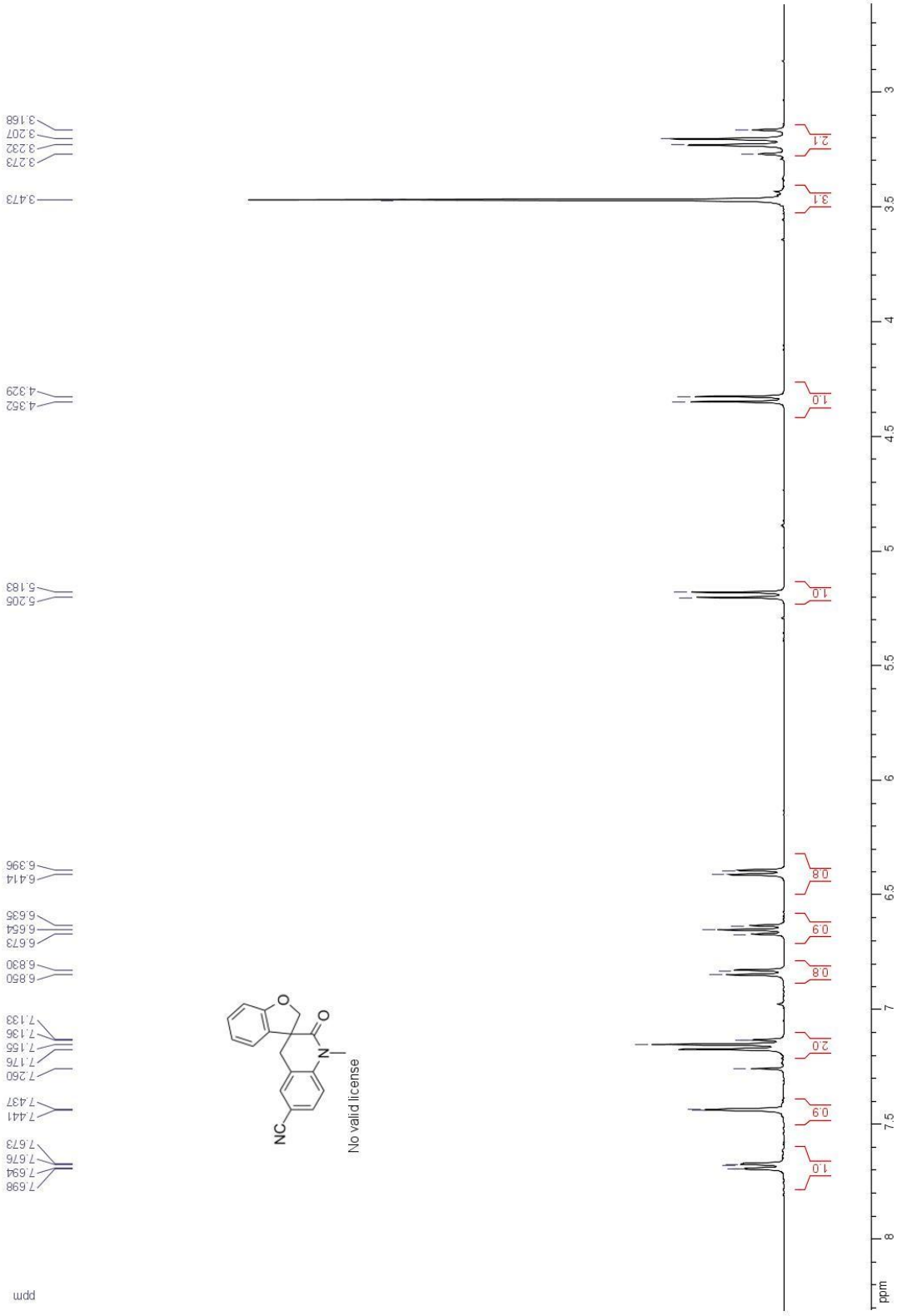
*1',6'-Dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2e***)*



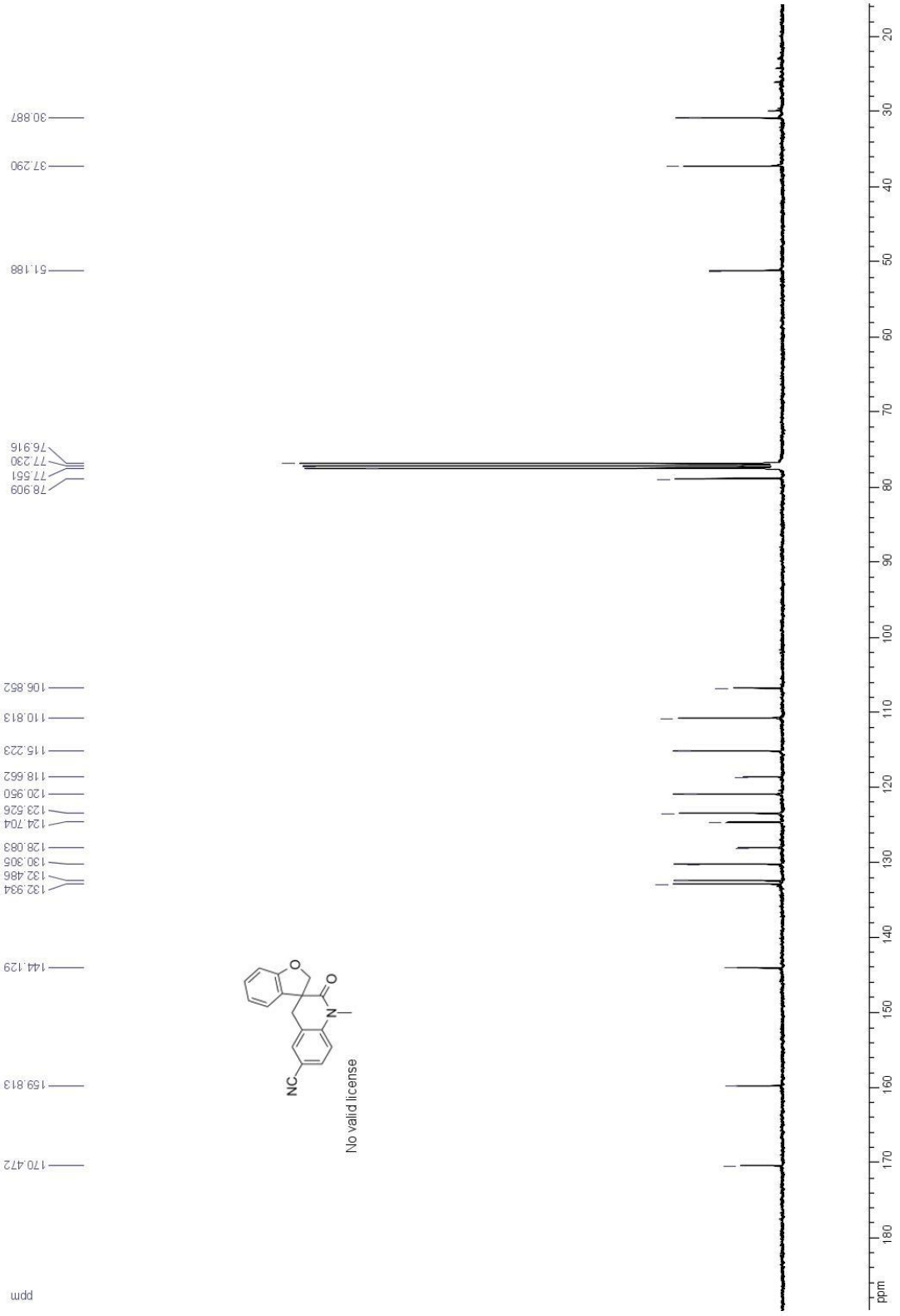
*1',6'-Dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2e***)*



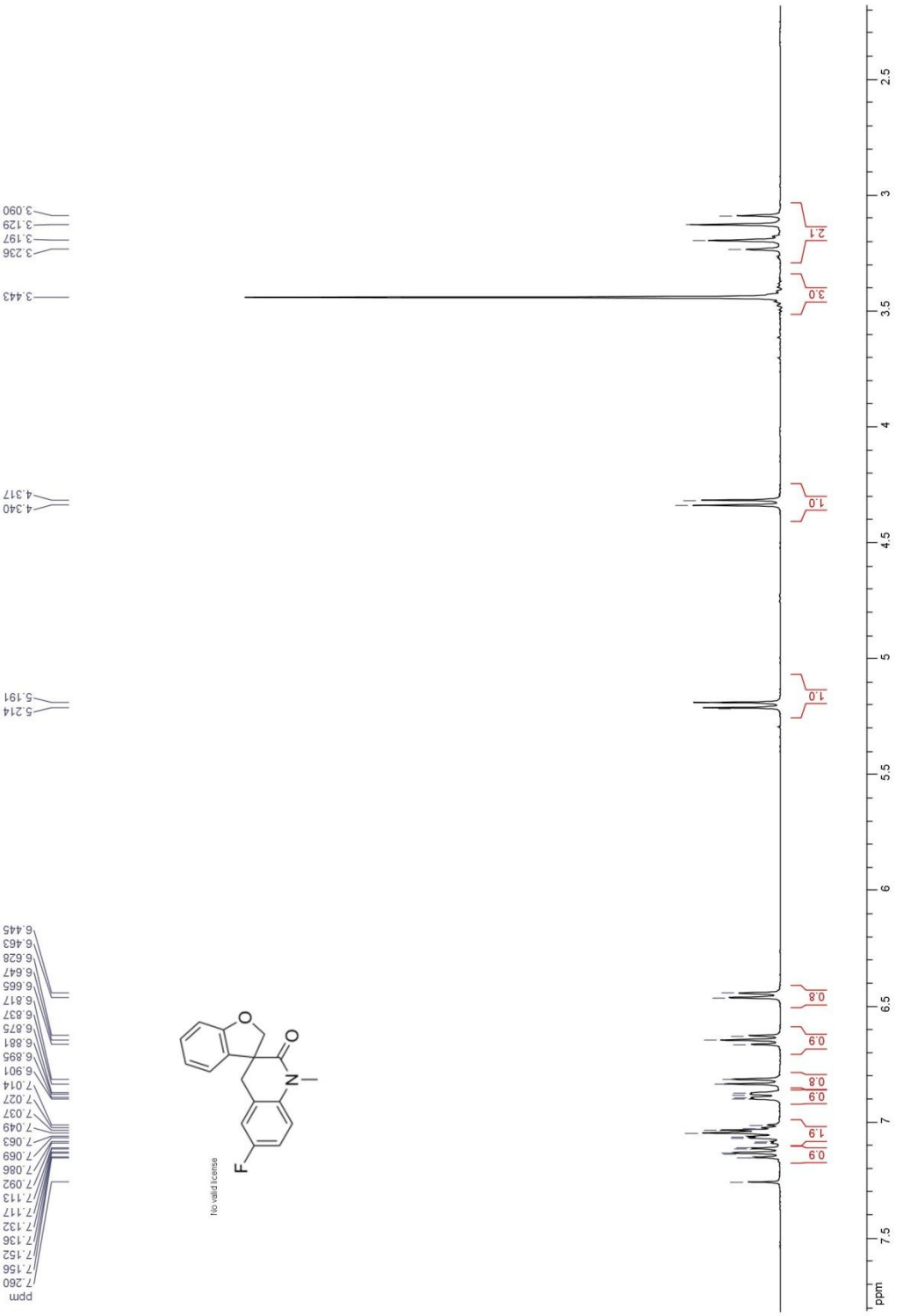
*1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-6'-carbonitrile (***2f***)*



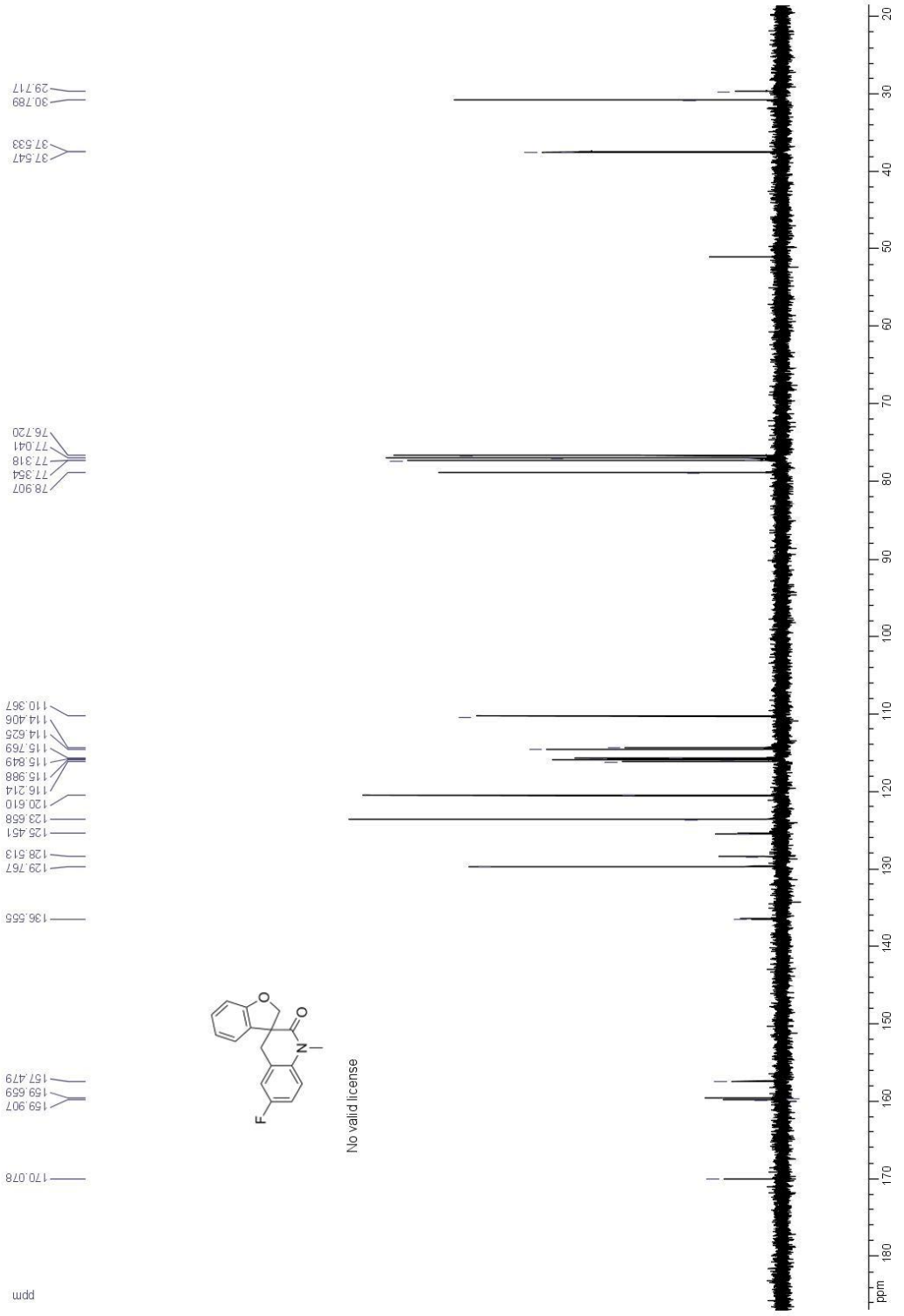
*1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-6'-carbonitrile (***2f***)*



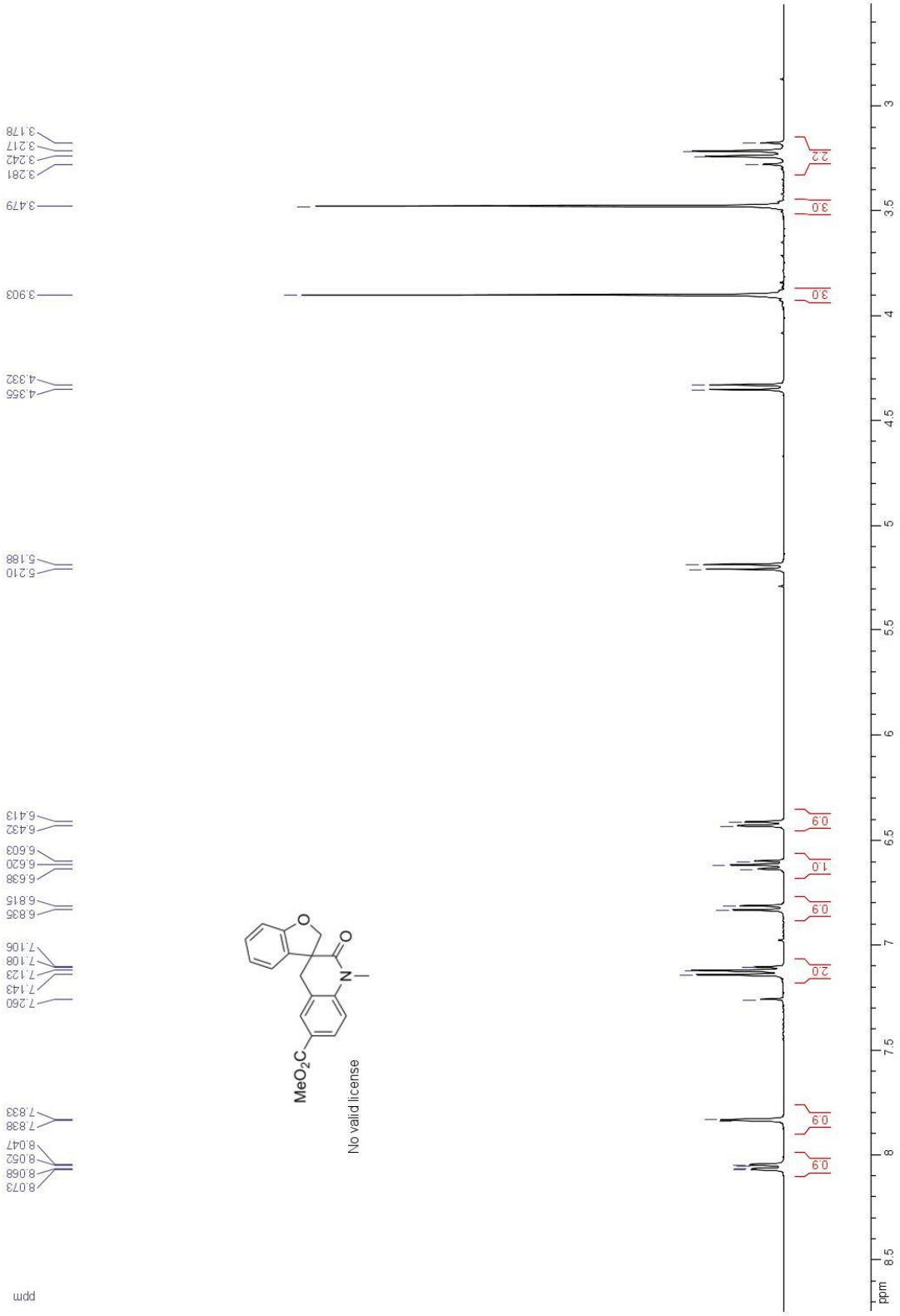
*1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-6'-carbonitrile (***2f***)*



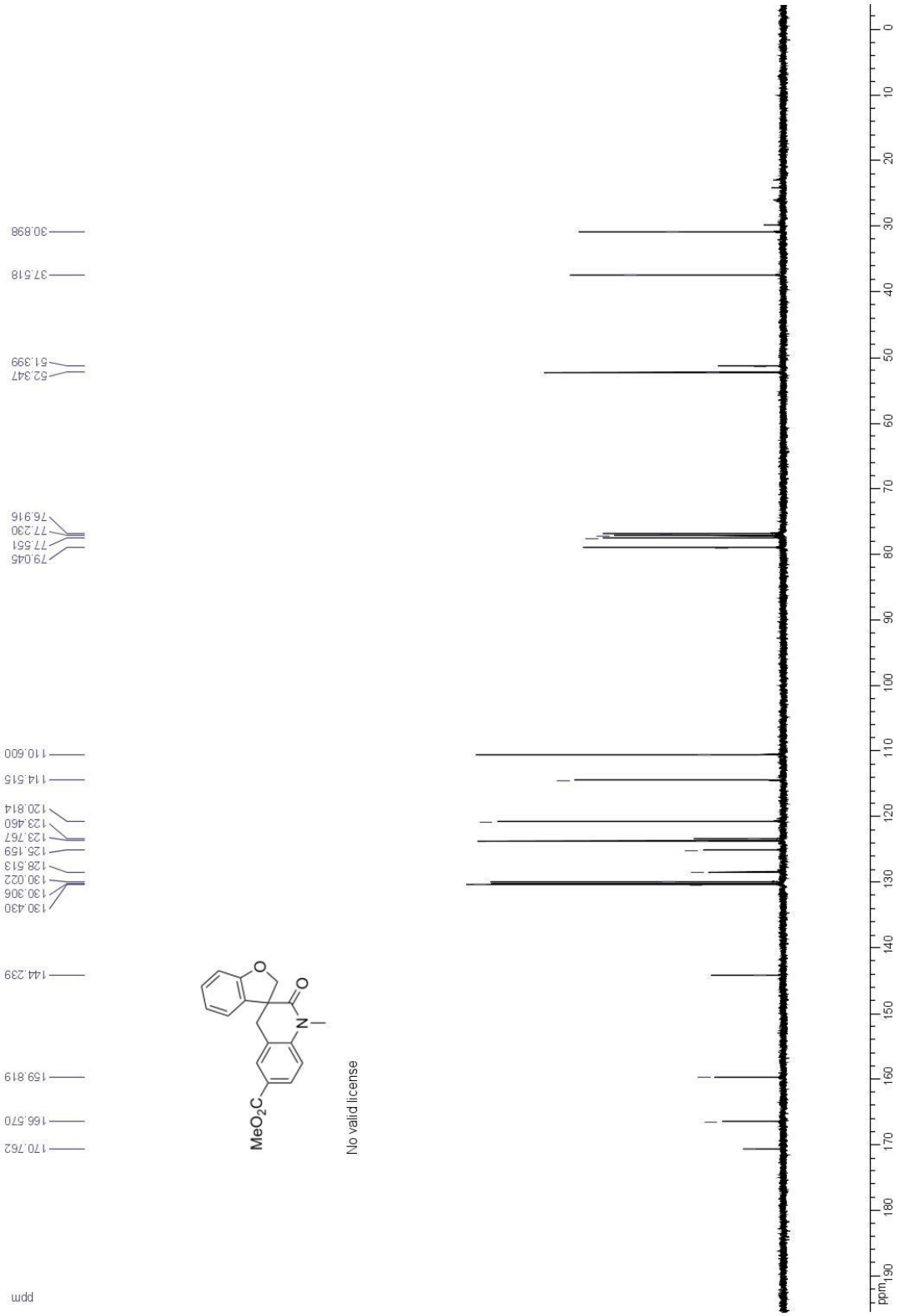
*1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-6'-carbonitrile (***2f***)*



*Methyl 1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-6'-carboxylate (***2h***)*

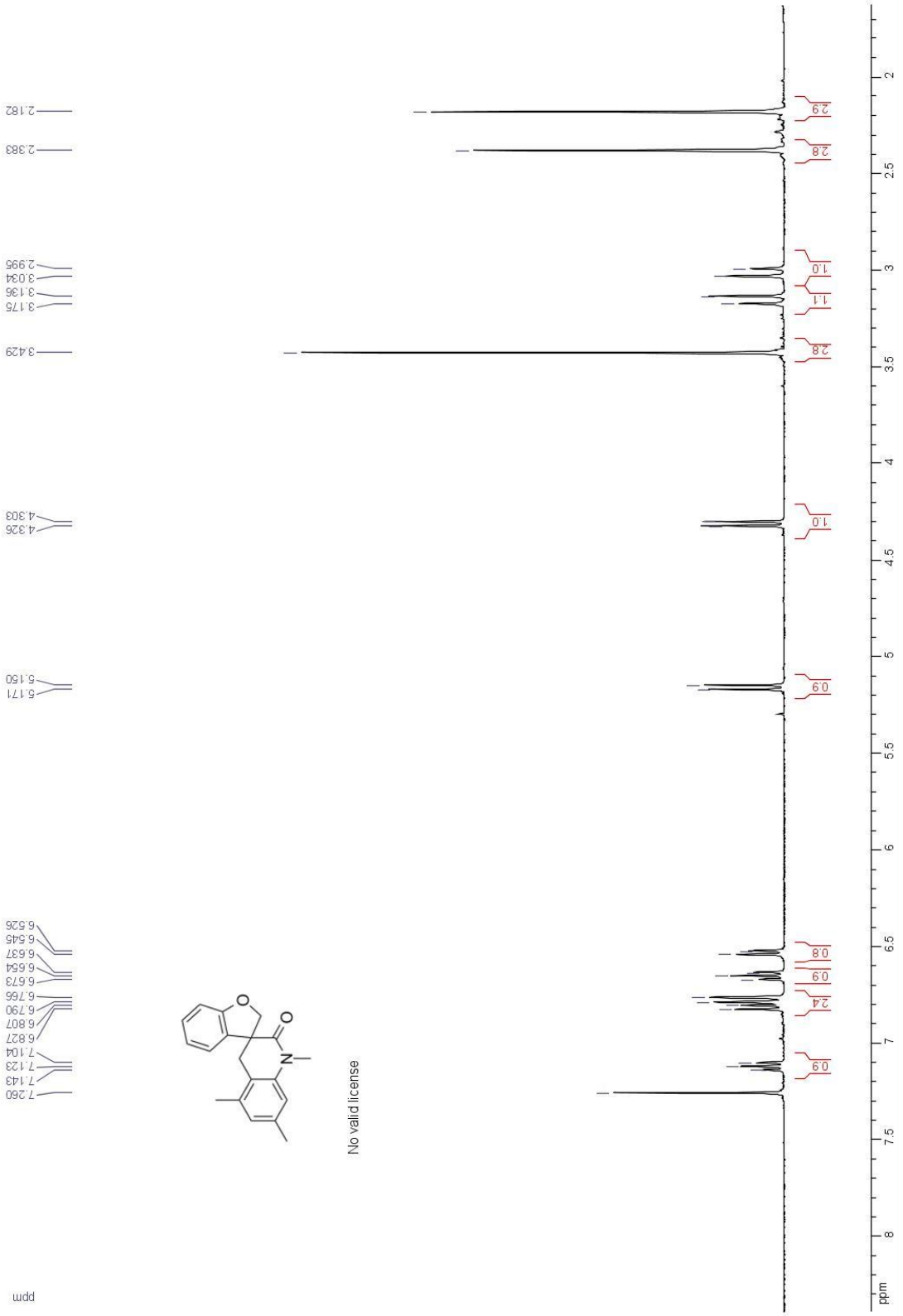


*Methyl 1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-6'-carboxylate (**2h**)*

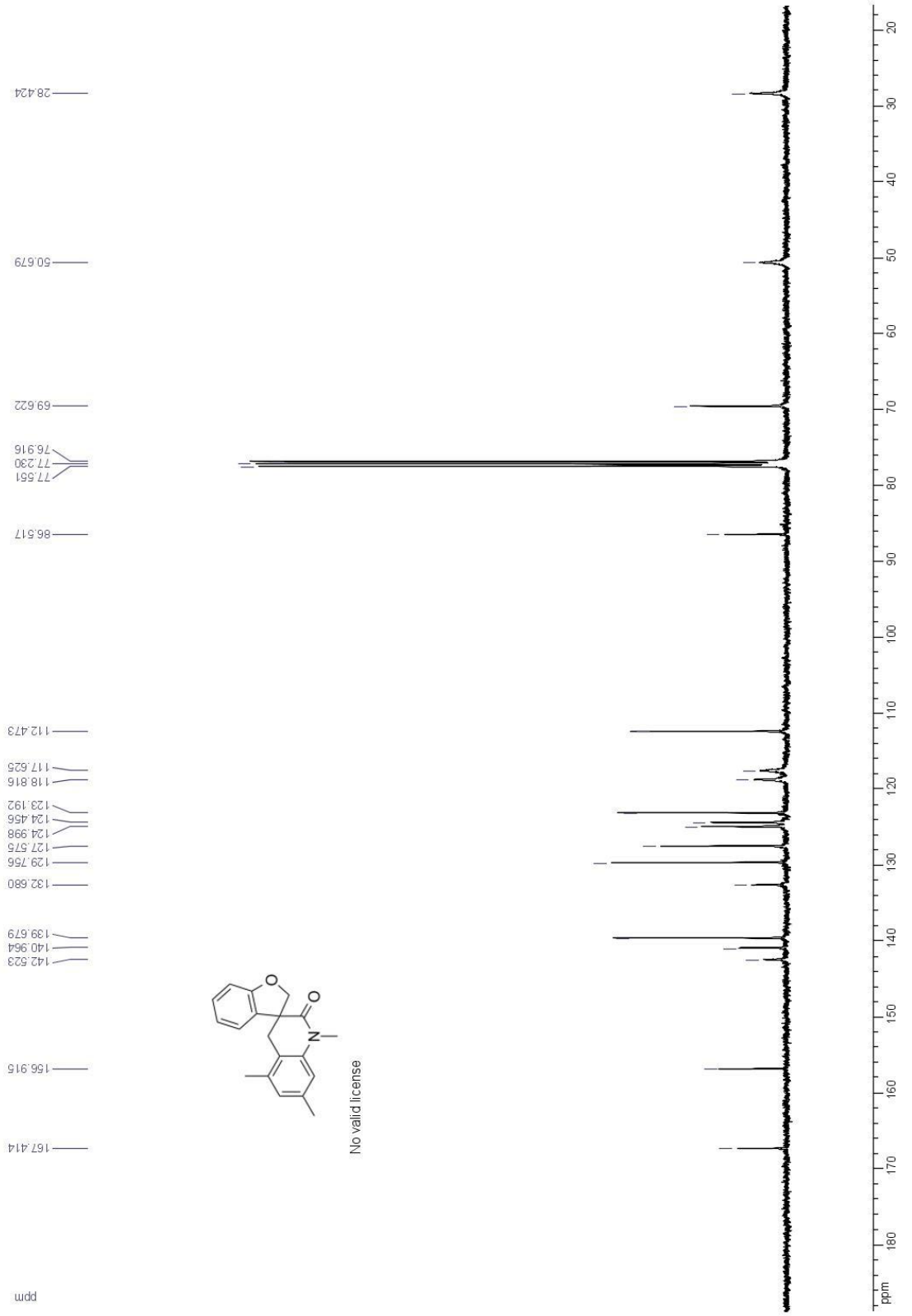


*1',5',7'-trimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (**2i**)*

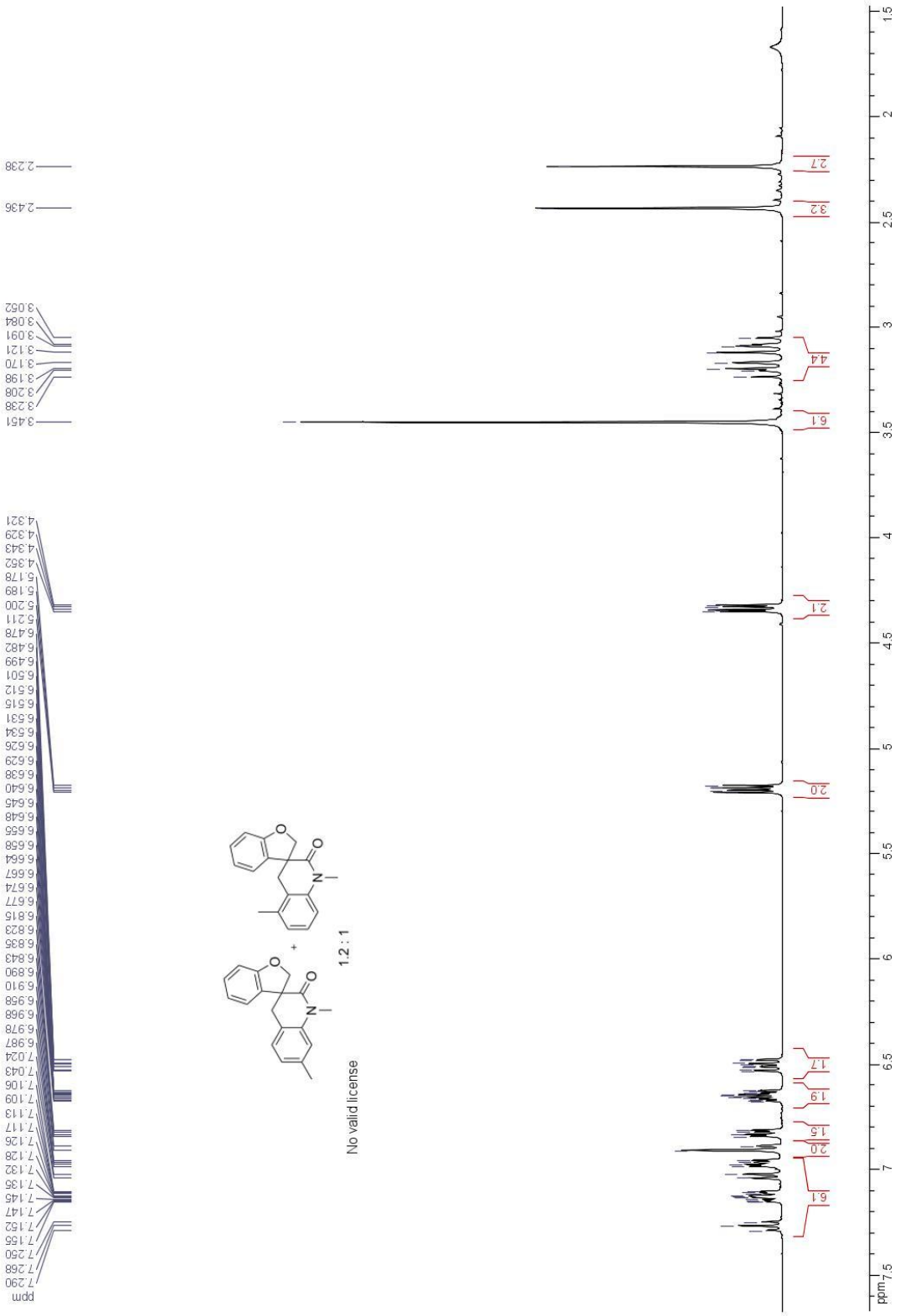
*1',5',7'-trimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (**2i**)*



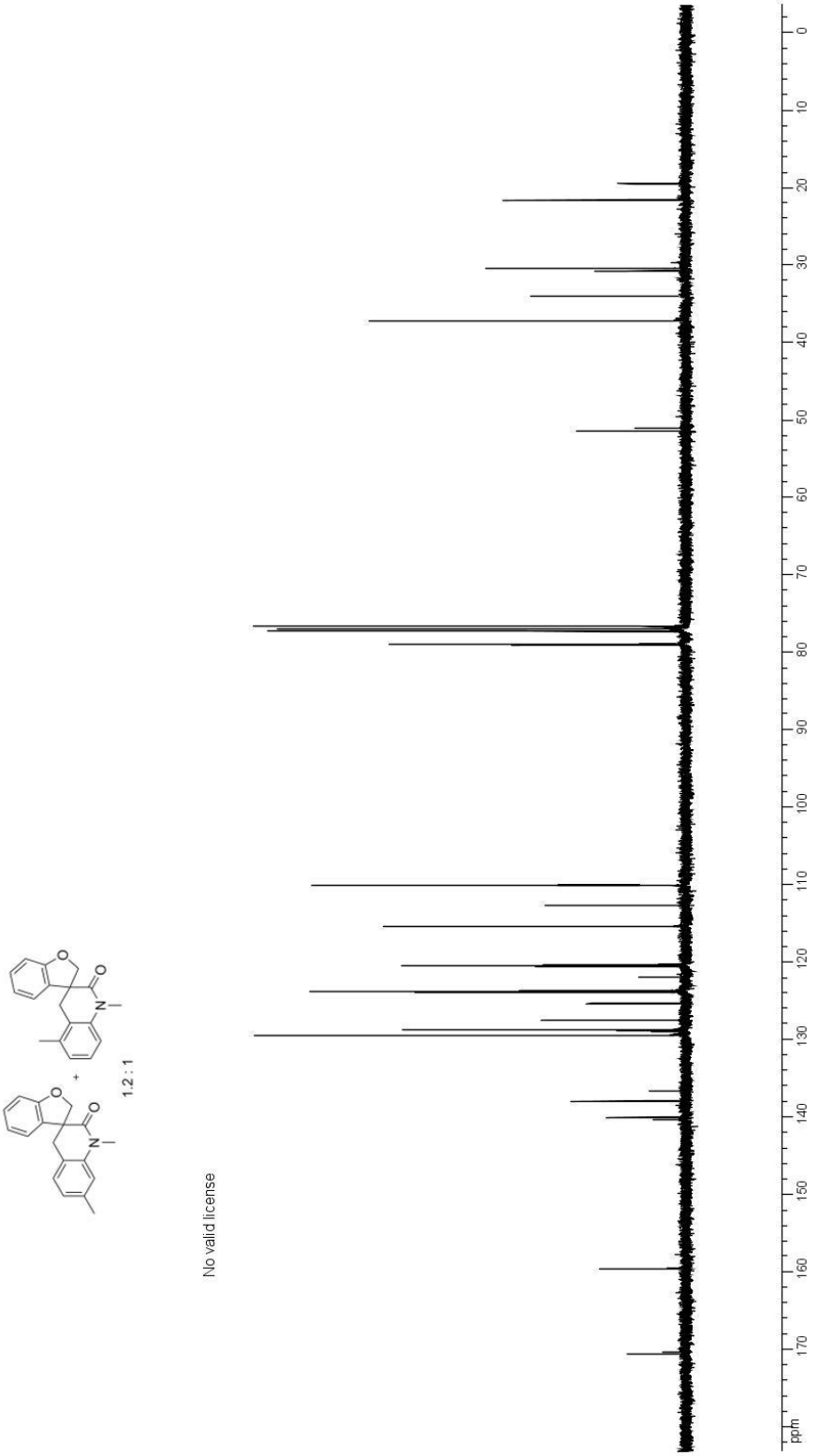
*1',5',7'-trimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (**2i**)*



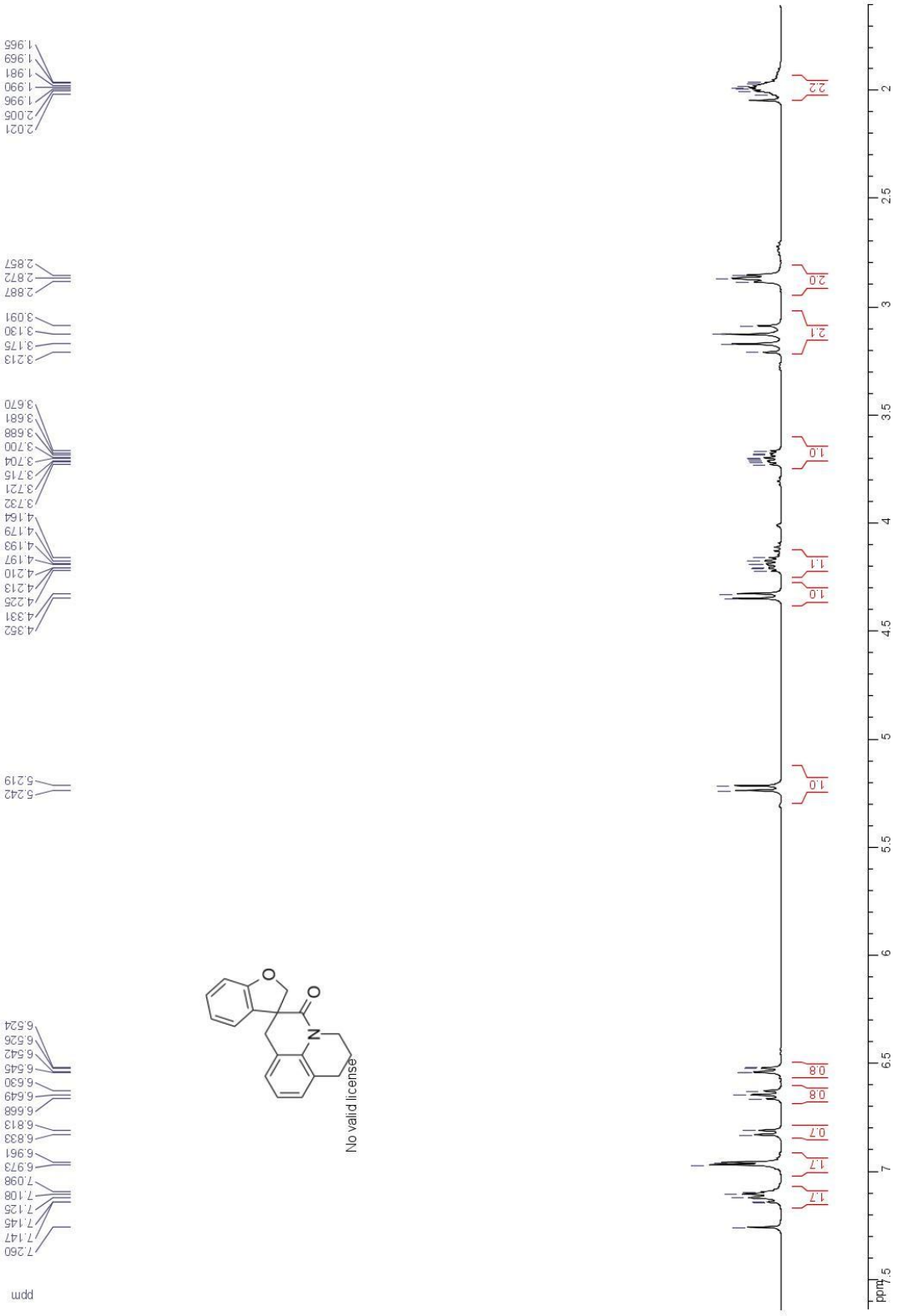
*1',7'-Dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one and 1',5'-dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2j** and **2j'***)*



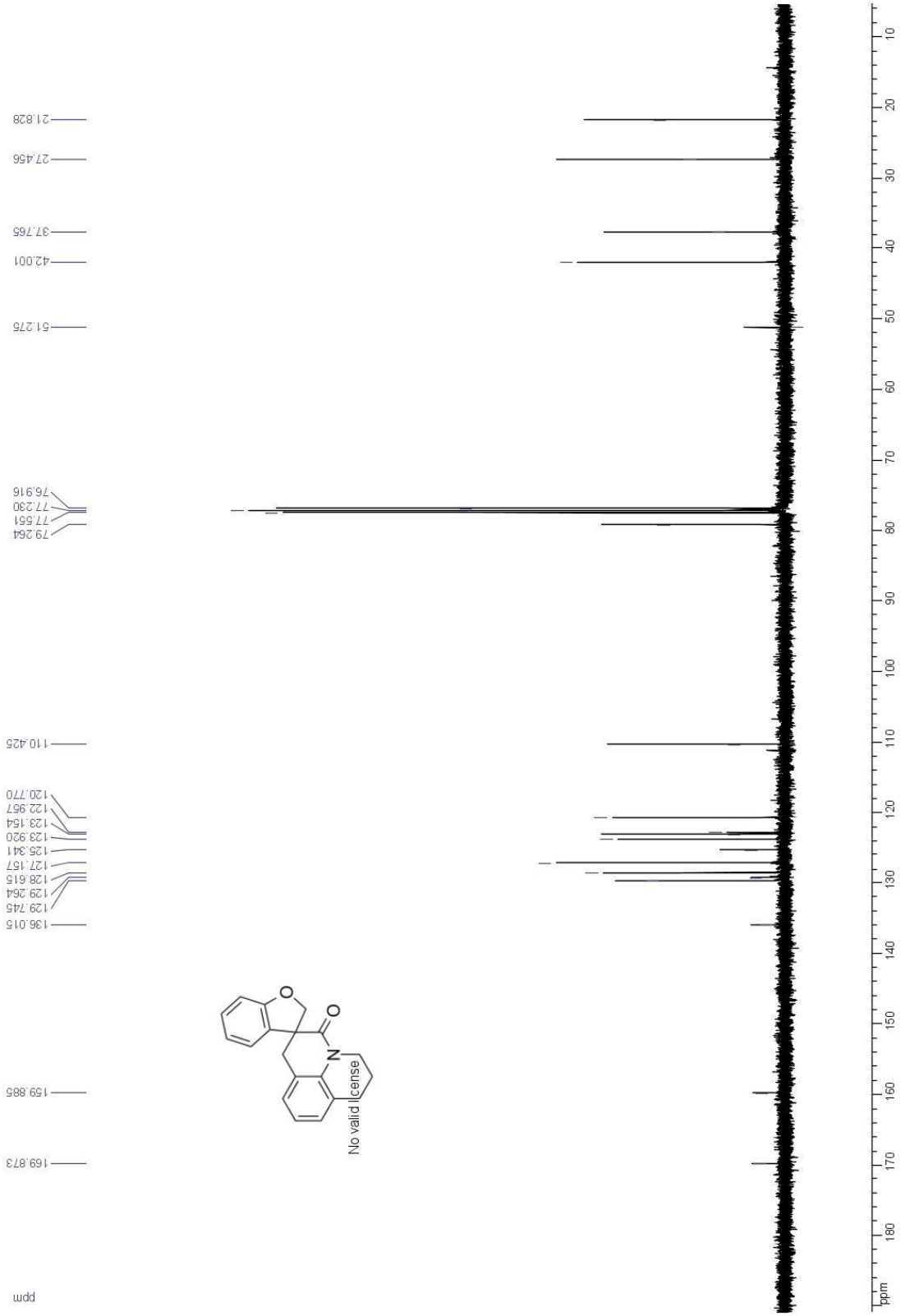
*1',7'-Dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one and 1',5'-dimethyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2j** and **2j'***)*



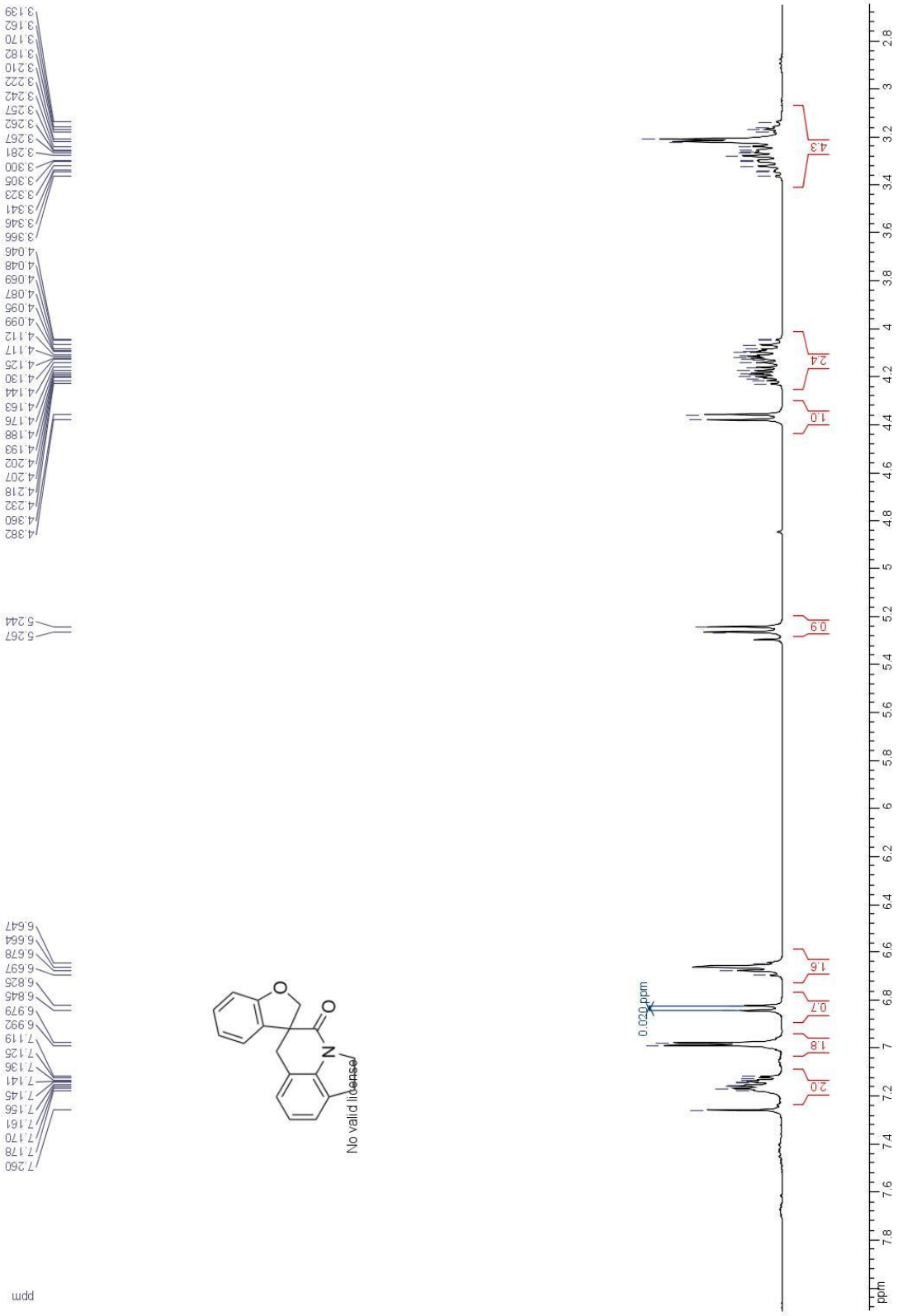
*6',7'-dihydro-1'H,2H-spiro[benzofuran-3,2'-pyrido[3,2,1-ij]quinolin]-3'(5'H)-one (***2k***)*



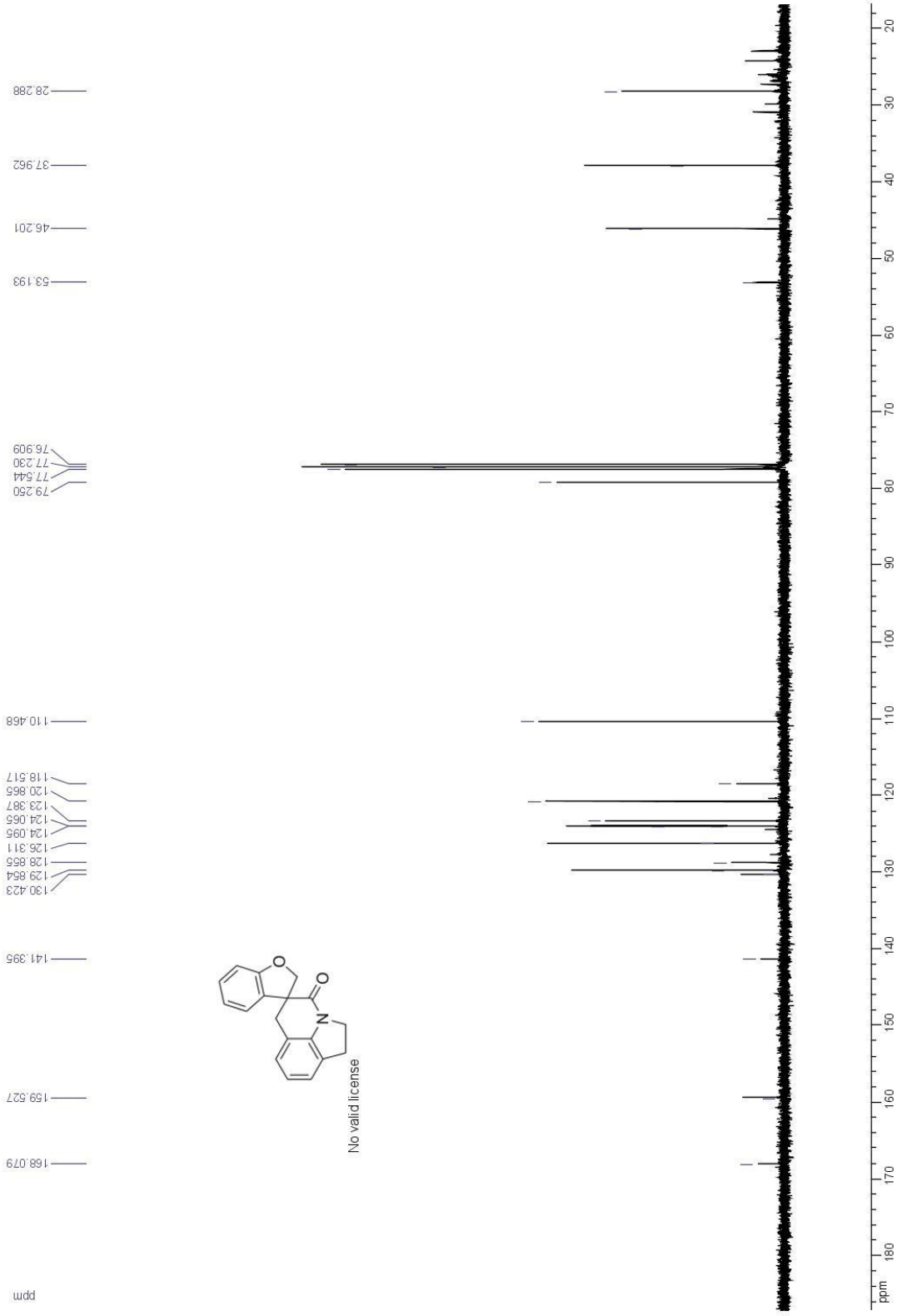
*6',7'-dihydro-1'H,2H-spiro[benzofuran-3,2'-pyrido[3,2,1-ij]quinolin]-3'(5'H)-one (***2k***)*



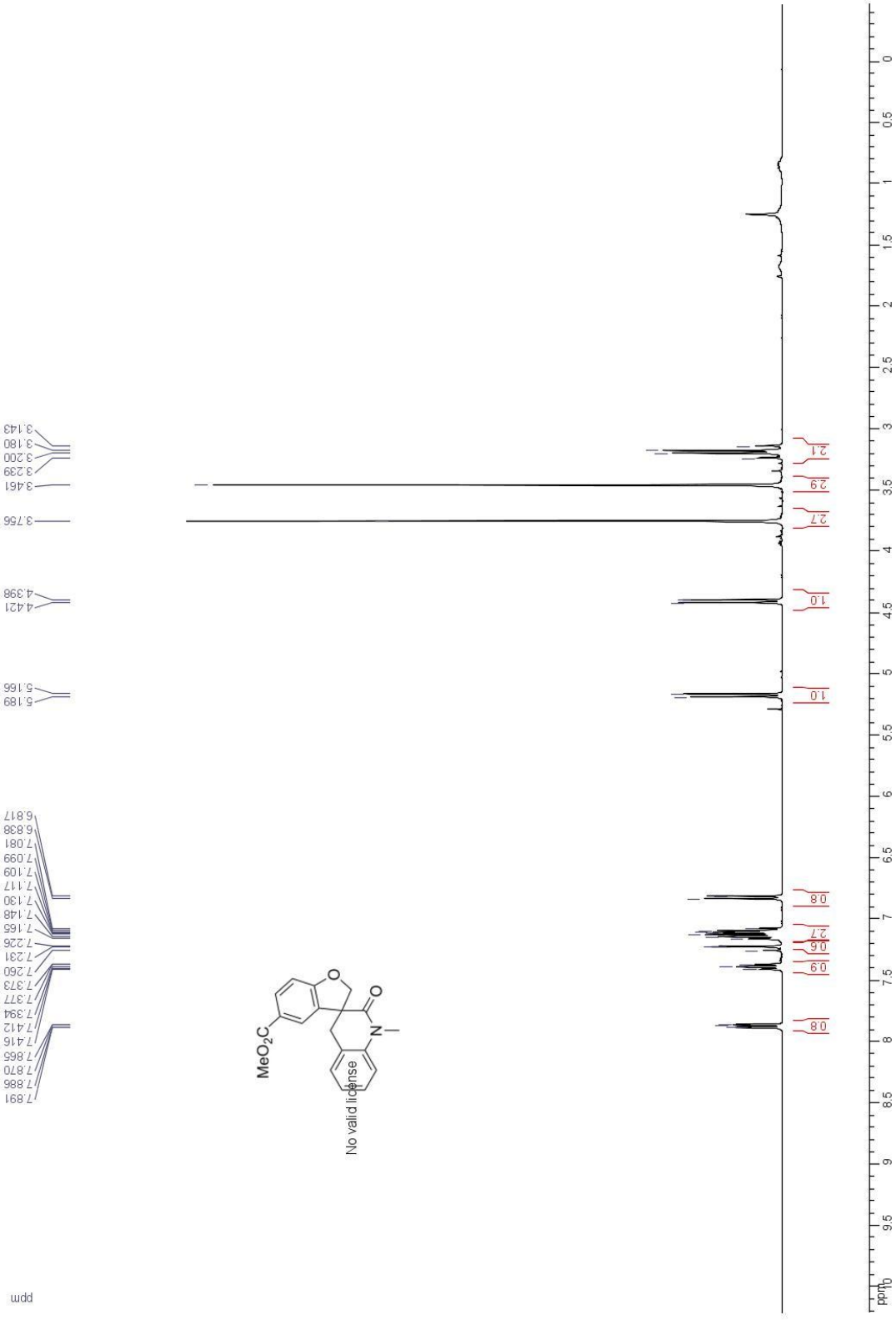
*1',2'-dihydro-2H-spiro[benzofuran-3,5'-pyrrolo[3,2,1-ij]quinolin]-4'(6'H)-one (**2l**)*



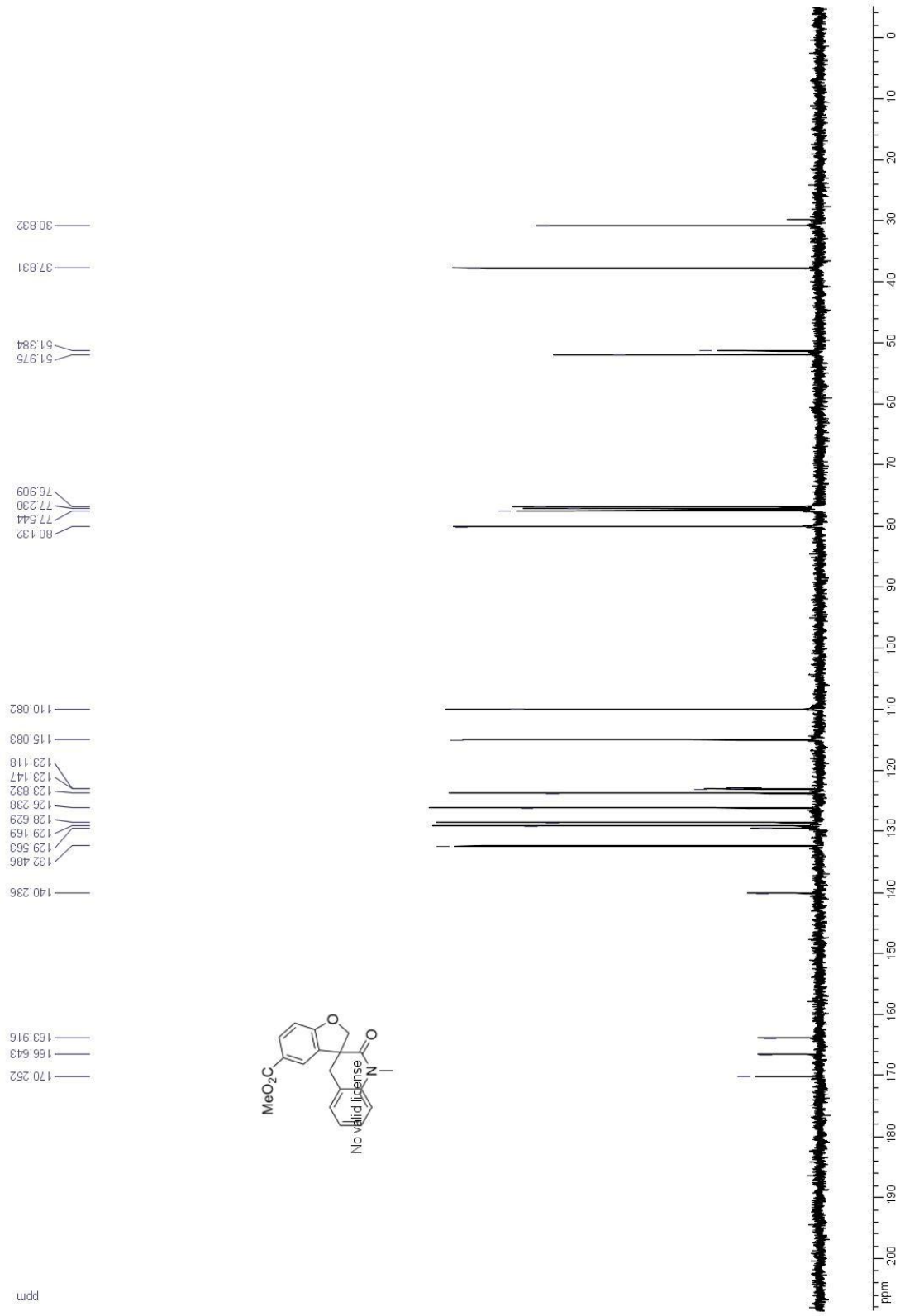
*1',2'-dihydro-2H-spiro[benzofuran-3,5'-pyrrolo[3,2,1-ij]quinolin]-4'(6'H)-one (***2l***)*



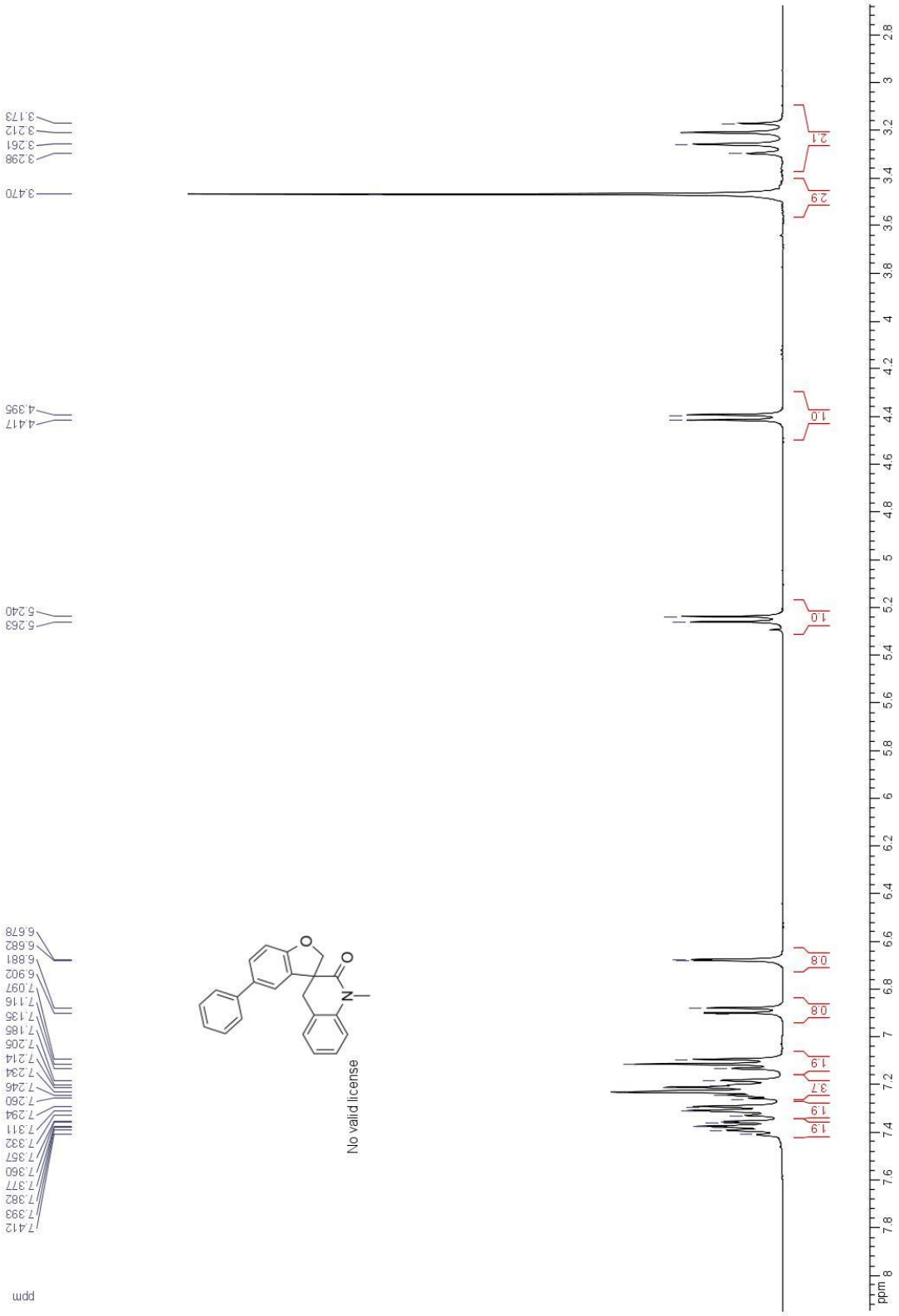
*methyl 1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-5-carboxylate (***2p***)*



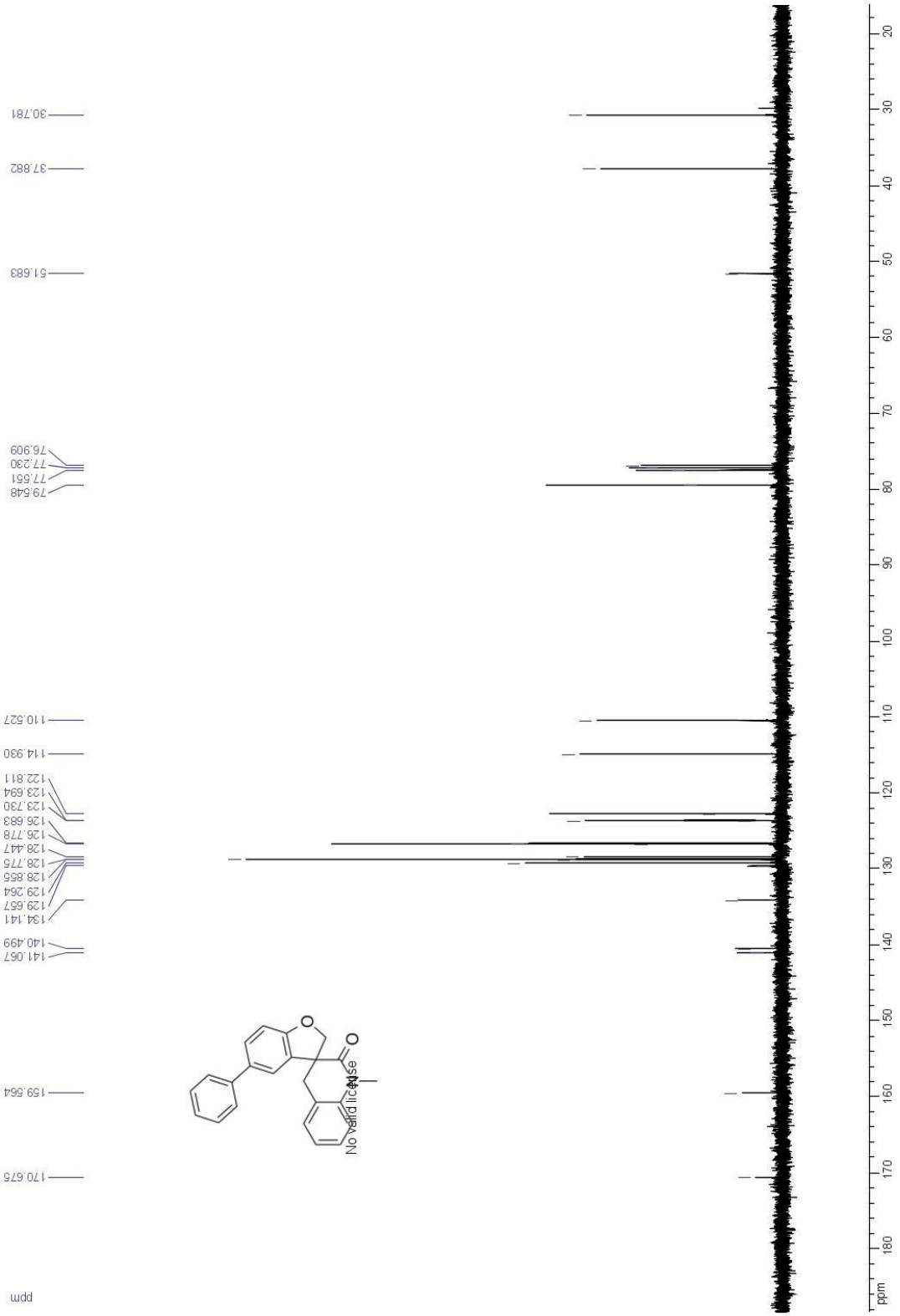
*methyl 1'-methyl-2'-oxo-2',4'-dihydro-1'H,2H-spiro[benzofuran-3,3'-quinoline]-5-carboxylate (***2p***)*



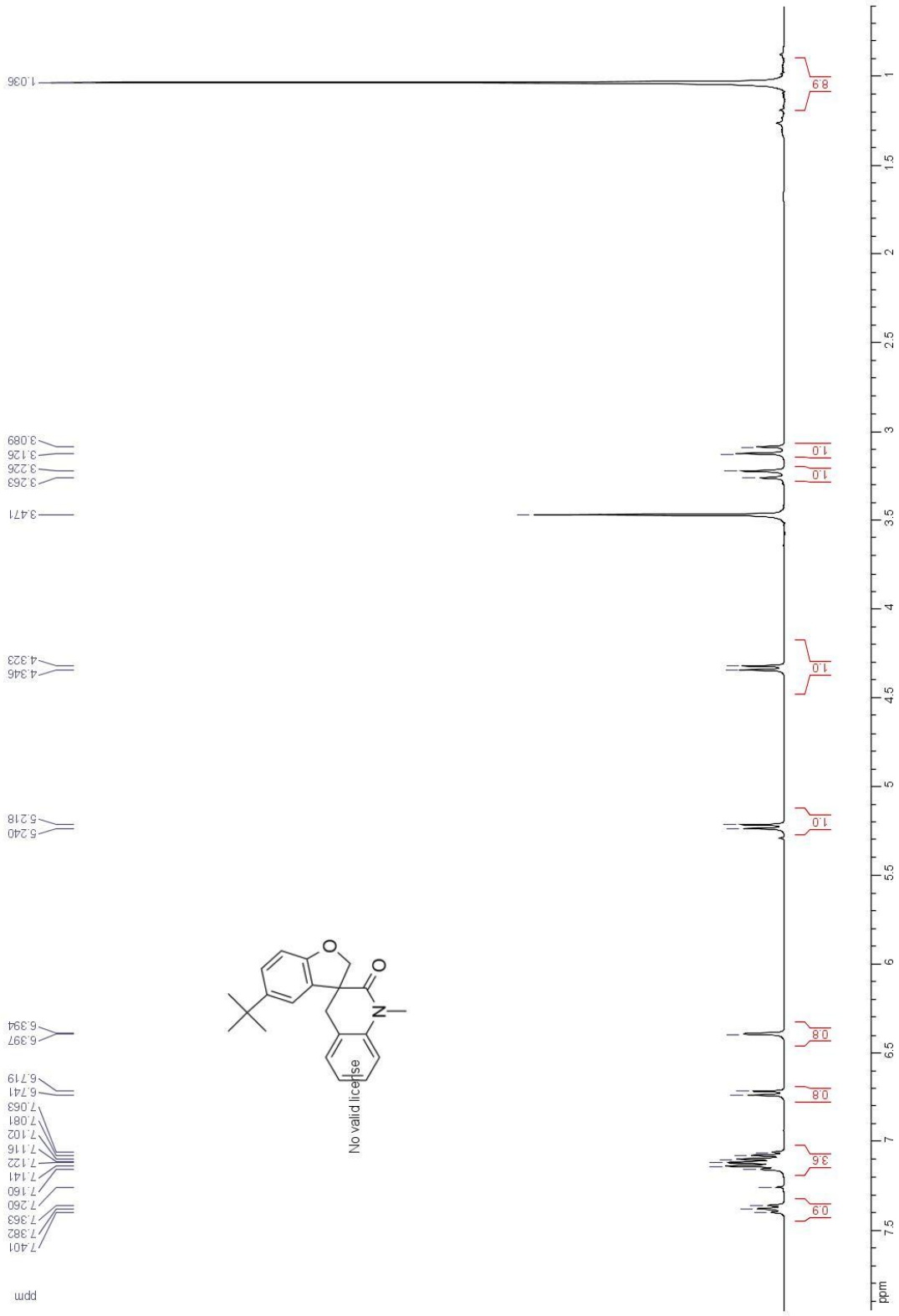
*1'-methyl-5-phenyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2q***)*



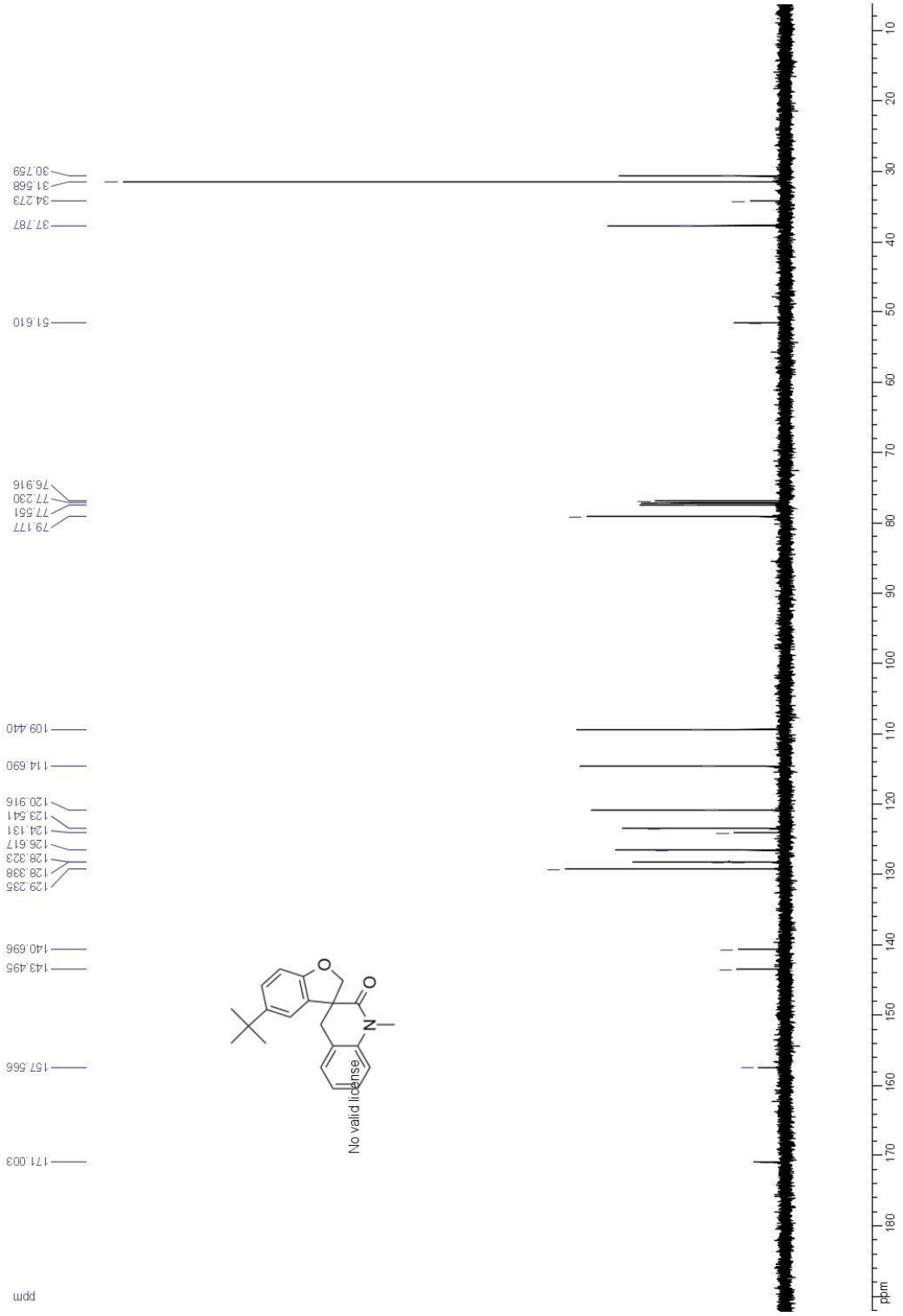
*1'-methyl-5-phenyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2q***)*



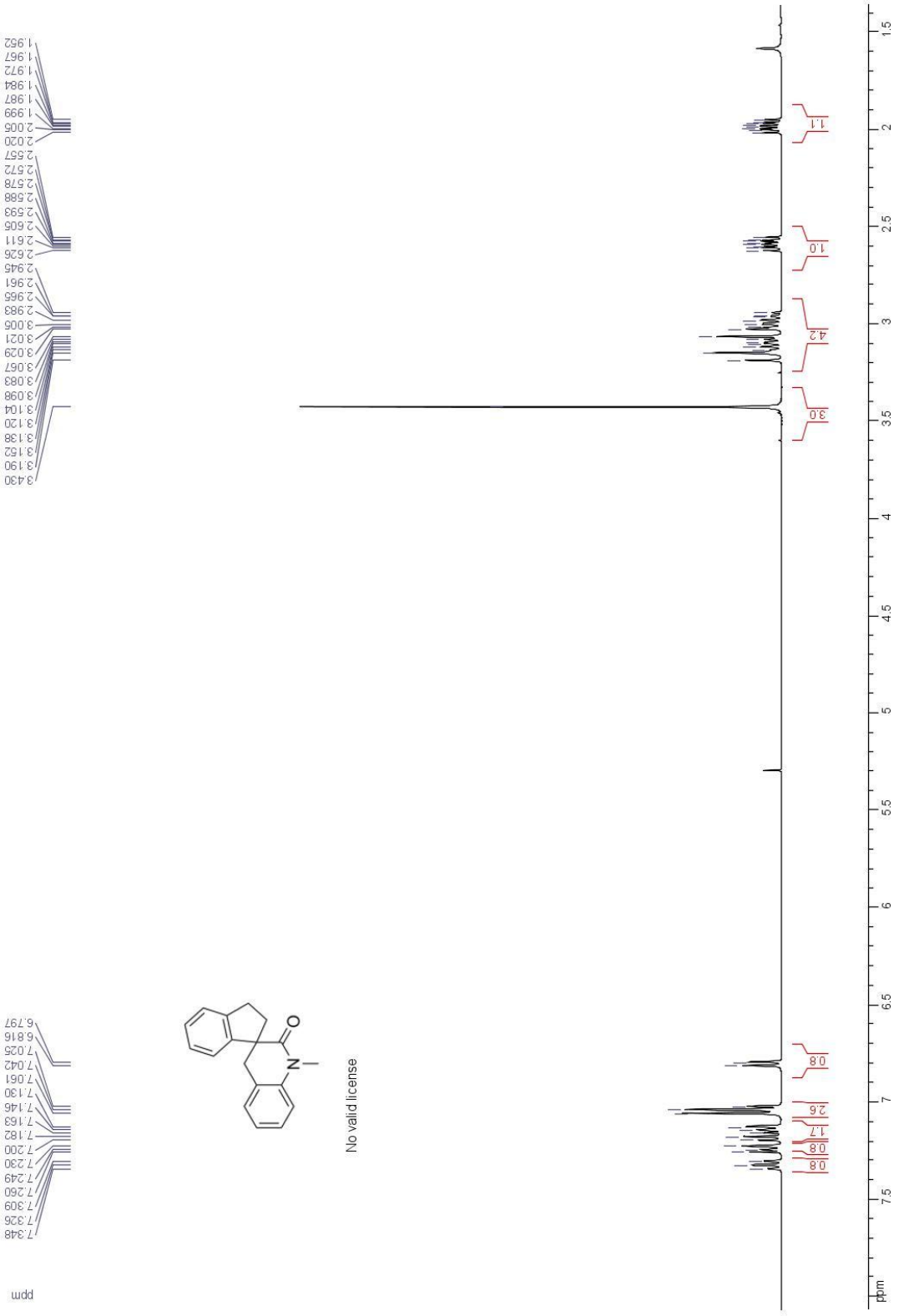
*5-(tert-butyl)-1'-methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2r***)*



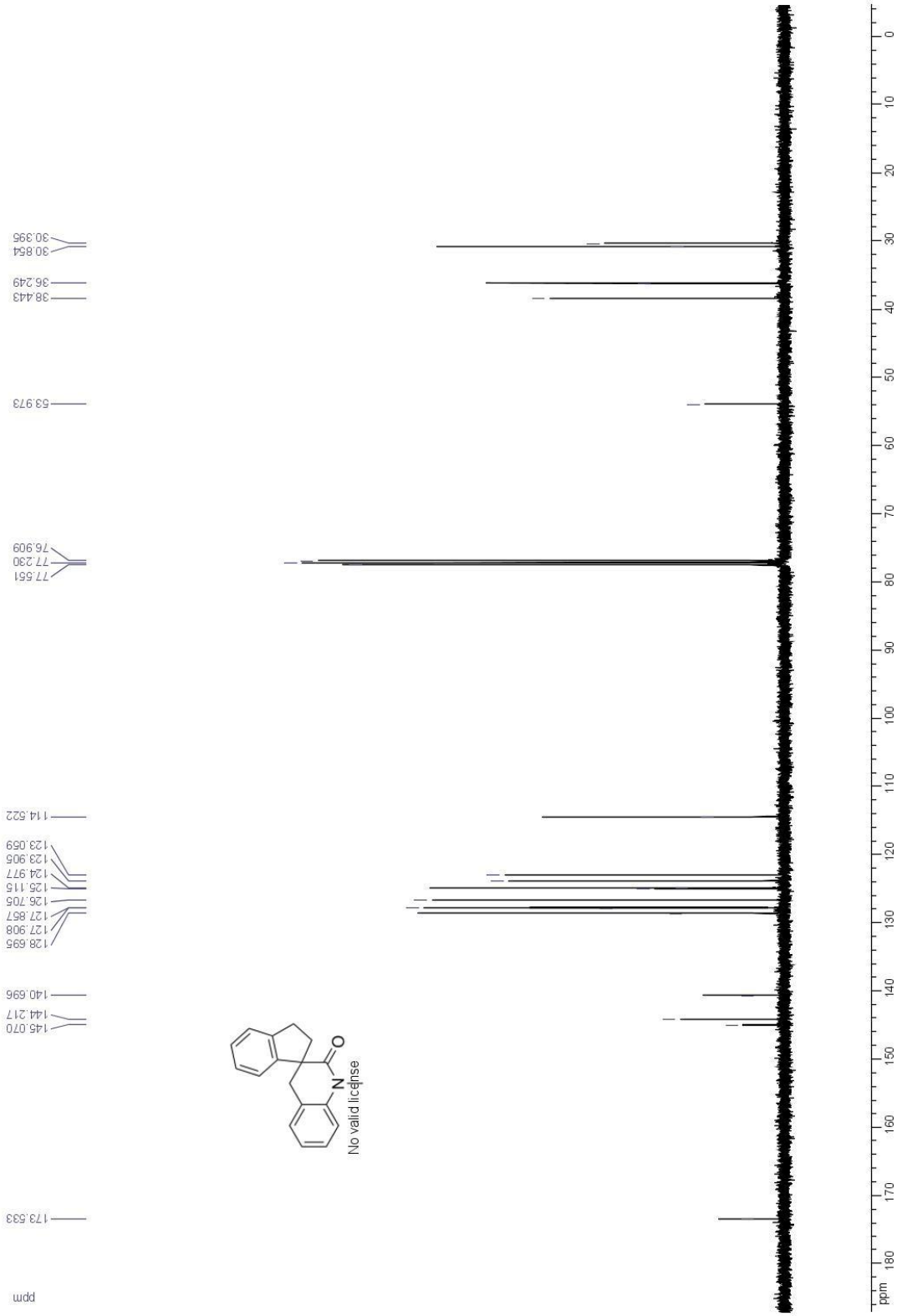
*5-(tert-butyl)-1'-methyl-1'H,2H-spiro[benzofuran-3,3'-quinolin]-2'(4'H)-one (***2r***)*



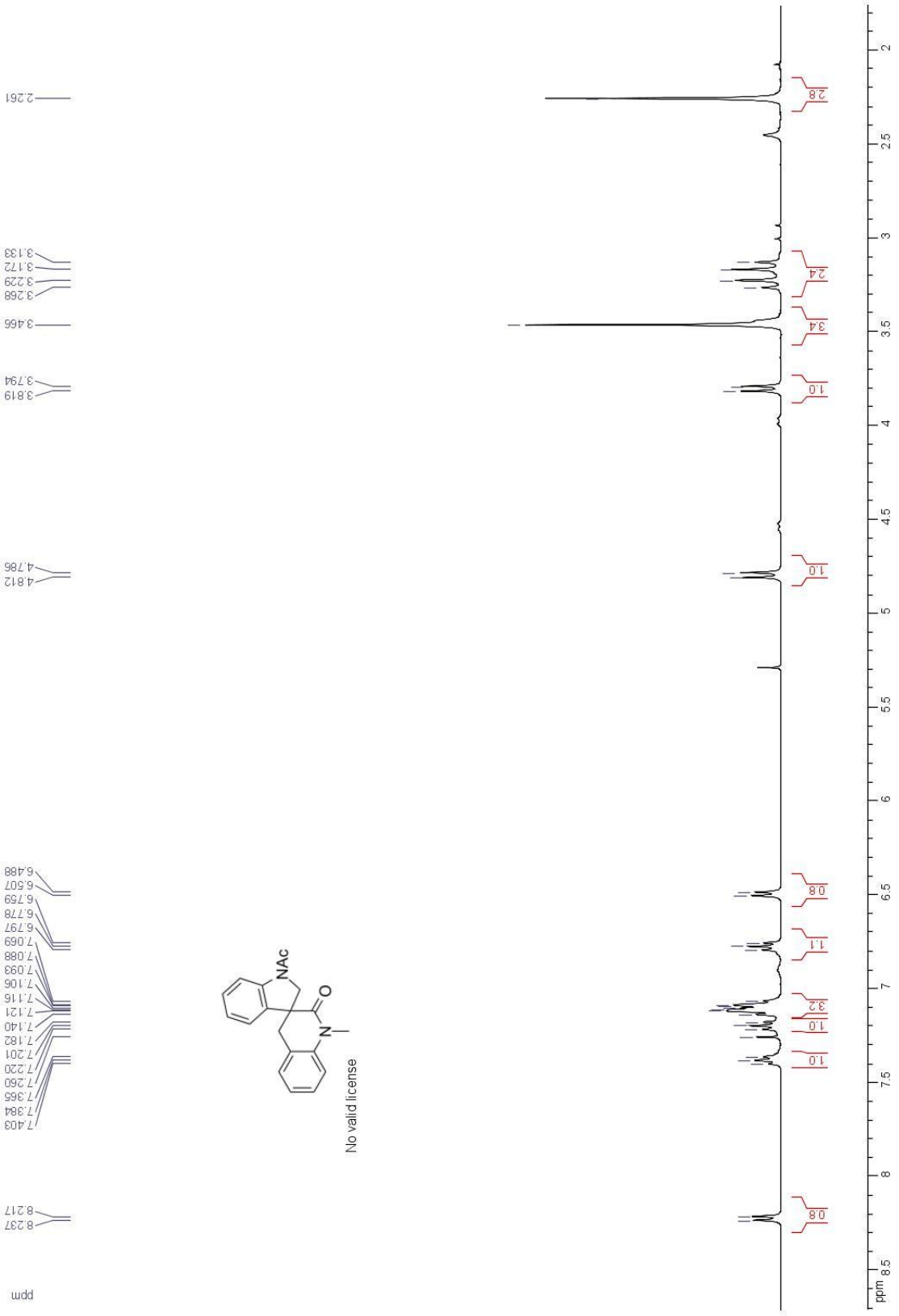
*1'-Methyl-2,3-dihydro-1'H-spiro[indene-1,3'-quinolin]-2'(4'H)-one (***2s***)*



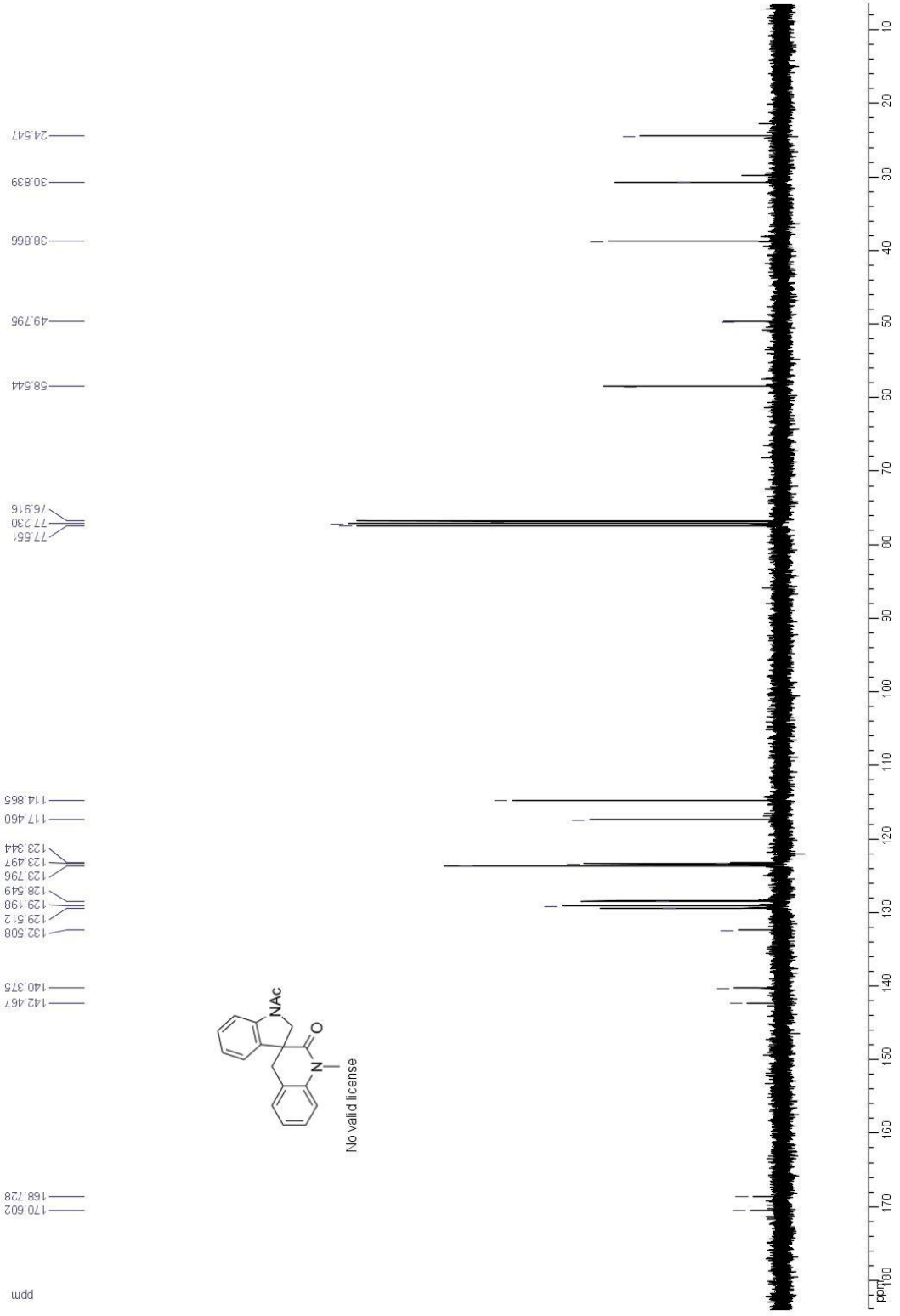
*1'-Methyl-2,3-dihydro-1'H-spiro[indene-1,3'-quinolin]-2'(4'H)-one (***2s***)*



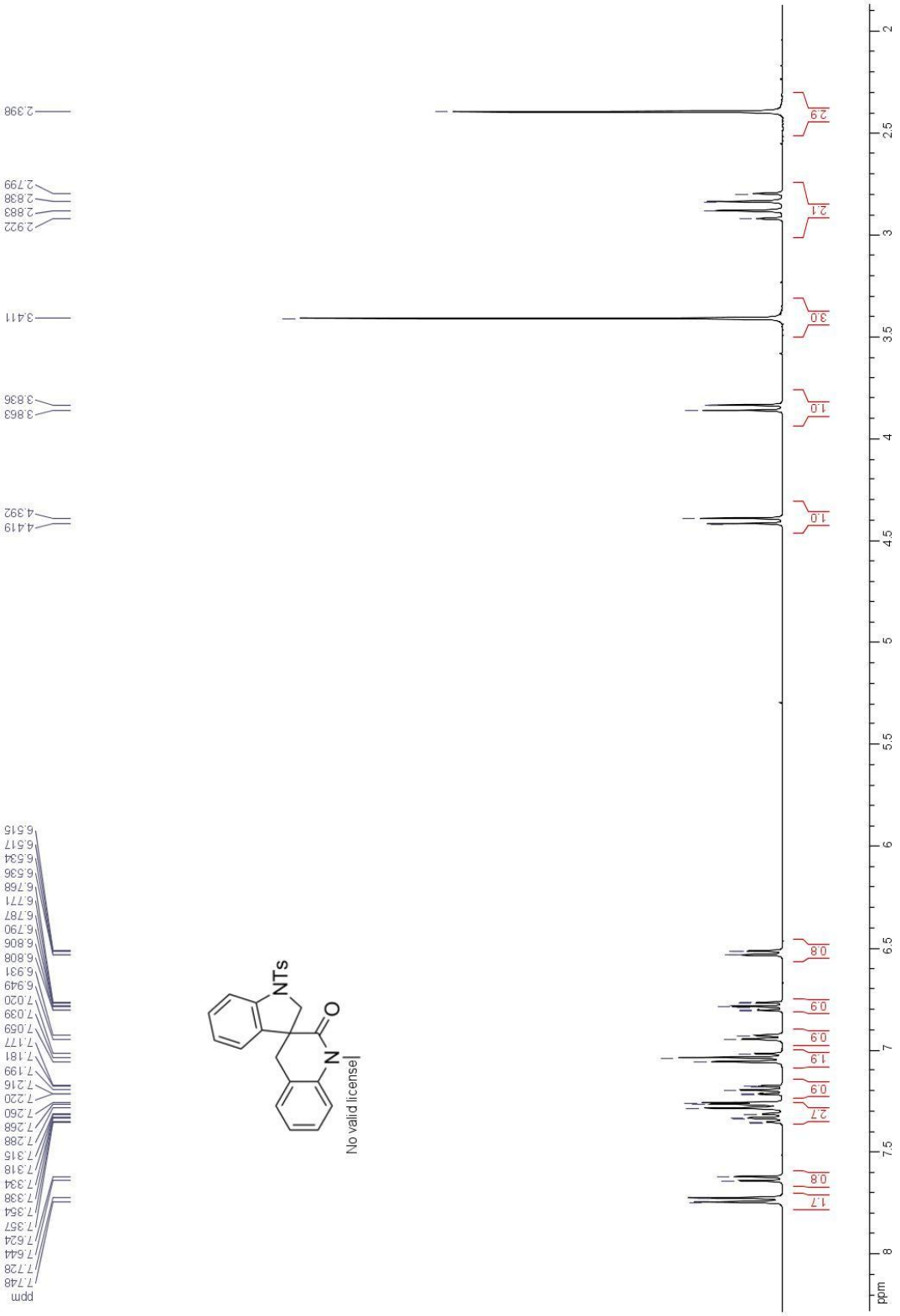
*1-Acetyl-1'-methyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2t***)*



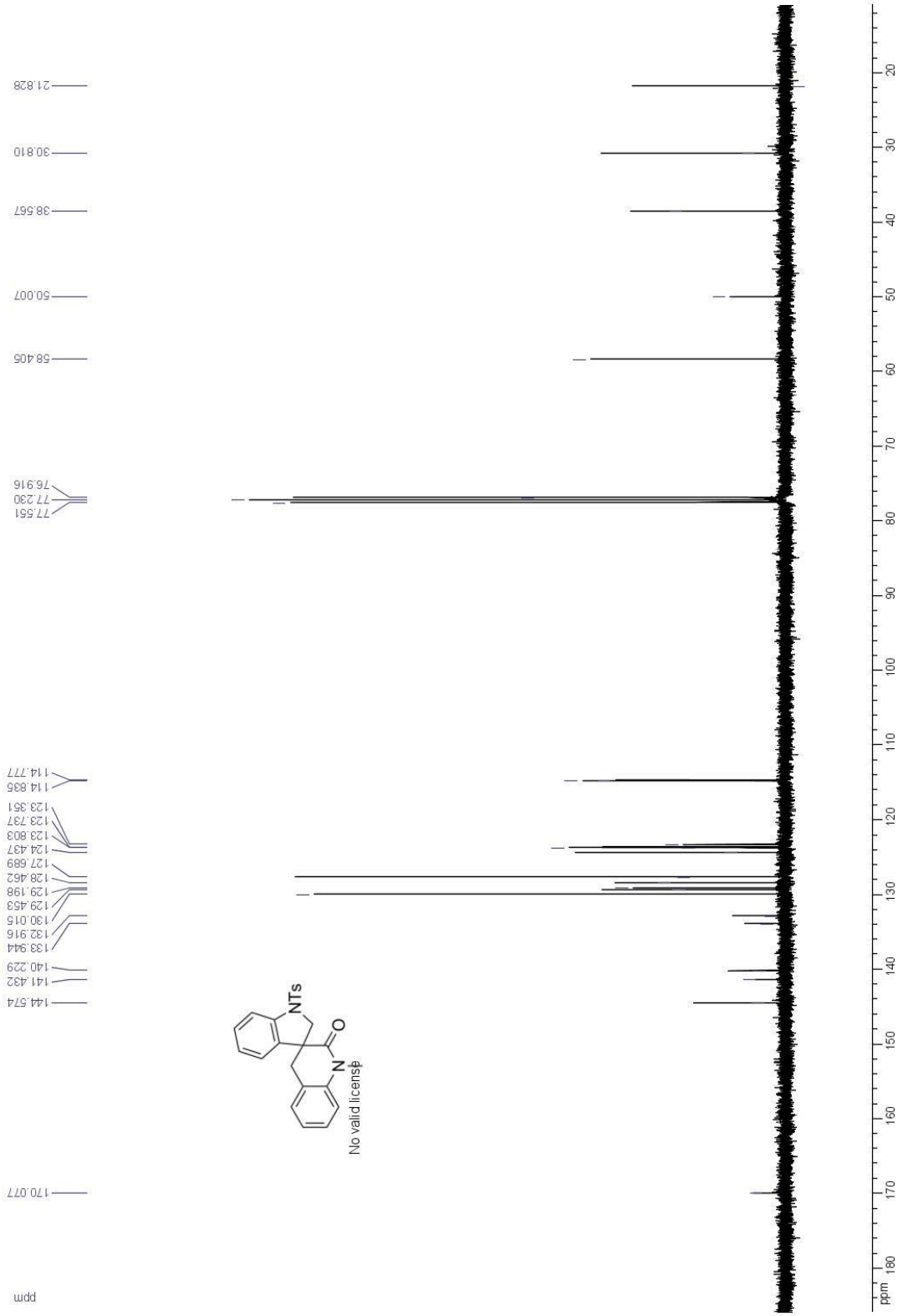
*1-Acetyl-1'-methyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2t***)*



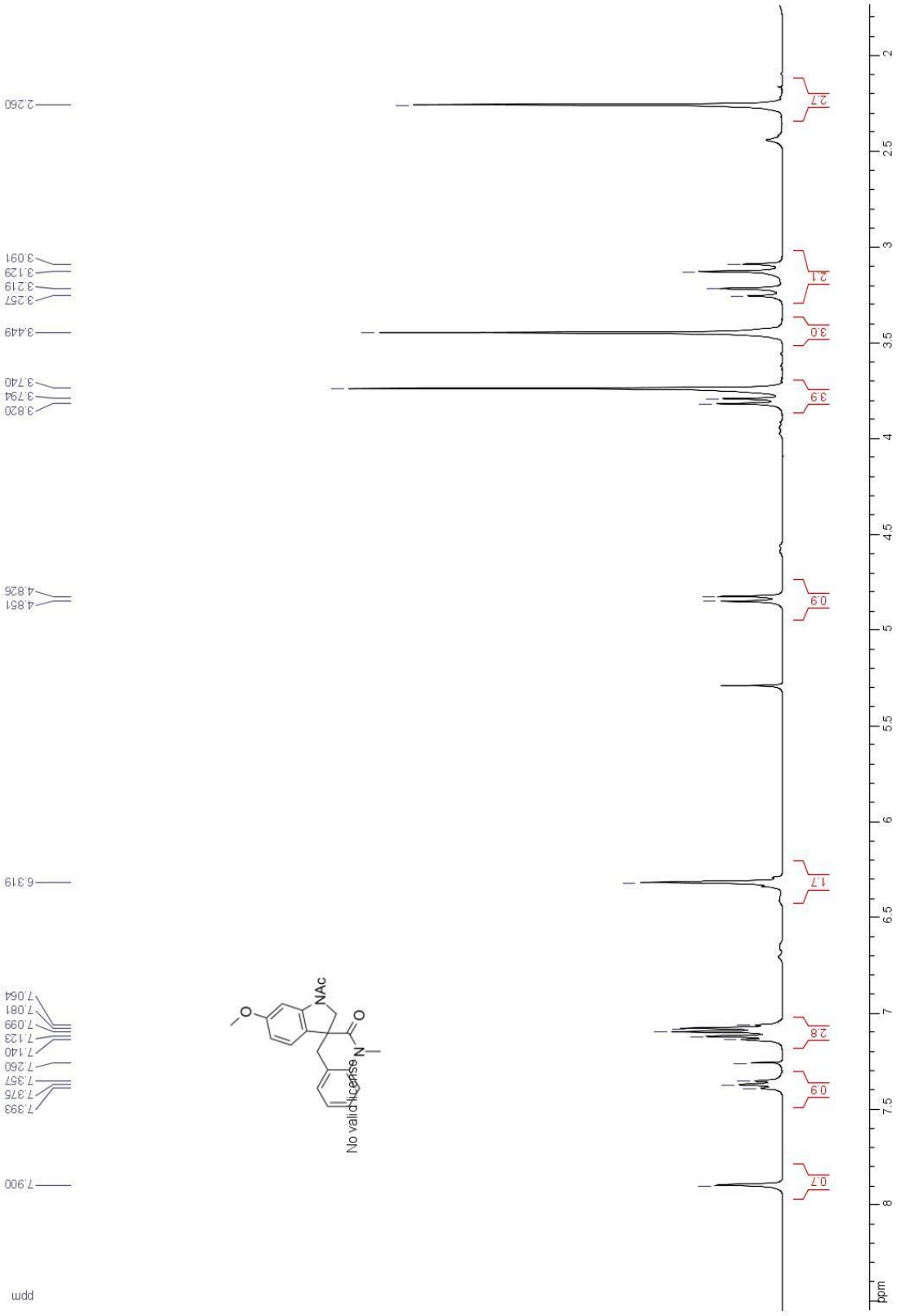
*1'-methyl-1-tosyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2u***)*



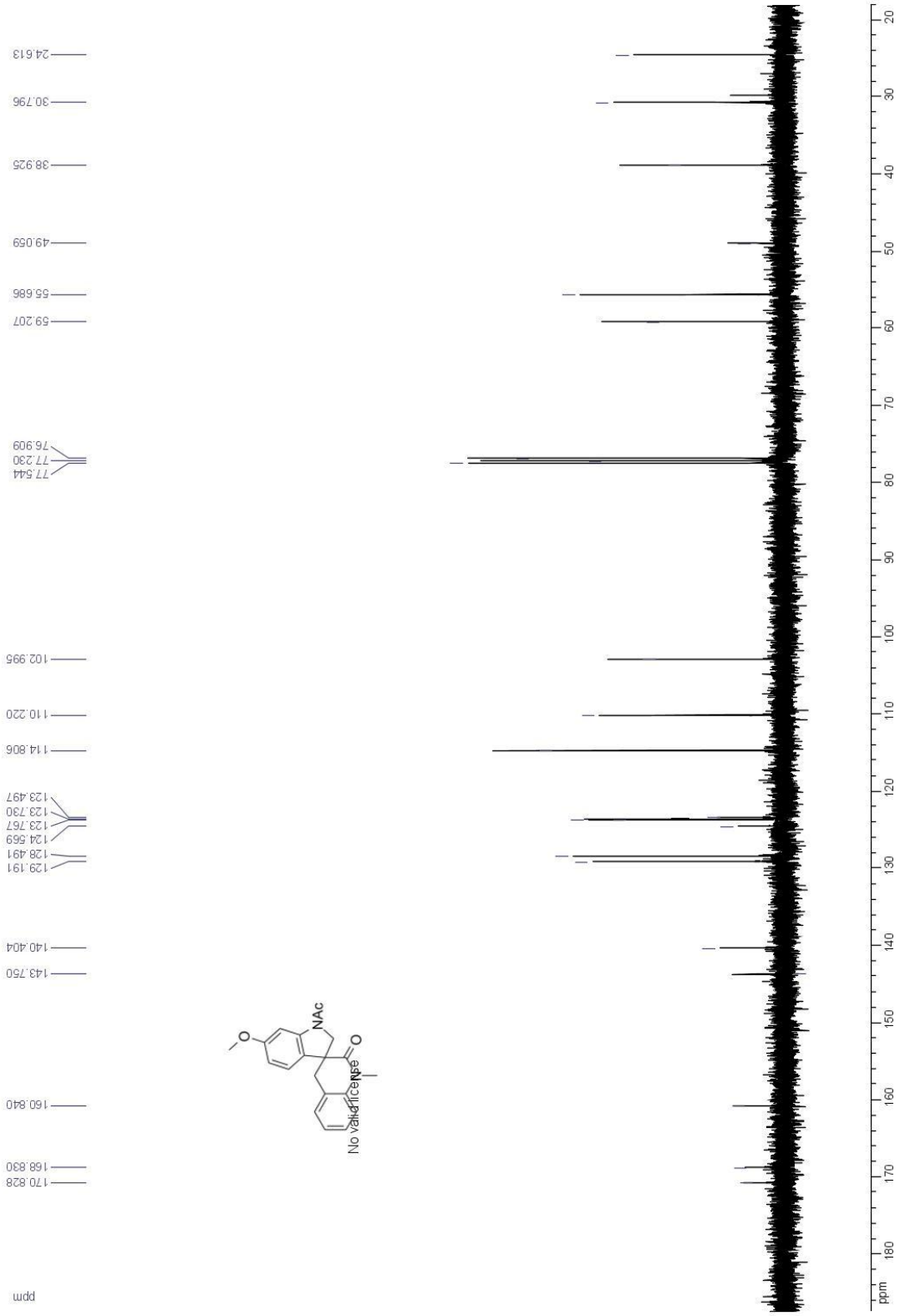
*1'-methyl-1-tosyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2u***)*



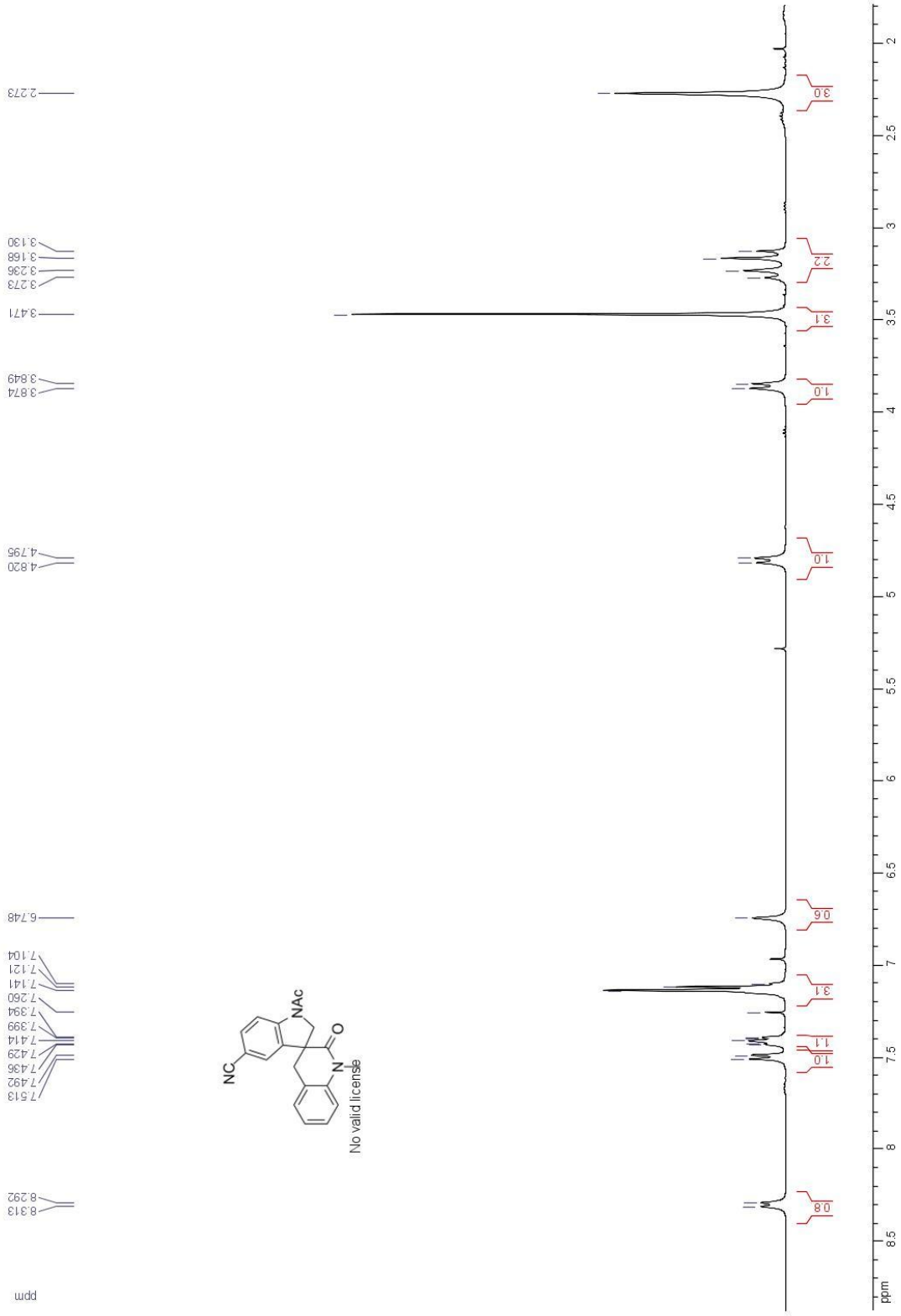
*1-Acetyl-6-methoxy-1'-methyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2v***)*



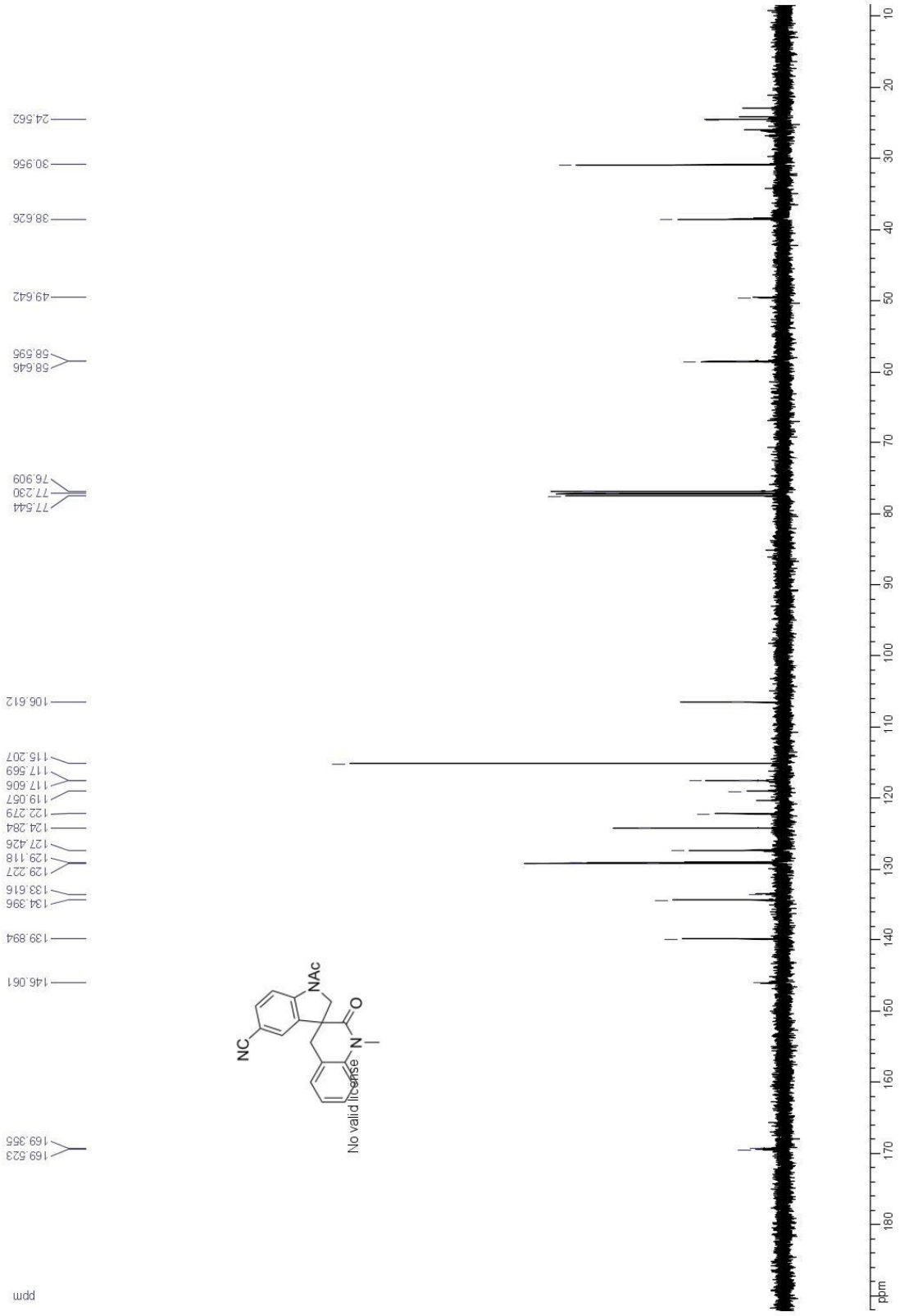
*1-Acetyl-6-methoxy-1'-methyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2v***)*



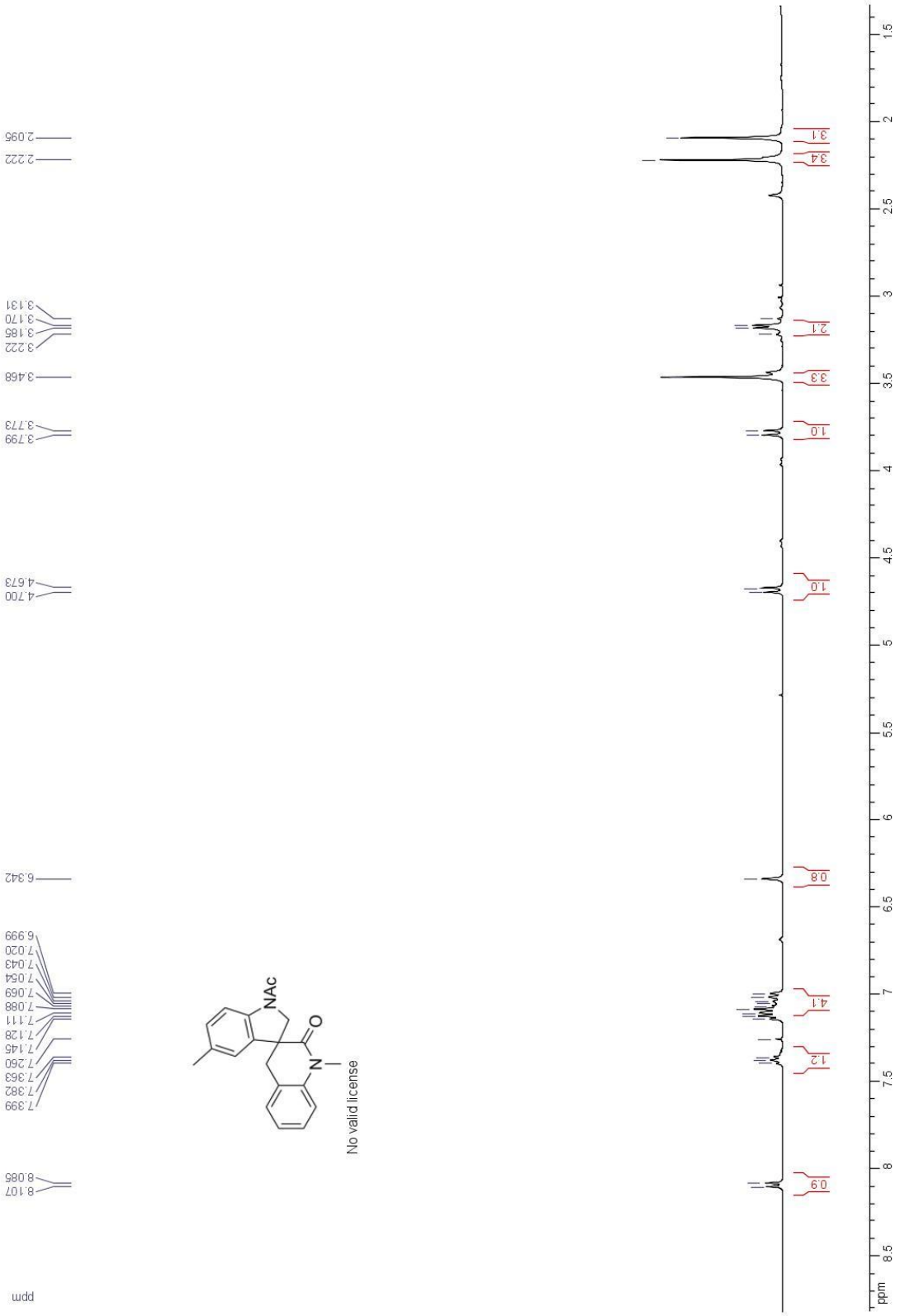
*1-Acetyl-1'-methyl-2'-oxo-2',4'-dihydro-1'H-spiro[indoline-3,3'-quinoline]-5-carbonitrile (***2w***)*



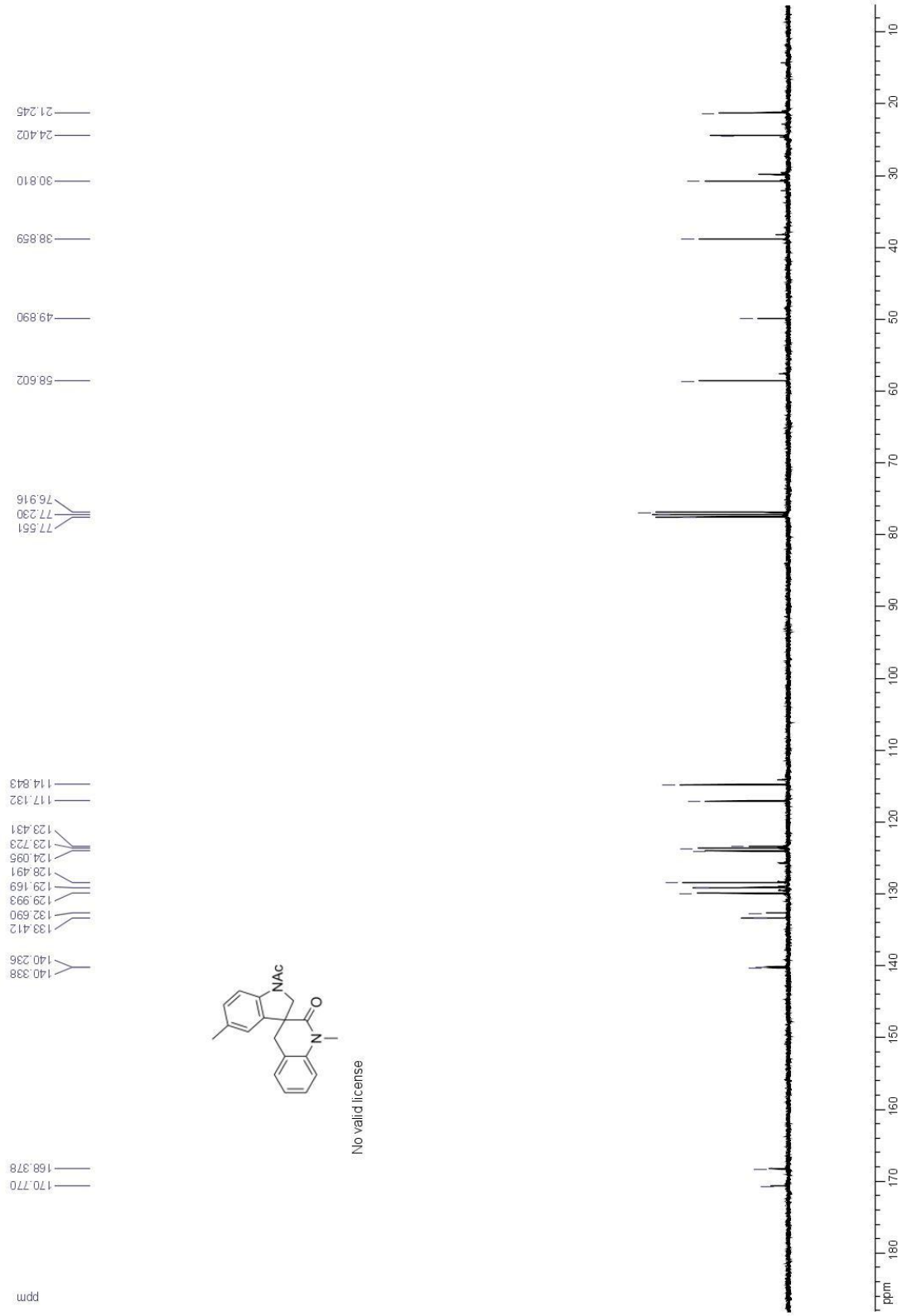
*1-Acetyl-1'-methyl-2'-oxo-2',4'-dihydro-1'H-spiro[indoline-3,3'-quinoline]-5-carbonitrile (***2w***)*



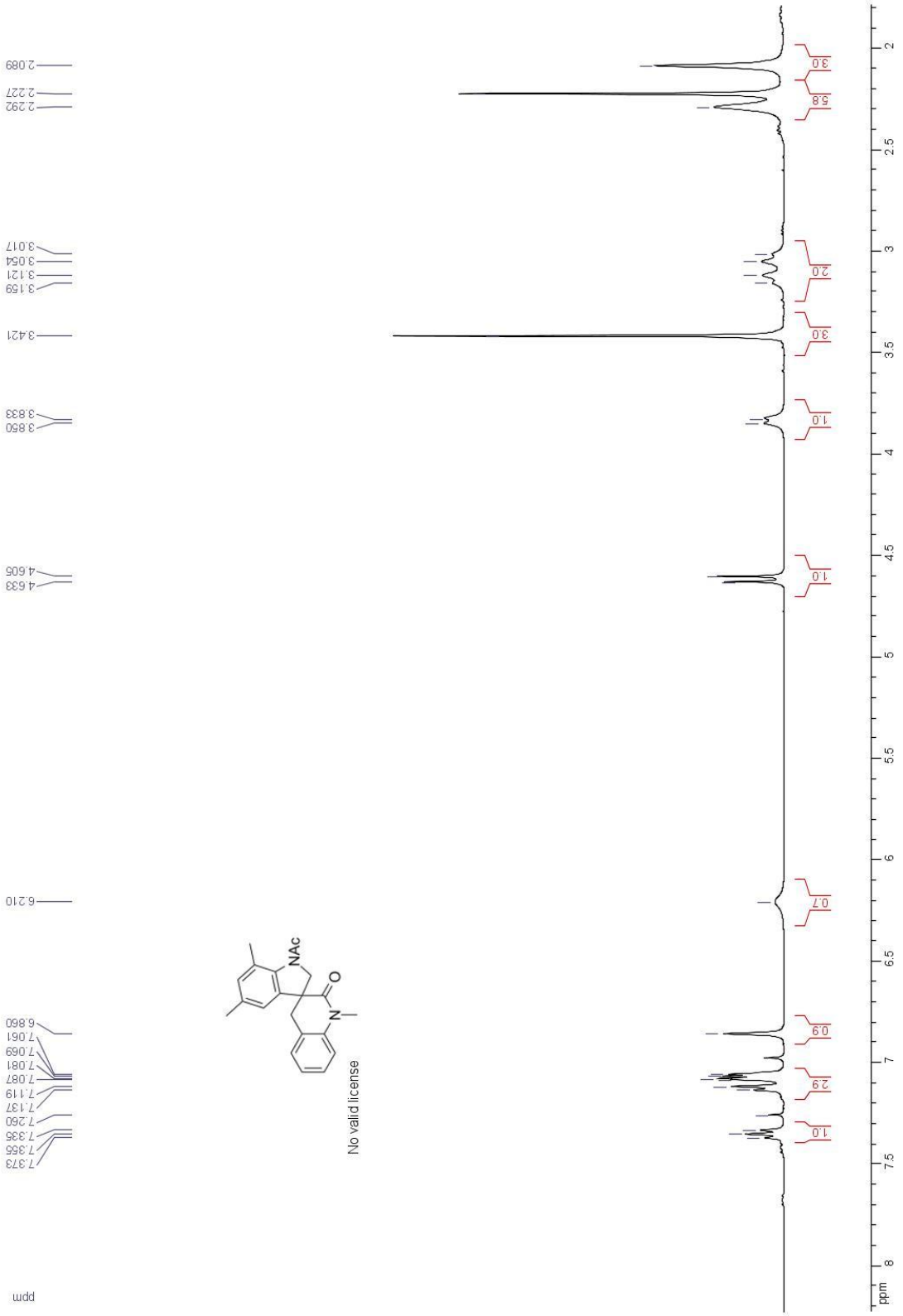
*1-Acetyl-1',5-dimethyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2w***)*



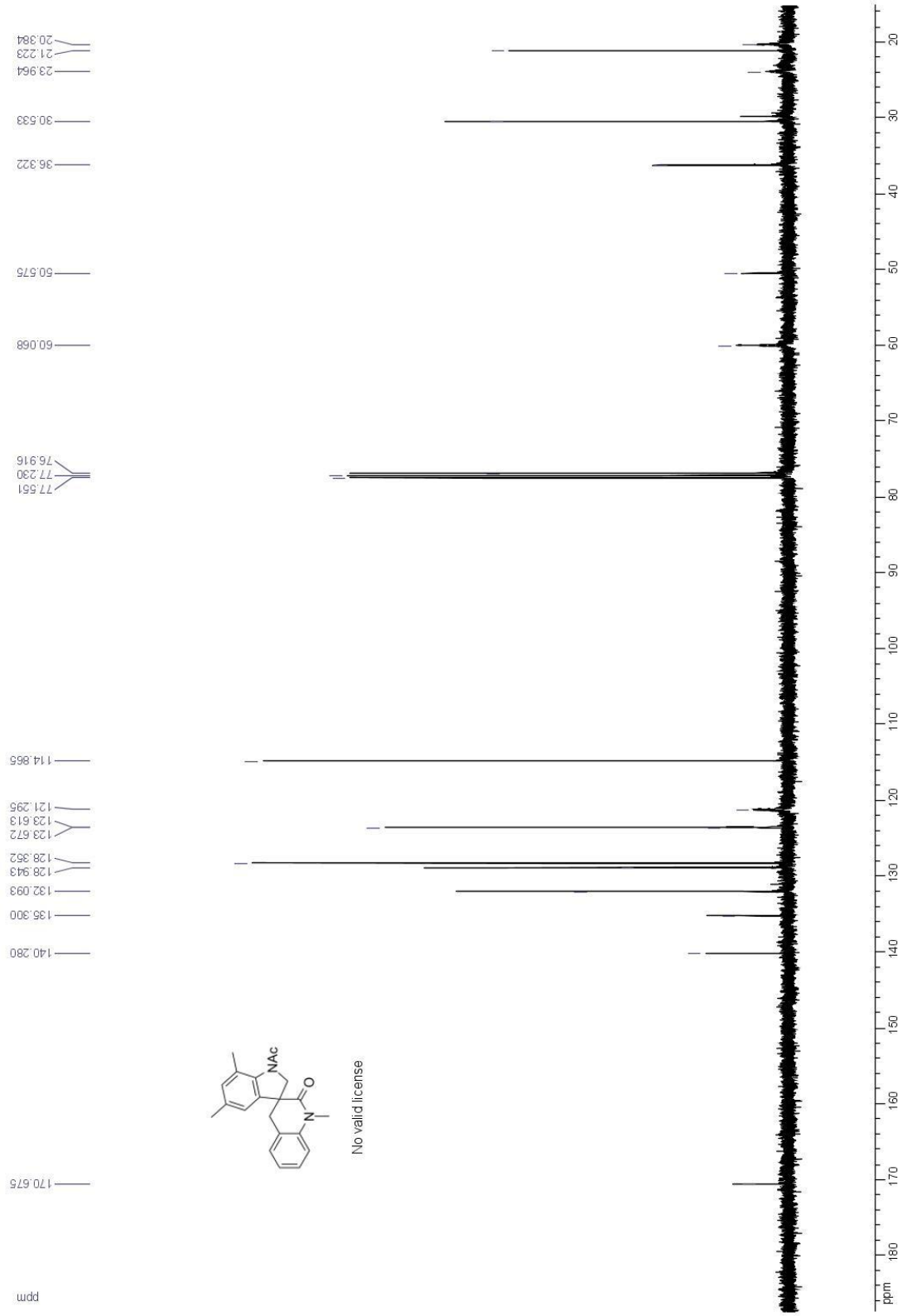
*1-Acetyl-1',5-dimethyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2w***)*



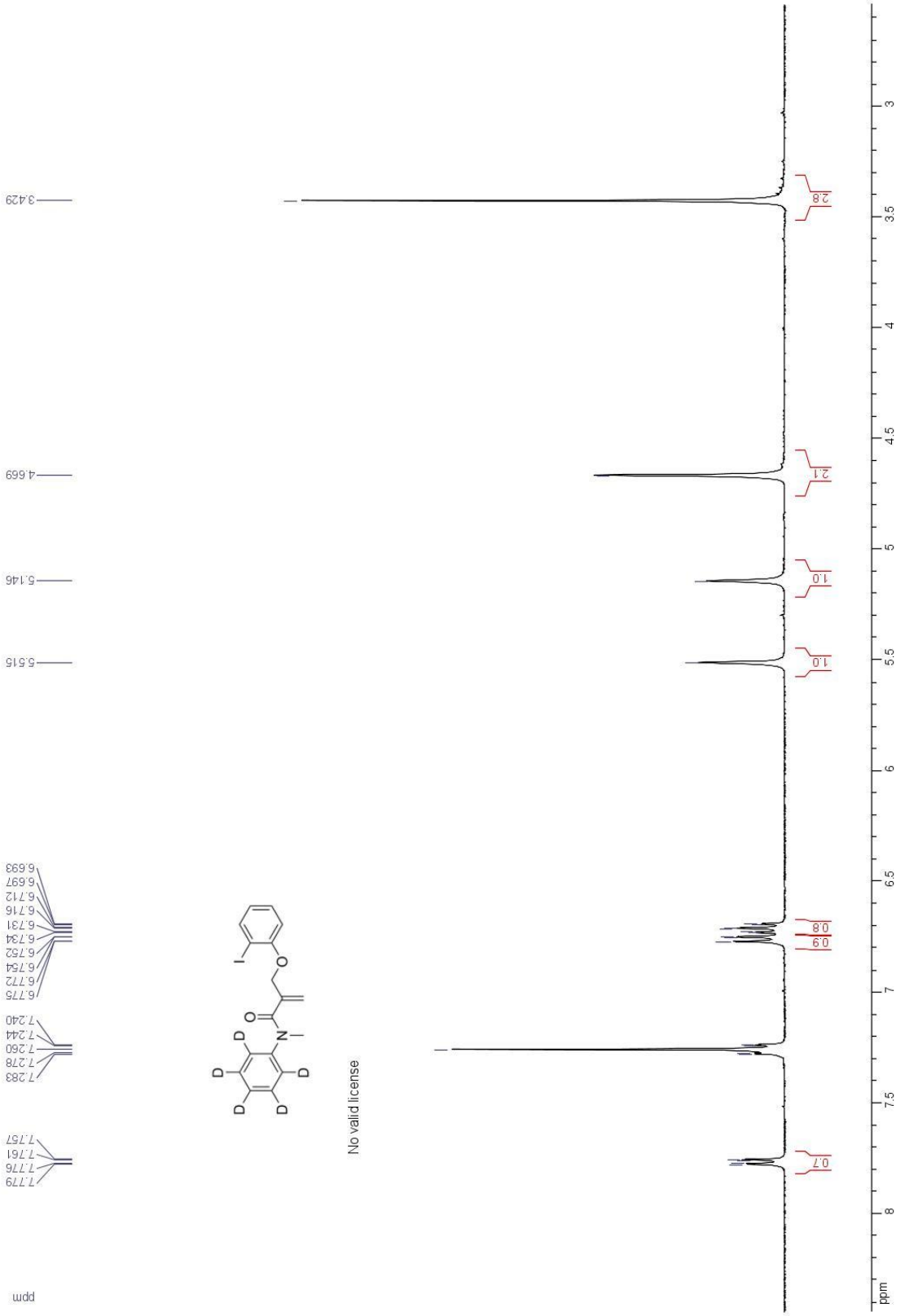
*1-Acetyl-1',5,7-trimethyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (***2y***)*



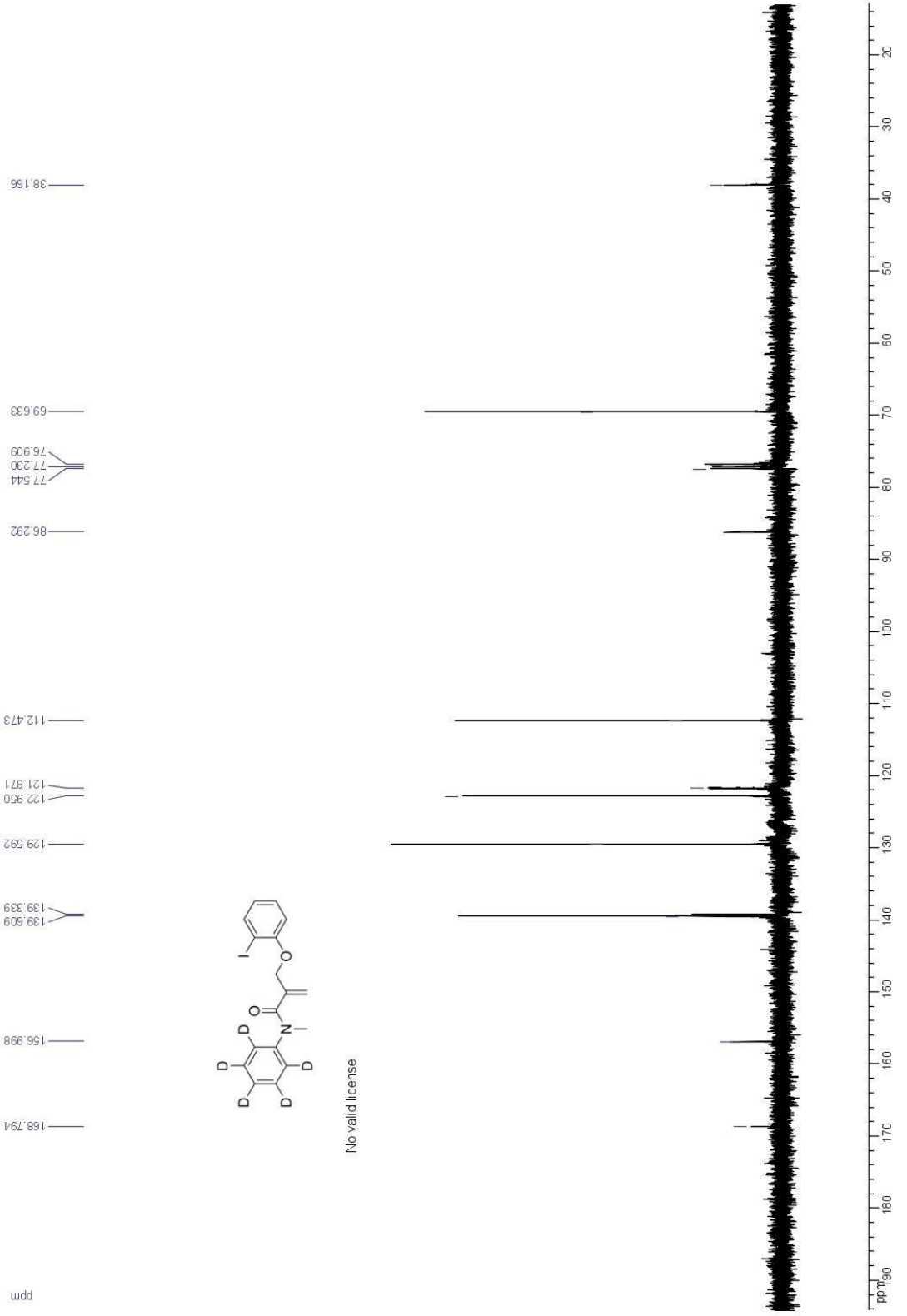
*1-Acetyl-1',5,7-trimethyl-1'H-spiro[indoline-3,3'-quinolin]-2'(4'H)-one (**2y**)*



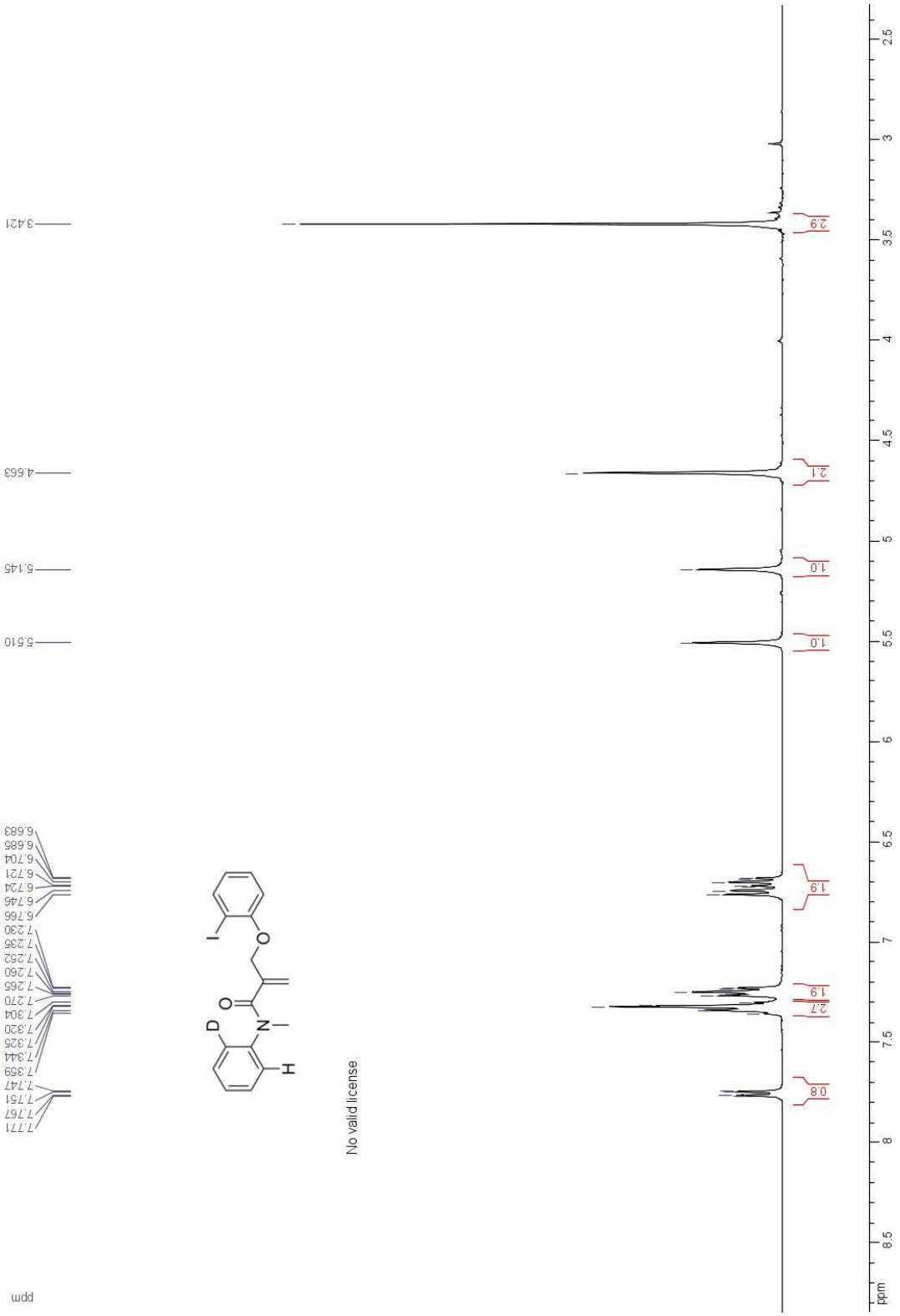
2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide-d_5 (**1a-D_5**)



2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide-d_5 (**1a-D_5**)



2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide-d_1 (**1a-D_1**)



2-((2-Iodophenoxy)methyl)-N-methyl-N-phenylacrylamide-d_1 (**1a-D_1**)