Electronic Supplementary Material (ESI) for Reaction Chemistry & Engineering.

Supporting Information

Sustainable and selective Ni-catalyzed allylation of 2-oxindoles and 2-coumaranones in batch and flow chemistry

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1- General experimental information

Chemicals were purchased from Aldrich, TCI (reagents and solvents) and Strem (nickel precursor and ligands). Common solvents were distilled and degassed before use. The reactions were performed under nitrogen atmosphere using standard Schlenk line techniques. $Ni(cod)_2$ is stored and weighted under argon atmosphere in a glovebox. Conversions were determined by gas chromatography on Shimadzu 2010 equipped with a column HT-5 (30 m, i.d. = 0.32 mm). NMR spectra were recorded using a Bruker AC 300 spectrometer. ^{1}H and ^{13}C NMR chemical shifts are reported to the solvent resonance [$CDCl_3$: 7.27 ppm (^{1}H), 77.0 ppm (^{13}C)].

2- General procedures

*2.1- Allylation of oxindole **1a** with allyl alcohol **2a** in batch mode*

A Schlenk tube closed with a Rotaflo® stopcock was filled in a glove box under argon atmosphere with the precursor $Ni(cod)_2$ (0.054 mmol, 14.8 mg, 3 mol%). The other reactants were added out the glove box by using Schlenk tube techniques (vacuum / nitrogen line). The dppf ligand (0.108 mmol, 60 mg, 6 mol%) and oxindole (1.8 mmol, 240 mg, 1 eq.) were subsequently added followed by freshly distilled and degassed allyl alcohol (5.4 mmol, 0.36 mL, 3 eq.). DMSO (0.5 mL) was then added, and the reaction mixture was stirred at 100 °C for 17 h. For determination of oxindole conversion by GC, the crude was homogenized with 0.5 mL methanol and a precise quantity of anisole (1 mmol) was added as internal standard. Conversions were calculated from the GC analysis of the homogeneous mixture. *N*-allylindole **3a** was purified by silica gel column chromatography using petroleum ether/ethyl acetate (90/10) as eluent.

2.2- Allylation of oxindole derivatives with allylic alcohols and diallylether

The products were synthesized according to the same procedure as those for allylation of oxindole with allyl alcohol. The products were purified by silica gel column chromatography using petroleum ether/ethyl acetate (90/10) as eluent.

2.3- C-allylation of coumaranones with allyl alcohol

The products were synthesized according to the same procedure as those for allylation of oxindole. The products were purified by silica gel column chromatography using petroleum ether/ethyl acetate (95/5) as eluent.

2.4- Allylation of oxindole or coumaranone in flow

We set up a reactor with a FEP tubing (length = 1 meter; tubing i.d. = 800 µm; heated volume = 0.5 mL). The catalyst was prepared in a Schlenk tube by mixing $Ni(cod)_2$ (3 mol%) and dppf (6 mol%) in THF (5 mL) then oxindole or coumaranone (18 mmol, 1 eq.) and allyl alcohol (54 mmol, 3.6 mL, 3 eq.) were added. This solution was introduced thanks to a unique syringe in the reactor. The tube was immersed in a water bath at 80 °C. A Back Pressure Regulator (20 Psi) should be installed because the reaction temperature was superior to the boiling point of the solvent.

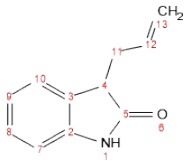
*

Depending the desired residence time, the flow was adjusted according the following table.

Flow (µL/min)	Residence time (min)
4	125.66
10	50.26
20	25.13
40	12.57
100	5.03
200	2.51

3- NMR Characterization of the products and HR-MS of the new ones

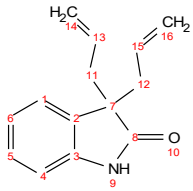
3-allylindolin-2-one (3a)[1]



^{1}H NMR (300 MHz, Chloroform-*d*) δ 8.73 (s, 1H, H_1), 7.26 (d, *J* = 7.4 Hz, 1H, H_{10}), 7.21 (m, 1H, H_8),), 7.01 (td, *J* = 7.6, 1.0 Hz, 1H, H_9), 6.90 (d, *J* = 7.7 Hz, 1H, H_7), 5.87 – 5.65 (m, 1H, H_{12}),), 5.18 – 4.99 (m, 2H, H_{13}), 3.53 (dd, *J* = 7.4, 5.0 Hz, 1H, H_4), 2.83 (m, 6.3, 5.1, 1.4 Hz, 1H, H_{11}), 2.60 (m, 1H, H_{11}).

^{13}C NMR (75 MHz, Chloroform-d) δ 180.49 (C5), 142.16 (C2), 134.53 (C12), 129.90 (C3), 128.65 (C8), 125.13 (C10), 122.92 (C9), 118.78 (C13), 110.40 (C7), 46.39 (C4), 35.43 (C11).

3,3-diallylindolin-2-one (4a), Yield : 60 %

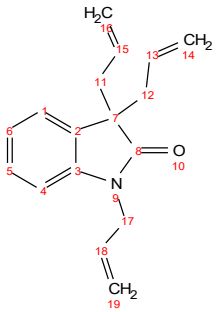


^{1}H NMR (300 MHz, Chloroform-*d*) δ 9.28 (s, 1H, H9), 7.26 – 6.81 (m, 4H, H1+H4+H5+H6), 5.47 (ddt, *J* = 17.5, 10.1, 7.3 Hz, 2H, H13+H15), 5.08 – 4.78 (m, 4H, H14+H16), 2.58 (dt, *J* = 9.1, 4.8 Hz, 4H, H11+H12).

13C NMR (75 MHz, $CDCl_3$) δ 182.02 (C8), 141.16 (C3), 132.26 (C13+C15), 131.89 (C2), 127.95 (C5), 123.70 (C6), 122.28 (C1), 118.94 (C14+C16), 109.94 (C4), 53.40 (C7), 41.31 (C11+C12).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{14}H_{15}NO$: 214.1154; Found 214.1075.

1,3,3-triallylindolin-2-one (5a), Yield: 20 %

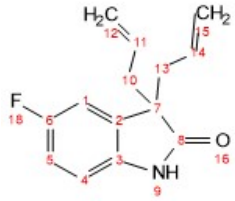


^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.20 (t, *J* = 7.3 Hz, 2H, H_6+H_5), 7.03 (t, *J* = 7.5 Hz, 1H, H_1), 6.81 – 6.72 (m, 1H, H_4), 5.99 – 5.61 (m, 1H,H_{18}), 5.40 (ddt, *J* = 17.2, 10.0, 7.3 Hz, 2H, H_{13}+H_{15}), 5.25 – 5.09 (m, 2H, $1H_{14}$+$1H_{16}$), 5.03 – 4.78 (m, 4H, $1H_{14}$+$1H_{16}$+H_{19}), 4.29 (d, *J* = 5.1 Hz, 2H, H_{17}), 2.75 – 2.35 (m, 4H, H_{11}+H_{12}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 178.55 (C8), 142.94 (C3), 132.32 (C11+C12), 131.59 (C18), 131.22 (C2), 127.77 (C5), 123.40 (C6), 122.22 (C1), 118.87 (C14+C16), 117.24 (C4), 108.83 (C19), 52.66 (C7), 42.11 (C12), 41.40 (C11), 41.04 (C17).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{17}H_{19}NO$: 254.1467; Found 254.1545.

3,3-diallyl-5-fluoroindolin-2-one **(4b), Yield: 67 %**



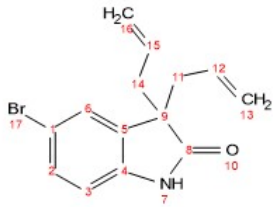
^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.95 (s, 1H, H_9), 7.00 – 6.86 (m, 2H, H_4+H_5), 6.81 (dd, $J_{H1\text{-}F}$ = 8.1 Hz, 1H, H_1), 5.48 (ddt, J = 17.0, 10.0, 7.6 Hz, 2H, H_{11}+H_{14}), 5.11 – 4.91 (m, 4H, H_{12}+H_{15}), 2.68 – 2.49 (m, 4H, H_{10}+H_{13}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 181.71 (C8), 159.17 (d, $J_{C\text{-}F}$ = 240.2 Hz, C6), 136.96 (C3), 133.69 (d, $J_{C\text{-}F}$ = 7.8 Hz, C2), 131.82 (C11+C14), 119.38 (C12+C15), 114.37 (d, $J_{C\text{-}F}$ = 23.5 Hz, C5), 111.7 (d, $J_{C\text{-}F}$ = 24.4 Hz, C1), 110.39 (d, $J_{C\text{-}F}$ = 8 Hz, C4), 54.03 (C7), 41.25 (C10+C13).

^{19}F NMR (282 MHz, Chloroform-d) δ -121.08 (td, $J_{F\text{-}H1}$ = $J_{F\text{-}H5}$ = 8.7, $J_{F\text{-}H4}$ = 4.3 Hz).

HR-MS (ESI) m/z: Calcd for $[M+H]^+$ $C_{14}H_{14}FNO$: 232.1059; Found 232.0981.

3,3-diallyl-5-bromoindolin-2-one **(4c), Yield: 70 %**

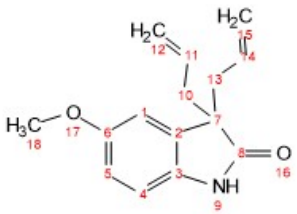


^{1}H NMR (300 MHz, Chloroform-*d*) δ 9.31 (s, 1H, H_7), 7.37 – 7.30 (m, 2H, H_2+H_3), 6.82 (d, J = 8.2 Hz, 1H, H_6), 5.54 – 5.39 (m, 2H, H_{12}+H_{15}), 5.10 – 4.92 (m, 4H, H_{13}+H_{16}), 2.58 (ddt, J = 6.9, 4.8, 1.1 Hz, 4H, H_{11}+H_{14}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 181.39 (C8), 140.06 (C17+C18), 134.05 (C3), 131.59 (C5), (C13 + C16), 130.83 (C1), 126.79 (C2), 119.38 (C12 + C15), 115.01 (C6), 111.34 (C3), 53.70 (C9), 41.10 (C11 + C14).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{14}H_{14}BrNO$: 292.0259; Found 292.0181.

3,3-diallyl-5-methoxyindolin-2-one **(4d), Yield: 95%**

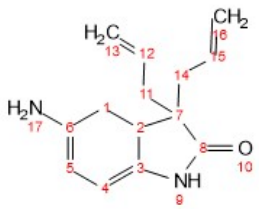


^{1}H NMR (300 MHz, Chloroform-*d*) δ 8.01 (s, 1H, H_9), 6.83 – 6.67 (m, 3H, H_{Ar}), 5.47 (ddd, J = 17.0, 10.1, 7.7 Hz, 2H, H_{11}+H_{14}), 5.11 – 4.89 (m, 4H, H_{12}+H_{16}), 3.79 (s, 3H, H_{18}), 2.67 – 2.44 (m, 4H, H_{10}+H_{13}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 180.82 (C8), 155.69 (C6), 134.14 (C2), 133.27 (C3), 132.12 (C11+C14), 118.92 (C12+C15), 112.04 (C5), 111.18 (C4), 109.71, (C1), 55.79 (C18), 53.64 (C7), 41.30 (C10+C13).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{15}H_{17}NO_2$: 244.1259; Found 244.1338.

3,3-diallyl-5-aminoindolin-2-one **(4e), Yield: 63 %**

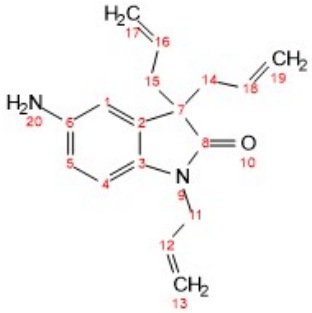


^{1}H NMR (300 MHz, Chloroform-*d*) δ 9.28 (s, 1H, H_8), 6.74 – 6.42 (m, 3H, H_{Ar}), 5.46 (ddt, *J* = 17.1, 10.1, 7.3 Hz, 2H, H_{12}+H_{15}), 5.01 (dd, *J* = 17.0, 1.9 Hz, 3H, H_{12}), 4.89 (dd, *J* = 10.1, 2.0 Hz, 3H, H_{16}), 3.55 (s, 2H, H_{17}), 2.66 – 2.38 (m, 4H, H_{11}+H_{14}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 180.59 (C8), 141.61 (C6), 133.15 (C3), 132.83 (C5), 132.30 (C12+C15), 118.75 (C13+C16), 114.32 (C2), 111.92 (C1), 109.96 (C4), 53.41 (C7), 41.32 (C11+C14).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{14}H_{16}N_2O$: 229.1263; Found 229.1184.

1,3,3-triallyl-5-aminoindolin-2-one **(5e), Yield: 32 %**

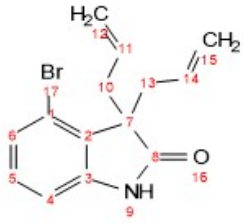


^{1}H NMR (300 MHz, Chloroform-*d*) δ 6.70 – 6.52 (m, 4H, H_{Ar}), 5.75 (ddt, *J* = 17.2, 10.3, 5.1 Hz, 1H, H_{12}), 5.43 (ddd, *J* = 17.0, 10.1, 7.7 Hz, 2H, H_{16}+H_{18}), 5.21 – 5.11 (m, 2H, H_{13}), 5.06 – 4.85 (m, 4H, H_{17}+H_{19}), 4.26 (dt, *J* = 5.1, 1.8 Hz, 2H, H_{11}), 3.54 (s, 2H, H_{20}), 2.65 – 2.45 (m, 4H, H_{10}+H_{14}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 178.02 (C8), 141.75 (C6), 135.45 (C3), 135.19 (C5), 132.46 (C16+C18), 131.88 (C12), 118.70 (C17+C19), 117.06 (C13), 113.99 (C2), 111.76 (C1), 109.33 (C4), 52.89 (C7), 42.16 (C11), 41.41 (C14+C15).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{17}H_{20}N_2O$: 269.1576; Found 269.1654.

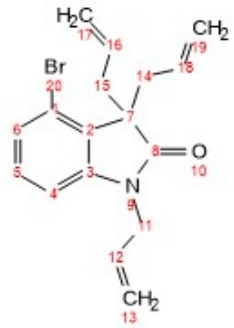
3,3-diallyl-4-bromoindolin-2-one **(4f), Yield: 36%**



^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.63 (s, 1H, H_9), 7.18 (dd, *J* = 8.2, 1.0 Hz, 1H, H_6), 7.09 (dd, *J* = 8.2, 7.6 Hz, 1H, H_5), 6.79 (dd, *J* = 7.6, 1.0 Hz, 1H, H_4), 5.37 (ddt, *J* = 17.2, 10.0, 7.3 Hz, 2H, H_{11}+H_{14}), 5.07 (ddt, *J* = 17.0, 2.3, 1.2 Hz, 2H, H_{12}), 4.87 (ddt, *J* = 10.0, 2.0, 0.8 Hz, 2H, H_{15}), 3.12 (dd, *J* = 13.5, 7.1 Hz, 2H, H_{10}), 2.62 (ddt, *J* = 13.5, 7.6, 1.0 Hz, 2H, H_{13}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 180.59 (C8), 141.61 (C1), 133.15 (C3), 132.83 (C5), 132.30 (C11+C14), 118.75 (C12+C15), 114.32 (C2), 111.92 (C6), 109.96 (C4), 53.41 (C7), 41.32 (C10+C12).

1,3,3-triallyl-4-bromoindolin-2-one **(5f), Yield: 30 %**

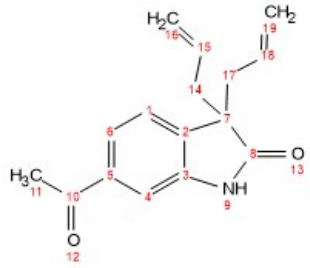


^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.25 – 7.00 (m, 3H, H_5+H_6), 6.72 (dd, *J* = 7.6, 1.1 Hz, 1H, H_4), 5.76 (ddt, *J* = 17.4, 10.1, 5.1 Hz, 1H, H_{12}), 5.29 (m, 2H, H_{16}+H_{18}), 5.19 (m, 2H, H_{13}), 5.04 (ddt, *J* = 17.0, 2.2, 1.2 Hz, 2H, H_{17}), 4.91 – 4.78 (m, 2H, H_{19}), 3.13 (ddt, *J* = 13.4, 7.1, 1.0 Hz, 1H, H_{11}), 2.64 (ddt, *J* = 13.5, 7.6, 1.0 Hz, 1H, H_{15}+H_{14}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 177.81 (C8), 145.19 (C3), 132.03 (C16+C18), 131.15 (C12), 129.33 (C5), 128.63 (C2), 126.45 (C6), 119.08 (C1), 118.60 (C17+C19), 117.49 (C13), 107.75 (C4), 55.87 (C7), 42.23 (C11), 38.68 (C14+C15).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{17}H_{19}NO$: 332.0572; Found 332.0650.

6-acetyl-3,3-diallylindolin-2-one **(4g), Yield: 73 %**

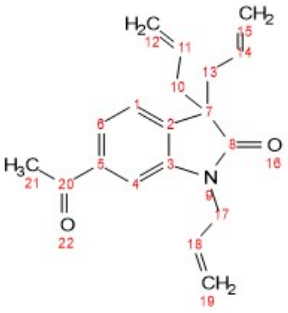


^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.78 (dd, *J* = 7.8, 1.5 Hz, 1H, H_{16}), 7.69 (s, 1H, H_4), 7.51 (dd, *J* = 1.5, 0.6 Hz, 1H, H_1), 5.56 – 5.31 (m, 2H, H_{15}+H_{18}), 5.09 – 4.87 (m, 4H, H_{16}+$_{19}$), 3.92 (d, *J* = 2.1 Hz, 2H, H_{11}), 2.60 (m, 2H, H_{14}+H_{17}).

^{13}C NMR (75 MHz, $CDCl_3$) ^{13}C NMR (75 MHz, $CDCl_3$) δ 180.76 (C10), 166.73 (C8), 141.11 (C3), 137.12 (C5), 131.60 (C15 + C18), 130.09 (C1), 124.11 (C6), 123.57 (C4), 119.34 (C16 + C19), 110.31 (C2), 53.54 (C7), 52.26 (C14 + C17), 41.13 (C11).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{16}H_{17}NO_2$: 256.1259; Found 256.1338.

6-acetyl-1,3,3-triallylindolin-2-one **(5g), Yield: 17 %**

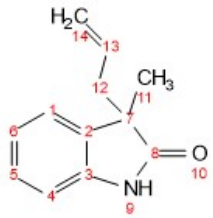


^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.72 (dd, *J* = 7.7, 1.4 Hz, 1H, H_6), 7.35 (d, *J* = 1.5 Hz, 1H, H_4), 7.29 – 7.11 (m, 1H, H_1), 5.71 (ddt, *J* = 17.4, 10.1, 5.0 Hz, 1H, H_{10}), 5.30 (ddt, *J* = 17.3, 10.1, 7.3 Hz, 2H, H_{11}+H_{14}), 5.18 – 5.06 (m, 2H, H_{19}), 4.97 – 4.75 (m, 4H, H_{12}+H_{15}), 4.28 (dt, *J* = 5.2, 1.8 Hz, 2H, H_{17}), 3.84 (s, 3H, H_{21}), 2.66 – 2.38 (m, 4H, H_{10}+H_{13}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 178.17 (C20), 166.78 (C8), 143.27 (C3), 136.62 (C5), 131.72 (C12+C15), 130.99 (C19), 130.04 (C1), 124.09 (C6), 123.21 (C4), 119.30 (C11+C14), 117.50 (C18), 109.34 (C2), 52.99 (C7), 52.25 (C10+C13), 42.13 (C17), 41.24 (C21).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{19}H_{21}NO_2$: 296.1572; Found 296.1651.

3-allyl-3-methylindolin-2-one **(4h), Yield: 85 %**

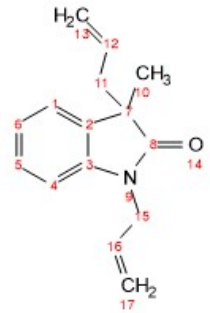


^{1}H NMR (300 MHz, Chloroform-*d*) δ 9.25 – 8.91 (m, 1H, H_9), 7.23 – 7.12 (m, 2H, H_1+H_6), 7.03 (td, *J* = 7.5, 1.1 Hz, 1H, H_5), 6.93 (d, *J* = 7.5Hz, 1H, H_4), 5.52 (dddd, *J* = 17.1, 10.1, 7.6, 7.0 Hz, 1H, H_{13}), 5.08 – 4.87 (m, 2H, $H_1$4), 2.69 – 2.42 (m, 2H, H_{12}), 1.41 (s, 3H, H_{11}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 183.13 (C8), 140.45 (C3), 134.09 (C2), 132.48 (C13), 127.76 (C5), 123.19 (C6), 122.30 (C14), 118.76 (C1), 109.90 (C4), 48.84 (C7), 42.39 (C12), 22.75 (C11).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{12}H_{13}NO$; 188.0997; Found 188.0919.

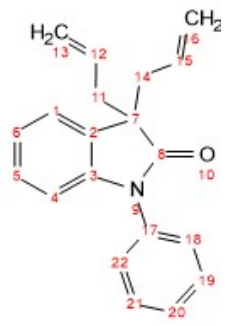
1,3-diallyl-3-methylindolin-2-on**e (5h), Yield: 10 %**[2]



^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.30 – 7.12 (m, 2H, H_1+H_6), 7.12 – 6.96 (m, 1H, H_5), 6.81 (dt, *J* = 7.4, 1.0 Hz, 1H, H_4), 5.81 (ddt, *J* = 17.4, 10.3, 5.1 Hz, 1H, H_{16}), 5.45 (ddd, *J* = 17.0, 10.1, 7.7, 6.9 Hz, 1H, H_{12}), 5.27 – 5.13 (m, 2H, H_{17}), 5.05 – 4.86 (m, 2H, H_{13}), 4.51 – 4.16 (m, 2H, H_{15}), 2.67 – 2.45 (m, 2H, H_{11}), 1.39 (s, 3H, H_{10}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 179.87 (C8), 142.34 (C3), 133.55 (C2), 132.60 (C12), 131.59 (C16), 127.65 (C1), 122.92 (C14), 122.30 (C17), 118.75 (C5), 117.24 (C6), 108.87 (C4), 48.25 (C7), 42.52 (C11), 42.13 (C15), 23.04 (C14).

3,3-diallyl-N-phenylindolin-2-one **(4i), Yield: 82 %**

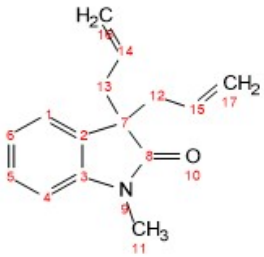


^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.55 – 7.03 (m, 8H, H_{Ar}), 6.77 (dt, *J* = 7.9, 0.8 Hz, 1H, H_6), 5.51 (dddd, *J* = 17.0, 10.1, 7.8, 6.8 Hz, 2H, H_{12}+H_{15}), 5.15 – 4.90 (m, 4H, H_{13}+H_{16}), 2.67 (qdt, *J* = 13.5, 6.8, 1.1 Hz, 4H, H_{11}+H_{14}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 178.35 (C8), 143.86 (C3), 134.66 (C2), 132.24 (C12+C15), 131.00 (17), 129.54 (C19+C21), 127.77 (C5), 127.77 (C20), 126.75 (C18+C22), 123.59 (C6), 122.72 (C1), 119.00 (C13+C16), 109.09 (C4), 52.77 (C7), 41.62 (C11+C14).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{20}H_{19}NO$: 290.1467; Found 290.1545.

3,3-diallyl-N-methylindolin-2-one **(4j), Yield: 51 %**

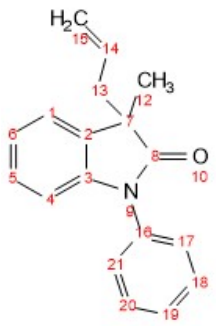


^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.34 – 7.16 (m, 2H, H_1+H_6), 7.06 (td, *J* = 7.5, 1.0 Hz, 1H, H_5), 6.81 (dt, *J* = 7.7, 0.8 Hz, 1H, H_4), 5.41 (ddt, *J* = 17.3, 10.1, 7.3 Hz, 2H, H_{14}+H_{15}), 5.07 – 4.81 (m, 2H, H_{16}+H_{17}), 3.18 (s, 3H, H_{11}), 2.65 – 2.45 (m, 2H,H_{12}+H_{13}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 178.90 (C8), 143.76 (C3), 132.31 (C13+C16), 131.33 (C2), 127.85 (C5), 123.37 (C6), 122.24 (C1), 118.70 (C14+C17), 107.79 (C4), 52.65 (C7), 41.21 (C11+C12), 26.02 (C15).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{15}H_{17}NO$: 228.1310; Found 228.1388.

3-allyl-3-methyl-1-phenylindolin-2-one **(4k), Yield: 100 %**



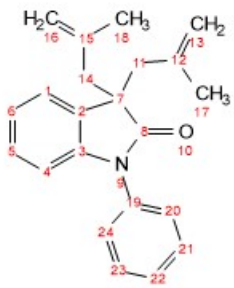
^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.47 – 7.01 (m, 8H, H_{Ar}), 5.44 (dd, *J* = 16.9, 10.1, 1H, H_{14}), 5.04 – 4.84 (m, 2H, H_{15}), 2.67 – 2.39 (m, 2H, H_{13}), 1.42 (s, 3H, H_{12}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 179.58 (C8), 143.20 (C3), 134.68 (C2), 133.32 (C15), 132.51 (C13), 129.55 (C17+C19), 127.94 (C6), 127.69 (C5),

126.67 (C16+C20), 123.16 (C18), 122.83 (C14), 118.93 (C1), 109.23 (C4), 48.41 (C7), 43.07 (C12), 23.03 (C11).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{18}H_{17}NO$; 264.1310; Found 264.1388.

3,3-bis(2-methylallyl)-1-phenylindolin-2-one **(5ic), Yield: 70 %**

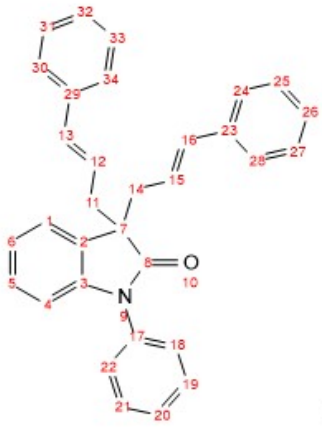


^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.55 – 7.00 (m, 2H, H_{Ar}), 6.81 – 6.65 (m, 1H, H_6), 4.71 – 4.57 (m, 4H, H_{13}+H_{16}), 2.85 (dd, *J* = 13.3, 1.0 Hz, 2H, $H_{11,14}$), 2.63 – 2.48 (m, 2H, $H_{11,14}$),), 1.40 (t, *J* = 1.2 Hz, 1H, H_{17}+H_{16}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 178.48 (C8), 144.12 (C3), 140.78 (C12+C15), 134.83 (C2), 131.15 (C19), 129.58 (C21+C23), 127.75 (C5), 127.92 (C22), 126.50 (C20+C24), 124.30 (C1), 122.37 (C6), 114.84 (C13+C16), 109.12 (C4), 53.31 (C7), 46.30 (C11+C14), 23.92 (C17+C18).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{12}H_{23}NO$: 318.1780; Found 318.1858.

3,3-dicinnamyl-1-phenylindolin-2-one **(5id), Yield: 65 %**



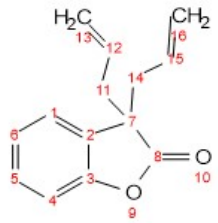
^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.44 – 7.05 (m, 18H, H_{Ar}), 6.77 – 6.58 (m, 1H, H_4), 6.42 (dd, *J* = 15.8, 1.2 Hz, 2H, H_{12}+H_{15}), 5.95 (ddd, *J* = 15.5, 8.1, 7.0 Hz, 2H, H_{13}+H_{16}), 2.98 – 2.77 (m, 4H, H_{11}+H_{14}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 178.455 (C8), 143.92 (C 3), 137.29 (C23+C29), 134.58 (C2), 134.14 (C12+C15), 131.06 (C17),

129.59 (C18+C22), 128.53 (C24+C28+C30+C34), 128.05 (C5), 127.34 (C32+C26), 126.88 (C19+C21), 126.24 (C25+C27+C31+C33), 123.91 (C13+C16), 123.68 (C6), 122.94 (C1), 109.34 (C4), 53.74 (C7), 40.74 (C11+C14).

HR-MS (ESI) m/z : Calcd for $[M+H]^+$ $C_{32}H_{27}NO$: 442.2093; Found 442.2171.

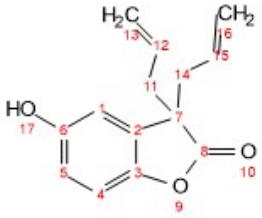
3,3-diallylbenzofuran-2(3H)-one **(3a'), Yield: 70 %**[3]



^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.22 – 7.02 (m, 4H, H_{Ar}), 5.47 (m, 2H, H_{12}+H_{15}), 5.21 – 4.94 (d, *J* = 16.9 Hz, 4H, H_{13}+H_{16}), 2.55 (m, 4H, H_{11}+H_{14}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 178.58 (C8), 152.97 (C3), 131.18 (C2), 129.41 (C12+C15), 128.78 (C1), 124.11 (C5), 123.65 (C6), 120.06 (C13+C16), 110.68 (C4), 52.37 (C7), 41.73 (C11+C14).

3,3-diallyl-5-hydroxybenzofuran-2(3H)-one **(3b'), Yield: 80 %**



^{1}H NMR (300 MHz, Chloroform-*d*) δ 7.00 – 6.89 (m, 1H, H_4), 6.81 – 6.68 (m, 2H, H_1+H_5), 5.47 (dddd, *J* = 16.9, 10.1, 7.7, 6.9 Hz, 2H, H_{12}+H_{15}), 5.21 – 4.94 (m, 5H, H_{13}+H_{16}+H_{17}), 2.75 – 2.48 (m, 4H, H_{11}+H_{14}).

^{13}C NMR (75 MHz, $CDCl_3$) δ 179.08 (C8), 152.51 (C6), 146.78 (C3), 131.08 (C12+C15), 130.64 (C2), 120.13 (C13+C16), 115.22 (C4), 111.24 (C1), 111.00 (C5), 53.07 (C7), 41.69 (C11+C14).

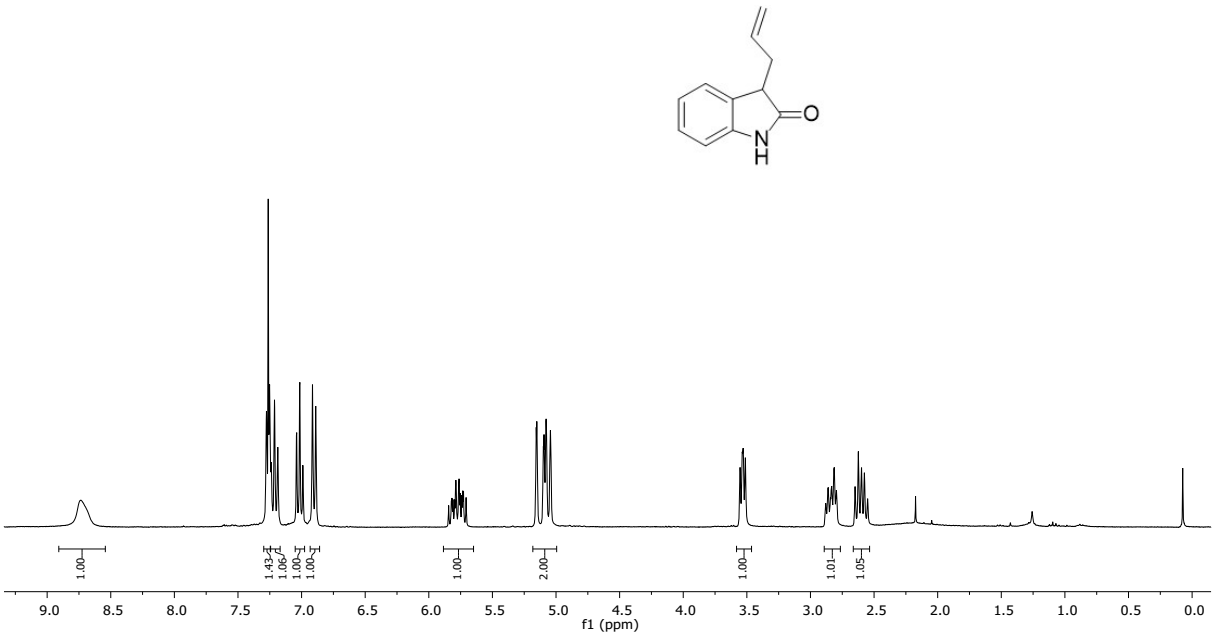
4- References

[1] B. M. Trost, Y. Zhang, T. Zhang, *J. Org. Chem.* **2009**, *74*, 14, 5115–5117.

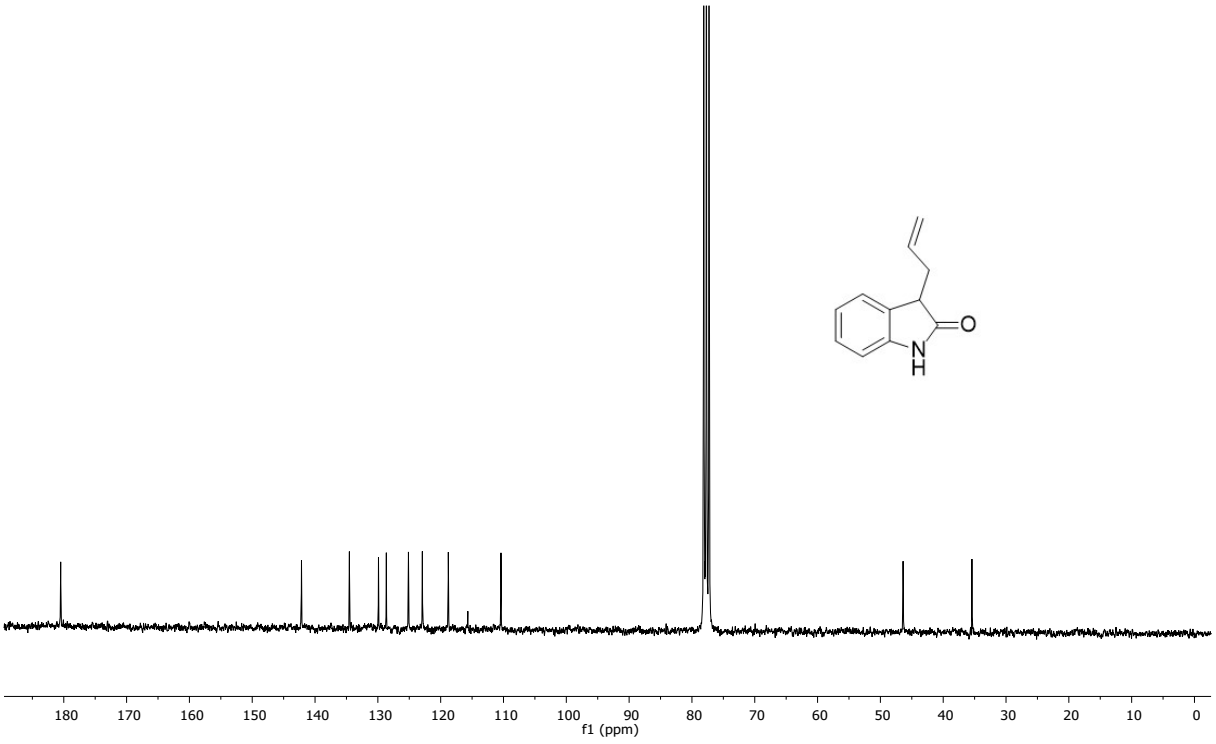
[2] N. Kumar, M. K. Das, S. Ghosh, A. Bisai, *Chem. Commun*. **2017**, *53*, 2170-2173.

[3] T.-Y. Zhao, K. Li, L.-L. Yang, S.-F. Zhu, Q.-L. Zhou, *Org. Lett.* **2021**, *23*, 3814−3817.

3-allylindolin-2-one (3a)

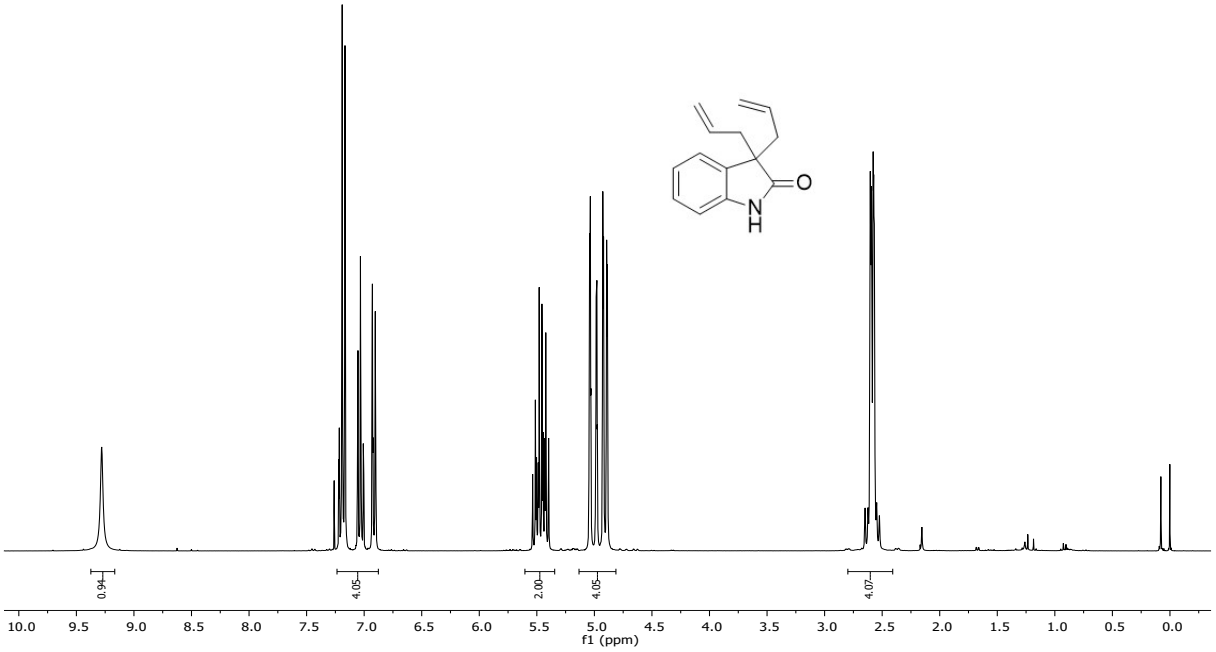


^{1}H NMR spectrum of 3-allylindolin-2-one (3a)

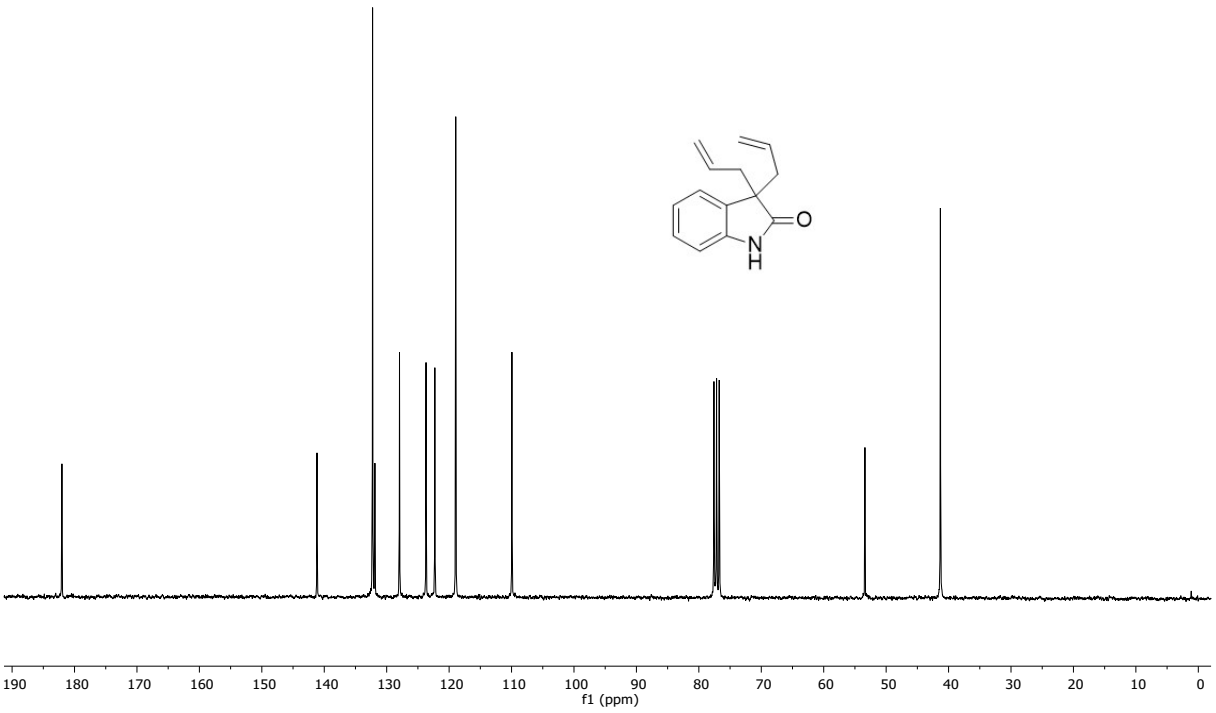


^{13}C NMR spectrum of 3-allylindolin-2-one (3a)

3,3-diallylindolin-2-one (4a)

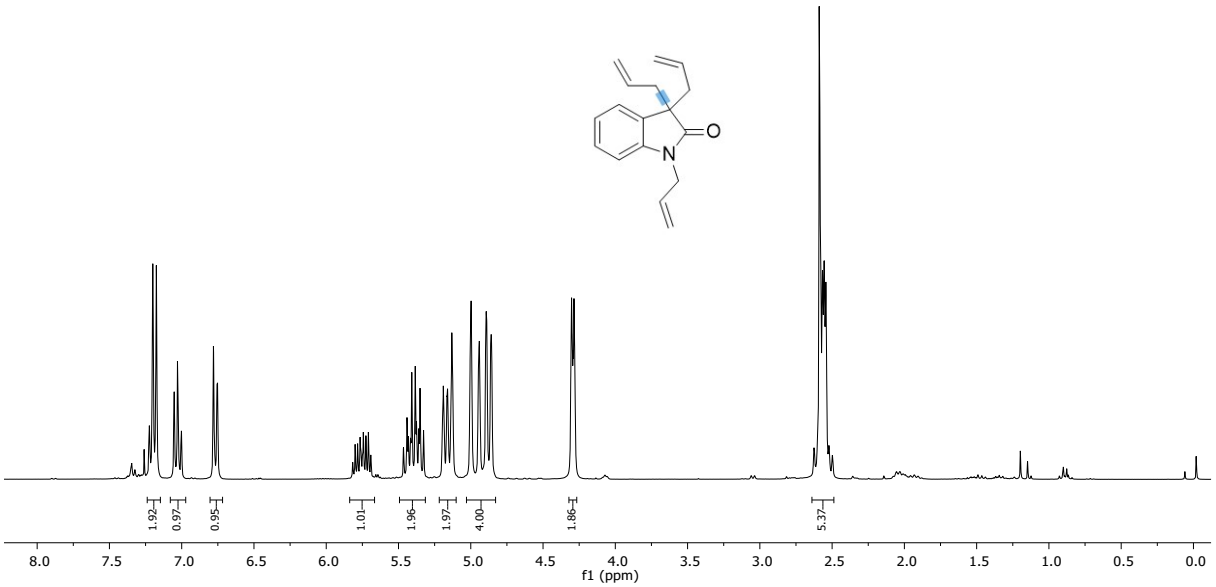


^{1}H NMR spectrum of 3,3-diallylindolin-2-one (4a)

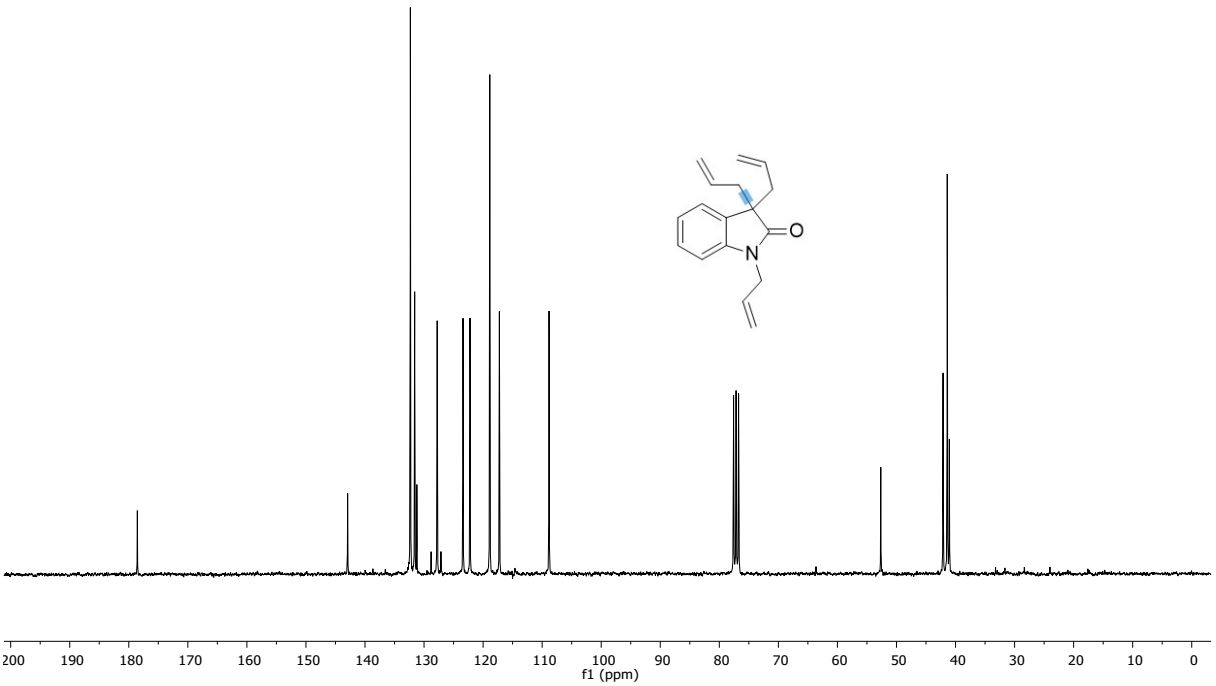


^{13}C NMR spectrum of 3,3-diallylindolin-2-one (4a)

1,3,3-triallylindolin-2-one (4a)

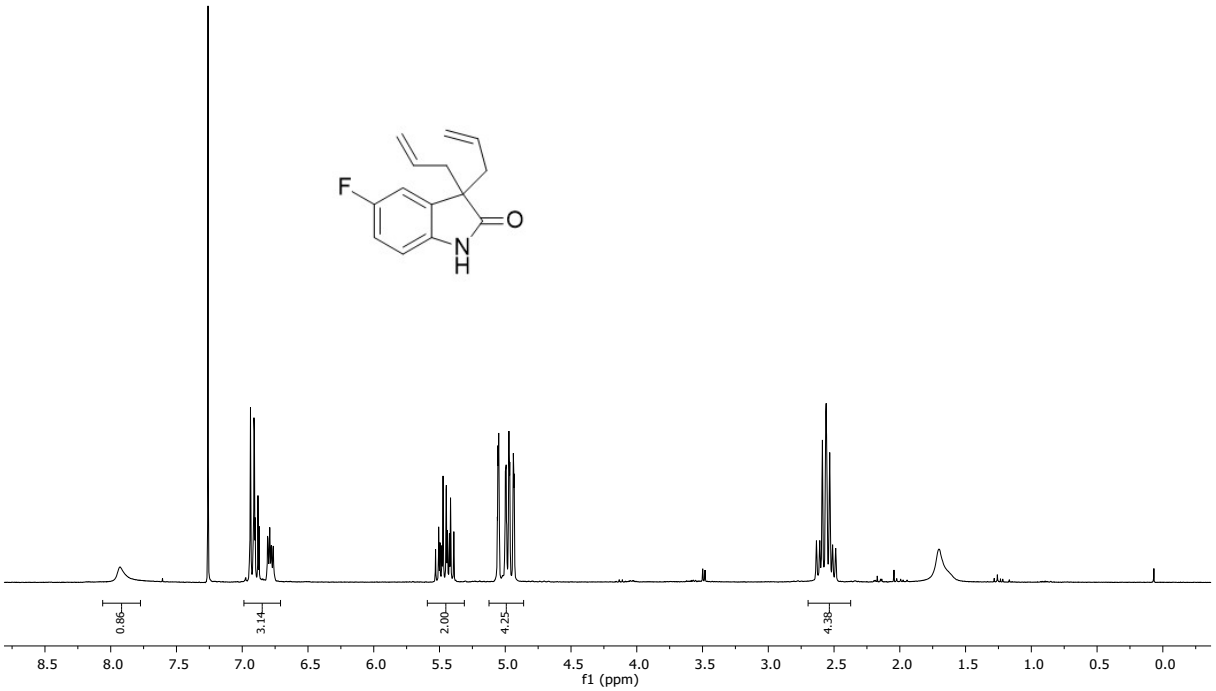


^{1}H NMR spectrum of 1,3,3-triallylindolin-2-one (5a)

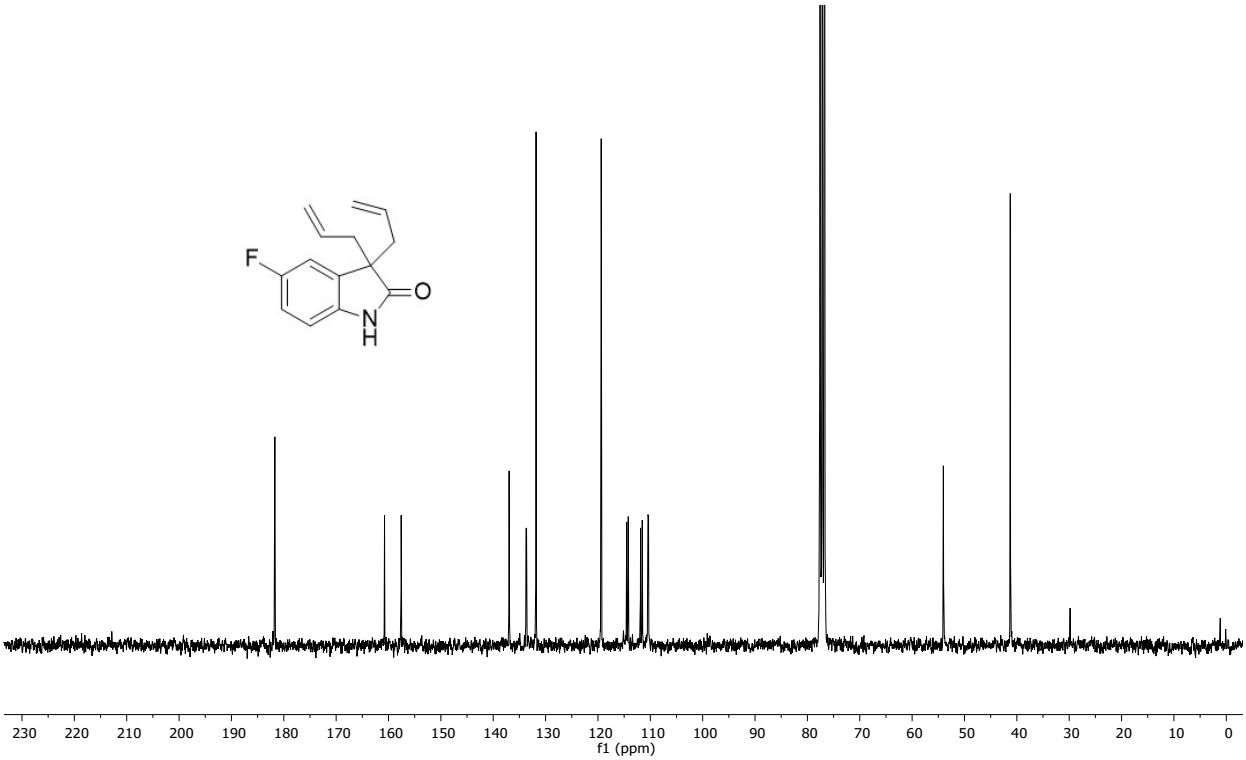


^{13}C NMR spectrum of 1,3,3-triallylindolin-2-one (5a)

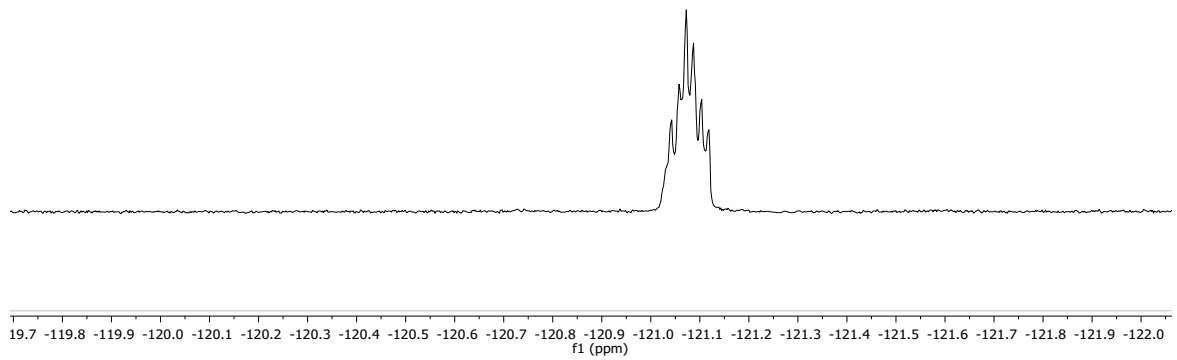
3,3-diallyl-5-fluoroindolin-2-one (4b)



^{1}H NMR spectrum of 3,3-diallyl-5-fluoroindolin-2-one (4b)

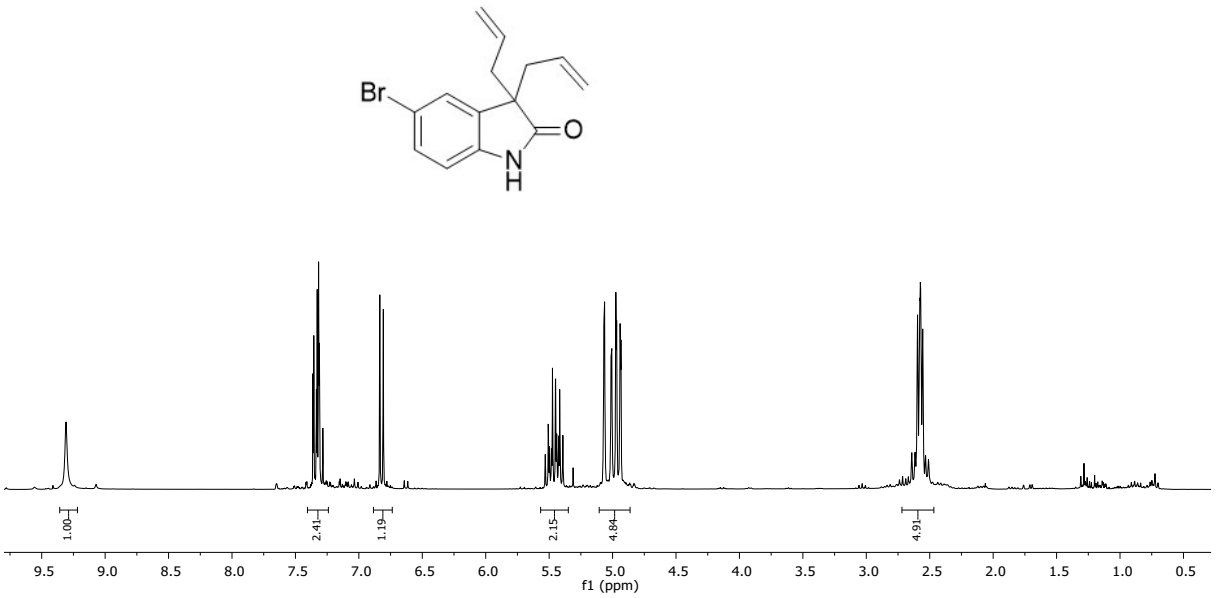


^{13}C NMR spectrum of 3,3-diallyl-5-fluoroindolin-2-one (4b)

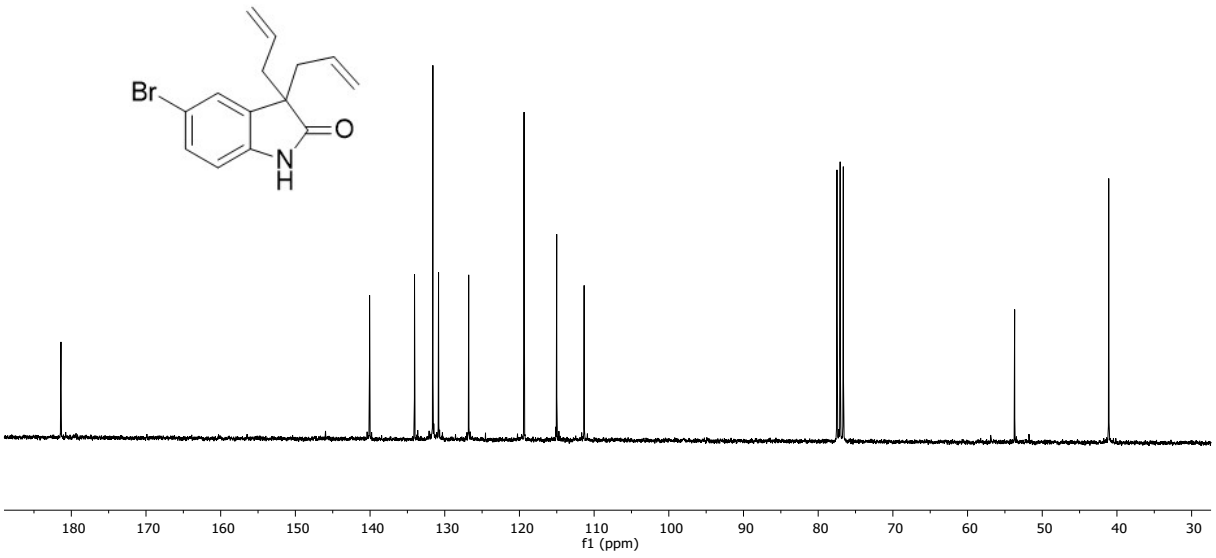


^{19}F NMR spectrum of 3,3-diallyl-5-fluoroindolin-2-one (4b)

3,3-diallyl-4-bromoindolin-2-one (4c)

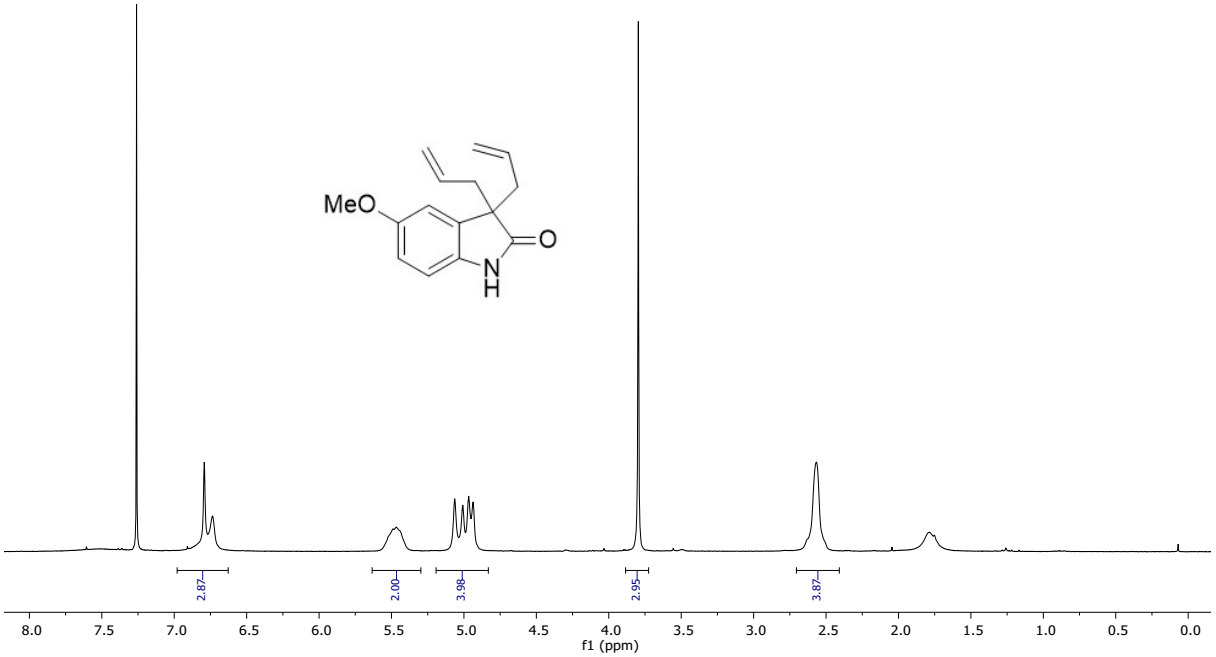


^{1}H NMR spectrum of 3,3-diallyl-4-bromoindolin-2-one (4c)

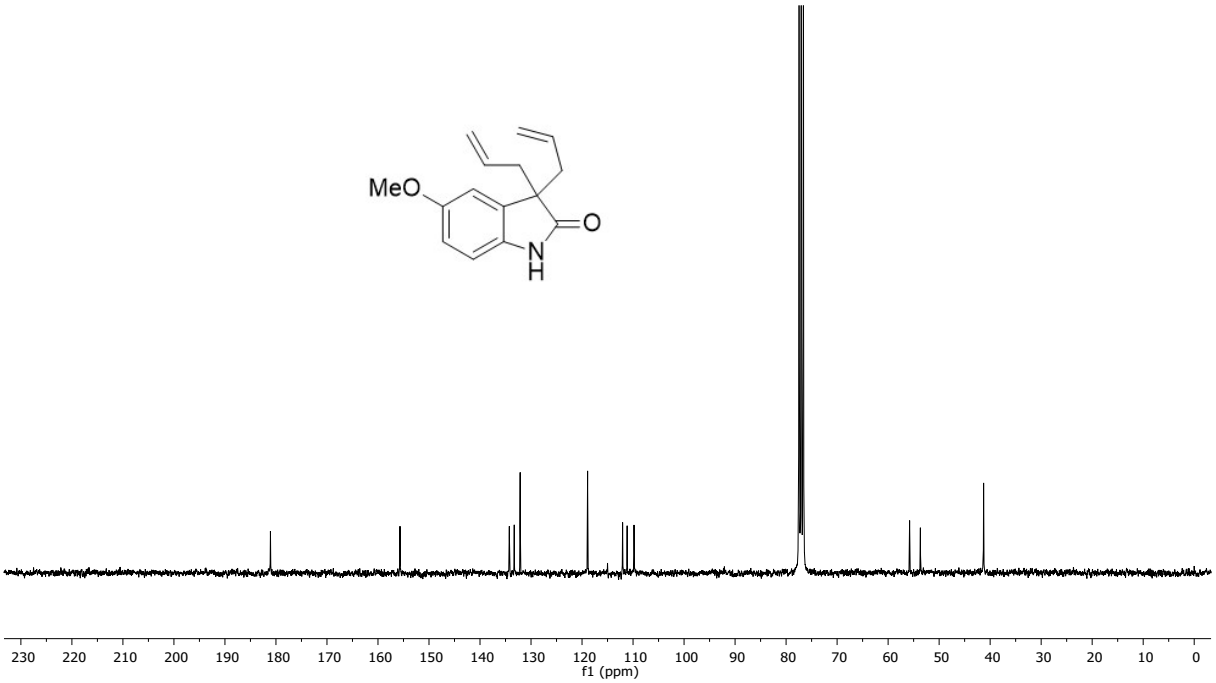


^{13}C NMR spectrum of 3,3-diallyl-4-bromoindolin-2-one (4c)

3,3-diallyl-5-methoxyindolin-2-one (4d)

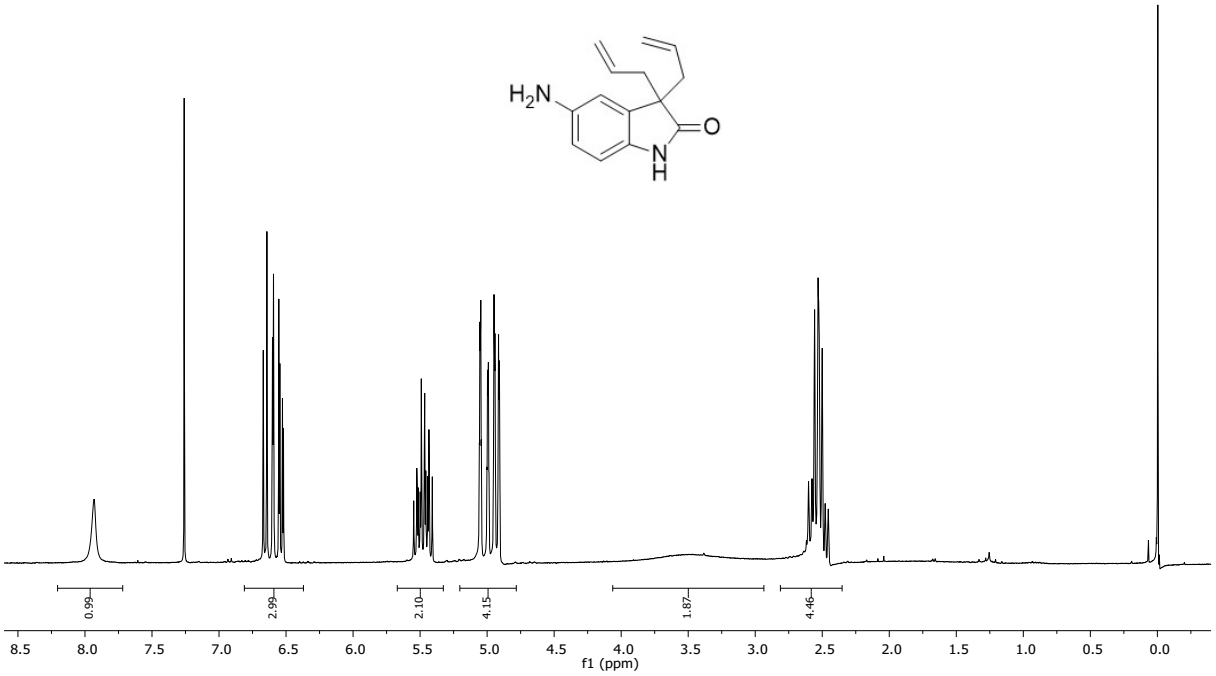


^{1}H NMR spectrum of 3,3-diallyl-5-methoxyindolin-2-one (4d)

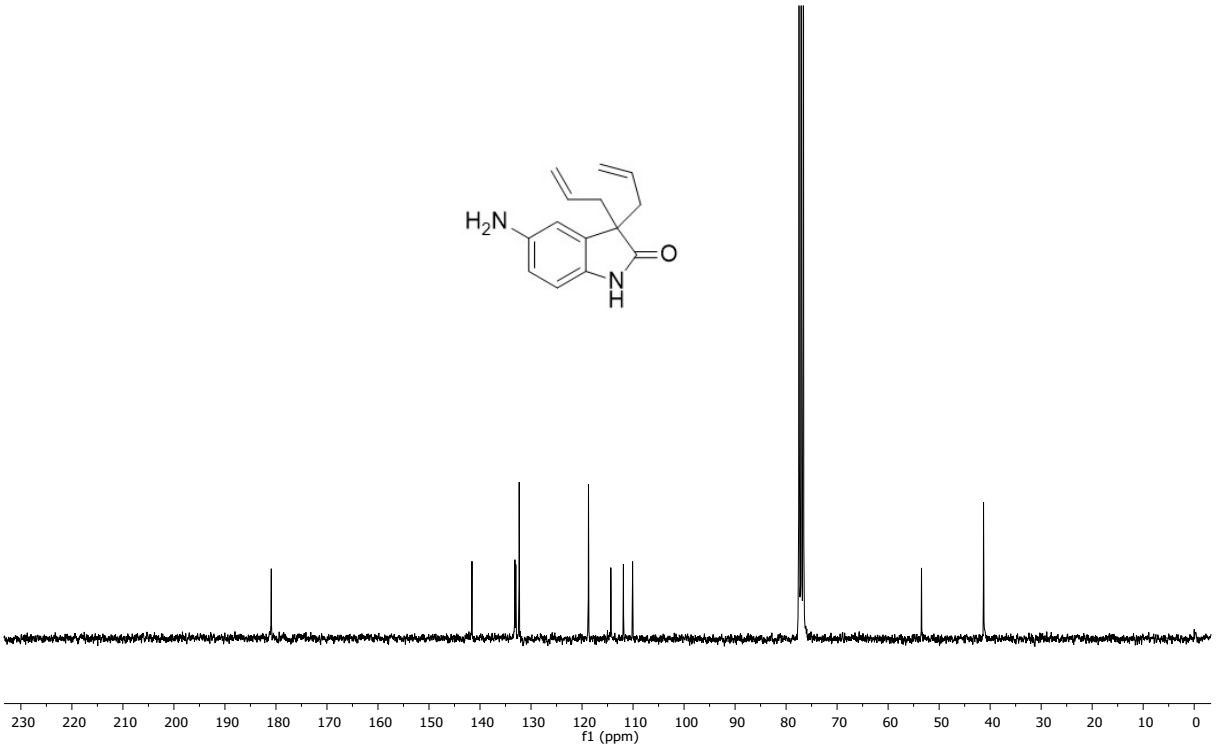


^{13}C NMR spectrum of 3,3-diallyl-5-methoxyindolin-2-one (4d)

3,3-diallyl-5-aminoindolin-2-one (4e)

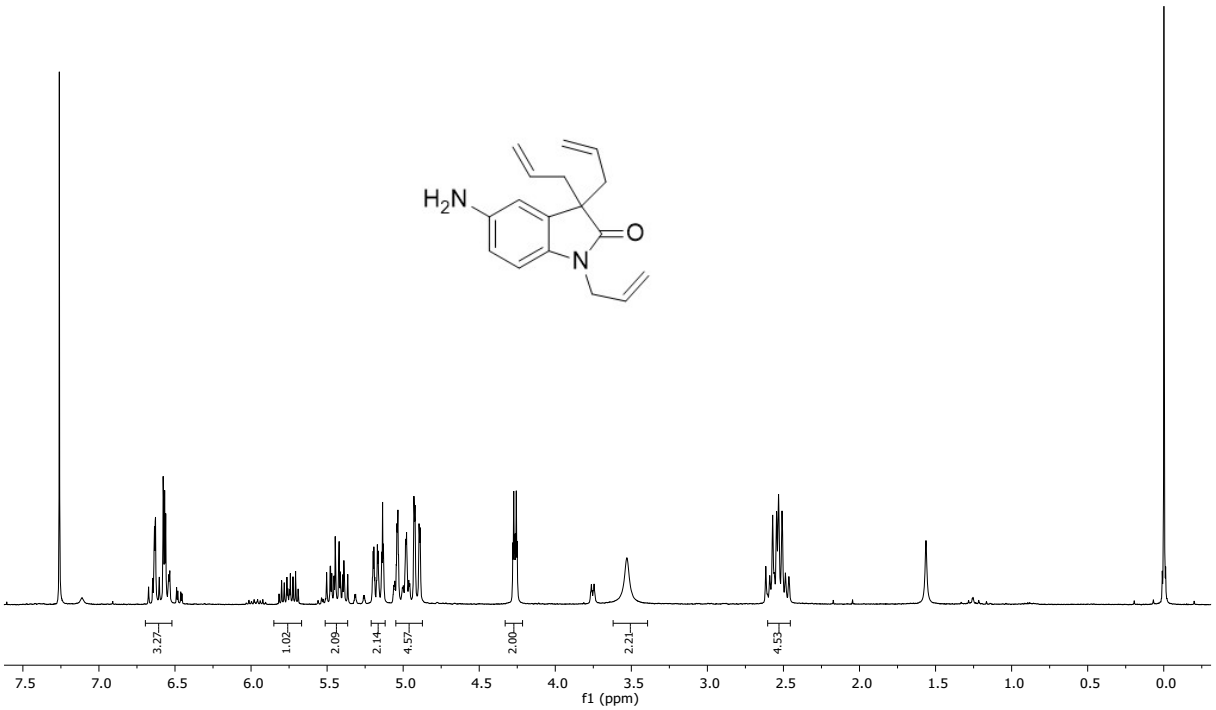


^{1}H NMR spectrum of 3,3-diallyl-5-aminoindolin-2-one (4e)

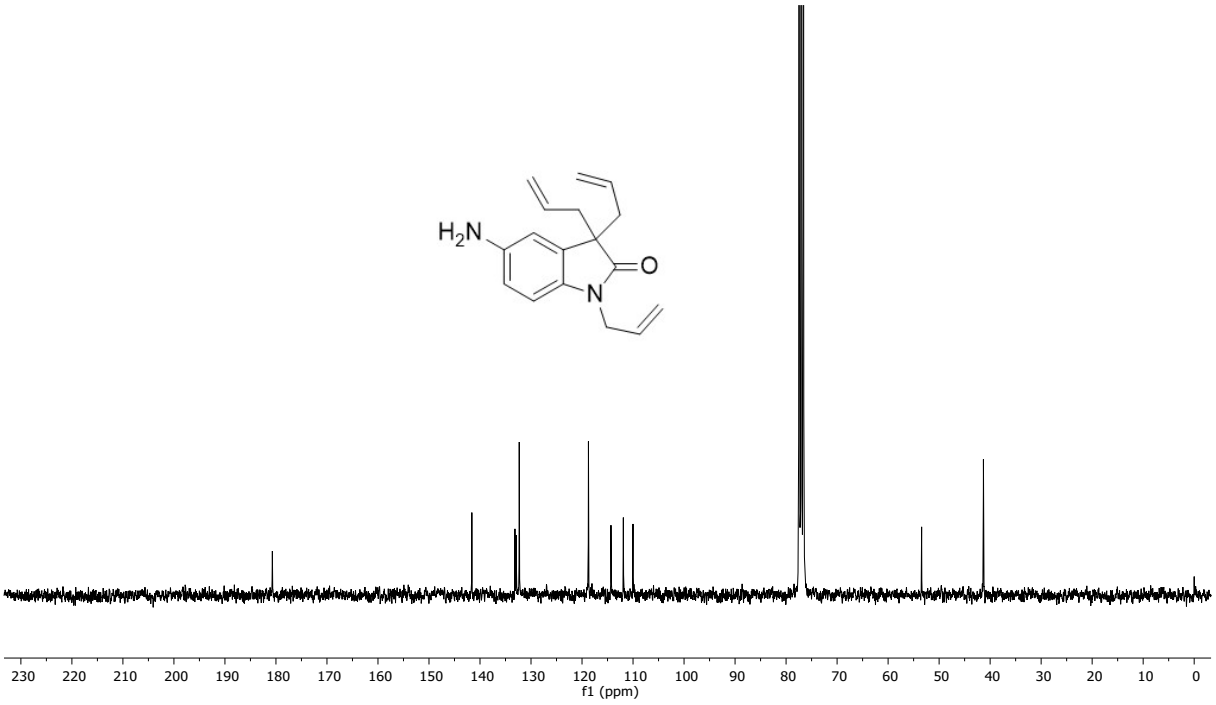


^{13}C NMR spectrum of 3,3-diallyl-5-aminoindolin-2-one (4e)

1,3,3-triallyl-5-aminoindolin-2-one (5e)

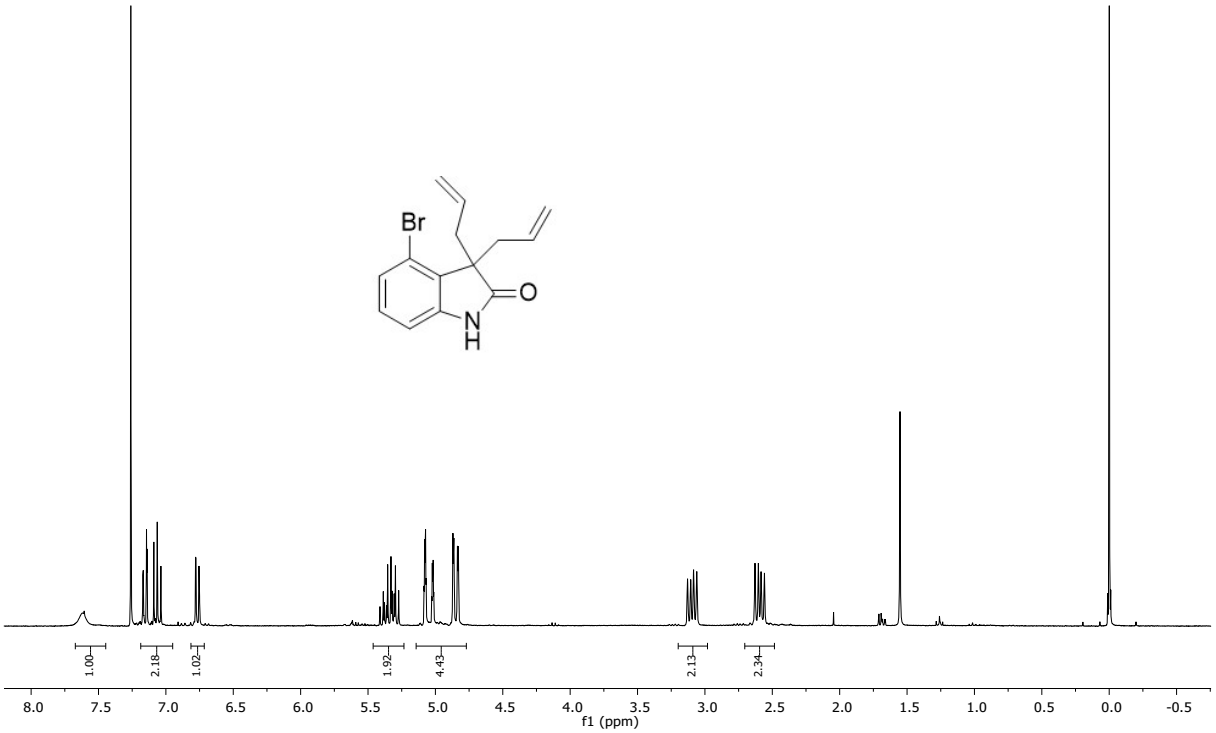


^{1}H NMR spectrum of 1,3,3-triallyl-5-aminoindolin-2-one (5e)

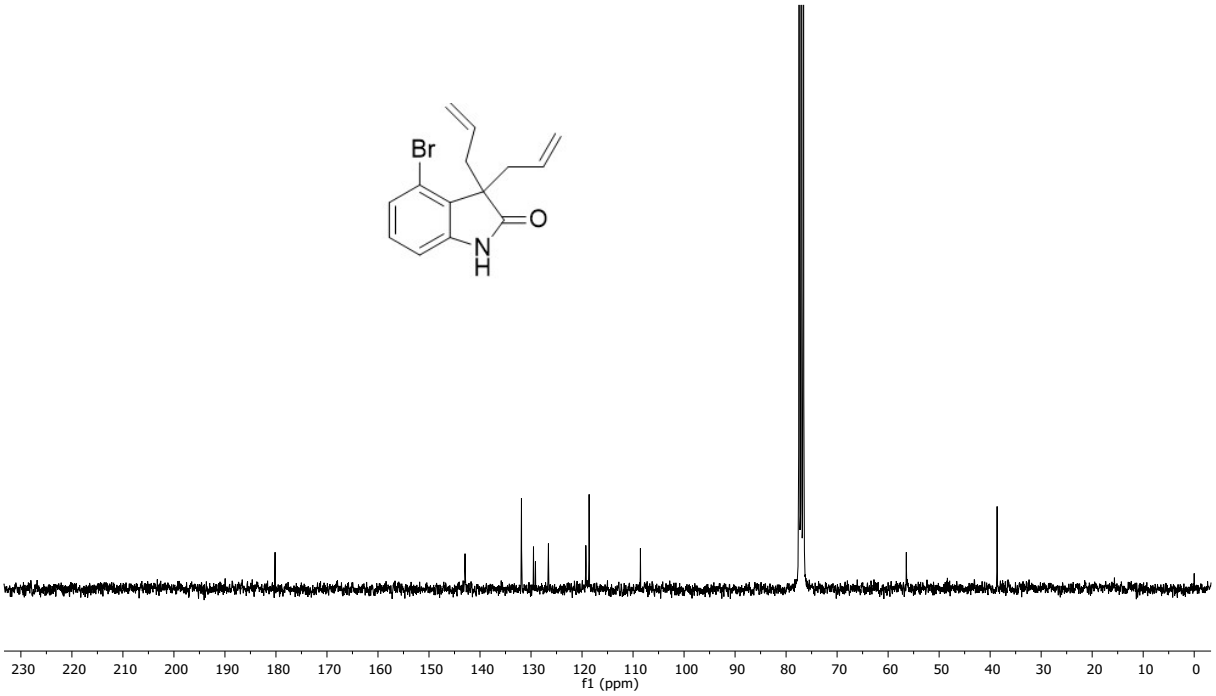


^{13}C NMR spectrum of 1,3,3-triallyl-5-aminoindolin-2-one (5e)

3,3-diallyl-4-bromoindolin-2-one (4f)

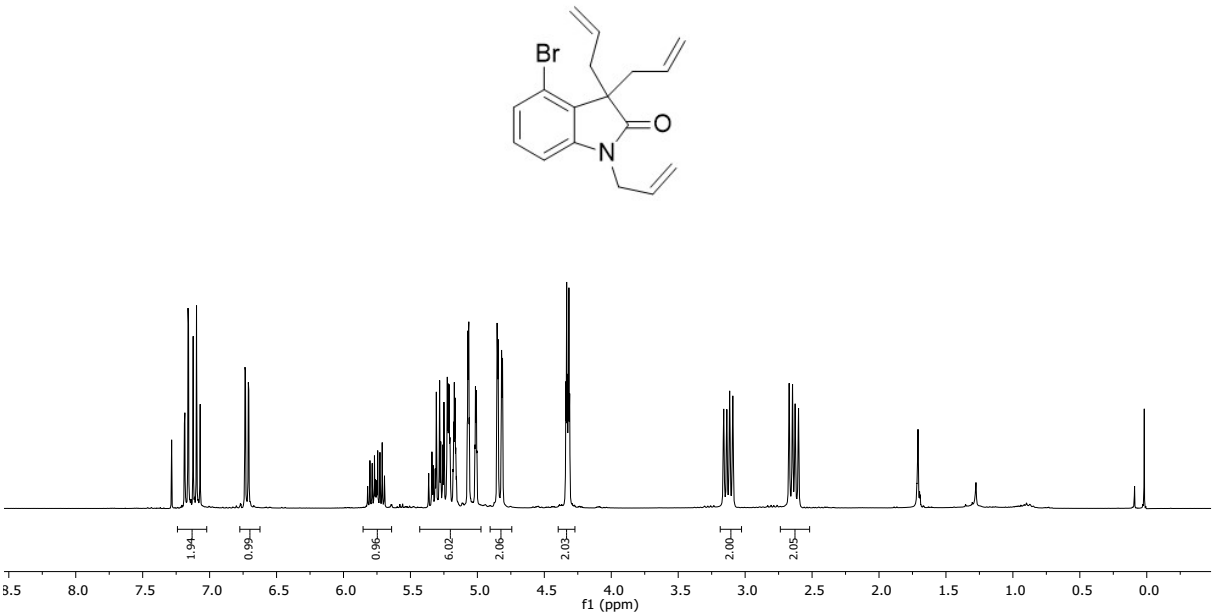


^{1}H NMR spectrum of 3,3-diallyl-4-bromoindolin-2-one (4f)

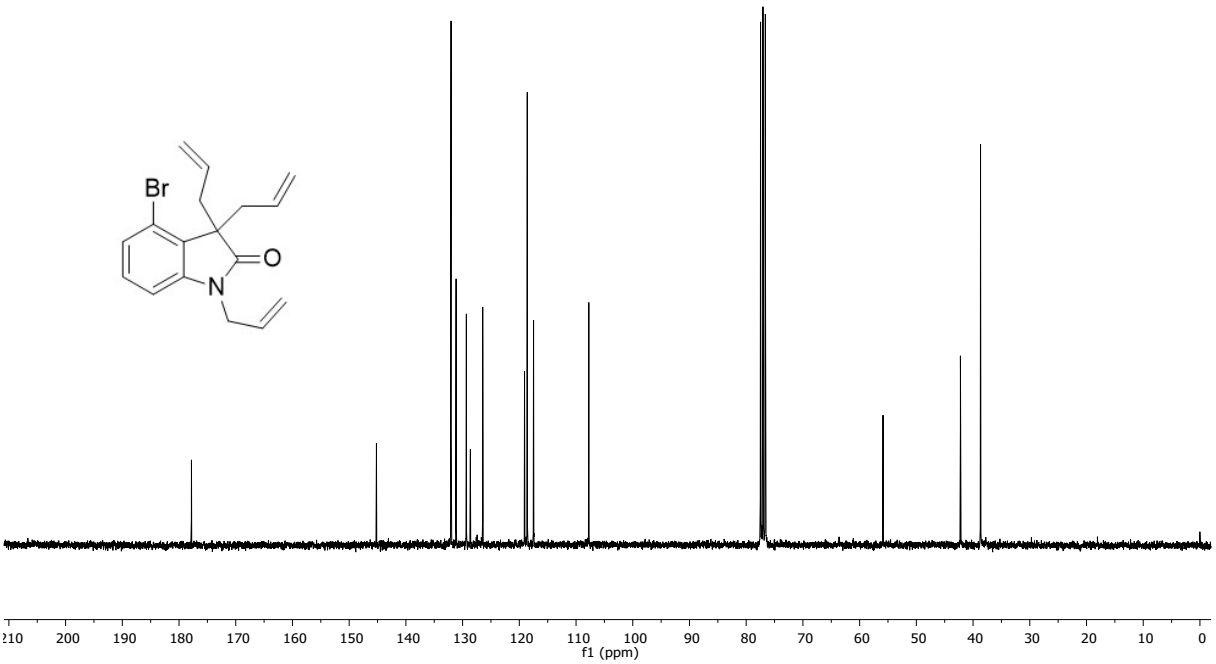


^{13}C NMR spectrum of 3,3-diallyl-4-bromoindolin-2-one (4f)

N,3,3-triallyl-4-bromoindolin-2-one (5f)

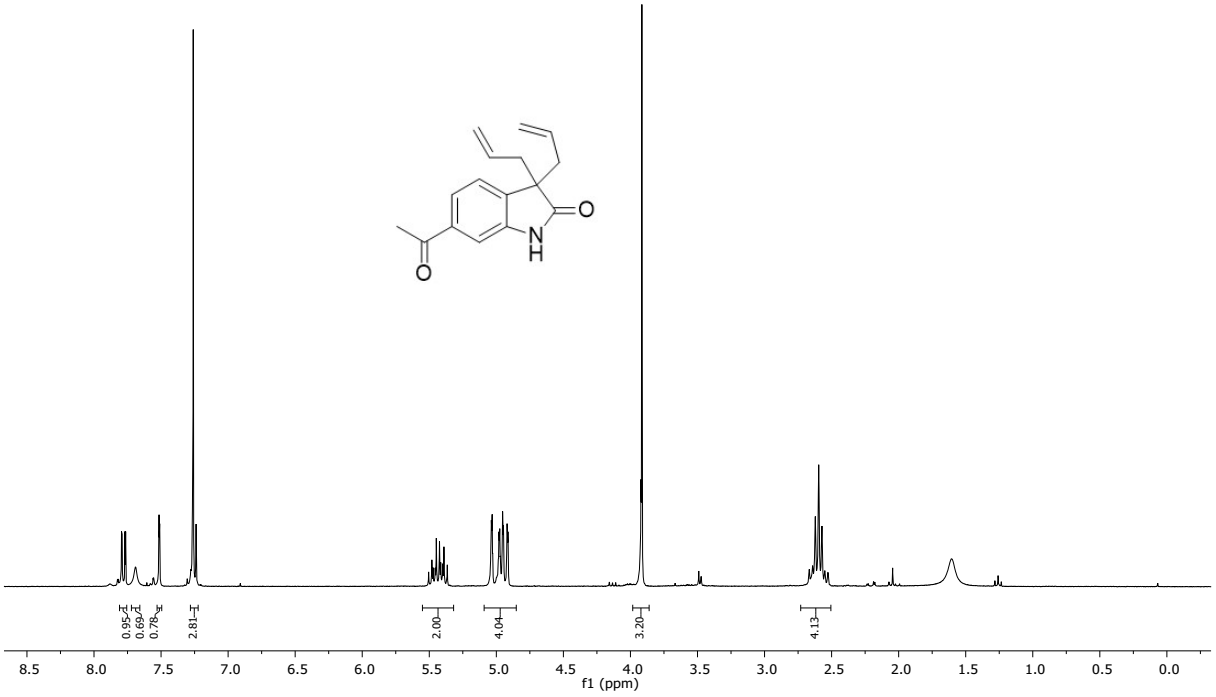


^{1}H NMR spectrum of N,3,3-triallyl-4-bromoindolin-2-one (5f)

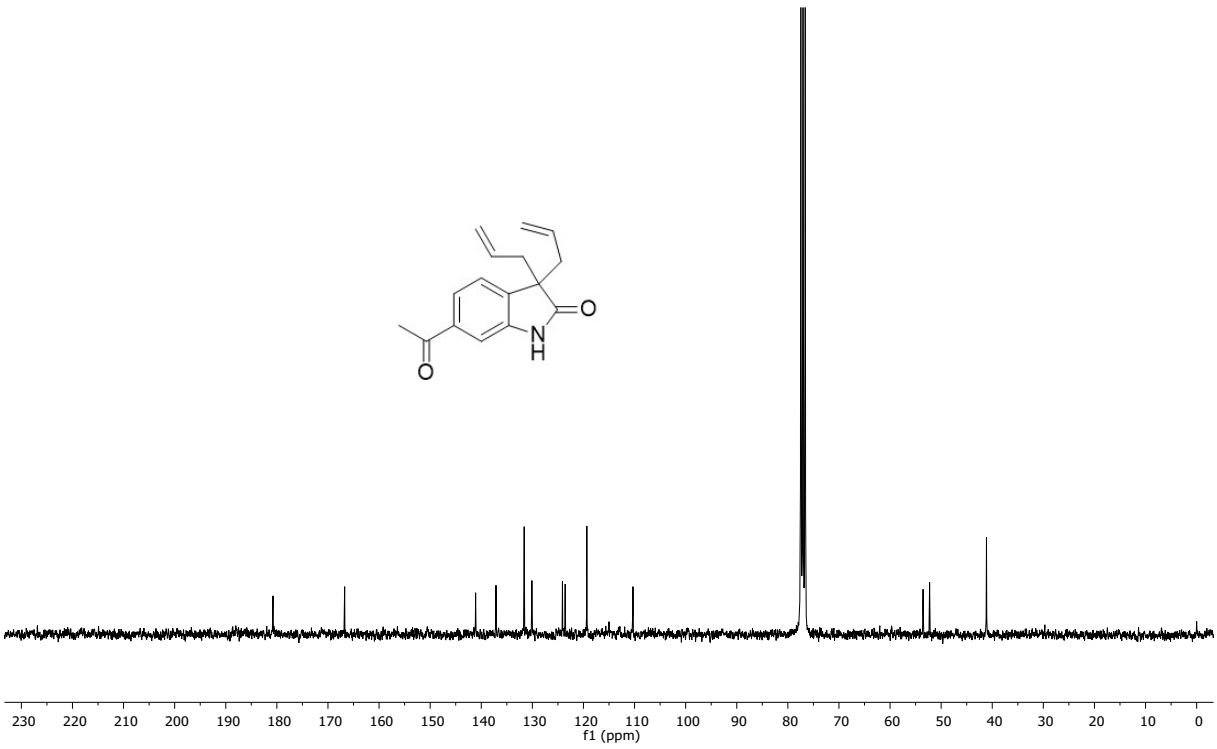


^{13}C NMR spectrum of N,3,3-triallyl-4-bromoindolin-2-one (5f)

6-acetyl-3,3-diallylindolin-2-one (4g)

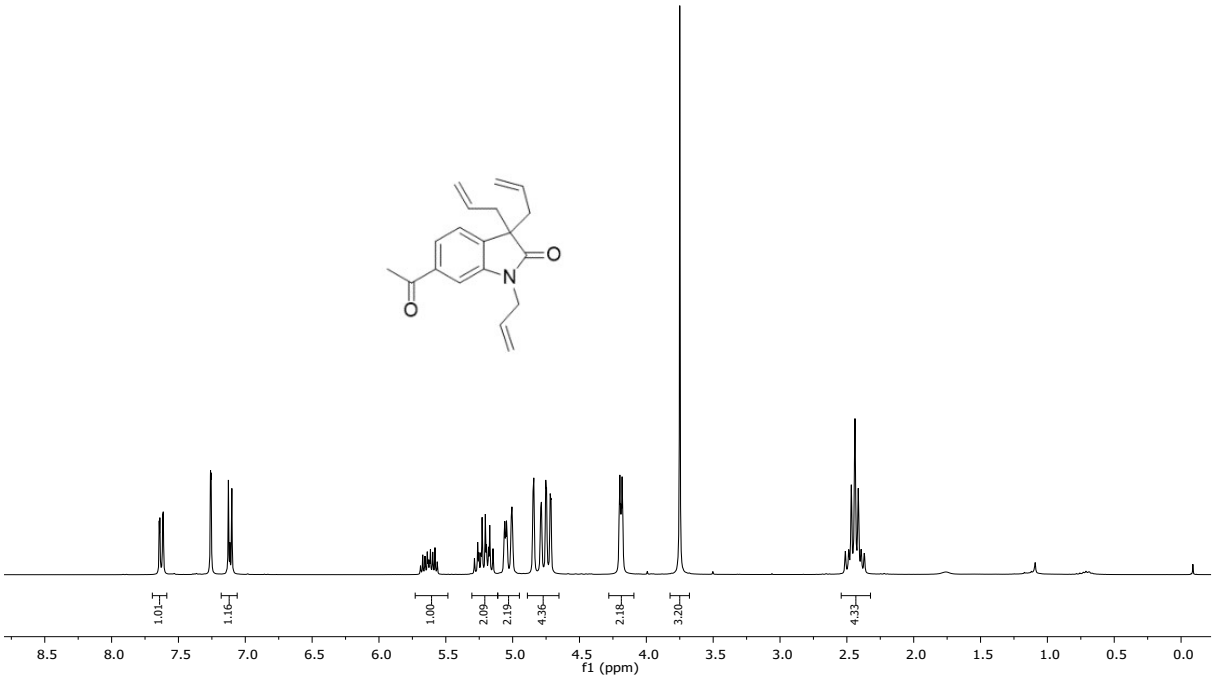


^{1}H NMR spectrum of 6-acetyl-3,3-diallylindolin-2-one (4g)

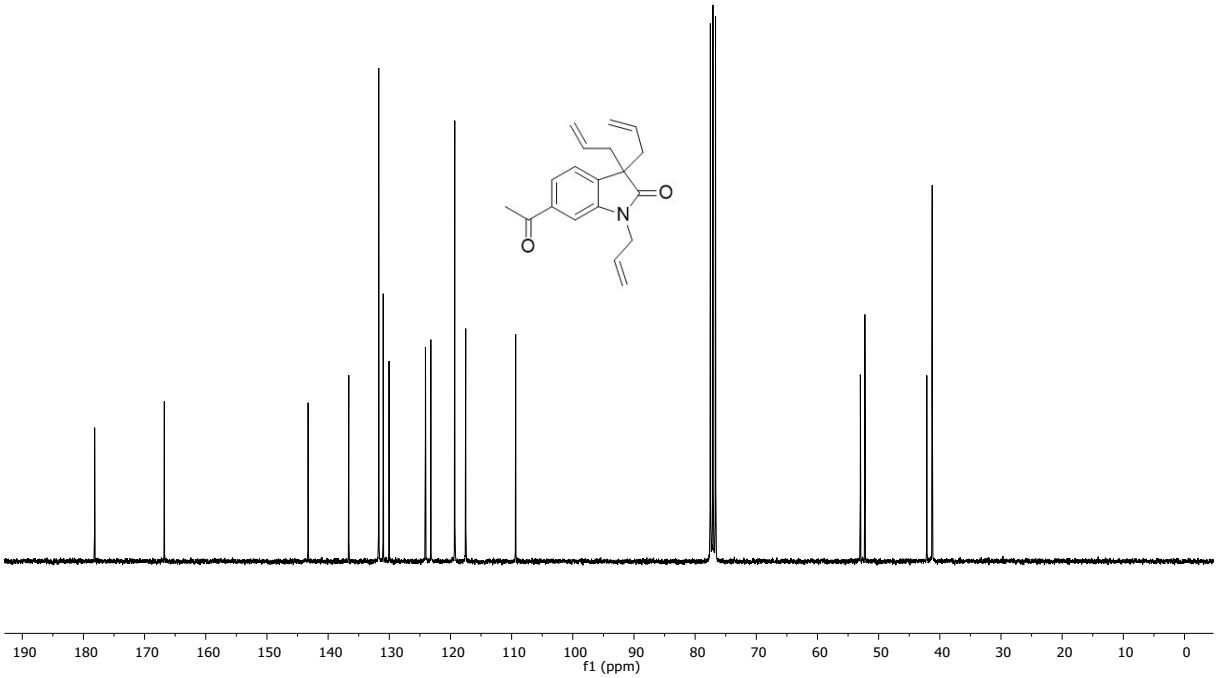


^{13}C NMR spectrum of 6-acetyl-3,3-diallylindolin-2-one (4g)

6-acetyl-1,3,3-triallylindolin-2-one (5g)

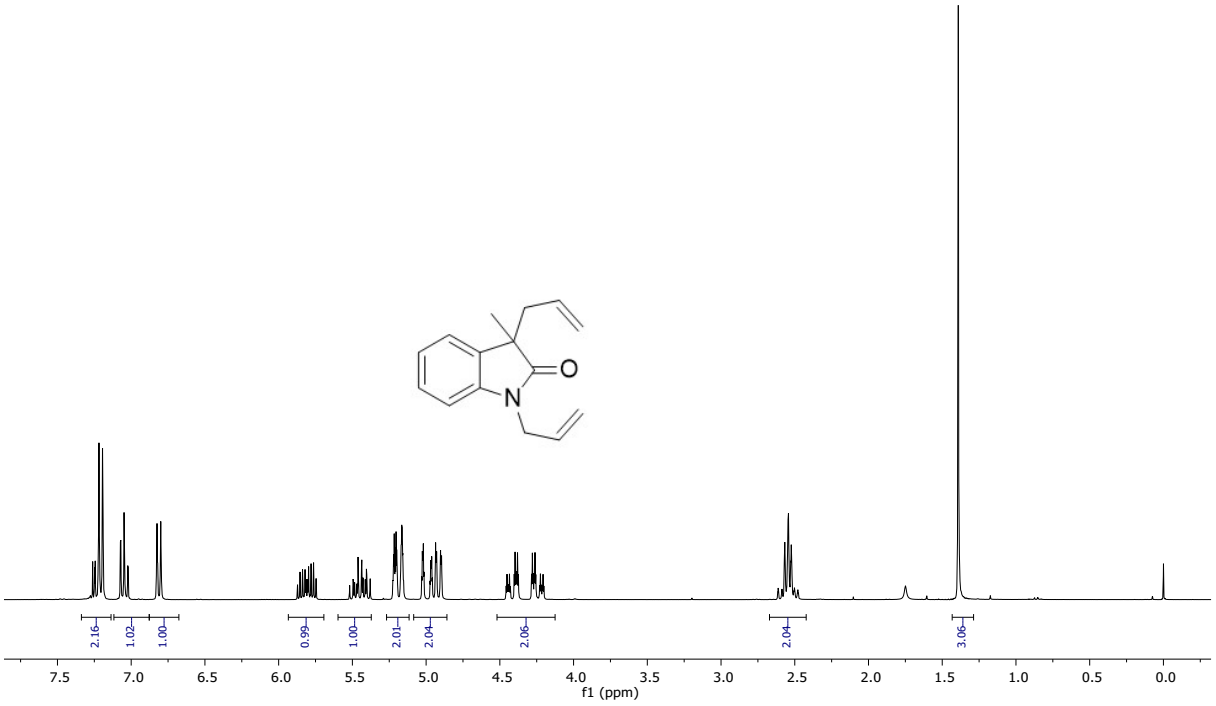


^{1}H NMR spectrum of 6-acetyl-1,3,3-triallylindolin-2-one (5g)

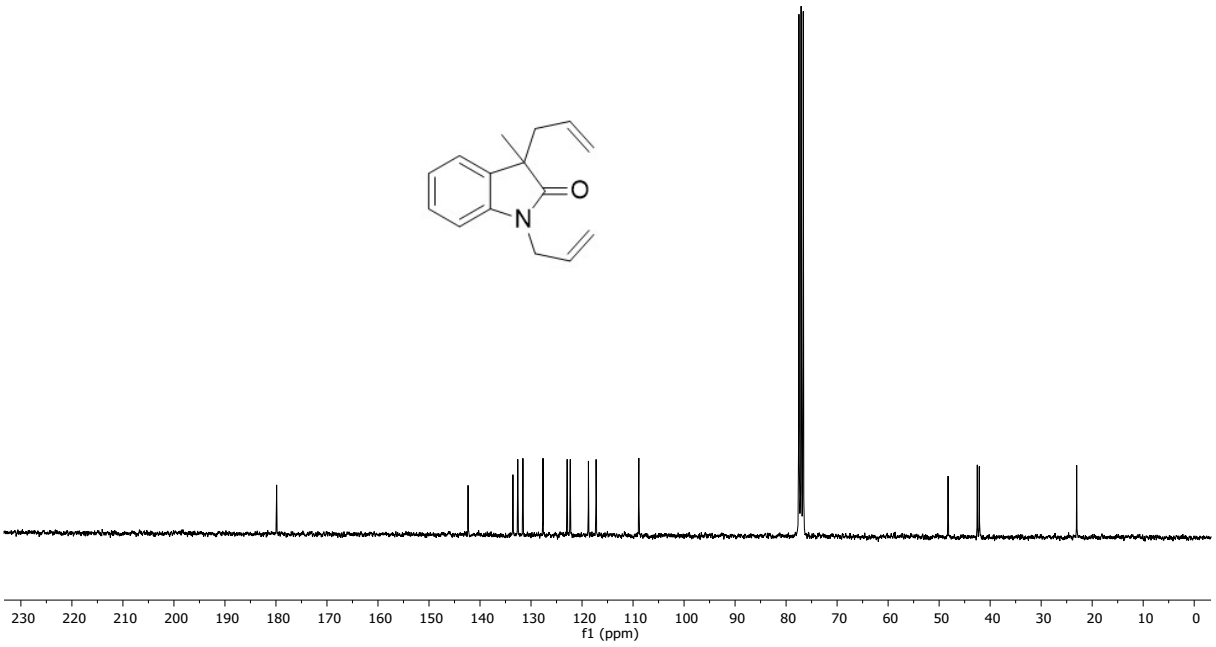


^{13}C NMR spectrum of 6-acetyl-1,3,3-triallylindolin-2-one (5g)

1,3-diallyl-3-methylindolin-2-one (5h)

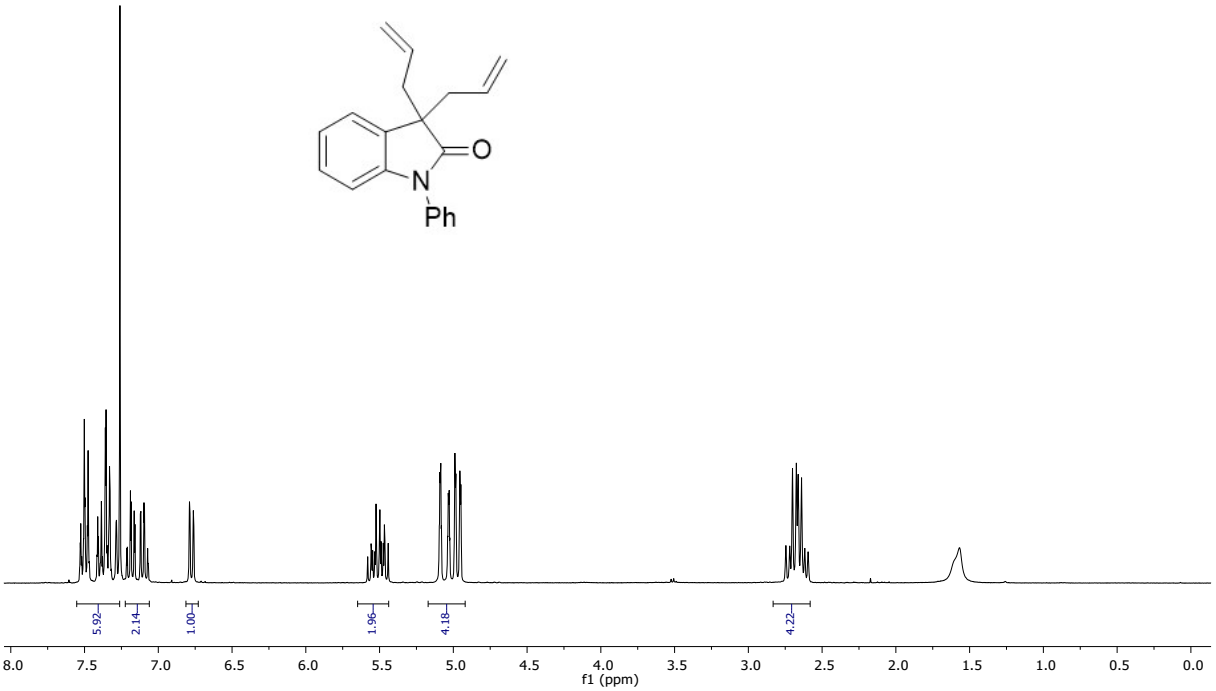


^{1}H NMR spectrum of 1,3-diallyl-3-methylindolin-2-one (5h)

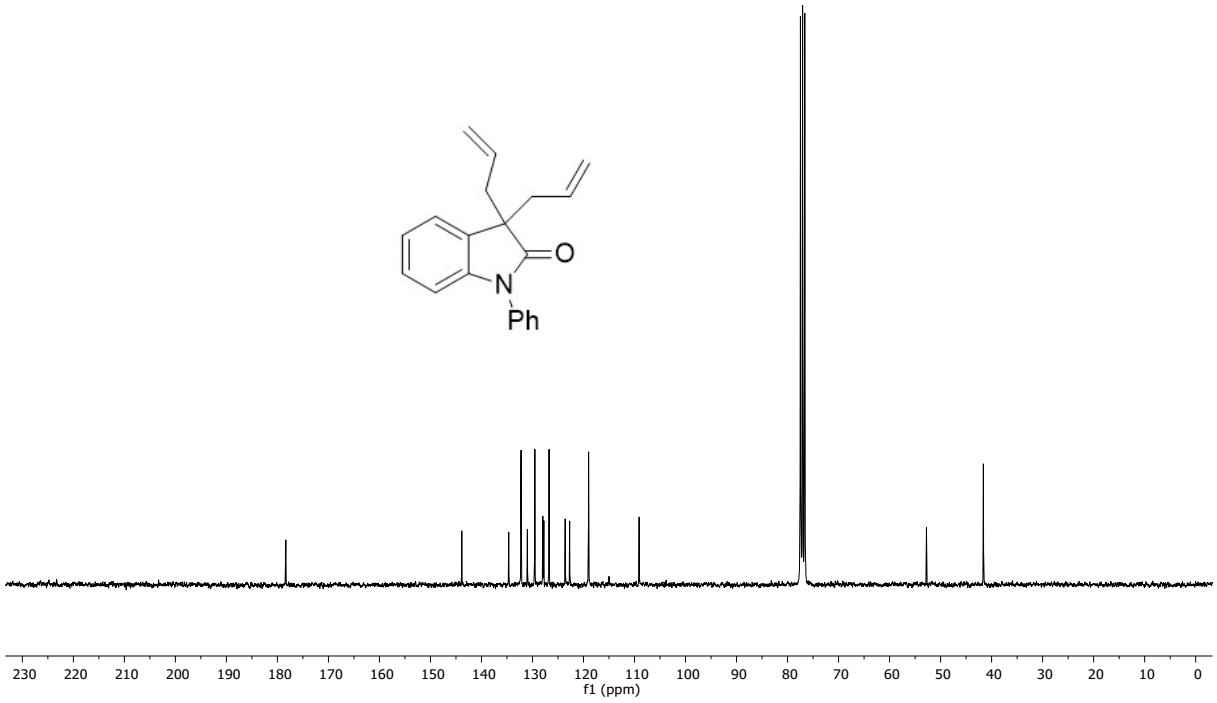


^{13}C NMR spectrum of 1,3-diallyl-3-methylindolin-2-one (5h)

3,3-diallyl-1-phenylindolin-2-one (4i)

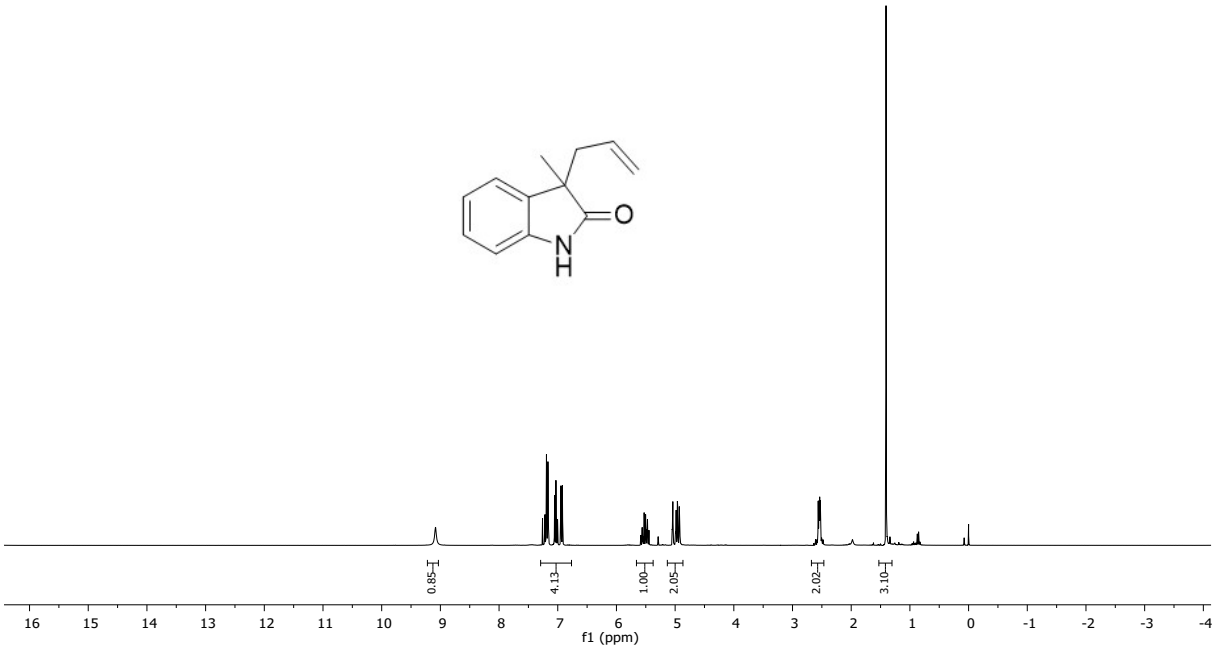


^{1}H NMR spectrum of 3,3-diallyl-1-phenylindolin-2-one (4i)

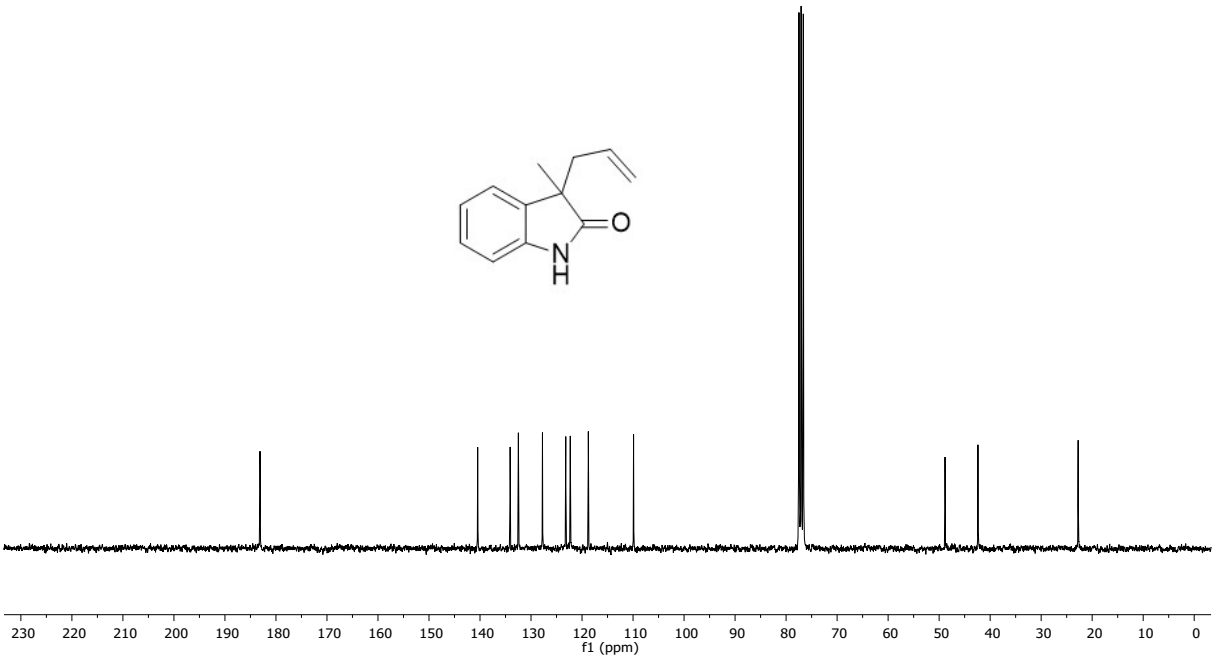


^{13}C NMR spectrum of 3,3-diallyl-1-phenylindolin-2-one (4i)

3-allyl-3-methylindolin-2-one (4j)

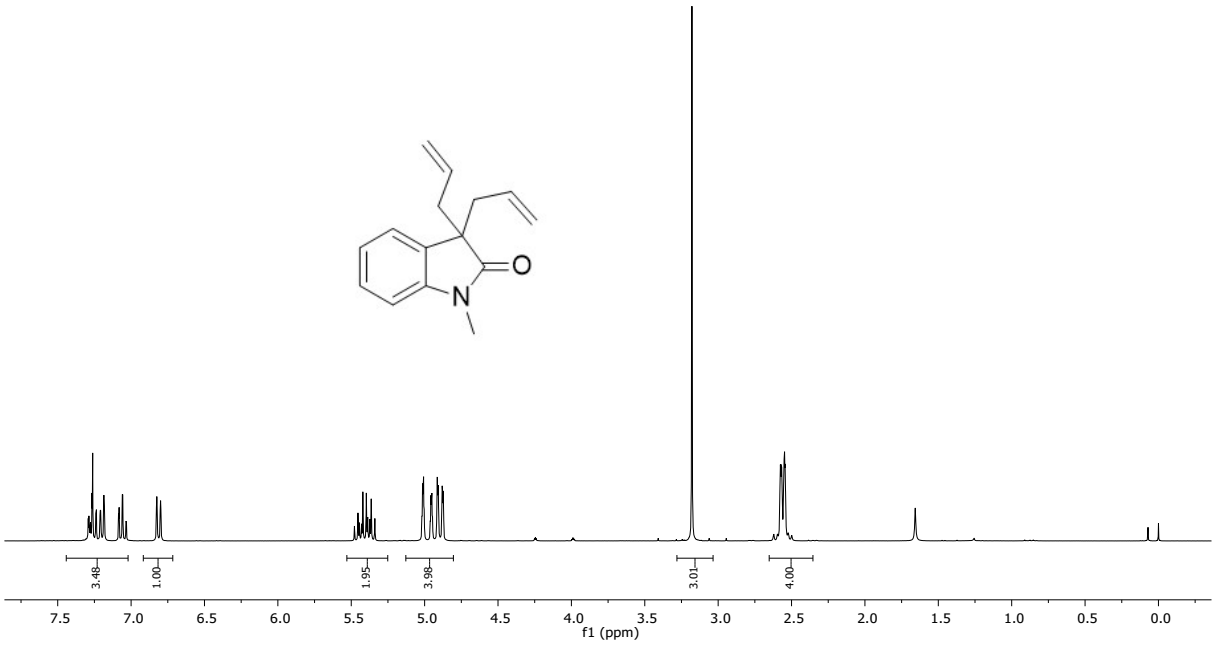


^{1}H NMR spectrum of 3-allyl-3-methylindolin-2-one (4j)

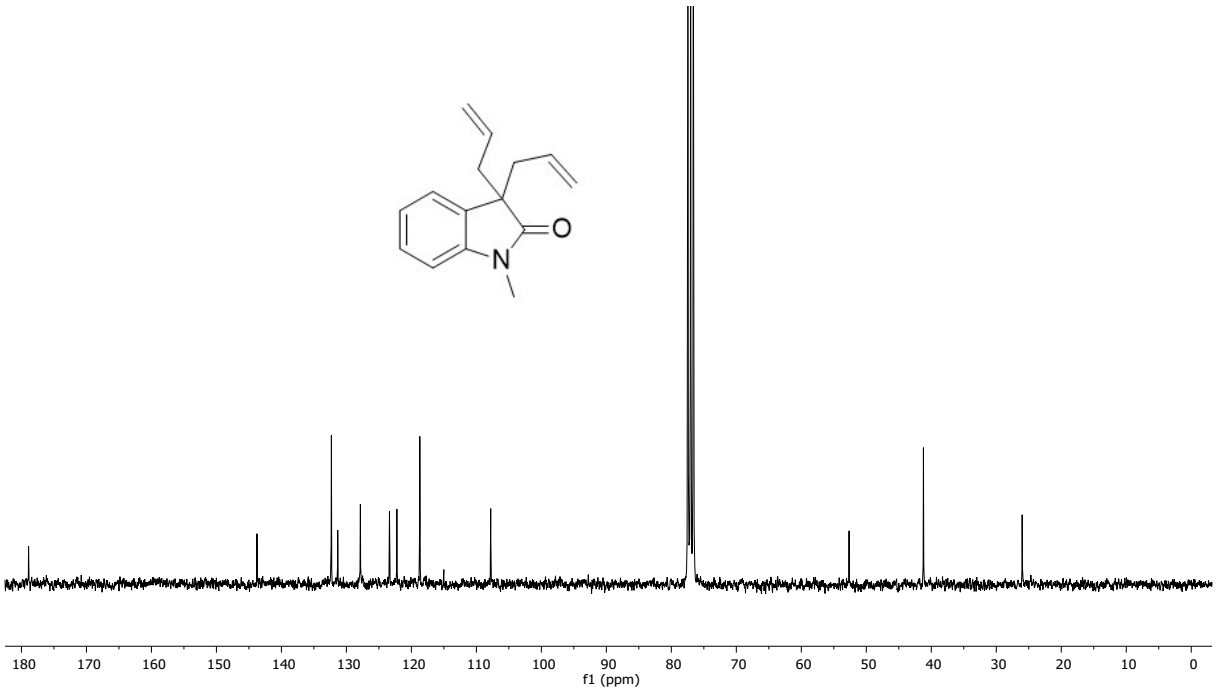


^{13}C NMR spectrum of 3-allyl-3-methylindolin-2-one (4j)

3,3-diallyl-1-methylindolin-2-one (4j)

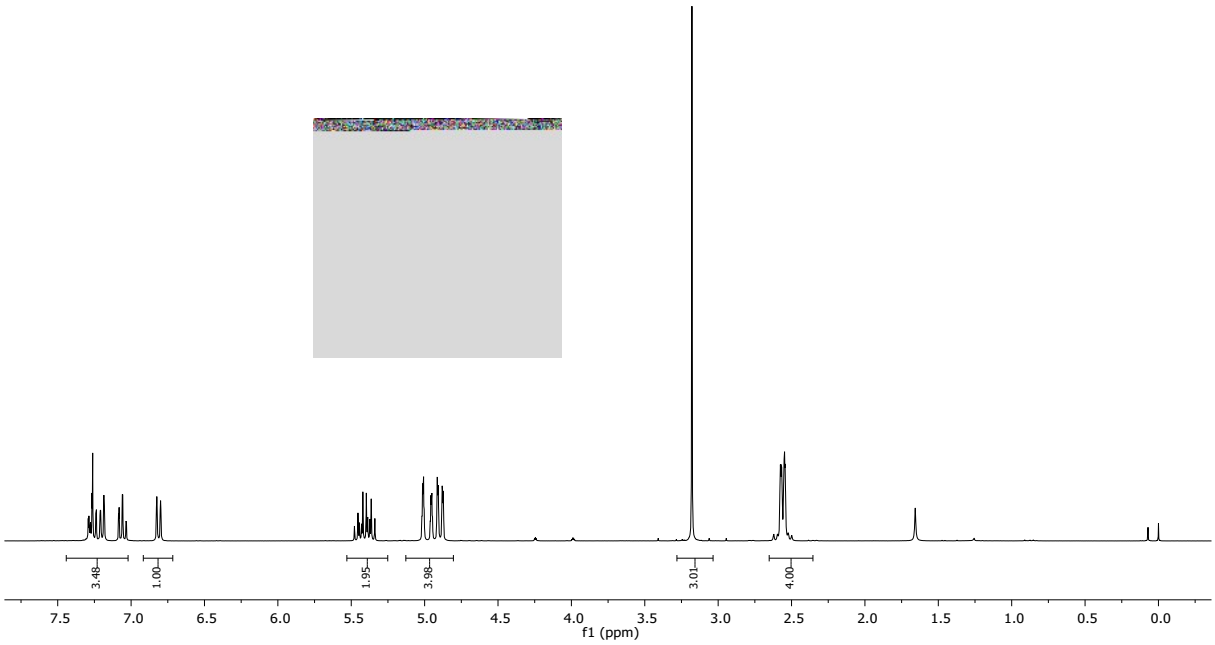


^{1}H NMR spectrum of 3,3-diallyl-1-methylindolin-2-one (4j)

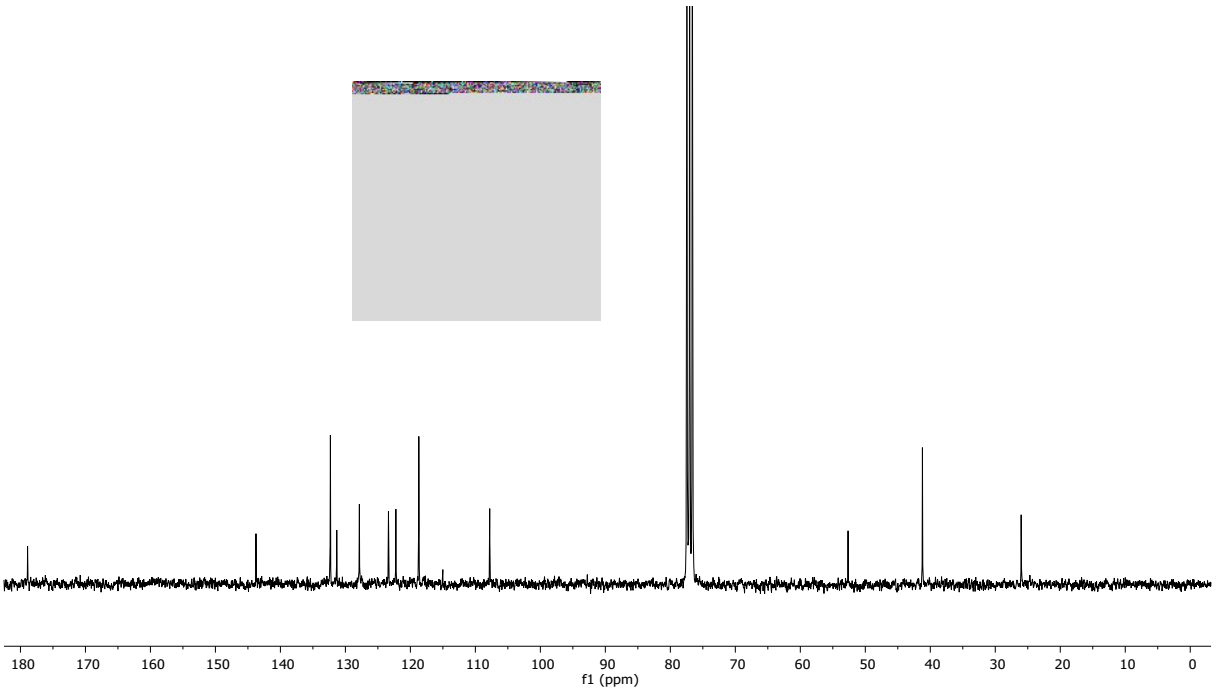


^{13}C NMR spectrum of 3,3-diallyl-1-methylindolin-2-one (4j)

3,3-diallyl-1-methylindolin-2-one (4j)

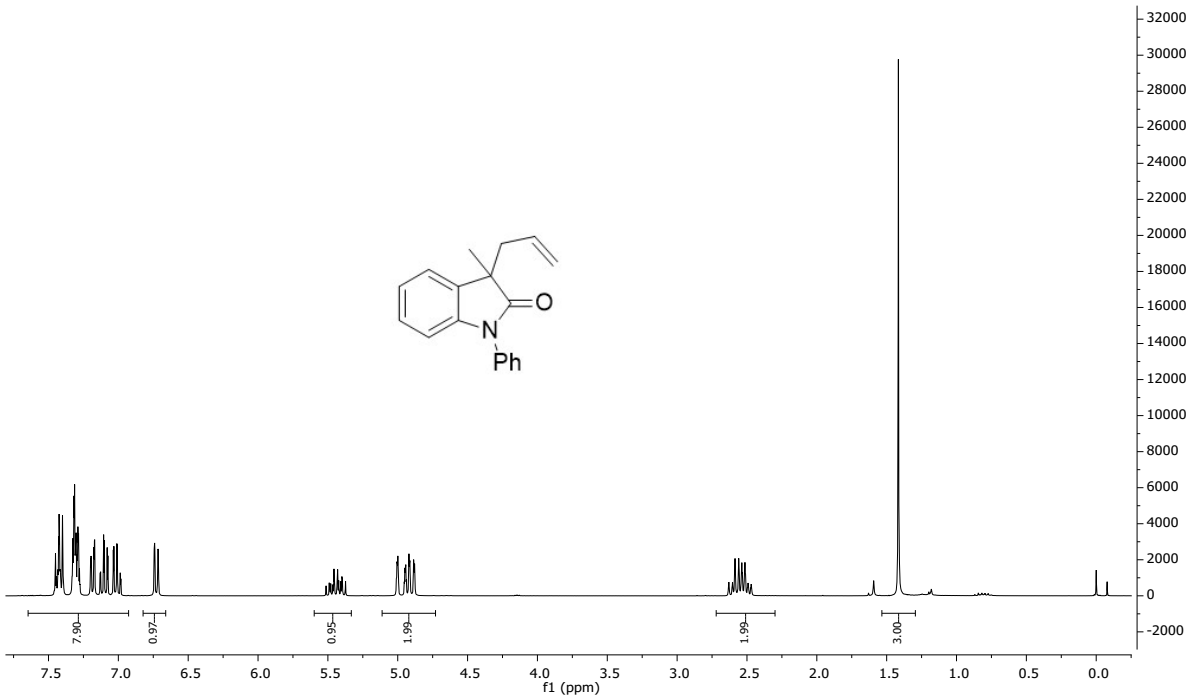


^{1}H NMR spectrum of 3,3-diallyl-1-methylindolin-2-one (4j)

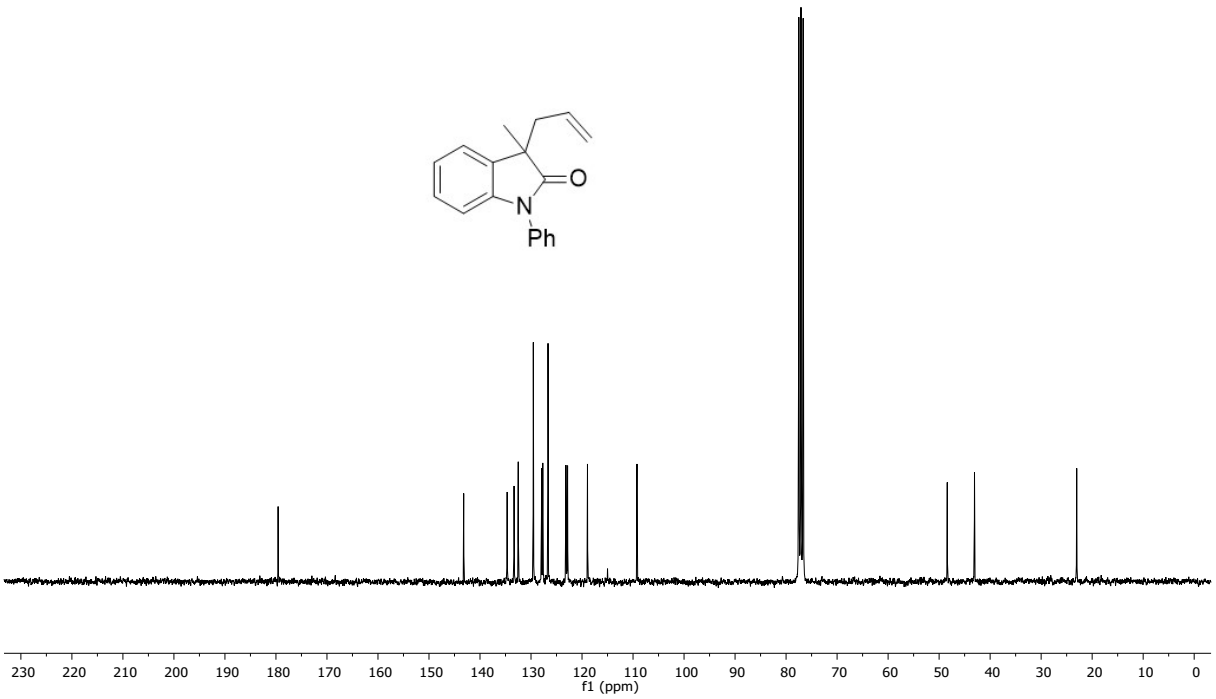


^{13}C NMR spectrum of 3,3-diallyl-1-methylindolin-2-one (4j)

3-allyl-3-methyl-1-phenylindolin-2-one (4k)

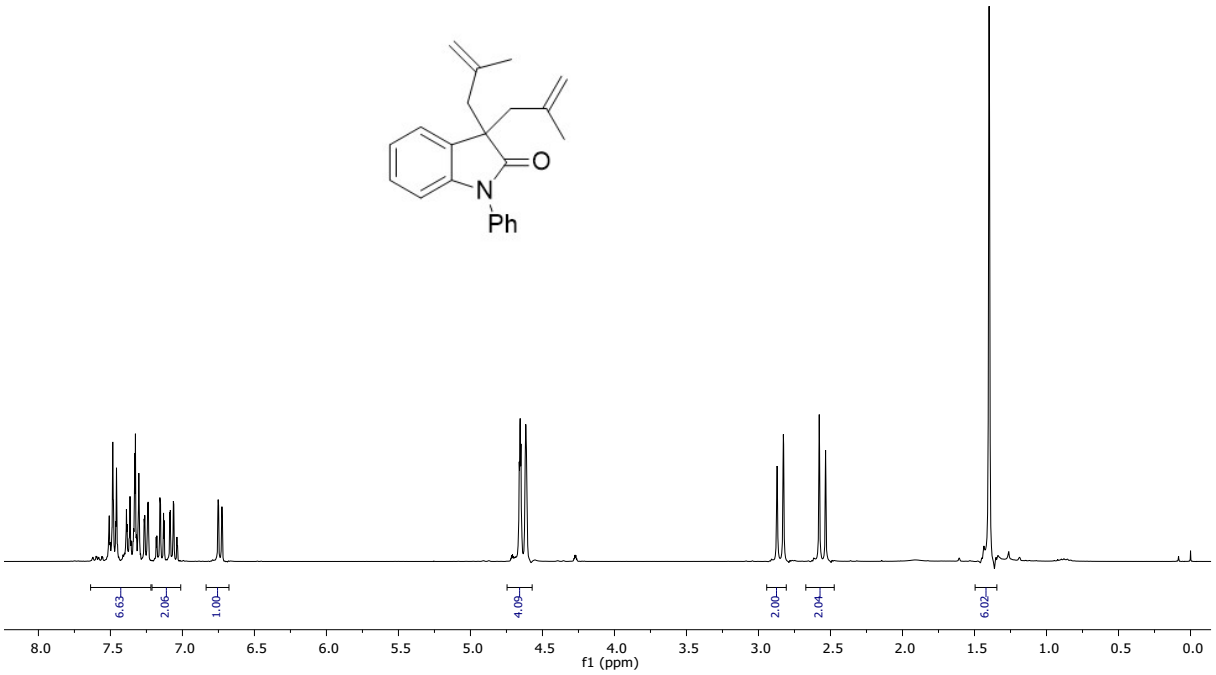


^{1}H NMR spectrum of 3-allyl-3-methyl-1-phenylindolin-2-one (4k)

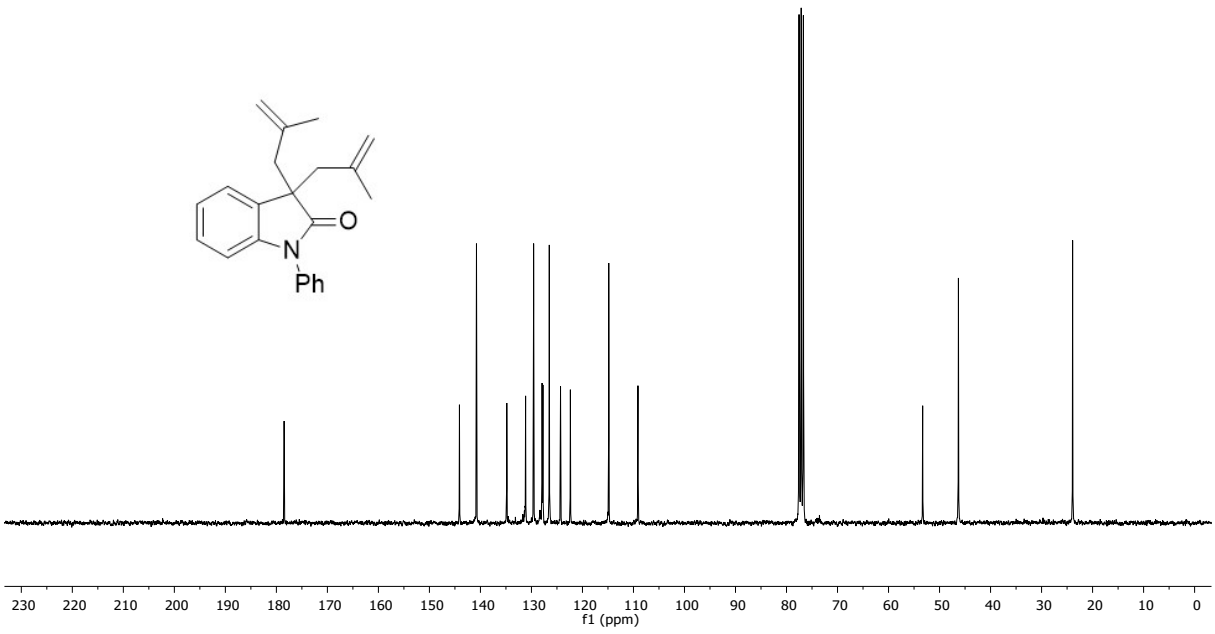


^{13}C NMR spectrum of 3-allyl-3-methyl-1-phenylindolin-2-one (4k)

3,3-bis(2-methylallyl)-1-phenylindolin-2-one (5ic)

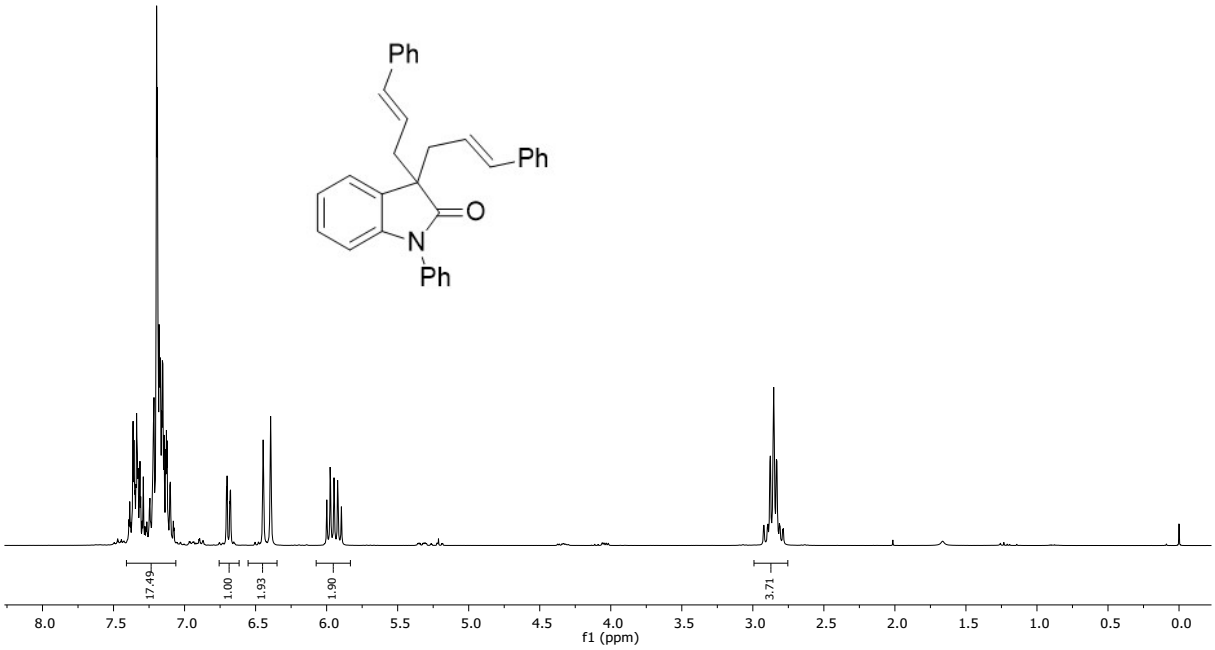


^{1}H NMR spectrum of 3,3-bis(2-methylallyl)-1-phenylindolin-2-one (5ic)

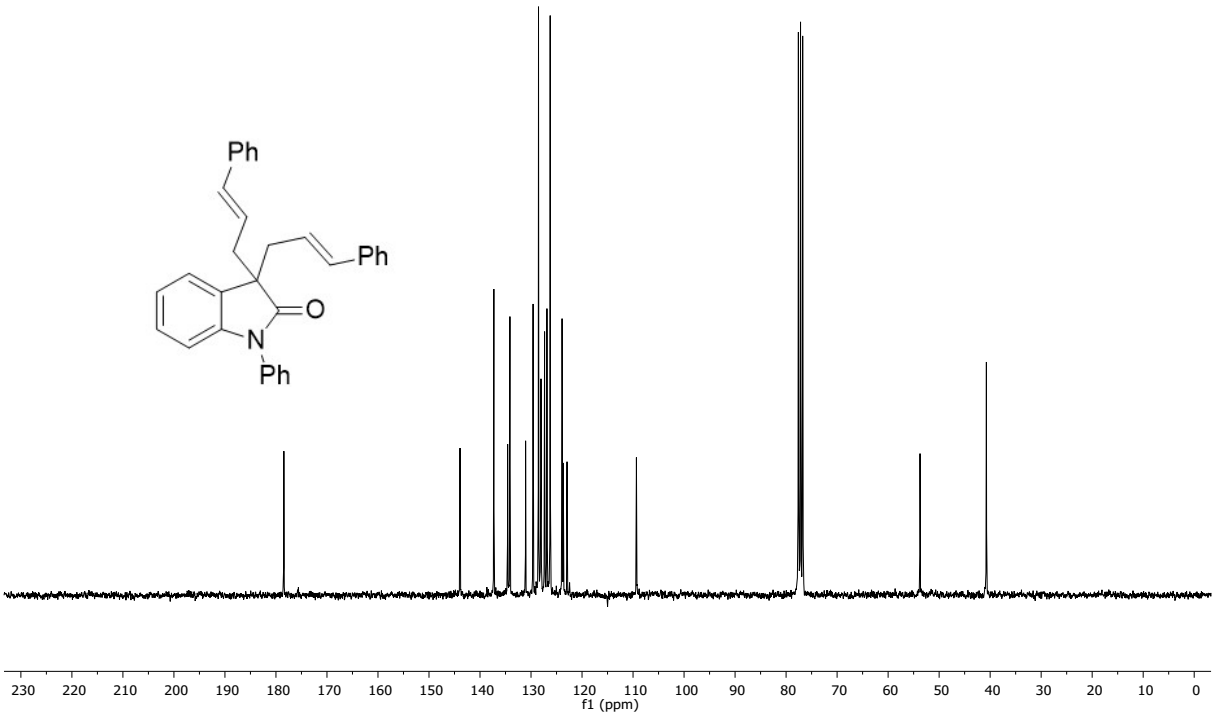


^{13}C NMR spectrum of 3,3-bis(2-methylallyl)-1-phenylindolin-2-one (5ic)

3,3-dicinnamyl-1-phenylindolin-2-one (5id)

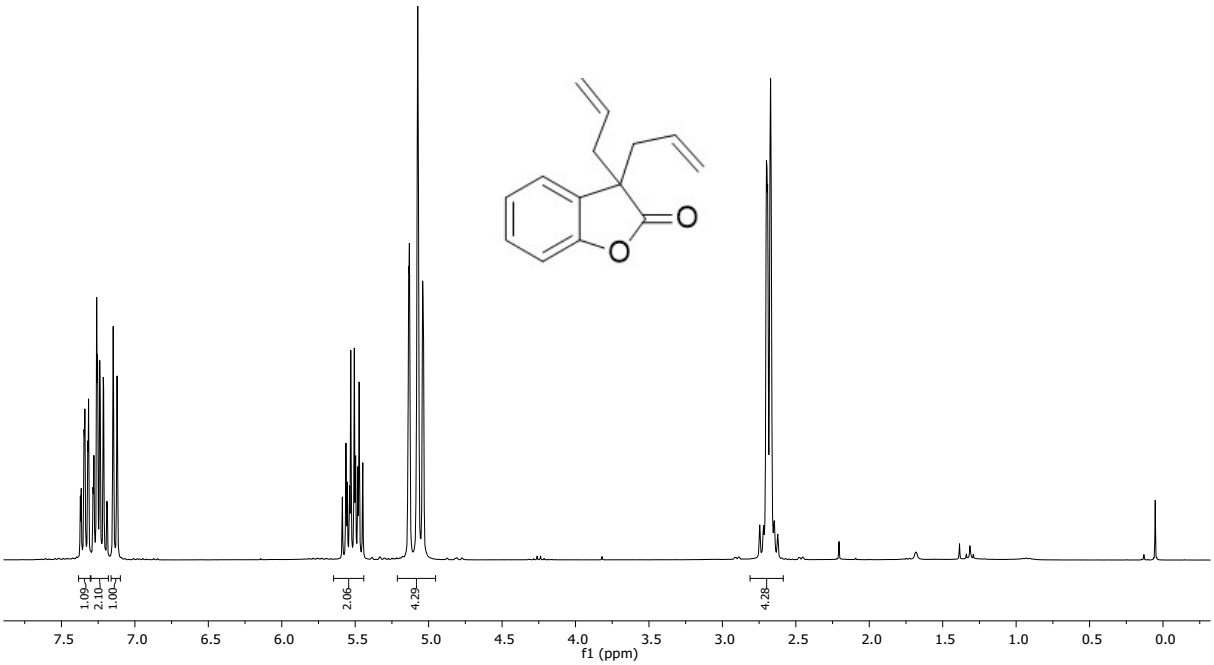


^{1}H NMR spectrum of 3,3-dicinnamyl-1-phenylindolin-2-one (5id)

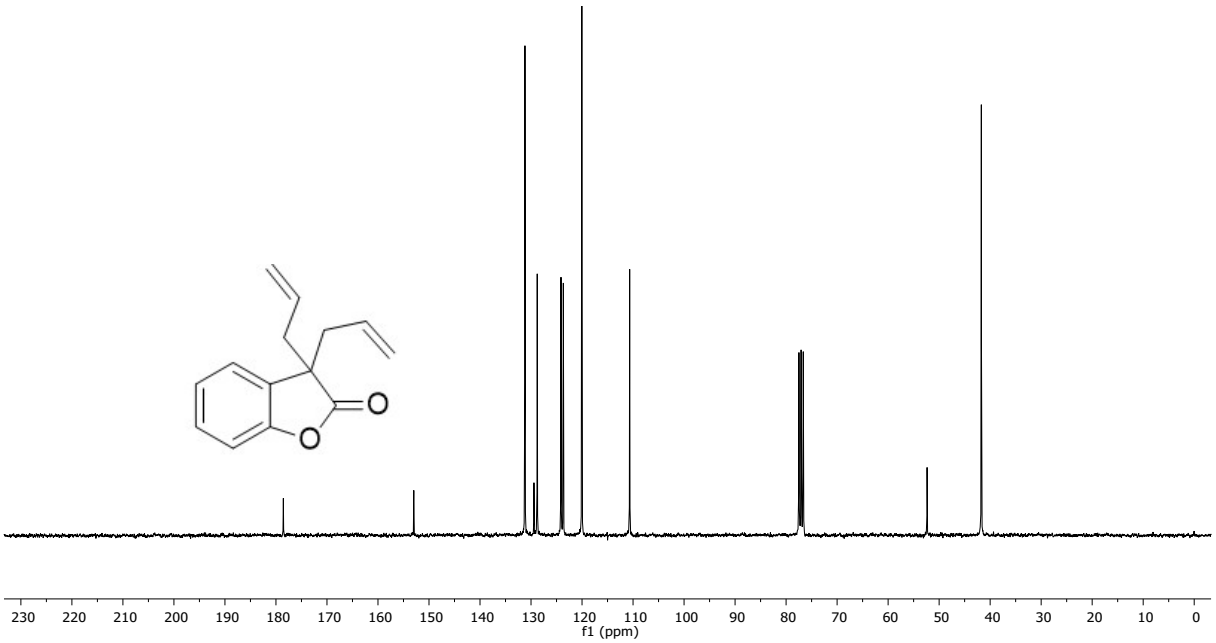


^{13}C NMR spectrum of 3,3-dicinnamyl-1-phenylindolin-2-one (5id)

3,3-diallyl-5-hydroxybenzofuran-2(3H)-one (3a')

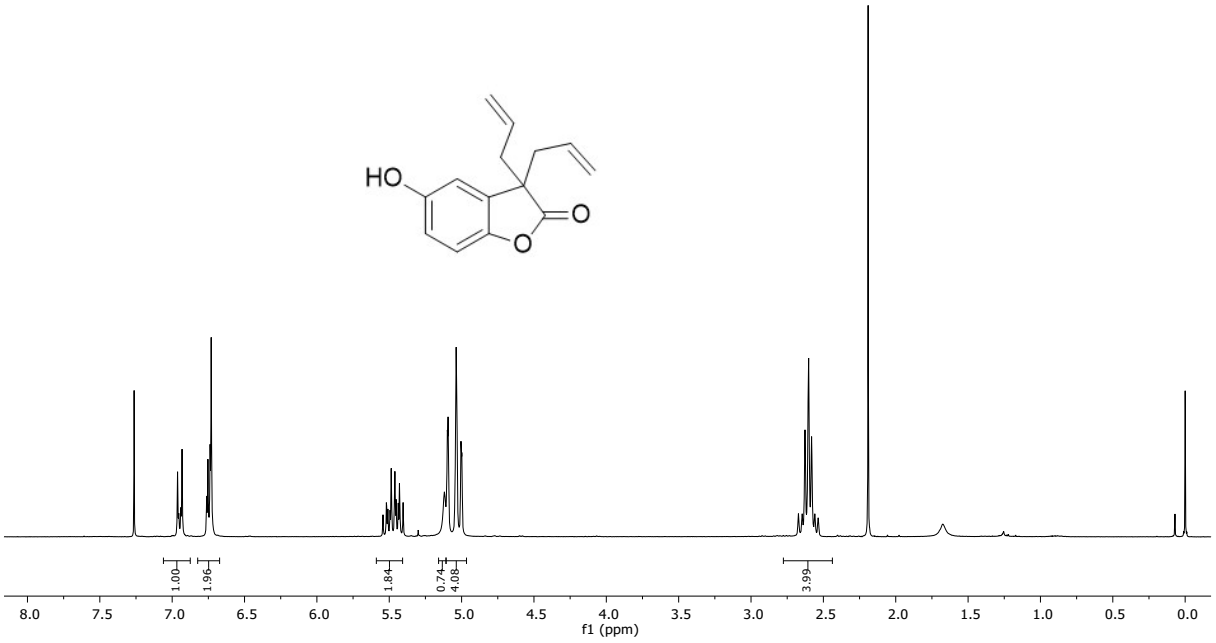


^{1}H NMR spectrum of 3,3-diallyl-5-hydroxybenzofuran-2(3H)-one (3a')

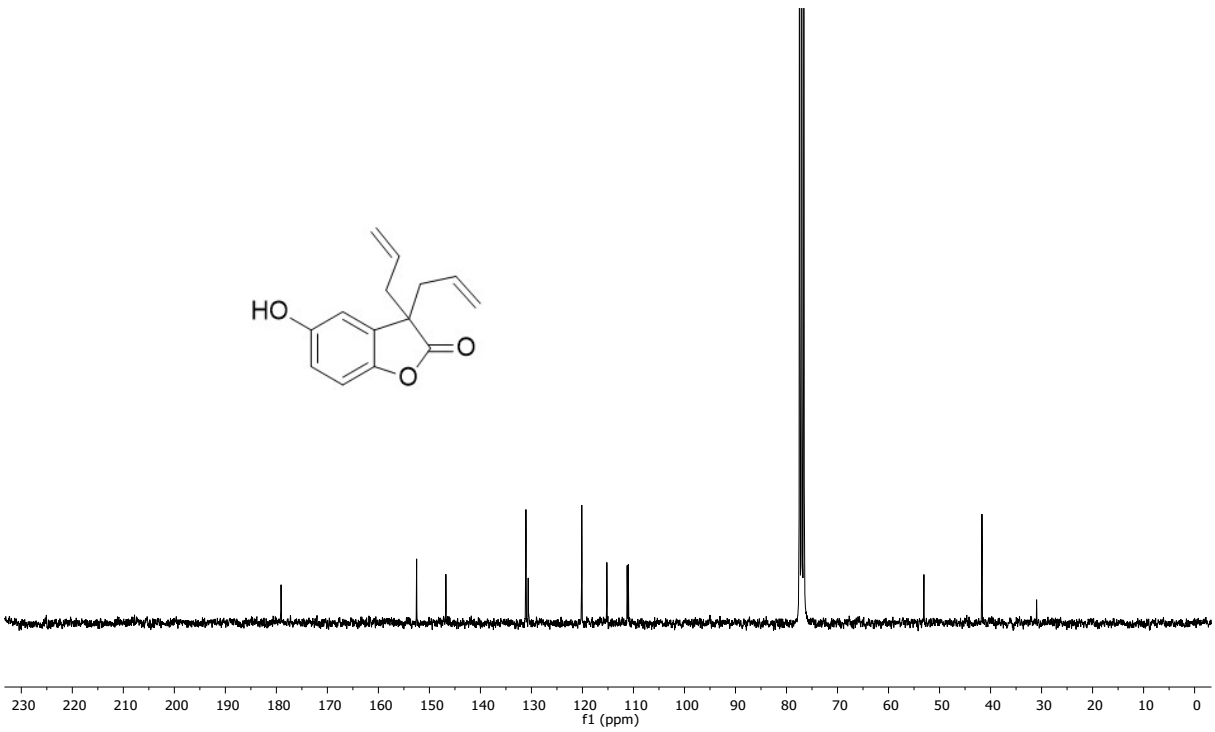


^{13}C NMR spectrum of 3,3-diallyl-5-hydroxybenzofuran-2(3H)-one (3a')

3,3-diallyl-5-hydroxybenzofuran-2(3H)-one (3b')



^{1}H NMR spectrum of 3,3-diallyl-5-hydroxybenzofuran-2(3H)-one (3b')



^{13}C NMR spectrum of 3,3-diallyl-5-hydroxybenzofuran-2(3H)-one (3b')