

Supporting Information

Combination of NDESs and Microflow Technology: A Sustainable Innovation for the Tandem Synthesis of 3-Aminohexahydrocoumarins

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Experimental

All reagents are commercially available and used without further purification. ^1H NMR (400 MHz) and ^{13}C NMR (100 MHz) spectra were obtained at 25 °C using CDCl_3 as solvent and chemical shifts are reported as δ values relative to TMS as internal standard. The FT-IR spectra were taken on a Nicolet-Impact 400D spectrophotometer in KBr pellets and reported in cm^{-1} . Melting points were determined using Stuart Scientific SMP2 apparatus and are uncorrected.

General procedure for the preparation of NADES (ChCl: urea)^[1]

A mixture of choline chloride (6.98 gr, 50 mmol) and urea (6.00 gr, 100 mmol) in a 1:2 molar ratio, were taken in a round bottom task and heated to 80 °C for 30 min to give a colorless transparent liquid. The resulting eutectic solvent, was then allowed to cool at room temperature and was used for the synthesis of 3-aminohexahydrocoumarins without further purification.

General procedure for the preparation of NADES (ChCl: thiourea)^[1]

A mixture of choline chloride (6.98 gr, 50 mmol) and thiourea (7.61 gr, 100 mmol) in a 1:2 molar ratio, placed in a 50 mL round-bottom flask. The resulting mixture was heated to 80 °C for 40 min with string until a clear colorless liquid was obtained.

General procedure for the preparation of NADES (ChCl: glycerol)^[1]

A mixture of choline chloride (6.98 gr, 50 mmol) and glycerol (9.21 gr, 100 mmol) in a 1:2 molar ratio, was added in a round-bottom flask and stirred during 60 min at 80 °C for obtaining the corresponding DES.

General procedure for the preparation of NADES (ChCl: benzoic acid) ^[1]

This DES is formed by mixing the ionic solid choline chloride (ChCl) with a hydrogen-bond donor benzoic acid in a 1:1 molar ratio. To prepare, a mixture of choline chloride (13.96 gr, 100 mmol) and benzoic acid (12.21 gr, 100 mmol) was slowly heated to 80 °C for 60 min under constant stirring to generate a colorless liquid.

General procedure for the preparation of NADES (ChCl: benzamide) ^[1]

A mixture of choline chloride (6.98 gr, 50 mmol) and benzamide (12.11 gr, 100 mmol) in a 1:2 molar ratio, was heated at 80 °C with stirring for 50 min the resulting eutectic solvent, was then allowed to cool at room temperature and was used for the synthesis of 3-aminohexahydrocoumarins without further purification.

General procedure for the synthesis of 3-aminohexahydrocoumarin derivatives 3 in a microreactor system

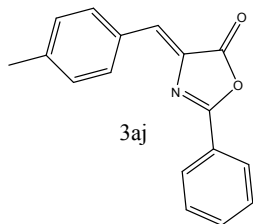
A flow microreactor system composed of two T-shaped micromixers (M1 and M2) and two microtube reactors (Q1 and Q2) was used. The whole flow microreactor system was dipped in an oil bath of 120 °C. A solution of hippuric acid (0.2 M in ChCl:urea) and acetic anhydride (0.2 M in ChCl:urea) and the binary solution of 4-methylbenzaldehyde (0.2 M in ChCl:urea) were introduced to M1 at 120 °C by peristaltic pumps. The resulting solution was passed through Q1 (internal diameter, 0.5 mm, internal volume, 0.6 ml, length, 200cm) (flow rate: 0.90 ml/h). The resulting solution was mixed with a solution of dimedone (0.2 M in ChCl:urea) in M2 at 120 °C. The resulting solution was passed through Q2 (flow rate: 3.6 ml/h), after the residence time was reached (10 min), the discharge was collected in a glass vessel. Water (5 ml) was added to it and stirred for 10 min until a white solid precipitated. After filtration, the pure products were recrystallized from ethanol in 80-97% yields. The pure product was characterized by FT-IR, ¹H NMR and ¹³CNMR.

Recover and reuse of the catalyst and DES

Reusability and recyclability of NADES as bio-renewable catalyst and solvent has the positive effect in minimizing the amount of waste. It is interesting to note that

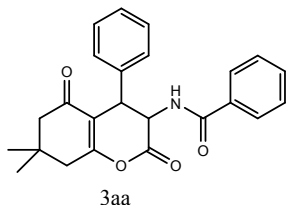
the work-up of the product could be performed also under eco-friendly condition. The outlet of the reactor was directly into a stirring collection vessel containing water. The reaction mixture was stirred for 0.5 h at room temperature in the vessel and the achieved supernatant was separated and crystallized in ethanol. The aqueous phase (which containing of the NADES) was separated and heated at 80 °C for 0.5 min. The DES was dried under vacuum and subjected to the next cycles without further purification.

Spectra data of (Z)-4-(4-methylbenzylidene)-2-phenyloxazol-5(4H)-one



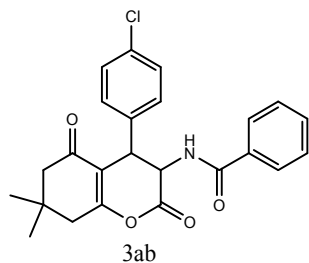
^1H NMR (400 MHz, CDCl_3): δ =2.44 (s, 3H), 7.30 (d, $J(\text{H,H})$ =8 Hz, 2H), 7.56 (t, $J(\text{H,H})$ =7.8 Hz, 2H), 7.64 (tt, $J(\text{H,H})$ =1.2, 8 Hz, 2H), 8.11 (d, $J(\text{H,H})$ =8 Hz, 2H), 8.19 ppm (d, $J(\text{H,H})$ =8 Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.83, 125.72, 128.28, 128.91, 129.74, 130.91, 132.03, 132.42, 132.56, 133.17, 142.11, 162.98, 167.79 ppm.

Spectra data of 3-aminohexahydrocoumarin products



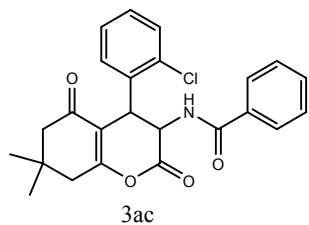
***N*-(3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-4-phenyl-2*H*-chromen-3-yl)benzamide:[1]**

IR (potassium bromide, ν , cm^{-1}): 1007, 1084, 1111, 1159, 1286, 1348, 1370, 1442, 1455, 1491, 1546, 1641, 1662, 1679, 1795, 1818, 2915, 2965, 3032, 3056, 3266, 3439. ^1H NMR (400 MHz, CDCl_3): δ =1.16 (s, 3H), 1.22 (s, 3H), 2.34 (d, $J(\text{H,H})$ =16 Hz, 1H), 2.4 (d, $J(\text{H,H})$ =16 Hz, 1H), 2.64 (s, 2H), 4.67 (d, $J(\text{H,H})$ =7.5 Hz, 1H), 5.41 (t, $J(\text{H,H})$ =7.5 Hz, 1H), 6.29 (d, $J(\text{H,H})$ =7.5 Hz, 1H), 7.13 (d, $J(\text{H,H})$ =9 Hz, 2H), 7.33 (m, 3H), 7.44 (d, $J(\text{H,H})$ =7.8 Hz, 2H), 7.54 (t, $J(\text{H,H})$ =7.4 Hz, 1H), 7.65 ppm (d, $J(\text{H,H})$ =8.4 Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.16, 28.61, 32.69, 40.74, 50.39, 52.61, 116.13, 127.08, 127.96, 128.39, 128.71, 129.28, 132.15, 133.23, 135.33, 164.78, 166.52, 167.11, 195.29 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{24}\text{NO}_4$ $[\text{M}+\text{H}]^+$: 390.1705, found: 390.1709.



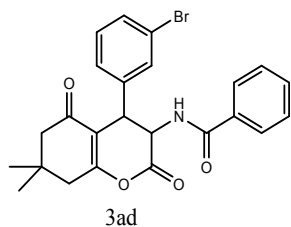
N*-(4-(4-chlorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide^[2]:**[2]*

IR (potassium bromide, ν , cm^{-1}): 1014, 1085, 1118, 1160, 1241, 1293, 1325, 1368, 1465, 1490, 1544, 1639, 1667, 1781, 1898, 2825, 2871, 2895, 2945, 3025, 3062, 3242, 3431, 3547. ^1H NMR (400 MHz, CDCl_3): δ =1.16 (s, 3H), 1.22 (s, 3H), 2.34 (d, $J(\text{H,H})=16$ Hz, 1H), 2.41 (d, $J(\text{H,H})=16$ Hz, 1H), 2.64 (d, $J(\text{H,H})=18$ Hz, 1H), 2.69 (d, $J(\text{H,H})=18$ Hz, 1H), 4.72 (d, $J(\text{H,H})=7.2$ Hz, 1H), 5.37 (t, $J(\text{H,H})=7.2$ Hz, 1H), 6.35 (d, $J(\text{H,H})=7.2$ Hz, 1H), 7.05 (d, $J(\text{H,H})=8$ Hz, 2H), 7.31 (d, $J(\text{H,H})=8$ Hz, 2H), 7.47 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.57 (t, $J(\text{H,H})=7.4$ Hz, 1H), 7.68 ppm (d, $J(\text{H,H})=7.2$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.1, 28.63, 32.69, 38.64, 40.70, 50.33, 52.66, 115.87, 127.08, 128.77, 129.36, 129.39, 133.28, 133.08, 133.99, 134.25, 164.87, 164.87, 166.37, 167.13, 195.29 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{23}\text{ClNO}_4$ $[\text{M}+\text{H}]^+$: 424.1311, found: 424.1313.



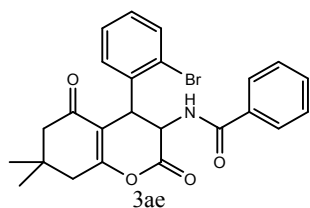
N*-(2-chlorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide^[2]:**[3]*

IR (potassium bromide, ν , cm^{-1}): 1007, 1056, 1086, 1113, 1164, 1208, 1292, 1347, 1374, 1442, 1475, 1541, 1601, 1653, 1805, 2832, 2872, 2958, 3060, 3193, 3274, 3439, 3583. ^1H NMR (400 MHz, CDCl_3): δ =1.15 (s, 3H), 1.22 (s, 3H), 2.33 (d, $J(\text{H,H})=16$ Hz, 1H), 2.40 (d, $J(\text{H,H})=16$ Hz, 1H), 2.63 (d, $J(\text{H,H})=18$ Hz, 1H), 2.69 (d, $J(\text{H,H})=18$ Hz, 1H), 5.29 (d, $J(\text{H,H})=7.6$ Hz, 1H), 5.56 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.24 (d, $J(\text{H,H})=7.6$ Hz, 1H), 7.01 (m, 1H), 7.25 (m, 2H), 7.44 (t, $J(\text{H,H})=7.2$ Hz, 2H), 7.54 (tt, $J(\text{H,H})=1.2, 7.6$ Hz, 1H), 7.67 ppm (d, $J(\text{H,H})=7.6$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.12, 28.58, 32.71, 38.98, 40.82, 50.30, 52.20, 115.87, 127.16, 127.77, 128.65, 129.57, 130.50, 133.02, 133.47, 135.44, 165.41, 166.18, 167.53, 194.99 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{23}\text{ClNO}_4$ $[\text{M}+\text{H}]^+$: 424.1311, found: 424.1313.



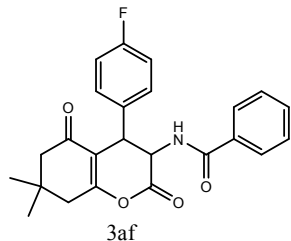
***N*-(4-(3-bromophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide:[4]**

IR (potassium bromide, ν , cm^{-1}): 1086, 1134, 1161, 1204, 1285, 1365, 1477, 1512, 1535, 1583, 1661, 1792, 2864, 2963, 3059, 3085, 3296, 3434. ^1H NMR (400 MHz, CDCl_3): δ =1.14 (s, 3H), 1.22 (s, 3H), 2.32 (d, $J(\text{H,H})=16$ Hz, 1H), 2.40 (d, $J(\text{H,H})=16$ Hz, 1H), 2.62 (d, $J(\text{H,H})=18$ Hz, 1H), 2.69 (d, $J(\text{H,H})=18$ Hz, 1H), 5.24 (d, $J(\text{H,H})=7.8$ Hz, 1H), 5.53 (t, $J(\text{H,H})=7.8$ Hz, 1H), 6.32 (d, $J(\text{H,H})=7.6$ Hz, 1H), 6.93 (d, $J(\text{H,H})=8.4$ Hz, 1H), 7.23 (dd, $J(\text{H,H})=2, 8.4$ Hz, 1H), 7.45 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.56 (t, $J(\text{H,H})=7.2$ Hz, 1H), 7.68 ppm (d, $J(\text{H,H})=8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.07, 28.61, 32.72, 35.75, 40.80, 50.25, 52.14, 115.74, 127.15, 128.00, 128.71, 130.27, 132.150, 132.27, 133.28, 134.74, 136.26, 165.50, 166.05, 167.57, 194.91 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{23}\text{BrINO}_4$ $[\text{M}+\text{H}]^+$: 468.0760, found: 468.0761.



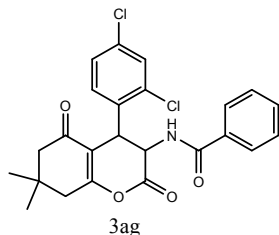
***N*-(4-(2-bromophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide^[2]:[5]**

IR (potassium bromide, ν , cm^{-1}): 1082, 1115, 1162, 1207, 1242, 1292, 1347, 1373, 1439, 1471, 1540, 1652, 1804, 2826, 2871, 2956, 3059, 3190, 3262, 3439, 3673. ^1H NMR (400 MHz, CDCl_3): δ =1.15 (s, 3H), 1.23 (s, 3H), 2.32 (d, $J(\text{H,H})=16$ Hz, 1H), 2.40 (d, $J(\text{H,H})=16$ Hz, 1H), 2.63 (d, $J(\text{H,H})=18$ Hz, 1H), 2.69 (d, $J(\text{H,H})=18$ Hz, 1H), 5.27 (d, $J(\text{H,H})=7.9$ Hz, 1H), 5.57 (t, $J(\text{H,H})=7.8$ Hz, 1H), 6.19 (d, $J(\text{H,H})=8$ Hz, 1H), 6.99 (dd, $J(\text{H,H})=2, 7.9$ Hz, 1H), 7.19 (td, $J(\text{H,H})=2, 7.9$ Hz, 1H), 7.31 (td, $J(\text{H,H})=1.2, 7$ Hz, 1H), 7.45 (t, $J(\text{H,H})=7.8$ Hz, 2H), 7.55 (tt, $J(\text{H,H})=1.2, 7.4$ Hz, 1H), 7.65 (dd, $J(\text{H,H})=1.2, 8$ Hz, 2H), 7.69 ppm (d, $J(\text{H,H})=7.8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.12, 28.57, 32.74, 38.48, 40.82, 50.29, 52.17, 116.38, 126.18, 127.24, 127.66, 128.47, 128.65, 129.83, 132.03, 133.95, 135.31, 165.33, 166.15, 167.57, 194.99 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{23}\text{BrINO}_4$ $[\text{M}+\text{H}]^+$: 468.0760, found: 468.0761.



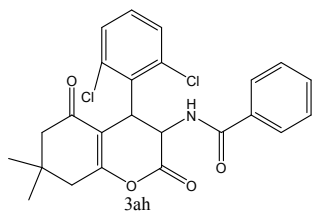
N*-(4-(4-fluorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide^[2]:**[6]*

IR (potassium bromide, ν , cm^{-1}): 1009, 1086, 1159, 1227, 1370, 1484, 1511, 1579, 1602, 1664, 1785, 2822, 2874, 2961, 3066, 3316, 3446, 3544. ^1H NMR (400 MHz, CDCl_3): δ =1.16 (s, 3H), 1.22 (s, 3H), 2.34 (d, $J(\text{H,H})=16$ Hz, 1H), 2.41 (d, $J(\text{H,H})=16$ Hz, 1H), 2.63 (s, 2H), 4.72 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.36 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.34 (d, $J(\text{H,H})=7.2$ Hz, 1H), 7.03 (t, $J(\text{H,H})=8.5$ Hz, 2H), 7.10 (t, $J(\text{H,H})=5$ Hz, 2H), 7.46 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.56 (tt, $J(\text{H,H})=1.5, 7.6$ Hz, 1H), 7.67 ppm (d, $J(\text{H,H})=7.6$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.14, 28.59, 32.69, 38.59, 40.72, 50.37, 50.77, 116.09, 116.31, 127.04, 128.77, 129.58, 129.66, 131.17, 131.20, 132.25, 133.11, 161.34, 163.80, 164.71, 166.45, 167.11, 195.19 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{23}\text{FNO}_4$ $[\text{M}+\text{H}]^+$: 408.1606, found: 408.1607.



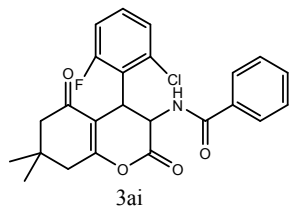
N*-(4-(2,4-dichlorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide:**[7]*

IR (potassium bromide, ν , cm^{-1}): 688, 1014, 1987, 1117, 1161, 1204, 1290, 1366, 1470, 1544, 1583, 1637, 1666, 1786, 2826, 2895, 2947, 3029, 3061, 3240, 3435, 3555. ^1H NMR (400 MHz, CDCl_3): δ =1.15 (s, 3H), 1.22 (s, 3H), 2.32 (d, $J(\text{H,H})=16$ Hz, 1H), 2.41 (d, $J(\text{H,H})=16$ Hz, 1H), 2.63 (d, $J(\text{H,H})=18$ Hz, 1H), 2.69 (d, $J(\text{H,H})=18$ Hz, 1H), 5.24 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.53 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.31 (d, $J(\text{H,H})=7.6$ Hz, 1H), 6.93 (d, $J(\text{H,H})=8$ Hz, 1H), 7.23 (dd, $J(\text{H,H})=2.1, 7.65$ Hz, 1H), 7.46 (t, $J(\text{H,H})=8$ Hz, 2H), 7.56 (tt, $J(\text{H,H})=1.2, 7.6$ Hz, 1H), 7.68 ppm (d, $J(\text{H,H})=7.6$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.06, 28.62, 32.73, 35.69, 40.77, 50.22, 52.13, 116.09, 127.17, 128.01, 128.7, 130.26, 132.15, 132.27, 133.25, 134.71, 165.54, 166.08, 167.58, 194.96 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{22}\text{Cl}_2\text{NO}_4$ $[\text{M}+\text{H}]^+$: 458.0833, found: 458.0831.



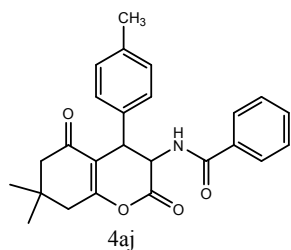
***N*-(4-(2,6-dichlorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide:[8]**

IR (potassium bromide, ν , cm^{-1}): 1019, 1063, 1094, 1111, 1164, 1301, 1375, 1455, 1549, 1576, 1603, 1644, 1796, 2835, 2893, 2958, 3028, 3064, 3193, 3256, 3565. ^1H NMR (400 MHz, CDCl_3): δ =1.11 (s, 3H), 1.20(s, 3H), 2.31 (d, $J(\text{H,H})=16$ Hz, 1H), 2.38 (d, $J(\text{H,H})=16$ Hz, 1H), 2.56 (d, $J(\text{H,H})=18$ Hz, 1H), 2.64 (d, $J(\text{H,H})=18$ Hz, 1H), 5.45 (d, $J(\text{H,H})=9$ Hz, 1H) 5.52 (t, $J(\text{H,H})=9$ Hz, 1H), 6.35 (d, $J(\text{H,H})=7.2$ Hz, 1H), 6.98 (t, $J(\text{H,H})=8$ Hz, 1H), 7.24 (m, 2H), 7.42 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.53 (t, $J(\text{H,H})=7.2$ Hz, 1H), 7.60 ppm (d, $J(\text{H,H})=7.6$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =27.91, 28.48, 32.59, 34.66, 41.01, 50.16, 50.41, 114.56, 114.79 126.39, 127.06, 128.66, 129.80, 129.90, 132.10, 133.41, 163.21, 165.60, 166.01, 167.49, 195.82 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{22}\text{Cl}_2\text{NO}_4$ $[\text{M}+\text{H}]^+$: 458.0833, found: 458.0831.



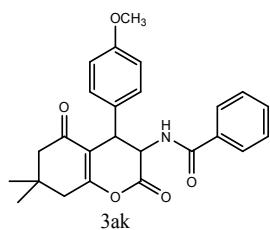
***N*-(4-(2-chloro-6-fluorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide:[9]**

IR (potassium bromide, ν , cm^{-1}): 1019, 1164, 1111, 1214, 1302, 1332, 1375, 1455, 1548, 1603, 1645, 1796, 2867, 2958, 3032, 3063, 3197, 3258. ^1H NMR (400 MHz, CDCl_3): δ =1.11 (s, 3H), 1.20(s, 3H), 2.31 (d, $J(\text{H,H})=16$ Hz, 1H), 2.37 (d, $J(\text{H,H})=16$ Hz, 1H), 2.56 (d, $J(\text{H,H})=18$ Hz, 1H), 2.64 (d, $J(\text{H,H})=18$ Hz, 1H), 5.45 (d, $J(\text{H,H})=9$ Hz, 1H) 5.52 (t, $J(\text{H,H})=9$ Hz, 1H), 6.34 (d, $J(\text{H,H})=7.2$ Hz, 1H), 6.98 (t, $J(\text{H,H})=8$ Hz, 1H), 7.26 (m, 2H), 7.42 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.53 (t, $J(\text{H,H})=7.6$ Hz, 1H), 7.60 ppm (d, $J(\text{H,H})=7.6$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =27.91, 28.47, 32.59, 34.66, 41.01, 50.16, 50.41, 114.56, 114.79, 126.36, 127.06, 128.66, 129.79, 129.89, 132.05, 133.41, 165.61, 166.01, 167.49, 195.82 pp; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{22}\text{ClFNO}_4$ $[\text{M}+\text{H}]^+$: 442.1265, found: 442.1262.



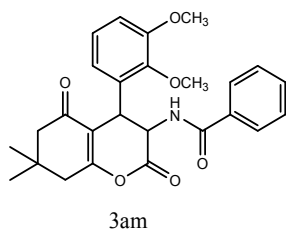
N*-(3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-4-*p*-tolyl-2*H*-chromen-3-yl)benzamide^[2]:**[10]*

IR (potassium bromide, ν , cm^{-1}): 1108, 1076, 1253, 1305, 1371, 1511, 1536, 1606, 1536, 1606, 1654, 1739, 1795, 2651, 2912, 2962, 3025, 3249, 3326, 3439. ^1H NMR (400 MHz, CDCl_3): δ =1.16 (s, 3H), 1.22(s, 3H), 2.32 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (d, $J(\text{H,H})=16$ Hz, 1H), 2.63 (s, 2H), 5.61 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.39 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.28 (d, $J(\text{H,H})=7.6$ Hz, 1H), 7.01 (d, $J(\text{H,H})=8$ Hz, 2H), 7.14 (d, $J(\text{H,H})=7.6$ Hz, 2H), 7.45 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.53 (t, $J(\text{H,H})=8$ Hz, 2H), 7.68 ppm (d, $J(\text{H,H})=7.6$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.09, 28.12, 28.62, 32.68, 38.96, 40.71, 50.39, 52.62, 116.28, 127.11, 127.82, 128.68, 129.98, 132.10, 133.29, 138.14, 164.69, 166.59, 167.07, 195.29 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{26}\text{NO}_4$ $[\text{M}+\text{H}]^+$: 404.1857, found: 404.1848.



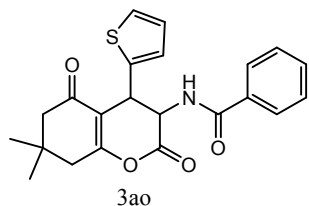
N*-(3,4,5,6,7,8-hexahydro-4(4-methoxyphenyl)-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide:**[11]*

IR (potassium bromide, ν , cm^{-1}): 1045, 1090, 1136, 1265, 1367, 1454, 1492, 1535, 1562, 1609, 1660, 1787, 2832, 2931, 2956, 3025, 3064, 3294, 3442. ^1H NMR (400 MHz, CDCl_3): δ =1.16 (s, 3H), 1.21(s, 3H), 2.33 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (d, $J(\text{H,H})=16$ Hz, 1H), 2.62 (d, $J(\text{H,H})=18$ Hz, 1H), 2.67 (d, $J(\text{H,H})=18$ Hz, 1H), 3.78 (s, 3H), 4.61 (d, $J(\text{H,H})=7.4$ Hz, 1H) 5.37 (t, $J(\text{H,H})=7.4$ Hz, 1H), 6.30 (d, $J(\text{H,H})=7.4$ Hz, 1H), 6.86 (d, $J(\text{H,H})=8.6$ Hz, 2H), 7.04 (d, $J(\text{H,H})=8.6$ Hz, 2H), 7.45 (t, $J(\text{H,H})=7.8$ Hz, 2H), 7.55 (t, $J(\text{H,H})=7.4$ Hz, 1H), 7.68 ppm (d, $J(\text{H,H})=8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.15, 28.61, 32.68, 38.57, 40.73, 50.41, 52.76, 55.28, 114.66, 116.37, 127.09, 128.71, 129.05, 132.13, 133.27, 159.53, 164.54, 166.62, 167.08, 195.29 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{25}\text{H}_{26}\text{NO}_4$ $[\text{M}+\text{H}]^+$: 404.1856, found: 404.1858.



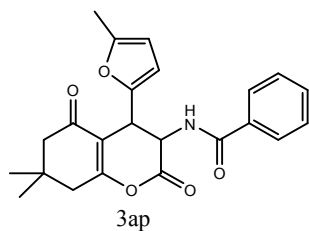
***N*-(3,4,5,6,7,8-hexahydro-4(2,3-dimethoxyphenyl)-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide:[12]**

IR (potassium bromide, ν , cm^{-1}): 1005, 1118, 1165, 1219, 1276, 1368, 1482, 1534, 1582, 1660, 1790, 2880, 2891, 2955, 3201, 3287, 3544. ^1H NMR (400 MHz, CDCl_3): δ =1.15 (s, 3H), 1.21(s, 3H), 2.31 (d, $J(\text{H,H})=16$ Hz, 1H), 2.37 (d, $J(\text{H,H})=16$ Hz, 1H), 2.61 (s, 2H), 3.86 (s, 3H), 3.91 (s, 3H), 4.96 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.35 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.43 (d, $J(\text{H,H})=7.2$ Hz, 1H), 6.71 (dd, $J(\text{H,H})=1.2, 7.6$ Hz, 1H), 6.89 (dd, $J(\text{H,H})=1.5, 4.2$ Hz, 1H), 7.01 (t, $J(\text{H,H})=7.6$ Hz, 1H), 7.41 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.50 (t, $J(\text{H,H})=7.6$ Hz, 1H), 7.63 ppm (d, $J(\text{H,H})=7.2$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.21, 28.56, 32.63, 34.97, 41.01, 50.58, 51.83, 55.79, 60.25, 112.75, 115.43, 121.31, 124.55, 127.15, 128.54, 128.93, 131.75, 133.67, 147.25, 152.79, 164.77, 165.74, 167.52, 195.65 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{26}\text{H}_{28}\text{NO}_6$ $[\text{M}+\text{H}]^+$: 450.1916, found: 450.1911.



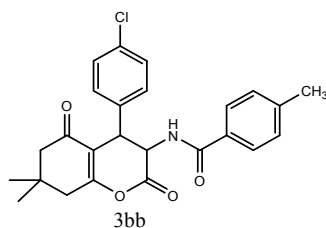
***N*-(3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-4(thiophen-2-yl)-2*H*-chromen-3-yl)benzamide^[2]:[13]**

IR (potassium bromide, ν , cm^{-1}): 693, 835, 933, 1000, 1081, 1115, 1153, 1204, 1288, 1355, 1541, 1544, 1787, 2815, 1892, 2932, 2960, 3065, 3254, 3561. ^1H NMR (400 MHz, CDCl_3): δ =1.18 (s, 3H), 1.21(s, 3H), 2.37 (d, $J(\text{H,H})=16$ Hz, 1H), 2.42 (d, $J(\text{H,H})=16$ Hz, 1H), 2.59 (d, $J(\text{H,H})=18$ Hz, 1H), 2.66 (d, $J(\text{H,H})=18$ Hz, 1H), 4.96 (d, $J(\text{H,H})=7.2$ Hz, 1H) 5.37 (t, $J(\text{H,H})=7.2$ Hz, 1H), 6.53 (d, $J(\text{H,H})=7.6$ Hz, 1H), 6.86 (d, $J(\text{H,H})=3.4$ Hz, 1H), 6.98 (d, $J(\text{H,H})=4.2$ Hz, 1H), 7.23 (dd, $J(\text{H,H})=1, 5.2$ Hz, 1H), 7.48 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.57 (tt, $J(\text{H,H})=1.2, 7.2$ Hz, 1H), 7.78 ppm (d, $J(\text{H,H})=7.6$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =27.92, 28.83, 32.63, 34.57, 40.72, 50.32, 52.93, 116.52, 125.67, 126.37, 127.17, 127.73, 128.75, 132.23, 133.18, 137.94, 164.80, 166.11, 167.08, 194.93 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{22}\text{H}_{22}\text{NO}_4\text{S}$ $[\text{M}+\text{H}]^+$: 396.1265, found: 396.1259.



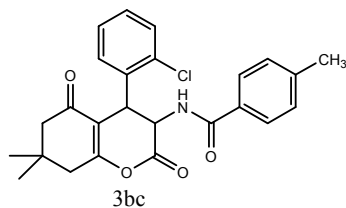
***N*-(3,4,5,6,7,8-hexahydro-7,7-dimethyl-4-(5-methylfuran-2-yl)-2,5-dioxo-2*H*-chromen-3-yl)benzamide:[14]**

IR (potassium bromide, ν , cm^{-1}): 1018, 1094, 1119, 1155, 1207, 1275, 1319, 1355, 1501, 1530, 1613, 1656, 1780, 1924, 2741, 2822, 2874, 2916, 2962, 3141, 3375, 3543. ^1H NMR (400 MHz, CDCl_3): δ =1.13 (s, 3H), 1.93 (s, 3H), 2.21 (s, 3H), 2.35 (s, 2H), 2.56 (s, 2H), 4.71 (d, $J(\text{H,H})=7$ Hz, 1H), 5.28 (t, $J(\text{H,H})=7$ Hz, 1H), 5.88 (d, $J(\text{H,H})=3$ Hz, 1H), 6.10 (d, $J(\text{H,H})=3$ Hz, 1H), 6.61 (d, $J(\text{H,H})=7.5$ Hz, 1H), 7.48 (t, $J(\text{H,H})=7.8$ Hz, 2H), 7.57 (tt, $J(\text{H,H})=1.2, 7.4$ Hz, 1H), 7.75 ppm (d, $J(\text{H,H})=7.8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =13.63, 27.99, 28.51, 32.71, 33.36, 40.78, 50.34, 51.14, 106.75, 110.09, 113.95, 127.14, 128.70, 132.13, 133.38, 147.01, 152.75, 165.21, 166.26, 167.18, 195.13 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{23}\text{H}_{24}\text{NO}_5$ $[\text{M}+\text{H}]^+$: 394.1654, found: 394.1649.



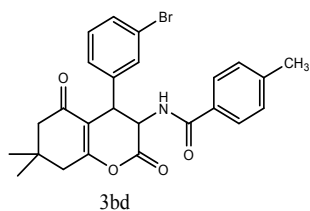
***N*-(4-(4-chlorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[15]**

IR (potassium bromide, ν , cm^{-1}): 1014, 1083, 1117, 1159, 1203, 1366, 1415, 1492, 1547, 1643, 1666, 1784, 2818, 2893, 2920, 2962, 3032, 3261, 3551. ^1H NMR (400 MHz, CDCl_3): δ =1.16 (s, 3H), 1.22 (s, 3H), 2.34 (d, $J(\text{H,H})=16$ Hz, 1H), 2.41 (d, $J(\text{H,H})=16$ Hz, 1H), 2.42 (s, 3H), 2.64 (d, $J(\text{H,H})=18$ Hz, 1H), 2.68 (d, $J(\text{H,H})=18$ Hz, 1H), 4.71 (d, $J(\text{H,H})=7.6$ Hz, 1H), 5.36 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.31 (d, $J(\text{H,H})=7$ Hz, 1H), 7.04 (d, $J(\text{H,H})=8.4$ Hz, 2H), 7.25 (d, $J(\text{H,H})=7.9$ Hz, 2H), 7.29 (t, $J(\text{H,H})=4.5$ Hz, 2H), 7.58 ppm (d, $J(\text{H,H})=8.2$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.52, 28.11, 28.62, 32.69, 38.68, 40.73, 50.35, 52.63, 115.9, 127.07, 129.34, 129.37, 129.42, 130.17, 134.01, 134.25, 142.89, 164.84, 166.44, 167.02, 195.15 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{25}\text{ClNO}_4$ $[\text{M}+\text{H}]^+$: 438.1458, found: 438.1449.



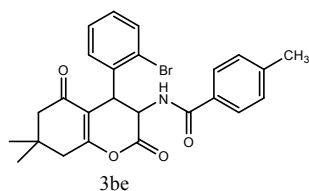
***N*-(4-(2-chlorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[16]**

IR (potassium bromide, ν , cm^{-1}): 1083, 1116, 1161, 1290, 1347, 1371, 1475, 1506, 1537, 1656, 1807, 2869, 2927, 2955, 3031, 3291, 3446. ^1H NMR (400 MHz, CDCl_3): δ =1.15 (s, 3H), 1.23(s, 3H), 2.33 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (s, 3H), 2.63 (d, $J(\text{H,H})=18$ Hz, 1H), 2.69 (d, $J(\text{H,H})=18$ Hz, 1H), 5.27 (d, $J(\text{H,H})=6.8$ Hz, 1H) 5.55 (t, $J(\text{H,H})=6.8$ Hz, 1H), 6.19 (d, $J(\text{H,H})=4.2$ Hz, 1H), 7.01 (t, $J(\text{H,H})=4.4$ Hz, 1H), 7.25 (t, $J(\text{H,H})=8$ Hz, 4H), 7.43 (m, 1H), 7.57 ppm (d, $J(\text{H,H})=8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.51, 28.13, 28.61, 32.71, 36.01, 40.85, 50.33, 52.22, 116.05, 127.17, 127.74, 127.93, 129.31, 129.53, 130.48, 130.59, 133.52, 135.46, 142.57, 165.41, 166.26, 167.44, 195.01 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{25}\text{ClNO}_4$ $[\text{M}+\text{H}]^+$: 438.1458, found: 438.1449.



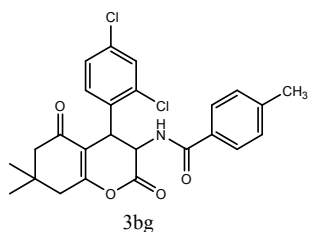
***N*-(4-(3-bromophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[17]**

IR (potassium bromide, ν , cm^{-1}): 1014, 1082, 1113, 1159, 1205, 1281, 1367, 1472, 1539, 1644, 1663, 1780, 2818, 2884, 2927, 2960, 3060, 3032, 3265, 3435. ^1H NMR (400 MHz, CDCl_3): δ =1.18 (s, 3H), 1.23 (s, 3H), 2.36 (d, $J(\text{H,H})=16$ Hz, 1H), 2.41 (d, $J(\text{H,H})=16$ Hz, 1H), 2.41 (s, 3H), 2.65 (s, 2H), 4.68 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.37 (t, $J(\text{H,H})=7.4$ Hz, 1H), 6.32 (d, $J(\text{H,H})=7.2$ Hz, 1H), 7.02 (d, $J(\text{H,H})=7.8$ Hz, 1H), 7.21 (t, $J(\text{H,H})=7.8$ Hz, 1H), 7.25 (m, 2H), 7.44 (d, $J(\text{H,H})=7.9$ Hz, 1H), 7.58 ppm (d, $J(\text{H,H})=6.4$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.54, 28.18, 28.57, 32.73, 38.89, 40.72, 50.33, 52.53, 115.54, 123.28, 126.57, 127.11, 129.83, 130.23, 130.69, 131.14, 131.54, 137.73, 142.88, 165.09, 166.27, 167.19, 195.20 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{25}\text{H}_{25}\text{BrNO}_4$ $[\text{M}+\text{H}]^+$: 482.0961, found: 482.0959.



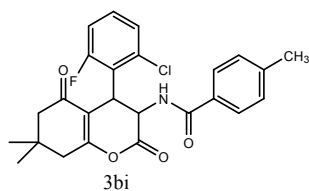
***N*-(4-(2-bromophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[18]**

1002, 1081, 1115, 1161, 1289, 1347, 1371, 1437, 1471, 1504, 1537, 1656, 1807, 2867, 2927, 2954, 3031, 3287, 3442, 3596. ^1H NMR (400 MHz, CDCl_3): δ =1.14 (s, 3H), 1.22 (s, 3H), 2.32 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (s, 3H), 2.63 (d, $J(\text{H,H})=18$ Hz, 1H), 2.69 (d, $J(\text{H,H})=18$ Hz, 1H), 5.25 (d, $J(\text{H,H})=7.8$ Hz, 1H) 5.57 (t, $J(\text{H,H})=8$ Hz, 1H), 6.17 (d, $J(\text{H,H})=8$ Hz, 1H), 6.98 (dd, $J(\text{H,H})=1.5, 7.8$ Hz, 1H), 7.19 (td, $J(\text{H,H})=1.5, 7.8$ Hz, 1H), 7.23 (d, $J(\text{H,H})=7.9$ Hz, 2H), 7.30 (t, $J(\text{H,H})=7.5$ Hz, 1H), 7.59 (d, $J(\text{H,H})=8$ Hz, 2H), 7.64 ppm (dd, $J(\text{H,H})=1.5, 6$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.53, 28.12, 28.59, 32.75, 38.48, 40.81, 50.28, 52.11, 116.38, 126.19, 127.25, 127.65, 128.45, 129.31, 129.80, 130.58, 133.93, 135.33, 142.59, 165.36, 166.25, 167.48, 195.07 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{25}\text{H}_{25}\text{BrNO}_4$ $[\text{M}+\text{H}]^+$: 482.0961, found: 482.0959.



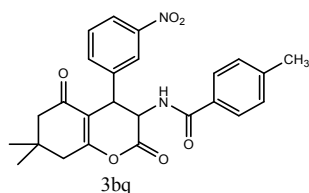
***N*-(4-(2,4-dichlorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[19]**

IR (potassium bromide, ν , cm^{-1}): 1005, 1055, 1086, 1112, 1132, 1161, 1285, 1365, 1485, 1534, 1582, 1661, 1791, 2871, 2963, 3057, 3292, 3439. ^1H NMR (400 MHz, CDCl_3): δ =1.05 (s, 3H), 1.13 (s, 3H), 2.23 (d, $J(\text{H,H})=16$ Hz, 1H), 2.31 (d, $J(\text{H,H})=16$ Hz, 1H), 2.31 (s, 3H), 2.53 (d, $J(\text{H,H})=18$ Hz, 1H), 2.59 (d, $J(\text{H,H})=18$ Hz, 1H), 5.15 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.44 (td, $J(\text{H,H})=2.4, 7.6$ Hz, 1H), 6.21 (dd, $J(\text{H,H})=1.2, 7.6$ Hz, 1H), 6.84 (dd, $J(\text{H,H})=1.5, 8.4$ Hz, 1H), 7.15 (t, $J(\text{H,H})=7.6$ Hz, 2H), 7.37 (t, $J(\text{H,H})=8.4$ Hz, 2H), 7.48 (d, $J(\text{H,H})=8.4$ Hz, 1H), 7.59 ppm (d, $J(\text{H,H})=7.6$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.07, 28.61, 32.72, 35.77, 40.80, 50.26, 52.09, 52.14, 115.74, 127.15, 128.01, 128.72, 129.37, 130.26, 130.41, 132.15, 142.73, 165.49, 166.05, 166.14, 167.45, 167.57, 195.91 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{25}\text{H}_{24}\text{Cl}_2\text{NO}_4$ $[\text{M}+\text{H}]^+$: 472.1019, found: 472.1008.



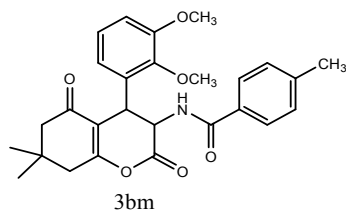
***N*-(4-(2-chloro-6-fluorophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[20]**

IR (potassium bromide, ν , cm^{-1}): 1017, 1062, 1091, 1118, 1161, 1213, 1240, 1276, 1349, 1372, 1454, 1536, 1574, 1608, 1657, 1808, 1932, 2871, 2960, 3088, 3286, 3435. ^1H NMR (400 MHz, CDCl_3): δ =1.09 (s, 3H), 1.19 (s, 3H), 2.95 (d, $J(\text{H,H})=16$ Hz, 1H), 2.36 (d, $J(\text{H,H})=16$ Hz, 1H), 2.38 (s, 3H), 2.58 (d, $J(\text{H,H})=18$ Hz, 1H), 2.63 (d, $J(\text{H,H})=18$ Hz, 1H), 5.42 (d, $J(\text{H,H})=9$ Hz, 1H) 5.51 (t, $J(\text{H,H})=9$ Hz, 1H), 6.35 (d, $J(\text{H,H})=7.4$ Hz, 1H), 6.97 (t, $J(\text{H,H})=8.8$ Hz, 1H), 7.22 (m, 4H), 7.49 ppm (d, $J(\text{H,H})=8$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.50, 27.90, 28.49, 32.60, 34.66, 40.98, 50.09, 50.39, 113.01, 114.54, 114.77, 121.83, 121.99, 126.33, 126.36, 127.09, 129.29, 129.74, 129.85, 130.49, 142.61, 160.74, 163.19, 165.63, 165.65, 166.13, 167.39, 195.89 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{25}\text{H}_{24}\text{ClFNO}_4$ $[\text{M}+\text{H}]^+$: 456.1325, found: 456.1320.



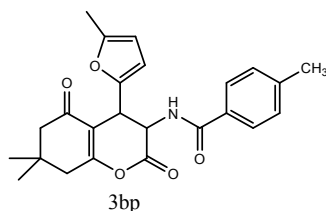
***N*-(4-(3-nitrophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[21]**

IR (potassium bromide, ν , cm^{-1}): 1015, 1083, 1112, 1160, 1283, 1352, 1530, 1641, 1662, 1782, 2818, 2888, 2928, 2964, 3032, 3269, 3375, 3551. ^1H NMR (400 MHz, CDCl_3): δ =1.19 (s, 3H), 1.25 (s, 3H), 2.37 (d, $J(\text{H,H})=16$ Hz, 1H), 2.41 (s, 3H), 2.44 (d, $J(\text{H,H})=16$ Hz, 1H), 2.69 (s, 2H), 4.95 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.37 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.43 (d, $J(\text{H,H})=6.2$ Hz, 1H), 7.24 (d, $J(\text{H,H})=7.9$ Hz, 1H), 7.44 (d, $J(\text{H,H})=6.4$ Hz, 1H), 7.51 (t, $J(\text{H,H})=7.8$ Hz, 1H), 7.58 (d, $J(\text{H,H})=8.2$ Hz, 2H), 7.94 (t, $J(\text{H,H})=2$ Hz, 2H), 8.17 ppm (d, $J(\text{H,H})=8$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.53, 28.06, 28.68, 32.78, 38.71, 40.73, 50.30, 52.77, 115.15, 122.51, 123.37, 127.05, 129.58, 129.92, 130.05, 134.61, 137.77, 143.12, 148.69, 165.50, 166.09, 167.26, 195.17 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{25}\text{H}_{25}\text{N}_2\text{O}_6$ $[\text{M}+\text{H}]^+$: 433.4764, found: 433.4751.



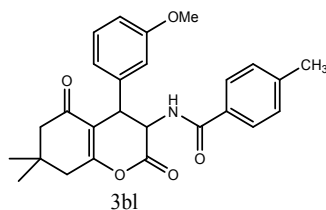
***N*-(3,4,5,6,7,8-hexahydro-4-(2,3-dimethoxyphenyl)-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[22]**

IR (potassium bromide, ν , cm^{-1}): 1005, 1117, 1165, 1218, 1280, 1367, 1482, 1534, 1661, 1790, 2880, 2891, 2955, 3197, 3289, 3544. ^1H NMR (400 MHz, CDCl_3): δ =1.14 (s, 3H), 1.19 (s, 3H), 2.29 (d, $J(\text{H,H})$ =16 Hz, 1H), 2.37 (d, $J(\text{H,H})$ =16 Hz, 1H), 2.37 (s, 3H), 2.59 (s, 2H), 3.85 (s, 3H), 3.89 (s, 3H), 4.94 (d, $J(\text{H,H})$ =7.6 Hz, 1H), 5.34 (t, $J(\text{H,H})$ =7.6 Hz, 1H), 6.41 (d, $J(\text{H,H})$ =7.6 Hz, 1H), 6.69 (d, $J(\text{H,H})$ =7.6 Hz, 1H), 6.87 (dd, $J(\text{H,H})$ =1.5, 8 Hz, 1H), 6.99 (t, $J(\text{H,H})$ =8 Hz, 1H), 7.19 (d, $J(\text{H,H})$ =8 Hz, 2H), 7.53 ppm (d, $J(\text{H,H})$ =8 Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.46, 28.20, 28.55, 32.61, 35.02, 41.00, 50.57, 51.77, 55.19, 60.24, 112.72, 115.43, 121.33, 124.51, 127.15, 129.18, 130.19, 142.22, 147.25, 152.78, 164.77, 165.81, 167.42, 195.88 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{27}\text{H}_{30}\text{NO}_6$ $[\text{M}+\text{H}]^+$: 464.2073, found: 464.2069.



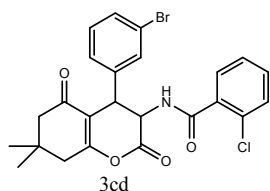
***N*-(3,4,5,6,7,8-hexahydro-7,7-dimethyl-4-(5-methylfuran-2-yl)-2,5-dioxo-2*H*-chromen-3-yl)methylbenzamide:[23]**

IR (potassium bromide, ν , cm^{-1}): 1018, 1094, 1117, 1156, 1206, 1275, 1319, 1355, 1501, 1530, 1655, 1780, 1924, 2822, 2874, 2915, 2962, 3141, 3292, 3375, 3544. ^1H NMR (400 MHz, CDCl_3): δ =1.13 (s, 3H), 1.92 (s, 3H), 2.21 (s, 3H), 2.35 (s, 2H), 2.42 (s, 3H), 2.56 (s, 2H), 4.69 (d, $J(\text{H,H})$ =7 Hz, 1H), 5.27 (t, $J(\text{H,H})$ =7.3 Hz, 1H), 5.87 (d, $J(\text{H,H})$ =3 Hz, 1H), 6.09 (d, $J(\text{H,H})$ =3 Hz, 1H), 6.57 (d, $J(\text{H,H})$ =7.6 Hz, 1H), 7.26 (d, $J(\text{H,H})$ =8 Hz, 2H), 7.65 ppm (d, $J(\text{H,H})$ =8 Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =13.62, 21.52, 21.98, 28.52, 32.70, 33.39, 40.78, 50.34, 51.40, 106.74, 110.06, 113.97, 127.14, 129.34, 130.51, 142.67, 147.06, 152.71, 165.21, 166.34, 167.09, 195.14 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{26}\text{NO}_5$ $[\text{M}+\text{H}]^+$: 408.1805, found: 408.1809.



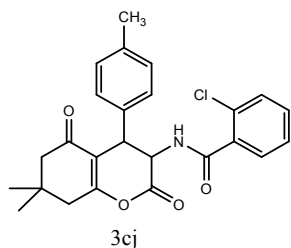
***N*-(3,4,5,6,7,8-hexahydro-4-(3-methoxyphenyl)-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[24]**

IR (potassium bromide, ν , cm^{-1}): 1011, 1046, 1090, 1135, 1205, 1265, 1367, 1454, 1492, 1535, 1582, 1609, 1660, 1687, 2832, 2871, 2931, 2956, 3025, 3293, 3439. ^1H NMR (400 MHz, CDCl_3): δ =1.17 (s, 3H), 1.22 (s, 3H), 2.34 (d, $J(\text{H,H})=16$ Hz, 1H), 2.40 (d, $J(\text{H,H})=16$ Hz, 1H), 2.4 (s, 3H), 2.63 (s, 2H), 3.7 (s, 3H), 4.63 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.39 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.28 (d, $J(\text{H,H})=7.6$ Hz, 1H), 6.64 (s, 1H), 6.69 (d, $J(\text{H,H})=7.6$ Hz, 1H), 7.25 (t, $J(\text{H,H})=7.6$ Hz, 3H), 7.6 ppm (d, $J(\text{H,H})=8.2$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.49, 28.14, 28.61, 32.66, 39.32, 40.75, 50.39, 52.51, 55.13, 113.66, 113.91, 116.05, 119.88, 127.09, 129.34, 130.24, 130.40, 136.91, 142.71, 160.12, 164.79, 166.49, 166.99, 195.27 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{26}\text{H}_{28}\text{NO}_5$ $[\text{M}+\text{H}]^+$: 434.1962, found: 434.1960.



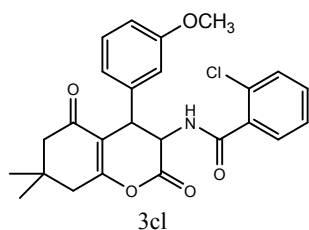
***N*-(4-(3-bromophenyl)-3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)-4-chlorobenzamide:[25]**

IR (potassium bromide, ν , cm^{-1}): 1055, 1085, 1112, 1135, 1161, 1204, 1285, 1364, 1485, 1534, 1582, 1661, 1791, 2826, 2871, 2964, 3026, 3057, 3197, 3292, 3438, 3551. ^1H NMR (400 MHz, CDCl_3): δ =1.51 (s, 3H), 1.21 (s, 3H), 2.34 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (d, $J(\text{H,H})=16$ Hz, 1H), 2.63 (s, 2H), 4.74 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.36 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.62 (d, $J(\text{H,H})=6.8$ Hz, 1H), 7.05 (d, $J(\text{H,H})=7.6$ Hz, 1H), 7.19 (t, $J(\text{H,H})=7.6$ Hz, 1H), 7.32 (m, 1H), 7.42 (m, 2H), 7.61 ppm (d, $J(\text{H,H})=7.2$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.19, 28.46, 32.71, 38.77, 40.71, 50.31, 52.96, 115.61, 123.27, 126.29, 127.14, 130.26, 130.46, 130.72, 130.97, 131.54, 131.66, 131.96, 133.39, 137.79, 164.90, 165.74, 166.24, 195.16 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{22}\text{BrClNO}_4$ $[\text{M}+\text{H}]^+$: 502.0360, found: 502.0361.



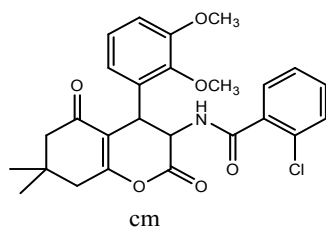
2-chloro-N-(3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-2H-chromen-3-yl)benzamide:[26]

IR (potassium bromide, ν , cm^{-1}): 1005, 1056, 1092, 1161, 1347, 1368, 1430, 1467, 1509, 1682, 1777, 1901, 2866, 2963, 3026, 3330, 3408, 3523. ^1H NMR (400 MHz, CDCl_3): δ =1.09 (s, 3H), 1.16 (s, 3H), 2.25 (s, 3H), 2.26 (d, $J(\text{H,H})=16$ Hz, 1H), 2.34 (d, $J(\text{H,H})=16$ Hz, 1H), 2.58 (s, 2H), 4.62 (d, $J(\text{H,H})=7.4$ Hz, 1H) 5.36 (t, $J(\text{H,H})=7.4$ Hz, 1H), 6.58 (d, $J(\text{H,H})=7.4$ Hz, 1H), 7.03 (d, $J(\text{H,H})=8$ Hz, 2H), 7.09 (d, $J(\text{H,H})=8$ Hz, 2H), 7.27 (m, 1H), 7.33 (s, 1H), 7.55 ppm (d, $J(\text{H,H})=7.2$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.06, 28.09, 28.50, 32.61, 38.82, 40.65, 50.32, 52.22, 116.29, 127.05, 127.95, 129.91, 130.17, 130.35, 130.94, 131.81, 132.29, 133.62, 138.06, 164.60, 166.12, 166.16, 195.38 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{25}\text{H}_{25}\text{ClNO}_4$ $[\text{M}+\text{H}]^+$: 438.1458, found: 438.1449.



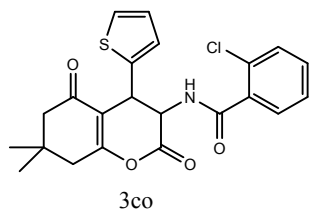
2-chloro-N-(3,4,5,6,7,8-hexahydro-4-(3-methoxyphenyl)-7,7-dimethyl-2,5-dioxo-2H-chromen-3-yl)benzamide:[27]

IR (potassium bromide, ν , cm^{-1}): 1046, 1090, 1136, 1265, 1367, 1535, 1582, 1609, 1660, 1787, 2832, 2871, 2931, 2956, 3064, 3294, 3442. ^1H NMR (400 MHz, CDCl_3): δ =1.15 (s, 3H), 1.21 (s, 3H), 2.34 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (d, $J(\text{H,H})=16$ Hz, 1H), 2.62 (s, 2H), 3.75 (s, 3H), 4.68 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.39 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.55 (d, $J(\text{H,H})=7.2$ Hz, 1H), 6.73 (m, 2H), 6.82 (dd, $J(\text{H,H})=3, 8.2$ Hz, 1H), 7.25 (t, $J(\text{H,H})=7.9$ Hz, 1H), 7.36 (m, 1H), 7.39 ppm (dd, $J(\text{H,H})=1.4, 8$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.18, 28.48, 32.68, 39.28, 40.75, 50.38, 52.85, 55.20, 113.81, 114.10, 116.15, 119.93, 127.10, 130.30, 130.32, 130.45, 131.02, 131.89, 133.50, 136.93, 160.13, 164.62, 165.97, 166.11, 195.26 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{25}\text{H}_{25}\text{ClNO}_5$ $[\text{M}+\text{H}]^+$: 454.1341, found: 454.1345.



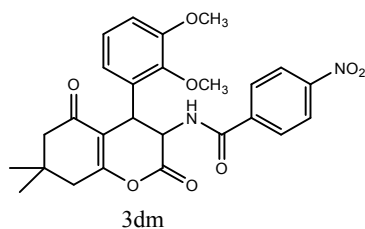
2-chloro-*N*-(3,4,5,6,7,8-hexahydro-4-(2,3-dimethoxyphenyl)-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)benzamide:[28]

IR (potassium bromide, ν , cm^{-1}): 875, 1065, 1106, 1167, 1318, 1278, 1369, 1479, 1533, 1584, 1657, 1792, 2512, 2829, 2878, 2938, 2963, 3063, 3287. ^1H NMR (400 MHz, CDCl_3): δ =1.14 (s, 3H), 1.19 (s, 3H), 2.29 (d, $J(\text{H,H})=16$ Hz, 1H), 2.36 (d, $J(\text{H,H})=16$ Hz, 1H), 2.58 (s, 2H), 2.87 (s, 3H), 3.89 (s, 3H), 4.98 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.36 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.57 (d, $J(\text{H,H})=7.2$ Hz, 1H), 6.79 (d, $J(\text{H,H})=7.6$ Hz, 1H), 6.86 (d, $J(\text{H,H})=8$ Hz, 1H), 7.00 (t, $J(\text{H,H})=8$ Hz, 1H), 7.28 (m, 1H), 7.34 (d, $J(\text{H,H})=5$ Hz, 2H), 7.55 ppm (d, $J(\text{H,H})=7.6$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.07, 28.65, 32.59, 35.60, 41.05, 50.63, 51.69, 55.78, 59.98, 112.66, 115.02, 124.28, 126.94, 128.97, 130.24, 130.37, 131.02, 131.55, 133.81, 147.33, 152.60, 164.29, 165.33, 166.19, 195.17 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{26}\text{H}_{27}\text{ClNO}_6$ $[\text{M}+\text{H}]^+$: 484.1493, found: 484.1489.



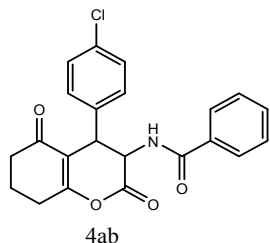
2-chloro-*N*-(3,4,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxo-4(thiophen-2-yl)-2*H*-chromen-3-yl)benzamide:[29]

IR (potassium bromide, ν , cm^{-1}): 1000, 1080, 1116, 1154, 1204, 1288, 1354, 1541, 1578, 1645, 1787, 2815, 2892, 2961, 3067, 3105, 3253, 3435. ^1H NMR (400 MHz, CDCl_3): δ =1.17 (s, 3H), 1.21 (s, 3H), 2.36 (d, $J(\text{H,H})=16$ Hz, 1H), 2.41 (d, $J(\text{H,H})=16$ Hz, 1H), 2.59 (d, $J(\text{H,H})=18$ Hz, 1H), 2.61 (d, $J(\text{H,H})=18$ Hz, 1H), 5.03 (d, $J(\text{H,H})=7.2$ Hz, 1H) 5.37 (t, $J(\text{H,H})=7.2$ Hz, 1H), 6.77 (d, $J(\text{H,H})=7.2$ Hz, 1H), 6.93 (d, $J(\text{H,H})=3.2$ Hz, 1H), 6.97 (dd, $J(\text{H,H})=3.6, 5.2$ Hz, 1H), 7.21 (d, $J(\text{H,H})=4.8$ Hz, 1H), 7.39 (m, 1H), 7.42 (t, $J(\text{H,H})=7.2$ Hz, 1H), 7.71 ppm (d, $J(\text{H,H})=7.6$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =27.95, 28.73, 32.66, 34.45, 40.71, 50.32, 53.25, 116.56, 125.64, 126.75, 127.15, 127.67, 130.37, 130.50, 131.08, 131.98, 133.44, 137.88, 164.66, 165.64, 166.15, 195.98 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{22}\text{H}_{21}\text{ClNO}_4\text{S}$ $[\text{M}+\text{H}]^+$: 430.0880, found: 430.0882.



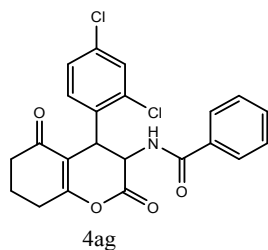
***N*-(3,4,5,6,7,8-hexahydro-4-(2,3-dimethoxyphenyl)-7,7-dimethyl-2,5-dioxo-2*H*-chromen-3-yl)nitrobenzamide:[30]**

IR (potassium bromide, ν , cm^{-1}): 998, 1065, 1100, 1223, 1284, 1346, 1368, 1481, 1526, 1600, 1650, 1785, 1792, 2832, 2944, 2963, 3004, 3071, 3105, 3283, 3551. ^1H NMR (400 MHz, CDCl_3): δ =1.17 (s, 3H), 1.22 (s, 3H), 2.32 (d, $J(\text{H,H})=16$ Hz, 1H), 2.39 (d, $J(\text{H,H})=16$ Hz, 1H), 2.63 (s, 2H), 3.87 (s, 3H), 3.911 (s, 3H), 4.99 (d, $J(\text{H,H})=7.6$ Hz, 1H), 5.32 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.54 (d, $J(\text{H,H})=7.2$ Hz, 1H), 6.66 (d, $J(\text{H,H})=7.6$ Hz, 1H), 6.90 (d, $J(\text{H,H})=7.2$ Hz, 1H), 7.03 (t, $J(\text{H,H})=7.6$ Hz, 1H), 7.79 (d, $J(\text{H,H})=9$ Hz, 2H), 8.24 ppm (d, $J(\text{H,H})=9$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =28.29, 28.49, 32.68, 40.93, 42.03, 50.51, 52.21, 55.75, 60.38, 112.69, 115.29, 123.82, 123.91, 124.90, 128.32, 128.41, 128.61, 139.27, 146.99, 149.71, 152.93, 165.02, 165.36, 165.60, 195.60 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{26}\text{H}_{27}\text{N}_2\text{O}_8$ $[\text{M}+\text{H}]^+$: 495.1767, found: 495.1765.



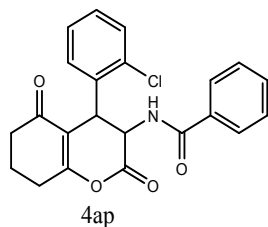
***N*-(4-(4-chlorophenyl)3,4,5,6,7,8-hexahydro-4-2,5-dioxo-2*H*-chromen-3-yl)benzamide^[2]:[31]**

IR (potassium bromide, ν , cm^{-1}): 1051, 1089, 1115, 1147, 1244, 1293, 1365, 1489, 1544, 1642, 1658, 1784, 1894, 2832, 2876, 2922, 3058, 3186, 3250, 3540. ^1H NMR (400 MHz, CDCl_3): δ =2.27 (m, 2H), 2.56 (m, 2H), 2.85 (m, 2H), 4.72 (d, $J(\text{H,H})=7.5$ Hz, 1H), 5.37 (t, $J(\text{H,H})=7.3$ Hz, 1H), 6.36 (d, $J(\text{H,H})=7$ Hz, 2H), 7.05 (d, $J(\text{H,H})=8$ Hz, 2H), 7.3 (d, $J(\text{H,H})=8$ Hz, 2H), 7.47 (t, $J(\text{H,H})=7.8$ Hz, 2H), 7.57 (tt, $J(\text{H,H})=1.2$, 8 Hz, 1H), 7.68 ppm (d, $J(\text{H,H})=7.8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.58, 27.01, 36.44, 38.73, 52.57, 117.13, 127.06, 128.79, 129.37, 132.29, 133.02, 133.92, 134.28, 166.22, 166.47, 167.11, 195.27 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{22}\text{H}_{19}\text{ClNO}_4$ $[\text{M}+\text{H}]^+$: 396.0998, found: 396.0994.



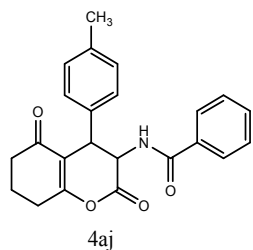
***N*-(4-(2,4-dichlorophenyl)3,4,5,6,7,8-hexahydro-4,2,5-dioxo-2*H*-chromen-3-yl)benzamide:[32]**

IR (potassium bromide, ν , cm^{-1}): 1058, 1093, 1116, 1146, 1287, 1365, 1427, 1473, 1541, 1637, 1665, 1780, 1893, 2822, 2916, 3057, 3246, 3435, 3547. ^1H NMR (400 MHz, CDCl_3): δ =2.27 (m, 2H), 2.56 (m, 2H), 2.84 (m, 2H), 5.24 (d, $J(\text{H,H})=7.8$ Hz, 1H) 5.53 (t, $J(\text{H,H})=7.8$ Hz, 1H), 6.33 (d, $J(\text{H,H})=7.6$ Hz, 1H), 6.93 (d, $J(\text{H,H})=8$ Hz, 1H), 7.23 (dd, $J(\text{H,H})=2, 8.4$ Hz, 2H), 7.46 (m, 3H), 7.56 (tt, $J(\text{H,H})=1.2, 7.4$ Hz, 1H) 7.68 ppm (d, $J(\text{H,H})=7.8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.55, 27.07, 35.82, 36.35, 52.11, 117.02, 127.16, 128.02, 128.12, 128.87, 130.19, 132.16, 132.21, 133.25, 134.72, 136.27, 165.91, 167.71, 167.58, 195.03 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{22}\text{H}_{18}\text{Cl}_2\text{NO}_4$ $[\text{M}+\text{H}]^+$: 430.0582, found: 430.0585.



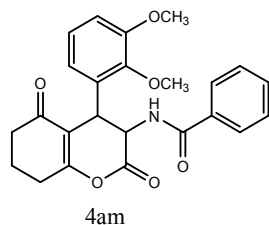
***N*-(4-(2-chlorophenyl)3,4,5,6,7,8-hexahydro-4,2,5-dioxo-2*H*-chromen-3-yl)benzamide^[2]:[33]**

IR (potassium bromide, ν , cm^{-1}): 1041, 1094, 1122, 1147, 1286, 1348, 1369, 1421, 1440, 1476, 1540, 1644, 1790, 1900, 2779, 2822, 2911, 3032, 3060, 3278, 3446, 3549. ^1H NMR (400 MHz, CDCl_3): δ =2.25 (m, 2H), 2.54 (m, 2H), 2.84 (m, 2H), 5.26 (d, $J(\text{H,H})=7.9$ Hz, 1H) 5.55 (t, $J(\text{H,H})=7.9$ Hz, 1H), 6.29 (d, $J(\text{H,H})=7.9$ Hz, 1H), 6.99 (m, 1H), 7.24 (m, 2H), 7.42 (m, 3H), 7.53 (t, $J(\text{H,H})=6.8$ Hz, 1H) 7.65 ppm (d, $J(\text{H,H})=7.8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.58, 27.08, 36.04, 36.38, 52.17, 117.28, 127.18, 127.78, 127.94, 128.63, 129.55, 130.40, 132.02, 133.45, 135.43, 166.06, 167.08, 167.53, 195.14 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{22}\text{H}_{19}\text{ClNO}_4$ $[\text{M}+\text{H}]^+$: 396.0998, found: 396.0994.



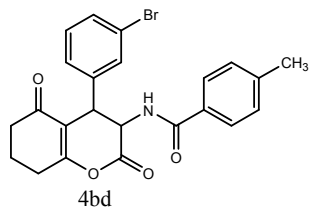
N*-(3,4,5,6,7,8-hexahydro-4-*p*-2*H*-chromen-3-yl)benzamide^[2]:**[34]*

IR (potassium bromide, ν , cm^{-1}): 1112, 1145, 1183, 1283, 1348, 1374, 1490, 1533, 1579, 1650, 1723, 1781, 1956, 2839, 2887, 2928, 3029, 3280, 3381, 3428, 3547. ^1H NMR (400 MHz, CDCl_3): δ =2.22 (m, 2H), 2.32 (s, 3H), 2.54 (m, 2H), 2.84 (m, 2H), 4.62 (d, $J(\text{H,H})=7.5$ Hz, 1H), 5.40 (t, $J(\text{H,H})=7.6$ Hz, 1H), 6.29 (d, $J(\text{H,H})=7.6$ Hz, 1H), 7.01 (d, $J(\text{H,H})=8$ Hz, 2H), 7.14 (d, $J(\text{H,H})=8$ Hz, 2H), 7.45 (t, $J(\text{H,H})=7.8$ Hz, 2H), 7.55 (tt, $J(\text{H,H})=1.2, 7.4$ Hz, 1H), 7.68 ppm (d, $J(\text{H,H})=7.6$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =21.09, 27.01, 36.49, 39.05, 52.51, 117.55, 127.01, 127.84, 128.69, 129.94, 132.11, 133.29, 138.17, 166.28, 166.46, 167.08, 195.40 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{23}\text{H}_{21}\text{NO}_4$ $[\text{M}+\text{H}]^+$: 376.1554, found: 376.1528.



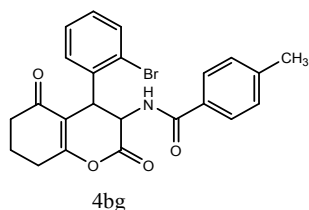
N*-(3,4,5,6,7,8-hexahydro-4-(2,3-dimethoxyphenyl)-2,5-dioxo-2*H*-chromen-3-yl)benzamide:**[35]*

IR (potassium bromide, ν , cm^{-1}): 1147, 1210, 1253, 1300, 1388, 1471, 1506, 1577, 1643, 1669, 1714, 1787, 2899, 2939, 2959, 3974, 3248, 3407. ^1H NMR (400 MHz, CDCl_3): δ =2.22 (m, 2H), 2.53 (m, 2H), 2.76 (t, $J(\text{H,H})=6.2$ Hz, 2H), 3.86 (s, 3H), 2.89 (s, 3H), 4.92 (d, $J(\text{H,H})=8.2$ Hz, 1H), 5.34 (t, $J(\text{H,H})=7.9$ Hz, 1H), 6.45 (d, $J(\text{H,H})=7.2$ Hz, 1H), 6.74 (d, $J(\text{H,H})=7.6$ Hz, 1H), 6.88 (dd, $J(\text{H,H})=1.2, 8.2$ Hz, 1H), 7.00 (t, $J(\text{H,H})=8$ Hz, 1H), 7.40 (t, $J(\text{H,H})=7.9$ Hz, 1H), 7.49 (t, $J(\text{H,H})=7$ Hz, 1H), 7.61 ppm (d, $J(\text{H,H})=7.9$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.46, 27.34, 35.70, 36.65, 51.63, 55.76, 60.05, 112.66, 116.46, 121.91, 124.51, 127.13, 128.54, 129.04, 131.77, 133.63, 147.16, 152.72, 165.58, 166.27, 167.49, 195.83 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{24}\text{H}_{23}\text{NO}_6$ $[\text{M}+\text{H}]^+$: 422.1598, found: 422.1596.



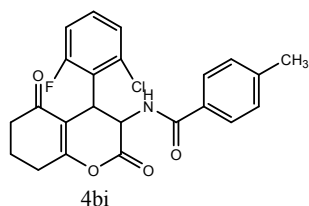
***N*-(4-(3-bromophenyl)-3,4,5,6,7,8-hexahydro-4,2,5-dioxo-2H-chromen-3-yl)-4-methylbenzamide:[36]**

IR (potassium bromide, ν , cm^{-1}): 1105, 1150, 1185, 1240, 1290, 1342, 1504, 1527, 1613, 1668, 1788, 1920, 2730, 2888, 2917, 2951, 3064, 3323, 3378, 3439. ^1H NMR (400 MHz, CDCl_3): δ =2.27 (m, 2H), 2.41 (s, 3H), 2.56 (m, 2H), 2.86 (m, 2H), 4.67 (d, $J(\text{H,H})=7.6$ Hz, 1H) 5.38 (t, $J(\text{H,H})=7.4$ Hz, 1H), 6.33 (d, $J(\text{H,H})=7$ Hz, 1H), 7.03 (d, $J(\text{H,H})=7.8$ Hz, 1H), 7.20 (t, $J(\text{H,H})=7.8$ Hz, 1H), 7.25 (d, $J(\text{H,H})=8.6$ Hz, 3H), 7.44 (d, $J(\text{H,H})=7.6$ Hz, 1H), 7.58 ppm (d, $J(\text{H,H})=8.2$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.57, 21.54, 27.01, 36.43, 38.98, 52.43, 116.77, 123.25, 126.69, 127.11, 129.24, 130.22, 130.65, 131.04, 131.53, 137.72, 142.87, 166.14, 166.73, 167.17, 195.32 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{23}\text{H}_{21}\text{BrNO}_4$ $[\text{M}+\text{H}]^+$: 454.0654, found: 454.0650.



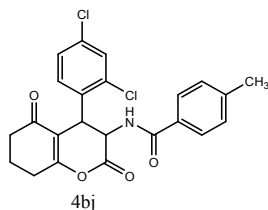
***N*-(4-(2-bromophenyl)-3,4,5,6,7,8-hexahydro-4,2,5-dioxo-2H-chromen-3-yl)-4-methylbenzamide:[37]**

IR (potassium bromide, ν , cm^{-1}): 1051, 1110, 1148, 1184, 1368, 1429, 1535, 1648, 1779, 2832, 2925, 2969, 3029, 3277, 3442. ^1H NMR (400 MHz, CDCl_3): δ =2.23 (m, 2H), 2.39 (s, 3H), 2.55 (m, 2H), 2.79 (m, 2H), 5.3 (d, $J(\text{H,H})=7.9$ Hz, 1H) 5.57 (t, $J(\text{H,H})=7.9$ Hz, 1H), 6.17 (d, $J(\text{H,H})=8$ Hz, 1H), 6.98 (dd, $J(\text{H,H})=1.5$, 7.9 Hz, 1H), 7.18 (td, $J(\text{H,H})=1.5$, 7.9 Hz, 1H), 7.23 (d, $J(\text{H,H})=7.9$ Hz, 2H), 7.30 (td, $J(\text{H,H})=1.2$, 7.4 Hz, 1H), 7.59 (d, $J(\text{H,H})=8.2$ Hz, 2H), 7.63 ppm (dd, $J(\text{H,H})=1.2$, 8 Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.59, 21.53, 27.09, 36.39, 38.60, 52.01, 117.67, 126.19, 127.25, 127.71, 128.46, 129.30, 129.78, 130.59, 133.84, 135.32, 142.57, 166.11, 166.96 167.46, 195.12 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{23}\text{H}_{21}\text{BrNO}_4$ $[\text{M}+\text{H}]^+$: 454.0654, found: 454.0650.



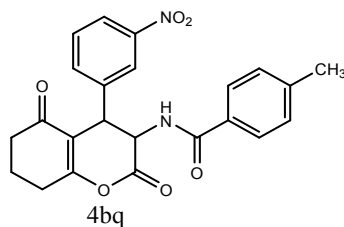
***N*-(4-(2-chloro-6-fluorophenyl)-3,4,5,6,7,8-hexahydro-4,2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[38]**

IR (potassium bromide, ν , cm^{-1}): 1115, 1150, 1187, 1287, 1304, 1367, 1454, 1538, 1613, 1642, 1658, 1799, 1941, 2839, 2927, 2954, 3025, 3253, 3439. ^1H NMR (400 MHz, CDCl_3): δ =2.18 (m, 2H), 2.38 (s, 3H), 2.53 (m, 2H), 2.73 (m, 2H), 5.41 (d, $J(\text{H,H})=9$ Hz, 1H) 5.51 (t, $J(\text{H,H})=7.5$ Hz, 1H), 6.32 (d, $J(\text{H,H})=7.5$ Hz, 1H), 6.98 (m, 1H), 7.22 (m, 3H), 7.49 ppm (d, $J(\text{H,H})=8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.54, 21.50, 27.29, 34.86, 36.50, 50.07, 114.57, 114.79, 121.83, 121.99, 126.27, 126.29, 127.07, 129.31, 129.70, 129.81, 130.50, 142.62, 165.99, 167.20, 167.36, 195.97 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{23}\text{H}_{20}\text{ClFNO}_4$ $[\text{M}+\text{H}]^+$: 428.1087, found: 428.1083.



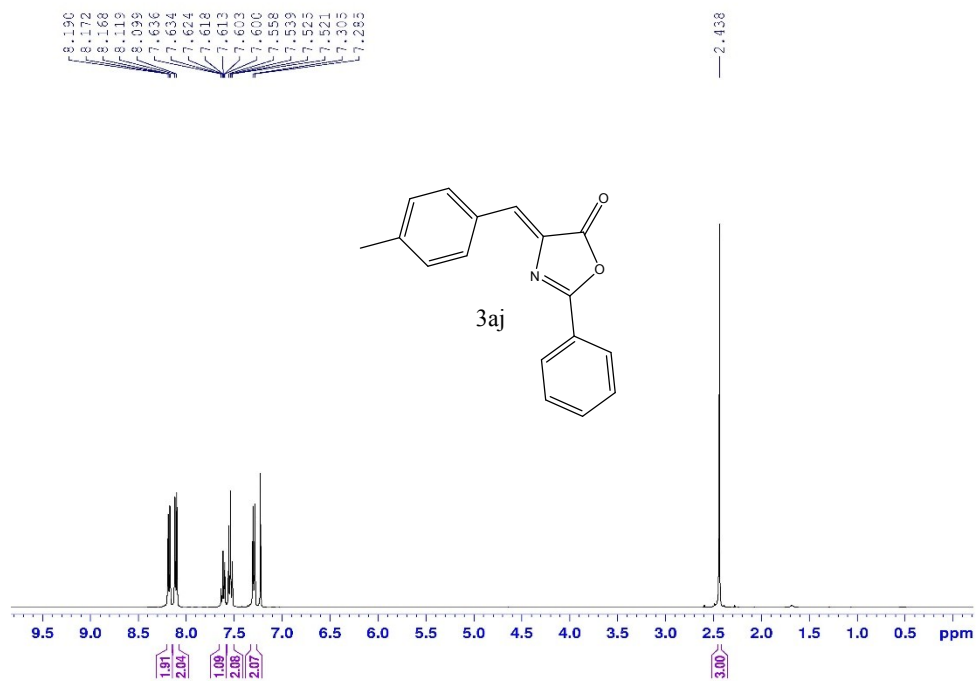
***N*-(4-(2,4-dichlorophenyl)-3,4,5,6,7,8-hexahydro-4,2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[39]**

IR (potassium bromide, ν , cm^{-1}): 1120, 1158, 1238, 1286, 1344, 1363, 1473, 1500, 1539, 1662, 1784, 1883, 2832, 2964, 3081, 3262, 3330, 3412, 3617. ^1H NMR (400 MHz, CDCl_3): δ =2.27 (m, 2H), 2.41 (s, 3H), 2.56 (m, 2H), 2.79 (m, 2H), 5.23 (d, $J(\text{H,H})=7.8$ Hz, 1H) 5.53 (t, $J(\text{H,H})=7.8$ Hz, 1H), 6.26 (d, $J(\text{H,H})=7.8$ Hz, 1H), 6.92 (d, $J(\text{H,H})=8.4$ Hz, 1H), 7.25 (m, 3H), 7.42 (d, $J(\text{H,H})=2.2$ Hz, 1H), 7.58 ppm (d, $J(\text{H,H})=8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.56, 21.53, 27.08, 35.87, 36.36, 52.06, 117.03, 127.16, 127.99, 128.88, 129.38, 130.18, 132.25, 134.68, 136.29, 142.75, 165.99, 167.14, 167.47, 195.02 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{23}\text{H}_{20}\text{Cl}_2\text{NO}_4$ $[\text{M}+\text{H}]^+$: 444.0769, found: 444.0778.

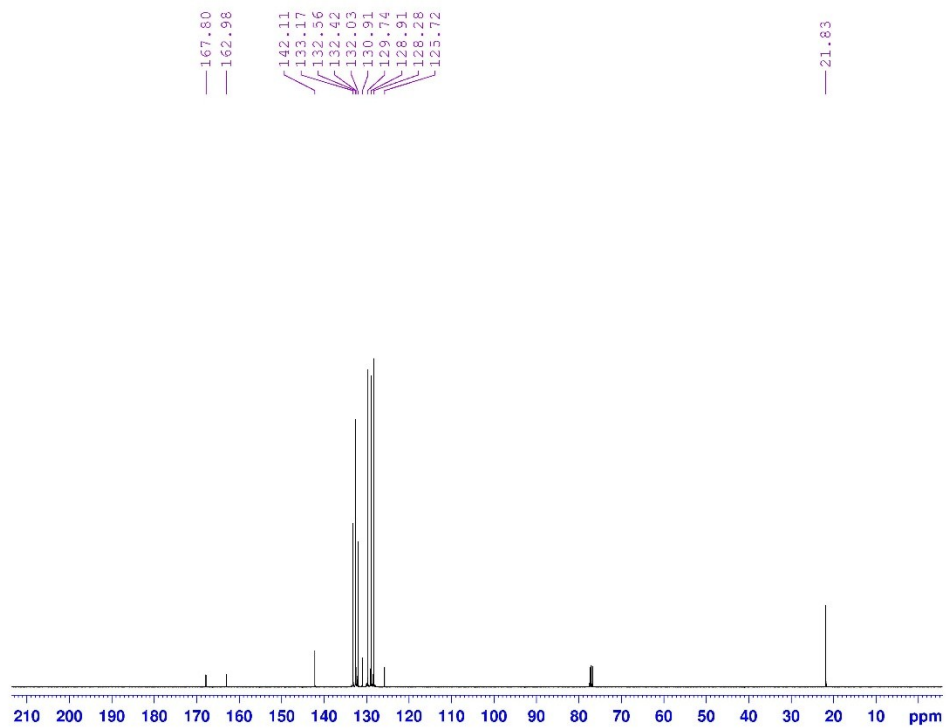


***N*-(4-(3-nitrophenyl)-3,4,5,6,7,8-hexahydro-4,2,5-dioxo-2*H*-chromen-3-yl)-4-methylbenzamide:[40]**

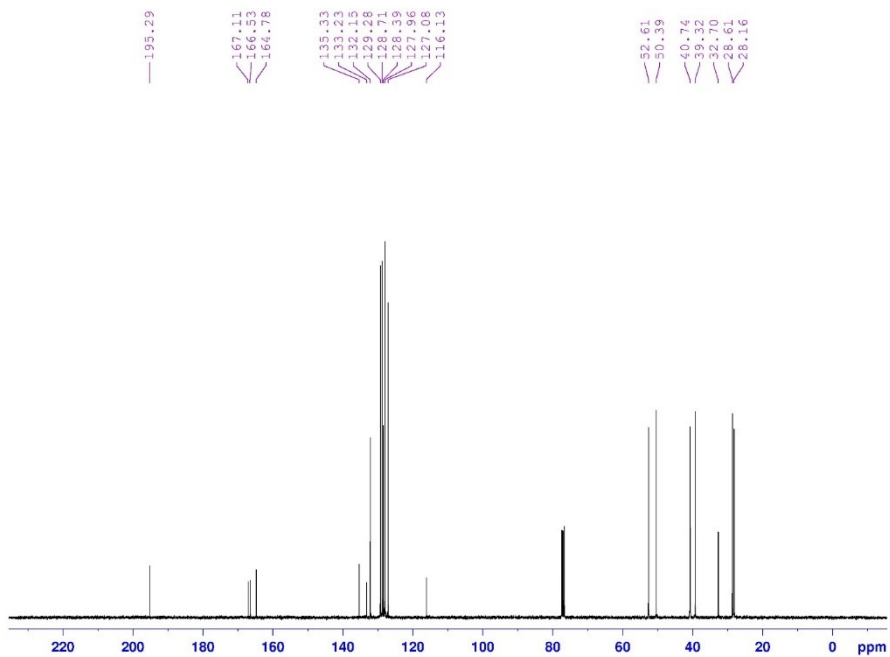
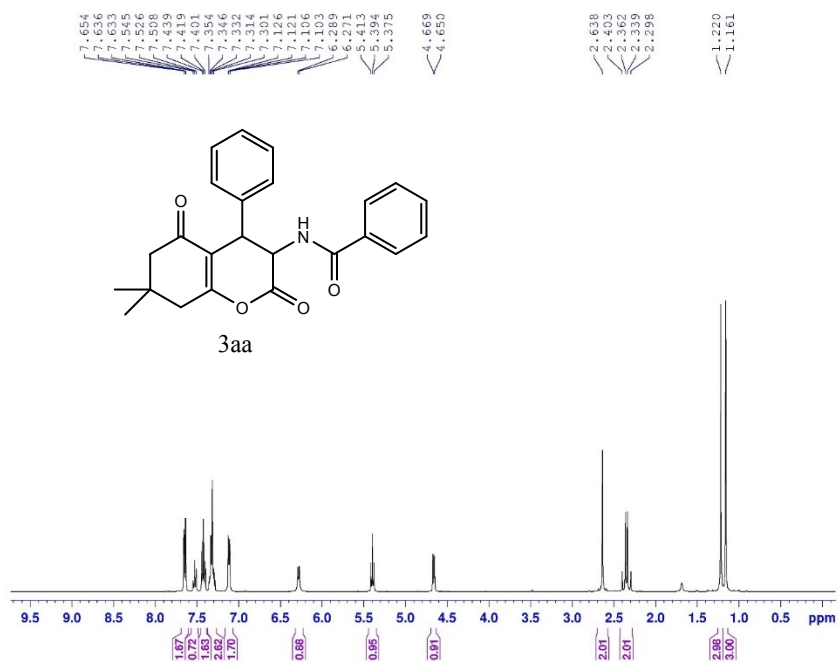
IR (potassium bromide, ν , cm^{-1}): 1087, 1112, 1151, 1186, 1353, 1369, 1536, 1657, 1780, 1795, 1921, 2867, 2927, 3029, 3067, 3266, 3435. ^1H NMR (400 MHz, CDCl_3): δ =2.27 (m, 2H), 2.41 (s, 3H), 2.58 (m, 2H), 2.91 (m, 2H), 4.95 (d, $J(\text{H,H})=7.6$ Hz, 1H), 5.37 (t, $J(\text{H,H})=7$ Hz, 1H), 6.43 (d, $J(\text{H,H})=6.2$ Hz, 1H), 7.24 (d, $J(\text{H,H})=8$ Hz, 2H), 7.44 (d, $J(\text{H,H})=7.8$ Hz, 2H), 7.50 (t, $J(\text{H,H})=7.8$ Hz, 1H), 7.58 (d, $J(\text{H,H})=8$ Hz, 2H), 7.92 (st, $J(\text{H,H})=1, 8$ Hz, 1H), 8.17 ppm (dq, $J(\text{H,H})=1.2, 8$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ =20.59, 21.53, 27.03, 36.39, 38.82, 52.64, 116.37, 122.55, 123.36, 127.05, 129.48, 129, 92, 130.01, 134.63, 137.74, 143.11, 148.67, 165.96, 167.13, 167.26, 195.27 ppm; HRMS (ESI) m/z calcd. for $\text{C}_{23}\text{H}_{20}\text{N}_2\text{O}_6$ $[\text{M}+\text{H}]^+$: 421.1350, found: 421.1384.

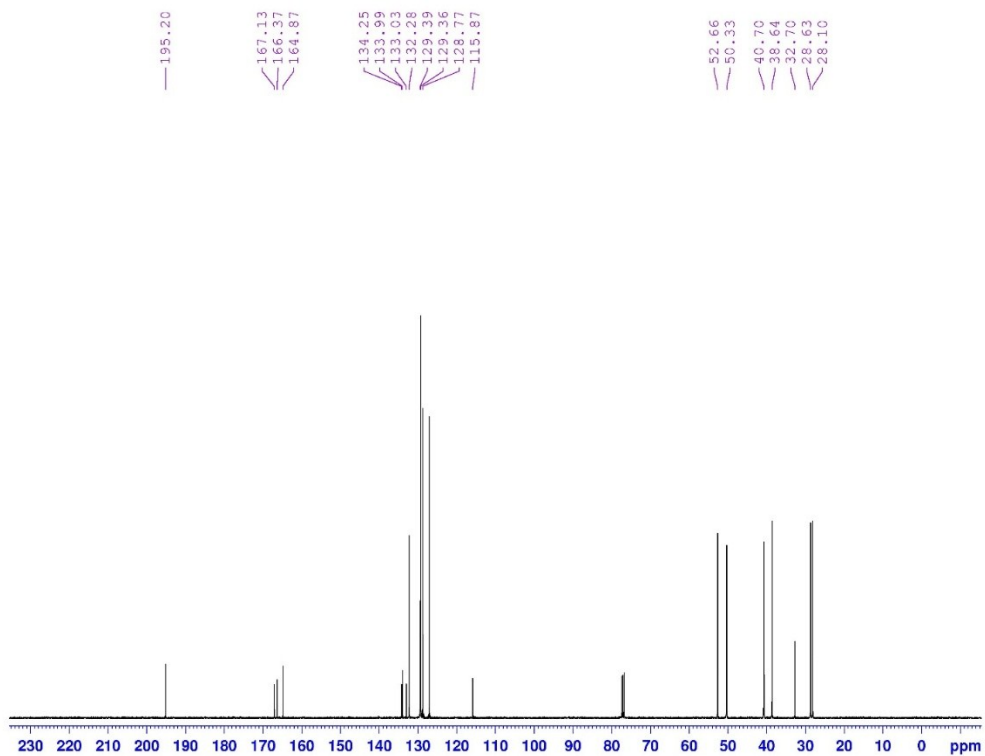
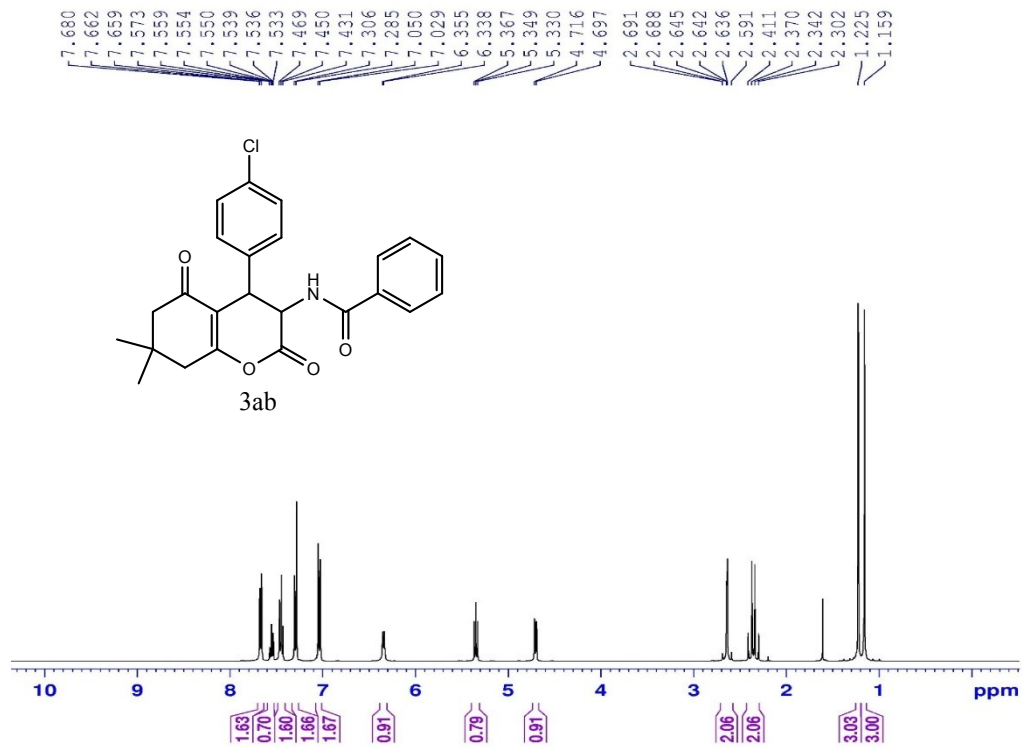


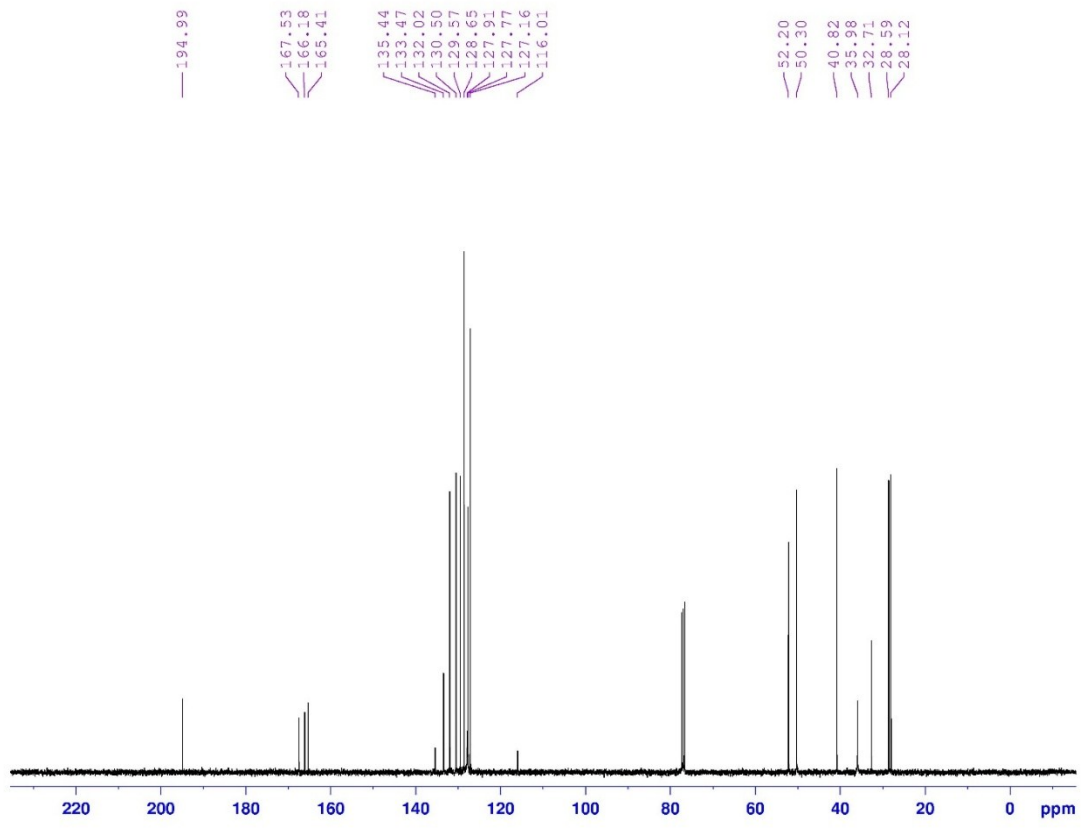
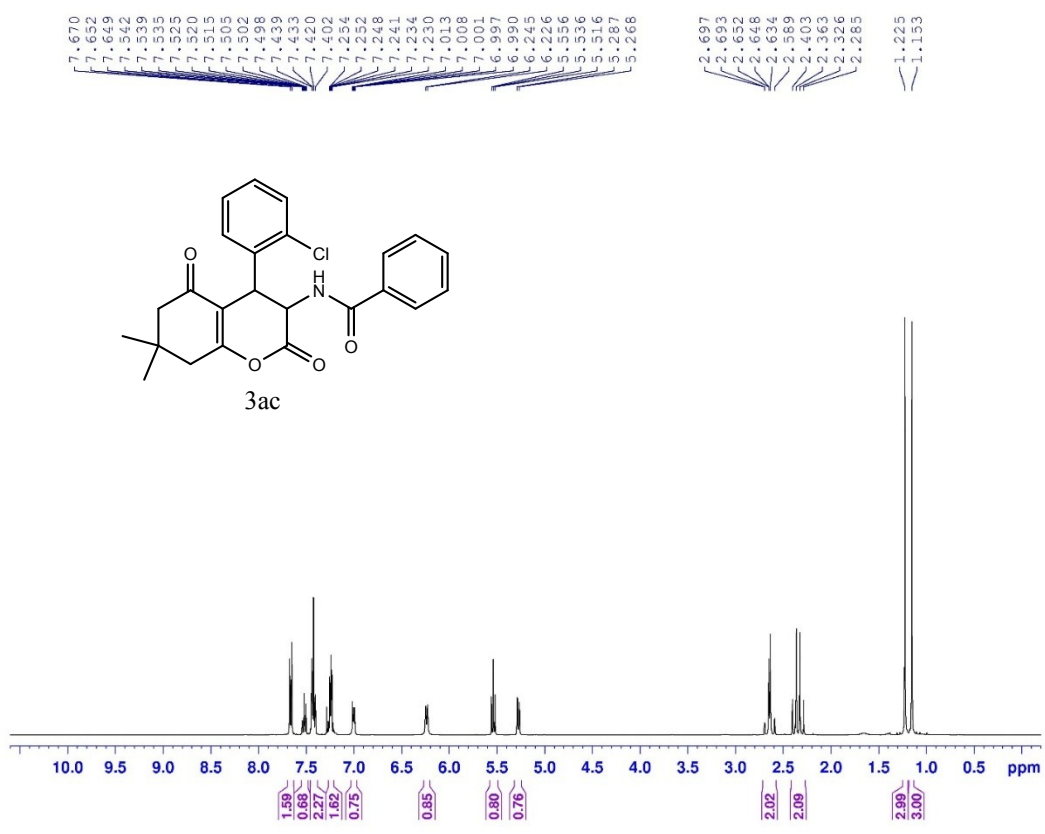
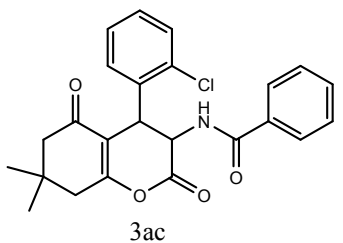
NMR spectra for (Z)-4-(4-methylbenzylidene)-2-phenyloxazol-5(4H)-one

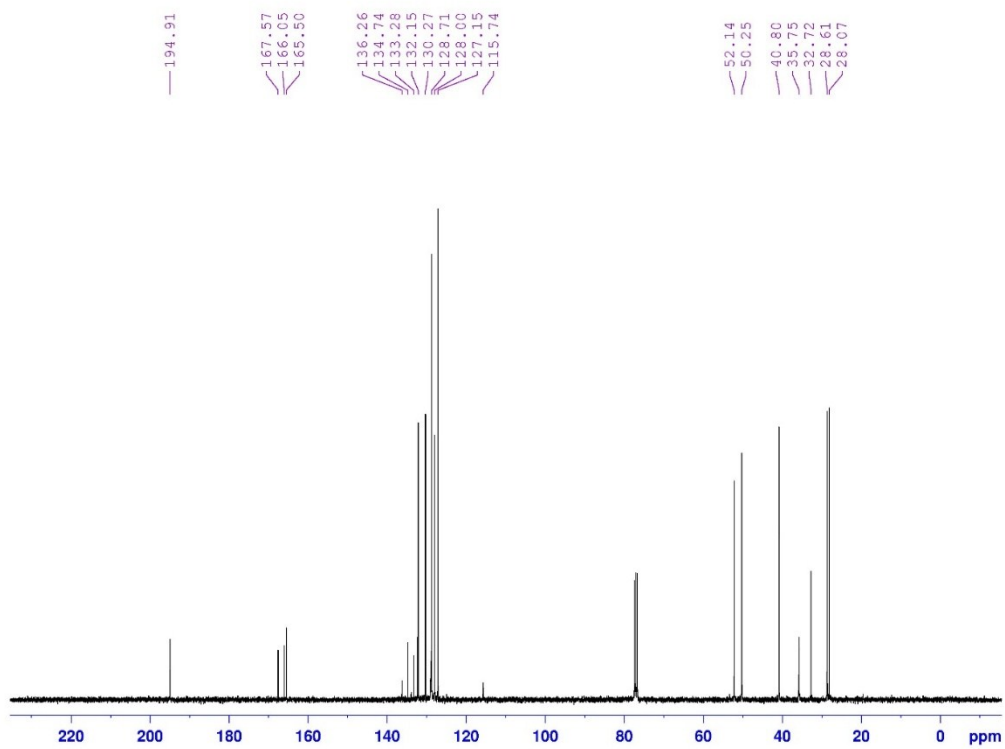
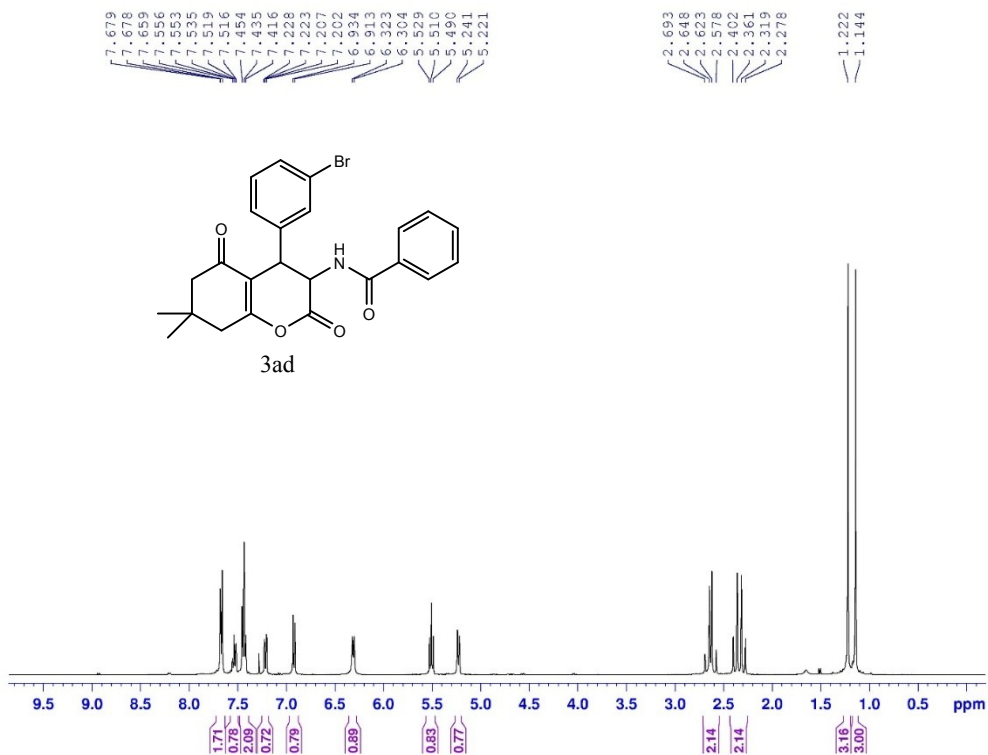


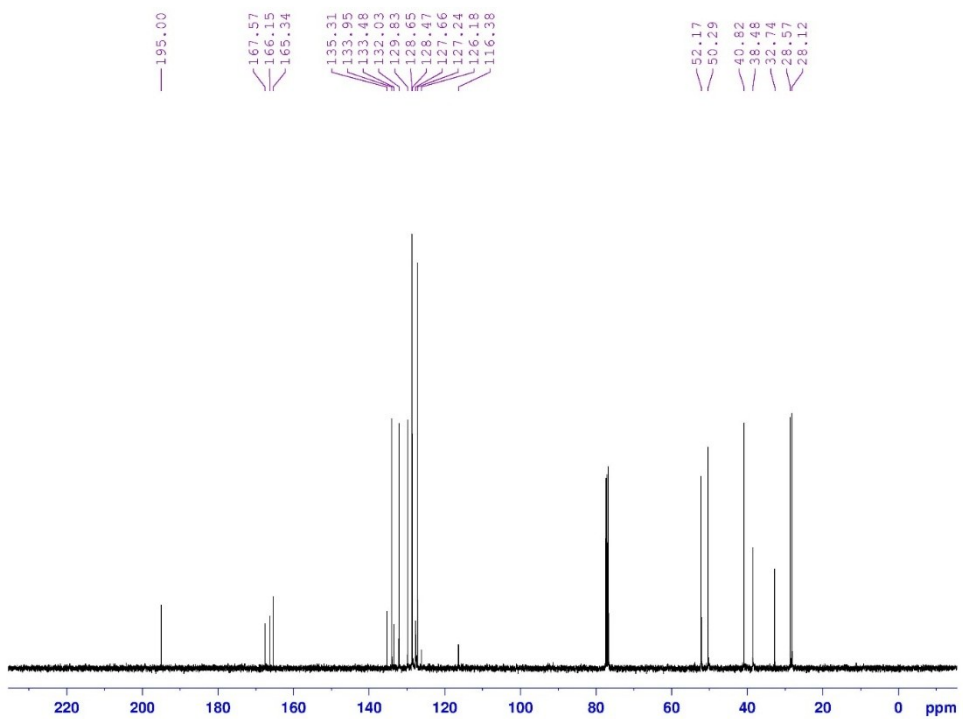
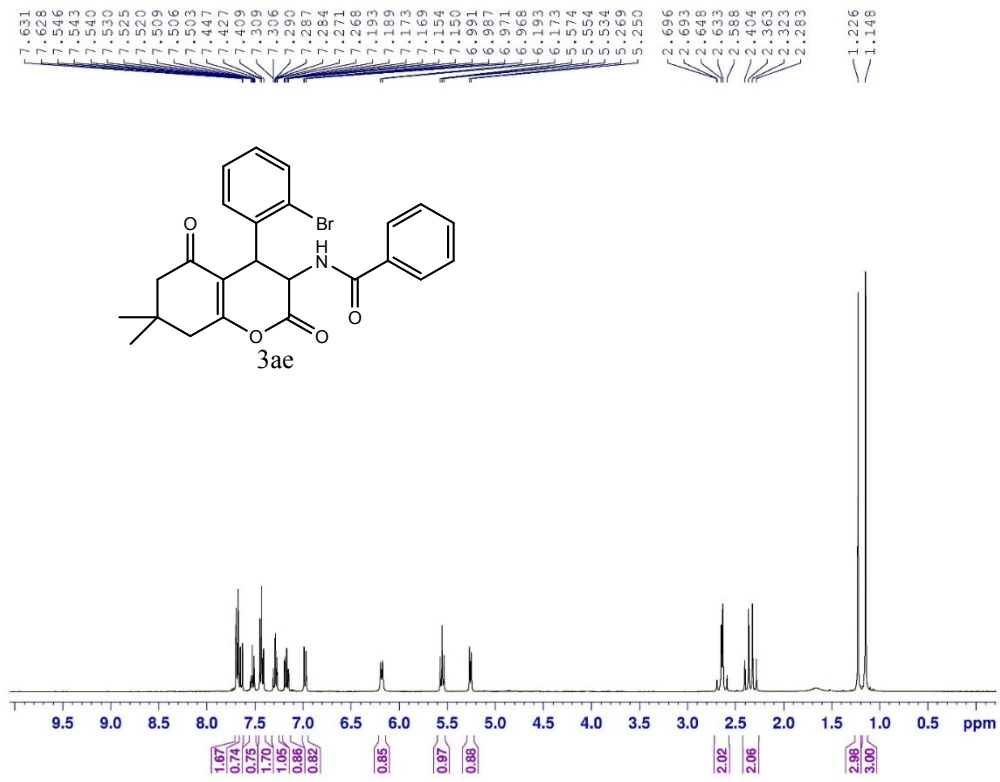
NMR spectra for 3-aminohexahydrocoumarin products

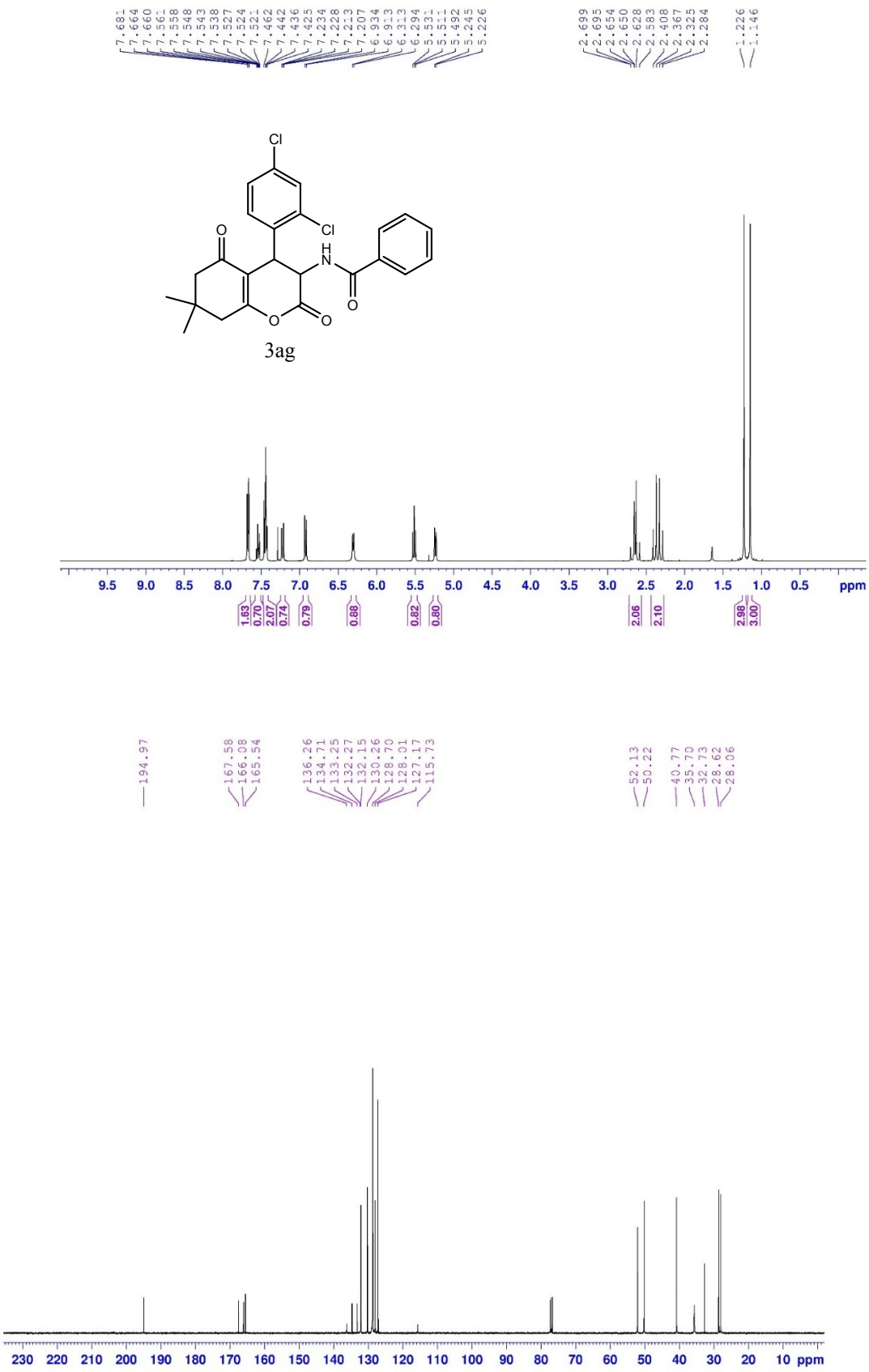


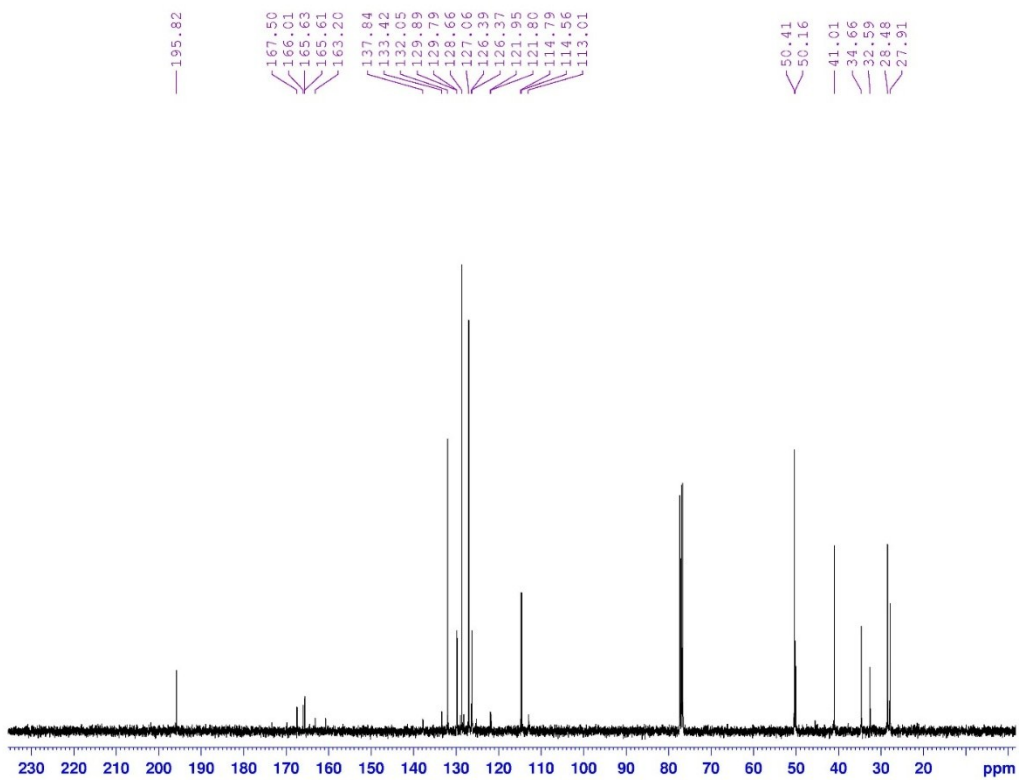
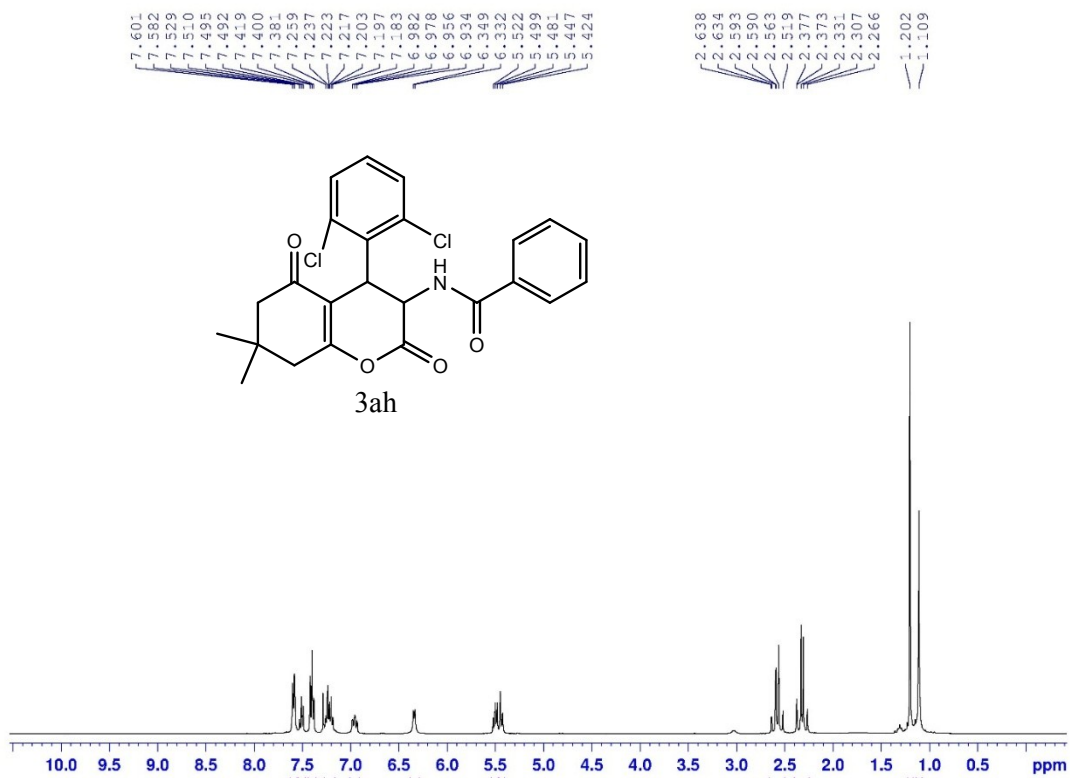


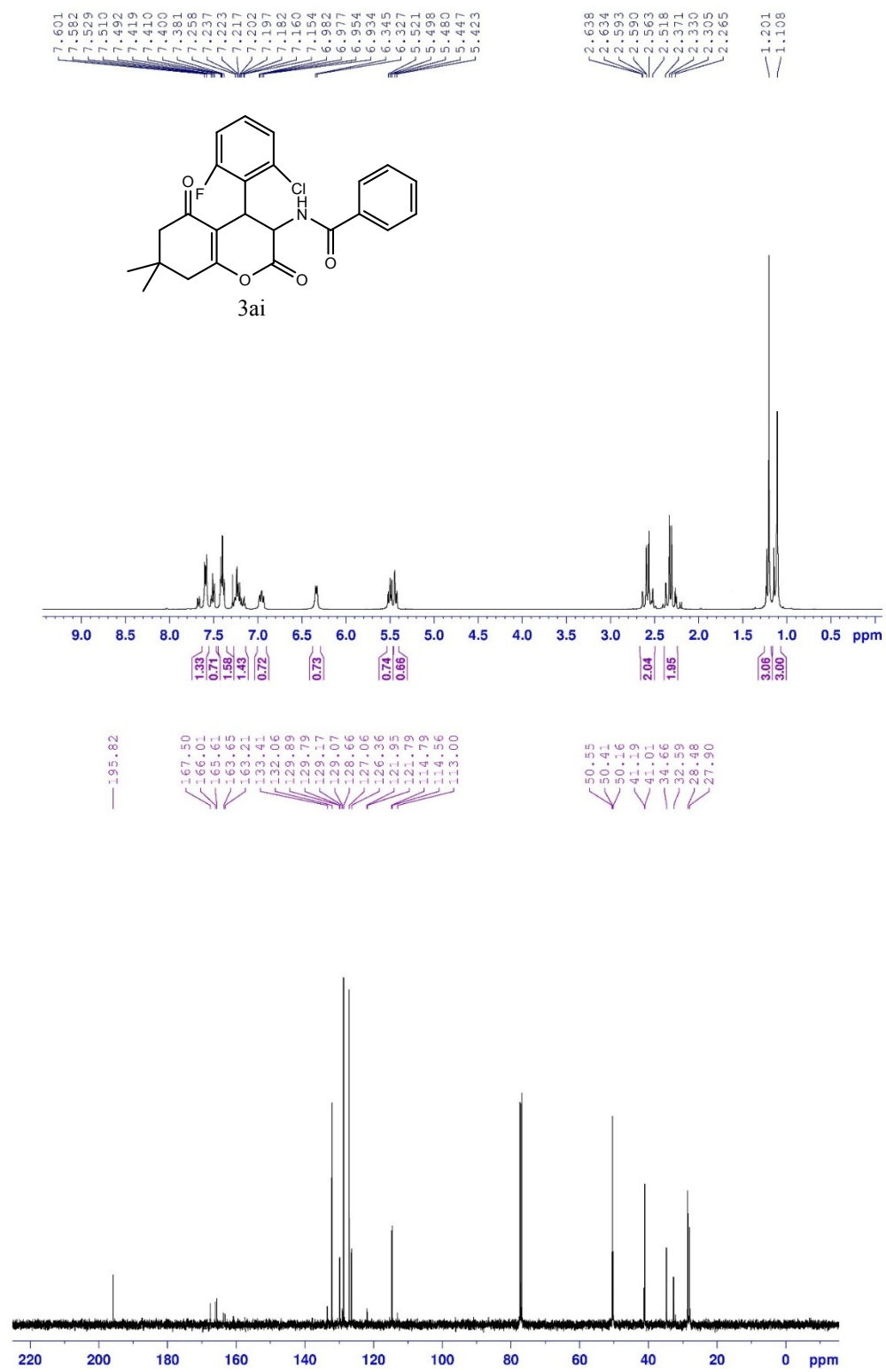


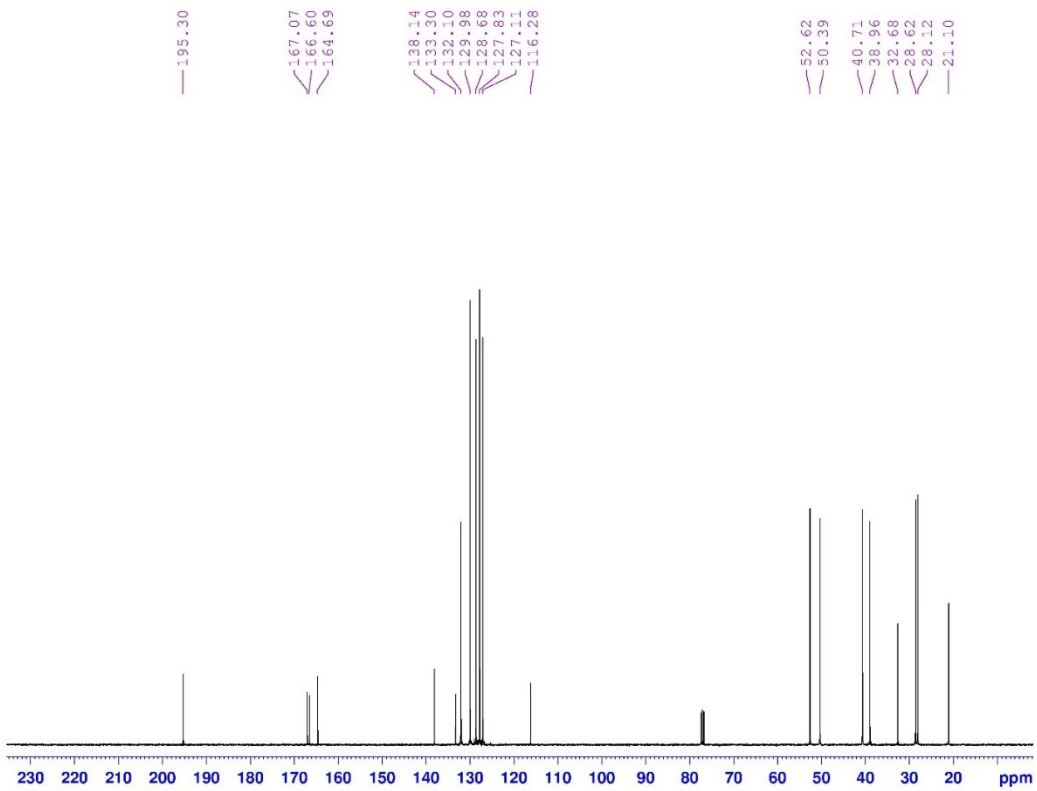
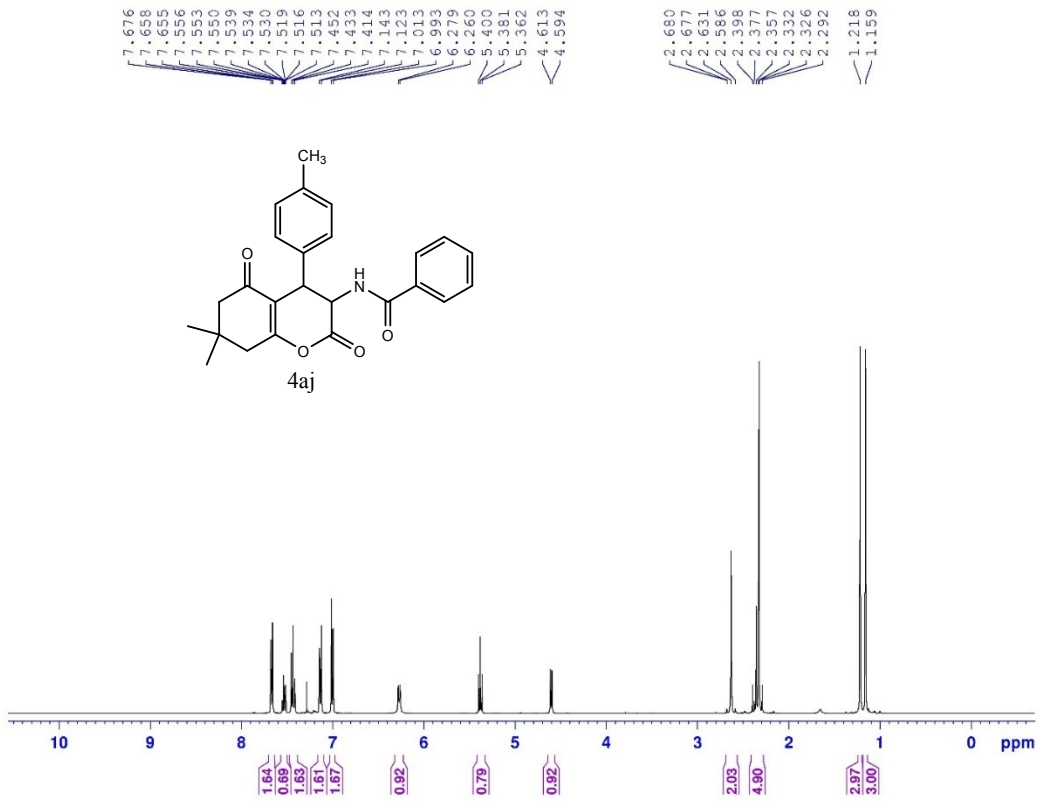


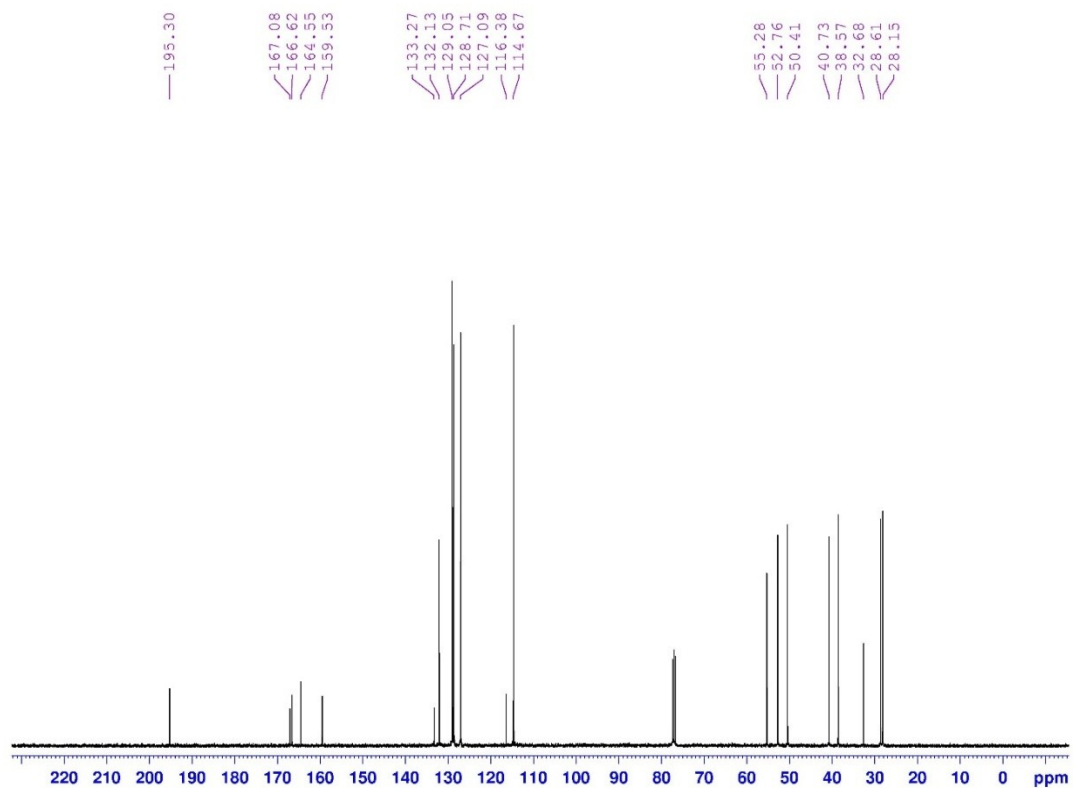
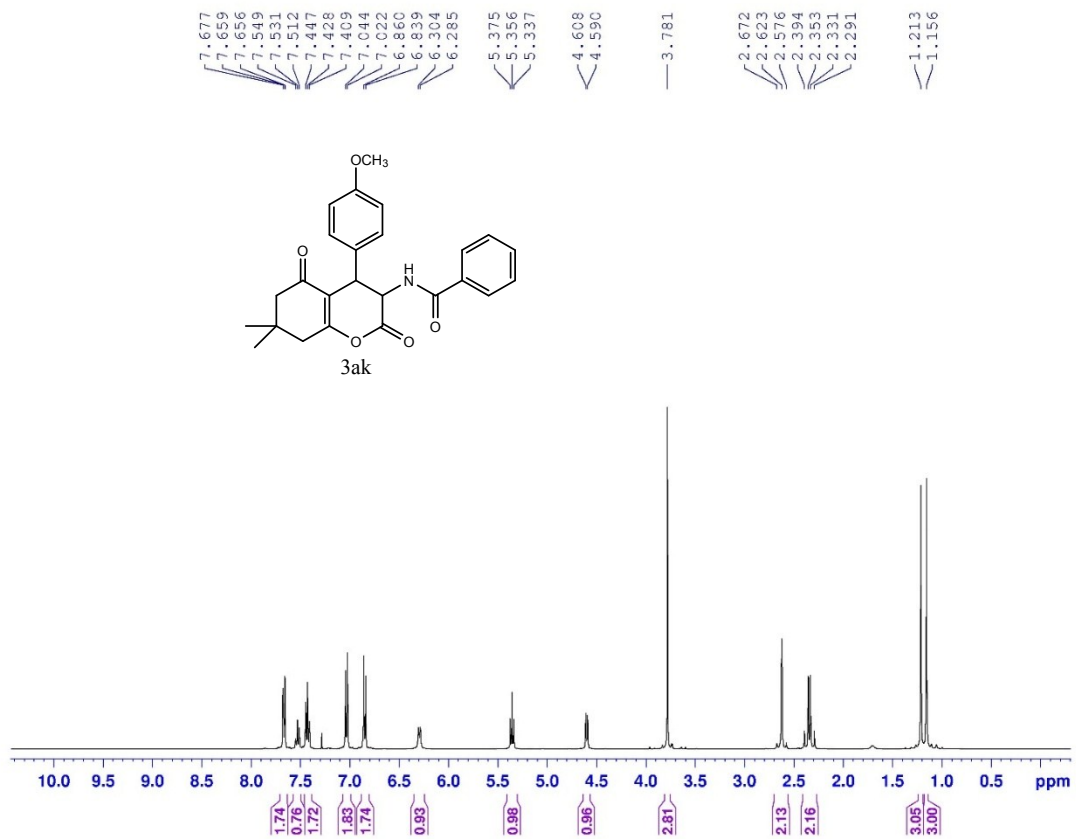


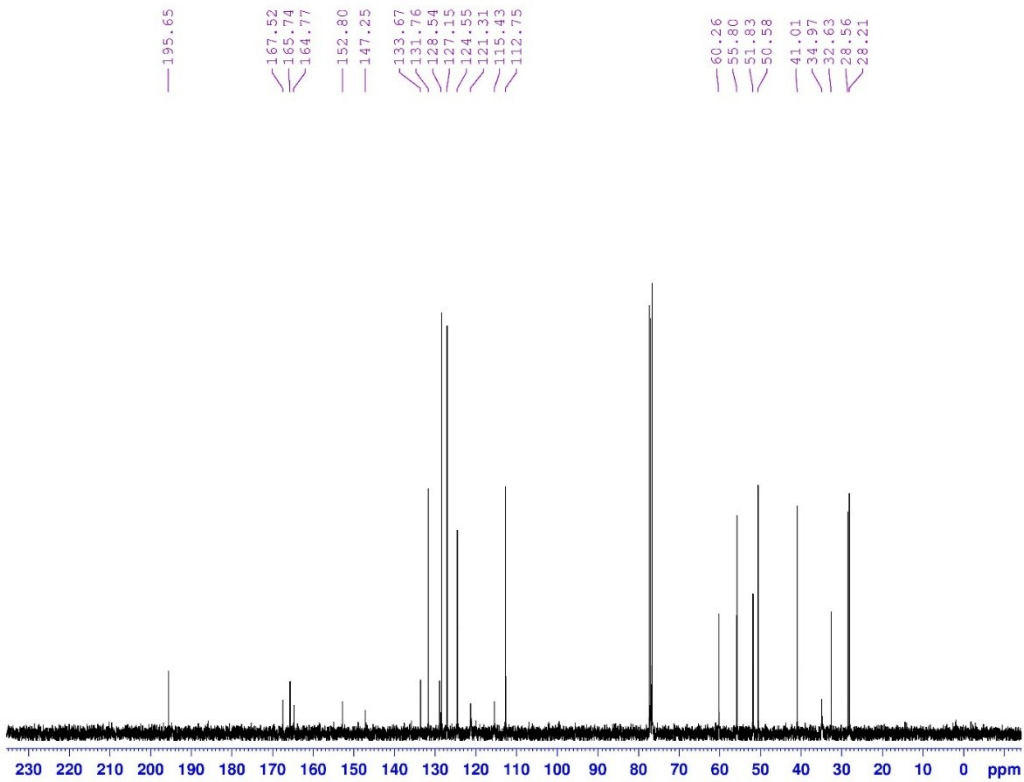
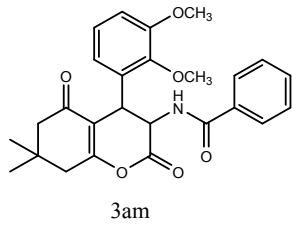
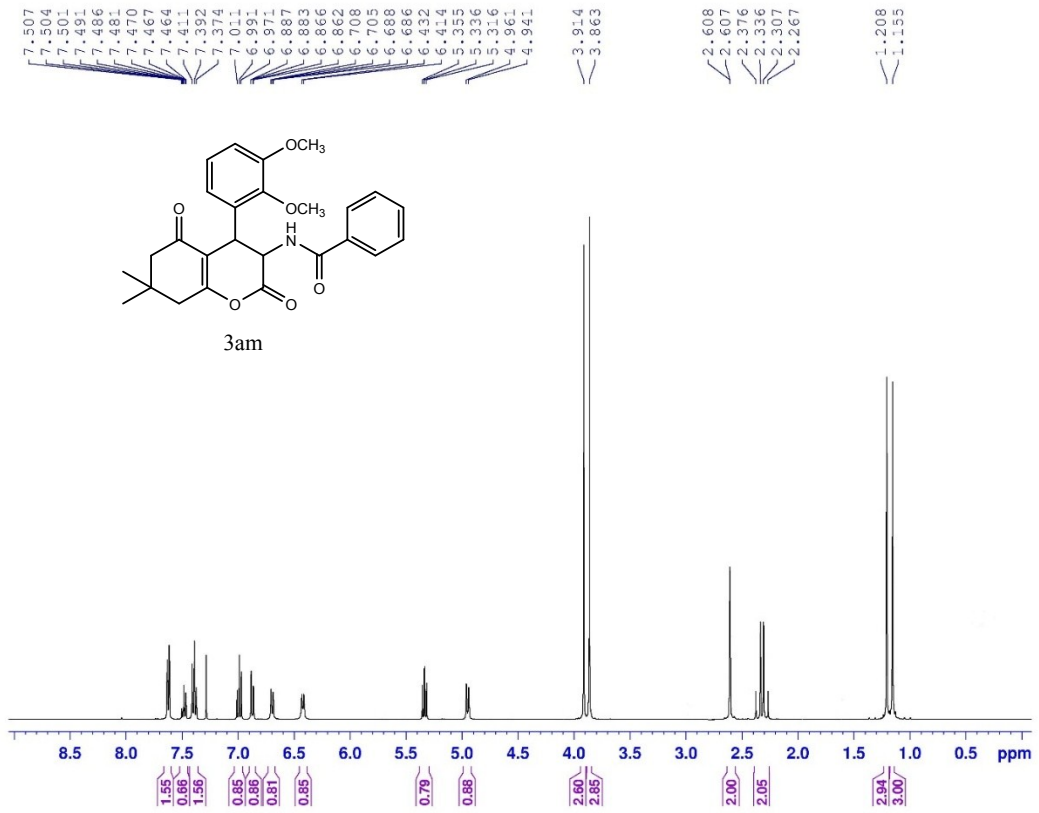




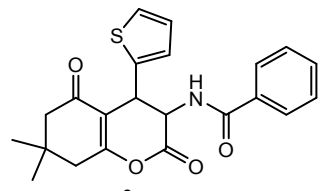




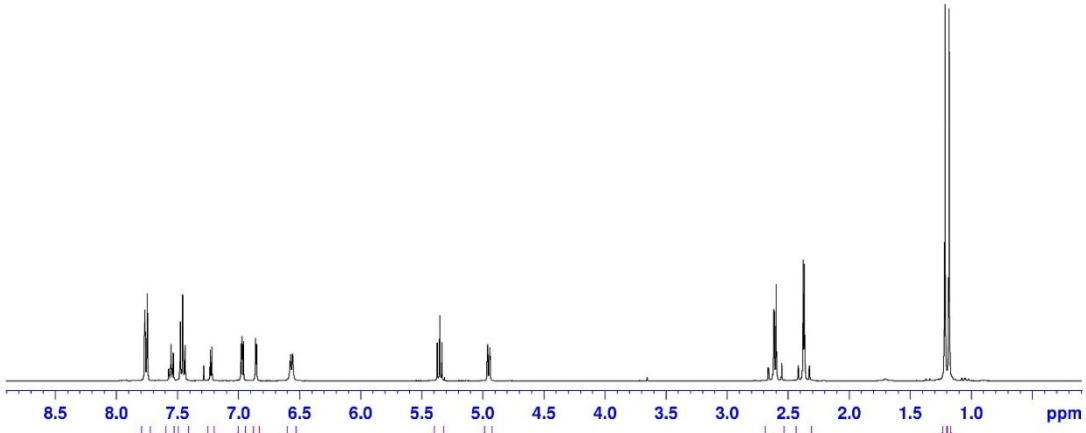




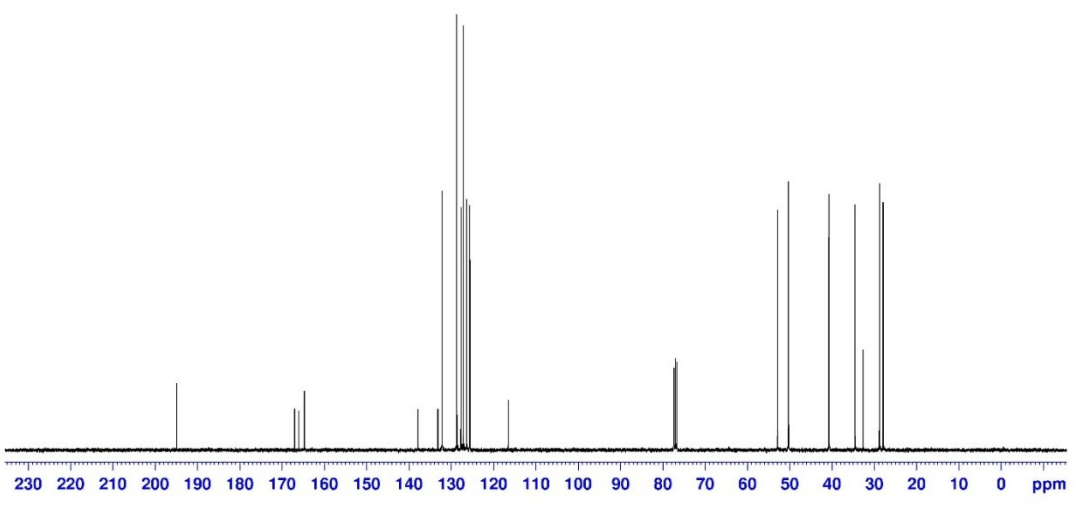
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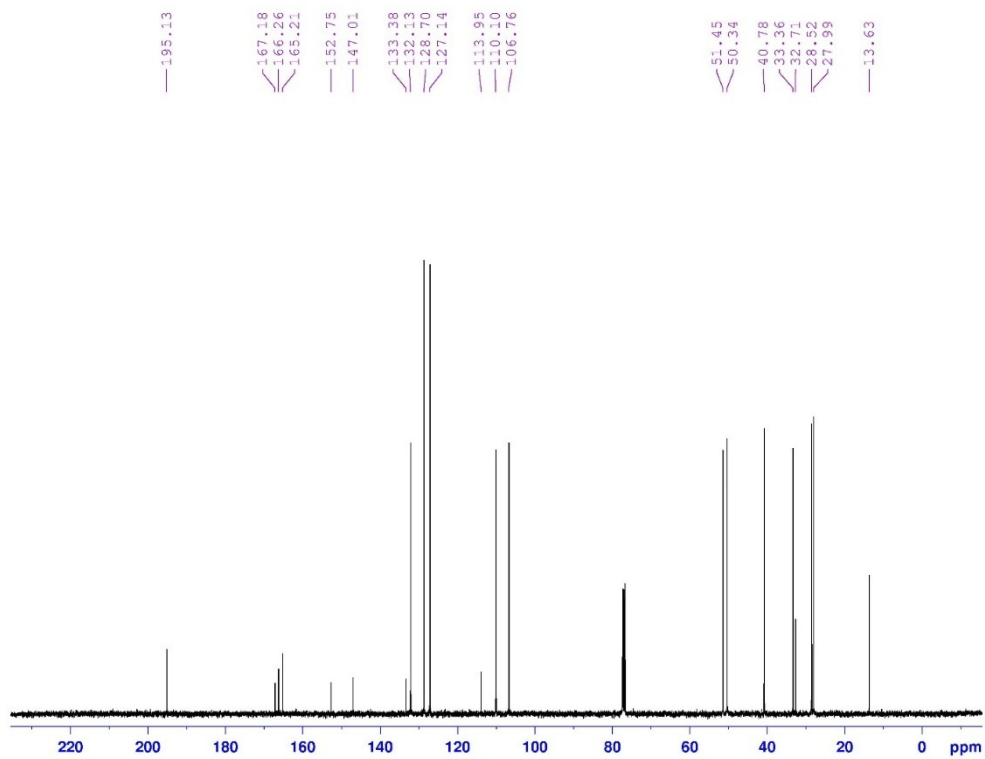
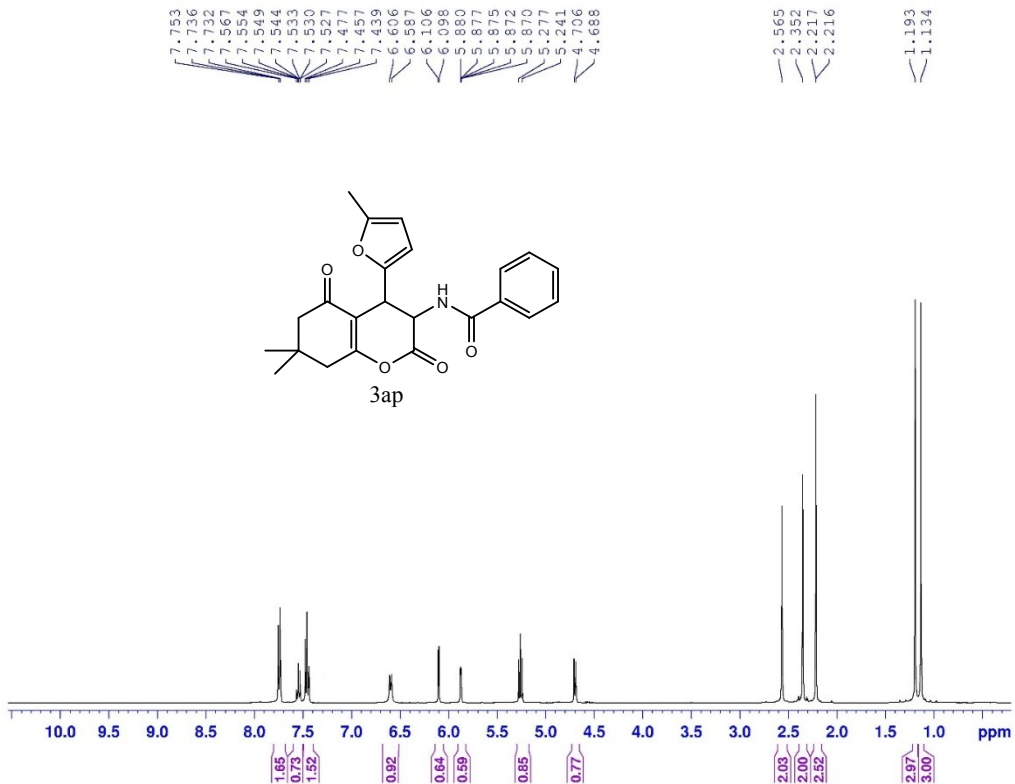


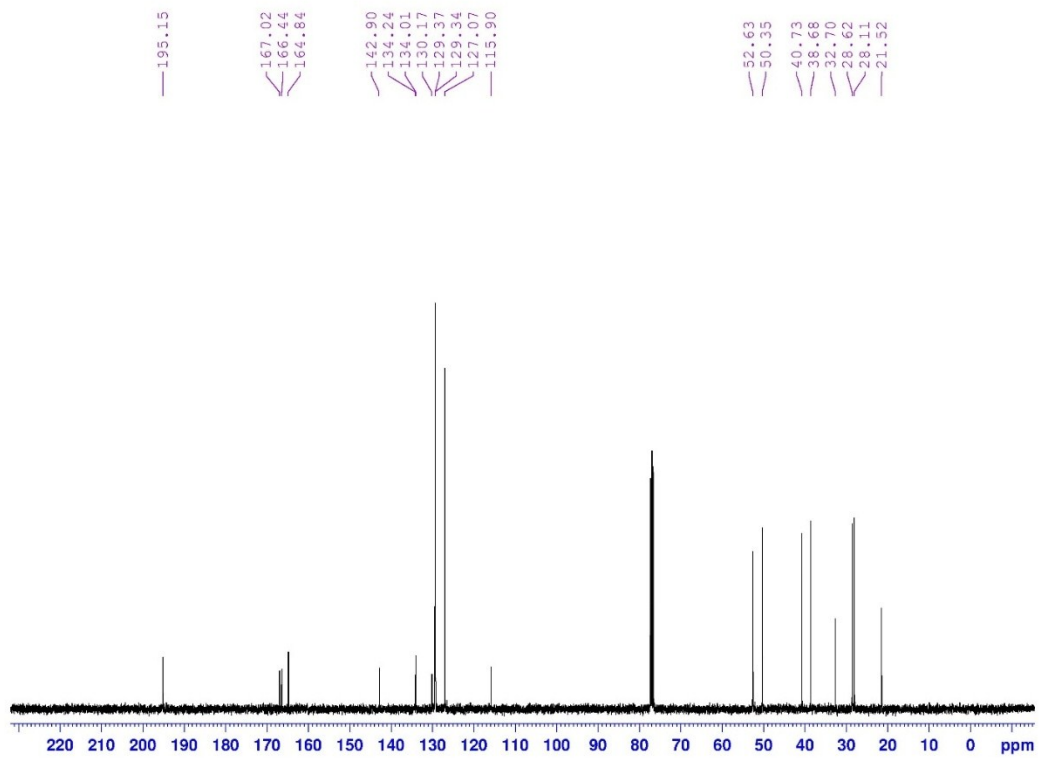
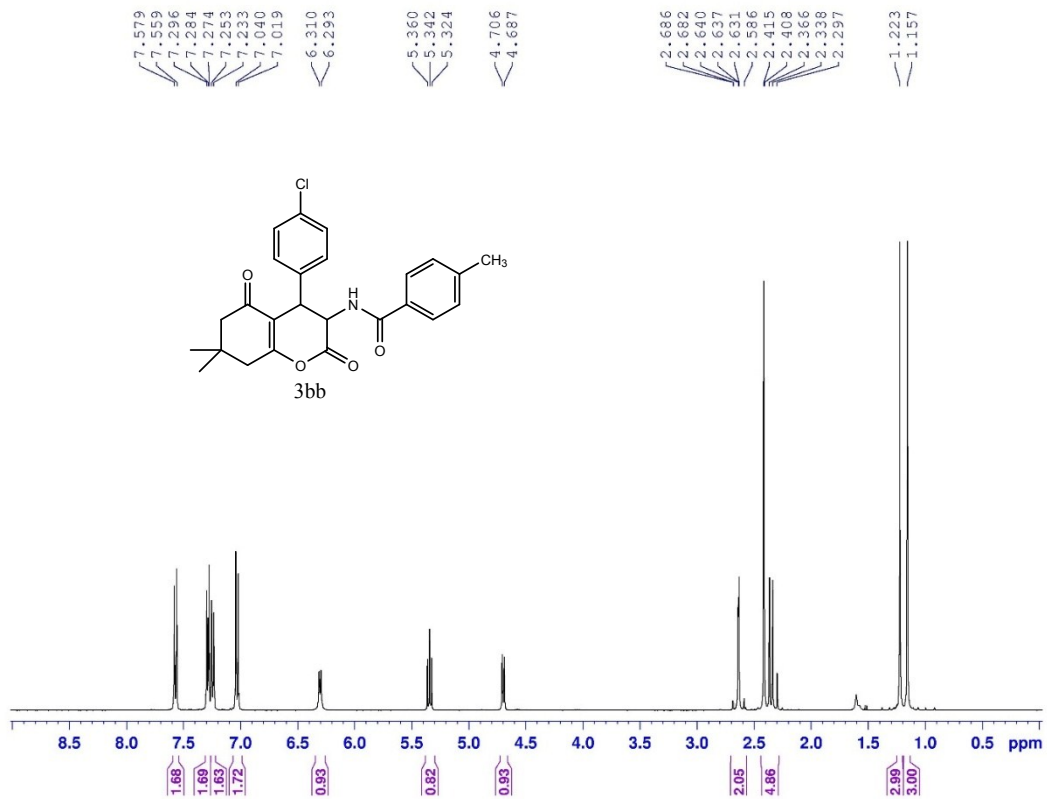
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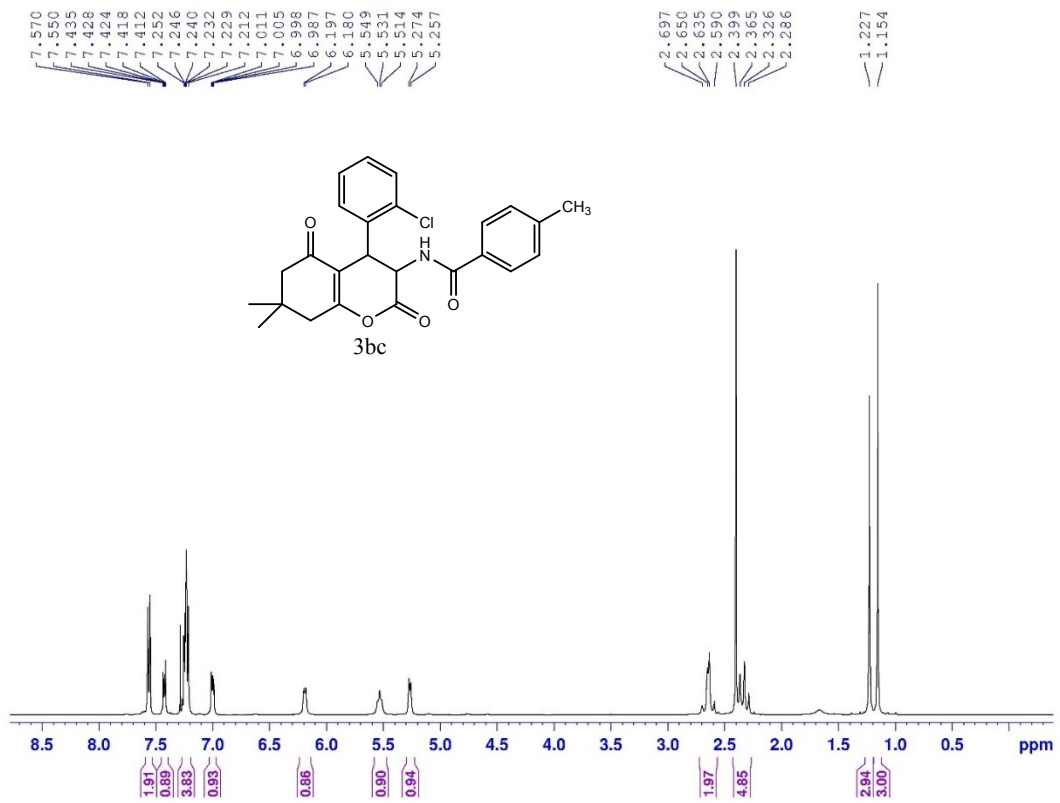


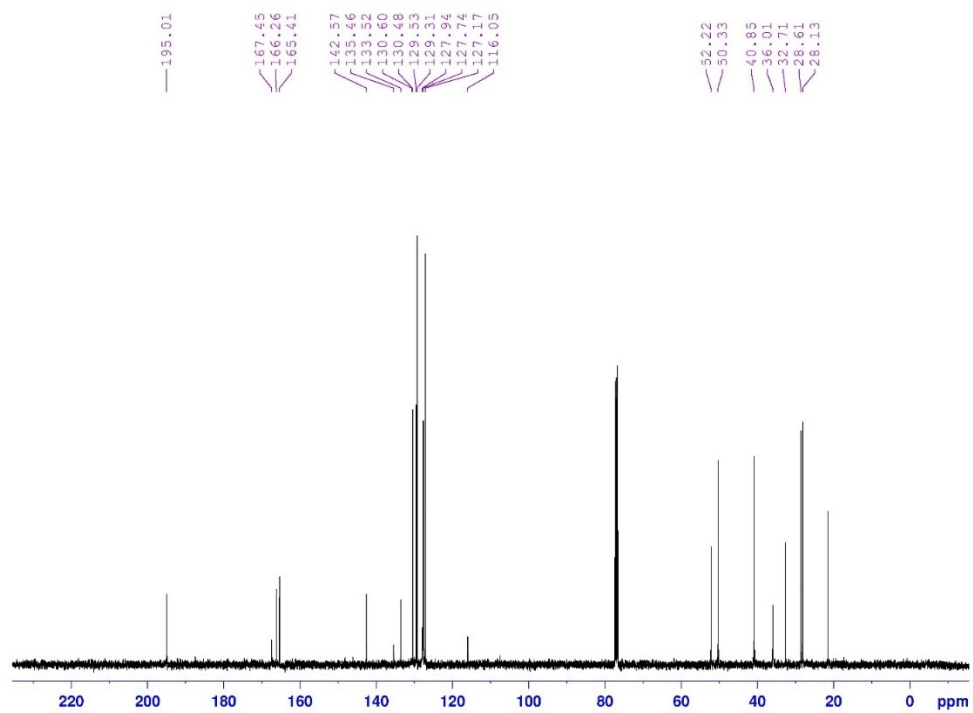
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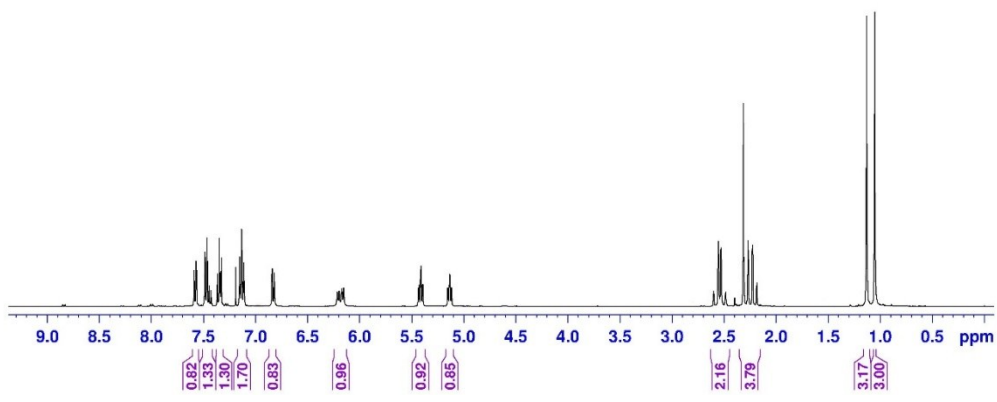
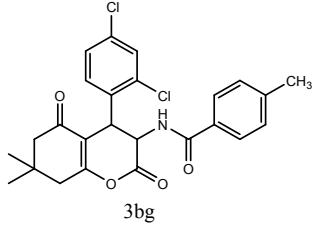






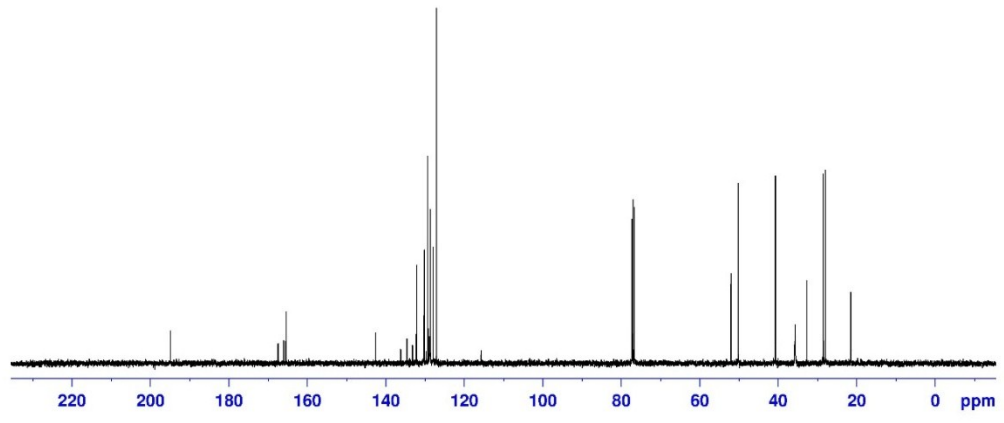
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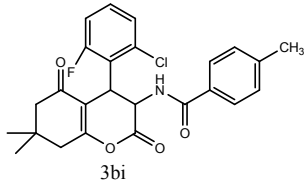
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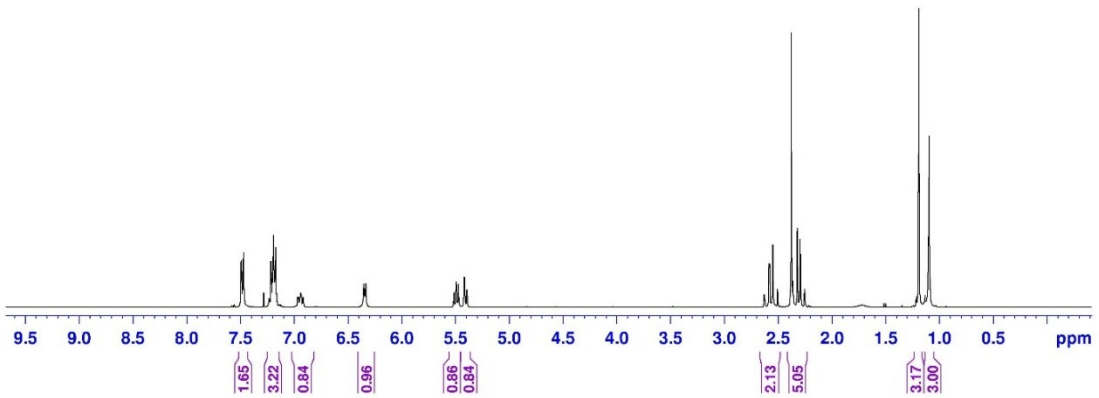


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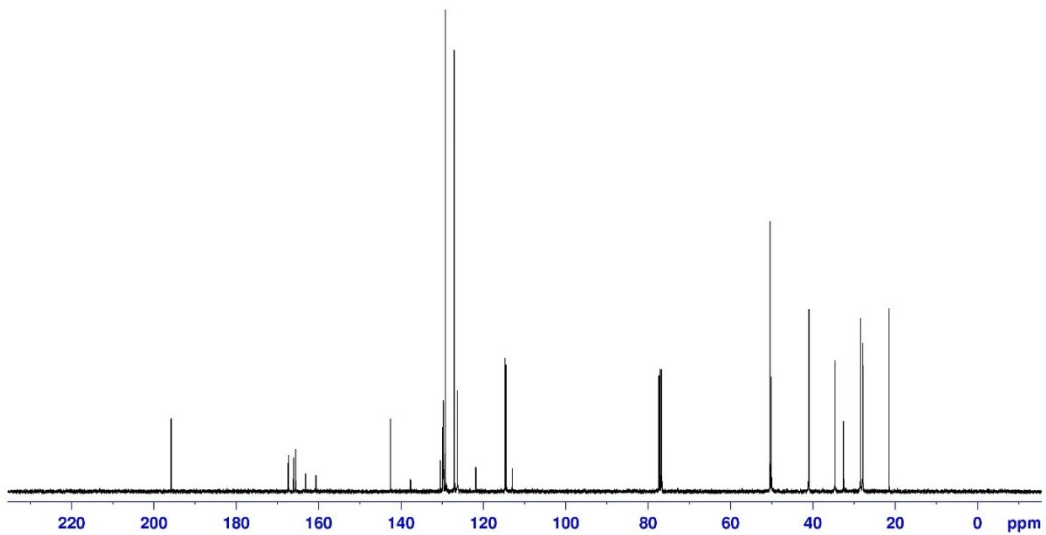
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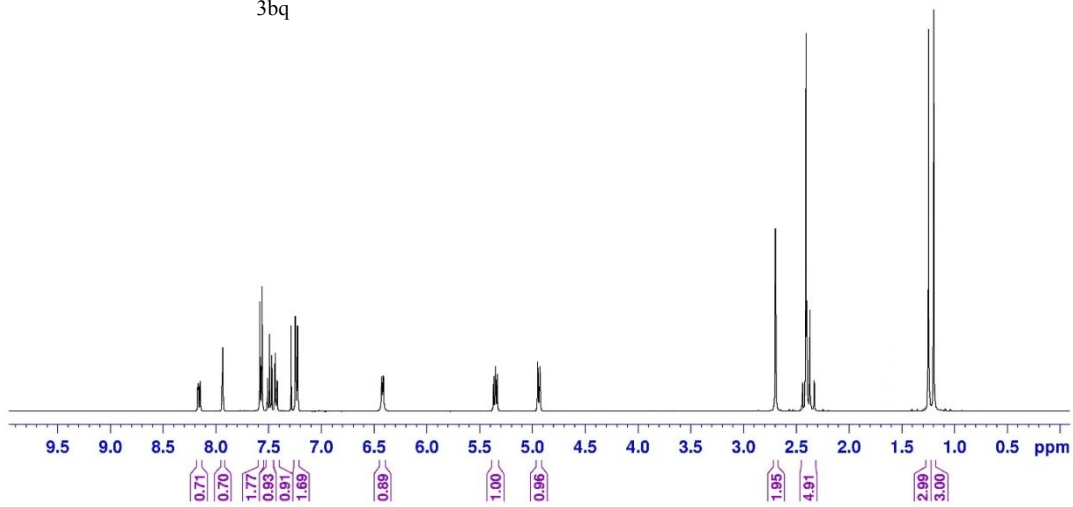
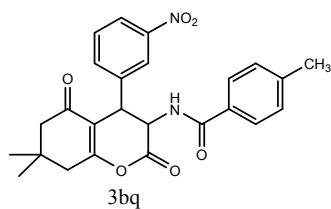
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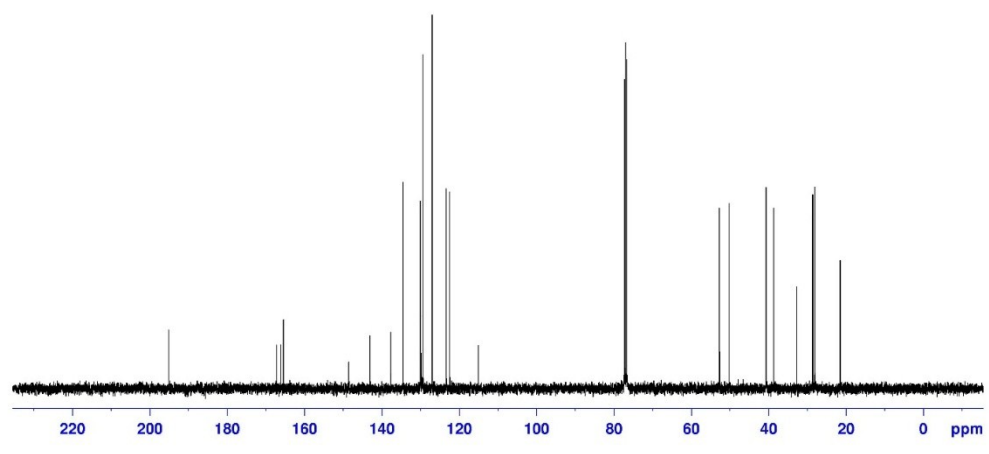
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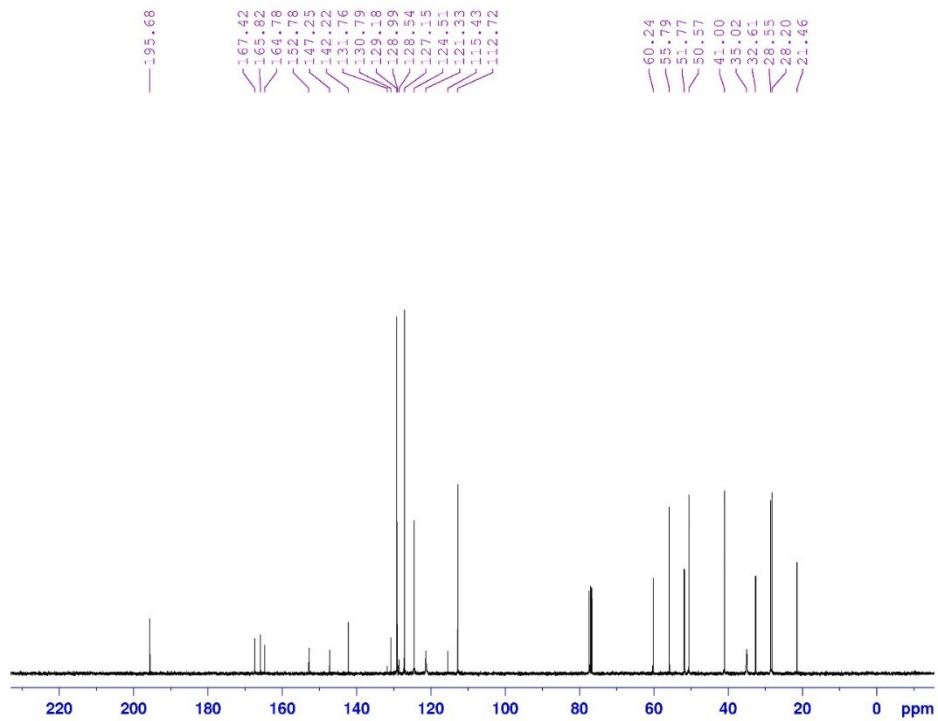
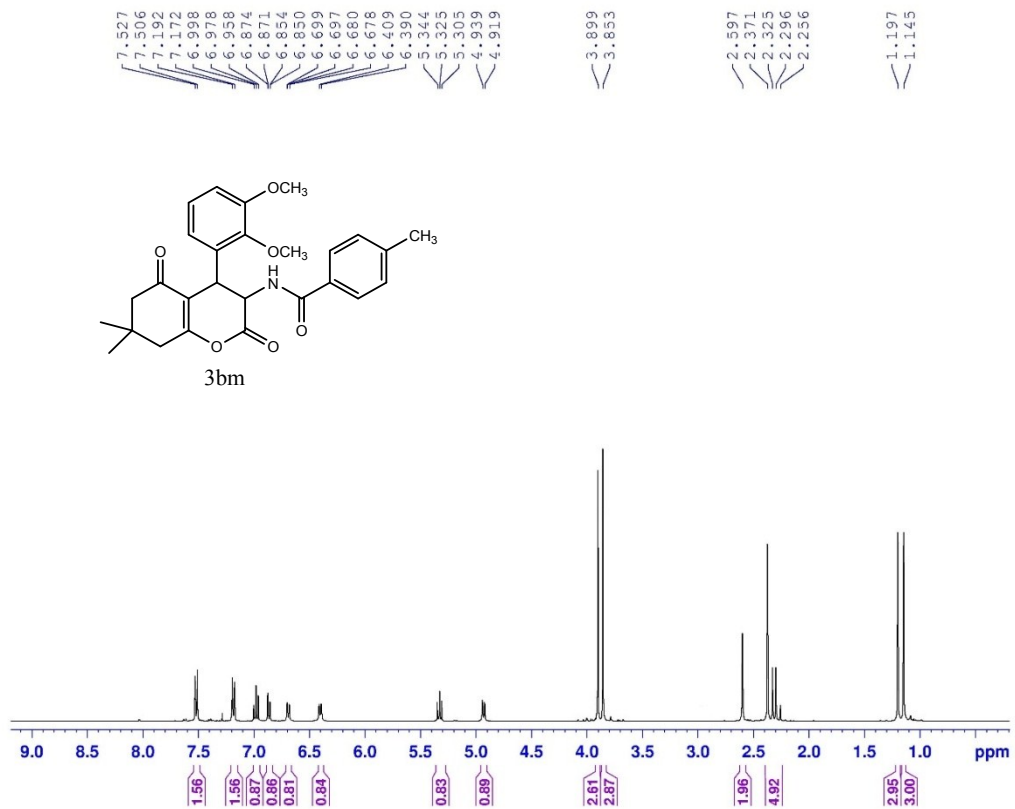
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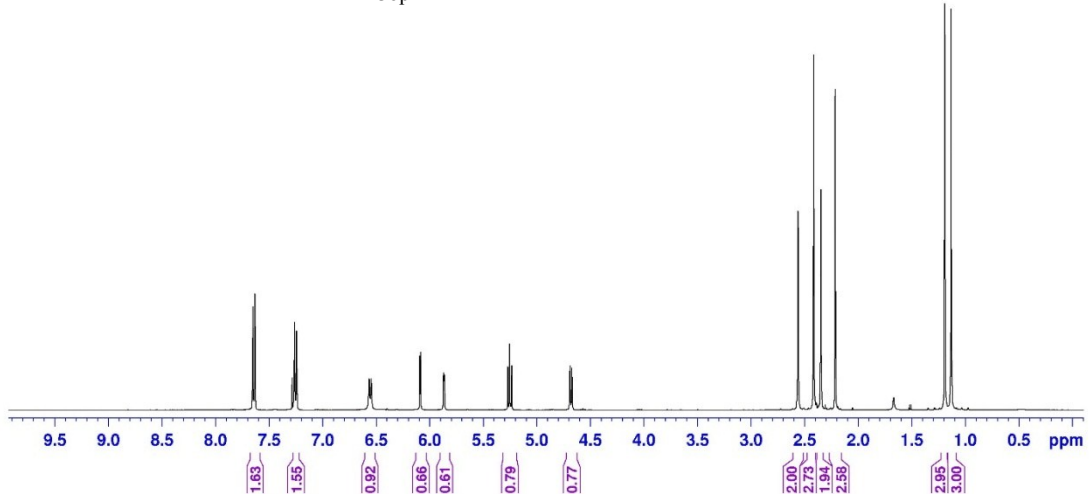
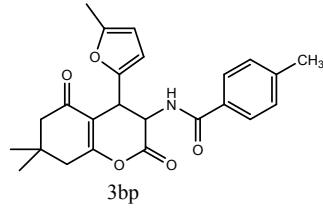




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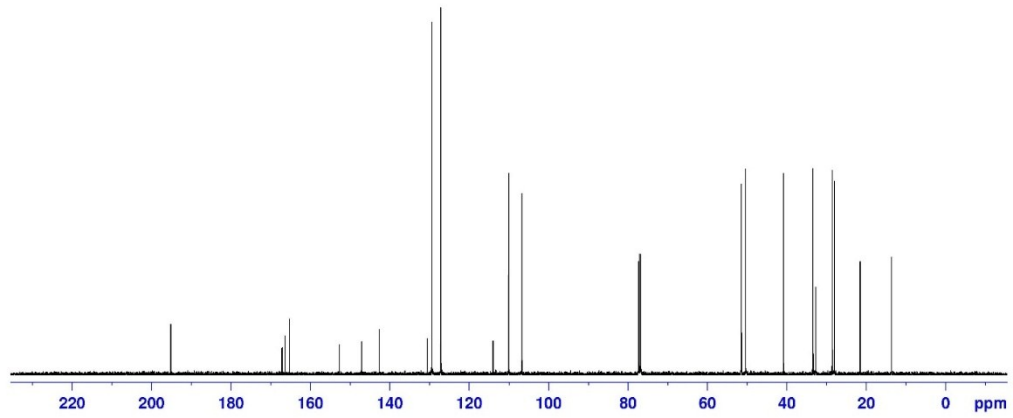
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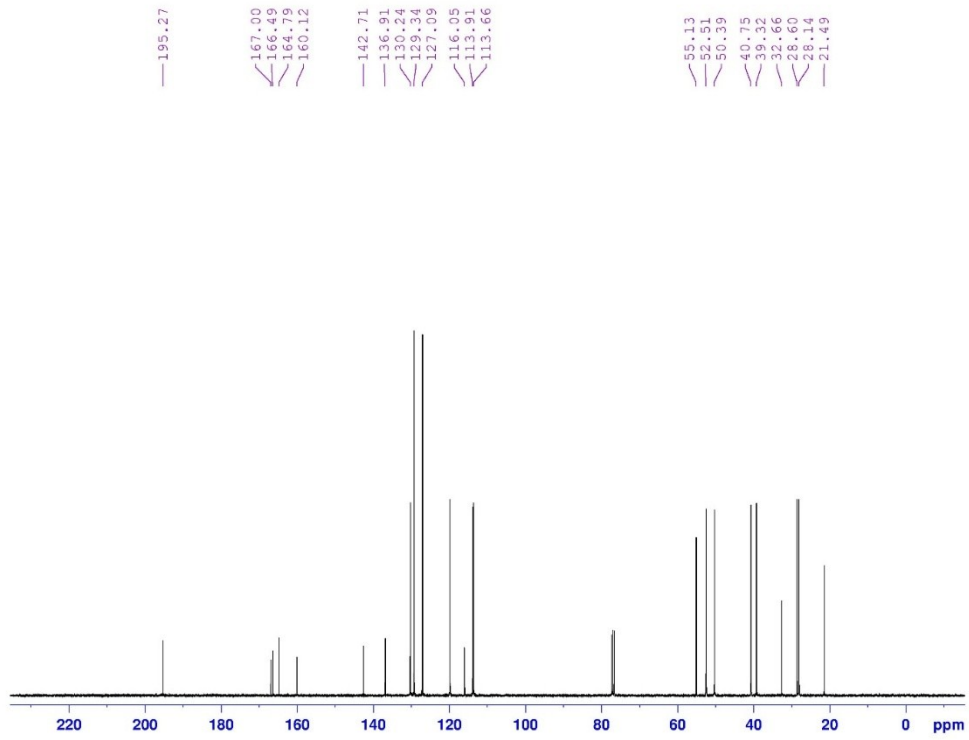
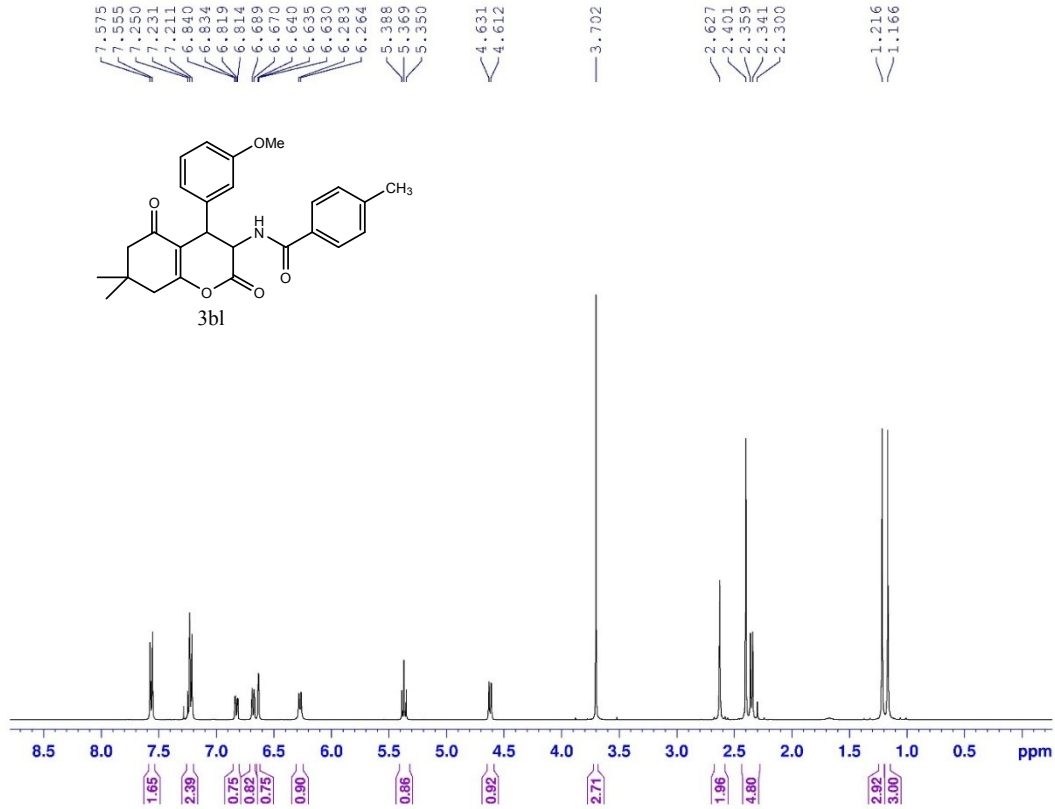
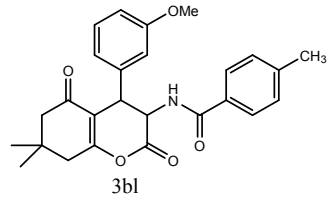
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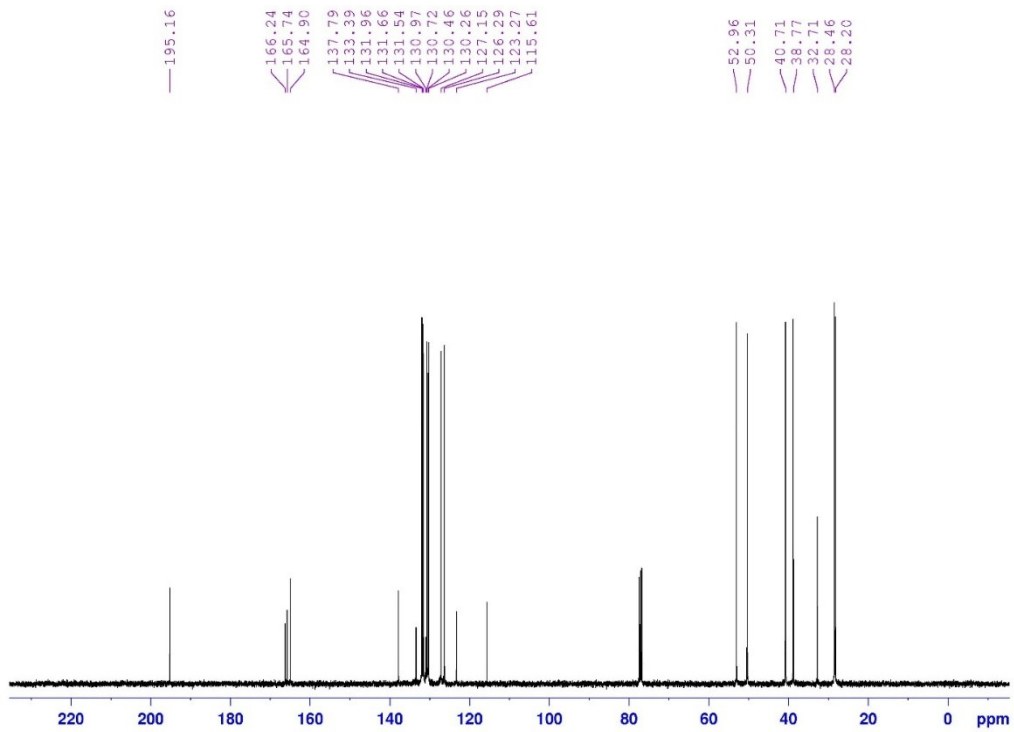
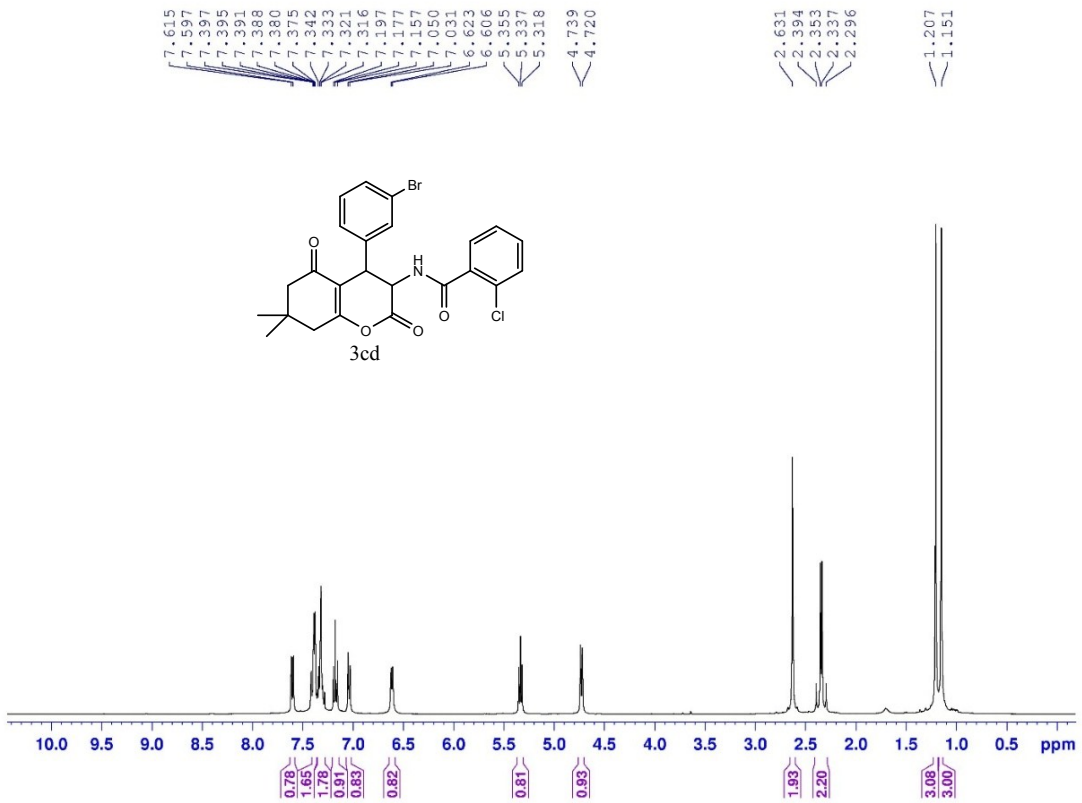


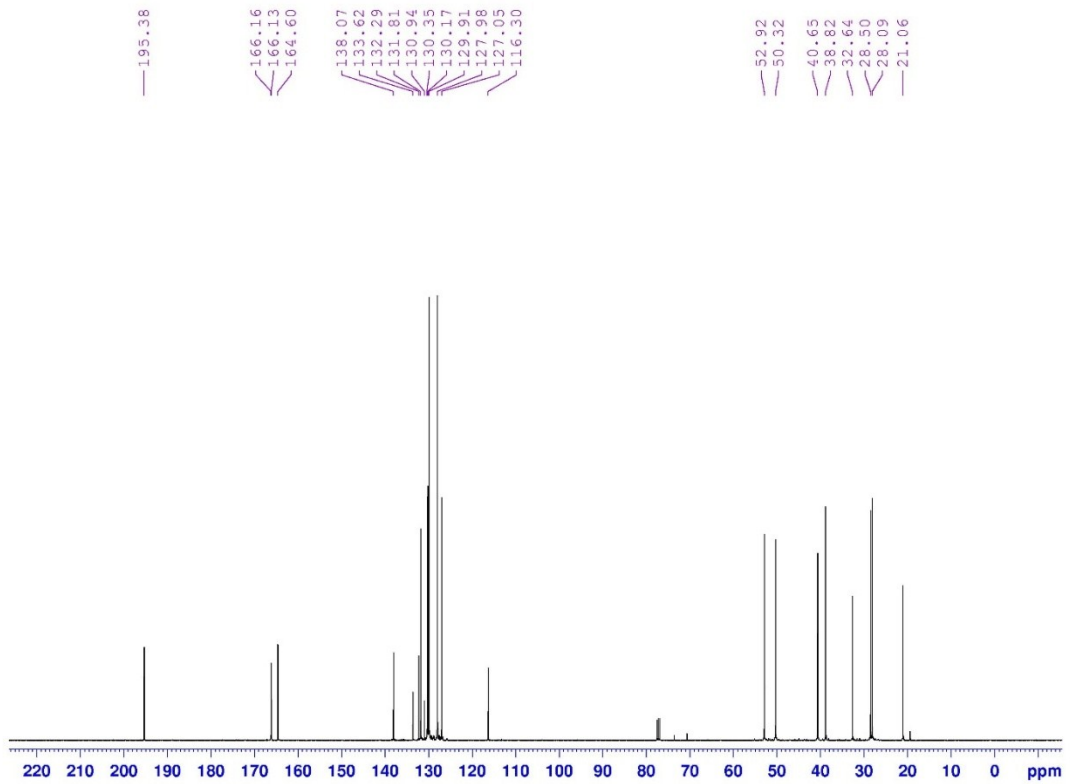
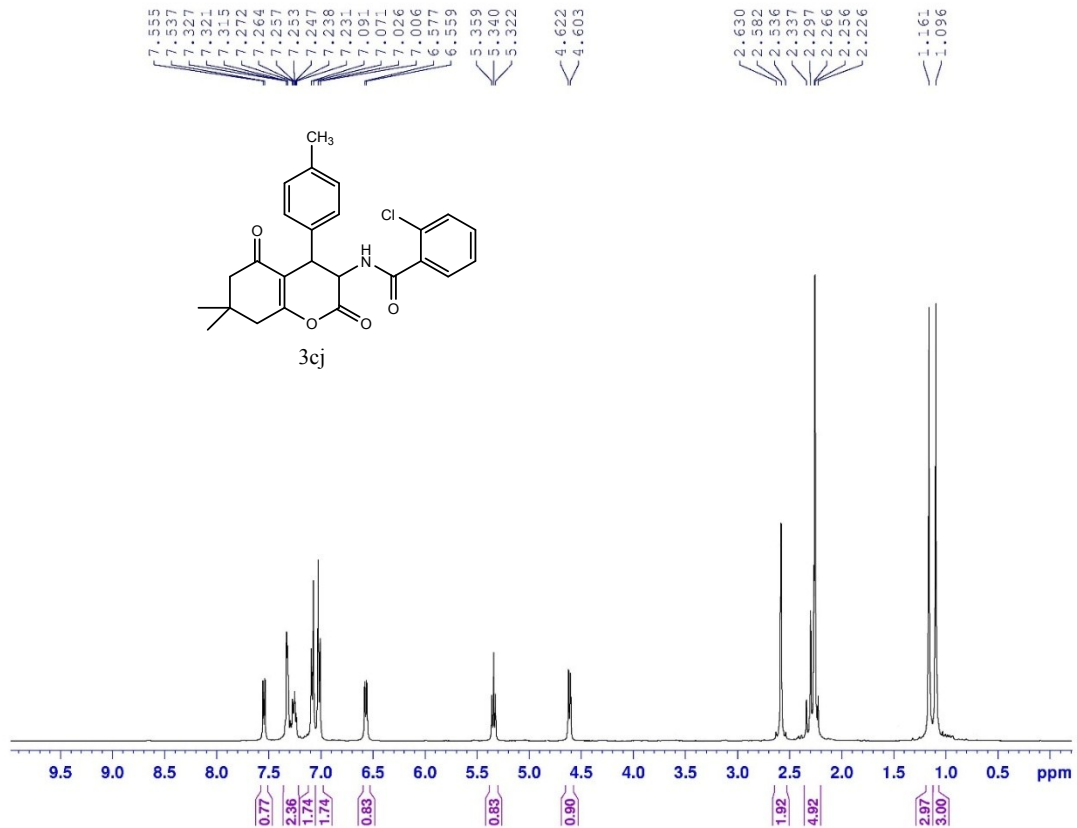
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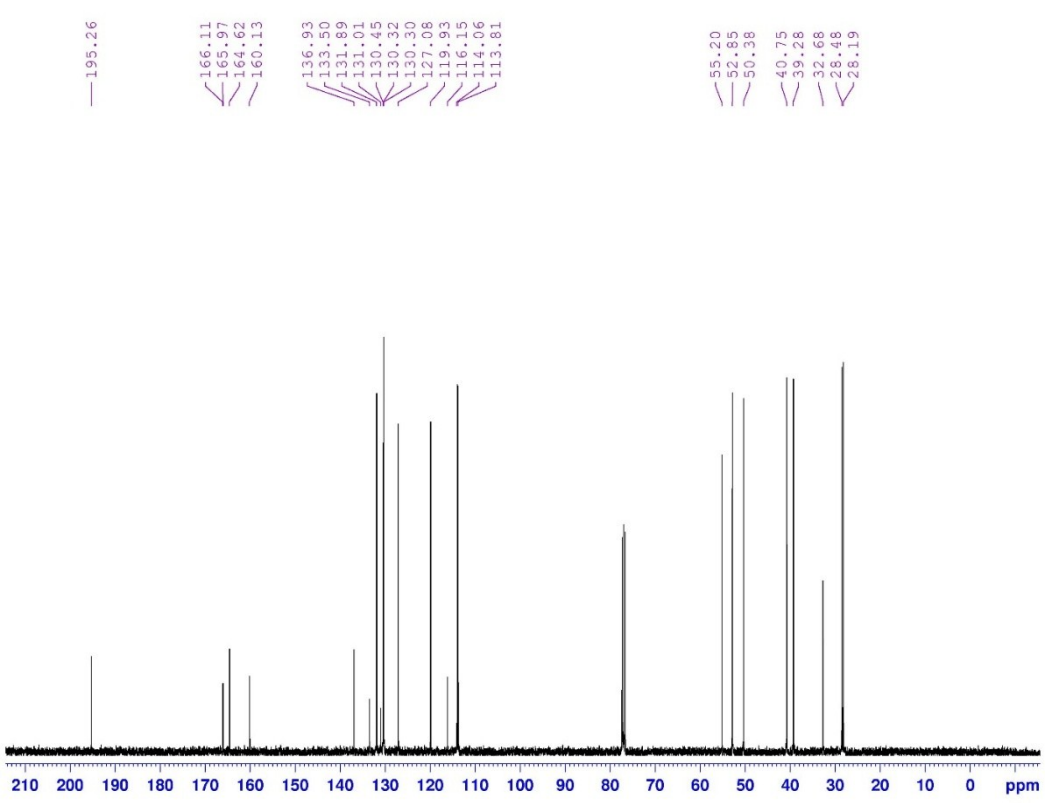
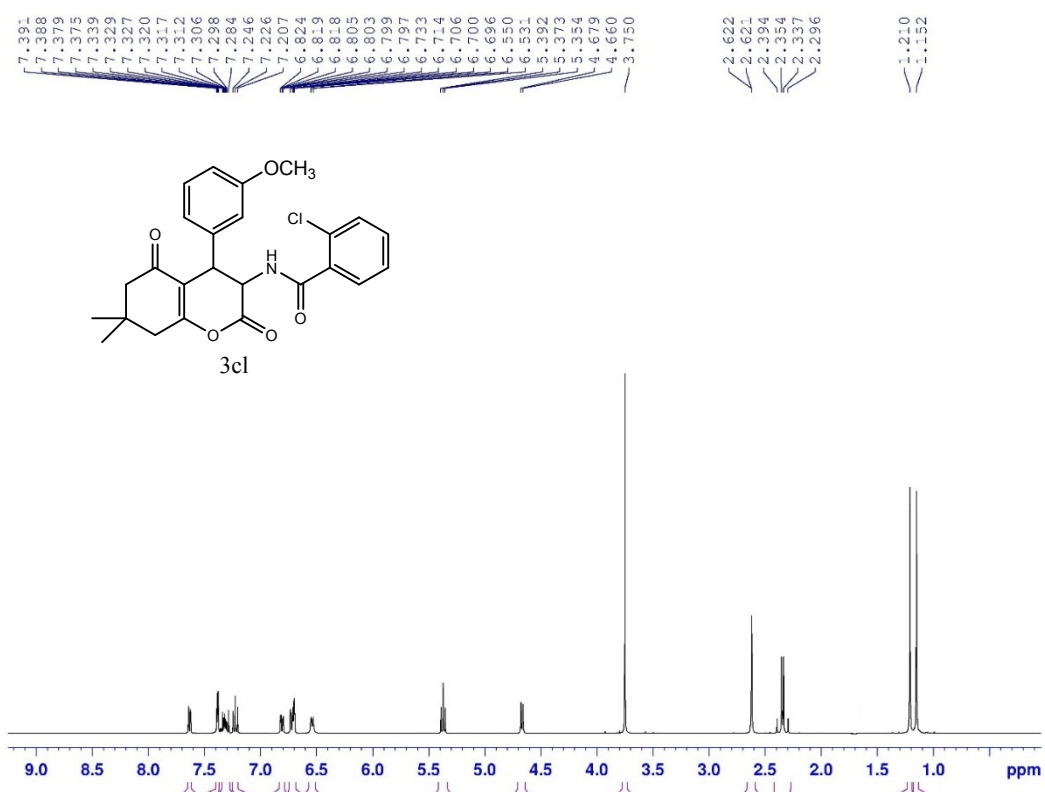
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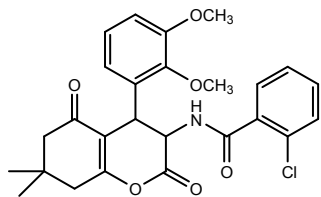


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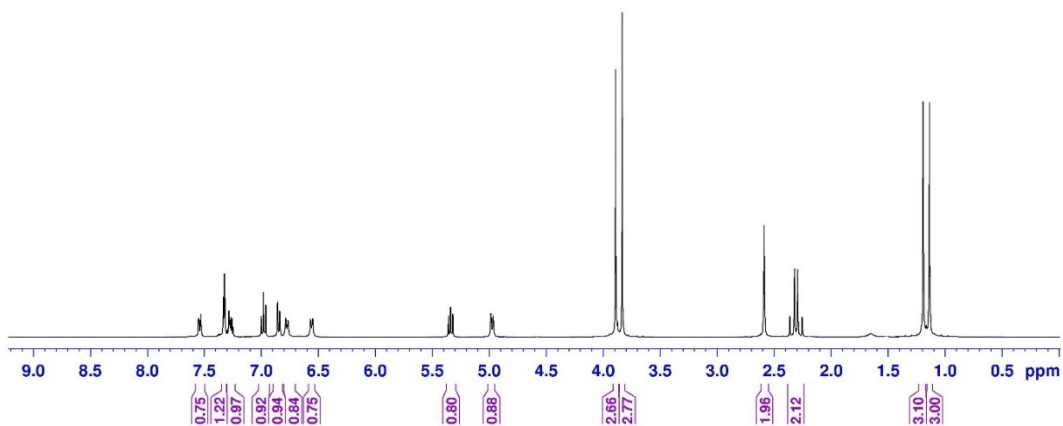
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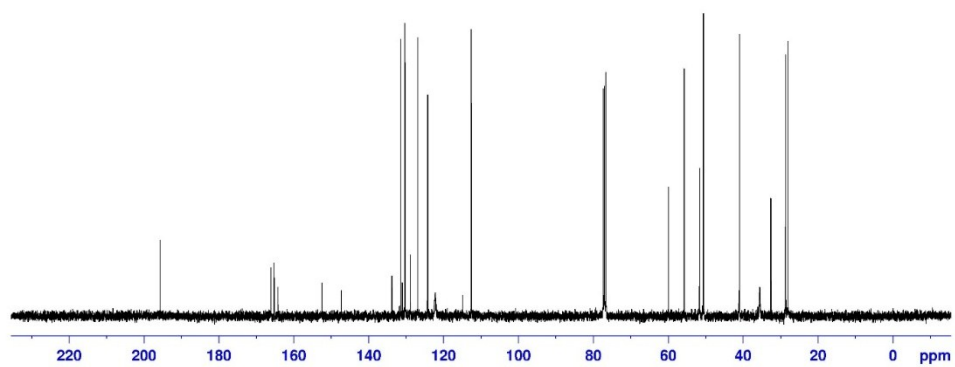
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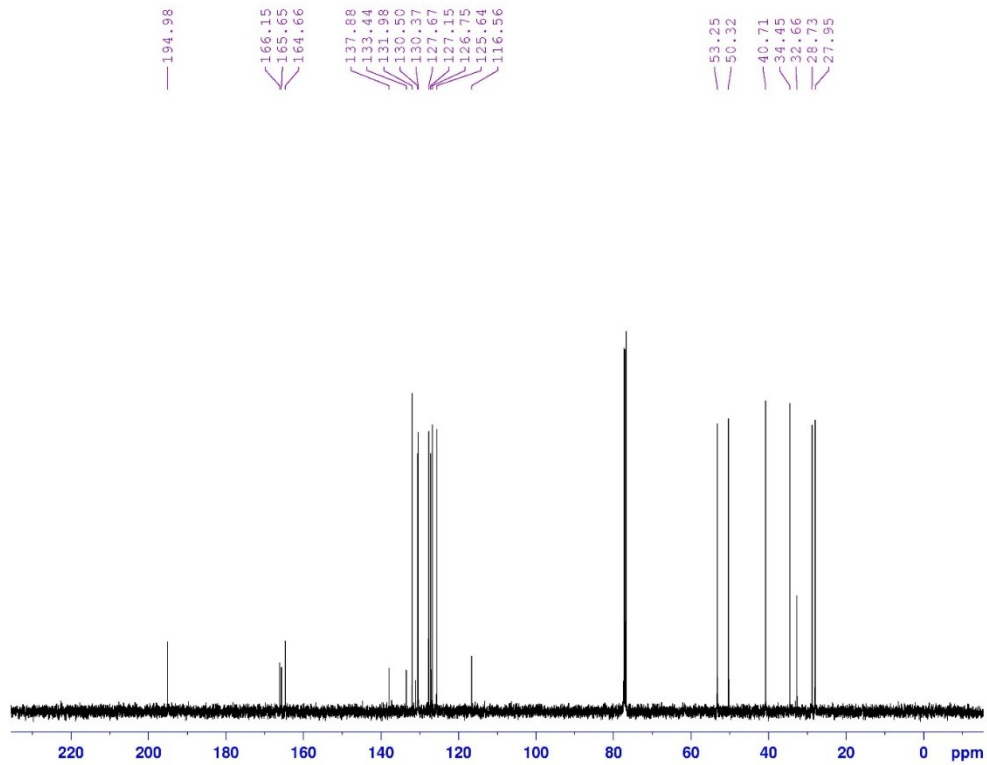
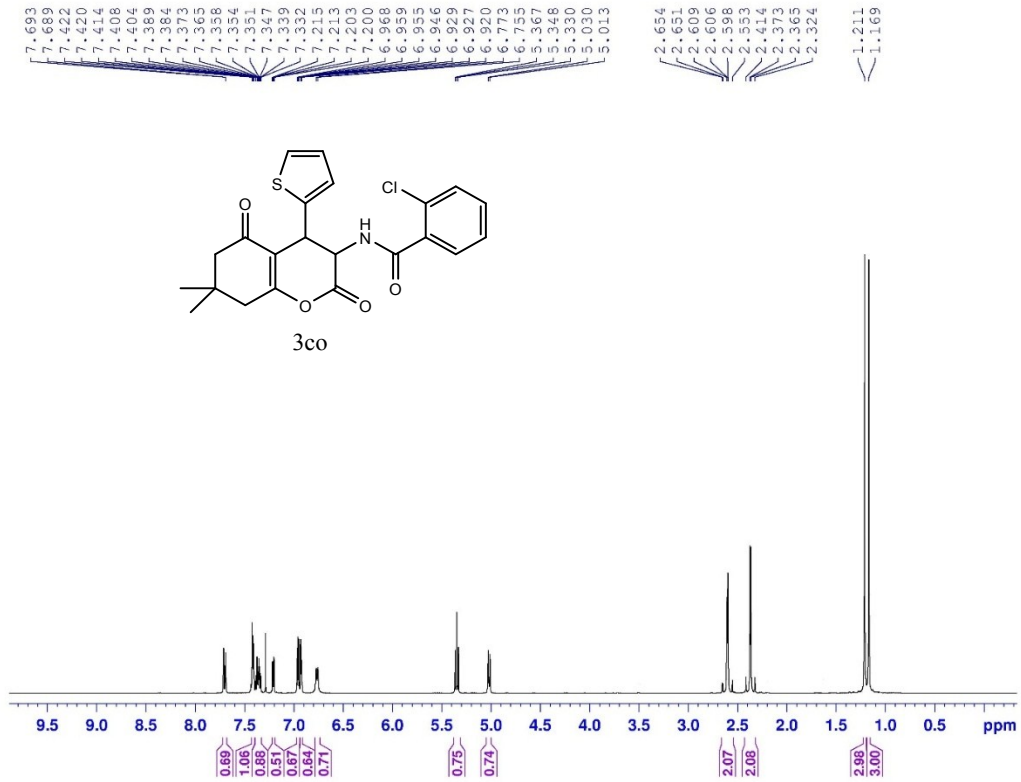


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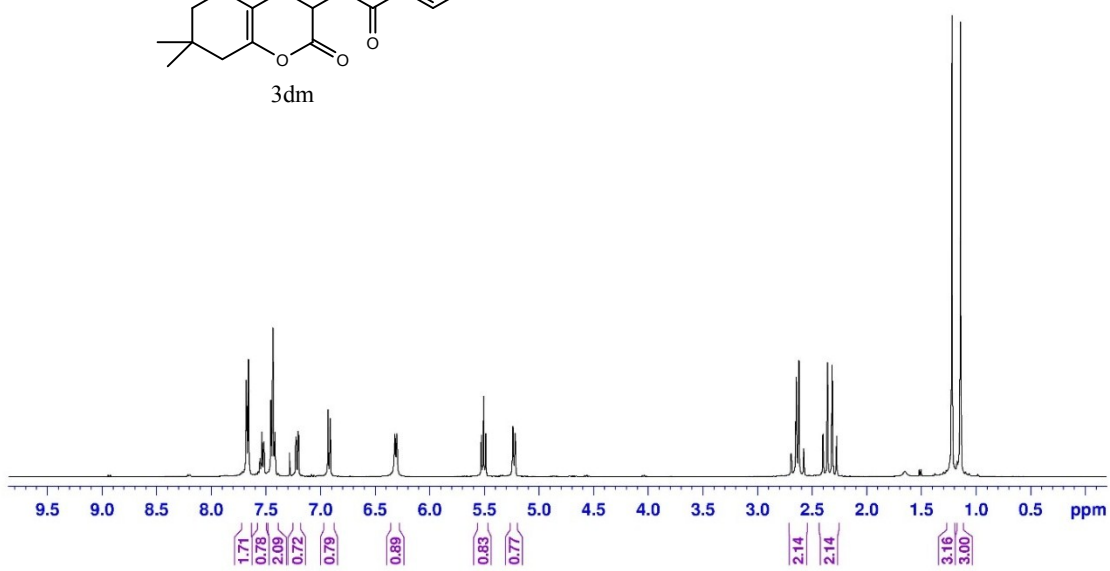
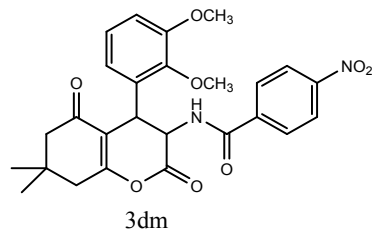
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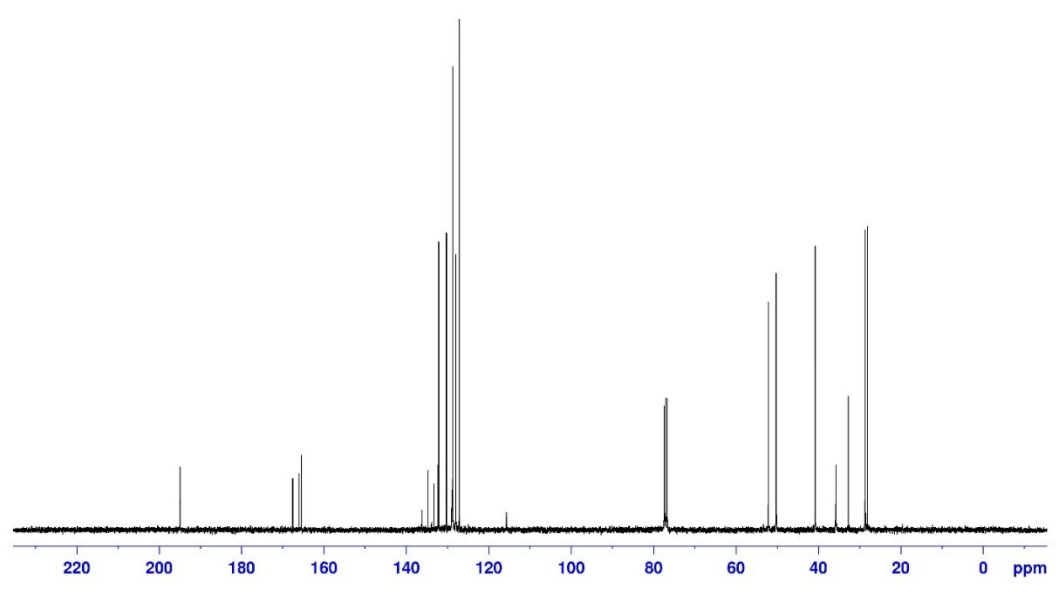


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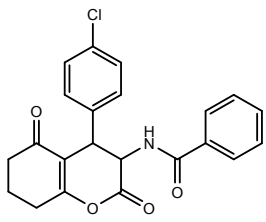
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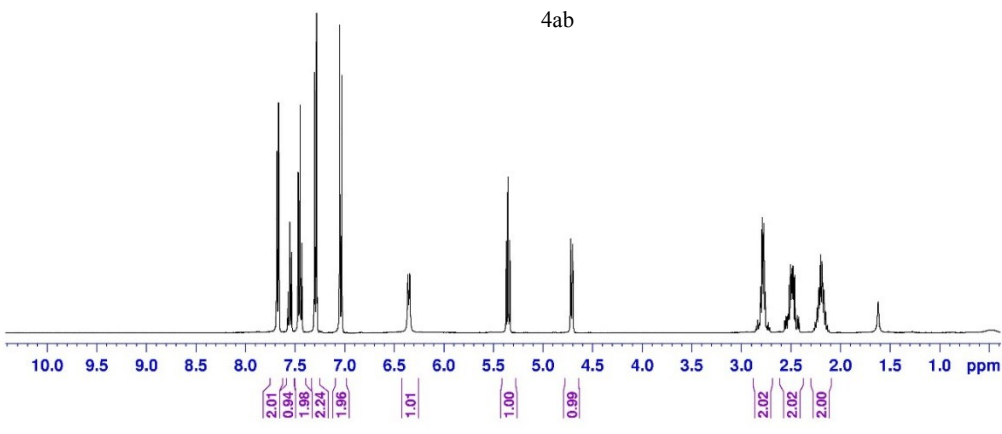
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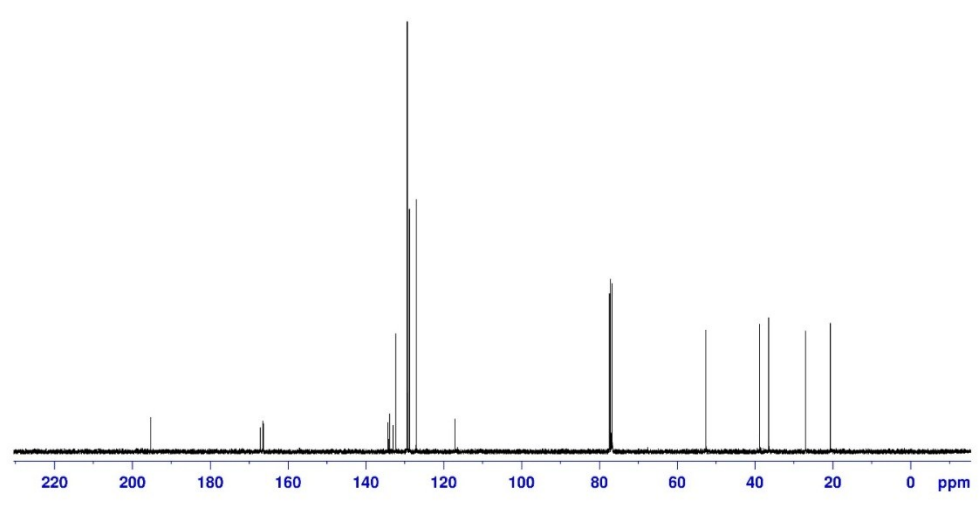
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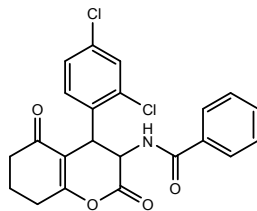
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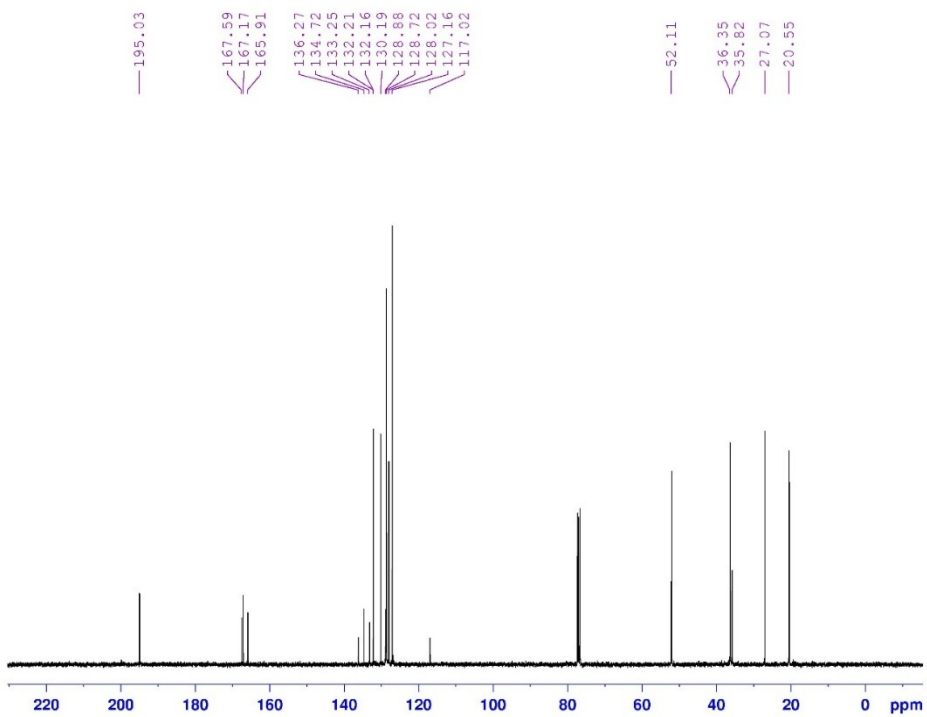
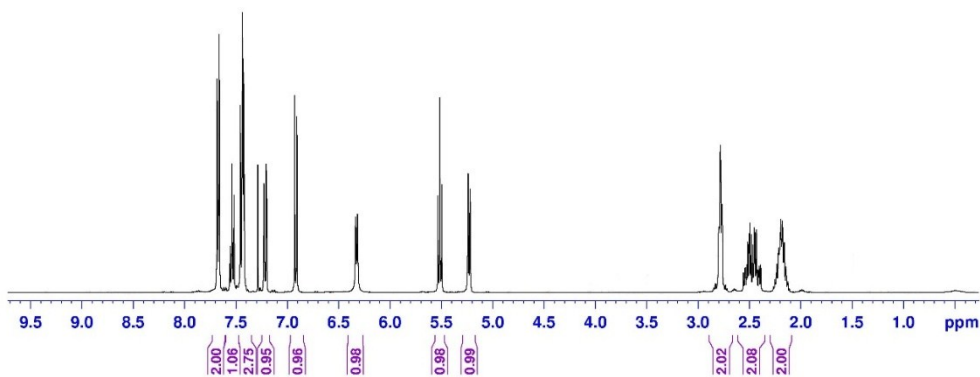
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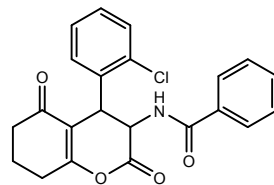
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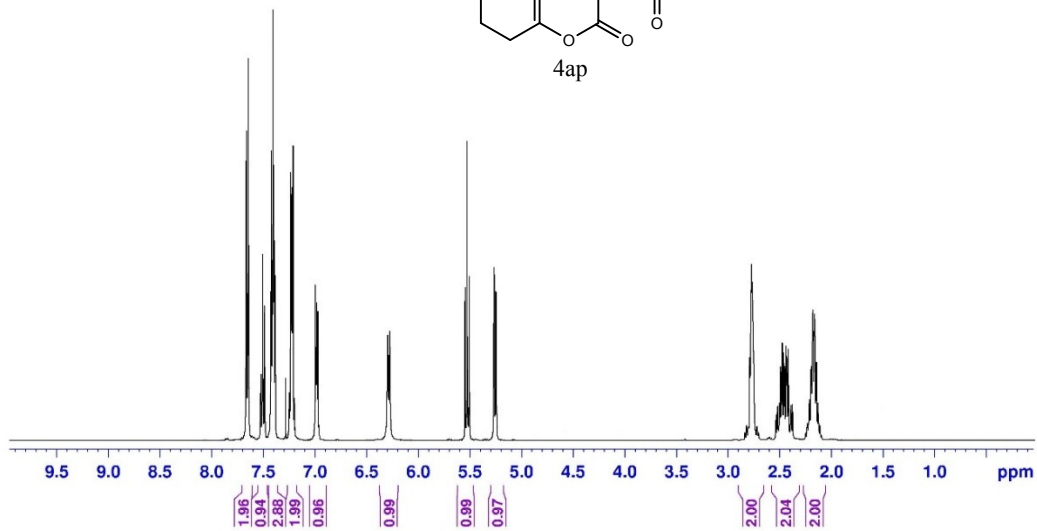
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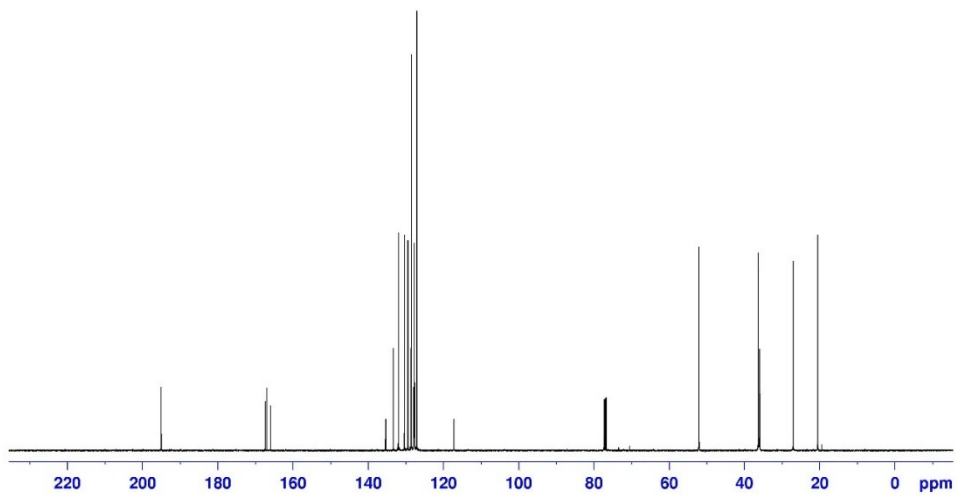
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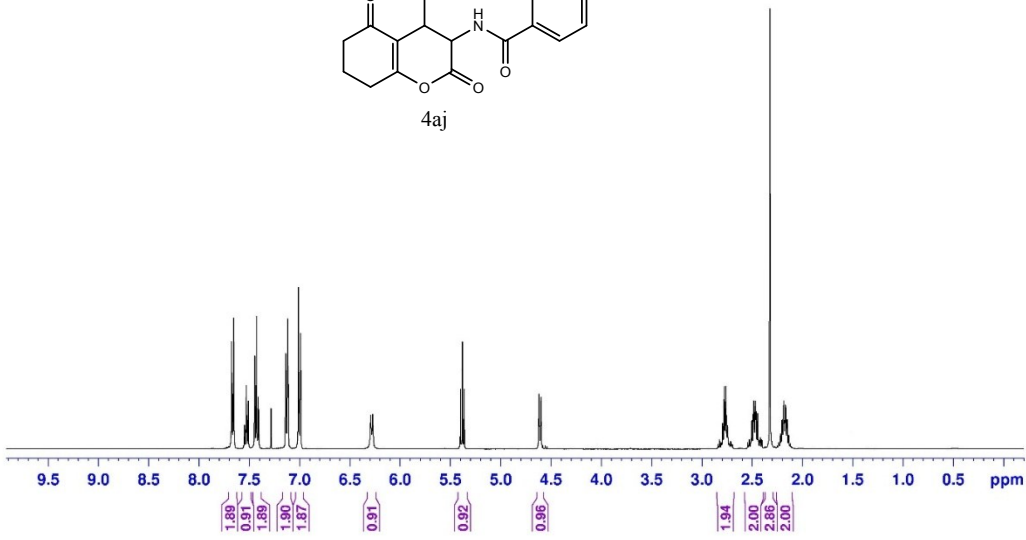
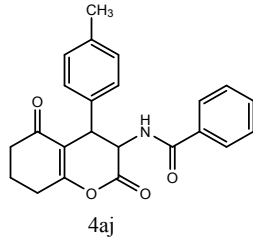
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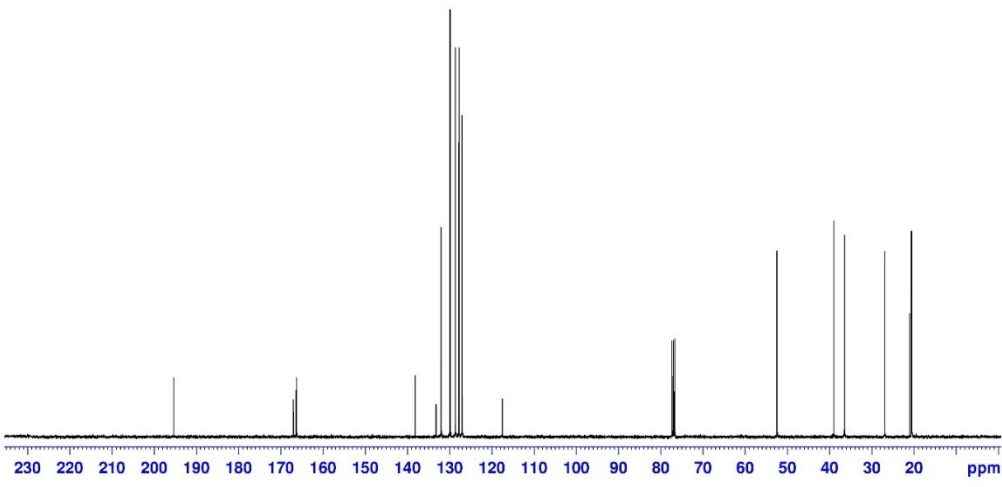
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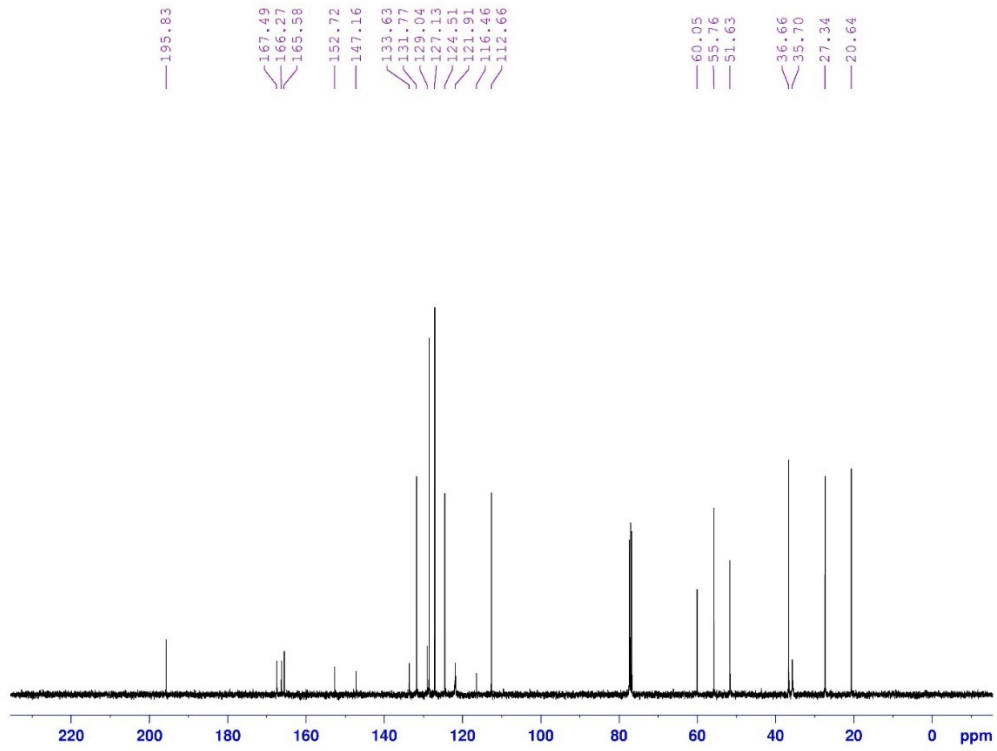
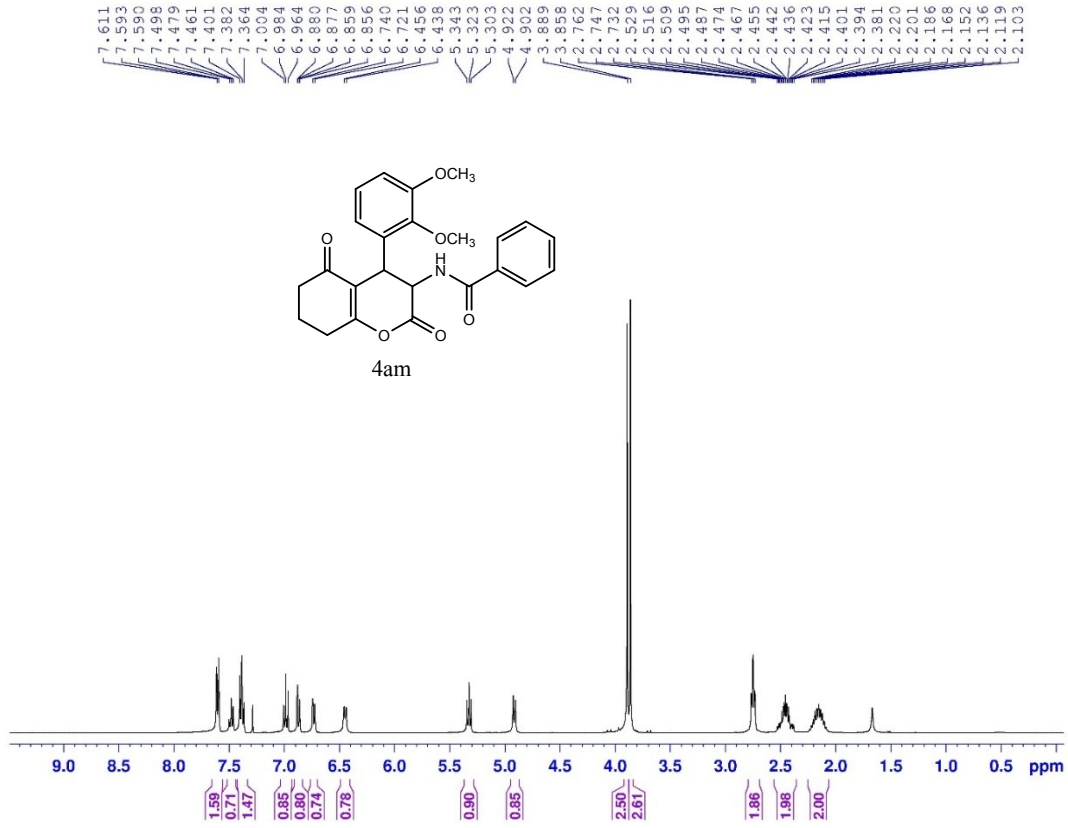
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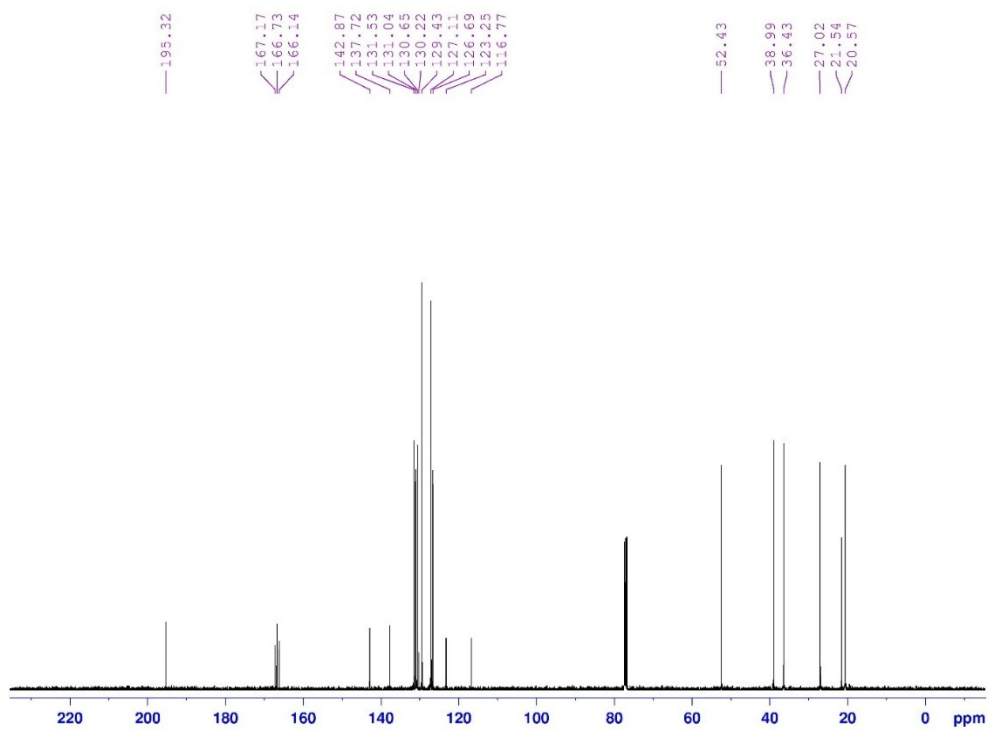
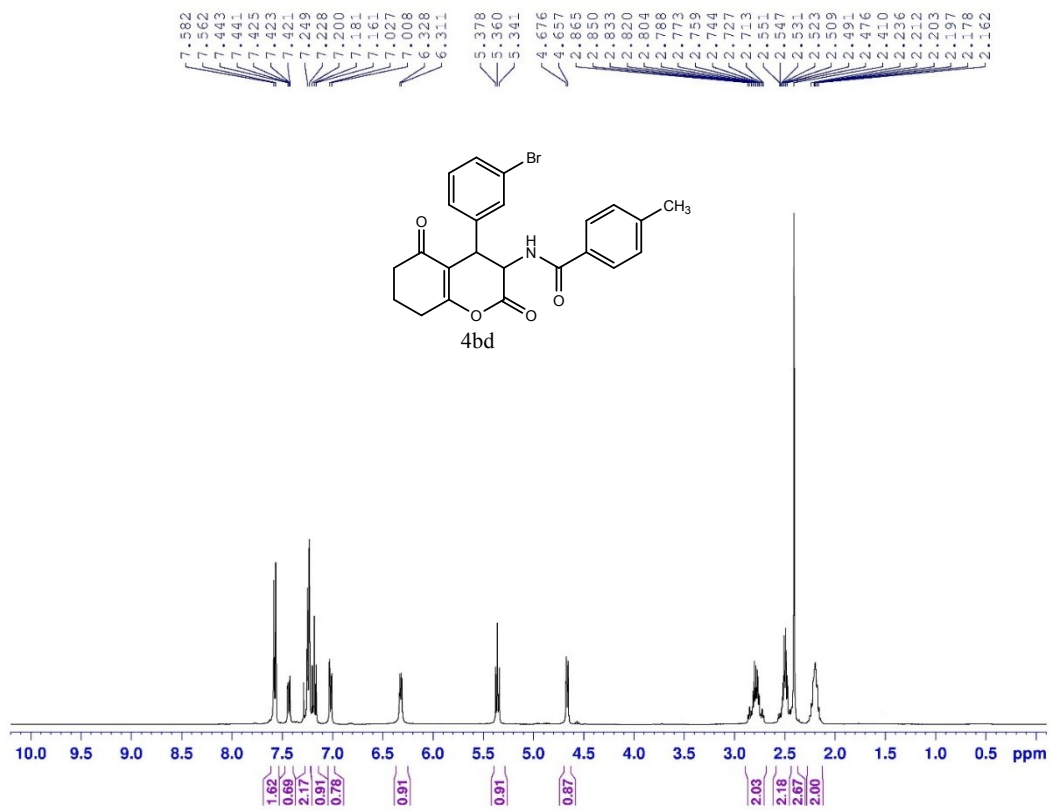
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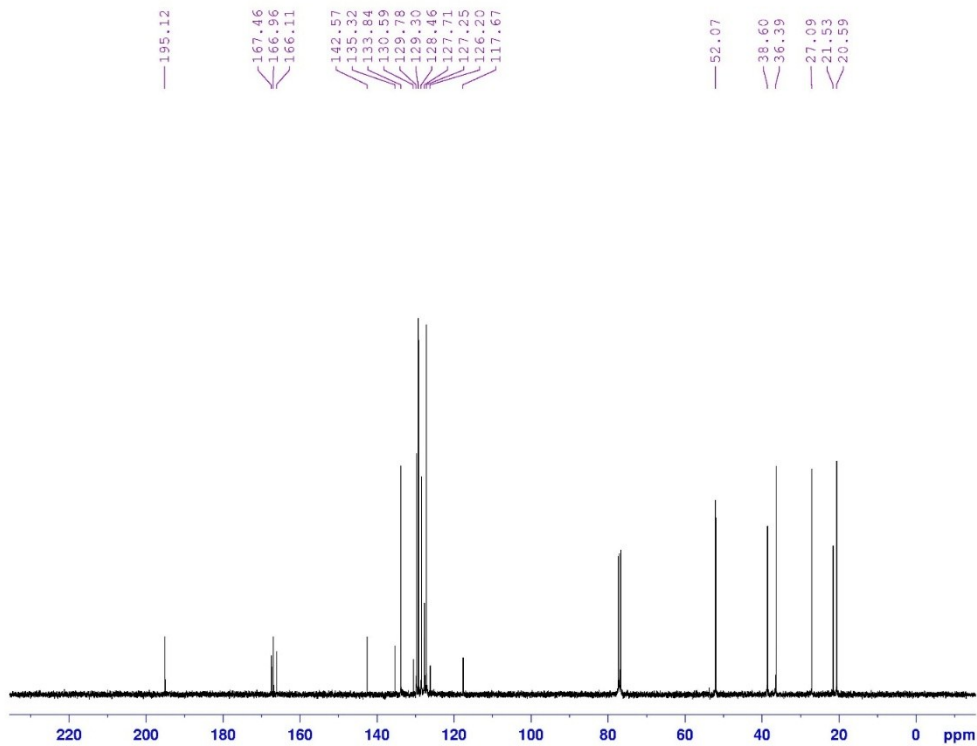
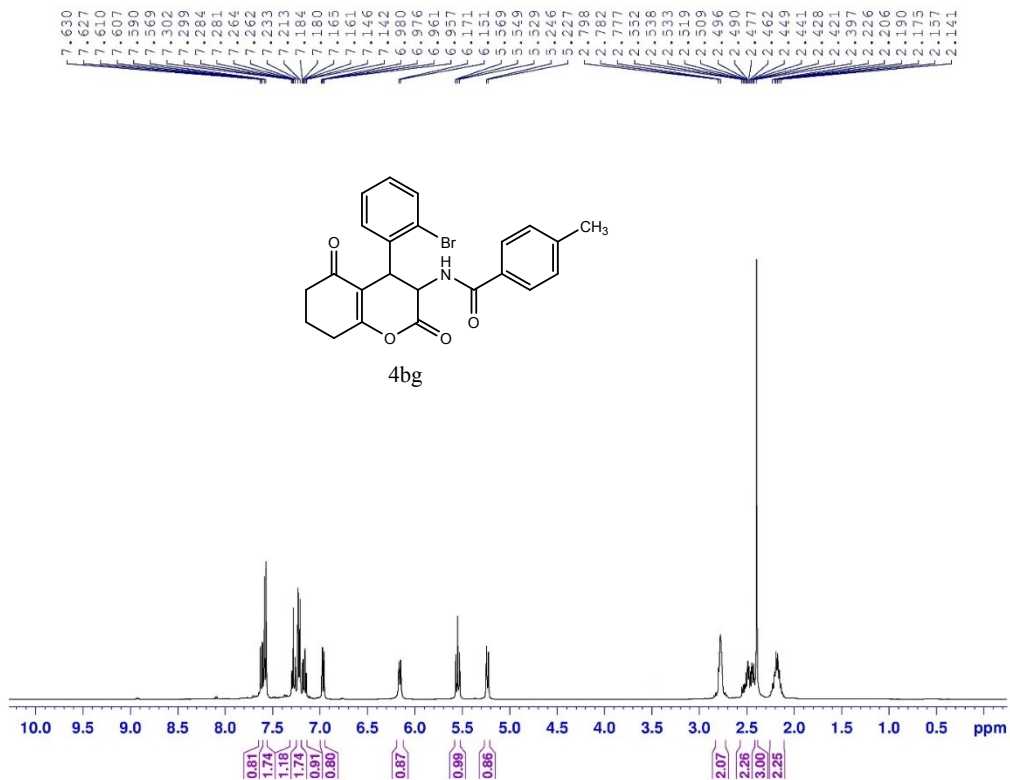
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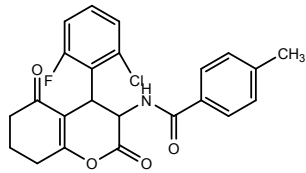




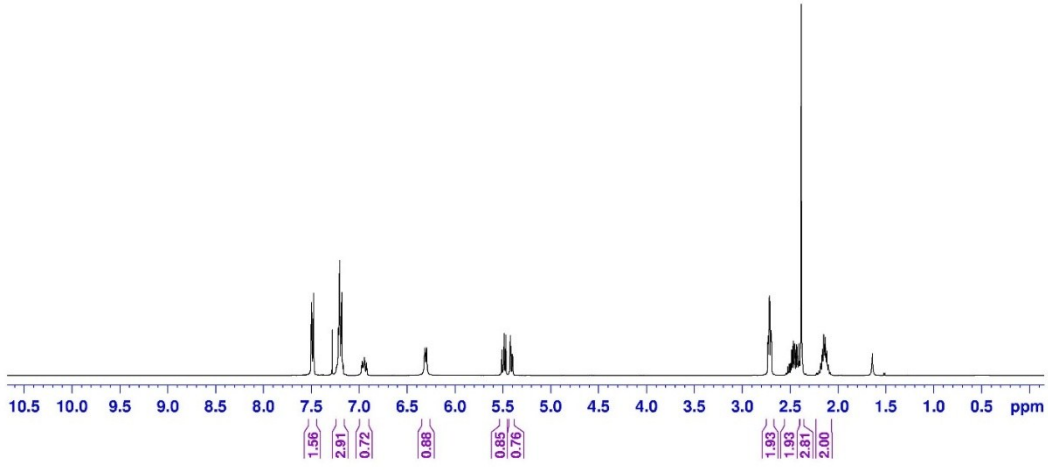




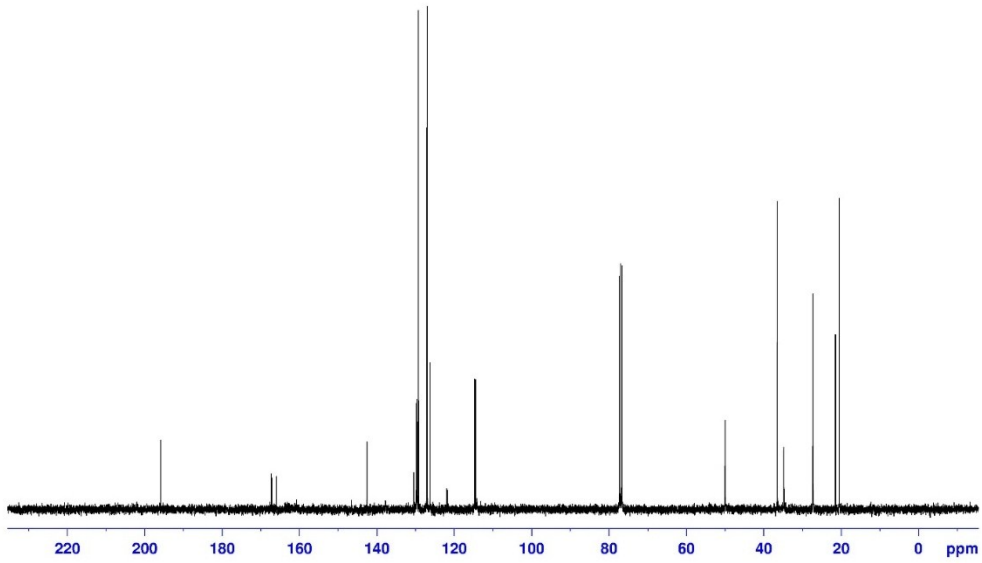
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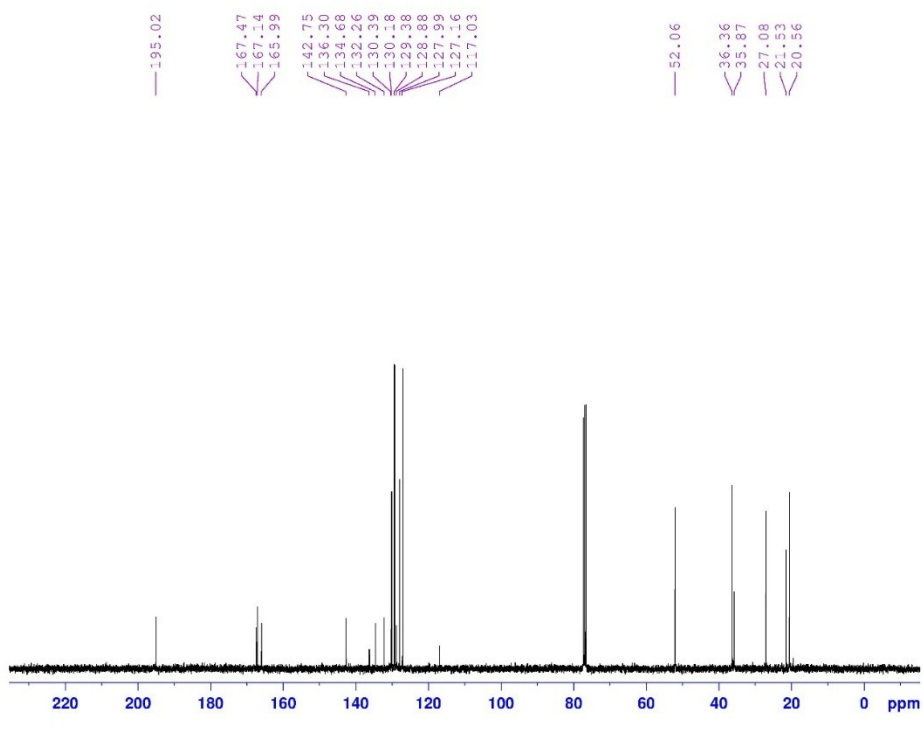
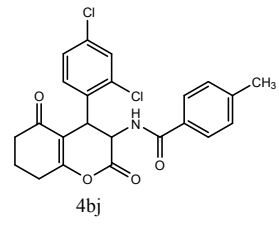
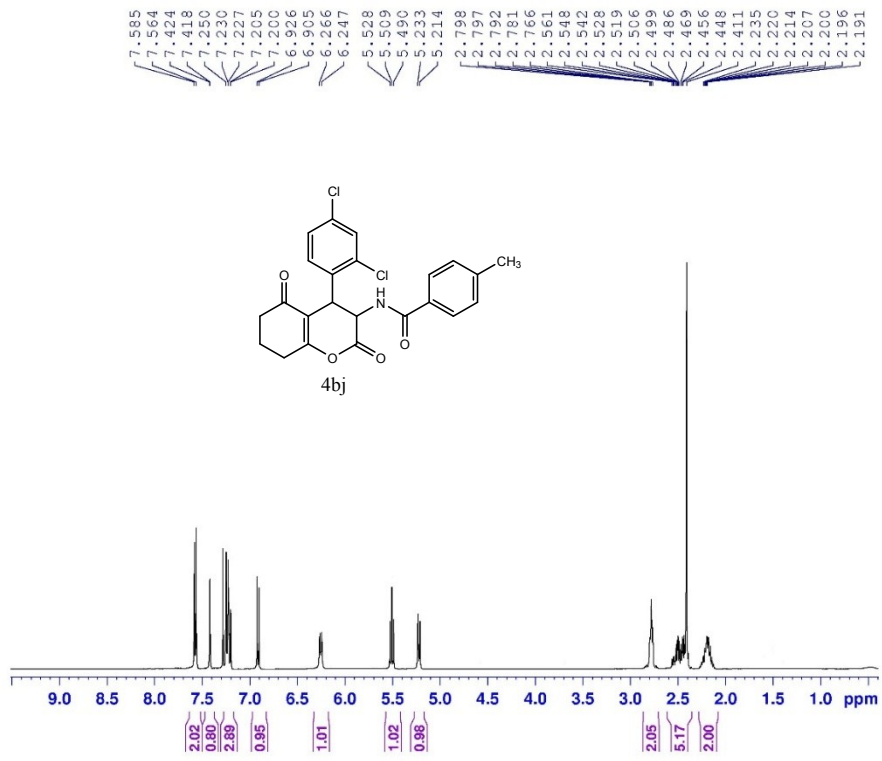


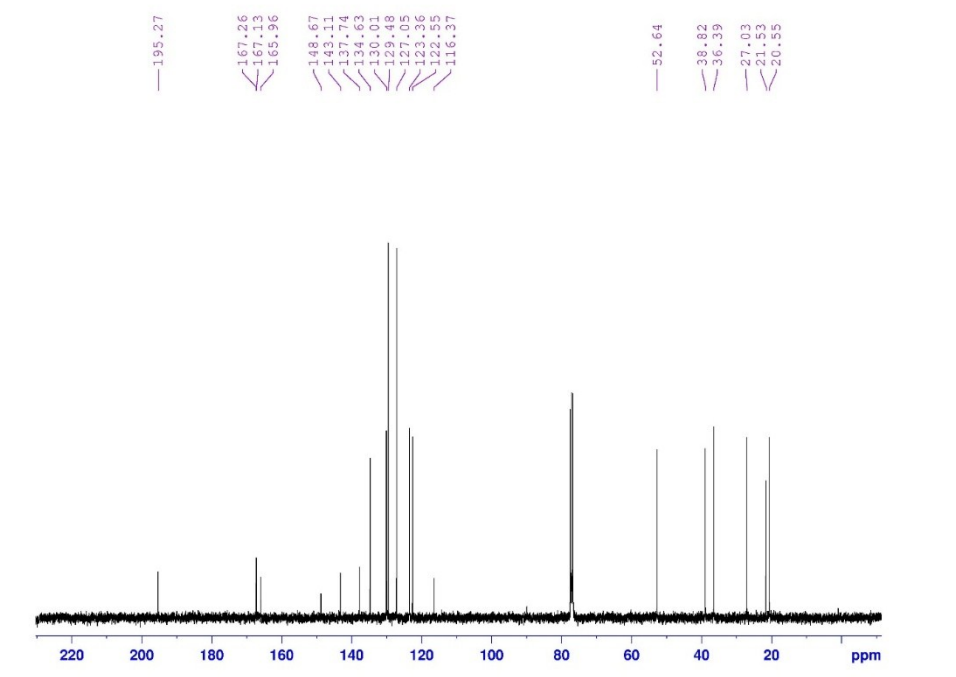
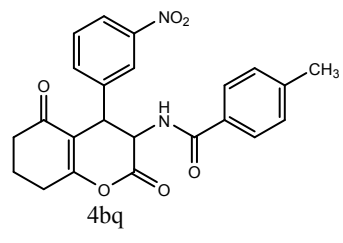
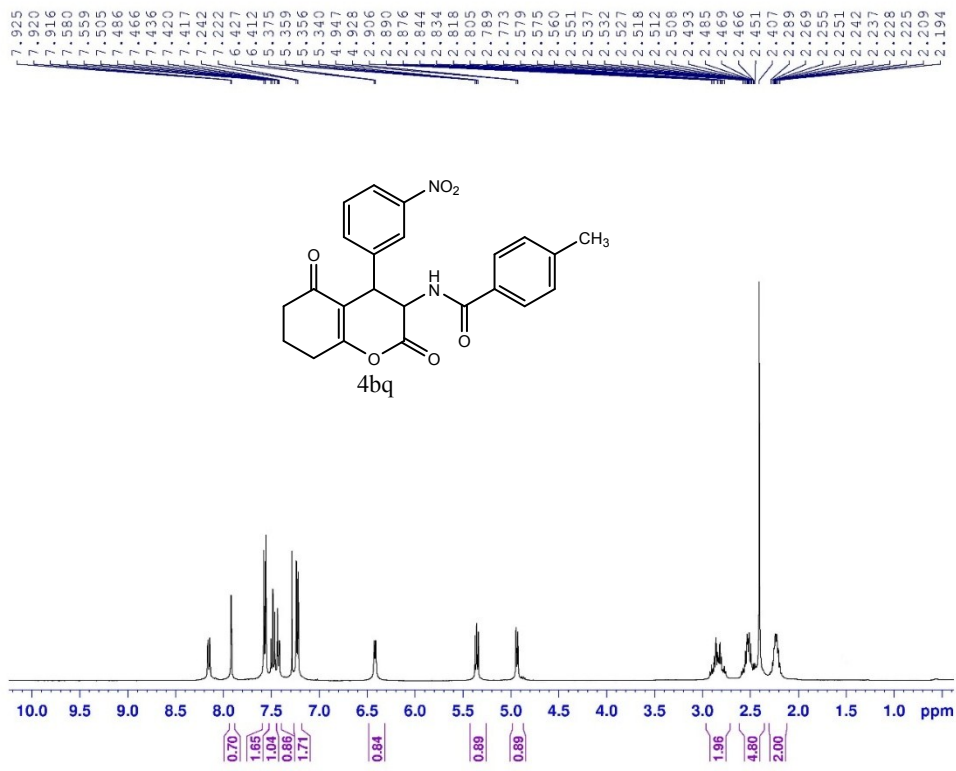
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