

**An Efficient Synthesis of Functionalized
Dihydro-1*H*-indol-4(5*H*)-ones via One-Pot Three-Component
Reaction under Catalyst-Free Conditions**

Huiyan Wang,^{1,2} Daqing Shi^{1,*}

†

¹ *Key Laboratory of Organic Synthesis of Jiangsu Province, College of Chemistry, Chemical Engineering and Materials Science, Soochow University, Suzhou 215123, P. R. China*

² *Department of Chemical Engineering, Huaihai Institute of Technology, Lianyungang 222005, P. R. China*

* *Corresponding author. E-mail: dqshi@suda.edu.cn*

Table of contents

Experimental Section.....	S3-S17
X-ray crystallographic information.....	S18-S21
Copies of ^1H NMR and ^{13}C NMR of compounds	S22-S58

General Experimental Methods

Melting points are uncorrected. IR spectra were recorded on Varian F-1000 spectrometer in KBr with absorptions in cm^{-1} . ^1H NMR and ^{13}C NMR were determined on Varian Inova-400 MHz or Inova-300 MHz spectrometer in $\text{DMSO-}d_6$ solution. J values are in Hz. Chemical shifts are expressed in ppm downfield from internal standard TMS. HRMS analyses were carried out using TOF-MS or GCT-TOF instrument. Phenylglyoxal monohydrate and 4-methoxy phenylglyoxal monohydrate were obtained from Alfa Aesar Company.

General procedure for the synthesis of arylglyoxal monohydrate **2**¹

SeO_2 (1390mg, 1.25 mmol) was suspended in a mixture of 1,4-dioxane (6 mL) and H_2O (0.4 mL) and aryl methyl ketones (5.0 mmol) was added just before microwave irradiation [ramp time (1 min) to a maximum power 250 W, hold time (15 min) at a set temperature of $100\text{ }^\circ\text{C}$]. then, the solution was cooled to room temperature. The solution was filtered in order to remove Se, and washed with MeOH (20 mL). The combined organic filtrate was concentrated under reduced pressure and the organic residue subsequently stirred in boiling H_2O (35 mL) and filtered hot to removed any insoluble precipitate. The aqueous filtrate was cooled on ice to afford crystals of the arylglyoxal monohydrate product were collected under vacuum filtration washed with cold H_2O and dried in a vacuum desiccator.

General procedure for the synthesis of pyrroles **4**

A dry 25 mL flask was charged with 1,3-dicarbonyl compounds **1** (1 mmol), phenylglyoxal monohydrate **2** (1 mmol), enaminones **3** (1 mmol) and ethanol (5 mL). The mixture was stirred at refluxing temperature for 0.7-5 h. After completion of the reaction (confirmed by TLC), the reaction mixture was cooled to room temperature. The crystalline solids were collected and washed with little cold ethanol to give the pure products **4**.

Reference:

1. Young, R. M.; Davies-Coleman, M. T. Microwave-assisted selenium dioxide oxidation of aryl methyl ketones to aryl glyoxals, *Tetrahedron Lett.* **2011**, *52*, 4036-4038.

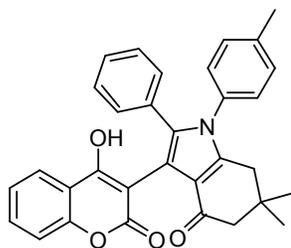
Characterizations for compounds

***p*-tolylglyoxal monohydrate (2b)**: white powder (76%); m.p.: 78-80 °C; IR (KBr, ν , cm^{-1}): 3407, 3351, 3050, 2917, 2858, 1689, 1607, 1432, 1383, 1301, 1228, 1188, 1110, 1036, 965, 859, 809, 788, 737, 693, 653; ^1H NMR (DMSO- d_6 , 400 MHz): δ (ppm): 7.94 (d, $J = 7.6$ Hz, 2H, ArH), 7.30 (d, $J = 8.0$ Hz, 2H, ArH), 6.68 (s, 2H, 2 \times OH), 5.65 (s, 1H, CH), 2.35 (s, 3H, CH₃).

4-chlorophenylglyoxal monohydrate (2d): white powder (80%); m.p.: 97-100 °C; IR (KBr, ν , cm^{-1}): 3465, 3428, 3043, 2944, 1693, 1589, 1569, 1489, 1429, 1399, 1321, 1294, 1223, 1118, 1092, 997, 976, 827, 717, 647; ^1H NMR (DMSO- d_6 , 400 MHz): δ (ppm) : 8.05 (d, $J = 8.4$ Hz, 2H, ArH), 7.56 (d, $J = 8.0$ Hz, 2H, ArH), 6.65-6.83 (m, 2H, 2 \times OH), 5.60 (t, $J = 7.2$ Hz, 1H, CH).

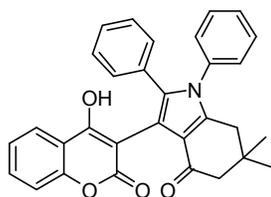
4-bromophenylglyoxal monohydrate (2e): white powder (82%); m.p.: 124-127 °C; IR (KBr, ν , cm^{-1}): 3468, 3430, 3097, 3039, 2944, 1693, 1584, 1565, 1485, 1428, 1395, 1292, 1220, 1170, 1118, 1070, 995, 972, 955, 892, 861, 821, 718, 643; ^1H NMR (DMSO- d_6 , 400 MHz): δ (ppm) : 7.97 (d, $J = 6.4$ Hz, 2H, ArH), 7.70 (d, $J = 6.4$ Hz, 2H, ArH), 6.83 (s, 2H, 2 \times OH), 5.60 (s, 1H, CH).

3-(4-hydroxy-2-oxo-2*H*-chromen-3-yl)-6,6-dimethyl-2-phenyl-1-(*p*-tolyl)-6,7-dihydro-1*H*-indol-4(5*H*)-one (4{*I,I,I*})



ol-4(5*H*)-one (4{*I,I,I*}): white powder (89%); m.p.: 162-163 °C; IR (KBr, ν , cm^{-1}): 3430, 3280, 3058, 2956, 2869, 1729, 1694, 1665, 1624, 1515, 1492, 1451, 1408, 1366, 1270, 1194, 1103, 1044, 1020, 961, 897, 837, 757, 699; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.81 (s, 1H, OH), 7.77 (d, $J = 8.0$ Hz, 1H, ArH), 7.54 (t, $J = 7.6$ Hz, 1H, ArH), 7.31-7.24 (m, 2H, ArH), 7.19 (d, $J = 7.6$ Hz, 2H, ArH), 7.09-6.99 (m, 7H, ArH), 2.56 (d, $J = 16.4$ Hz, 1H, CH₂), 2.47-2.45 (m, 1H, CH₂), 2.30-2.26 (m, 4H, CH₂+CH₃), 2.16 (d, $J = 15.6$ Hz, 1H, CH₂), 1.04 (s, 3H, CH₃), 1.01 (s, 3H, CH₃); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.80, 162.17, 161.17, 152.78, 144.37, 138.06, 135.37, 134.90, 132.36, 131.41, 130.19, 130.02, 129.77, 128.26, 127.96, 127.65, 124.25, 123.86, 118.27, 116.45, 109.36, 100.34, 52.62, 36.85, 35.38, 28.91, 28.38, 21.07; HRMS calcd for C₃₂H₂₇NNaO₄ [M+Na]⁺: 512.1838, found: 512.1842.

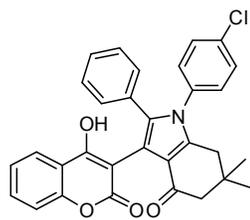
3-(4-hydroxy-2-oxo-2*H*-chromen-3-yl)-6,6-dimethyl-1,2-diphenyl-6,7-dihydro-1*H*-indol-4(5*H*)-one (4{*I,I,2*})



)-one (4{*I,I,2*}): white powder (91%); m.p.: 308-310 °C; IR (KBr, ν , cm^{-1}): 3425, 3282, 3165, 3002, 2948, 2875, 1688, 1664, 1640, 1578, 1496, 1411, 1199, 1117, 1045, 1021, 925, 806, 751, 704, 648; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.79 (s, 1H, OH), 7.74 (d, $J =$

7.6 Hz, 1H, ArH), 7.49 (t, $J = 7.6$ Hz, 1H, ArH), 7.36-7.16 (m, 7H, ArH), 7.02-6.94 (m, 5H, ArH), 2.53 (d, $J = 16.4$ Hz, 1H, CH₂), 2.44 (d, $J = 16.8$ Hz, 1H, CH₂), 2.25 (d, $J = 15.6$ Hz, 1H, CH₂), 2.13 (d, $J = 16.0$ Hz, 1H, CH₂), 1.00 (s, 3H, CH₃), 0.97 (s, 3H, CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.94, 162.24, 161.24, 152.78, 151.25, 144.34, 137.43, 135.37, 133.56, 132.41, 131.31, 129.74, 128.66, 128.25, 127.70, 124.30, 123.86, 118.35, 116.49, 116.42, 109.46, 100.28, 52.59, 36.82, 35.40, 28.85, 28.40; HRMS calcd for C₃₁H₂₅NNaO₄ [M+Na]⁺: 498.1681, found: 498.1685.

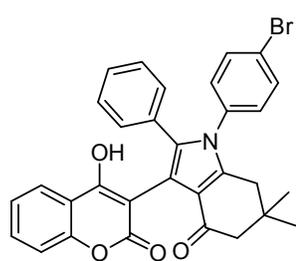
1-(4-chlorophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-2-phenyl-6,7-dihydro



-1H-indol-4(5H)-one (4{1,1,3}): light yellow powder (88%); m.p.: 170-172 °C; IR (KBr, ν , cm⁻¹): 3433, 3060, 3002, 2956, 2869, 1731, 1665, 1574, 1495, 1450, 1366, 1272, 1194, 1116, 1089, 1043, 1014, 962, 923, 896, 843, 757, 723, 698; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.85 (s, 1H, OH), 7.79 (d, $J = 6.4$ Hz, 1H, ArH), 7.52 (t, $J =$

8.0 Hz, 1H, ArH), 7.46 (d, $J = 7.6$ Hz, 2H, ArH), 7.30-7.23 (m, 4H, ArH), 7.08-7.02 (m, 5H, ArH), 2.61 (d, $J = 16.8$ Hz, 1H, CH₂), 2.49 (d, $J = 21.2$ Hz, 1H, CH₂), 2.30 (d, $J = 15.2$ Hz, 1H, CH₂), 2.18 (d, $J = 12.0$ Hz, 1H, CH₂), 1.05 (s, 3H, CH₃), 1.02 (s, 3H, CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.89, 162.18, 161.33, 152.83, 144.38, 136.37, 135.37, 133.23, 132.38, 131.09, 130.42, 130.35, 130.06, 129.83, 128.39, 127.85, 124.24, 123.88, 118.59, 116.43, 109.77, 100.15, 52.61, 36.71, 35.41, 28.91, 28.30; HRMS calcd for C₃₁H₂₄ClNNaO₄ [M+Na]⁺: 532.1292, found: 532.1294

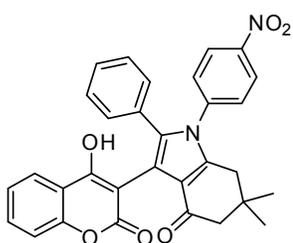
1-(4-bromophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-2-phenyl-6,7-dihydro



o-1H-indol-4(5H)-one (4{1,1,4}): white powder (90%); m.p.: 176-178 °C; IR (KBr, ν , cm⁻¹): 3432, 3058, 3002, 2956, 2869, 1731, 1665, 1573, 1494, 1450, 1411, 1365, 1271, 1194, 1115, 1100, 1070, 1044, 1012, 961, 923, 896, 842, 757, 698; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.84 (s, 1H, OH), 7.79 (d, $J =$

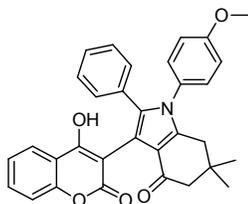
7.6 Hz, 1H, ArH), 7.61-7.52 (m, 3H, ArH), 7.31-7.25 (m, 2H, ArH), 7.18 (d, $J = 7.6$ Hz, 2H, ArH), 7.10-7.00 (m, 5H, ArH), 2.61 (d, $J = 16.8$ Hz, 1H, CH₂), 2.49 (d, $J = 16.4$ Hz, 1H, CH₂), 2.30 (d, $J = 16.0$ Hz, 1H, CH₂), 2.17 (d, $J = 16.0$ Hz, 1H, CH₂), 1.05 (s, 3H, CH₃), 1.03 (s, 3H, CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.88, 162.13, 161.34, 152.82, 144.32, 136.79, 135.28, 132.72, 132.41, 131.07, 130.36, 129.81, 128.42, 127.86, 124.27, 123.89, 121.72, 118.59, 116.49, 116.42, 109.80, 100.10, 52.61, 36.71, 35.43, 28.94, 28.28; HRMS calcd for C₃₁H₂₅BrNO₄ [M+H]⁺: 554.0967, found: 554.0937.

3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-1-(4-nitrophenyl)-2-phenyl-6,7-dihydro-



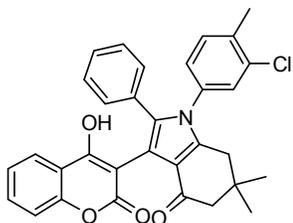
1H-indol-4(5H)-one (4{1,1,5}): yellow green powder (77%); m.p.: 185-186 °C; IR (KBr, ν , cm^{-1}): 3430, 3085, 3001, 2967, 2869, 1710, 1708, 1641, 1575, 1524, 1492, 1449, 1406, 1348, 1287, 1108, 1042, 1012, 923, 713, 649; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.98 (s, 1H, OH), 8.24 (d, J = 8.0 Hz, 2H, ArH), 7.78 (d, J = 7.2 Hz, 1H, ArH), 7.55-7.46 (m, 3H, ArH), 7.30-7.25 (m, 2H, ArH), 7.09-6.99 (m, 5H, ArH), 2.69 (d, J = 16.8 Hz, 1H, CH_2), 2.52 (d, J = 16.8 Hz, 1H, CH_2), 2.31 (d, J = 15.6 Hz, 1H, CH_2), 2.16 (d, J = 16.0 Hz, 1H, CH_2), 1.03 (s, 6H, $2\times\text{CH}_3$); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.88, 161.96, 161.44, 152.84, 146.90, 144.28, 143.08, 135.20, 132.46, 130.78, 129.76, 129.49, 128.58, 128.01, 125.08, 124.28, 123.91, 119.11, 116.51, 116.36, 110.58, 99.83, 52.54, 36.72, 35.50, 28.97, 28.10; HRMS calcd for $\text{C}_{31}\text{H}_{24}\text{N}_2\text{NaO}_6$ $[\text{M}+\text{Na}]^+$: 543.1532, found: 543.1508.

3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-1-(4-methoxyphenyl)-6,6-dimethyl-2-phenyl-6,7-dihydro-



ro-1H-indol-4(5H)-one (4{1,1,6}): white powder (90%); m.p.: 270-271 °C; IR (KBr, ν , cm^{-1}): 3418, 3057, 3001, 2956, 2869, 1693, 1658, 1572, 1513, 1493, 1451, 1408, 1367, 1298, 1249, 1199, 1172, 1104, 1025, 982, 963, 898, 795, 756, 701; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.76 (s, 1H, OH), 7.77 (d, J = 7.6 Hz, 1H, ArH), 7.54 (t, J = 7.6 Hz, 1H, ArH), 7.31-7.25 (m, 2H, ArH), 7.13 (d, J = 7.6 Hz, 2H, ArH), 7.07-6.93 (m, 7H, ArH), 3.71 (s, 3H, OCH_3), 2.54 (d, J = 16.8 Hz, 1H, CH_2), 2.44 (d, J = 15.6 Hz, 1H, CH_2), 2.27 (d, J = 16.0 Hz, 1H, CH_2), 2.15 (d, J = 16.0 Hz, 1H, CH_2), 1.05 (s, 3H, CH_3), 1.02 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.83, 162.20, 161.14, 159.07, 152.77, 144.63, 135.54, 132.38, 131.40, 130.11, 129.78, 129.64, 129.42, 128.26, 127.66, 124.28, 123.85, 118.09, 116.48, 114.78, 109.11, 100.37, 55.74, 52.60, 36.78, 35.37, 28.91, 28.38; HRMS calcd for $\text{C}_{32}\text{H}_{27}\text{NNaO}_5$ $[\text{M}+\text{Na}]^+$: 528.1787, found: 528.1760.

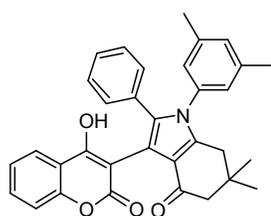
1-(3-chloro-4-methylphenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-2-phenyl-6,



7-dihydro-1H-indol-4(5H)-one (4{1,1,7}): light red crystal (94%); m.p.: 152-154 °C; IR (KBr, ν , cm^{-1}): 3427, 3058, 3002, 2957, 2935, 2870, 1664, 1579, 1496, 1446, 1268, 1191, 1116, 1044, 1013, 966, 923, 896, 831, 758, 717, 700, 649; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.82 (s, 1H, OH), 7.80 (d, J = 7.6 Hz, 1H, ArH), 7.51 (t, J = 7.2 Hz, 1H, ArH), 7.35 (d, J = 7.6 Hz, 1H, ArH), 7.29-7.22 (m, 3H, ArH), 7.15-7.05 (m, 6H,

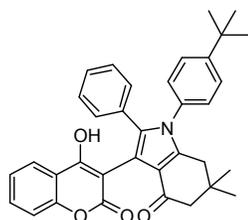
ArH), 2.64 (d, $J = 16.4$ Hz, 1H, CH₂), 2.47 (d, $J = 16.0$ Hz, 1H, CH₂), 2.30 (d, $J = 16.0$ Hz, 1H, CH₂), 2.24 (s, 3H, CH₃), 2.15 (d, $J = 15.6$ Hz, 1H, CH₂), 1.03 (s, 3H, CH₃), 1.02 (s, 3H, CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.73, 162.00, 161.29, 152.84, 144.44, 136.48, 136.12, 135.41, 133.72, 132.32, 132.07, 131.12, 129.79, 128.37, 127.82, 127.04, 124.18, 123.87, 118.50, 116.46, 116.40, 109.70, 100.16, 52.63, 36.70, 35.41, 29.04, 28.15, 19.65; HRMS calcd for C₃₂H₂₆ClNO₄[M]⁺: 523.1550, found: 523.1553.

1-(3,5-dimethylphenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-2-phenyl-6,7-dihydro-1H-indol-4(5H)-one (4{1,1,8}):



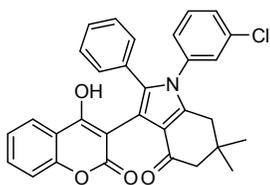
hydro-1H-indol-4(5H)-one (4{1,1,8}): white powder (87%); m.p.: 294-295 °C; IR (KBr, ν , cm⁻¹): 3440, 3058, 3002, 2953, 1691, 1661, 1612, 1573, 1493, 1449, 1409, 1365, 1329, 1206, 1103, 1077, 1044, 970, 923, 900, 860, 756, 698; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.74 (s, 1H, OH), 7.78 (d, $J = 8.0$ Hz, 1H, ArH), 7.54 (t, $J = 7.6$ Hz, 1H, ArH), 7.31-7.25 (m, 2H, ArH), 7.09-6.96 (m, 6H, ArH), 6.82 (s, 2H, ArH), 2.57 (d, $J = 16.4$ Hz, 1H, CH₂), 2.48 (d, $J = 15.6$ Hz, 1H, CH₂), 2.27 (d, $J = 16.0$ Hz, 1H, CH₂), 2.20-2.14 (m, 7H, 1×CH₂+2×CH₃), 1.05 (s, 3H, CH₃), 1.02 (s, 3H, CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.69, 162.10, 161.12, 152.80, 144.22, 138.84, 137.37, 135.29, 132.31, 131.46, 130.61, 130.08, 129.69, 128.20, 127.60, 125.77, 124.21, 123.88, 118.25, 116.48, 109.41, 100.39, 52.66, 36.83, 35.41, 28.89, 28.40, 21.19; HRMS calcd for C₃₃H₂₉NNaO₄ [M+Na]⁺: 526.1994, found: 526.1996.

1-(4-(tert-butyl)phenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-2-phenyl-6,7-dihydro-1H-indol-4(5H)-one (4{1,1,9}):



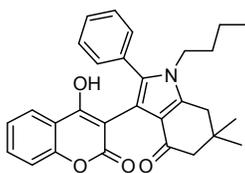
hydro-1H-indol-4(5H)-one (4{1,1,9}): light red powder (88%); m.p.: 268-269 °C; IR (KBr, ν , cm⁻¹): 3470, 3059, 3001, 2954, 2869, 1698, 1692, 1664, 1630, 1572, 1518, 1491, 1450, 1409, 1366, 1265, 1188, 1120, 1101, 1044, 1014, 962, 897, 756, 699; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.76 (s, 1H, OH), 7.76 (d, $J = 7.6$ Hz, 1H, ArH), 7.54 (t, $J = 7.6$ Hz, 1H, ArH), 7.41 (d, $J = 7.6$ Hz, 2H, ArH), 7.31-7.24 (m, 2H, ArH), 7.13 (d, $J = 7.6$ Hz, 2H, ArH), 7.04-6.96 (m, 5H, ArH), 2.56 (d, $J = 16.8$ Hz, 1H, CH₂), 2.48 (d, $J = 13.2$ Hz, 1H, CH₂), 2.27 (d, $J = 16.0$ Hz, 1H, CH₂), 2.15 (d, $J = 16.0$ Hz, 1H, CH₂), 1.22 (s, 9H, 3×CH₃), 1.05 (s, 3H, CH₃), 1.02 (s, 3H, CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.91, 162.27, 161.26, 152.77, 151.03, 144.36, 135.38, 134.83, 133.48, 132.38, 131.37, 129.74, 128.17, 127.74, 126.42, 124.28, 123.86, 118.25, 116.48, 109.85, 109.43, 100.28, 52.62, 36.82, 35.40, 34.85, 31.47, 28.82, 28.41; HRMS calcd for C₃₅H₃₃NNaO₄ [M+Na]⁺: 554.2307, found: 554.2319

1-(3-chlorophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-2-phenyl-6,7-dihydro



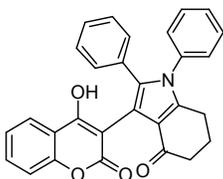
-1H-indol-4(5H)-one (4{1,1,10}): white powder (82%); m.p.: 278-280 °C; IR (KBr, ν , cm^{-1}): 3425, 3076, 3002, 2951, 2876, 1683, 1667, 1578, 1490, 1446, 1365, 1271, 1156, 1104, 1046, 972, 924, 899, 828, 794, 755, 717, 703, 649; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.83 (s, 1H, OH), 7.79 (d, $J = 7.6$ Hz, 1H, ArH), 7.53 (t, $J = 7.6$ Hz, 1H, ArH), 7.30-7.24 (m, 4H, ArH), 7.41 (s, 2H, ArH), 7.09-7.00 (m, 5H, ArH), 2.65 (d, $J = 16.4$ Hz, 1H, CH_2), 2.47 (d, $J = 15.6$ Hz, 1H, CH_2), 2.30 (d, $J = 16.0$ Hz, 1H, CH_2), 2.15 (d, $J = 16.0$ Hz, 1H, CH_2), 1.03 (s, 3H, CH_3), 1.02 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.79, 161.99, 161.33, 152.84, 144.40, 138.86, 135.37, 133.70, 132.39, 131.31, 131.02, 129.76, 128.78, 128.41, 128.16, 127.87, 127.21, 124.24, 123.89, 118.59, 116.50, 116.38, 109.82, 100.07, 52.62, 36.64, 35.44, 29.04, 28.13; HRMS calcd for $\text{C}_{31}\text{H}_{24}\text{ClNO}_4$ $[\text{M}]^+$: 509.1394, found: 509.1403.

butyl-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-2-phenyl-6,7-dihydro-1H-indol-4(



5H)-one (4{1,1,12}): red crystal (81%); m.p.: 114-116 °C; IR (KBr, ν , cm^{-1}): 3430, 3059, 3000, 2958, 2871, 1729, 1577, 1493, 1408, 1367, 1268, 1187, 1104, 1080, 1044, 968, 923, 897, 824, 757, 703, 649; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.59 (s, 1H, OH), 7.71 (d, $J = 7.6$ Hz, 1H, ArH), 7.51 (t, $J = 7.6$ Hz, 1H, ArH), 7.31-7.23 (m, 7H, ArH), 3.85-3.71 (m, 2H, CH_2), 2.75 (dd, $J_1 = 25.6$ Hz, $J_2 = 16.4$ Hz, 2H, CH_2), 2.22 (d, $J = 16.0$ Hz, 1H, CH_2), 2.12 (d, $J = 16.0$ Hz, 1H, CH_2), 1.48-1.36 (m, 2H, CH_2), 1.11-1.08 (m, 8H, $2 \times \text{CH}_3 + 1 \times \text{CH}_2$), 0.68 (t, $J = 7.2$ Hz, 3H, CH_3); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.32, 162.15, 160.66, 152.67, 143.23, 134.97, 132.15, 131.78, 130.18, 128.68, 128.38, 124.13, 123.76, 117.71, 116.50, 116.39, 108.72, 100.62, 52.61, 43.99, 35.93, 35.40, 32.58, 28.95, 28.69, 19.56, 13.80; HRMS calcd for $\text{C}_{29}\text{H}_{29}\text{NO}_4$ $[\text{M}]^+$: 455.2097, found: 455.2113.

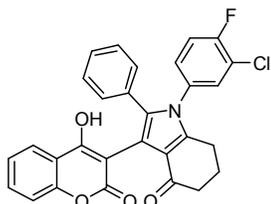
3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-1,2-diphenyl-6,7-dihydro-1H-indol-4(5H)-one



(4{1,1,13}): white powder (87%); m.p.: 282-284 °C; IR (KBr, ν , cm^{-1}): 3217, 3087, 2949, 2885, 1728, 1696, 1530, 1499, 1459, 1411, 1366, 1328, 1268, 1236, 1174, 1103, 1050, 1014, 965, 923, 898, 853, 793, 755, 697; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.82 (s, 1H, OH), 7.76 (d, $J = 7.6$ Hz, 1H, ArH), 7.54 (t, $J = 8.0$ Hz, 1H, ArH), 7.40-7.20 (m, 7H, ArH), 7.07-6.97 (m, 5H, ArH), 2.70-2.57 (m, 2H, CH_2), 2.38-2.28 (m, 2H, CH_2), 2.04-1.99 (m, 2H, CH_2); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 193.43, 162.22, 161.14, 152.77, 145.44, 137.42, 135.26, 132.36, 131.26,

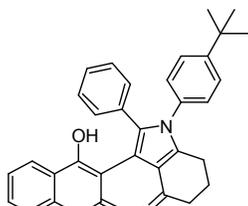
129.82, 129.66, 128.62, 128.26, 128.15, 127.76, 124.27, 123.96, 123.85, 119.52, 116.46, 109.71, 100.35, 38.57, 23.61, 23.05; HRMS calcd for C₂₉H₂₁NNaO₄ [M+Na]⁺: 470.1368, found: 470.1370.

1-(3-chloro-4-fluorophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-2-phenyl-6,7-dihydro-1H-



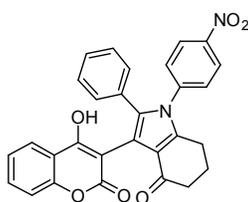
indol-4(5H)-one (4{1,1,14}): light yellow powder (90%); m.p.: 172-174 °C; IR (KBr, ν , cm⁻¹): 3430, 3065, 3001, 2937, 1729, 1573, 1502, 1409, 1362, 1260, 1116, 1102, 1074, 1019, 967, 923, 897, 835, 755, 699; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.82 (s, 1H, OH), 7.77 (d, *J* = 8.0 Hz, 1H, ArH), 7.55 (t, *J* = 7.6 Hz, 2H, ArH), 7.44 (t, *J* = 8.8 Hz, 1H, ArH), 7.31-7.25 (m, 3H, ArH), 7.13-7.05 (m, 3H, ArH), 7.00 (d, *J* = 6.8 Hz, 2H, ArH), 2.75-2.60 (m, 2H, CH₂), 2.35-2.26 (m, 2H, CH₂), 2.05-2.01 (m, 2H, CH₂); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 193.42, 162.02, 161.26, 158.73, 155.43, 152.78, 145.80, 135.29, 134.56, 132.45, 130.79, 130.35, 129.86, 129.31, 129.21, 128.48, 128.05, 124.31, 123.85, 120.44, 120.19, 119.63, 117.99, 117.69, 116.50, 116.34, 109.94, 100.04, 38.51, 23.52, 22.78; HRMS calcd for C₂₉H₁₈ClFNO₄ [M-H]⁺: 498.0908, found: 498.0909.

1-(4-(tert-butyl)phenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-2-phenyl-6,7-dihydro-1H-ind-



ol-4(5H)-one (4{1,1,15}): white powder (94%); m.p.: 278-279 °C; IR (KBr, ν , cm⁻¹): 3426, 3001, 2963, 2935, 1684, 1682, 1660, 1604, 1575, 1518, 1492, 1451, 1364, 1266, 1241, 1220, 1012, 969, 924, 761, 698; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.82 (s, 1H, OH), 7.74 (d, *J* = 7.6 Hz, 1H, ArH), 7.53 (t, *J* = 8.0 Hz, 1H, ArH), 7.39 (d, *J* = 8.0 Hz, 2H, ArH), 7.30-7.23 (m, 2H, ArH), 7.13 (d, *J* = 8.0 Hz, 2H, ArH), 7.03-6.98 (m, 5H, ArH), 2.62-2.59 (m, 2H, CH₂), 2.34-2.29 (m, 2H, CH₂), 2.01-1.98 (m, 2H, CH₂), 1.21 (s, 9H, 3×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 193.51, 162.29, 161.11, 152.72, 151.02, 145.62, 135.25, 134.82, 132.38, 131.28, 129.79, 128.19, 127.79, 127.65, 126.36, 124.30, 123.83, 119.35, 116.46, 116.43, 109.57, 100.35, 38.54, 34.83, 31.45, 23.59, 23.10; HRMS calcd for C₃₃H₂₉NNaO₄ [M+Na]⁺: 526.1994, found: 526.2000.

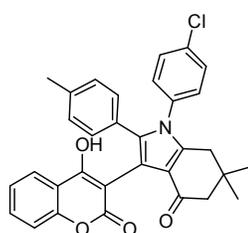
3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-1-(4-nitrophenyl)-2-phenyl-6,7-dihydro-1H-indol-4(5



H)-one (4{1,1,16}): light yellow powder (72%); m.p.: 167-170 °C; IR (KBr, ν , cm⁻¹): 3394, 3109, 3079, 2976, 2885, 1678, 1652, 1523, 1494, 1454, 1409, 1345, 1268, 1197, 1101, 1042, 1014, 970, 924, 897, 875, 858, 755, 697; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.91 (s, 1H,

OH), 8.26-8.22 (m, 2H, ArH), 7.76 (s, 1H, ArH), 7.54-7.47 (m, 3H, ArH), 7.30-7.27 (m, 2H, ArH), 7.08-6.96 (m, 5H, ArH), 2.74-2.67 (m, 2H, CH₂), 2.37-2.31 (m, 2H, CH₂), 2.04-2.00 (m, 2H, CH₂); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 193.5, 162.0, 161.4, 152.8, 146.9, 145.5, 143.1, 135.1, 132.5, 130.7, 129.8, 129.4, 128.6, 128.1, 125.1, 124.3, 123.9, 120.2, 116.5, 116.4, 110.8, 99.9, 38.5, 23.6, 23.1. HRMS calcd for C₂₉H₂₀N₂O₆ [M]⁺: 492.1321, found: 492.1335.

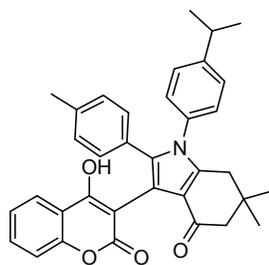
1-(4-chlorophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-2-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{1,2,3}): white powder (83%); m.p.:



172-174 °C; IR (KBr, ν, cm⁻¹): 3435, 3066, 2954, 2868, 1689, 1664, 1622, 1566, 1552, 1494, 1446, 1415, 1363, 1324, 1191, 1100, 1089, 1040, 1014, 958, 875, 759, 657; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.80 (s, 1H, OH), 7.76 (s, 1H, ArH), 7.51-7.43 (m, 3H, ArH), 7.26-7.21 (m, 4H, ArH), 6.88-6.84 (m, 4H, ArH), 2.58 (d, *J* = 14.4 Hz,

1H, CH₂), 2.46 (d, *J* = 12.8 Hz, 1H, CH₂), 2.27 (d, *J* = 14.8 Hz, 1H, CH₂), 2.14 (d, *J* = 15.6 Hz, 1H, CH₂), 2.04 (s, 3H, CH₃), 1.00 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.76 162.10, 161.20, 152.84, 144.19, 137.12, 136.45, 135.40, 133.17, 132.35, 130.09, 129.78, 129.69, 129.07, 128.16, 124.23, 123.89, 118.56, 116.45, 109.48, 100.27, 52.62, 36.72, 35.43, 28.98, 28.25, 21.08; HRMS (ESI) calcd for C₃₂H₂₆ClNNaO₄ [M+Na]⁺: 546.1448, found: 546.1443.

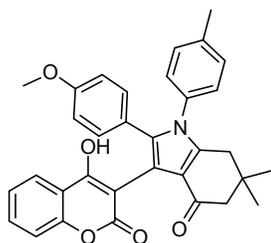
3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-1-(4-isopropylphenyl)-6,6-dimethyl-2-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{1,2,11}): white powder (90%); m.p.:



250-251 °C; IR (KBr, ν, cm⁻¹): 3426, 3080, 2956, 2867, 1680, 1659, 1610, 1565, 1548, 1502, 1449, 1418, 1386, 1364, 1200, 1172, 1102, 1042, 844, 754, 667; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.75 (s, 1H, OH), 7.77 (d, *J* = 6.8 Hz, 1H, ArH), 7.51 (t, *J* = 6.4 Hz, 3H, ArH), 7.29-7.14 (m, 6H, ArH), 6.87-6.80 (m, 4H, ArH),

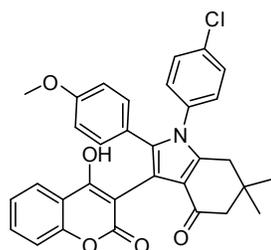
2.87-2.82 (m, 1H, CH), 2.55 (d, *J* = 16.4 Hz, 1H, CH₂), 2.47 (d, *J* = 11.6 Hz, 1H, CH₂), 2.27 (d, *J* = 15.6 Hz, 1H, CH₂), 2.15 (d, *J* = 15.6 Hz, 1H, CH₂), 2.02 (s, 3H, CH₃), 1.13-1.00 (m, 12H, 4×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.66 162.18, 161.05, 152.83, 148.59, 144.15, 136.81, 135.46, 135.24, 132.27, 129.65, 128.86, 128.50, 128.08, 127.52, 124.18, 123.88, 118.29, 116.51, 116.46, 109.17, 100.54, 52.69, 36.90, 35.40, 33.39, 28.93, 28.38, 24.11, 21.09; HRMS (ESI) calcd for C₃₅H₃₃NNaO₄ [M+Na]⁺: 554.2307, found: 554.2302.

3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-2-(4-methoxyphenyl)-6,6-dimethyl-1-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{1,3,I}):



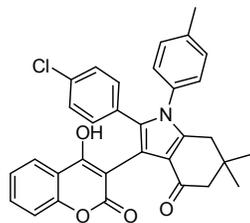
white powder (85%); m.p.: 264-265 °C; IR (KBr, ν , cm^{-1}): 3406, 3068, 2954, 2835, 1690, 1666, 1652, 1611, 1575, 1555, 1516, 1502, 1448, 1412, 1364, 1290, 1247, 1176, 1105, 1056, 1043, 1029, 962, 842, 760; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.75 (s, 1H, OH), 7.77 (d, $J = 6.8$ Hz, 1H, ArH), 7.53 (t, $J = 7.2$ Hz, 1H, ArH), 7.35-7.05 (m, 6H, ArH), 6.92 (d, $J = 7.6$ Hz, 2H, ArH), 6.63 (d, $J = 7.6$ Hz, 2H, ArH), 3.53 (s, 3H, OCH₃), 2.53 (d, $J = 16$ Hz, 1H, CH₂), 2.44 (d, $J = 14.4$ Hz, 1H, CH₂), 2.35-2.20 (m, 4H, 1 \times CH₂+1 \times CH₃), 2.14 (d, $J = 15.6$ Hz, 1H, CH₂), 1.03 (s, 3H, CH₃), 1.00 (s, 3H, CH₃); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.72, 162.21, 161.05, 158.70, 152.83, 144.04, 138.00, 135.37, 135.03, 132.32, 131.16, 130.20, 128.04, 124.24, 123.89, 123.61, 118.19, 116.51, 113.75, 108.85, 100.54, 55.23, 52.67, 36.90, 35.41, 28.95, 28.42, 21.10; HRMS (ESI) calcd for C₃₃H₂₉NNaO₅ [M+Na]⁺: 542.1943, found: 542.1963.

1-(4-chlorophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-2-(4-methoxyphenyl)-6,6-dimethyl-6,7-dihydro-1H-indol-4(5H)-one (4{1,3,3}):



white powder (92%); m.p.: 172-173 °C; IR (KBr, ν , cm^{-1}): 3436, 3068, 2956, 2870, 1715, 1672, 1626, 1610, 1574, 1447, 1412, 1363, 1247, 1191, 1174, 1103, 1090, 1028, 1015, 844, 754, 659; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.79 (s, 1H, OH), 7.76 (d, $J = 4.8$ Hz, 1H, ArH), 7.51-7.44 (m, 3H, ArH), 7.28-7.20 (m, 4H, ArH), 6.92-6.90 (m, 2H, ArH), 6.67-6.63 (m, 2H, ArH), 3.53 (s, 3H, OCH₃), 2.57 (d, $J = 15.6$ Hz, 1H, CH₂), 2.45 (d, $J = 9.6$ Hz, 1H, CH₂), 2.26 (d, $J = 15.6$ Hz, 1H, CH₂), 2.13 (d, $J = 15.2$ Hz, 1H, CH₂), 1.02 (s, 3H, CH₃), 1.00 (s, 3H, CH₃); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 193.08, 162.44, 161.47, 159.13, 153.14, 144.32, 136.77, 135.61, 133.45, 132.66, 131.48, 130.42, 130.07, 124.55, 124.20, 123.53, 118.77, 116.76, 114.19, 109.50, 100.59, 55.55, 52.92, 37.03, 35.73, 29.26, 28.59; HRMS (ESI) calcd for C₃₂H₂₆ClNNaO₅ [M+Na]⁺: 562.1397, found: 562.1418.

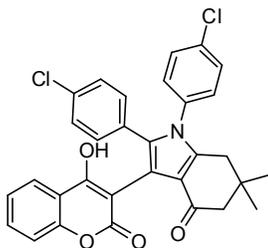
2-(4-chlorophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-1-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{1,4,I}):



white powder (89%); m.p.: > 300 °C; IR (KBr, ν , cm^{-1}): 3437, 3219, 3064, 3003, 2954, 2871, 1687, 1664, 1618, 1552, 1516, 1486, 1448, 1365, 1193, 1155, 1104, 1042, 1014, 844, 757, 648; ^1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 10.85 (s, 1H, OH), 7.76 (d, $J = 6.4$ Hz, 1H, ArH), 7.51 (t, $J = 7.2$ Hz, 1H, ArH), 7.29-6.96 (m, 10H, ArH), 2.55-2.44 (m, 2H, CH₂), 2.25-2.22 (m, 4H, 1 \times CH₂+1 \times CH₃), 2.15 (d, $J = 14.0$ Hz, 1H, CH₂), 1.01 (s, 3H, CH₃), 0.98 (s, 3H, CH₃); ^{13}C NMR

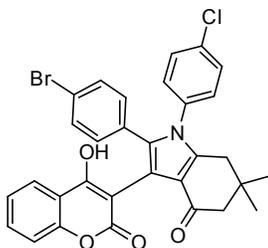
(DMSO-*d*₆, 75 MHz) δ (ppm) : 192.74, 162.12, 161.29, 152.86, 151.74, 144.61, 138.26, 134.68, 134.12, 132.50, 132.45, 131.46, 130.35, 128.47, 127.97, 124.27, 123.93, 118.35, 116.52, 116.46, 109.92, 100.04, 52.64, 36.81, 35.38, 28.89, 28.36, 21.09; HRMS (ESI) calcd for C₃₂H₂₆ClNNaO₄ [M+Na]⁺: 546.1448, found: 546.1468.

1,2-bis(4-chlorophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-6,7-dihydro-1H-indol-4(5H)-one (4{1,4,3}):



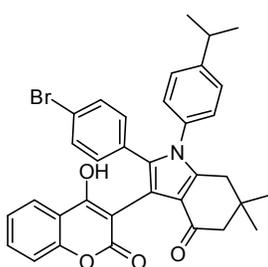
white powder (94%); m.p.: > 300 °C; IR (KBr, ν , cm⁻¹): 3448, 3080, 2956, 2866, 1684, 1669, 1611, 1546, 1529, 1492, 1446, 1433, 1410, 1396, 1366, 1257, 1239, 1224, 1195, 1103, 1089, 1013, 829, 752, 661; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.92 (s, 1H, OH), 7.80 (s, 1H, ArH), 7.57-7.50 (m, 3H, ArH), 7.32-7.19 (m, 6H, ArH), 7.00-6.97 (m, 2H, ArH), 2.62 (d, *J* = 17.6 Hz, 1H, CH₂), 2.50 (d, *J* = 16.0 Hz, 1H, CH₂), 2.30 (d, *J* = 16.4 Hz, 1H, CH₂), 2.17 (d, *J* = 17.6 Hz, 1H, CH₂), 1.05 (s, 3H, CH₃), 1.04 (s, 3H, CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 193.07, 162.30, 161.70, 153.15, 144.89, 136.39, 134.32, 133.65, 132.96, 132.80, 131.74, 130.33, 130.27, 130.22, 128.91, 124.60, 124.23, 118.88, 116.84, 116.68, 110.56, 100.05, 52.87, 36.92, 35.71, 29.19, 28.51; HRMS (ESI) calcd for C₃₁H₂₃Cl₂NNaO₄ [M+Na]⁺: 566.0902, found: 566.0896.

2-(4-bromophenyl)-1-(4-chlorophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-6,6-dimethyl-6,7-dihydro-1H-indol-4(5H)-one (4{1,5,3}):



white powder (92%); m.p.: > 300 °C; IR (KBr, ν , cm⁻¹): 3535, 1684, 1668, 1610, 1546, 1528, 1493, 1446, 1433, 1410, 1392, 1365, 1258, 1239, 1225, 1195, 1102, 1091, 1012, 846, 752, 669; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.84 (s, 1H, OH), 7.73 (s, 1H, ArH), 7.49-7.20 (m, 9H, ArH), 6.87-6.83 (m, 2H, ArH), 2.54 (d, *J* = 16.4 Hz, 1H, CH₂), 2.43 (d, *J* = 16.4 Hz, 1H, CH₂), 2.22 (d, *J* = 14.4 Hz, 1H, CH₂), 2.10 (d, *J* = 14.4 Hz, 1H, CH₂), 0.97 (s, 3H, CH₃), 0.95 (s, 3H, CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.73, 161.94, 161.35, 152.84, 151.64, 144.60, 136.07, 134.02, 133.33, 132.49, 131.69, 131.51, 130.33, 129.92, 124.29, 123.93, 121.36, 118.58, 116.53, 116.35, 110.23, 99.74, 52.55, 36.60, 35.39, 28.90, 28.18; HRMS (ESI) calcd for C₃₁H₂₃BrClNNaO₄ [M+Na]⁺: 610.0397, found: 610.0411.

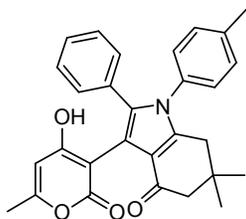
2-(4-bromophenyl)-3-(4-hydroxy-2-oxo-2H-chromen-3-yl)-1-(4-isopropylphenyl)-6,6-dimethyl-6,7-dihydro-1H-indol-4(5H)-one (4{1,5,11}):



white powder (93%); m.p.: > 300 °C; IR (KBr, ν , cm⁻¹): 3435, 3035, 2956, 2868, 1680, 1667, 1612, 1562, 1548, 1528, 1516, 1486, 1449, 1409, 1388, 1365, 1197, 1157, 1102, 1058, 1011, 848, 757, 659; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.87 (s, 1H, OH), 7.79-6.89 (m, 12H, ArH), 2.88-2.84 (m, 1H, CH), 2.58-2.47 (m, 2H, CH₂), 2.29 (d, *J* = 16.4 Hz,

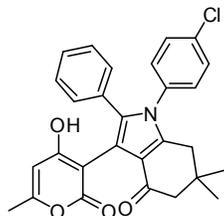
1H, CH₂), 2.17 (d, *J* = 14.8 Hz, 1H, CH₂), 1.15-0.95 (m, 12H, 4×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.69, 162.10, 161.27, 152.86, 148.85, 144.58, 134.88, 134.15, 132.41, 131.69, 131.29, 130.70, 128.03, 127.68, 124.25, 123.93, 121.17, 118.35, 116.45, 109.95, 100.03, 52.64, 36.80, 35.37, 33.40, 28.85, 28.33, 24.10;; HRMS (ESI) calcd for C₃₄H₃₀BrNNaO₄ [M+Na]⁺: 618.1256, found: 618.1269.

3-(4-hydroxy-6-methyl-2-oxo-2*H*-pyran-3-yl)-6,6-dimethyl-2-phenyl-1-(*p*-tolyl)-6,7-dihydro-



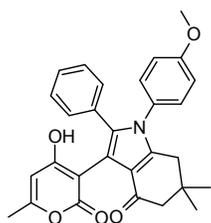
1*H*-indol-4(5*H*)-one (4{2,*I*,*I*)}: light red powder (81%); m.p.: 268-270 °C; IR (KBr, ν, cm⁻¹): 3427, 3063, 3035, 2938, 2867, 2735, 2691, 1662, 1637, 1576, 1515, 1492, 1447, 1404, 1367, 1279, 1233, 1209, 1107, 1125, 1081, 1050, 1000, 934, 923, 828, 804, 766, 726, 699, 650; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.64 (s, 1H, OH), 7.17-6.97 (m, 9H, ArH), 5.91 (s, 1H, =CH), 2.50-2.45 (m, 2H, CH₂), 2.25-2.18 (m, 5H, 1×CH₂+1×CH₃), 2.10 (s, 3H, CH₃), 1.00 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.46, 166.34, 164.14, 160.89, 143.64, 137.88, 134.96, 134.27, 131.88, 130.18, 129.67, 128.19, 127.91, 127.33, 118.14, 110.74, 100.51, 97.07, 52.75, 36.89, 35.19, 28.67, 28.59, 21.05, 19.75; HRMS calcd for C₂₉H₂₇NO₄ [M]⁺: 453.1940, found: 453.1929.

1-(4-chlorophenyl)-3-(4-hydroxy-6-methyl-2-oxo-2*H*-pyran-3-yl)-6,6-dimethyl-2-phenyl-6,7-



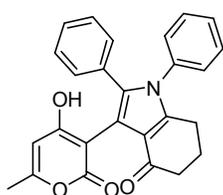
dihydro-1*H*-indol-4(5*H*)-one (4{2,*I*,3}): white powder (88%); m.p.: 282-283 °C; IR (KBr, ν, cm⁻¹): 3430, 3089, 3062, 2938, 2867, 1691, 1633, 1525, 1494, 1398, 1364, 1274, 1231, 1206, 1171, 1125, 1078, 1054, 1002, 974, 937, 843, 828, 765, 699; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.77 (s, 1H, OH), 7.42 (d, *J* = 7.6 Hz, 2H, ArH), 7.21-7.17 (m, 2H, ArH), 7.10-7.08 (m, 3H, ArH), 6.95 (d, *J* = 5.6 Hz, 2H, ArH), 5.91 (s, 1H, =CH), 2.52-2.44 (m, 2H, CH₂), 2.25-2.16 (m, 2H, CH₂), 2.09 (s, 3H, CH₃), 0.99 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.63, 166.46, 164.15, 161.04, 143.70, 136.38, 135.37, 134.25, 133.03, 131.51, 130.00, 129.72, 128.33, 127.57, 118.41, 111.04, 100.50, 96.84, 52.70, 36.74, 35.22, 28.62, 28.52, 19.74; HRMS calcd for C₂₈H₂₃ClNO₄ [M-H]⁺: 472.1316, found: 472.1319.

3-(4-hydroxy-6-methyl-2-oxo-2*H*-pyran-3-yl)-1-(4-methoxyphenyl)-6,6-dimethyl-2-phenyl-6,



7-dihydro-1*H*-indol-4(5*H*)-one (4{2,*I*,6}): white powder (85%); m.p.: 142-144 °C; IR (KBr, ν, cm⁻¹): 3422, 3058, 2957, 2870, 2733, 2682, 1662, 1578, 1514, 1491, 1448, 1367, 1298, 1251, 1210, 1169, 1119, 1079, 1029, 993, 924, 768, 735, 720, 700, 648; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.67 (s, 1H, OH), 7.09-7.04 (m, 5H, ArH), 6.93 (dd, *J* = 28.6, 7.4 Hz 4H,

ArH), 5.90 (s, 1H, =CH), 3.68 (s, 3H, OCH₃), 2.47 (d, *J* = 9.6 Hz, 2H, CH₂), 2.19 (q, 2H, CH₂), 2.08 (s, 3H, CH₃), 0.98 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.48, 166.34, 164.19, 160.88, 158.96, 143.87, 134.46, 131.88, 130.19, 129.70, 129.35, 128.16, 127.33, 117.99, 114.75, 110.48, 100.52, 97.14, 55.71, 52.76, 36.85, 35.16, 28.68, 28.59, 19.73; HRMS calcd for C₂₉H₂₇NO₅[M]⁺: 469.1889, found: 469.1894.

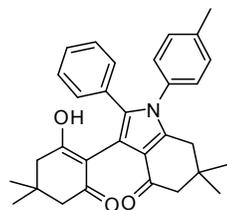


3-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-1,2-diphenyl-6,7-dihydro-1H-indol-4(5H)-one (4{2,I,12}): white powder (80%); m.p.: 279-281 °C;

IR (KBr, ν , cm⁻¹): 3425, 3003, 2974, 2865, 1649, 1494, 1458, 1399, 1367, 1210, 1130, 1044, 935, 837, 799, 771, 710, 649; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.68 (s, 1H, OH), 7.36-7.31 (m, 2H, ArH), 7.16-7.05 (m, 5H, ArH), 6.95-6.91 (m, 2H, ArH), 5.89 (s, 1H, =CH), 2.63-2.59 (m, 2H, CH₂), 2.31-2.28 (m, 2H, CH₂), 2.09 (s, 3H, CH₃), 1.99-1.95 (m, 2H, CH₂); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) :

193.06, 166.32, 164.23, 160.94, 144.85, 137.48, 134.12, 131.74, 129.70, 129.64, 128.47, 128.22, 128.13, 127.45, 119.36, 111.04, 100.50, 97.08, 38.71, 23.59, 23.08, 19.75; HRMS calcd for C₂₆H₂₀NO₄[M-H]⁺: 410.1392, found: 410.1424.

3-(2-hydroxy-4,4-dimethyl-6-oxocyclohex-1-en-1-yl)-6,6-dimethyl-2-phenyl-1-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{3,I,I}): white powder (80%); m.p.:



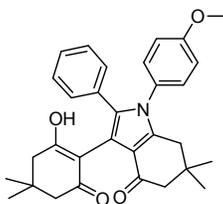
196-198 °C; IR (KBr, ν , cm⁻¹): 3426, 3058, 2957, 2866, 1668, 1573,

1515, 1490, 1452, 1408, 1367, 1198, 1116, 1046, 1013, 922, 803, 773,

699; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 9.72 (s, 1H, OH), 7.14-6.91 (m, 9H, ArH), 2.47-2.07 (m, 11H, 1×CH₃+4×CH₂), 1.09-0.87

(m, 12H, 4×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 194.60, 192.48, 163.88, 143.45, 137.67, 135.21, 133.79, 132.11, 130.10, 129.68, 127.92, 127.04, 124.85, 118.44, 112.42, 109.61, 52.83, 52.63, 36.98, 36.69, 36.63, 35.18, 32.06, 28.78, 28.70, 28.25, 21.05; HRMS calcd for C₃₁H₃₃NO₃[M]⁺: 467.2460, found: 467.2446.

3-(2-hydroxy-4,4-dimethyl-6-oxocyclohex-1-en-1-yl)-1-(4-methoxyphenyl)-6,6-dimethyl-2-phenyl-6,7-dihydro-1H-indol-4(5H)-one (4{3,I,6}): white powder (82%); m.p.:



183-184 °C; IR (KBr, ν , cm⁻¹): 3588, 3425, 3056, 3030, 2957, 2892,

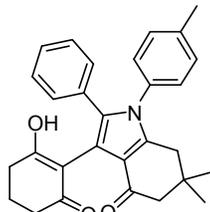
2644, 1670, 1585, 1562, 1513, 1486, 1451, 1367, 1299, 1257, 1211, 1166,

1148, 1046, 1028, 1013, 921, 845, 807, 775, 718, 702; ¹H NMR

(DMSO-*d*₆, 400 MHz) δ (ppm) : 9.63 (s, 1H, OH), 7.08-7.03 (m, 5H, ArH), 6.92-6.89 (m, 4H, ArH), 3.71 (s, 3H, OCH₃), 2.46-1.96 (m, 8H, 4×CH₂), 1.09-0.87 (m, 12H,

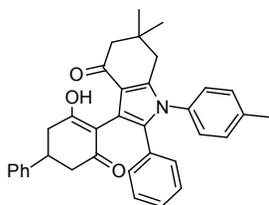
4×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 196.51, 192.26, 170.40, 158.83, 143.57, 133.94, 132.17, 130.50, 129.71, 129.32, 127.92, 126.99, 118.32, 114.69, 112.19, 109.66, 56.49, 55.72, 52.88, 36.93, 35.17, 32.06, 28.77, 28.29, 19.02; HRMS calcd for C₃₁H₃₃NO₄ [M]⁺: 483.2410, found: 483.2398.

3-(2-hydroxy-6-oxocyclohex-1-en-1-yl)-6,6-dimethyl-2-phenyl-1-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{4,I,I}):



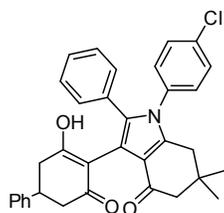
ol-4(5H)-one (4{4,I,I}): white powder (75%); m.p.: 174-175 °C; IR (KBr, ν, cm⁻¹): 3431, 3059, 3033, 2952, 2875, 2620, 1664, 1578, 1515, 1491, 1448, 1422, 1381, 1334, 1280, 1209, 1183, 1160, 1117, 1071, 1049, 985, 923, 833, 811, 769, 730, 697, 649; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 9.76 (s, 1H, OH), 7.15-6.90 (m, 9H, ArH), 2.47-1.70 (m, 13H, 1×CH₃+5×CH₂), 0.98 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.36, 143.35, 137.63, 135.27, 133.55, 132.12, 130.10, 129.61, 129.53, 128.07, 127.95, 127.89, 126.92, 118.40, 112.67, 110.82, 52.88, 36.98, 36.95, 35.16, 28.67, 21.04, 20.91, 19.01.; HRMS calcd for C₂₉H₂₈NO₃ [M-H]⁺: 438.2069, found: 438.2073.

3-(5-hydroxy-3-oxo-1,2,3,6-tetrahydro-[1,1'-biphenyl]-4-yl)-6,6-dimethyl-2-phenyl-1-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{5,I,I}):



-6,7-dihydro-1H-indol-4(5H)-one (4{5,I,I}): grey powder (77%); m.p.: 137-138 °C; IR (KBr, ν, cm⁻¹): 3425, 3059, 3029, 3002, 2956, 2935, 2895, 1579, 1515, 1489, 1447, 1386, 1368, 1284, 1116, 1045, 1015, 924, 836, 764, 699, 649; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.06 (s, 1H, OH), 7.25-6.97 (m, 14H, ArH), 3.30-3.10 (m, 1H, CH), 2.63-2.03 (m, 11H, 1×CH₃+4×CH₂), 0.99 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 192.54, 192.42, 144.12, 143.97, 143.49, 143.43, 137.67, 135.29, 133.89, 133.65, 132.23, 132.10, 130.13, 129.69, 129.62, 129.03, 128.97, 128.06, 127.93, 127.32, 118.60, 118.32, 112.51, 112.45, 110.76, 110.67, 52.96, 52.89, 38.93, 38.55, 37.05, 35.19, 28.72, 28.44, 21.08. HRMS calcd for C₃₅H₃₃NO₃ [M]⁺: 515.2460, found: 515.2455.

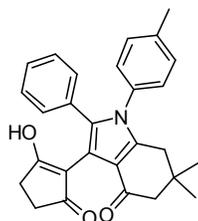
1-(4-chlorophenyl)-3-(5-hydroxy-3-oxo-1,2,3,6-tetrahydro-[1,1'-biphenyl]-4-yl)-6,6-dimethyl-2-phenyl-6,7-dihydro-1H-indol-4(5H)-one (4{5,I,3}):



2-phenyl-6,7-dihydro-1H-indol-4(5H)-one (4{5,I,3}): white powder (88%); m.p.: 154-156 °C; IR (KBr, ν, cm⁻¹): 3432, 3089, 3028, 2955, 2892, 1049, 1620, 1578, 1448, 1417, 1383, 1363, 1244, 1116, 1091, 1069, 1050, 1014, 923, 888, 843, 760, 724, 649; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 10.04 (s, 1H, OH), 7.04-6.93 (m, 14H, ArH), 3.35-3.10 (m, 1H, CH), 2.61-2.19 (m, 8H, 4×CH₂), 0.99 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) :

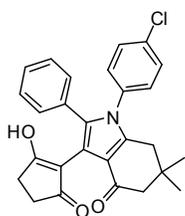
192.57, 192.44, 144.09, 143.95, 143.47, 143.42, 136.71, 133.81, 133.56, 132.83, 131.86, 131.74, 130.00, 129.70, 129.65, 129.02, 128.96, 128.22, 128.19, 127.33, 127.15, 118.59, 112.84, 112.79, 110.51, 110.42, 52.90, 52.84, 38.87, 38.53, 36.86, 35.22; 31.14, 28.67; HRMS calcd for $C_{34}H_{30}ClNO_3$ $[M]^+$: 535.1914, found: 535.1907.

3-(2-hydroxy-5-oxocyclopent-1-en-1-yl)-6,6-dimethyl-2-phenyl-1-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{6,I,I}):



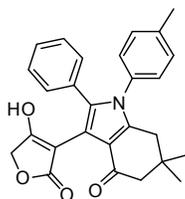
white powder (84%); m.p.: 293-295 °C; IR (KBr, ν , cm^{-1}): 3427, 3031, 2955, 2932, 2869, 2590, 1665, 1576, 1515, 1487, 1410, 1313, 1208, 1116, 1045, 1018, 924, 840, 802, 771, 721, 699, 665, 649; 1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 11.36 (s, 1H, OH), 7.15-6.95 (m, 9H, ArH), 2.47-1.93 (m, 11H, $1 \times CH_3 + 4 \times CH_2$), 0.97 (s, 6H, $2 \times CH_3$); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.42, 143.61, 137.88, 135.07, 134.15, 133.95, 132.03, 131.87, 130.21, 129.73, 128.08, 127.92, 127.18, 118.32, 112.52, 110.16, 52.88, 36.95, 36.87, 35.12, 28.66, 28.58, 21.06; HRMS calcd for $C_{28}H_{26}NO_3$ $[M-H]^+$: 424.1913, found: 424.1940.

1-(4-chlorophenyl)-3-(2-hydroxy-5-oxocyclopent-1-en-1-yl)-6,6-dimethyl-2-phenyl-6,7-dihydro-1H-indol-4(5H)-one (4{6,I,3}):



grey powder (89%); m.p.: 290-292 °C; IR (KBr, ν , cm^{-1}): 3422, 3058, 2957, 2935, 2870, 2838, 2733, 2682, 1578, 1514, 1496, 1448, 1367, 1298, 1251, 1210, 1169, 1119, 1079, 1029, 993, 924, 768, 735, 720, 700, 648; 1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 11.36 (s, 1H, OH), 7.42 (d, $J = 7.6$ Hz, 2H, ArH), 7.17-7.08 (m, 5H, ArH), 6.95 (d, $J = 6.0$ Hz, 2H, ArH), 2.50-2.18 (m, 8H, $4 \times CH_2$), 0.98 (s, 6H, $2 \times CH_3$); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.44, 143.58, 136.47, 133.79, 132.97, 131.50, 130.15, 130.00, 129.86, 129.74, 128.23, 127.37, 118.54, 114.67, 112.24, 110.49, 52.80, 36.74, 35.15, 31.15, 28.59, 19.01; HRMS calcd for $C_{27}H_{23}ClNO_3$ $[M-H]^+$: 444.1366, found: 444.1366.

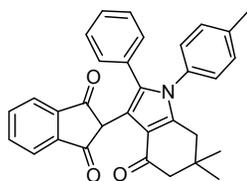
3-(4-hydroxy-2-oxo-2,5-dihydrofuran-3-yl)-6,6-dimethyl-2-phenyl-1-(p-tolyl)-6,7-dihydro-1H-indol-4(5H)-one (4{7,I,I}):



grey powder (76%); m.p.: 142-144 °C; IR (KBr, ν , cm^{-1}): 3427, 3001, 2958, 2869, 1755, 1663, 1578, 1515, 1490, 1423, 1281, 1117, 1045, 1014, 924, 831, 768, 728, 698, 649; 1H NMR (DMSO- d_6 , 400 MHz) δ (ppm) : 11.82 (s, 1H, OH), 7.14-7.03 (m, 9H, ArH), 4.57-4.00 (m, 2H, CH_2), 2.49-2.23 (m, 7H, $CH_3 + 2 \times CH_2$), 0.98 (s, 6H, $2 \times CH_3$); ^{13}C NMR (DMSO- d_6 , 75 MHz) δ (ppm) : 192.58, 174.58, 173.84, 143.84, 138.07, 134.88, 134.81, 131.43, 130.25, 129.89, 128.25, 127.92, 127.52, 118.34, 107.80, 95.79, 67.08, 52.73, 36.86, 35.17, 28.63,

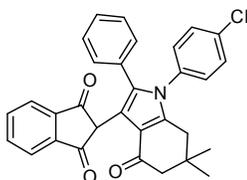
28.41, 21.05; HRMS calcd for C₂₇H₂₄NO₄ [M-H]⁺: 426.1705, found: 426.1714.

2-(6,6-dimethyl-4-oxo-2-phenyl-1-(p-tolyl)-4,5,6,7-tetrahydro-1H-indol-3-yl)-1H-indene-1,3(2



H)-dione (4{8,1,1}): yellow powder (82%); m.p.: 239-240 °C; IR (KBr, ν , cm⁻¹): 3433, 3001, 2955, 2869, 1751, 1651, 1516, 1490, 1465, 1445, 1417, 1367, 1321, 1246, 1206, 1154, 1114, 1040, 1014, 924, 886, 843, 778, 764, 746, 701, 649; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 7.88 (s, 4H, ArH), 7.20-7.15 (m, 7H, ArH), 7.07 (d, *J* = 7.6 Hz, 2H, ArH), 4.45 (s, 1H, CH), 2.47 (s, 2H, CH₂), 2.25 (s, 3H, CH₃), 2.05 (s, 2H, CH₂), 0.90 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 198.95, 193.07, 144.01, 142.27, 138.21, 137.54, 135.54, 134.56, 130.50, 130.27, 129.98, 128.83, 128.26, 127.87, 122.98, 116.47, 110.78, 53.86, 51.35, 36.46, 35.53, 28.47, 21.04; HRMS calcd for C₃₂H₂₆NO₃ [M-H]⁺: 472.1913, found: 472.1923.

2-(1-(4-chlorophenyl)-6,6-dimethyl-4-oxo-2-phenyl-4,5,6,7-tetrahydro-1H-indol-3-yl)-1H-ind



ene-1,3(2H)-dione (4{8,1,3}): ocher powder (85%); m.p.: 224-226 °C; IR (KBr, ν , cm⁻¹): 3428, 3001, 2979, 2935, 1718, 1654, 1573, 1494, 1421, 1247, 1114, 1043, 1013, 923, 846, 649; ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm) : 7.90 (s, 4H, ArH), 7.44 (d, *J* = 7.6 Hz, 2H, ArH), 7.26-7.19 (m, 7H, ArH), 4.50 (s, 1H, CH), 2.53-2.46 (m, 2H, CH₂), 2.08-1.94 (m, 2H, CH₂), 0.92 (s, 6H, 2×CH₃); ¹³C NMR (DMSO-*d*₆, 75 MHz) δ (ppm) : 198.88, 193.52, 144.01, 142.30, 137.46, 136.71, 136.04, 135.57, 133.32, 130.57, 130.01, 129.81, 128.96, 128.48, 122.98, 116.76, 111.16, 53.79, 51.35, 36.30, 36.25, 35.56, 28.44; HRMS calcd for C₃₁H₂₃ClNO₃ [M-H]⁺: 492.1366, found: 492.1373.

Crystal structures of compound

Crystal data for 4{I,I,I}:

C₃₂H₂₇NO₄; M = 489.55, colorless block crystals, 0.30 × 0.28 × 0.25 mm, Triclinic, space group P1, a = 10.9476(13) Å, b = 11.9290(15) Å, c = 13.8496(19) Å, α = 107.328(2)°, β = 93.9780(10)°, γ = 104.690(2)°, V = 1649.5(4) Å³, Z = 2, D_c = 0.986 g·cm⁻³, F(000) = 516, μ (MoKα) = 0.065 mm⁻¹. Intensity data were collected on a diffractometer with graphite monochromated MoKα radiation (λ = 0.71073 Å) using ω scan mode with 2.00° < θ < 25.02°. 10582 unique reflections were measured and 4017 reflections with I > 2σ(I) were used in the refinement. The structure was solved by direct methods and expanded using Fourier techniques. The final cycle of full-matrix least squares technique to R = 0.0699 and wR = 0.1369.

Table 1. Bond lengths (Å) for 4{I,I,I}

Bond	Bond Lengths	Bond	Bond Lengths	Bond	Bond Lengths
N(1)-C(19)	1.387(8)	C(10)-C(11)	1.428(9)	C(36)-C(37)	1.556(10)
N(1)-C(1)	1.399(8)	C(11)-C(12)	1.316(9)	C(37)-C(38)	1.522(9)
N(1)-C(20)	1.414(9)	C(12)-C(13)	1.443(9)	C(38)-C(42)	1.472(10)
N(2)-C(40)	1.359(8)	C(13)-C(14)	1.340(10)	C(38)-C(41)	1.541(9)
N(2)-C(33)	1.385(8)	C(13)-C(18)	1.385(9)	C(38)-C(39)	1.570(10)
N(2)-C(52)	1.433(10)	C(14)-C(15)	1.429(10)	C(39)-C(40)	1.484(9)
O(1)-C(10)	1.377(8)	C(15)-C(16)	1.392(12)	C(43)-C(44)	1.471(10)
O(1)-C(14)	1.395(9)	C(16)-C(17)	1.335(11)	C(44)-C(45)	1.343(9)
O(2)-C(10)	1.221(8)	C(17)-C(18)	1.305(11)	C(45)-C(46)	1.446(9)
O(3)-C(12)	1.330(8)	C(20)-C(25)	1.316(10)	C(46)-C(51)	1.330(9)
O(4)-C(4)	1.241(8)	C(20)-C(21)	1.377(10)	C(46)-C(47)	1.395(10)
O(5)-C(47)	1.325(10)	C(21)-C(22)	1.410(10)	C(47)-C(48)	1.427(10)
O(5)-C(43)	1.409(9)	C(22)-C(23)	1.277(13)	C(48)-C(49)	1.395(11)
O(6)-C(43)	1.177(9)	C(23)-C(24)	1.379(14)	C(49)-C(50)	1.348(11)
O(7)-C(45)	1.334(8)	C(23)-C(32)	1.574(15)	C(50)-C(51)	1.378(10)
O(8)-C(36)	1.209(8)	C(24)-C(25)	1.420(12)	C(52)-C(53)	1.341(10)
C(1)-C(2)	1.345(8)	C(26)-C(31)	1.346(9)	C(52)-C(57)	1.391(11)
C(1)-C(26)	1.467(9)	C(26)-C(27)	1.421(10)	C(53)-C(54)	1.389(11)
C(2)-C(3)	1.403(9)	C(27)-C(28)	1.405(11)	C(54)-C(55)	1.370(13)
C(2)-C(11)	1.491(9)	C(28)-C(29)	1.350(10)	C(55)-C(56)	1.318(13)
C(3)-C(19)	1.352(8)	C(29)-C(30)	1.403(11)	C(55)-C(58)	1.580(14)

C(3)-C(4)	1.464(9)	C(30)-C(31)	1.321(10)	C(56)-C(57)	1.384(13)
C(4)-C(5)	1.474(9)	C(33)-C(34)	1.386(9)	C(59)-C(64)	1.319(9)
C(5)-C(6)	1.559(9)	C(33)-C(59)	1.500(10)	C(59)-C(60)	1.401(9)
C(6)-C(7)	1.481(9)	C(34)-C(35)	1.414(9)	C(60)-C(61)	1.445(11)
C(6)-C(8)	1.542(10)	C(34)-C(44)	1.441(9)	C(61)-C(62)	1.242(11)
C(6)-C(9)	1.557(9)	C(35)-C(40)	1.393(8)	C(62)-C(63)	1.359(11)
C(7)-C(19)	1.480(9)	C(35)-C(36)	1.447(10)	C(63)-C(64)	1.448(12)

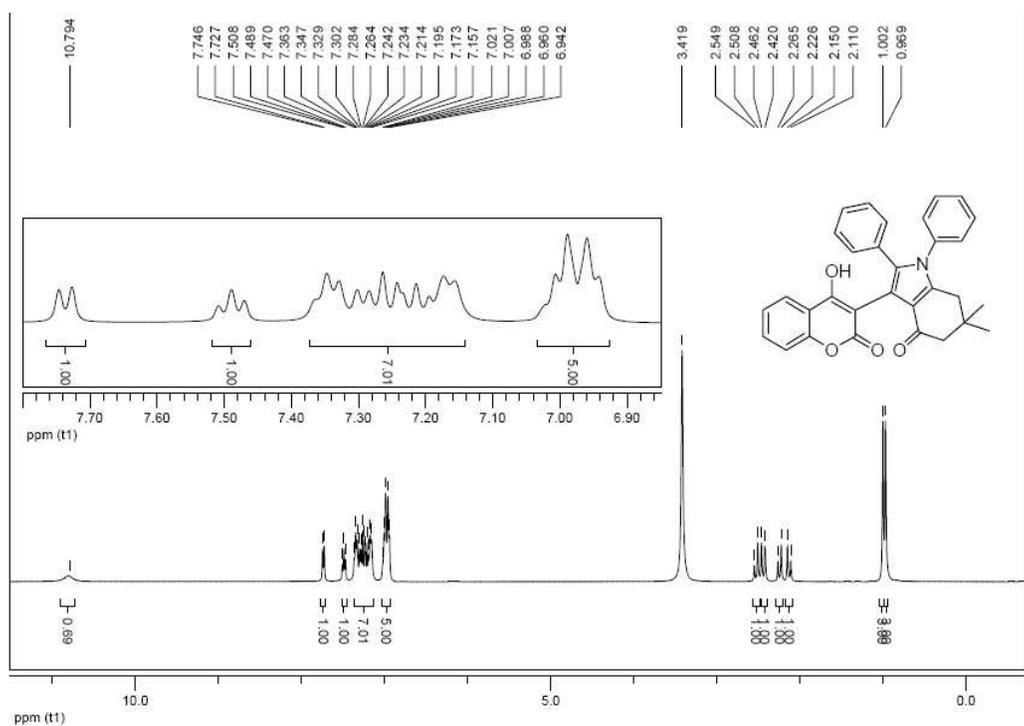
Table 2. Bond angles ($^{\circ}$) for **4{I,I,I}**

Angles	($^{\circ}$)	Angles	($^{\circ}$)
C(19)-N(1)-C(1)	107.8(6)	C(29)-C(28)-C(27)	119.5(10)
C(19)-N(1)-C(20)	123.5(7)	C(28)-C(29)-C(30)	120.8(10)
C(1)-N(1)-C(20)	128.6(7)	C(31)-C(30)-C(29)	118.3(9)
C(40)-N(2)-C(33)	109.7(7)	C(30)-C(31)-C(26)	125.1(9)
C(40)-N(2)-C(52)	124.5(6)	N(2)-C(33)-C(34)	108.2(7)
C(33)-N(2)-C(52)	125.6(7)	N(2)-C(33)-C(59)	123.0(7)
C(10)-O(1)-C(14)	118.2(7)	C(34)-C(33)-C(59)	128.8(7)
C(47)-O(5)-C(43)	121.6(7)	C(33)-C(34)-C(35)	106.4(7)
C(2)-C(1)-N(1)	108.7(7)	C(33)-C(34)-C(44)	125.5(7)
C(2)-C(1)-C(26)	130.7(7)	C(35)-C(34)-C(44)	128.1(8)
N(1)-C(1)-C(26)	120.6(7)	C(40)-C(35)-C(34)	108.3(7)
C(1)-C(2)-C(3)	106.7(7)	C(40)-C(35)-C(36)	120.5(7)
C(1)-C(2)-C(11)	127.3(7)	C(34)-C(35)-C(36)	131.1(8)
C(3)-C(2)-C(11)	126.0(7)	O(8)-C(36)-C(35)	123.7(7)
C(19)-C(3)-C(2)	109.9(7)	O(8)-C(36)-C(37)	121.9(9)
C(19)-C(3)-C(4)	118.6(7)	C(35)-C(36)-C(37)	114.4(8)
C(2)-C(3)-C(4)	131.4(7)	C(38)-C(37)-C(36)	115.6(7)
O(4)-C(4)-C(3)	121.8(7)	C(42)-C(38)-C(37)	113.5(7)
O(4)-C(4)-C(5)	121.5(8)	C(42)-C(38)-C(41)	110.7(7)
C(3)-C(4)-C(5)	116.7(8)	C(37)-C(38)-C(41)	108.9(7)
C(4)-C(5)-C(6)	114.9(7)	C(42)-C(38)-C(39)	108.4(7)
C(7)-C(6)-C(8)	111.8(7)	C(37)-C(38)-C(39)	108.0(6)
C(7)-C(6)-C(9)	109.3(7)	C(41)-C(38)-C(39)	107.0(7)
C(8)-C(6)-C(9)	108.4(6)	C(40)-C(39)-C(38)	110.3(6)

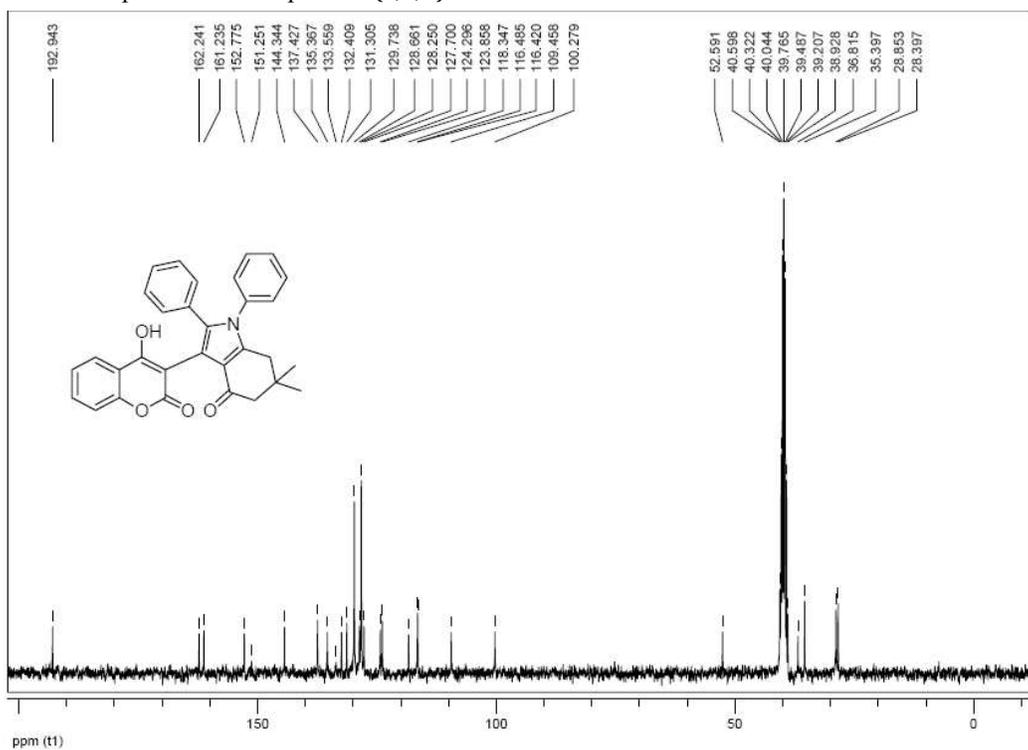
C(7)-C(6)-C(5)	108.6(6)	N(2)-C(40)-C(35)	107.4(7)
C(8)-C(6)-C(5)	109.1(6)	N(2)-C(40)-C(39)	126.1(7)
C(9)-C(6)-C(5)	109.7(6)	C(35)-C(40)-C(39)	126.5(8)
C(19)-C(7)-C(6)	112.2(6)	O(6)-C(43)-O(5)	117.5(8)
O(2)-C(10)-O(1)	113.8(8)	O(6)-C(43)-C(44)	126.2(9)
O(2)-C(10)-C(11)	126.6(8)	O(5)-C(43)-C(44)	116.3(8)
O(1)-C(10)-C(11)	119.7(6)	C(45)-C(44)-C(34)	123.4(7)
C(12)-C(11)-C(10)	119.4(8)	C(45)-C(44)-C(43)	119.9(8)
C(12)-C(11)-C(2)	125.1(8)	C(34)-C(44)-C(43)	116.5(8)
C(10)-C(11)-C(2)	115.5(7)	O(7)-C(45)-C(44)	124.7(7)
C(11)-C(12)-O(3)	124.2(8)	O(7)-C(45)-C(46)	112.7(8)
C(11)-C(12)-C(13)	122.7(8)	C(44)-C(45)-C(46)	122.6(8)
O(3)-C(12)-C(13)	113.0(8)	C(51)-C(46)-C(47)	118.9(8)
C(14)-C(13)-C(18)	118.5(9)	C(51)-C(46)-C(45)	126.0(9)
C(14)-C(13)-C(12)	116.1(8)	C(47)-C(46)-C(45)	115.0(8)
C(18)-C(13)-C(12)	125.4(10)	O(5)-C(47)-C(46)	124.6(8)
C(13)-C(14)-O(1)	124.0(8)	O(5)-C(47)-C(48)	116.0(9)
C(13)-C(14)-C(15)	122.9(8)	C(46)-C(47)-C(48)	119.5(9)
O(1)-C(14)-C(15)	113.2(10)	C(49)-C(48)-C(47)	118.9(9)
C(16)-C(15)-C(14)	113.6(10)	C(50)-C(49)-C(48)	119.1(9)
C(17)-C(16)-C(15)	122.5(11)	C(49)-C(50)-C(51)	121.3(10)
C(18)-C(17)-C(16)	122.1(10)	C(46)-C(51)-C(50)	122.2(9)
C(17)-C(18)-C(13)	120.5(10)	C(53)-C(52)-C(57)	116.0(9)
C(3)-C(19)-N(1)	106.8(6)	C(53)-C(52)-N(2)	123.4(10)
C(3)-C(19)-C(7)	126.3(8)	C(57)-C(52)-N(2)	120.6(10)
N(1)-C(19)-C(7)	126.8(7)	C(52)-C(53)-C(54)	123.7(10)
C(25)-C(20)-C(21)	119.4(9)	C(55)-C(54)-C(53)	115.8(11)
C(25)-C(20)-N(1)	121.6(10)	C(56)-C(55)-C(54)	124.6(13)
C(21)-C(20)-N(1)	118.9(9)	C(56)-C(55)-C(58)	115.1(15)
C(20)-C(21)-C(22)	118.0(9)	C(54)-C(55)-C(58)	120.0(14)
C(23)-C(22)-C(21)	124.7(12)	C(55)-C(56)-C(57)	116.8(13)
C(22)-C(23)-C(24)	116.7(12)	C(56)-C(57)-C(52)	122.7(11)
C(22)-C(23)-C(32)	123.8(15)	C(64)-C(59)-C(60)	120.3(9)
C(24)-C(23)-C(32)	119.5(14)	C(64)-C(59)-C(33)	119.3(8)
C(23)-C(24)-C(25)	121.2(11)	C(60)-C(59)-C(33)	120.4(9)

C(20)-C(25)-C(24)	119.8(11)	C(59)-C(60)-C(61)	118.0(9)
C(31)-C(26)-C(27)	117.2(8)	C(62)-C(61)-C(60)	120.1(11)
C(31)-C(26)-C(1)	125.1(9)	C(61)-C(62)-C(63)	124.2(14)
C(27)-C(26)-C(1)	117.7(8)	C(62)-C(63)-C(64)	117.8(11)
C(28)-C(27)-C(26)	119.1(8)	C(59)-C(64)-C(63)	119.4(10)

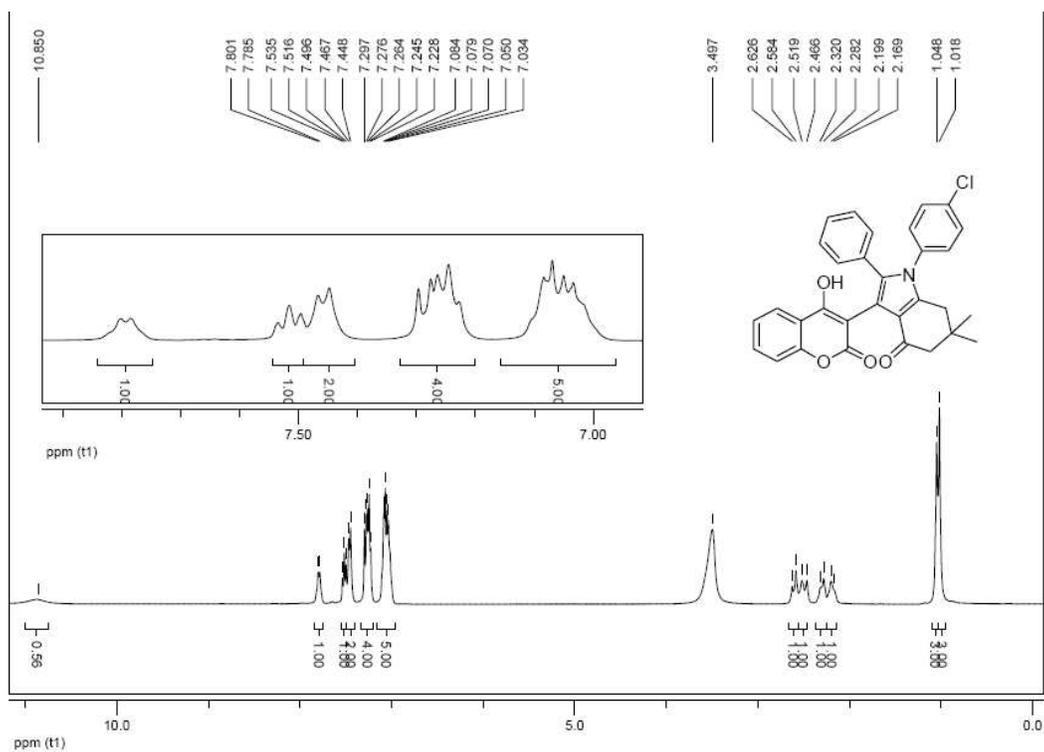
¹H NMR spectrum of compound 4{1,1,2}



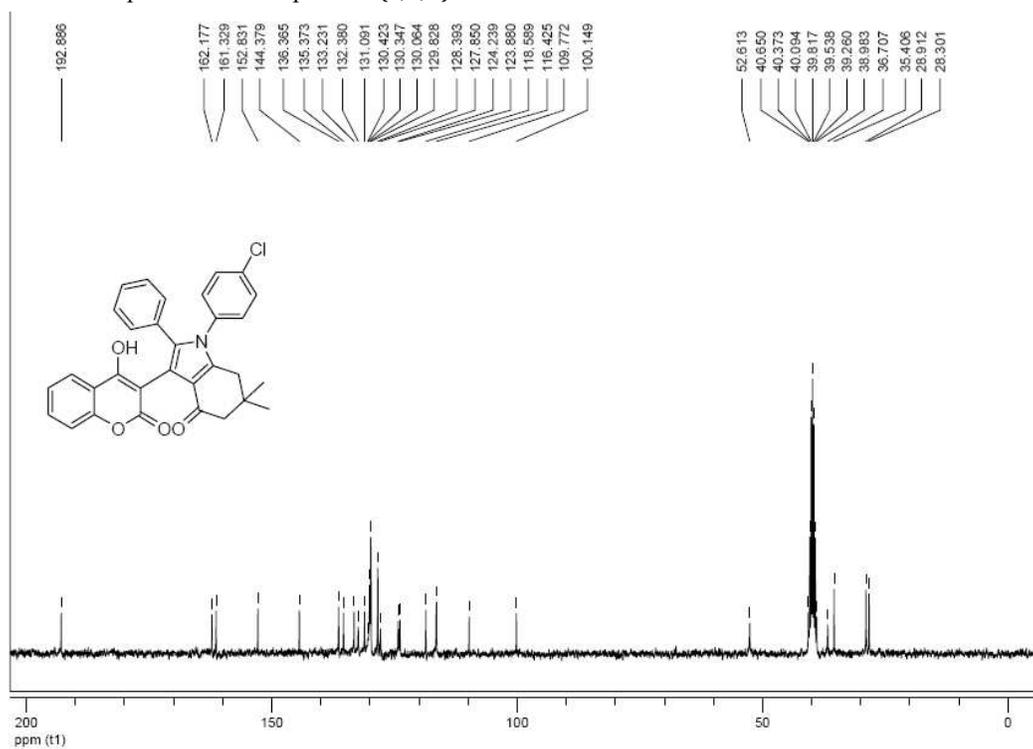
¹³C NMR spectrum of compound 4{1,1,2}



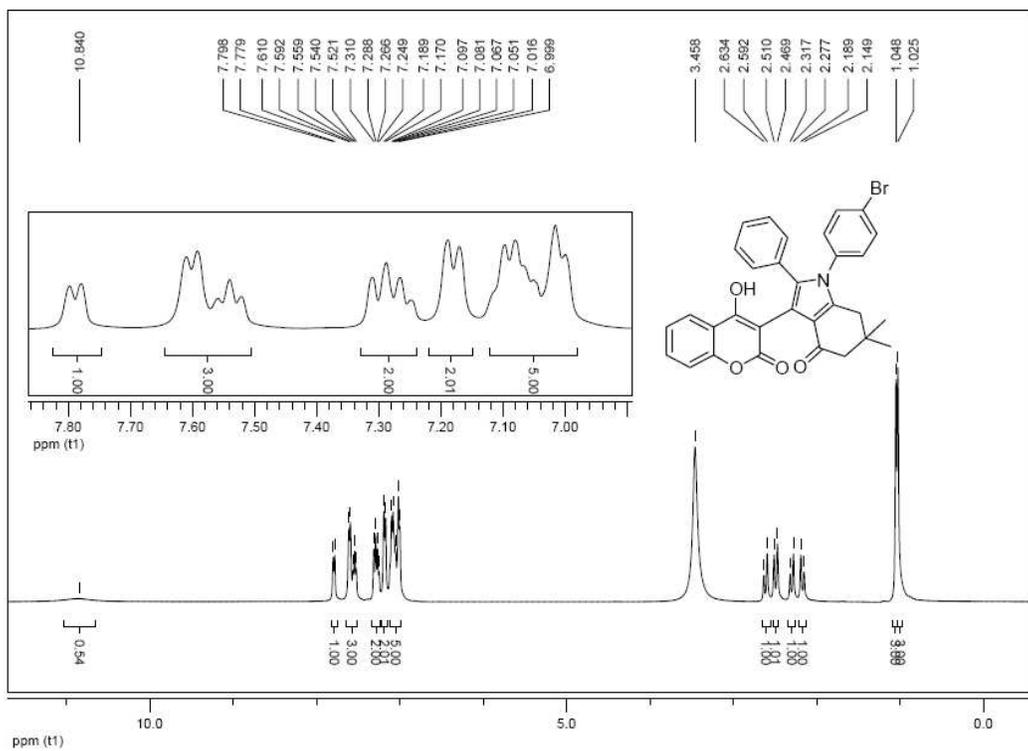
¹H NMR spectrum of compound **4{1,1,3}**



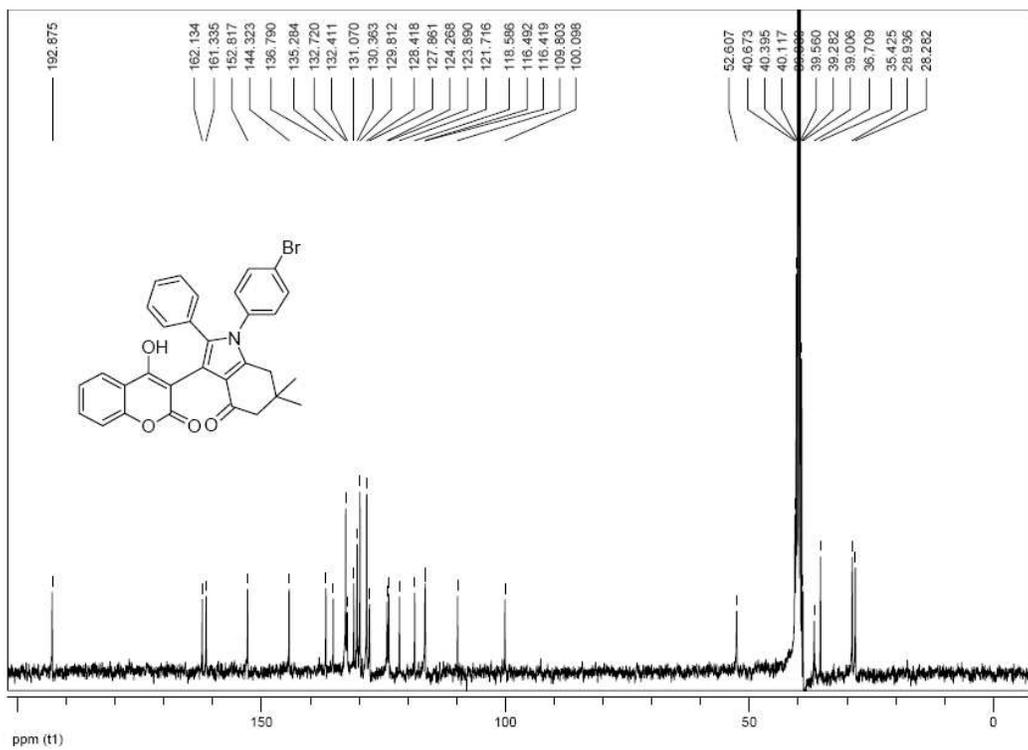
¹³C NMR spectrum of compound **4{1,1,3}**



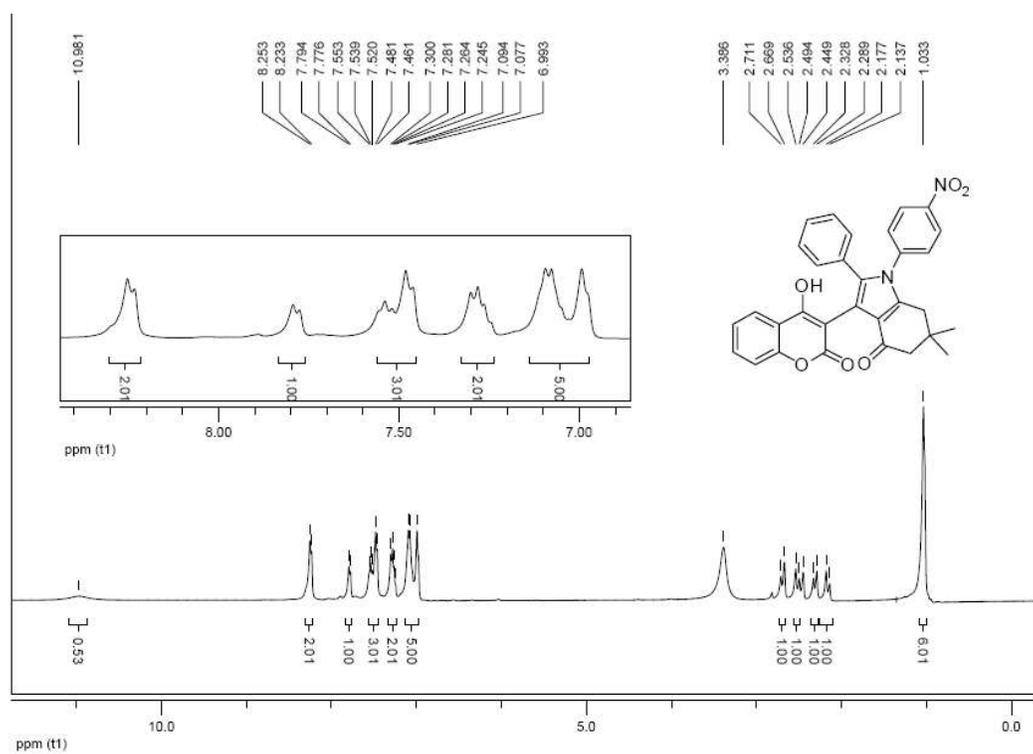
¹H NMR spectrum of compound **4{1,1,4}**



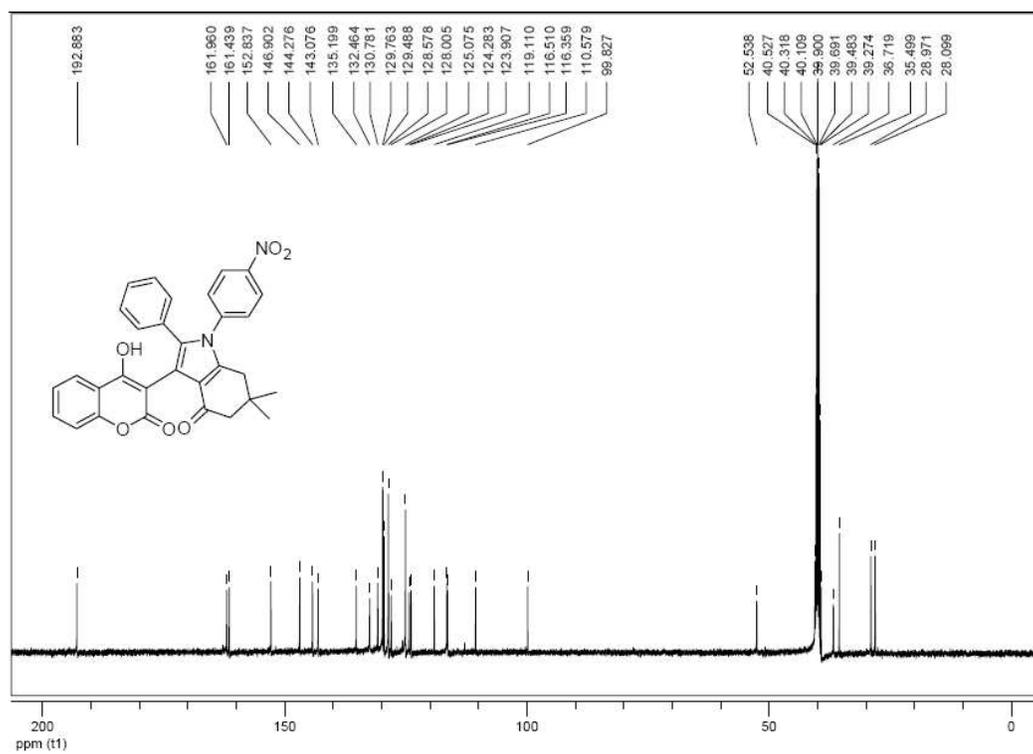
¹³C NMR spectrum of compound **4{1,1,4}**



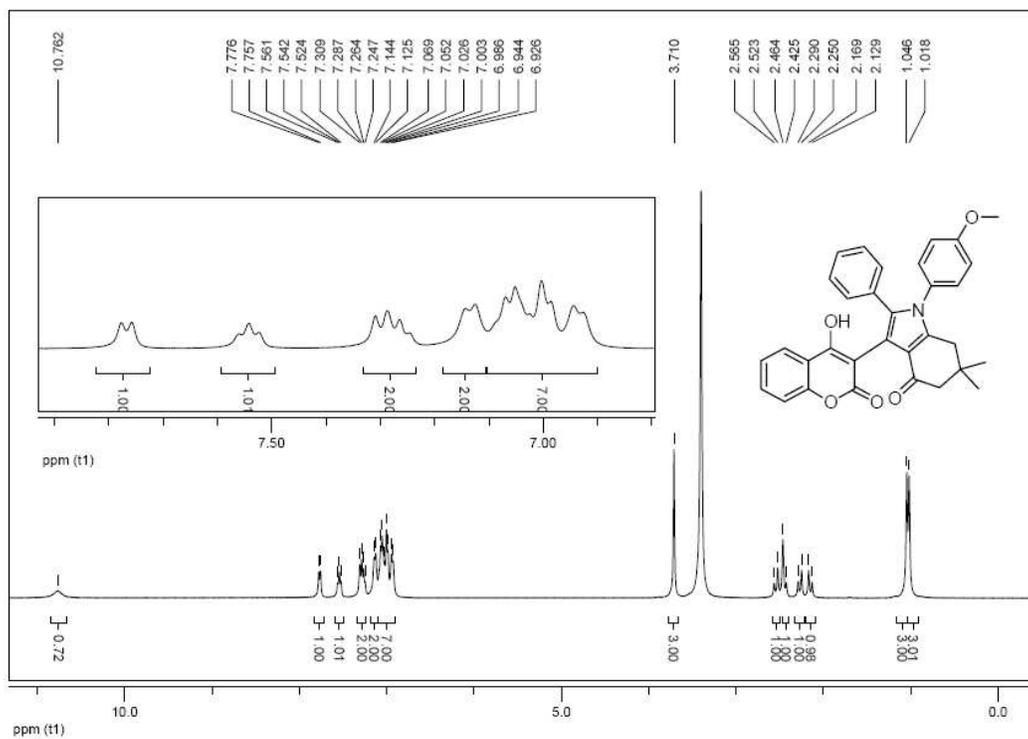
¹H NMR spectrum of compound **4{1,1,5}**



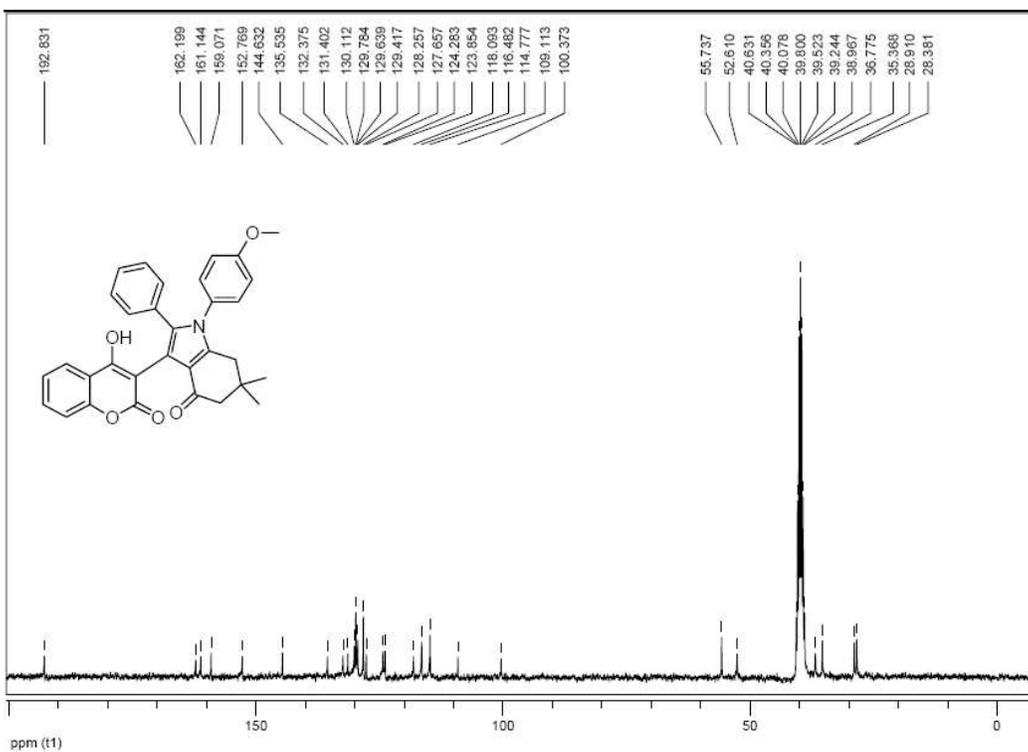
¹³C NMR spectrum of compound **4{1,1,5}**



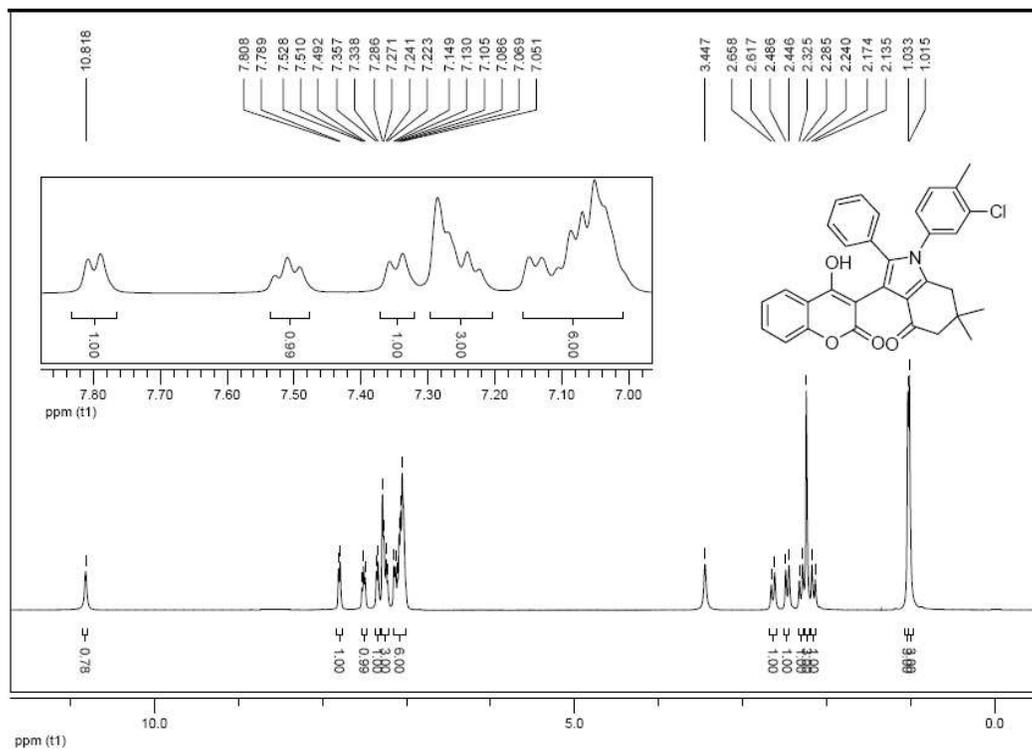
¹H NMR spectrum of compound **4{1,1,6}**



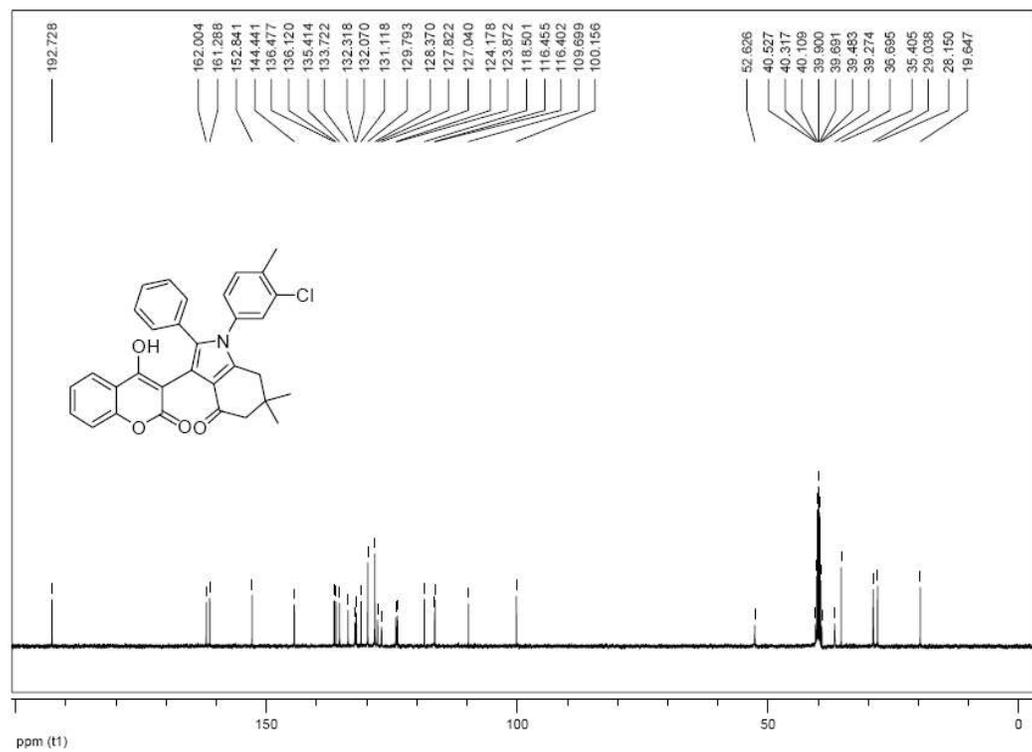
¹³C NMR spectrum of compound **4{1,1,6}**



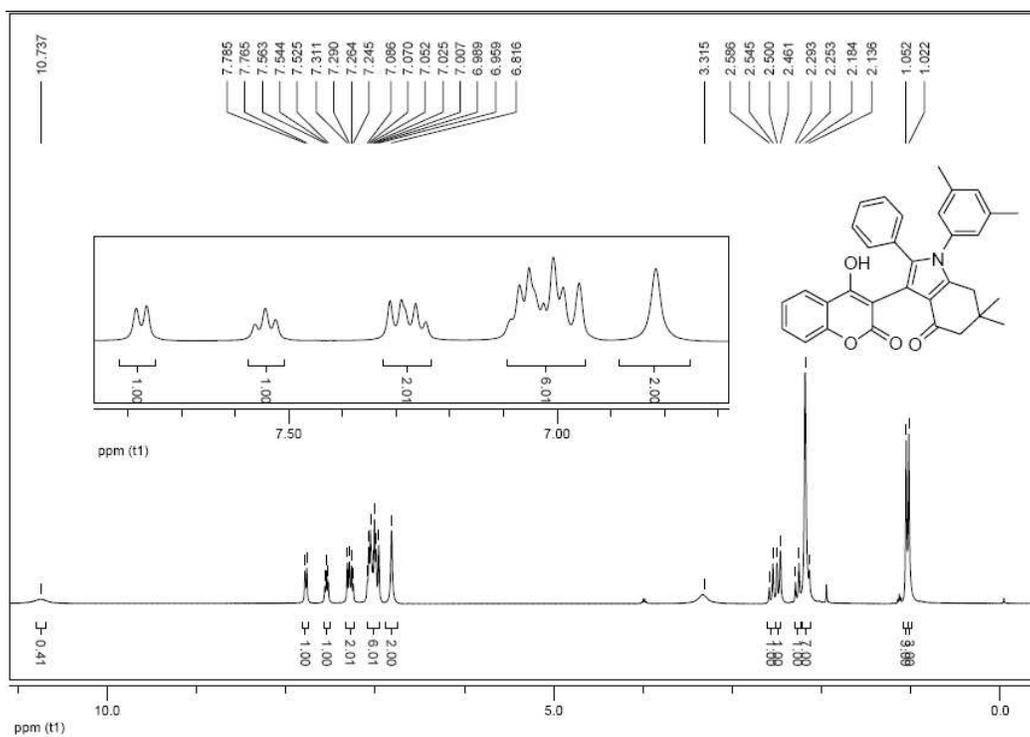
¹H NMR spectrum of compound **4**{1,1,7}



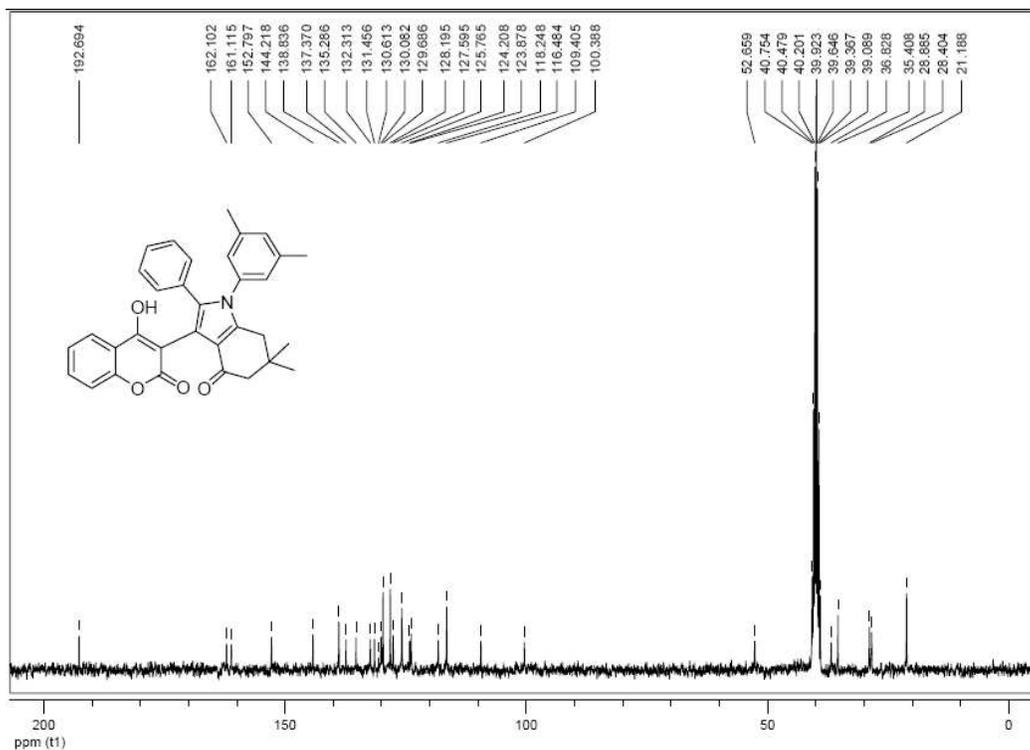
¹³C NMR spectrum of compound **4**{1,1,7}



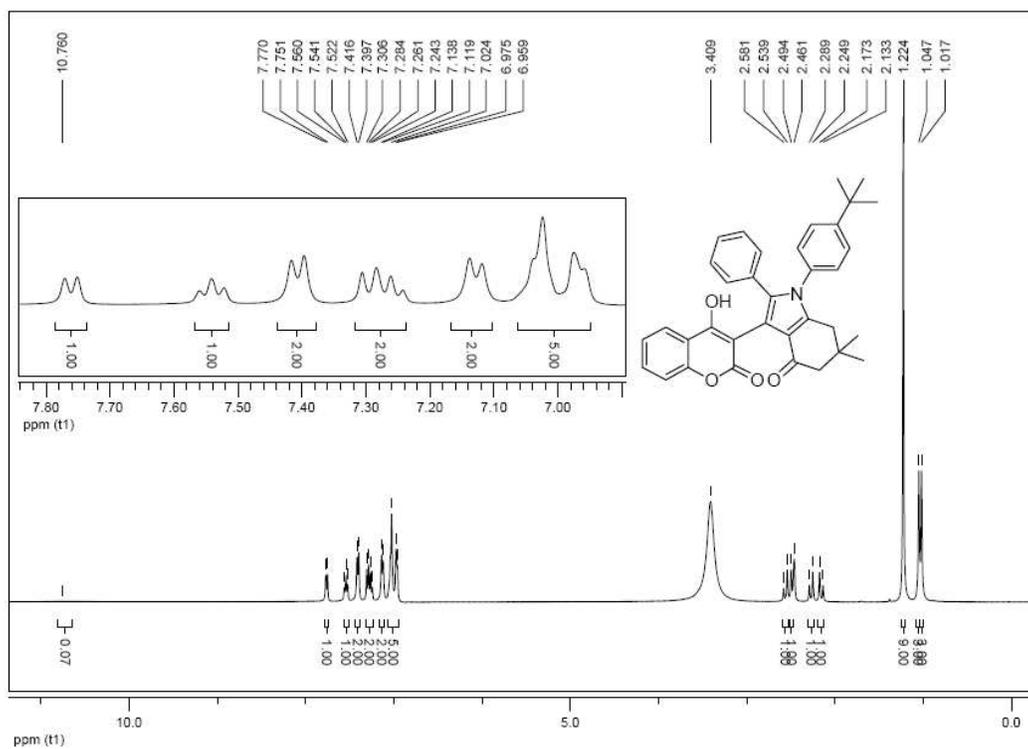
¹H NMR spectrum of compound **4{1,1,8}**



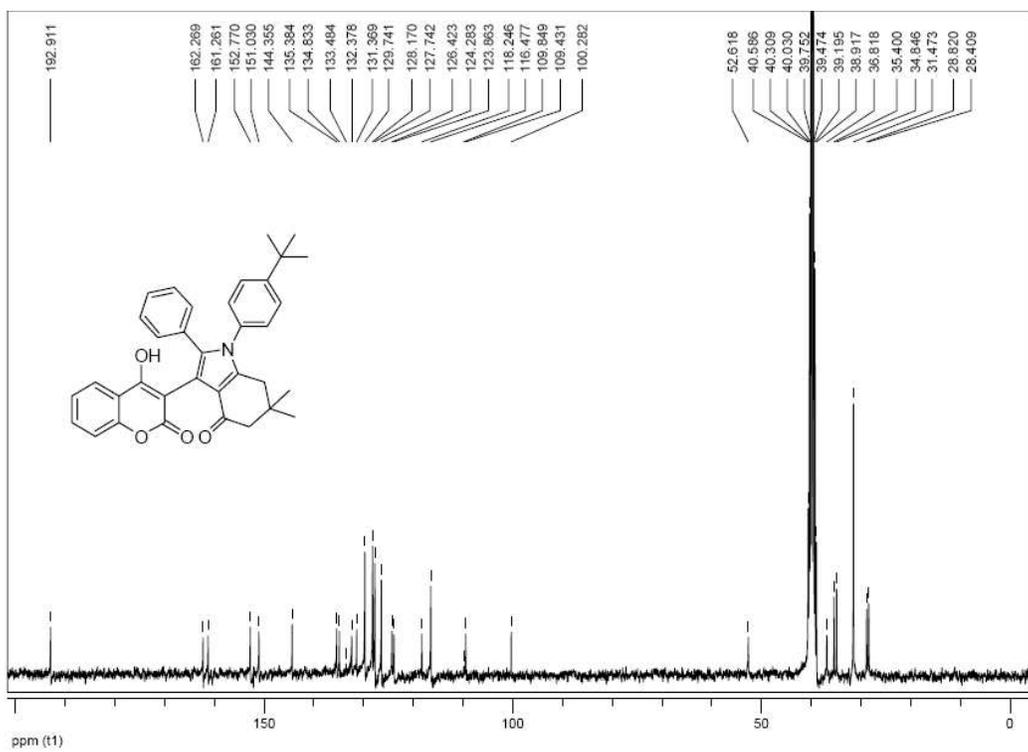
¹³C NMR spectrum of compound **4{1,1,8}**



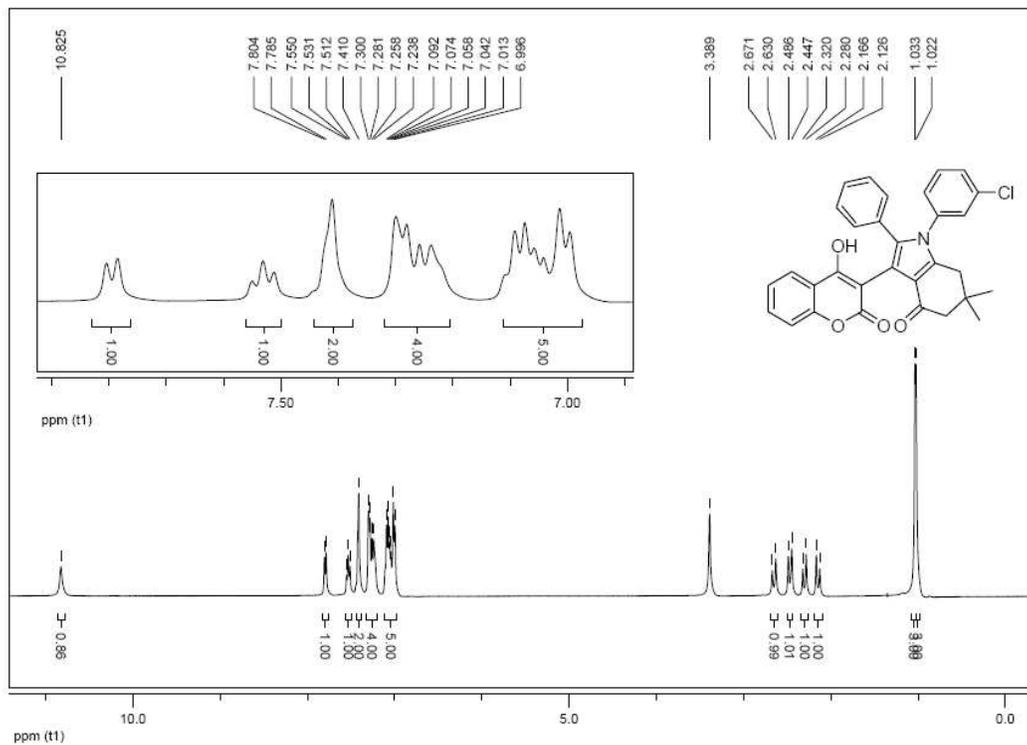
^1H NMR spectrum of compound **4{1,1,9}**



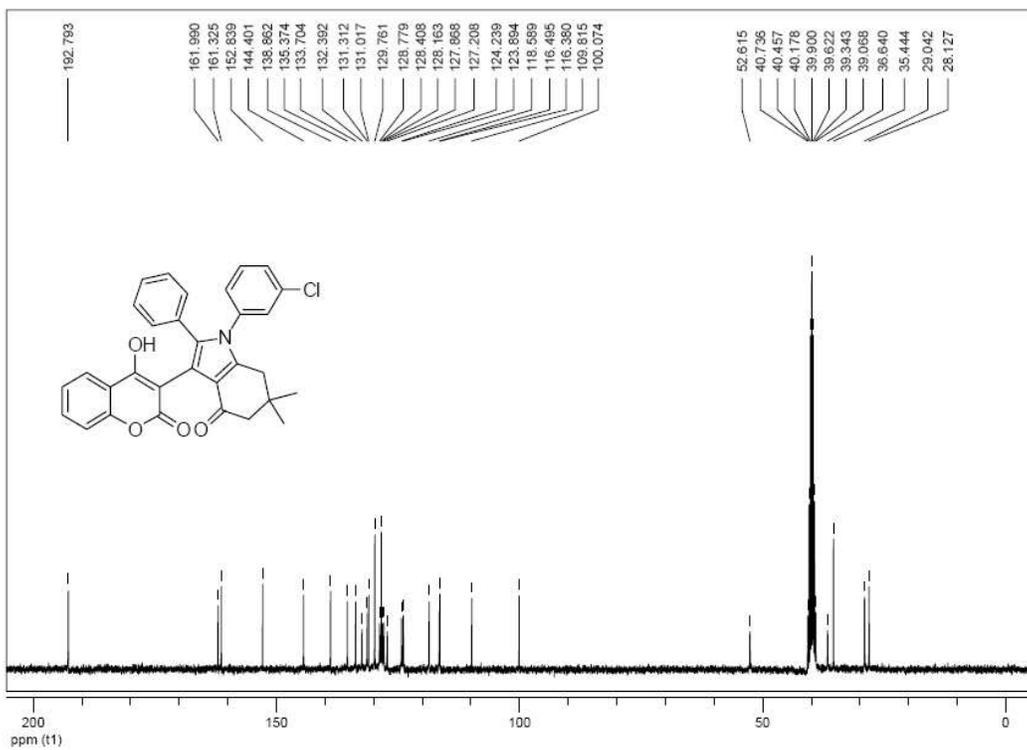
^{13}C NMR spectrum of compound **4{1,1,9}**



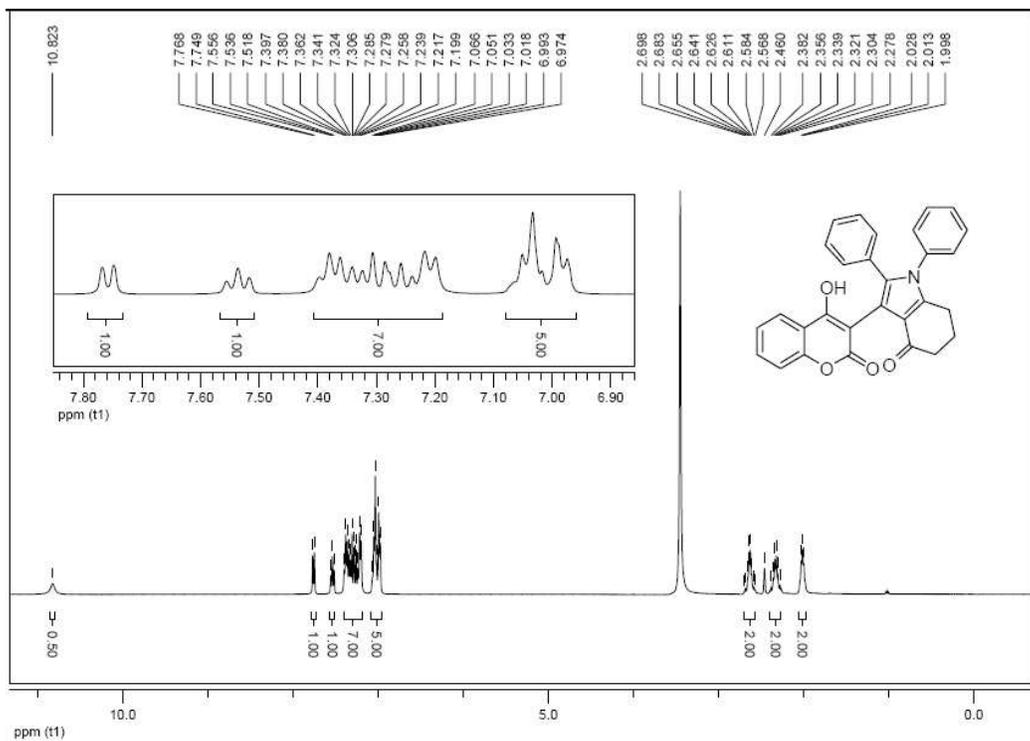
¹H NMR spectrum of compound **4{1,1,10}**



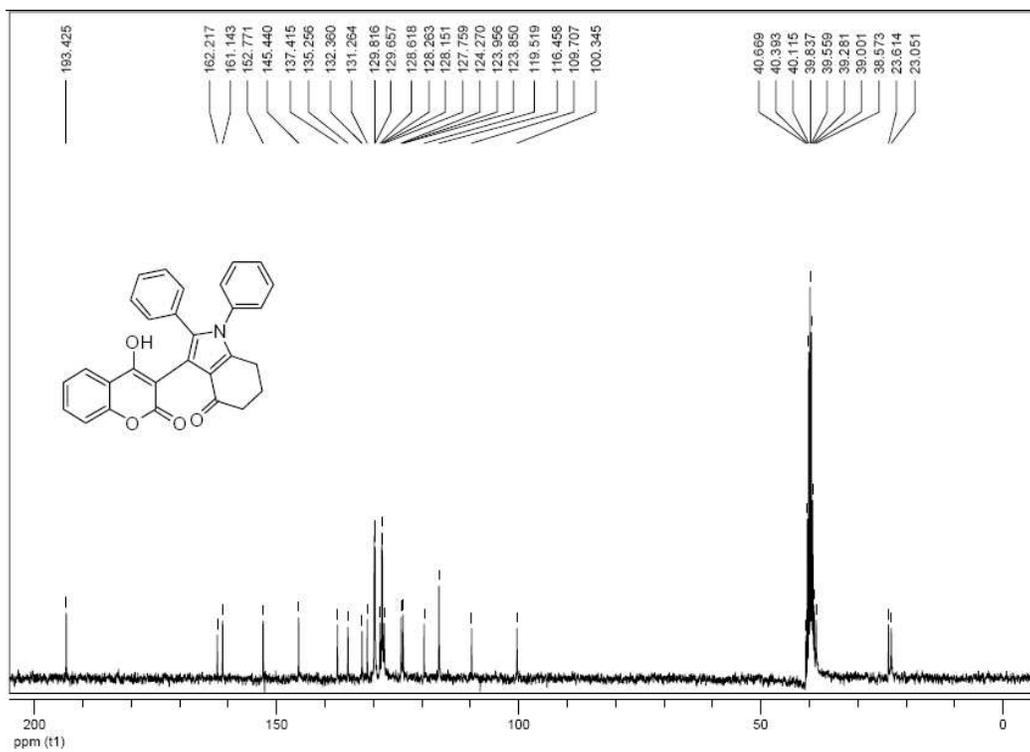
¹³C NMR spectrum of compound **4{1,1,10}**



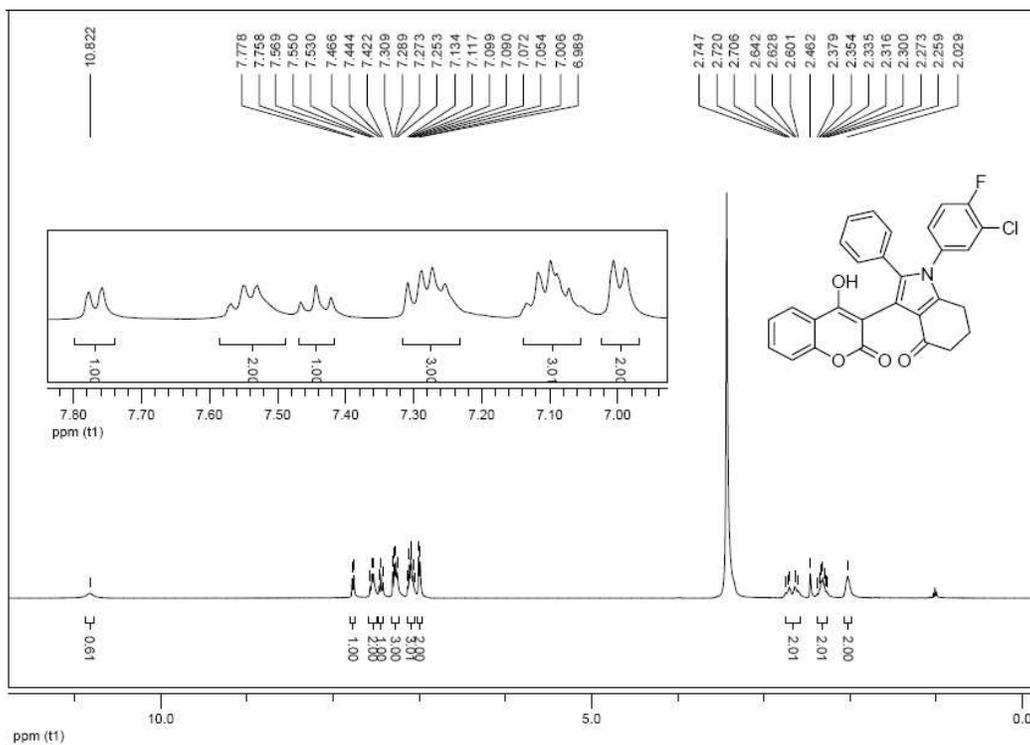
¹H NMR spectrum of compound 4{1,1,13}



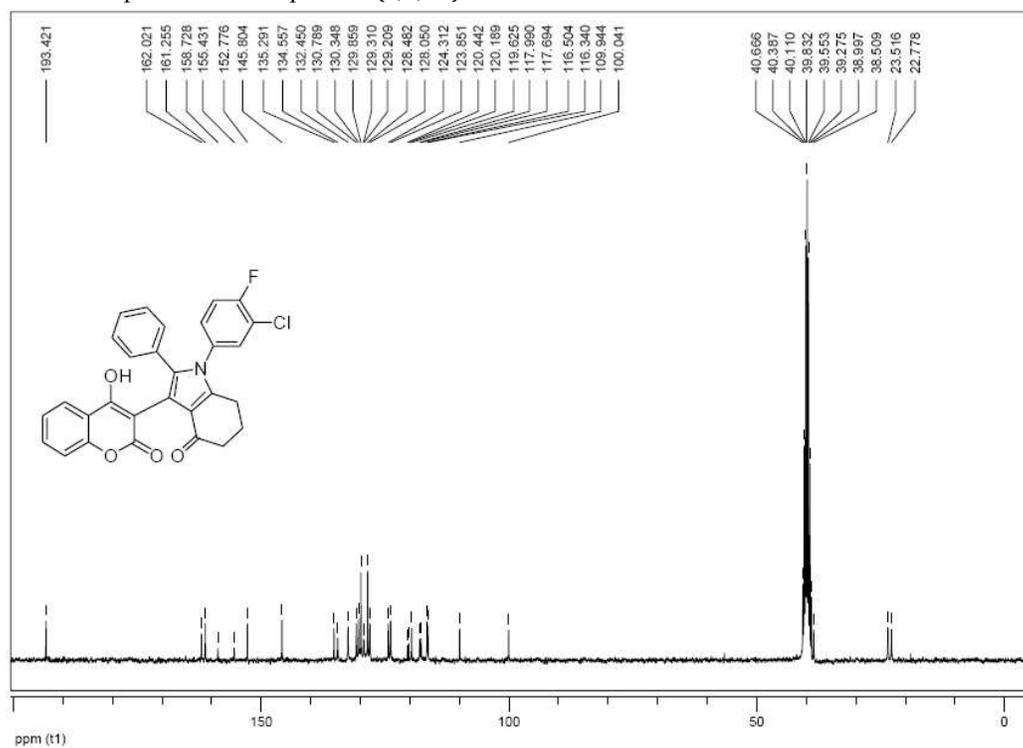
¹³C NMR spectrum of compound 4{1,1,13}



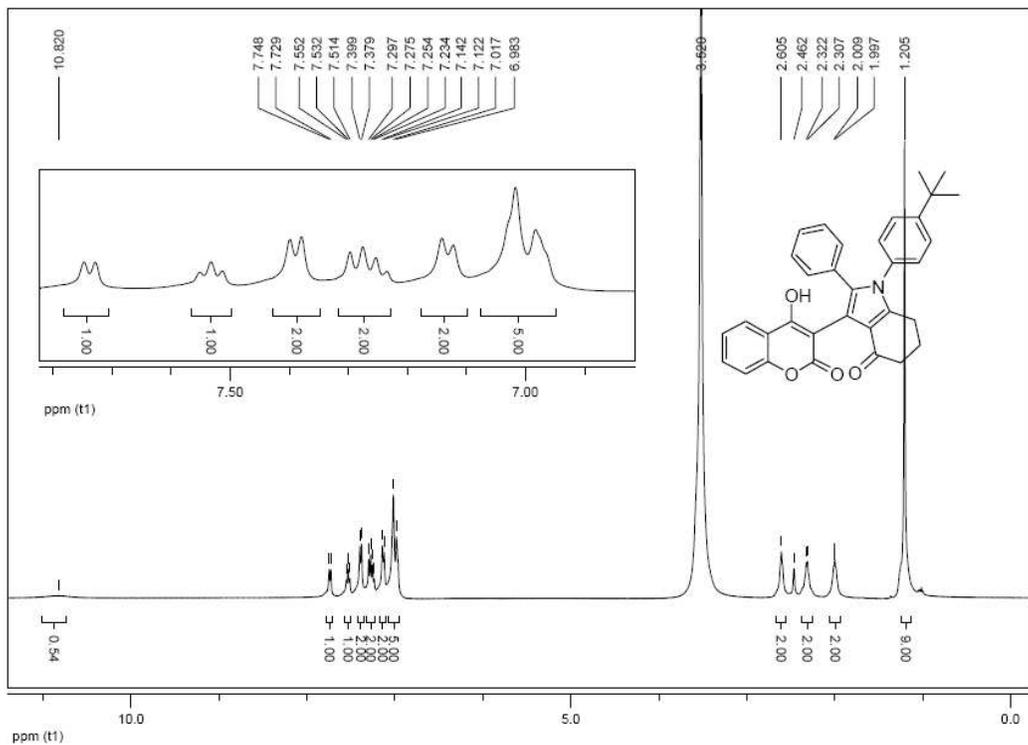
¹H NMR spectrum of compound **4{1,1,14}**



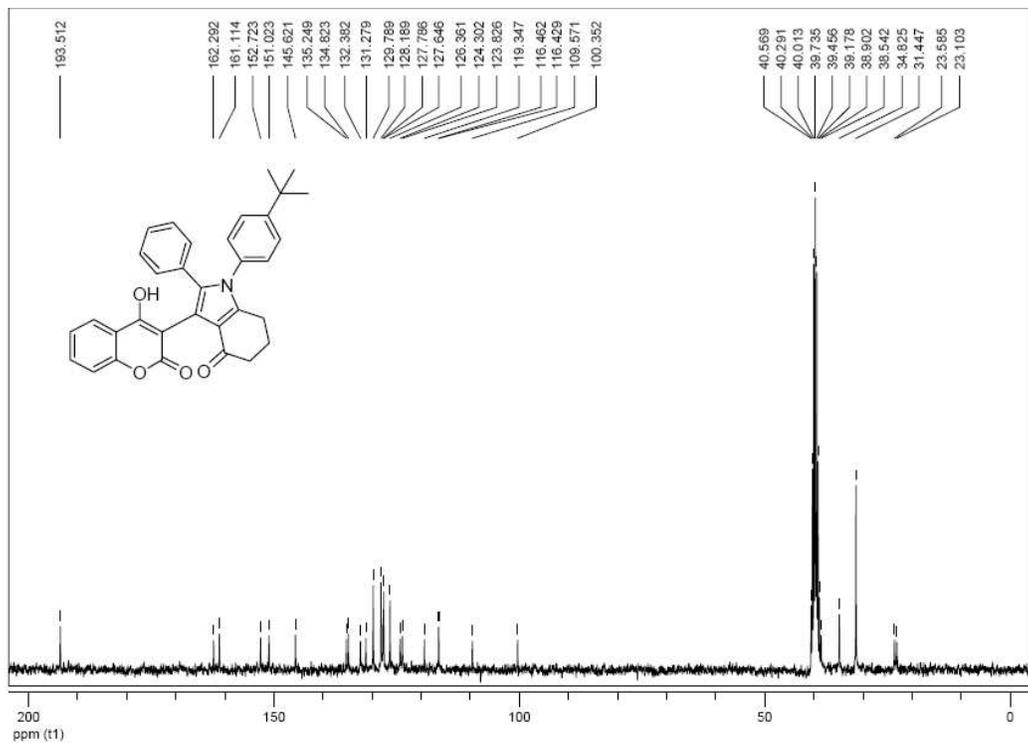
¹³C NMR spectrum of compound **4{1,1,14}**



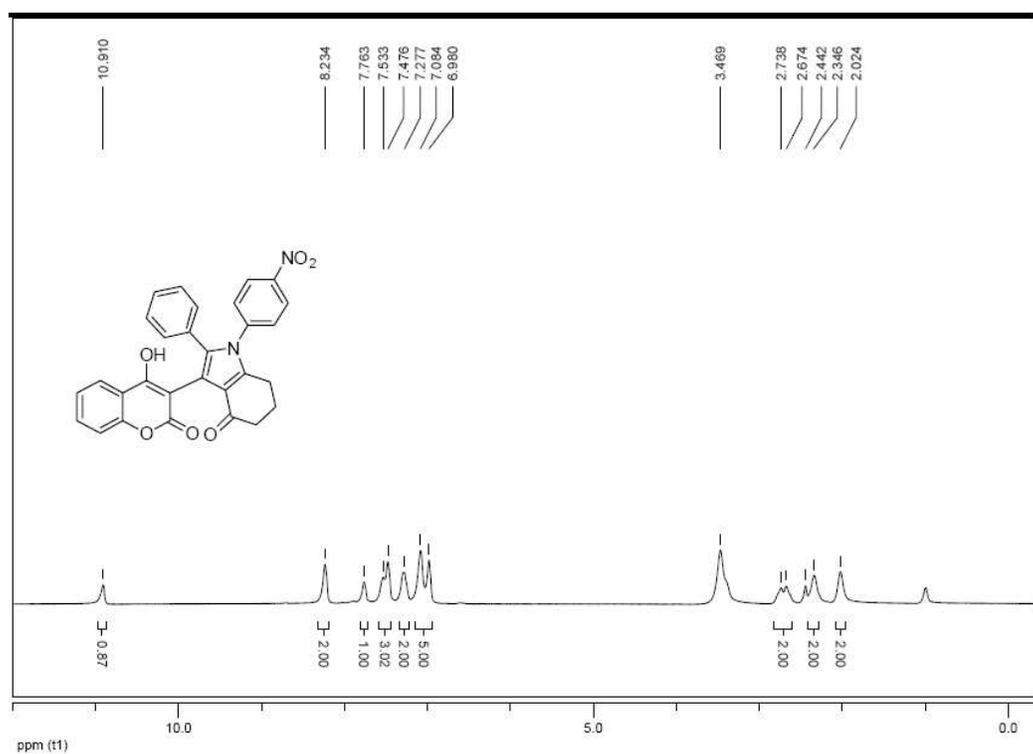
¹H NMR spectrum of compound **4**{*1,1,15*}



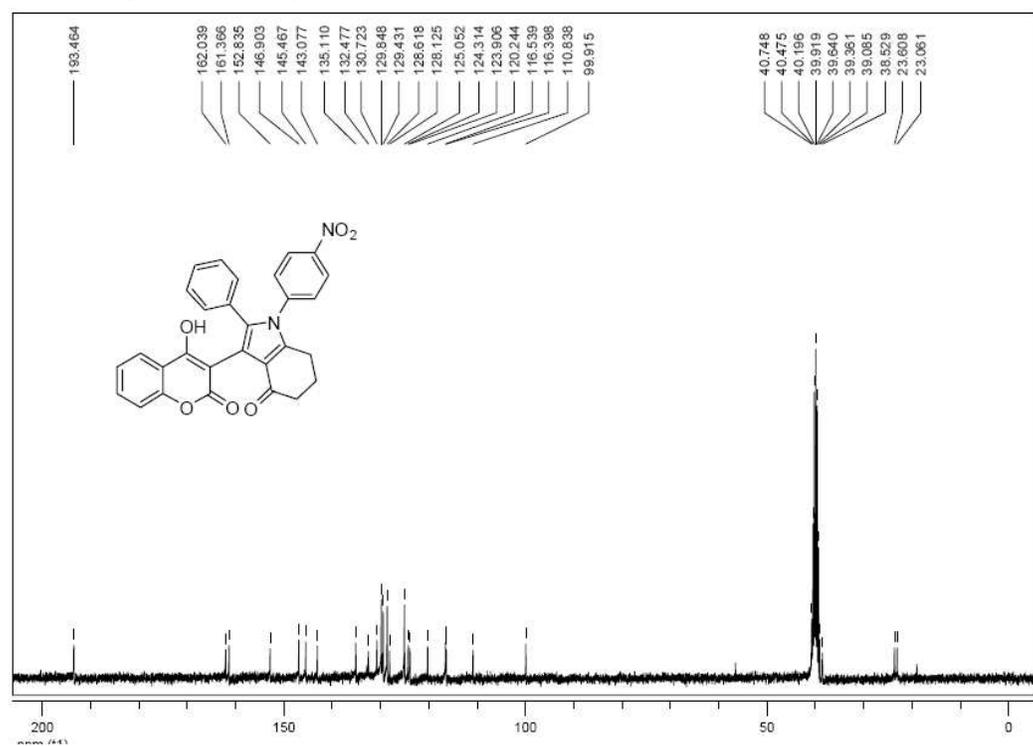
¹³C NMR spectrum of compound **4**{*1,1,15*}



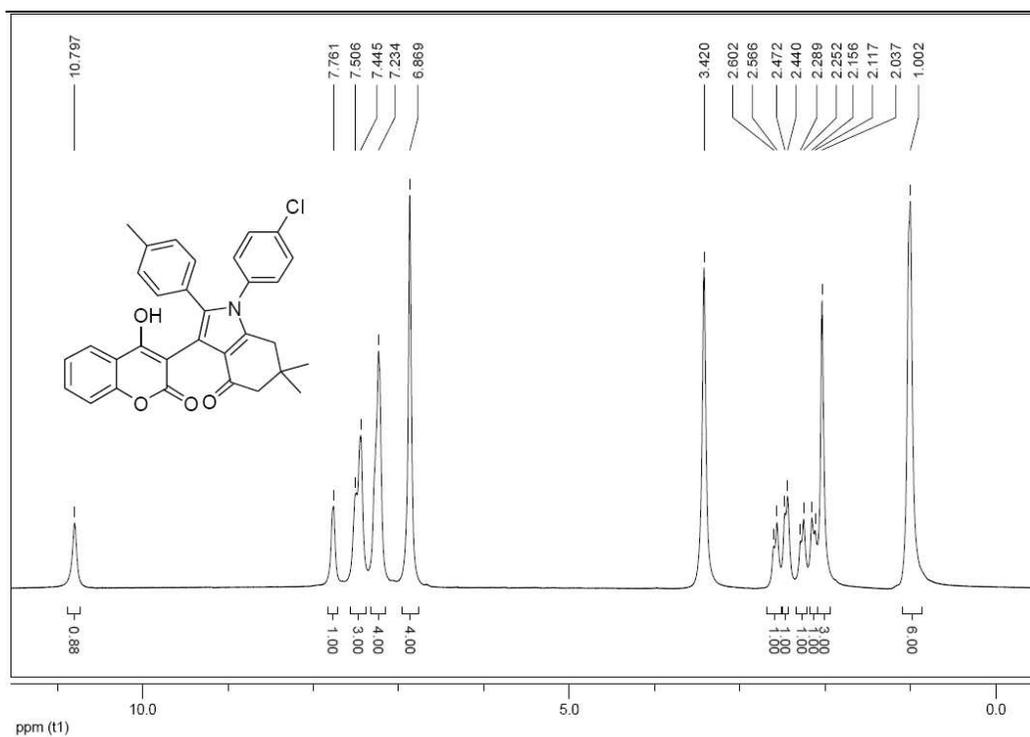
^1H NMR spectrum of compound **4{1,1,16}**



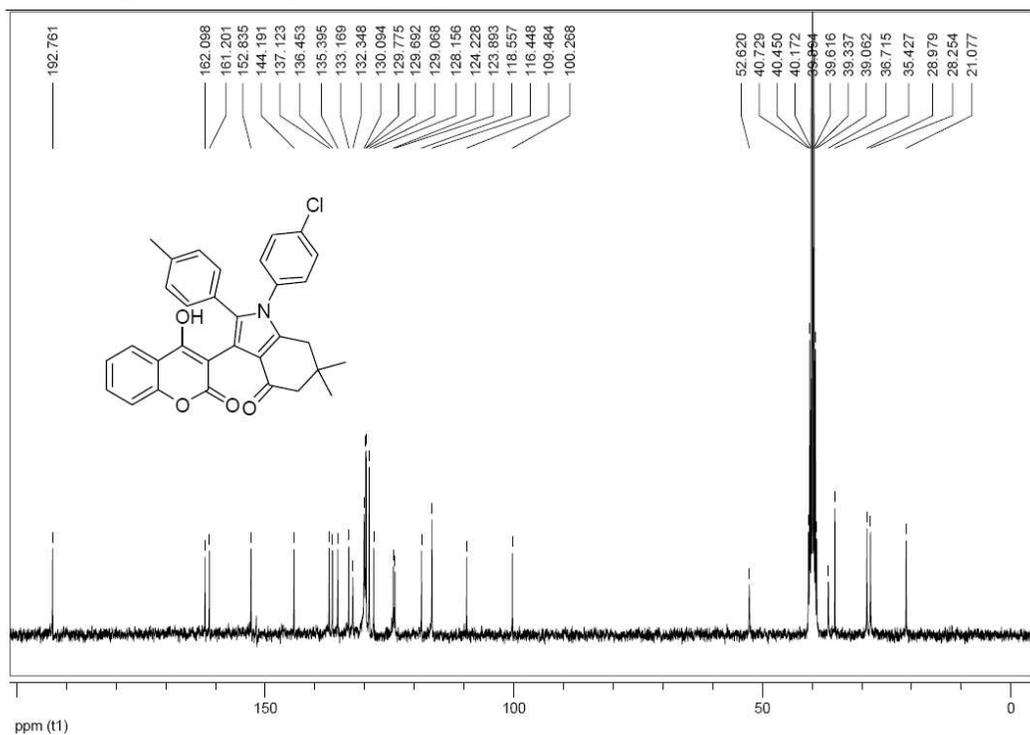
^{13}C NMR spectrum of compound **4{1,1,16}**



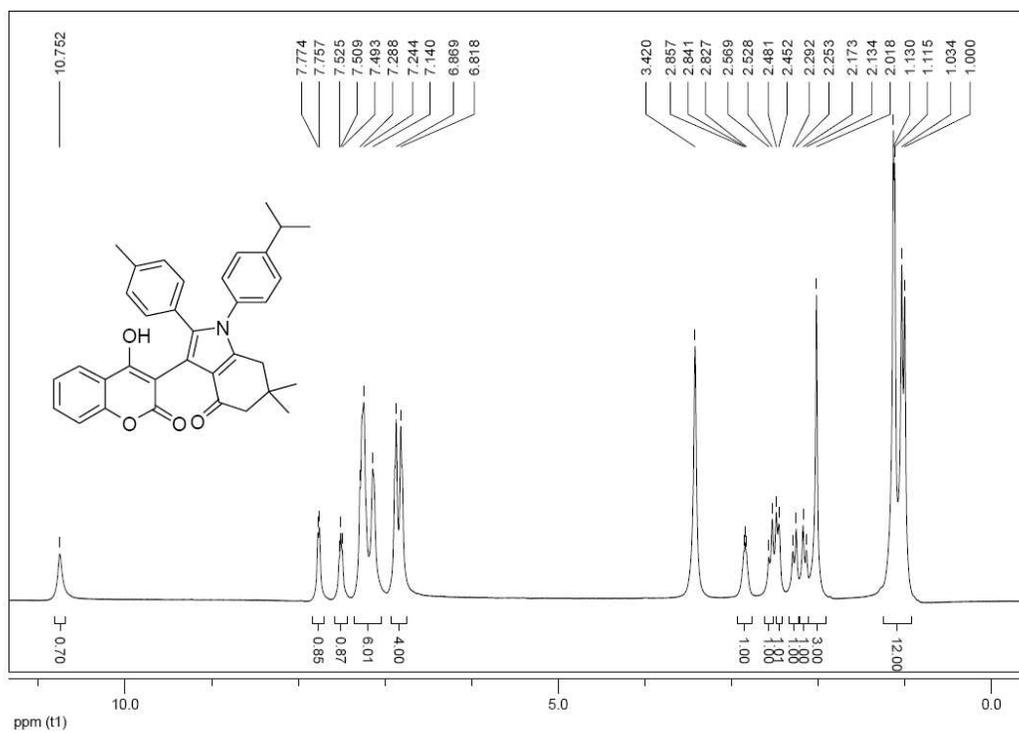
¹H NMR spectrum of compound 4{1,2,3}



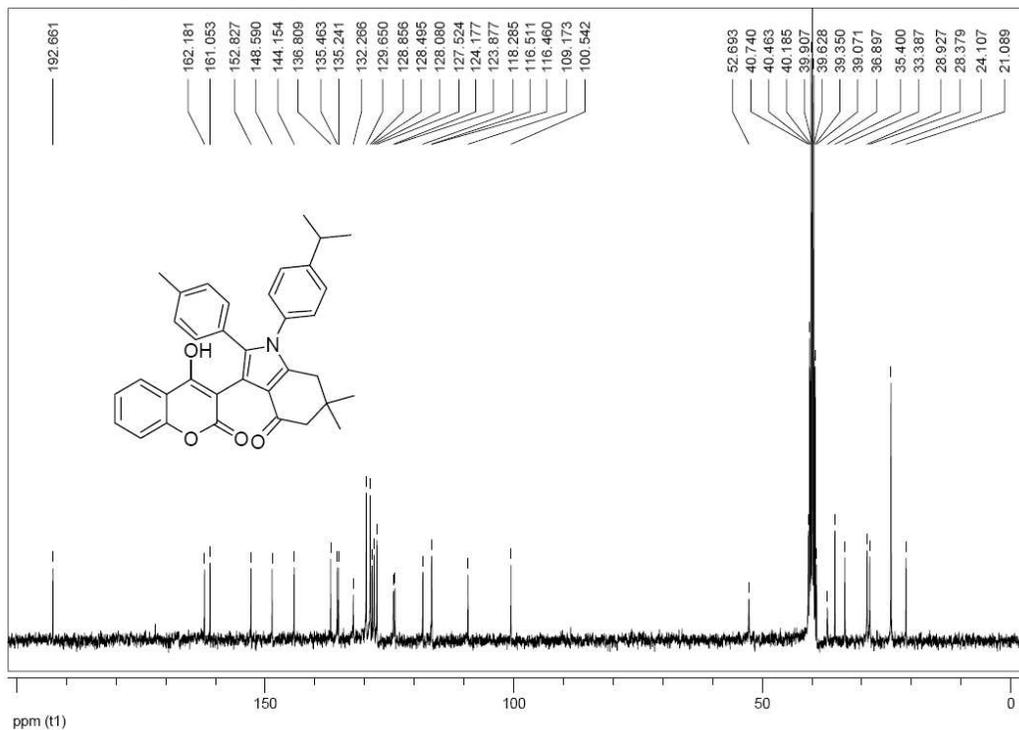
¹³C NMR spectrum of compound 4{1,2,3}



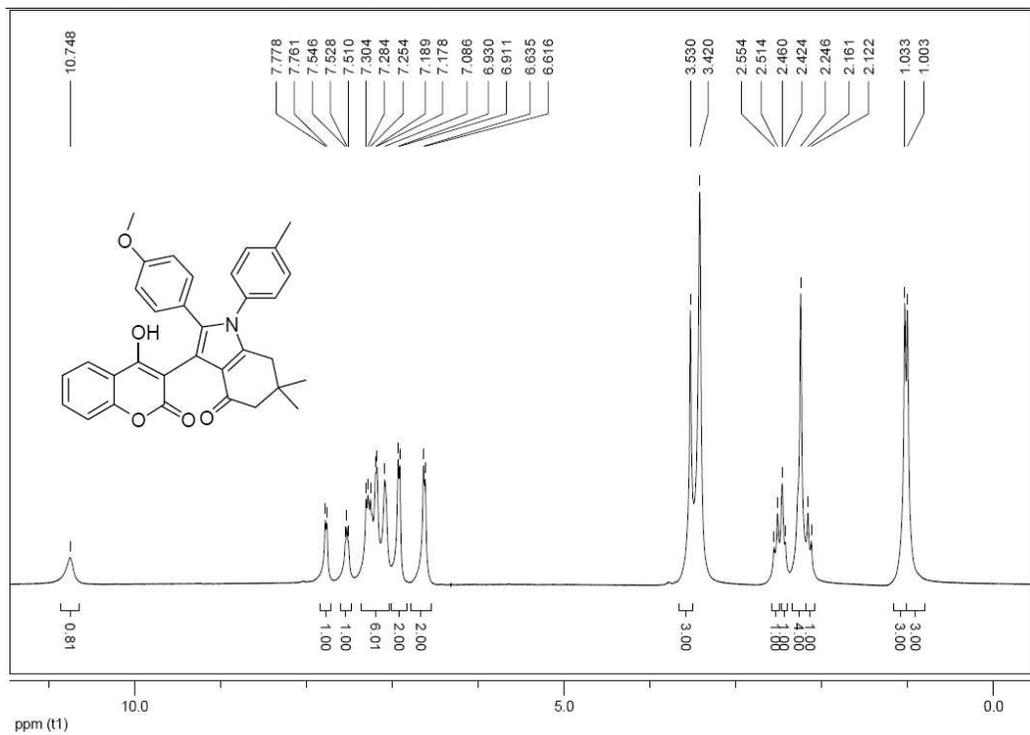
¹H NMR spectrum of compound **4**{*1,2,11*}



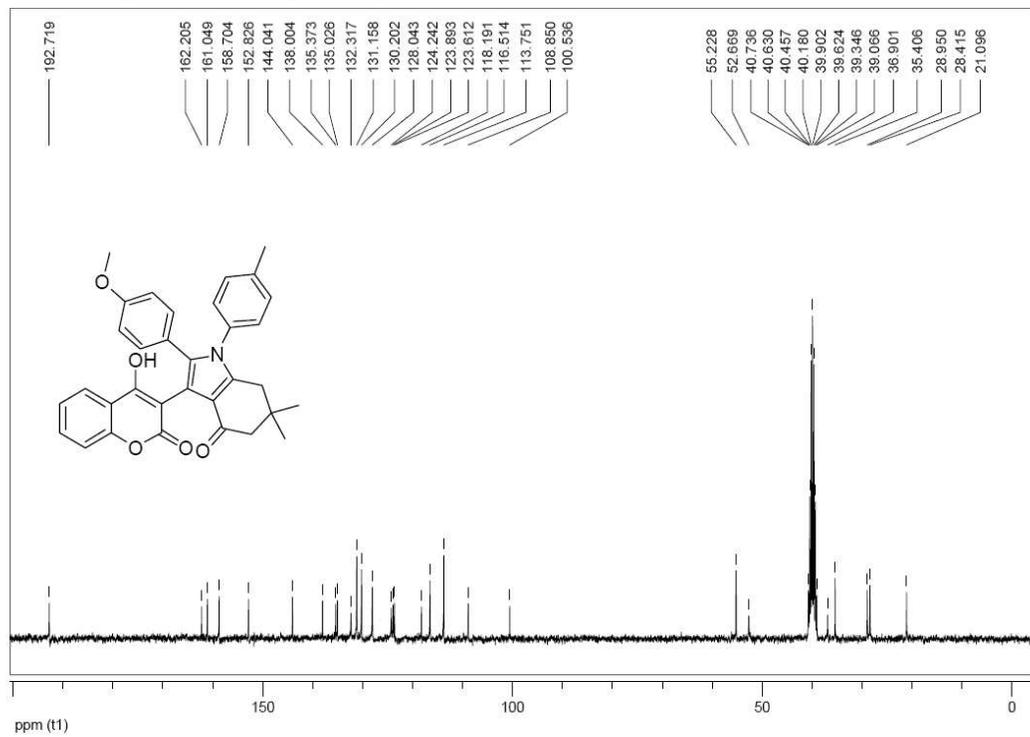
¹³C NMR spectrum of compound **4**{*1,2,11*}



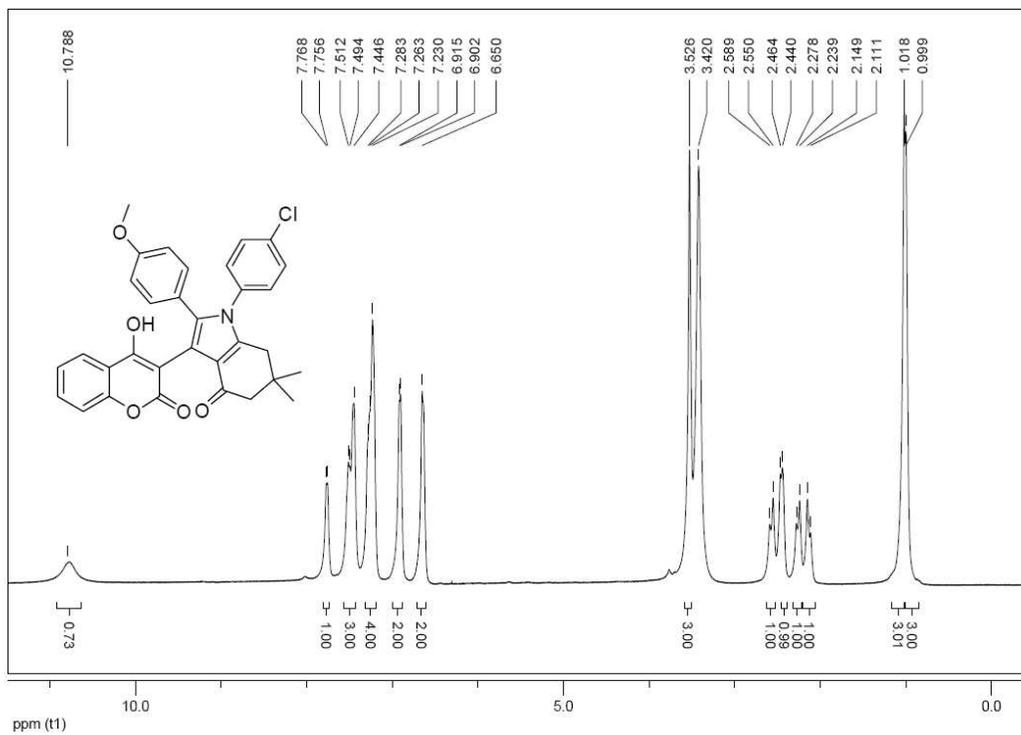
¹H NMR spectrum of compound **4**{*1,3,1*}



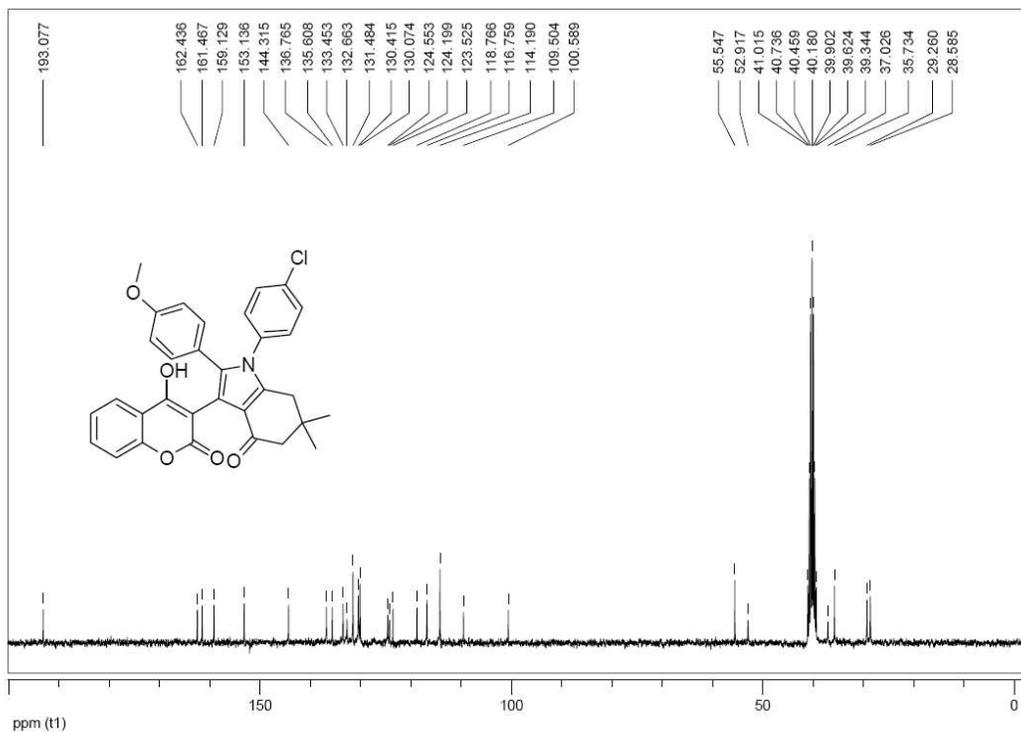
¹³C NMR spectrum of compound **4**{*1,3,1*}



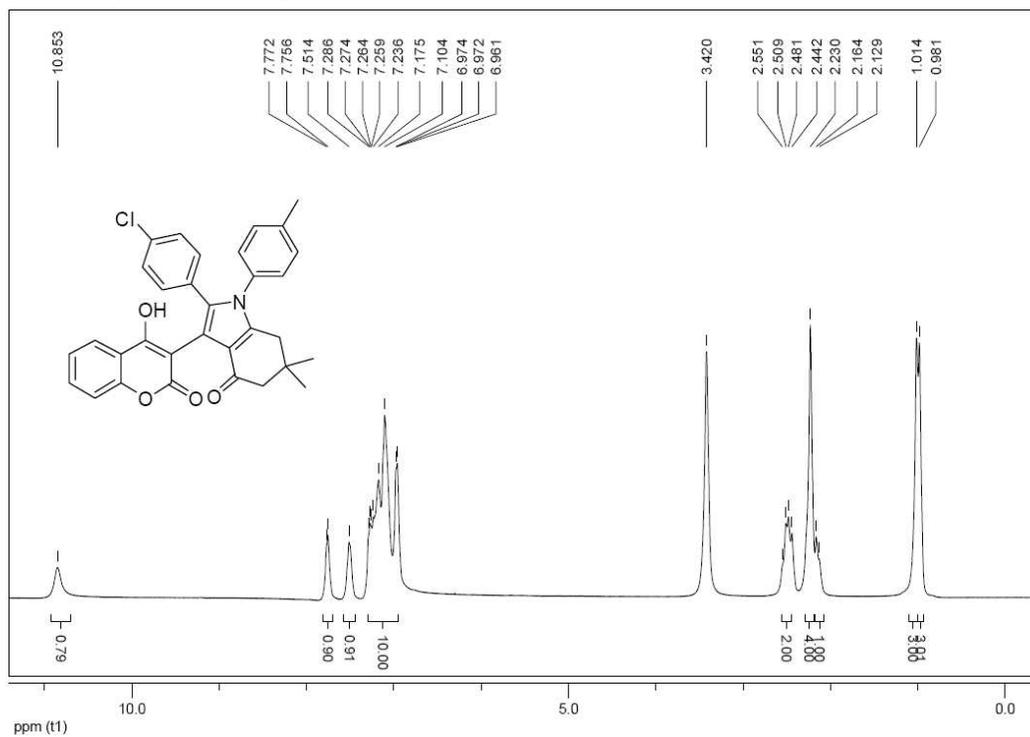
¹H NMR spectrum of compound **4**{1,3,3}



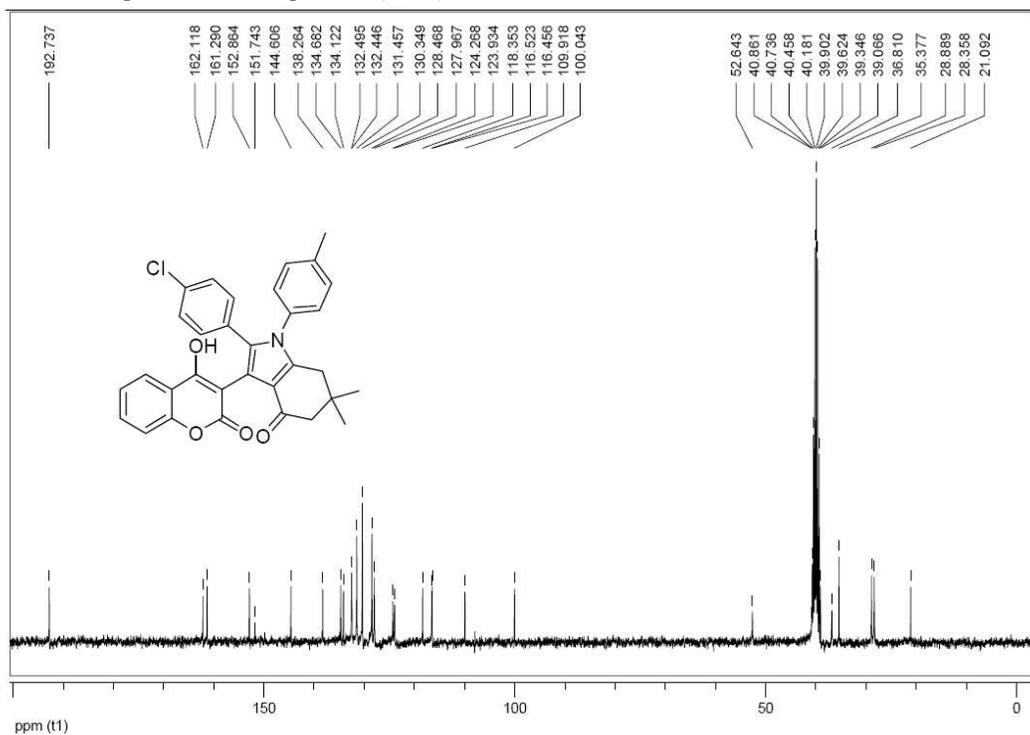
¹³C NMR spectrum of compound **4**{1,3,3}



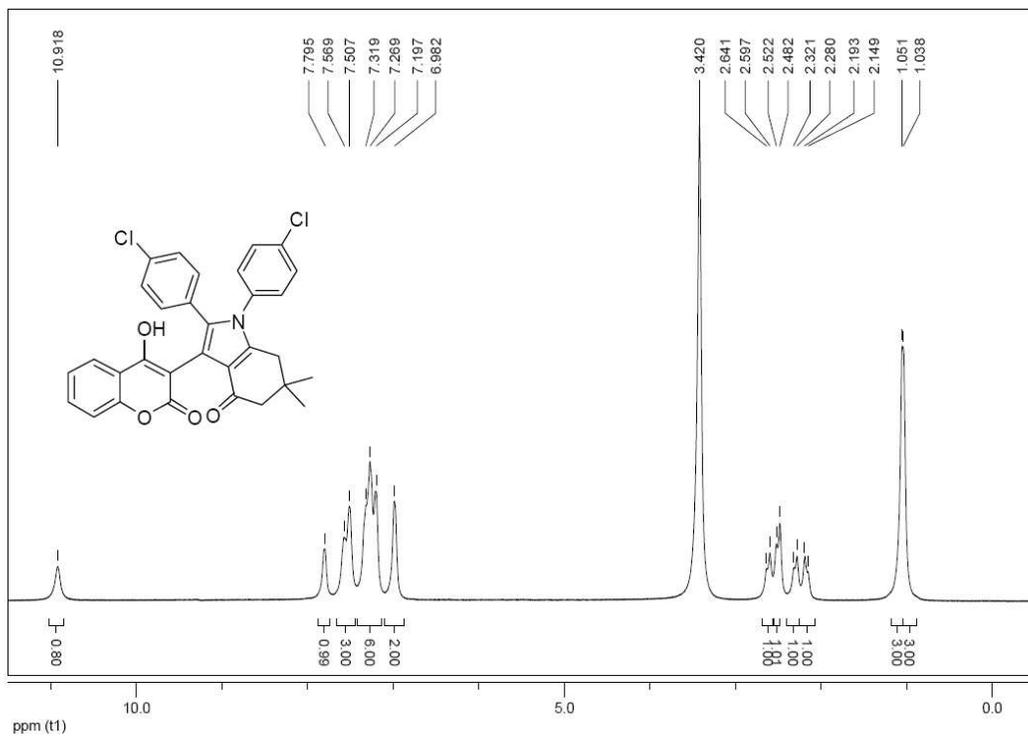
¹H NMR spectrum of compound **4{1,4,1}**



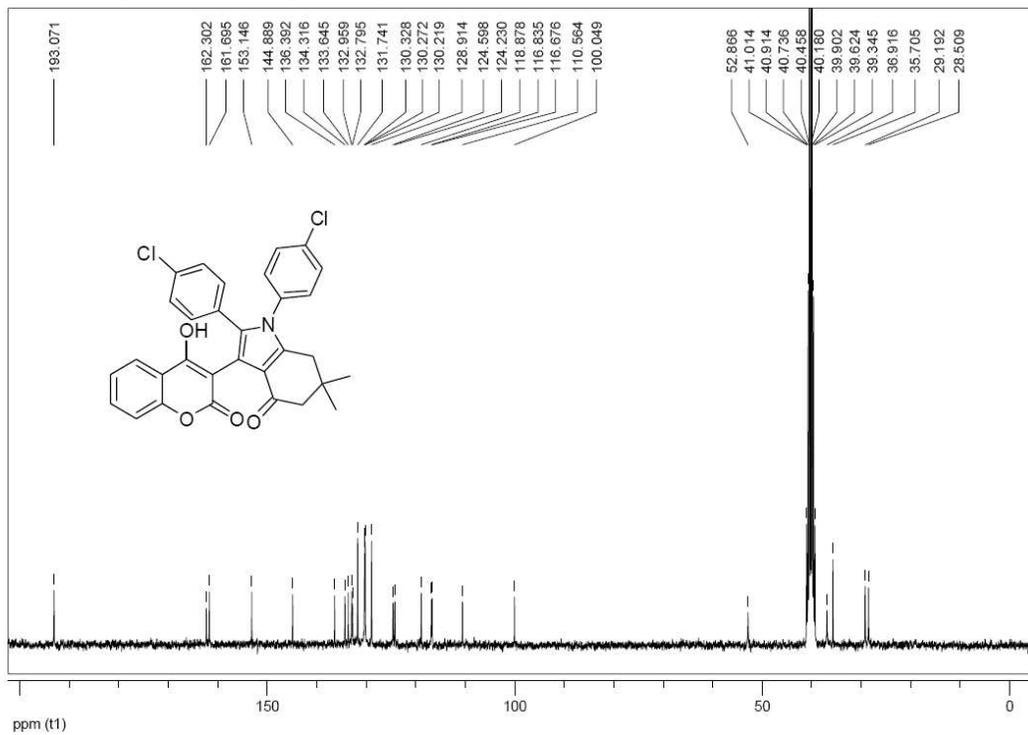
¹³C NMR spectrum of compound **4{1,4,1}**



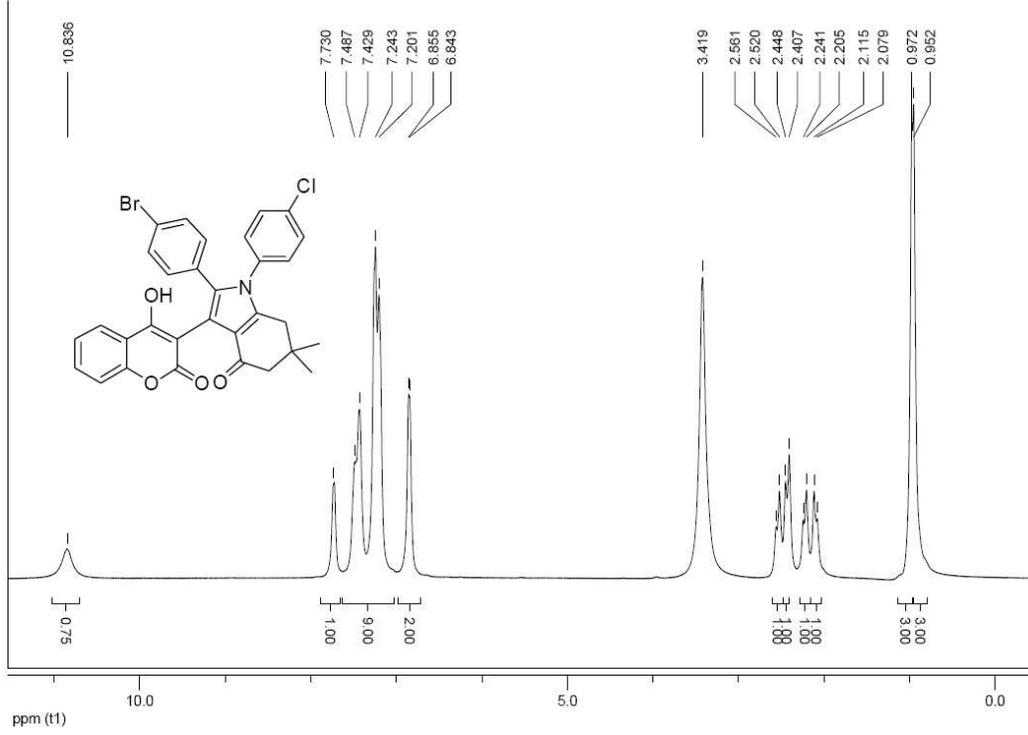
¹H NMR spectrum of compound 4{1,4,3}



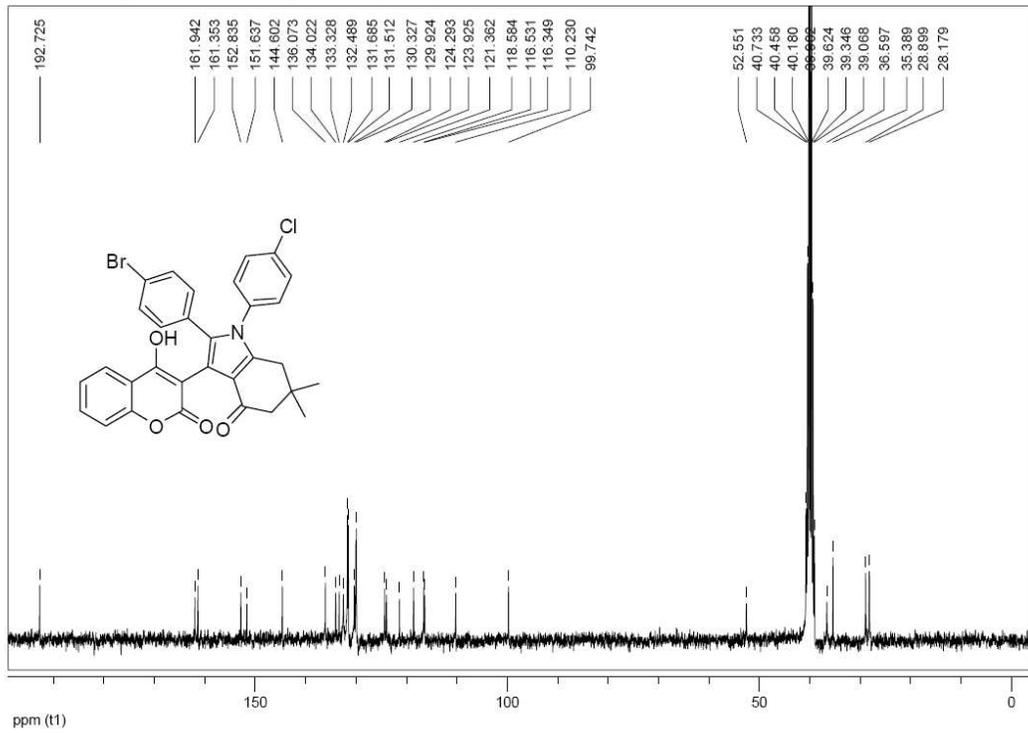
¹³C NMR spectrum of compound 4{1,4,3}



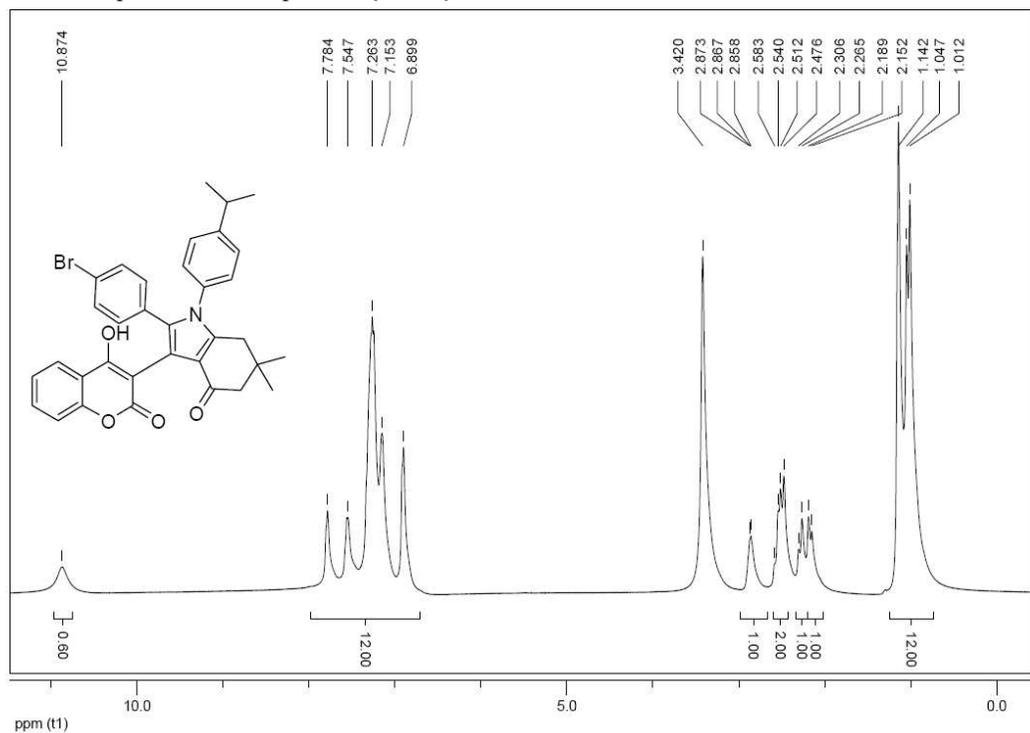
¹H NMR spectrum of compound 4{1,5,3}



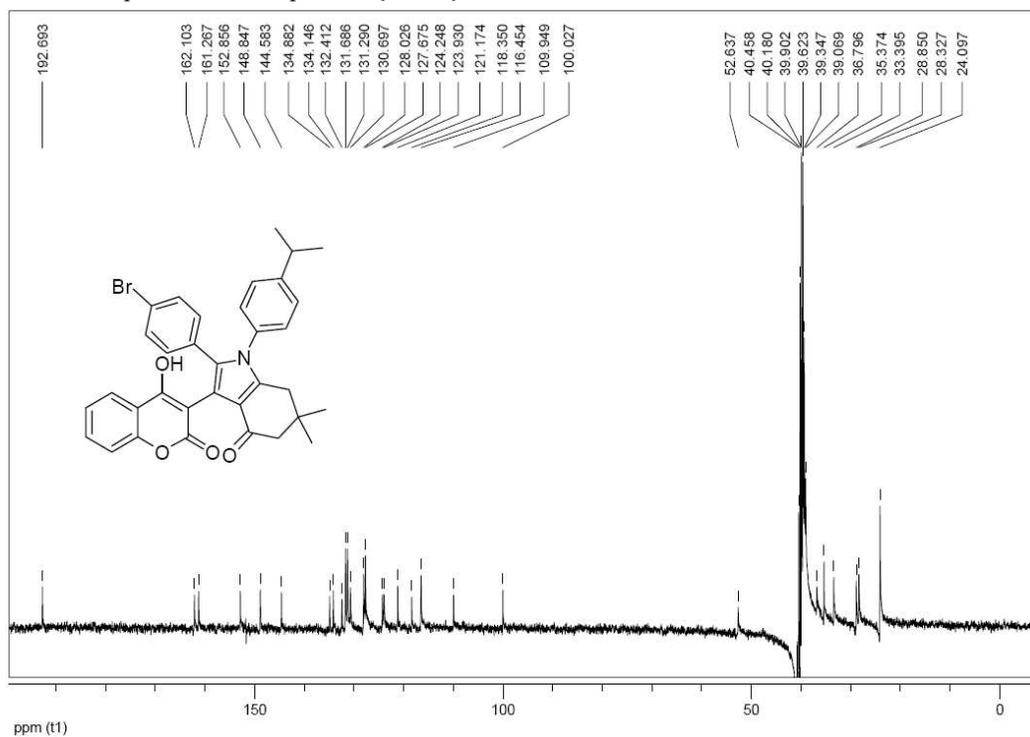
¹³C NMR spectrum of compound 4{1,5,3}



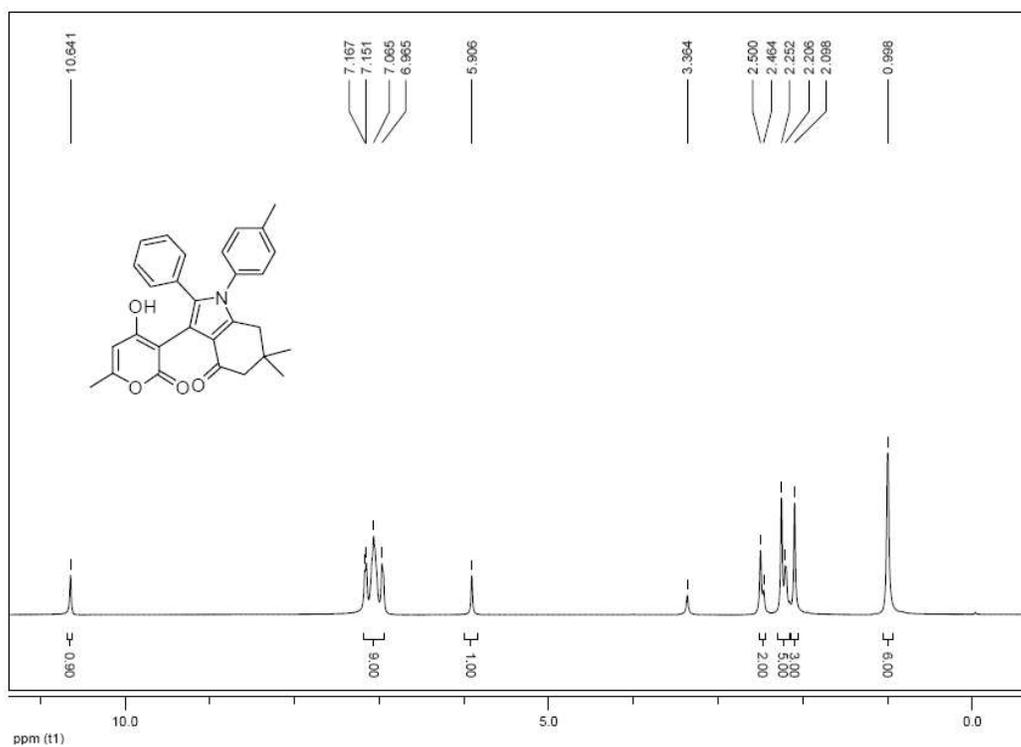
¹H NMR spectrum of compound 4{1,5,11}



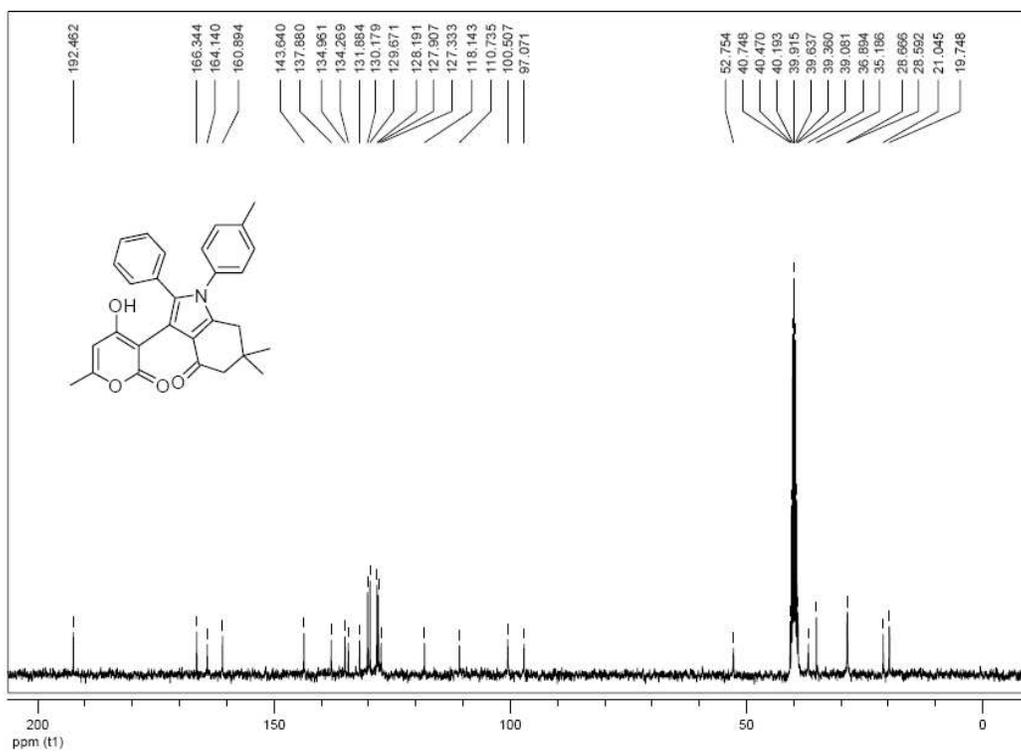
¹³C NMR spectrum of compound 4{1,5,11}



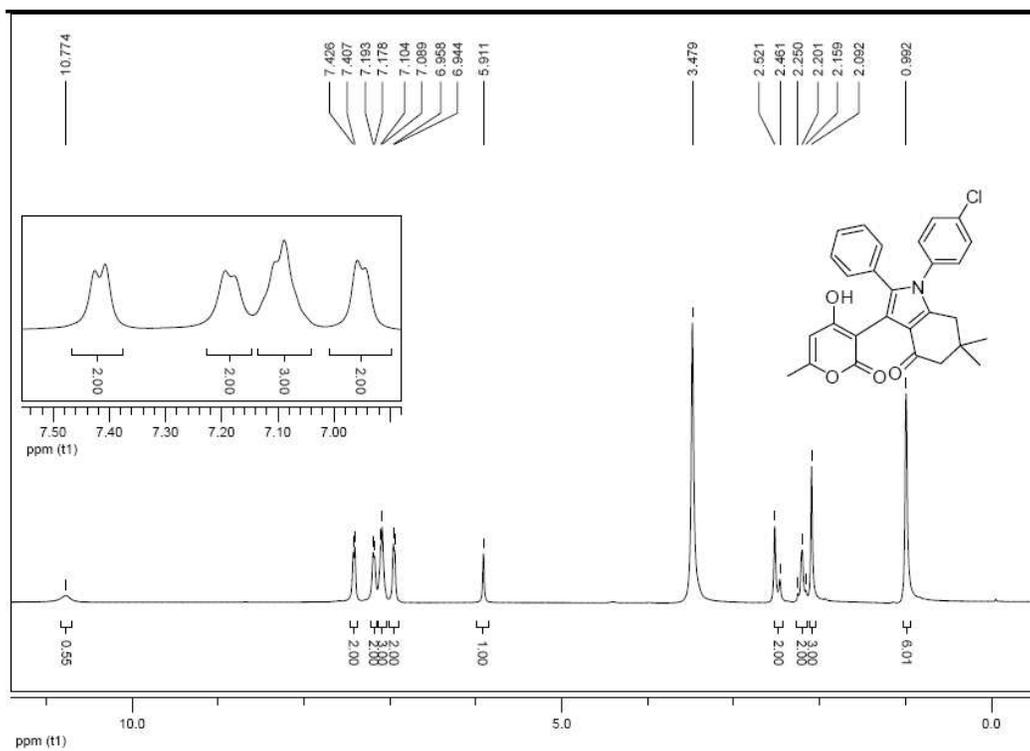
^1H NMR spectrum of compound **4**{2,1,1}



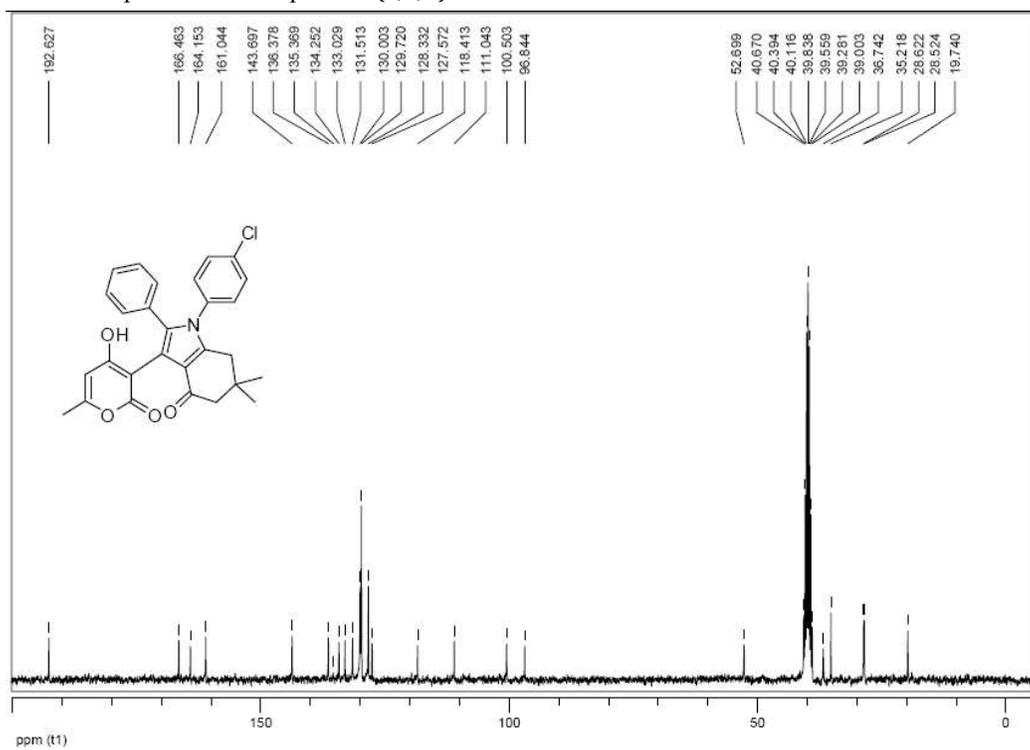
^{13}C NMR spectrum of compound **4**{2,1,1}



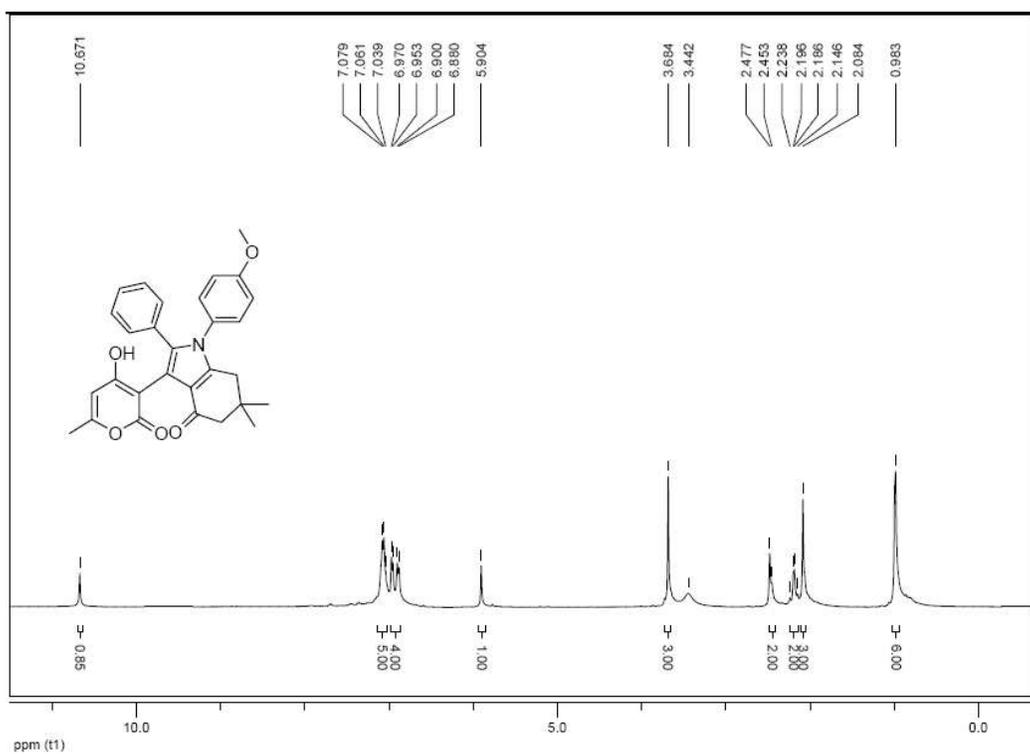
^1H NMR spectrum of compound **4**{2,1,3}



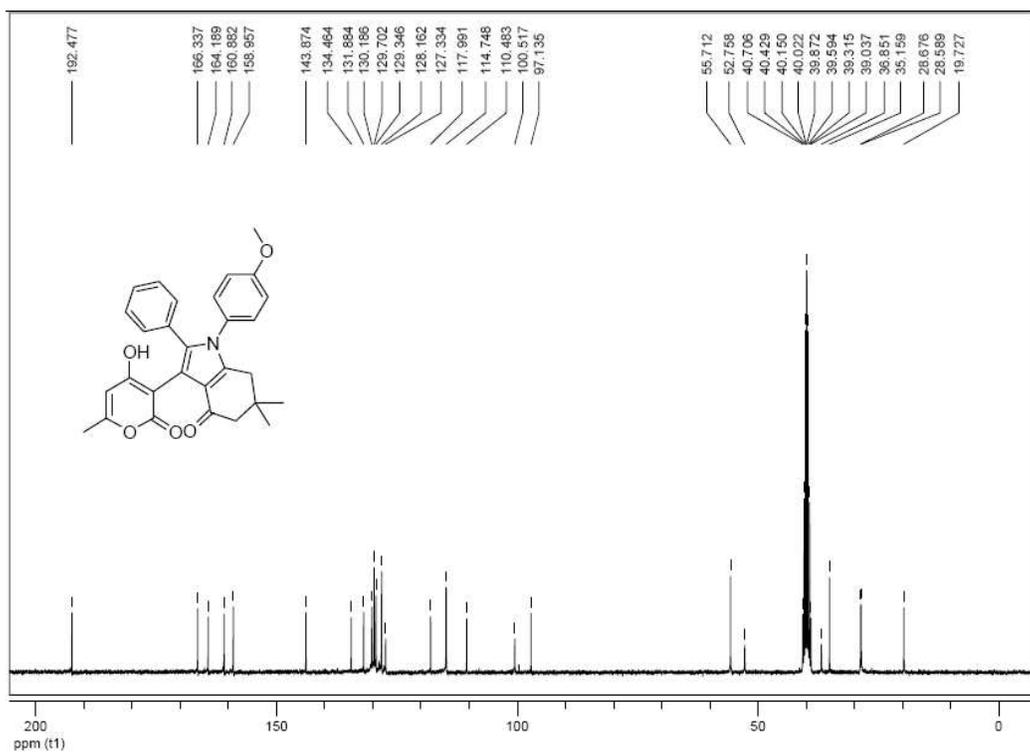
^{13}C NMR spectrum of compound **4**{2,1,3}



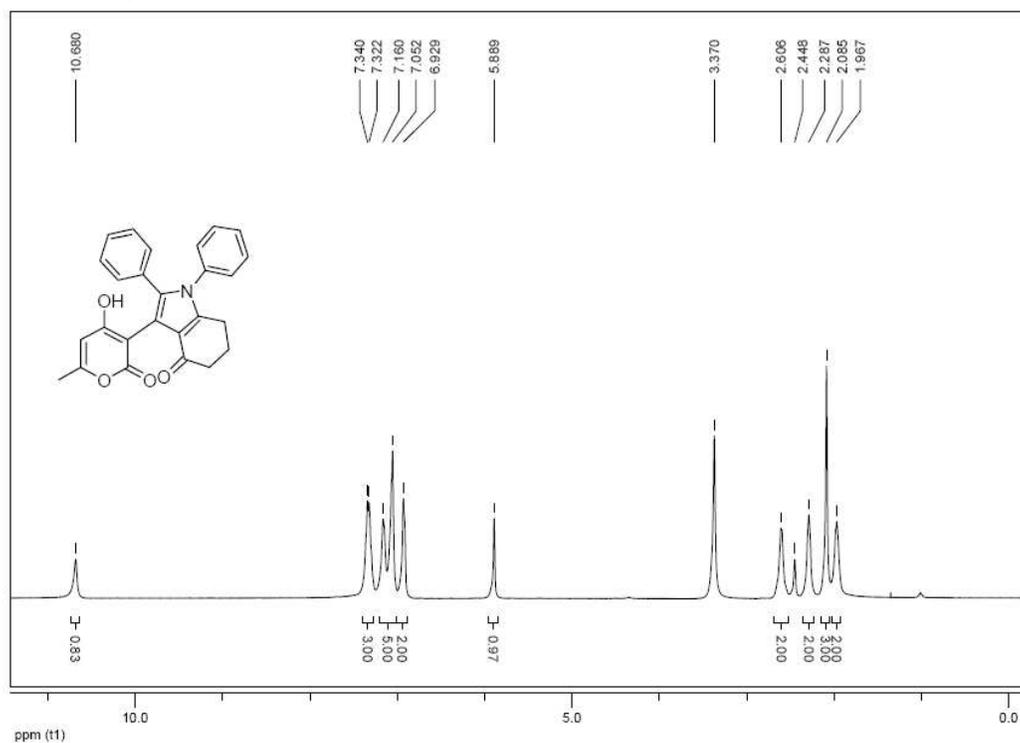
^1H NMR spectrum of compound **4**{2,1,6}



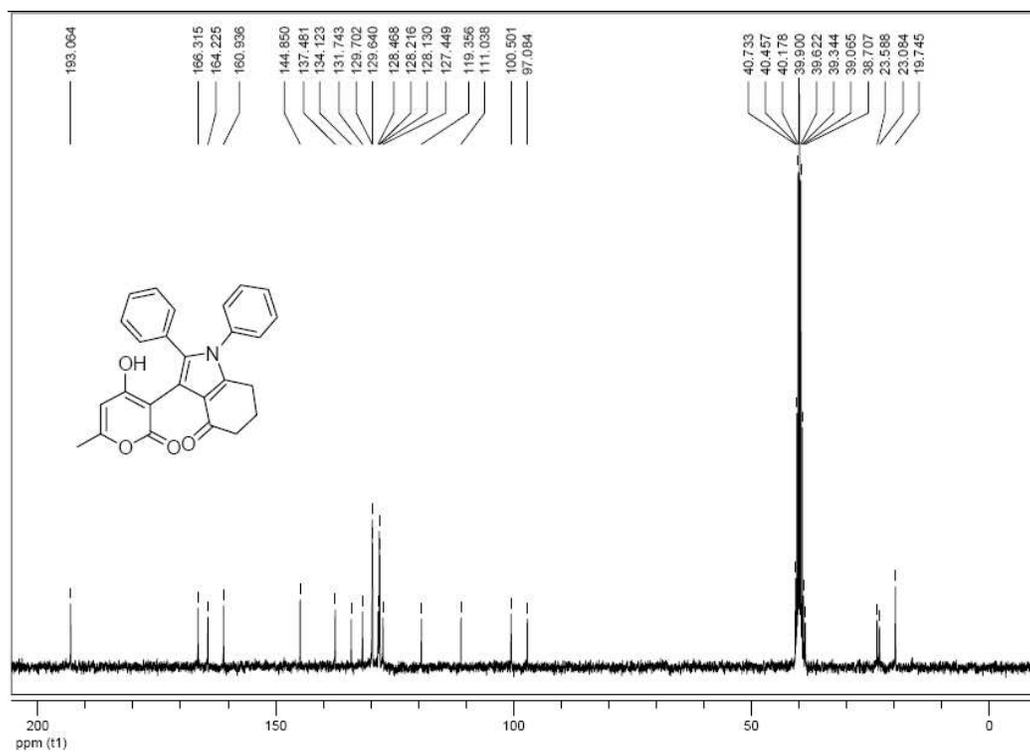
^{13}C NMR spectrum of compound **4**{2,1,6}



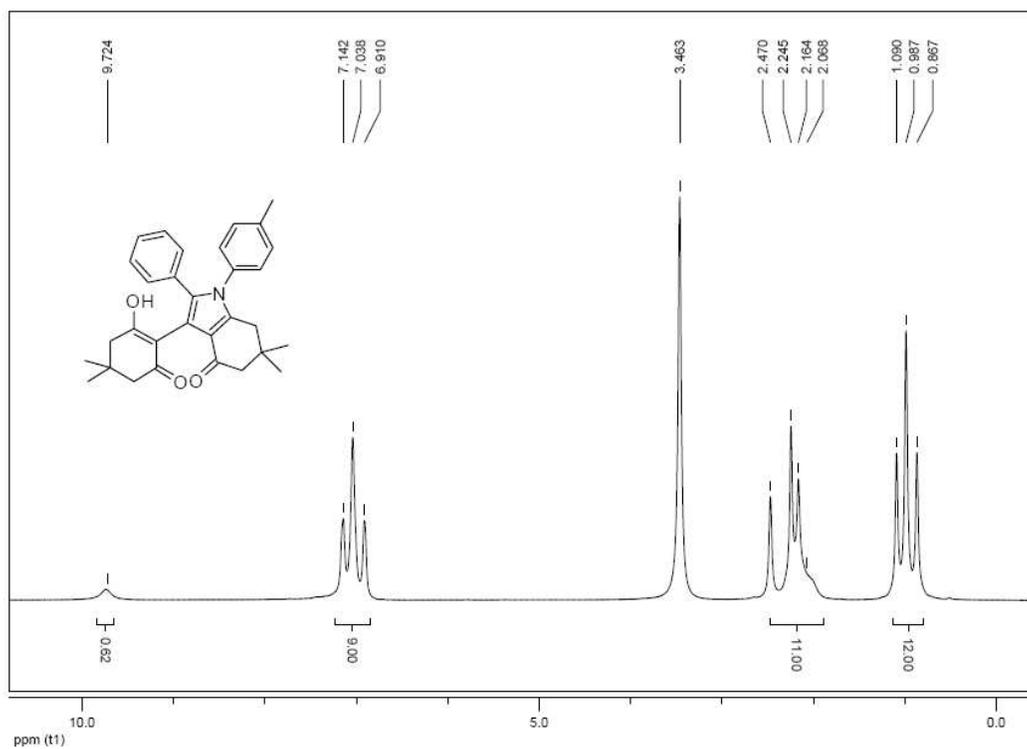
¹H NMR spectrum of compound 4{2,1,12}



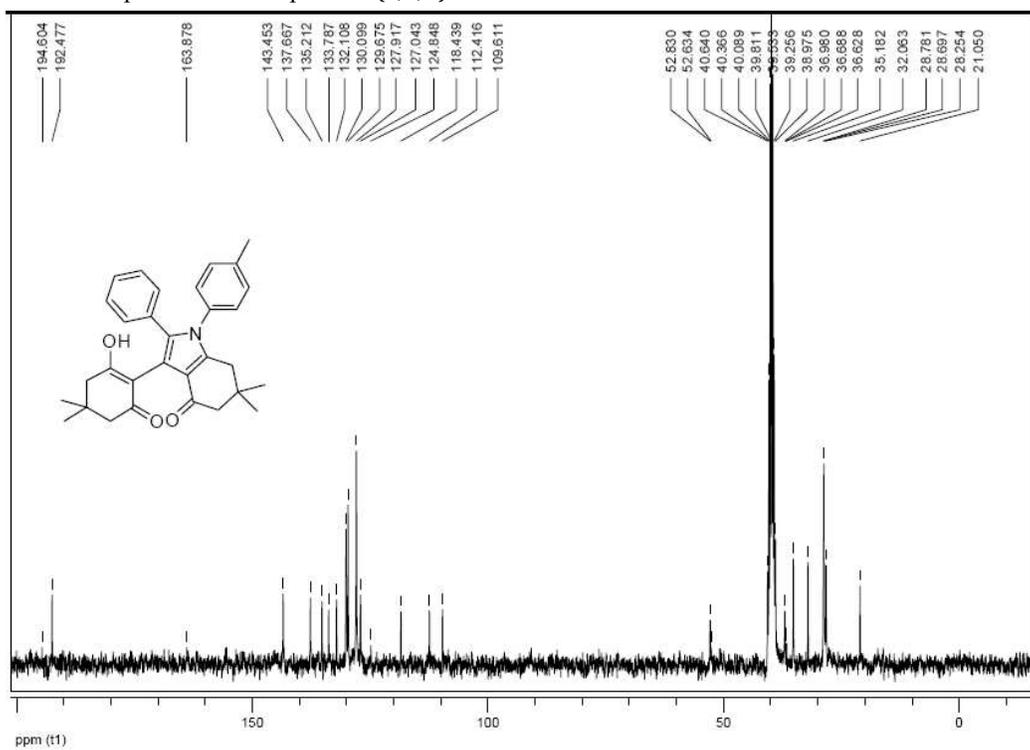
¹³C NMR spectrum of compound 4{2,1,12}



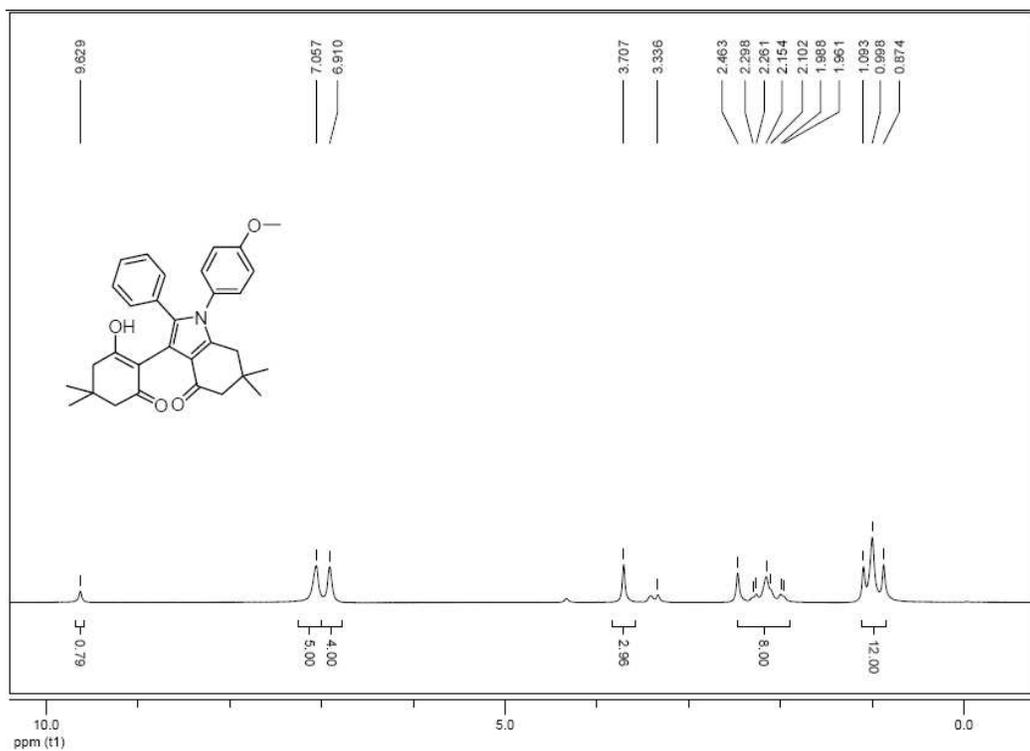
¹H NMR spectrum of compound **4{3,1,1}**



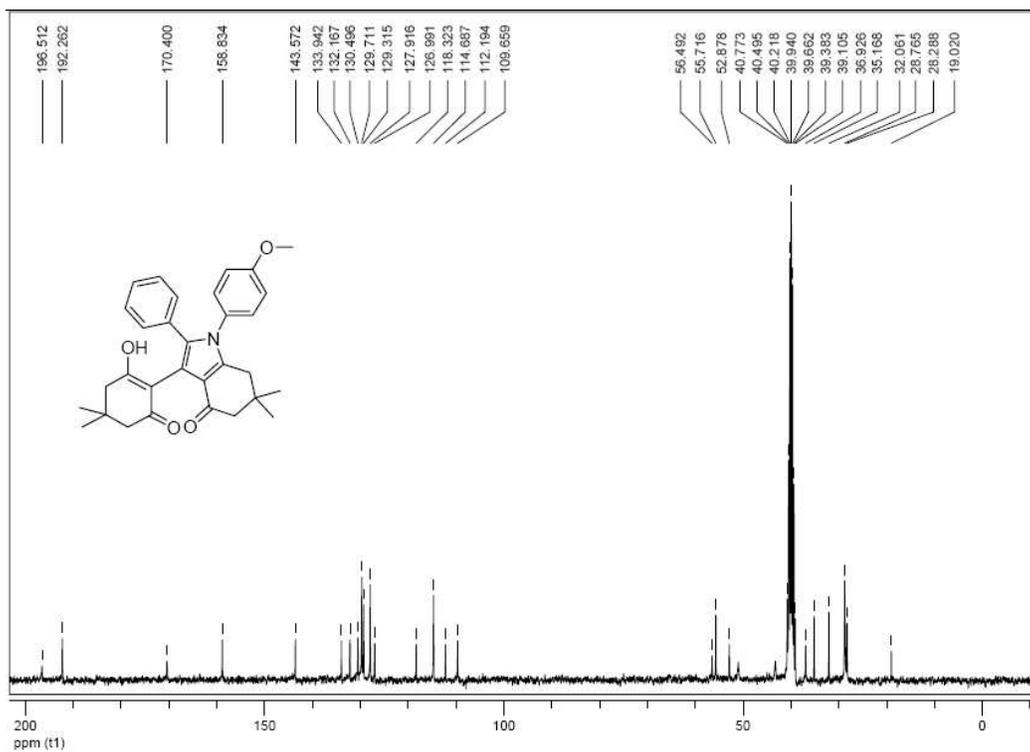
¹³C NMR spectrum of compound **4{3,1,1}**



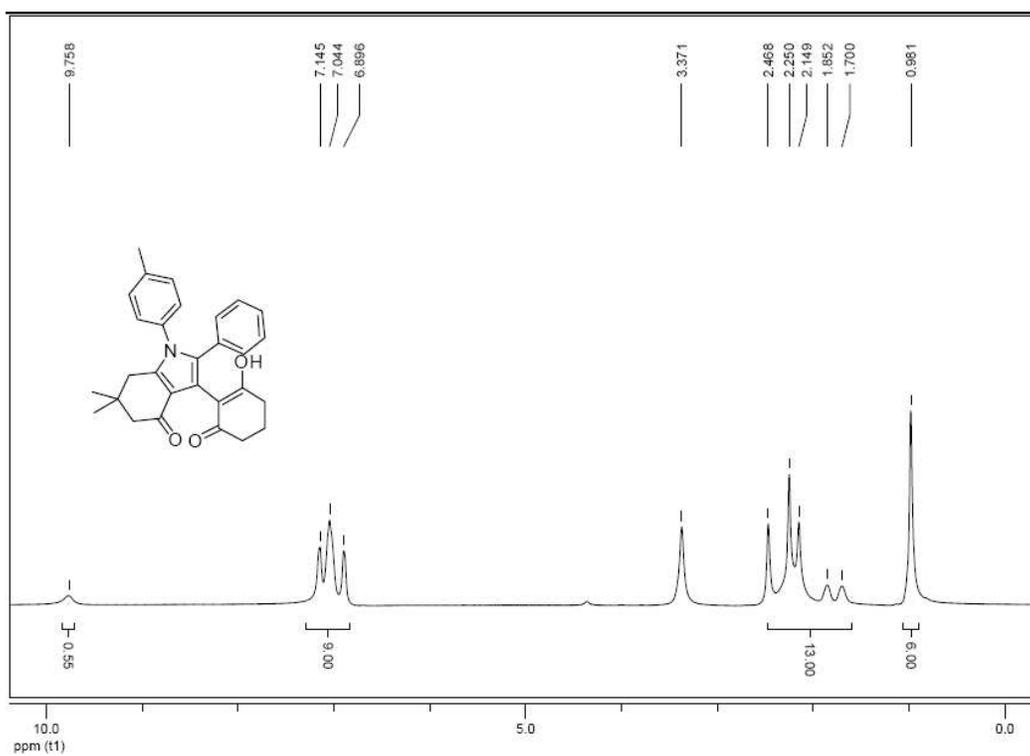
^1H NMR spectrum of compound **4**{*3,1,6*}



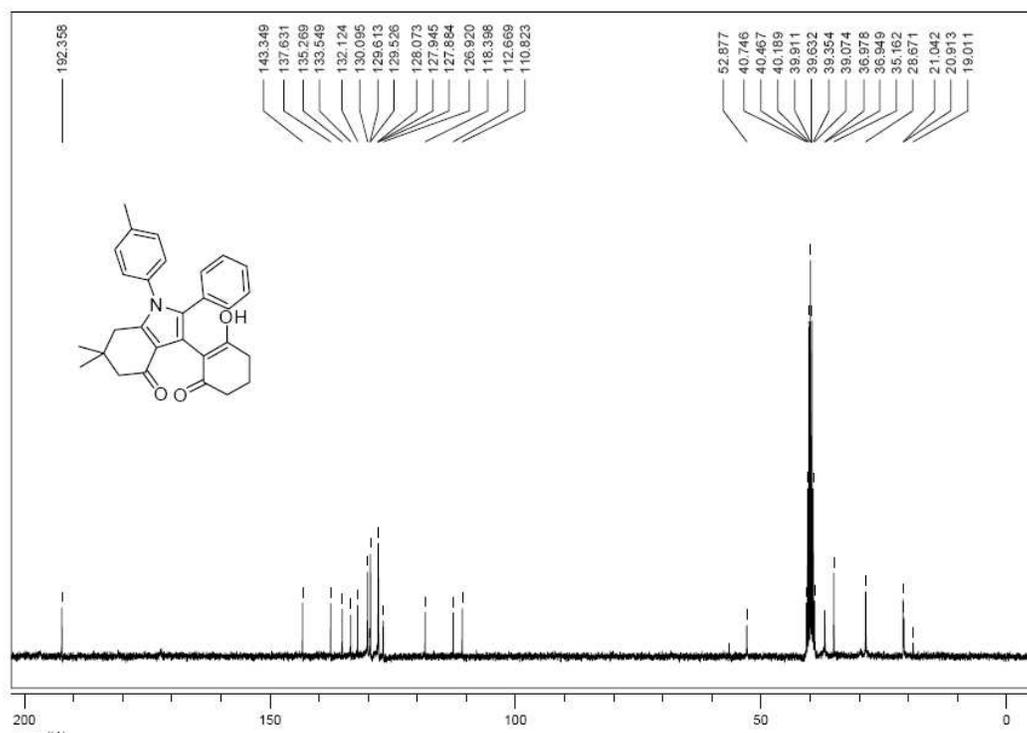
^{13}C NMR spectrum of compound **4**{*1,1,3*}



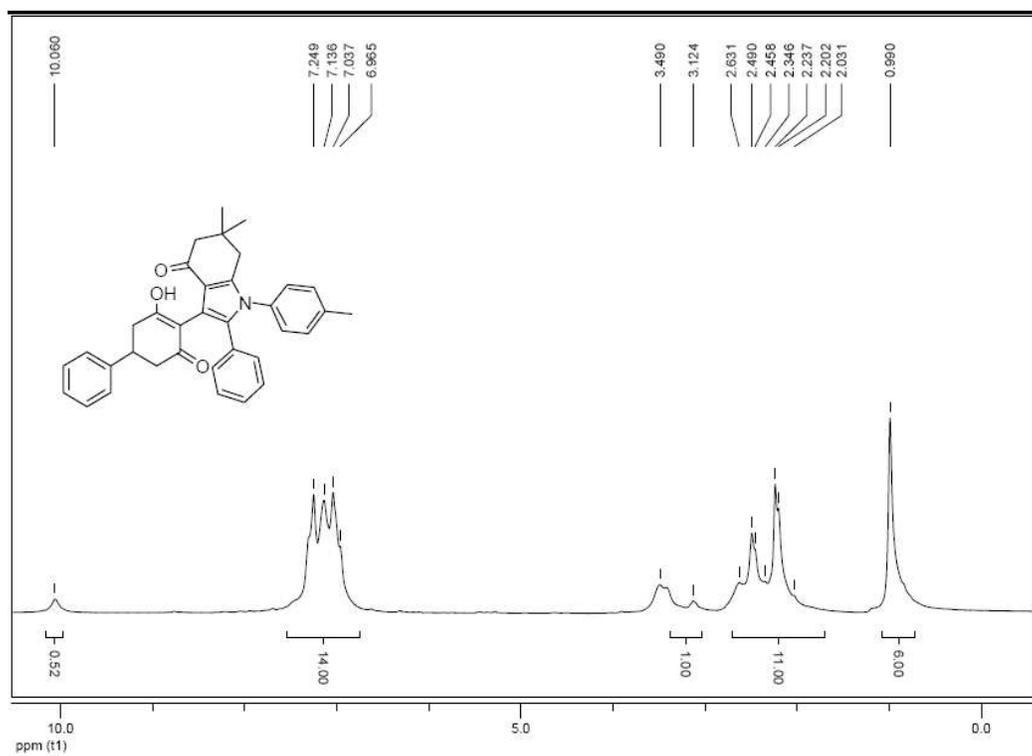
^1H NMR spectrum of compound **4{5,1,1}**



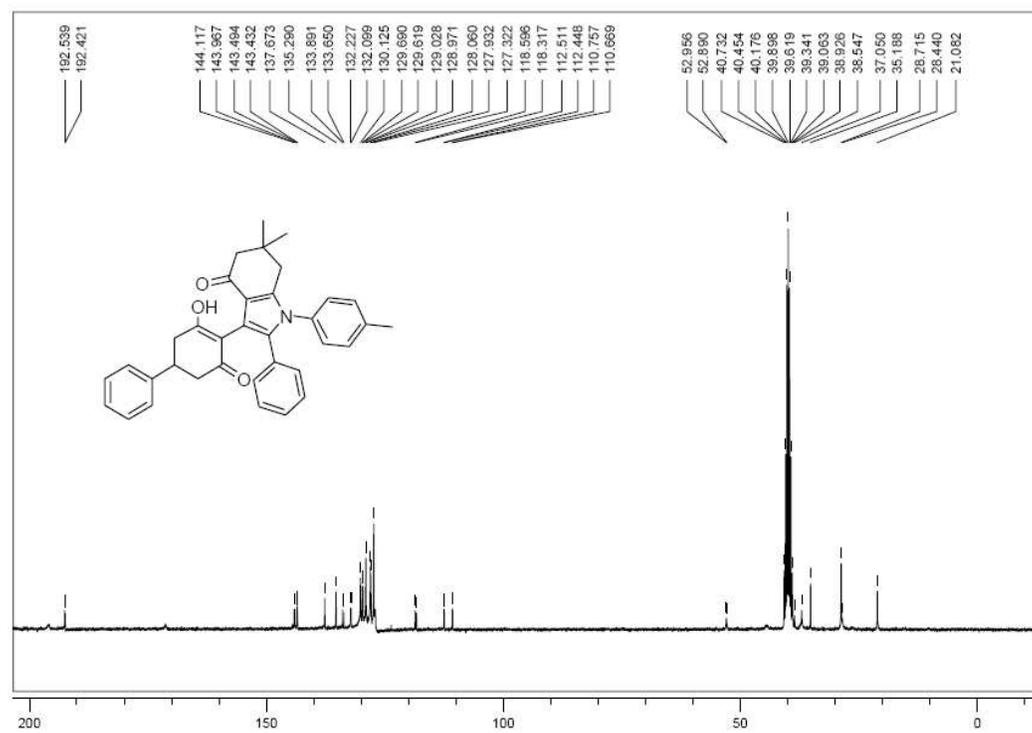
^{13}C NMR spectrum of compound **4{5,1,1}**



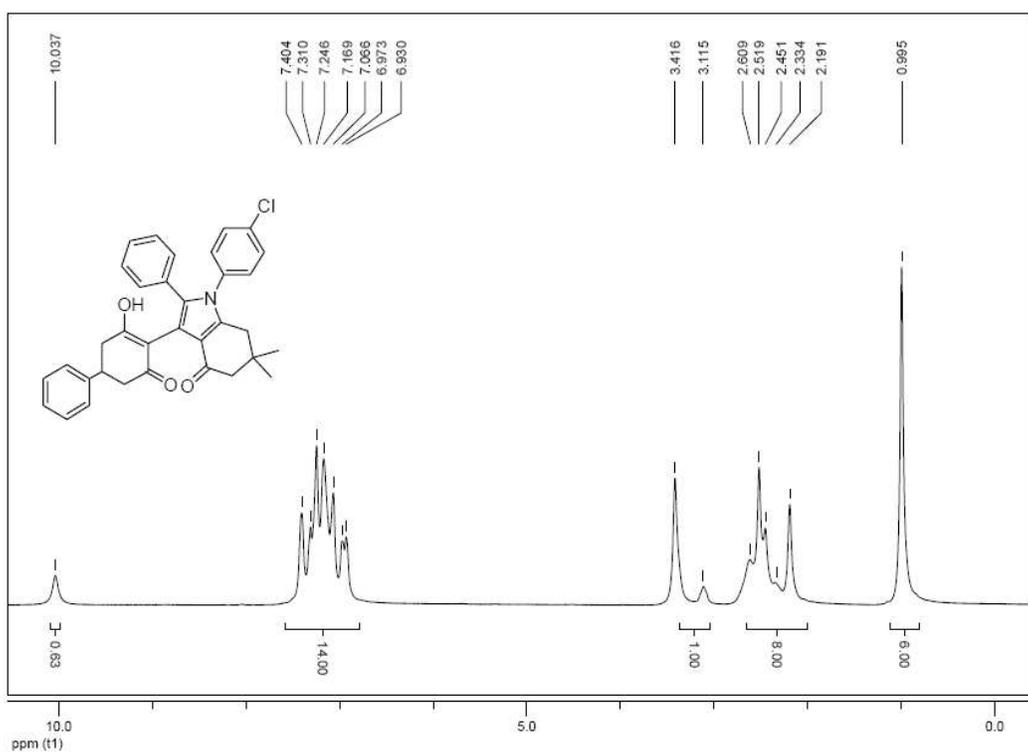
^1H NMR spectrum of compound **4{5,1,1}**



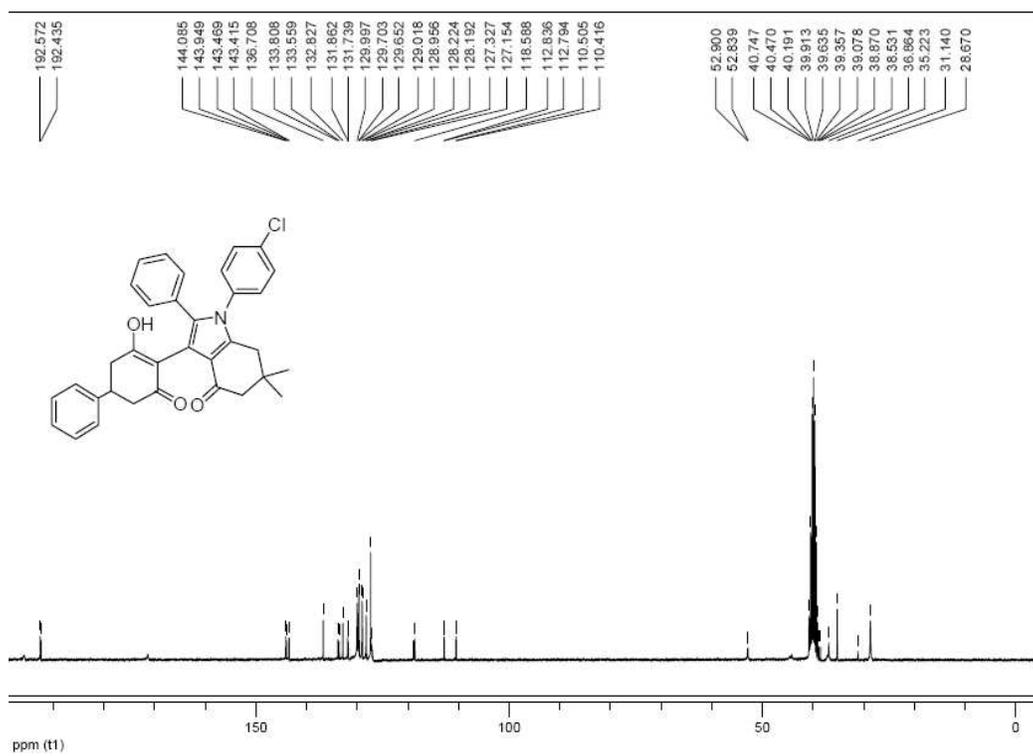
^{13}C NMR spectrum of compound **4{5,1,1}**



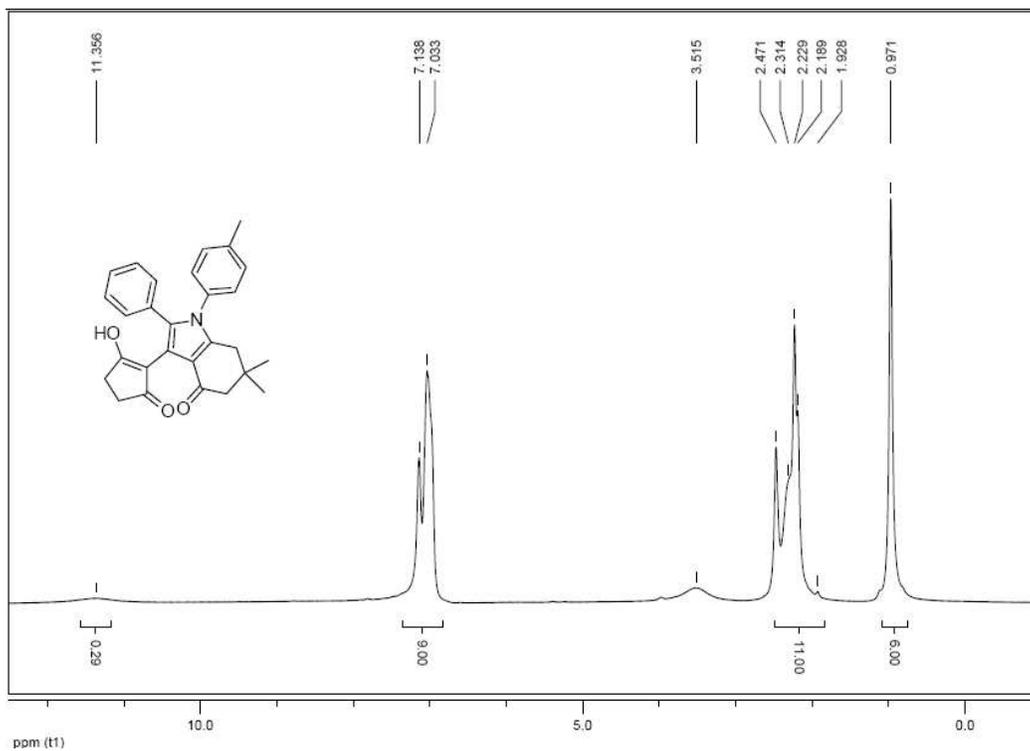
¹H NMR spectrum of compound **4{5,1,3}**



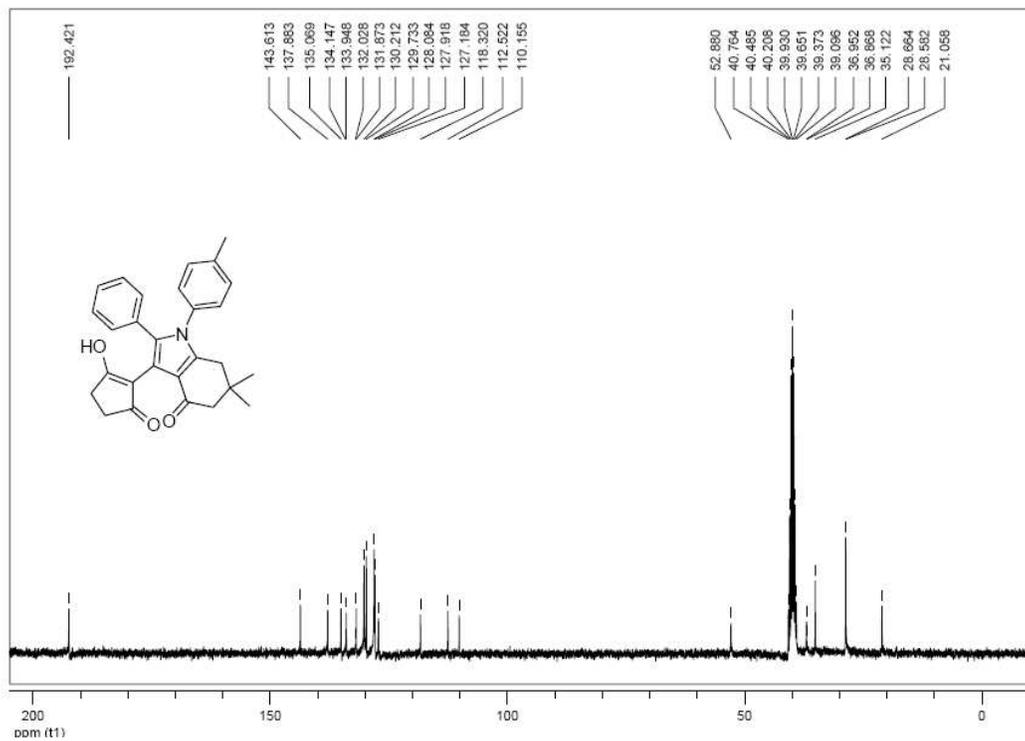
¹³C NMR spectrum of compound **4{5,1,3}**



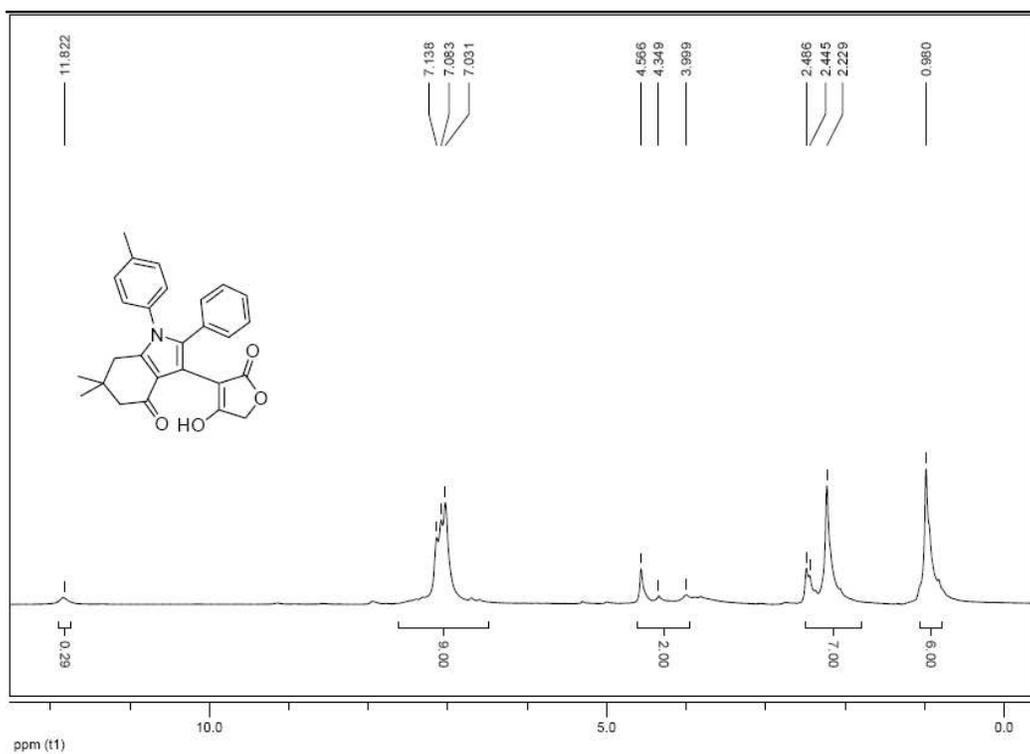
¹H NMR spectrum of compound **4{6,1,1}**



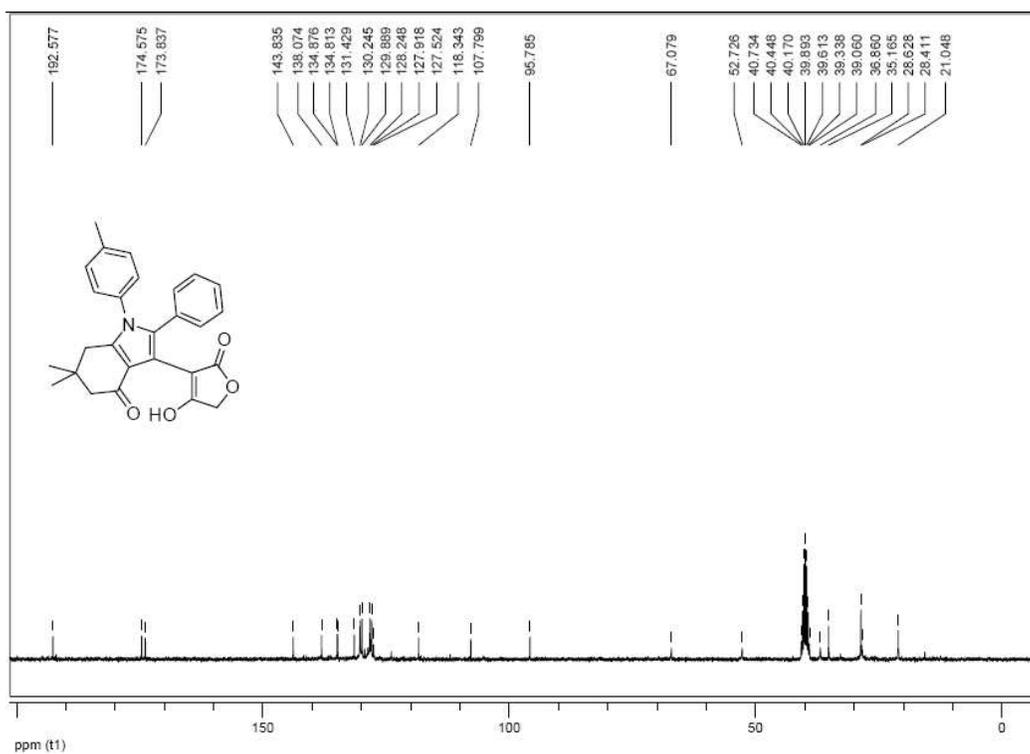
¹³C NMR spectrum of compound **4{6,1,1}**



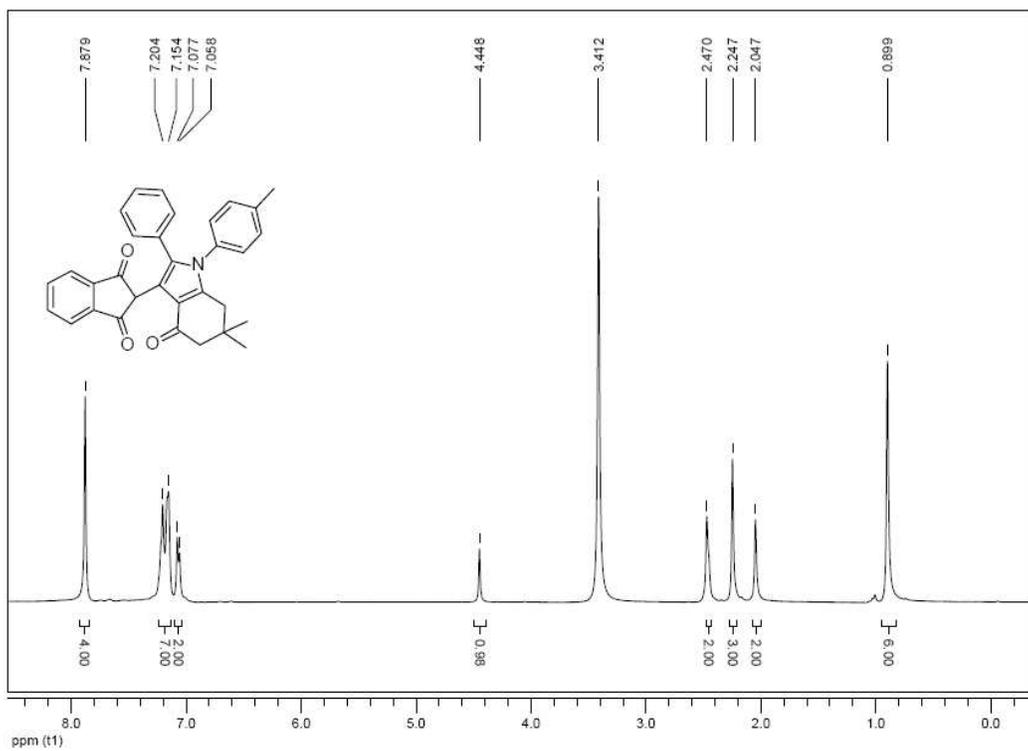
^1H NMR spectrum of compound **4**{7,1,1}



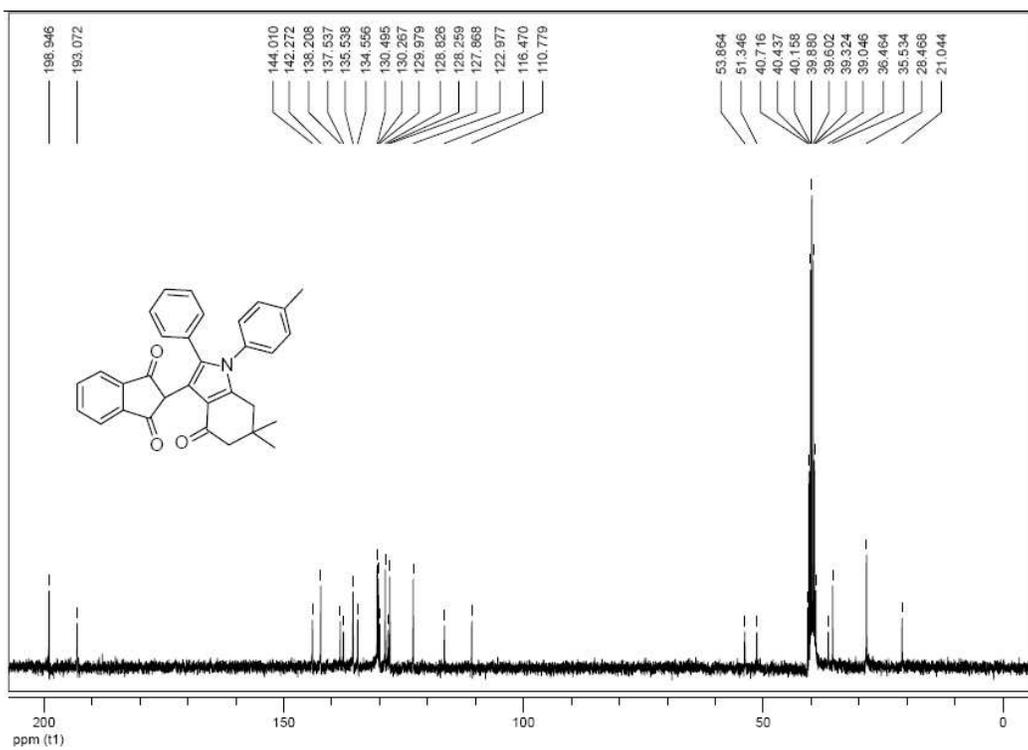
^{13}C NMR spectrum of compound **4**{7,1,1}



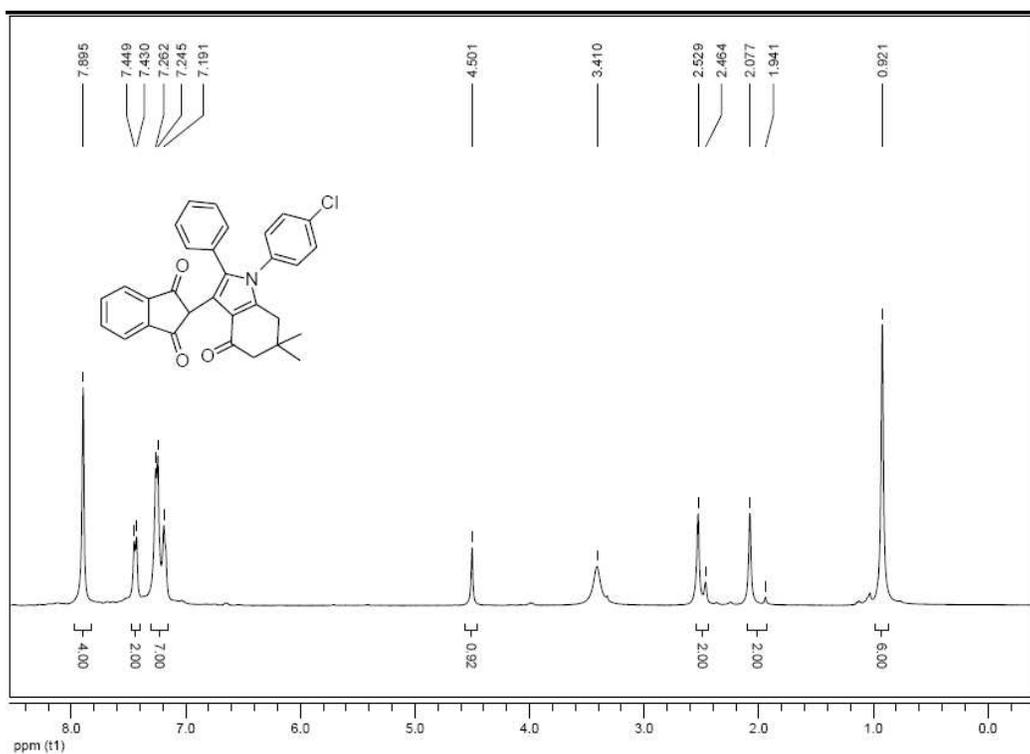
^1H NMR spectrum of compound **4{8,1,1}**



^{13}C NMR spectrum of compound **4{8,1,1}**



^1H NMR spectrum of compound **4{8,1,3}**



^{13}C NMR spectrum of compound **4{8,1,3}**

