# One-Pot, Three-Component Synthesis of a Library of Spirooxindole-Pyrimidines Catalyzed by Magnetic Nanoparticle Supported Dodecyl Benzenesulfonic Acid in Aqueous Media

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## Supporting Information

**General Information**. All solvents and chemicals were obtained commercially and were used as received. X-ray diffraction analysis was carried out using a PANalytical X'Pert Pro X-ray diffractometer. Surface morphology and particle size were studied using a Hitachi S-4800 SEM instrument. Transmission electron microscope (TEM) observation was performed using Hitachi H-7650 microscope at 80 KV. Elemental compositions were determined with a Hitachi S-4800 scanning electron microscope equipped with an INCA 350 energy dispersive spectrometer (SEM-EDS) presenting a 133 eV resolution at 5.9 keV. Melting points were determined using an X-4 apparatus and are uncorrected. IR spectra were recorded using a Bruker-TENSOR 27 spectrometer instrument. NMR spectra were taken with a Bruker DRX-500 spectrometer at 500 MHz ($^{1}$H) and 125 MHz ($^{13}$C) using DMSO-$d_6$ as the solvent with TMS as internal standard. Elemental analyses were obtained on a Vario EL III CHNOS elemental analyzer.

**Large Scale Synthesis of 4** {*1,1,1*}**.** A mixture of 5,5-dimethyl-cyclohexane-dione (7.0 g, 50 mmol), barbituric (6.5 g, 50 mmol), isatine (7.5 g, 50 mmol), and γ-$Fe_2O_3$@$SiO_2$-DDBSA (30 g) in refluxing water (250 mL) was stirred for 3.5 h. After completion of the reaction, catalyst could be placed on the side wall of the reaction vessel with the aid of an external magnet, and water was removed from the mixture to leave a residue (including the product and catalyst). Then, the product was dissolved in THF (50 ml) and the catalyst easily separated from the product by attaching an external magnet onto the reaction vessel, followed by decantation of the product solution. This solution was concentrated to generate the crude product. The crude product was washed with water (100 mL) and ethanol( 50 mL) to afford the pure **4**{*1,1,1*} as a white powder (17.4 g, yield 92%).

A B C

**Figure 1.** SEM image (A), TEM image (B) and EDS spectrum (C) of γ-$Fe_2O_3$@$SiO_2$-DDBSA.

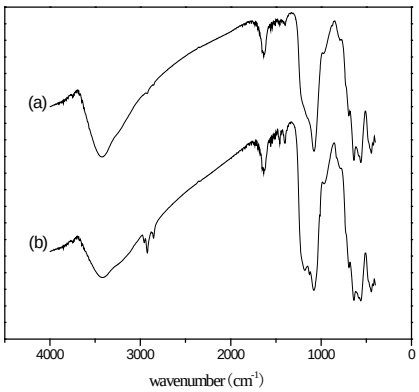



**Figure 2**. IR spectra of (a) γ-$Fe_2O_3$@$SiO_2$ and γ-$Fe_2O_3$@$SiO_2$-DDBSA.

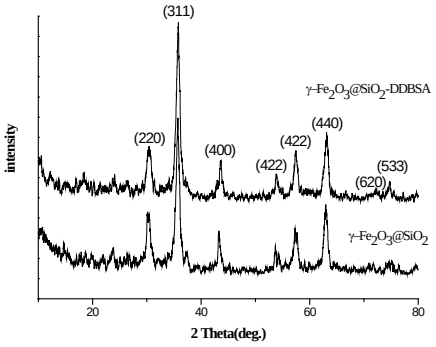



**Figure 3**. XRD pattern of γ-$Fe_2O_3$@$SiO_2$ and γ-$Fe_2O_3$@$SiO_2$-DDBSA.

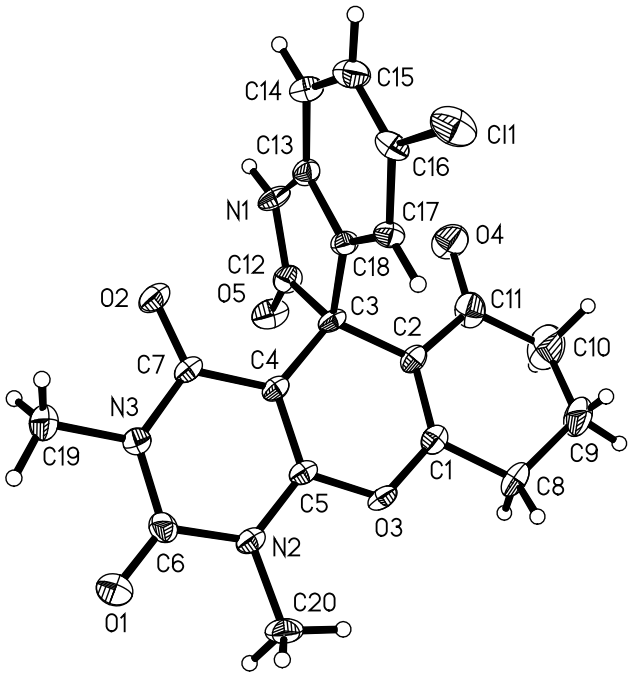



**Figure 4**. The crystal structure of product **4**{*2,2,5*} (CCDC 863099).

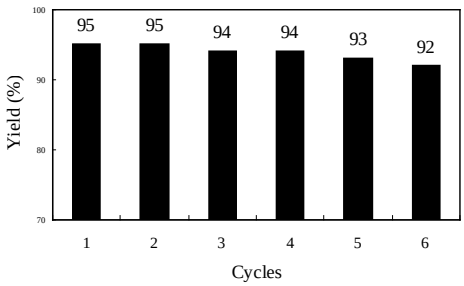
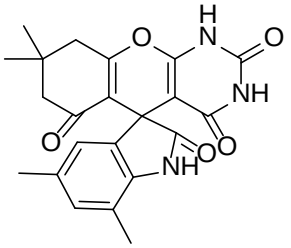



**Figure 5**. Reusability of catalyst.

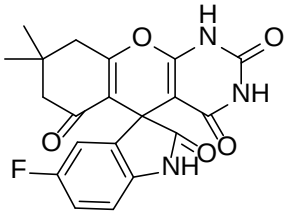
**Spectroscopic data for all new compoounds:**

5',7',8,8-Tetramethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*1*,*1*,*3*}).



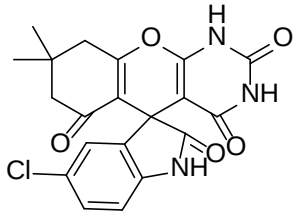
Colorless crystals, mp > 300 $^{o}$C; IR (KBr): 3319, 3244, 2958, 2929, 2813, 1720, 1662, 1624, 1558, 1485, 1305, 1228, 1184, 999, 790, 605, 563 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.97 (s, 3H, C$H_3$), 1.03 (s, 3H, C$H_3$), 2.07 and 2.16 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.11 (s, 3H, C$H_3$), 2.15 (s, 3H, C$H_3$), 2.57 and 2.64 (AB system, $J_{AB}$ = 17.5 Hz, 2H, C$H_2$), 6.58 (s, 1H, *H*Ar), 6.70 (s, 1H, *H*Ar), 10.27 (s, 1H, N*H*), 10.98 (s, 1H, N*H*), 12.16 (s,1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 16.8, 21.0, 27.2, 28.1, 32.2, 45.8, 50.9, 89.8, 113.8, 118.0, 121.4, 129.9, 130.3, 133.7, 140.4, 149.5, 153.4, 161.8, 163.4, 178.6, 195.3 ppm; Anal. Calcd for $C_{22}H_{21}N_3O_5$: C, 64.86; H, 5.20; N, 10.31. Found: C, 64.68; H, 5.01; N, 10.50.

5'-Fluoro-8,8-dimethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*1*,*1*,*4*}).



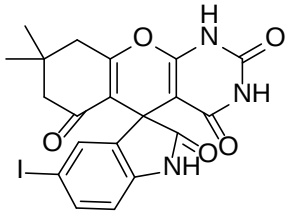
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3192, 2958, 2929, 1685, 1681, 1627, 1488, 1309, 1178, 802, 601, 561 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.99 (s, 3H, C$H_3$), 1.03 (s, 3H, C$H_3$), 2.10 and 2.19 (AB system, $J_{AB}$ = 15.5 Hz, 2H, C$H_2$), 2.56 and 2.64 (AB system, $J_{AB}$ = 17.5 Hz, 2H, C$H_2$), 6.67 (dd, *J* = 8.5, 4.0 Hz, 1H, *H*Ar), 6.87-6.92 (m, 1H, *H*Ar), 6.98 (dd, *J* = 8.5 Hz, 2.5 Hz, 1H, *H*Ar), 10.38 (s, 1H, N*H*), 10.99 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 27.4, 28.0, 29.7, 32.2, 46.0, 50.8, 89.0, 109.2 (d, $^{3}J_{FC}$ = 7.6 Hz), 111.3 (d, $^{2}J_{FC}$ = 24.5 Hz), 113.1, 114.2 (d, $^{2}J_{FC}$ = 22.7 Hz), 135.6 (d, $^{3}J_{FC}$ = 7.6 Hz), 140.6, 150.0, 154.0, 158.4 (d, $^{1}J_{FC}$ = 234.0 Hz), 162.0, 164.0, 178.2, 195.4 ppm; Anal. Calcd for $C_{20}H_{16}FN_3O_5$: C, 60.45; H, 4.06; N, 10.57. Found: C, 60.26; H, 3.91; N, 10.39.

5'-Chloro-8,8-dimethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*1*,*1*,*5*}).



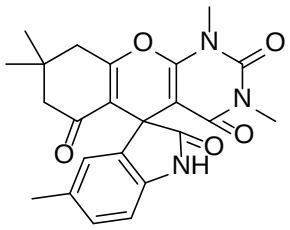

Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3365, 3176, 2956, 2837, 1685, 1681, 1624, 1558, 1477, 1436, 1307, 1236, 1190, 1164, 1002, 819, 559 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.97 (s, 3H, C$H_3$), 1.00 (s, 3H, C$H_3$), 2.10 and 2.16 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.56 and 2.60 (AB system, $J_{AB}$ = 18.0 Hz, 2H, C$H_2$), 6.70 (d, *J* = 8.0 Hz, 1H, *H*Ar), 7.10 (d, *J* = 8.0 Hz, 1H, *H*Ar), 7.12 (s, 1H, *H*Ar), 10.50 (s, 1H, N*H*), 10.99 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-*d*6, 125 MHz) δ: 27.5, 27.8, 32.2, 45.8, 50.7, 88.9, 110.1, 113.0, 123.6, 125.2, 128.1, 136.1, 143.3, 149.7, 154.0, 162.1, 164.1, 178.0, 195.6 ppm; Anal. Calcd for $C_{20}H_{16}ClN_3O_5$: C, 58.05; H, 3.90; N, 10.15. Found: C, 57.90; H, 4.08; N, 10.01.

5'-Iodo-8,8-dimethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*1*,*1*,*7*}).



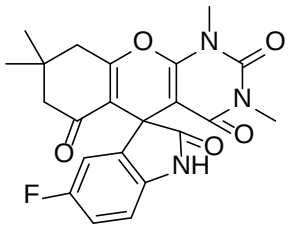

Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3197, 2958, 2821, 1710, 1681, 1616, 1558, 1473, 1307, 1234, 1186, 1164, 1002, 815, 690, 563 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.99 (s, 3H, C$H_3$), 1.02 (s, 3H, C$H_3$), 2.10 and 2.14 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.60 (s, 2H, C$H_2$), 6.58 (d, *J* = 8.0 Hz, 1H, *H*Ar), 7.32 (s, 1H, *H*Ar), 7.40 (d, *J* = 8.0 Hz, 1H, *H*Ar), 10.46 (s, 1H, N*H*), 10.88 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 27.4, 27.8, 32.2, 45.6, 50.7, 83.8, 88.7, 111.5, 112.9, 131.5, 136.7, 136.8, 144.1, 150.2, 154.5, 162.2, 164.3, 177.9, 195.7 ppm; Anal. Calcd for $C_{20}H_{16}IN_3O_5$: C, 47.54; H, 3.19; N, 8.32. Found: C, 47.72; H, 2.99; N, 8.51.

1,3,5',8,8-pentamethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*1*,*2*,*2*}).



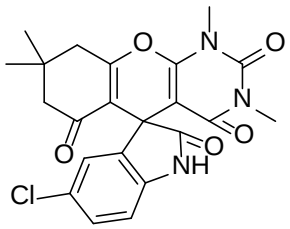
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3209, 2956, 2933, 2869, 1689, 1647, 1624, 1492, 1352, 1315, 1215, 1186, 1107, 1082, 966, 812, 748, 700, 684, 509 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.99 (s, 3H, C$H_3$), 1.04 (s, 3H, C$H_3$), 2.13 (s, 3H, C$H_3$), 2.10 and 2.18 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.65 and 2.70 (AB system, $J_{AB}$ = 18.0 Hz, 2H, C$H_2$), 3.02 (s, 3H, C$H_3$), 3.40 (s, 3H, C$H_3$), 6.59 (d, $J$ = 8.0 Hz, 1H, *H*Ar), 6.77 (s, 1H, *H*Ar), 6.88 (d, $J$ = 8.0 Hz, 1H, *H*Ar), 10.27 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 21.1, 28.0, 28.1, 29.7, 32.2, 46.3, 50.9, 90.0, 108.7, 113.7, 124.1, 128.6, 129.9, 134.0, 142.0, 150.0, 152.3, 159.8, 163.3, 178.1, 195.3 ppm; Anal. Calcd for $C_{23}H_{23}N_3O_5$: C, 65.55; H, 5.50; N, 9.97. Found: C, 65.38; H, 5.69; N, 10.16.

5'-Fluoro-1,3,8,8-tetramethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*1*,*2*,*4*}).



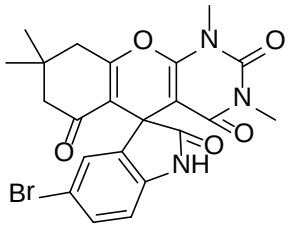
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3654, 3388, 3226, 2964, 1693, 1662, 1631, 1483, 1352, 1319, 1217, 1184, 1103, 966, 802, 754, 680, 599 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.00 (s, 3H, C$H_3$), 1.04 (s, 3H, C$H_3$), 2.14 and 2.22 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.66 and 2.72 (AB system, $J_{AB}$ = 16.5 Hz, 2H, C$H_2$), 3.03 (s, 3H, C$H_3$), 3.40 (s, 3H, C$H_3$), 6.70 (dd, $J$ = 8.0, 4.0 Hz, 1H, *H*Ar), 6.88-6.95 (m, 2H, *H*Ar), 10.43 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 27.4, 27.9, 28.1, 29.7, 32.2, 46.7, 50.8, 89.5, 109.3 (d, $^{3}J_{FC}$ = 7.6 Hz), 111.4 (d, $^{2}J_{FC}$ = 24.5 Hz), 113.5, 114.4 (d, $^{2}J_{FC}$ = 22.7 Hz), 135.4 (d, $^{3}J_{FC}$ = 7.6 Hz), 140.6, 150.0, 152.5, 158.2 (d, $^{1}J_{FC}$ = 233.8 Hz), 160.0, 163.7, 178.2, 195.4 ppm; Anal. Calcd for $C_{22}H_{20}FN_3O_5$: C, 62.11; H, 4.74; N, 9.88. Found: C, 61.92; H, 4.90; N, 10.06.

5'-Chloro-1,3,8,8-tetramethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*1*,*2*,*5*}).



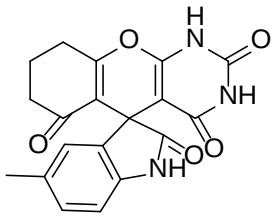

Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3249, 2960, 2929, 2879, 1700, 1689, 1678, 1631, 1477, 1442, 1357, 1311, 1213, 1178, 1089, 821, 773, 677 cm$^{-1}$; $^{1}$H NMR (DMSO-*d*6, 500 MHz) δ: 0.99 (s, 3H, C*H*$_3$), 1.02 (s, 3H, C*H*$_3$), 2.14 and 2.19 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C*H*$_2$), 2.68 (s, 2H, C*H*$_2$), 3.02 (s, 3H, C*H*$_3$), 3.37 (s, 3H, C*H*$_3$), 6.85 (d, *J* = 8.0 Hz, 1H, *H*Ar), 7.20 (s, 1H, *H*Ar), 7.24 (d, *J* = 8.0 Hz, 1H, *H*Ar), 10.54 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 27.5, 28.7, 28.1, 29.7, 32.2, 46.4, 50.8, 89.4, 110.8, 112.8, 113.1, 126.4, 131.1, 136.3, 143.8, 150.0, 152.6, 160.0, 163.8, 177.8, 195.5 ppm; Anal. Calcd for $C_{22}H_{20}ClN_3O_5$: C, 59.80; H, 4.56; N, 9.51. Found: C, 59.96; H, 4.40; N, 9.70.

5'-Bromo-1,3,8,8-tetramethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*1*,*2*,*6*}).



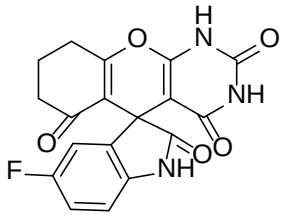

Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3257, 3043, 2964, 2871, 1689, 1636, 1616, 1475, 1355, 1311, 1213, 1176, 1087, 966, 821, 773, 754, 486 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.00 (s, 3H, C*H*$_3$), 1.03 (s, 3H, C*H*$_3$), 2.16 and 2.20 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C*H*$_2$), 2.69 (s, 2H, C*H*$_2$), 3.02 (s, 3H, C*H*$_3$), 3.39 (s, 3H, C*H*$_3$), 6.69 (d, *J* = 8.0 Hz, 1H, *H*Ar), 7.21 (s, 1H, *H*Ar), 7.25 (d, *J* = 8.0 Hz, 1H, *H*Ar), 10.55 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 27.5, 27.8, 28.1, 29.7, 32.2, 46.4, 50.8, 89.4, 110.8, 112.8, 113.1, 126.4, 131.1, 136.3, 143.8, 150.0, 152.6, 160.0, 163.8, 177.8, 195.5 ppm; Anal. Calcd for $C_{22}H_{20}BrN_3O_5$: C, 54.33; H, 4.15; N, 8.64. Found: C, 54.25; H, 3.96; N, 8.80.

5'-Methyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*2*,*1*,*2*}).



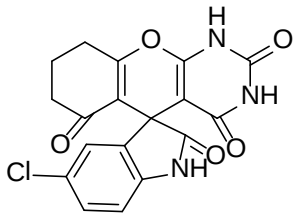
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3627, 3369, 3126, 2923, 2852, 1735, 1685, 1662, 1622, 1492, 1427, 1325, 1303, 1224, 1186, 1110, 1087, 989, 825, 592 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.80-2.00 (m, 2H, C$H_2$), 2.15 (s, 3H, C$H_3$), 2.20-2.30 (m, 2H, C$H_2$), 2.65-2.80 (m, 2H, C$H_2$), 6.58 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 6.79 (s, 1H, *H*Ar), 6.87 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 10.22 (s, 1H, N*H*), 10.96 (s, 1H, N*H*), 12.13 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.1, 21.1, 27.2, 37.3, 45.6, 89.7, 108.6, 114.7, 124.1, 128.5, 129.9, 134.2, 141.8, 149.4, 153.3, 161.8, 165.3, 178.2, 195.4 ppm; Anal. Calcd for $C_{19}H_{15}N_3O_5$: C, 62.46; H, 4.14; N, 11.50. Found: C, 62.28; H, 3.95; N, 11.76.

5'-Fluoro-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*2,1,4*}).



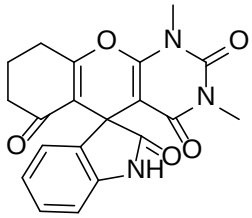
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3369, 3126, 3072, 2923, 1680, 1662, 1624, 1488, 1326, 1305, 1228, 1188, 808, 696, 599 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.88-1.95 (m, 2H, C$H_2$), 2.18-2.30 (m, 2H, C$H_2$), 2.64-2.75 (m, 2H, C$H_2$), 6.67 (dd, $J$ = 7.5, 4.0 Hz, 1H, *H*Ar), 6.87-6.99 (m, 2H, *H*Ar), 2.69 (dd, $J$ = 8.5 Hz, $J$ = 1.5 Hz, 2H, *H*Ar), 10.39 (s, 1H, N*H*), 11.03 (s, 1H, N*H*), 12.00 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-*d*6, 125 MHz) δ: 20.2, 27.3, 37.2, 46.1, 89.2, 109.2 (d, $^{3}J_{FC}$ = 7.6 Hz), 111.6 (d, $^{2}J_{FC}$ = 24.5 Hz) , 114.1, 114.3 (d, $^{2}J_{FC}$ = 22.7 Hz), 135.6 (d, $^{3}J_{FC}$ = 7.6 Hz), 140.6, 150.0, 153.5, 158.5 (d, $^{1}J_{FC}$ = 233.7 Hz), 162.0, 165.8, 178.3, 195.6 ppm; Anal. Calcd for $C_{18}H_{12}FN_3O_5$: C, 58.54; H, 3.28; N, 11.38. Found: C, 58.72; H, 3.09; N, 11.56.

5'-Chloro-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*2,1,5*}).



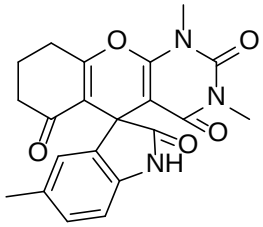
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3323, 3138, 2962, 2813, 1710, 1681, 1616, 1529, 1481, 1434, 1363, 1326, 1303, 1236, 1191, 997, 812, 559 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.89-1.95 (m, 2H, C$H_2$), 2.19-2.30 (m, 2H, C$H_2$), 2.66-2.75 (m, 2H, C$H_2$), 6.70 (d, $J$ = 8.0 Hz, 1H, $H$Ar), 7.11 (dd, $J$ = 8.0 Hz, $J$ = 2.0 Hz, 1H, $H$Ar), 7.14 (d, $J$ = 2.0 Hz, 1H, $H$Ar), 10.50 (s, 1H, N$H$), 11.02 (s, 1H, N$H$), 12.20 (s, 1H, N$H$) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.0, 27.2, 37.2, 45.8, 89.1, 110.1, 114.1, 123.8, 125.2, 128.1, 136.1, 143.3, 149.5, 153.6, 162.0, 166.0, 178.1, 195.6 ppm; Anal. Calcd for $C_{18}H_{12}ClN_3O_5$: C, 56.04; H, 3.14; N, 10.89. Found: C, 55.86; H, 2.05; N, 11.08.

1,3-Dimethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*2,2,1*}).



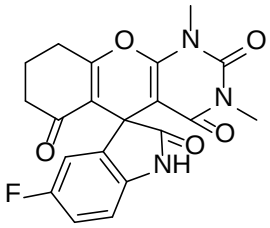
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3307, 3051, 2960, 2916, 2877, 1689, 1681, 1620, 1558, 1485, 1471, 1325, 1330, 1305, 1236, 1211, 1103, 1083, 968, 775, 752, 677, 617 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.85-2.00 (m, 2H, C$H_2$), 2.20-2.31 (m, 2H, C$H_2$), 2.75-2.87 (m, 2H, C$H_2$), 3.01 (s, 3H, C$H_3$), 3.40 (s, 3H, C$H_3$), 6.72 (d, $J$ = 7.5 Hz, 1H, $H$Ar), 6.77 (t, 1H, $J$ = 7.5 Hz, $H$Ar), 6.97 (d, $J$ = 7.5 Hz, 1H, $H$Ar), 7.07 (t, $J$ = 7.5 Hz, 1H, $H$Ar), 10.37 (s, 1H, N$H$) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.1, 27.1, 28.1, 29.7, 37.4, 46.4, 90.1, 108.9, 114.7, 121.2, 123.6, 128.3, 144.3, 150.0, 152.1, 159.8, 165.1, 178.1, 195.4 ppm; Anal. Calcd for $C_{20}H_{17}N_3O_5$: C, 63.32; H, 4.52; N, 11.08. Found: C, 63.15; H, 4.70; N, 10.92.

1,3,5'-Trimethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*2,2,2*}).



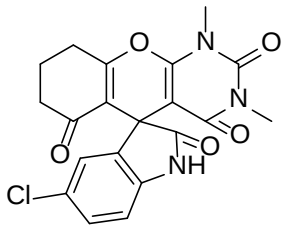
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3311, 2956, 2918, 2868, 2850, 1681, 1639, 1624, 1558, 1492, 1305, 1213, 1161, 1083, 970, 823, 790, 754 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.85-2.00 (m, 2H, C$H_2$), 2.14 (s, 3H, C$H_3$), 2.20-2.30 (m, 2H, C$H_2$), 2.75-2.85 (m, 2H, C$H_2$), 3.01 (s, 3H, C$H_3$), 3.17 (s, 3H, C$H_3$), 6.60 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 6.79 (s, 1H, *H*Ar), 6.88 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 10.28 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.1, 21.1, 28.1, 29.7, 37.4, 46.4, 90.1, 108.6, 114.7, 124.3, 128.6, 129.9, 134.0, 142.0, 150.0, 152.1, 159.8, 165.1, 178.1, 195.4 ppm; Anal. Calcd for $C_{21}H_{19}N_3O_5$: C, 64.12; H, 4.87; N, 10.68. Found: C, 63.95; H, 5.06; N, 10.86.

5'-Fluoro-1,3-dimethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*2*,*2*,*4*}).



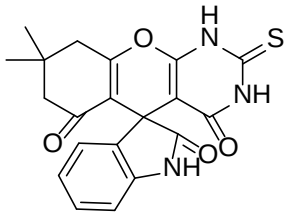
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3512, 3319, 1712, 1685, 1681, 1624, 1485, 1359, 1307, 1217, 1107, 1078, 970, 796, 596 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.90-1.95 (m, 2H, C$H_2$), 2.20-2.31 (m, 2H, C$H_2$), 2.71-2.82 (m, 2H, C$H_2$), 3.01 (s, 3H, C$H_3$), 3.39 (s, 3H, C$H_3$), 6.68 (q, $J$ = 4.0 Hz, 1H, *H*Ar), 6.87-6.94 (m, 2H, *H*Ar), 10.41(s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.0, 27.2, 28.1, 29.7, 37.3, 46.8, 89.6, 109.2 (d, $^{3}J_{FC}$ = 7.6 Hz), 111.7 (d, $^{2}J_{FC}$ = 24.5 Hz) , 114.2, 114.4 (d, $^{2}J_{FC}$ = 22.7 Hz), 135.5 (d, $^{3}J_{FC}$ = 37.6 Hz), 140.6, 150.0, 152.3, 158.2 (d, $^{1}J_{FC}$ = 235.6 Hz), 159.9, 165.5, 178.2, 195.6 ppm; Anal. Calcd for $C_{20}H_{16}FN_3O_5$: C, 60.45; H, 4.06; N, 10.57. Found: C, 60.26; H, 3.88; N, 10.75.

5'-Chloro-1,3-dimethyl-8,9-dihydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2,2',4,6(1*H*,3*H*,7*H*)-tetraone (**4**{*2*,*2*,*5*}).



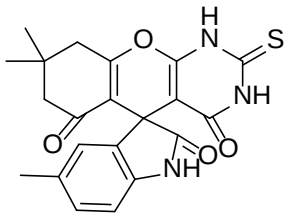

Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3336, 2960, 2871, 1716, 1685, 1639, 1624, 1477, 1307, 1215, 1186, 1093, 970, 775, 752 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.89-2.00 (m, 2H, C$H_2$), 2.20-2.30 (m, 2H, C$H_2$), 2.75-2.85 (m, 2H, C$H_2$), 3.03 (s, 3H, C$H_3$), 3.40 (s, 3H, C$H_3$), 6.72 (d, $J$ = 8.0 Hz, 1H, *H*Ar), 7.11 (s, 1H, *H*Ar), 7.13 (d, $J$ = 8.0 Hz, 1H, *H*Ar), 10.54 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.1, 21.1, 27.1, 28.1, 29.7, 37.4, 46.4, 90.1, 108.6, 114.7, 124.3, 128.6, 129.9, 134.0, 142.0, 150.0, 152.1, 159.8, 165.1, 178.1, 195.4 ppm; Anal. Calcd for $C_{20}H_{16}ClN_3O_5$: C, 58.05; H, 3.90; N, 10.15. Found: C, 57.88; H, 4.08; N, 9.96.

8,8-dimethyl-2-thioxo-2,3,8,9-tetrahydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2',4,6(1*H*,7*H*)-trione (**4**{*1,3,1*}).



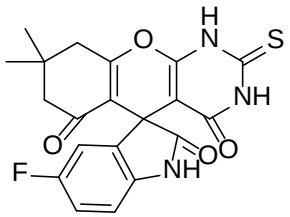

Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3207, 2956, 2871, 1681, 1618, 1556, 1471, 1311, 1188, 1164, 1041, 752, 680, 569 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.97 (s, 3H, C$H_3$), 1.03 (s, 3H, C$H_3$), 2.17 and 2.20 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.55 and 2.66 (AB system, $J_{AB}$ = 13.0 Hz, 2H, C$H_2$), 6.70-7.22 (m, 4H, *H*Ar), 10.40 (s, 1H, N*H*), 12.32 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 27.1, 28.1, 29.7, 32.2, 45.5, 50.8, 94.1, 109.1, 113.3, 121.4, 123.4, 128.5, 133.5, 144.2, 153.3, 159.7, 163.8, 174.2, 177.9, 195.4 ppm; Anal. Calcd for $C_{20}H_{17}N_3O_4S$: C, 60.75; H, 4.33; N, 10.63. Found: C, 60.58; H, 4.16; N, 10.80.

5',8,8-Trimethyl-2-thioxo-2,3,8,9-tetrahydrospiro[chromeno [2,3-*d*]pyrimidine-5,3'-indoline]-2',4,6(1*H*,7*H*)-trione (**4**{*1,3,2*}).



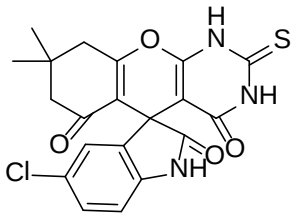
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3327, 2956, 2929, 2869, 1710, 1678, 1622, 1560, 1556, 1492, 1334, 1307, 1203, 1176, 1157, 1006, 1041, 813, 684, 569 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.99 (s, 3H, C$H_3$), 1.03 (s, 3H, C$H_3$), 2.10 and 2.18 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.15 (s, 3H, C$H_3$), 2.58 and 2.64 (AB system, $J_{AB}$ = 17.5 Hz, 2H, C$H_2$), 6.61 (d, $J$ = 8.0 Hz, 1H, *H*Ar), 6.82 (s, 1H, *H*Ar), 6.89 (d, $J$ = 8.0 Hz, 1H, *H*Ar), 10.29 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 21.1, 27.3, 28.0, 32.2, 45.6, 50.9, 94.3, 108.7, 113.4, 124.1, 128.7, 130.2, 133.5, 142.0, 152.9, 159.5, 163.5, 174.0, 177.7, 195.3 ppm; Anal. Calcd for $C_{21}H1_9N_3O_4S$: C, 61.60; H, 4.68; N, 10.26. Found: C, 61.42; H, 4.85; N, 10.28.

5'-Fluoro-8,8-dimethyl-2-thioxo-2,3,8,9-tetrahydrospiro [chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2',4,6(1*H*,7*H*)-trione (**4**{*1,3,4*}).



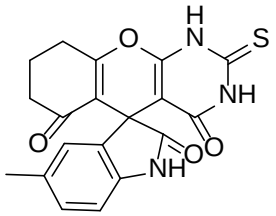
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3346, 2960, 2931, 2871, 1700, 1681, 1622, 1560, 1556, 1488, 1311, 1191, 1176, 1159, 1041, 813, 800, 684, 599 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.99 (s, 3H, C$H_3$), 1.03 (s, 3H, C$H_3$), 2.10 and 2.19 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.55 and 2.64 (AB system, $J_{AB}$ = 17.5 Hz, 2H, C$H_2$), 6.67 (dd, $J$ = 8.0, 4.0 Hz, 1H, *H*Ar), 6.89-6.93 (m, 1H, *H*Ar), 7.07 (d, $J$ = 7.0 Hz, 1H, *H*Ar), 10.46 (s, 1H, N*H*), 12.44 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 27.4, 28.0, 32.2, 46.0, 50.8, 93.7, 109.3 (d, $^{3}J_{FC}$ = 7.6 Hz), 111.6 (d, $^{2}J_{FC}$ = 24.5 Hz) , 113.0, 114.5 (d, $^{2}J_{FC}$ = 22.7 Hz), 135.0 (d, $^{3}J_{FC}$ = 7.6 Hz), 140.6, 153.1, 154.0, 158.2 (d, $^{1}J_{FC}$ = 234.1 Hz), 164.0, 174.1, 177.7, 195.4 ppm; Anal. Calcd for $C_{20}H_{16}FN_3O_4S$: C, 58.10; H, 3.90; N, 10.16. Found: C, 57.92; H, 4.08; N, 9.98.

5'-Chloro-8,8-dimethyl-2-thioxo-2,3,8,9-tetrahydrospiro [chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2',4,6(1*H*,7*H*)-trione (**4**{*1,3,5*}).



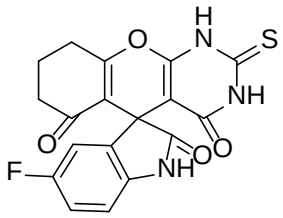
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3342, 3195, 2871, 1674, 1622, 1560, 1477, 1338, 1311, 1188, 1163, 1074, 1041, 819, 684, 586 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.98 (s, 3H, C$H_3$), 1.01 (s, 3H, C$H_3$), 2.12 and 2.17 (AB system, $J_{AB}$ =16.0 Hz, 2H, C$H_2$), 2.57 and 2.61 (AB system, $J_{AB}$ =17.5 Hz, 2H, C$H_2$), 6.70 (d, $J$ = 8.5 Hz, 1H, *H*Ar), 7.11 (dd, $J$ = 8.5 Hz, $J$ = 2.0 Hz, 1H, *H*Ar), 7.20 (s, 1H, *H*Ar), 10.53 (s, 1H, N*H*), 12.38 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 21.1, 28.0, 28.1, 29.7, 32.2, 46.3, 50.9, 90.0, 108.7, 113.7, 124.1, 128.6, 129.9, 134.0, 142.0, 150.0, 152.3, 159.8, 163.3, 178.1, 195.3 ppm; Anal. Calcd for $C_{20}H_{16}ClN_3O_4S$: C, 55.88; H, 3.75; N, 9.77. Found: C, 56.02; H, 3.93; N, 9.58.

5'-Methyl-2-thioxo-2,3,8,9-tetrahydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2',4,6(1*H*,7*H*)-trione (**4**{*2,3,2*}).



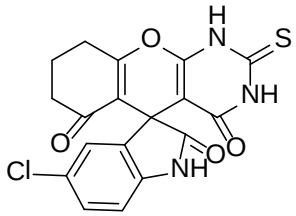
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3384, 2869, 1700, 1670, 1622, 1560, 1492, 1325, 1299, 1184, 1145, 1105, 1080, 1010, 821, 686, 569 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.80-2.00 (m, 2H, C$H_2$), 2.15 (s, 3H, C$H_3$), 2.18-2.30 (m, 2H, C$H_2$), 2.65-2.75 (m, 2H, C$H_2$), 6.60 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 6.84 (s, 1H, *H*Ar), 6.89 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 10.31 (s, 1H, N*H*), 12.38 (s, 1H, N*H*), 13.65 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.0, 21.1, 27.2, 37.3, 45.6, 94.4, 108.6, 114.5, 124.3, 128.7, 130.1, 133.6, 141.8, 152.7, 159.5, 165.4, 174.0, 177.7, 195.4 ppm; Anal. Calcd for $C_{19}H_{15}N_3O_4S$: C, 59.83; H, 3.96; N, 11.02. Found: C, 60.01; H, 4.15; N, 10.85.

5'-Fluoro-2-thioxo-2,3,8,9-tetrahydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2',4,6(1*H*,7*H*)-trione (**4**{*2,3,4*}).



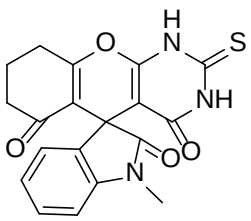
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3215, 3084, 2875, 1681, 1618, 1566, 1554, 1485, 1326, 1303, 1267, 1188, 1012, 804, 599 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.88-1.95 (m, 2H, C$H_2$), 2.10-2.30 (m, 2H, C$H_2$), 2.64-2.75 (m, 2H, C$H_2$), 6.68-6.70 (m, 1H, *H*Ar), 6.91 (t, *J* = 8.0 Hz,1H, *H*Ar), 7.05 (d, *J* = 7.0 Hz,1H, *H*Ar), 10.46 (s, 1H, N*H*), 12.42 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.0, 27.3, 37.2, 46.1, 93.8, 109.2 (d, $^{3}J_{FC}$ = 7.6 Hz), 111.8 (d, $^{2}J_{FC}$ = 24.5 Hz) , 114.0, 114.4 (d, $^{3}J_{FC}$ = 22.7 Hz), 135.0 (d, $^{2}J_{FC}$ = 7.6 Hz), 140.6, 153.0, 158.3 (d, $^{1}J_{FC}$ = 234.2 Hz), 159.6, 165.8, 174.1, 177.8, 195.6 ppm; Anal. Calcd for $C_{18}H_{12}FN_3O_4S$: C, 56.10; H, 3.14; N, 10.90. Found: C, 55.92; H, 2.96; N, 11.08.

5'-Chloro--2-thioxo-2,3,8,9-tetrahydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2',4,6(1*H*,7*H*)-trione (**4**{*2,3,5*}).



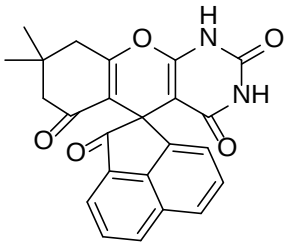
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3357, 2852, 2678, 1674, 1622, 1564, 1477, 1326, 1299, 1191, 1012, 825, 613, 576 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.89-1.95 (m, 2H, C$H_2$), 2.19-2.30 (m, 2H, C$H_2$), 2.66-2.75 (m, 2H, C$H_2$), 6.72 (d, *J* = 8.0 Hz, 1H, *H*Ar), 7.13 (d, *J* = 8.0 Hz, 1H, *H*Ar), 7.21 (s, 1H, *H*Ar), 10.56 (s, 1H, N*H*), 12.40 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.0, 27.3, 37.2, 45.8, 93.7, 110.1, 114.0, 124.1, 125.3, 128.3, 135.5, 143.3, 153.1, 159.7, 166.0, 174.2, 177.6, 195.6 ppm; Anal. Calcd for $C_{18}H_{12}ClN_3O_4S$: C, 53.80; H, 3.01; N, 10.46. Found: C, 53.98; H, 2.83; N, 10.62.

1'-Methyl-2-thioxo-2,3,8,9-tetrahydrospiro[chromeno[2,3-*d*]pyrimidine-5,3'-indoline]-2',4,6(1*H*,7*H*)-trione (**4**{*2,3,8*}).



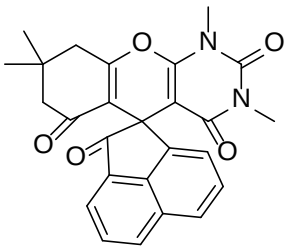
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3442, 3064, 2918, 2848, 1666, 1610, 1569, 1494, 1469, 1382, 1357, 1323, 1298, 1249, 1184, 1128, 1087, 1008, 761, 671, 569 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.85-1.95 (m, 2H, C$H_2$), 2.15-2.30 (m, 2H, C$H_2$), 2.66-2.77 (m, 2H, C$H_2$), 3.12 (s, 1H, C$H_3$), 6.86-6.91 (m, 2H, *H*Ar), 7.10 (d, $J$ = 7.0 Hz, 1H, *H*Ar), 7.19 (t, $J$ = 7.0 Hz, 1H, *H*Ar), 12.40 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 24.8, 31.6, 31.9, 41.8, 45.8, 53.8, 98.9, 112.5, 119.0, 126.8, 128.2, 135.5, 137.4, 150.4, 157.6, 164.2, 170.5, 178.8, 181.2, 200.2 ppm; Anal. Calcd for $C_{19}H_{15}N_3O_4S$: C, 59.83; H, 3.96; N, 11.02. Found: C, 60.02; H, 4.15; N, 10.85.

8',8'-Dimethyl-8',9'-dihydro-2*H*-spiro[acenaphthylene-1,5'-chromeno[2,3-*d*]pyrimidine]-2,2',4',6'(1'*H*,3'*H*,7'*H*)-tetraone (**6**{*1*,*1*,***5***}).




Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3442, 2956, 1683, 1652, 1577, 1496, 1363, 1307, 1203, 1186, 1159, 1107, 1041, 997, 785 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.97 (s, 3H, C$H_3$), 1.03 (s, 3H, C$H_3$), 2.00 and 2.13 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.62 and 2.73 (AB system, $J_{AB}$ = 18.0 Hz, 2H, C$H_2$), 7.38 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 7.55 (t, 1H, $J$ = 7.5 Hz, *H*Ar), 7.76 (t, $J$ = 7.5 Hz, 1H, *H*Ar), 7.80 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 7.85 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 8.13 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 10.94 (s, 1H, N*H*), 12.32 (s, 1H, N*H*) ppm.

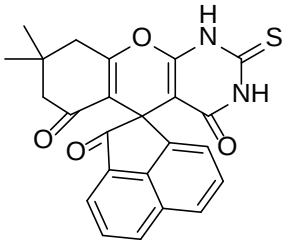
1',3',8',8'-Tetramethyl-8',9'-dihydro-2*H*-spiro[acenaphthylene-1,5'-chromeno[2,3-*d*]pyrimidine]-2,2',4',6'(1'*H*,3'*H*,7'*H*)-tetraone (**6**{*1*,*2*,***5***}).




Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3336, 2960, 1716, 1685, 1639, 1624, 1477, 1215, 1186, 1093, 775, 752 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.97 (s, 3H, C$H_3$), 1.03 (s, 3H, C$H_3$), 2.12 and 2.14 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C$H_2$), 2.70 and 2.77 (AB system, $J_{AB}$ = 18.0 Hz, 2H, C$H_2$), 2.88 (s, 3H, C$H_3$), 3.43 (s, 3H, C$H_3$), 7.38 (d, $J$ = 7.5 Hz,

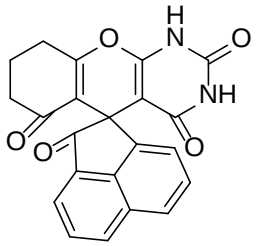
1H, *H*Ar), 7.55 (t, 1H, *J* = 7.5 Hz, *H*Ar), 7.77 (t, *J* = 7.5 Hz, 1H, *H*Ar), 7.83 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.85 (d, *J* = 7.5 Hz, 1H, *H*Ar), 8.13 (d, *J* = 7.5 Hz, 1H, *H*Ar) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 27.3, 28.0, 28.1, 29.8, 32.3, 50.3, 91.2, 115.2, 120.2, 120.4, 125.1, 128.5, 128.6, 129.7, 130.3, 135.6, 141.1, 143.3, 150.0, 152.6, 160.5, 164.0, 195.8, 203.5 ppm; Anal. Calcd for $C_{26}H_{22}N_2O_5$: C, 70.58; H, 5.01; N, 6.33. Found: C, 70.76; H, 4.85; N, 6.50.

8',8'-Dimethyl-2'-thioxo-2',3',8',9'-tetrahydro-2*H*-spiro[acenaphthylene-1,5'-chromeno[2,3-*d*]pyrimidine]-2,4',6'(1'*H*,7'*H*)-trione (**6**{*1,3,**5***}).




Colorless crystals; mp: 291-293 $^{o}$C; IR (KBr): 3419, 2956, 1681, 1622, 1536, 1492, 1465, 1363, 1307, 1242, 1180, 1157, 1103, 1037, 835, 788 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.97 (s, 3H, C*H*$_3$), 1.03 (s, 3H, C*H*$_3$), 2.00 and 2.14 (AB system, $J_{AB}$ = 16.0 Hz, 2H, C*H*$_2$), 2.62 and 2.73 (AB system, $J_{AB}$ = 18.0 Hz, 2H, C*H*$_2$), 7.44 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.55 (t, 1H, *J* = 7.5 Hz, *H*Ar), 7.77 (t, *J* = 7.5 Hz, 1H, *H*Ar), 7.82 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.87 (d, *J* = 7.5 Hz, 1H, *H*Ar), 8.14 (d, *J* = 7.5 Hz, 1H, *H*Ar), 12.35 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 18.5, 26.6, 27.6, 31.8, 56.0, 95.0, 114.5, 120.0, 120.4, 124.6, 128.0, 128.2, 129.2, 130.0, 134.8, 140.8, 142.4, 152.7, 159.5, 163.7, 173.7, 195.3, 202.7 ppm;

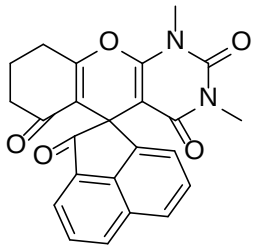
8',9'-Dihydro-2*H*-spiro[acenaphthylene-1,5'-chromeno[2,3-*d*]pyrimidine]-2,2',4',6'(1'*H*,3'*H*,7'*H*)-tetraone (**6**{*2,1,**5***}).




Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3446, 2929, 2810, 1685, 1666, 1624, 1521, 1419, 1359, 1298, 1234, 1186, 993, 790, 779, 754 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.85-1.95 (m, 2H, C*H*$_2$), 2.07-2.20 (m, 2H, C*H*$_2$), 2.70-2.80 (m, 2H, C*H*$_2$), 7.39 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.54 (t, 1H, *J* = 7.5 Hz, *H*Ar), 7.74 (t, *J* = 7.5 Hz, 1H, *H*Ar), 7.80 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.84 (d, *J* = 7.5 Hz, 1H, *H*Ar), 8.12 (d, *J* = 7.5 Hz, 1H, *H*Ar) ppm; $^{13}$C NMR

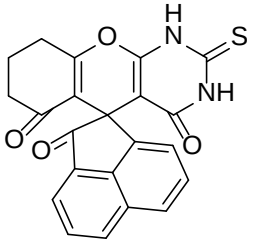
(DMSO-$d_6$, 125 MHz) δ: 20.1, 27.1, 36.7, 49.7, 90.9, 116.3, 120.1, 120.4, 125.0, 128.4, 128.7, 129.7, 130.3, 135.6, 141.1, 143.6, 150.0, 153.6, 162.4, 166.1, 195.9, 203.6 ppm; Anal. Calcd for $C_{22}H_{14}N_2O_5$: C, 68.39; H, 3.65; N, 7.25. Found: C, 68.20; H, 3.46; N, 7.43.

1',3'-Dimethyl-8',9'-dihydro-2*H*-spiro[acenaphthylene-1,5'-chromeno[2,3-*d*]pyrimidine]-2,2',4',6'(1'*H*,3'*H*,7'*H*)-tetraone (**6**{*2*,*2*,***5***}).



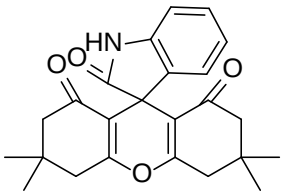
Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3421, 1720, 1647, 1618, 1558, 1490, 1355, 1305, 1215, 1112, 1082, 993, 786 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.88-1.98 (m, 2H, C$H_2$), 2.11-2.34 (m, 2H, C$H_2$), 2.82-2.85 (m, 2H, C$H_2$), 2.88 (s, 3H, C$H_3$), 3.44 (s, 3H, C$H_3$), 7.38 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.54 (t, 1H, *J* = 7.5 Hz, *H*Ar), 7.77 (t, *J* = 7.5 Hz, 1H, *H*Ar), 7.82 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.85 (d, *J* = 7.5 Hz, 1H, *H*Ar), 8.13 (d, *J* = 7.5 Hz, 1H, *H*Ar) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.1, 27.0, 28.0, 29.8, 36.8, 50.4, 91.3, 116.3, 120.1, 120.6, 125.0, 128.5, 128.6, 129.7, 130.3, 135.6, 141.1, 143.5, 150.0, 152.5, 160.5, 165.8, 195.9, 203.5 ppm; Anal. Calcd for $C_{24}H_{18}N_2O_5$: C, 69.56; H, 4.38; N, 6.76. Found: C, 69.74; H, 4.56; N, 6.58.

2'-Thioxo-2',3',8',9'-tetrahydro-2H-spiro[acenaphthylene-1,5'-chromeno[2,3-d]pyrimidine]-2,4',6'(1'*H*,7'*H*)-trione (**6**{*2*,*3*,***5***}).



Colorless crystals; mp >300 $^{o}$C; IR (KBr): 3627, 3350, 3134, 3018, 2854, 1724, 1681, 1643, 1612, 1556, 1560, 1371, 1311, 1184, 1103, 1080, 1020, 786, 795 cm$^{-1}$; $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 1.85-1.95 (m, 2H, C$H_2$), 2.07-2.20 (m, 2H, C$H_2$), 2.70-2.80 (m, 2H, C$H_2$), 7.44 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.54 (t, 1H, *J* = 7.5 Hz, *H*Ar), 7.76 (t, *J* = 7.5 Hz, 1H, *H*Ar), 7.82 (d, *J* = 7.5 Hz, 1H, *H*Ar), 7.85 (d, *J* = 7.5 Hz, 1H, *H*Ar), 8.13 (d, *J* = 7.5 Hz, 1H, *H*Ar), 12.43 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 20.1, 27.1, 36.7, 49.7,

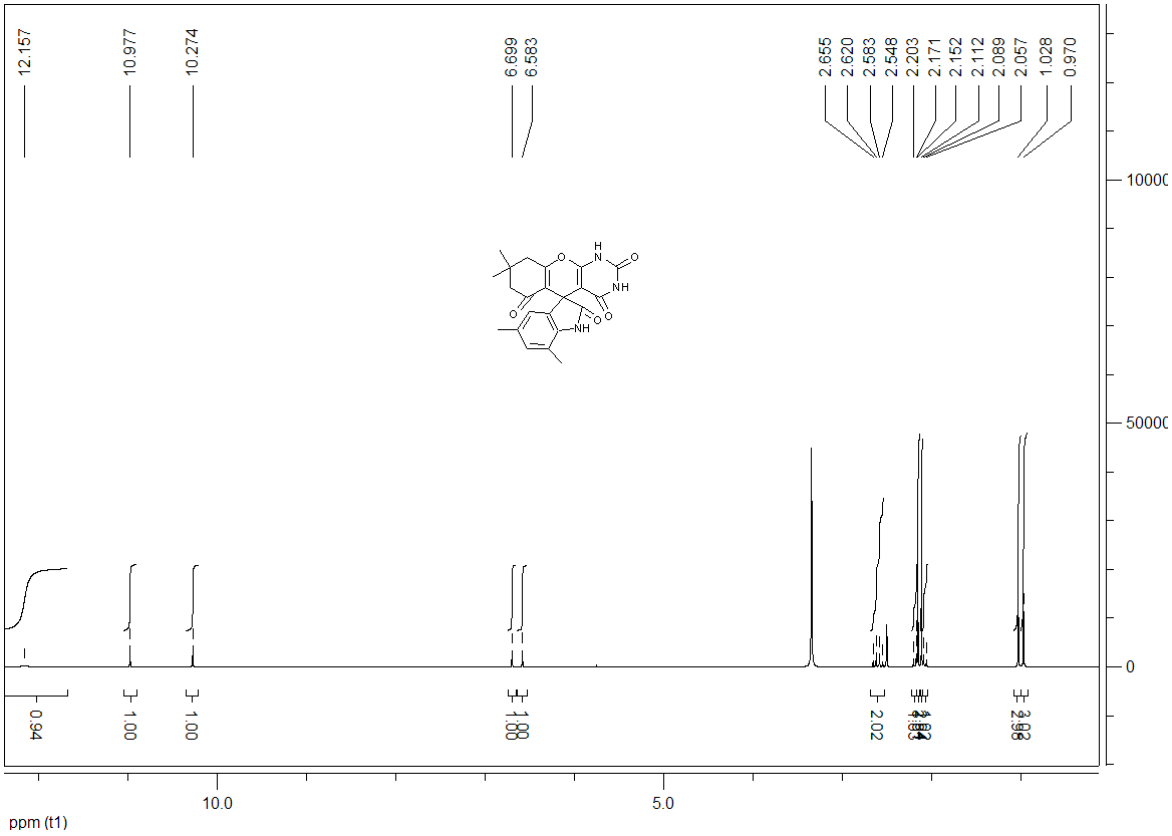
95.6, 116.0, 120.3, 120.7, 125.0, 128.5, 128.7, 129.7, 130.4, 135.3, 141.2, 143.0, 153.0, 160.0, 166.1, 174.1, 195.9, 203.1 ppm; Anal. Calcd for $C_{22}H_{14}N_2O_4S$: C, 65.66; H, 3.51; N, 6.96. Found: C, 65.83; H, 3.32; N, 7.12.

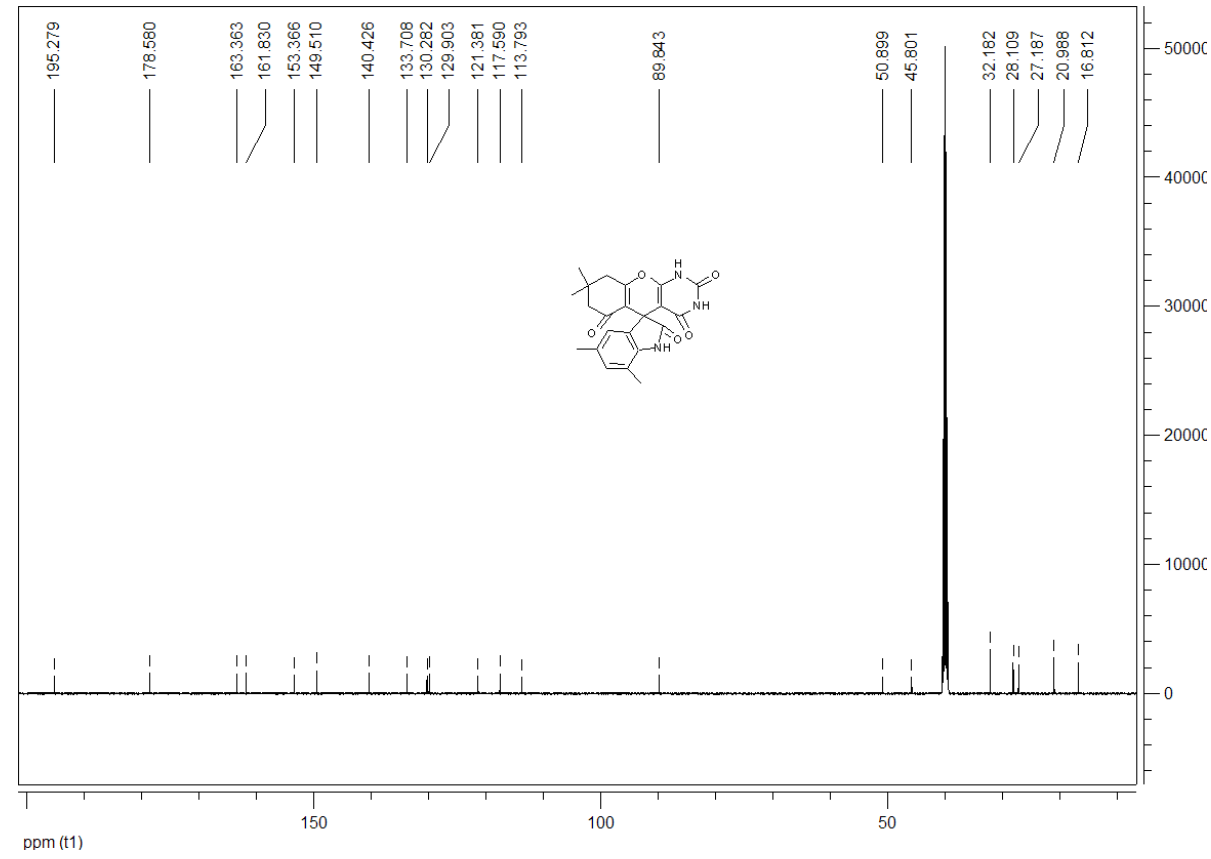



3',3',6',6'-tetramethyl-3',4',6',7'-tetrahydrospiro[indoline-3,9'-xanthene]-1',2,8'(2'*H*,5'*H*)-trione (**7**).

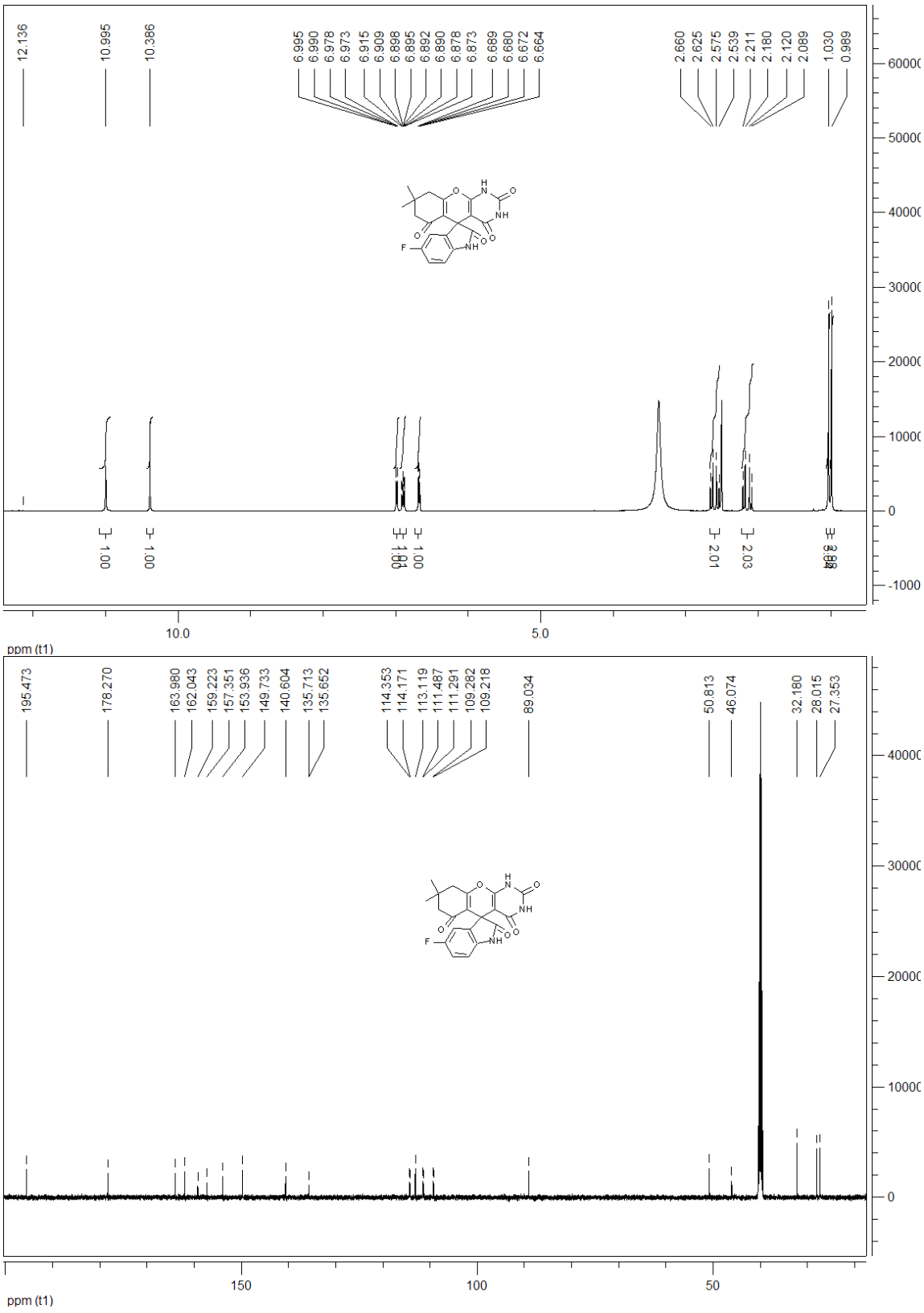
Colorless crystals; mp: 302 $^{o}$C; IR (KBr): 3431, 2962, 1732, 1712, 1666, 1618, 1483, 1469, 1346, 1315, 1294, 1222, 1168, 1134, 1031, 902, 746 cm$^{-1}$ $^{1}$H NMR (DMSO-$d_6$, 500 MHz) δ: 0.94 (s, 6H, C$H_3$), 1.02 (s, 6H, C$H_3$), 2.02 and 2.18 (AB system, $J_{AB}$ = 16.0 Hz, 4H, C$H_2$), 2.51 and 2.62 (AB system, $J_{AB}$ = 18.0 Hz, 4H, C$H_2$), 6.70 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 6.76 (t, 1H, $J$ = 7.5 Hz, *H*Ar), 6.83 (d, $J$ = 7.5 Hz, 1H, *H*Ar), 7.05 (t, $J$ = 7.5 Hz, 1H, *H*Ar), 10.29 (s, 1H, N*H*) ppm; $^{13}$C NMR (DMSO-$d_6$, 125 MHz) δ: 26.5, 27.9, 31.6, 45.1, 50.5, 108.4, 112.9, 120.6, 122.1, 127.7, 134.0, 143.8, 163.4, 178.2, 194.9 ppm.

$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,1,3*}

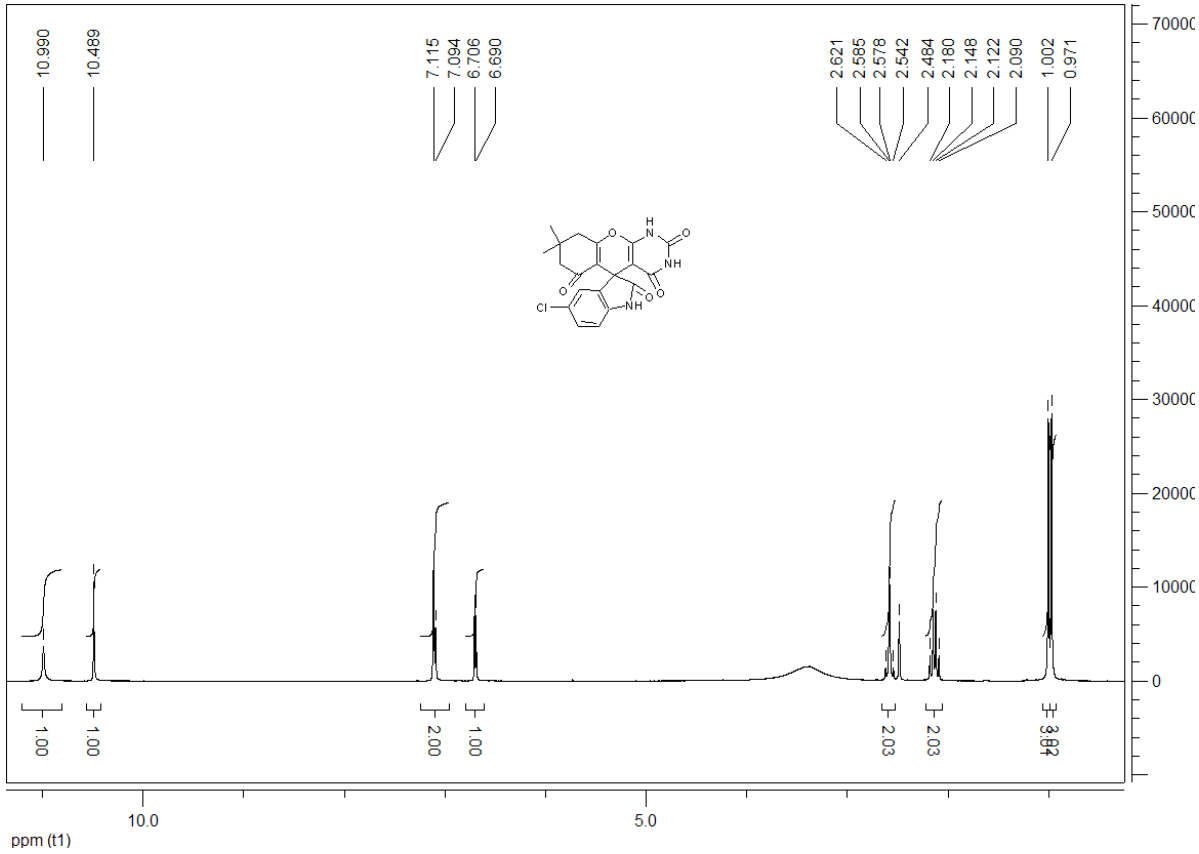


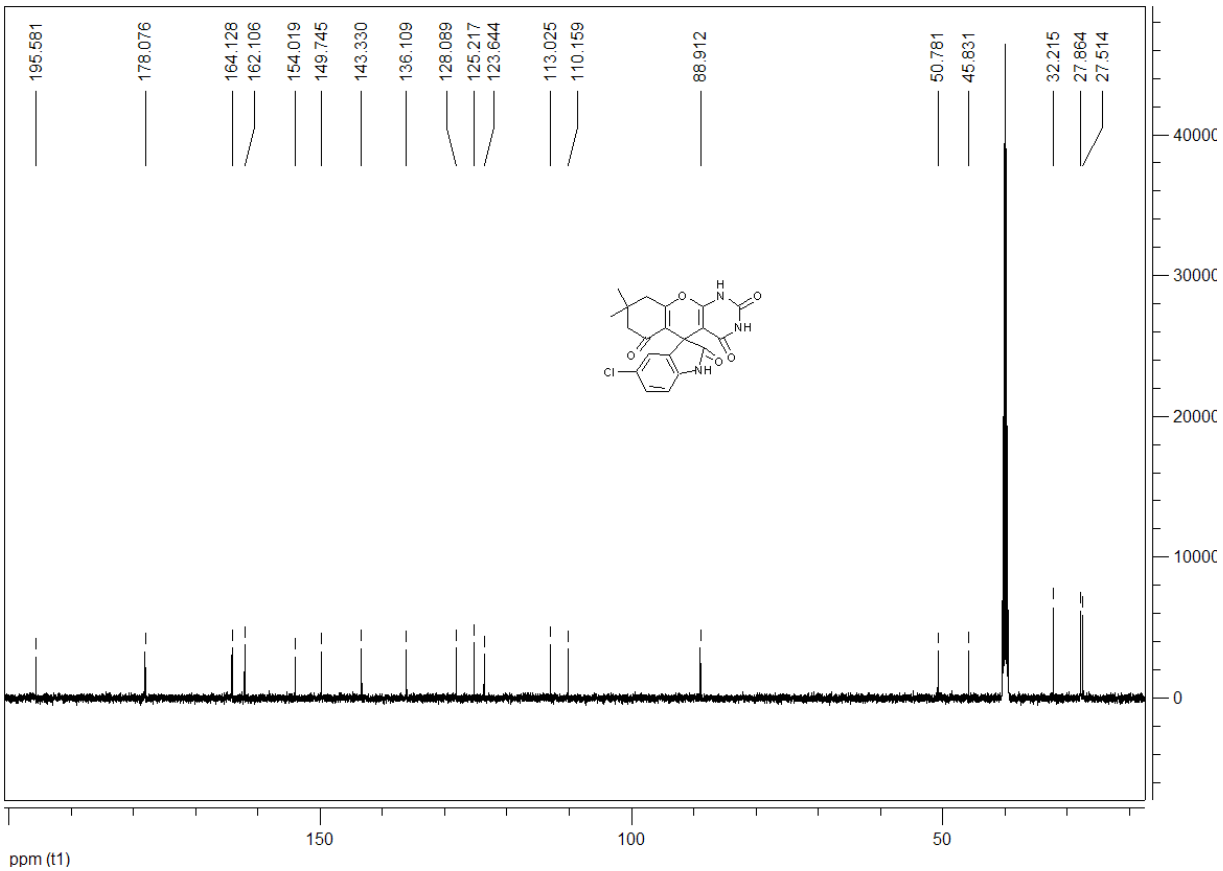



$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,1,4*}

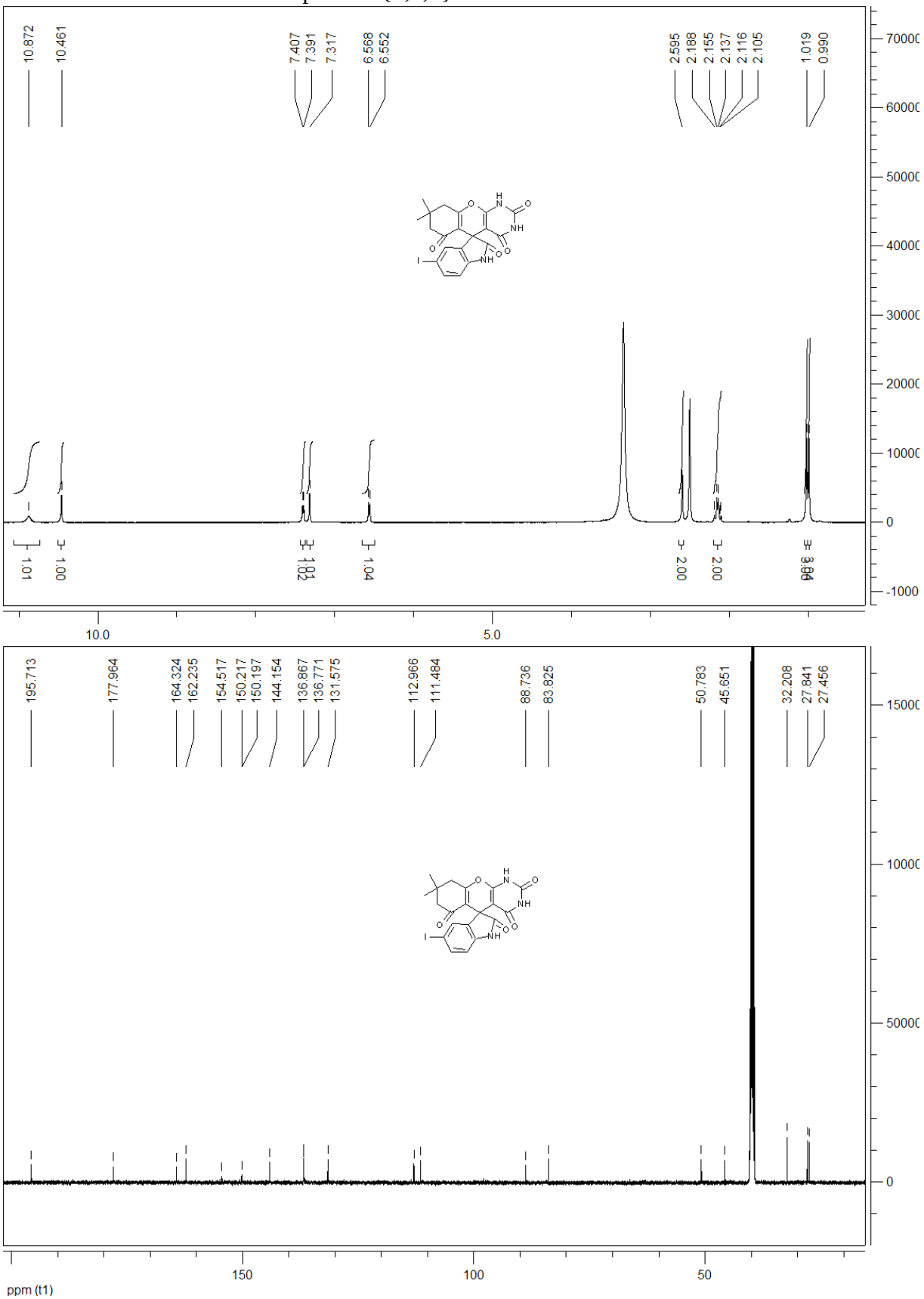


$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,1,5*}

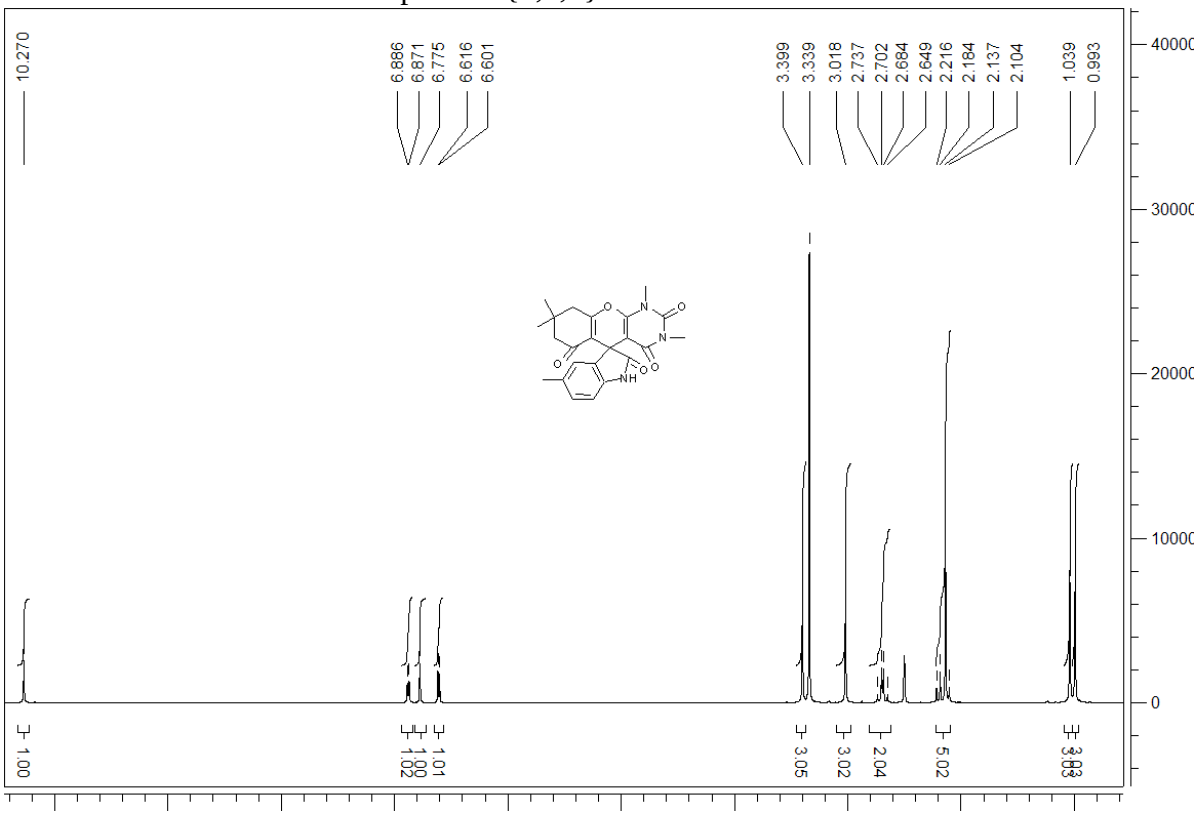


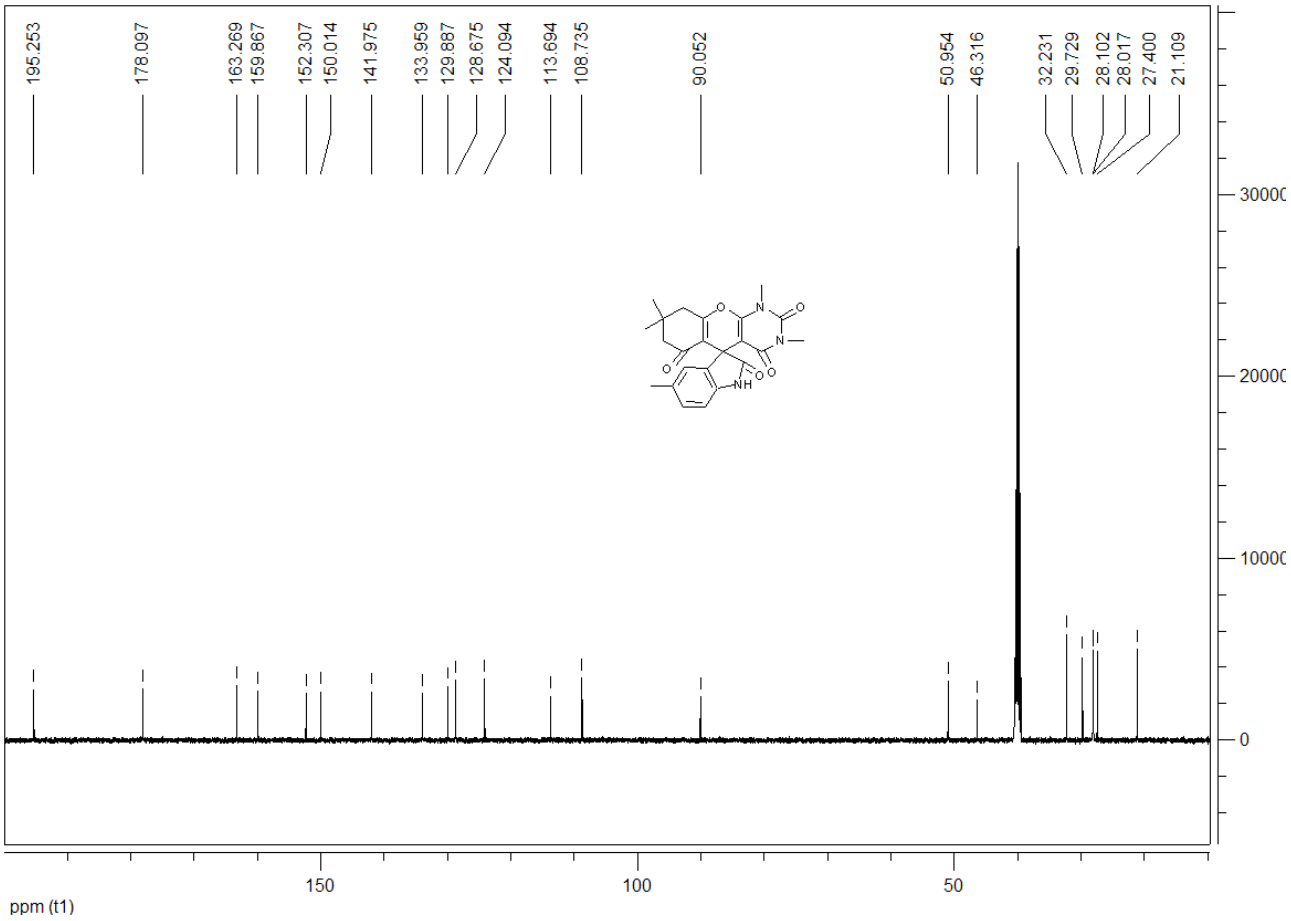



$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,1,7*}

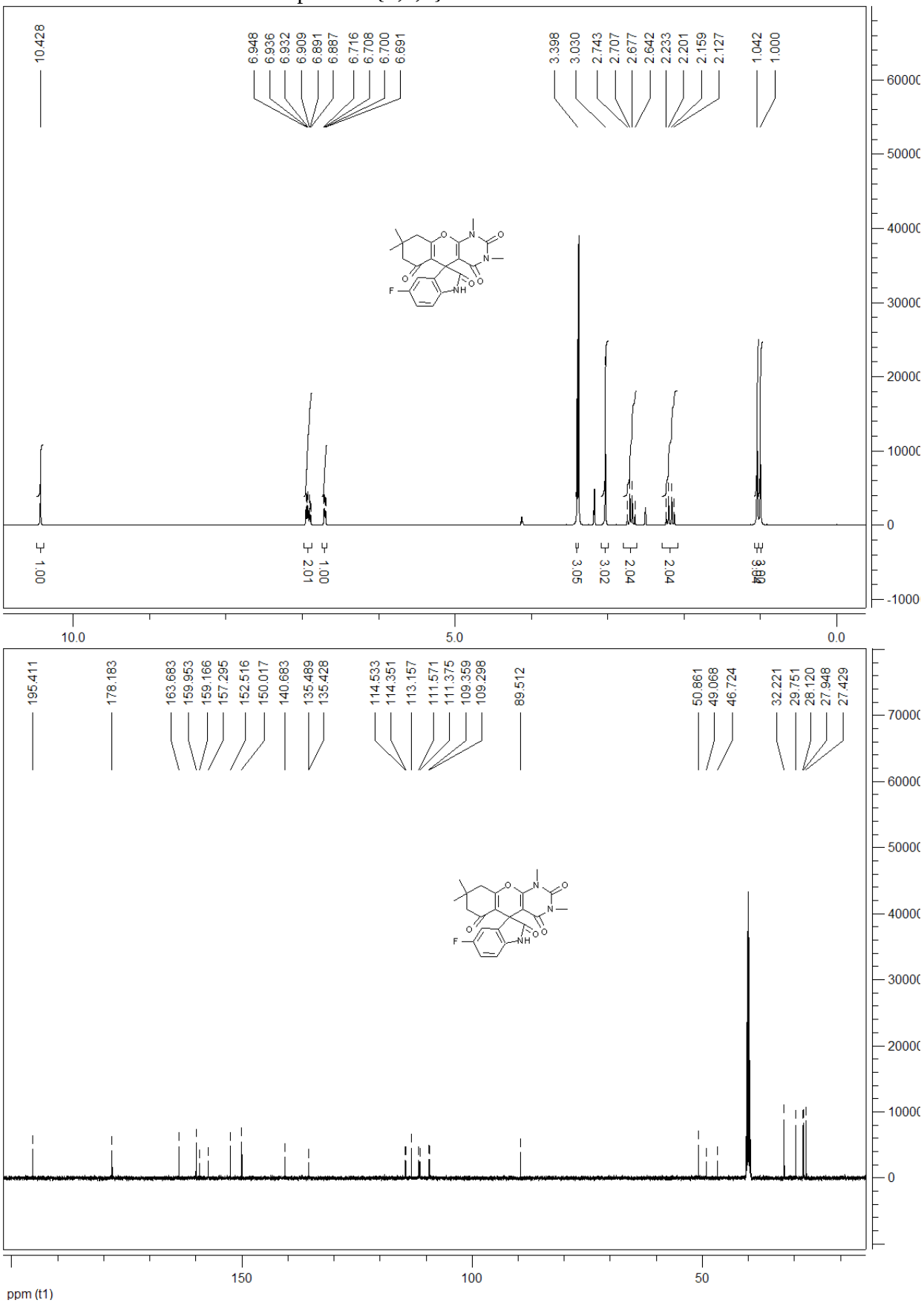


$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,2,2*}

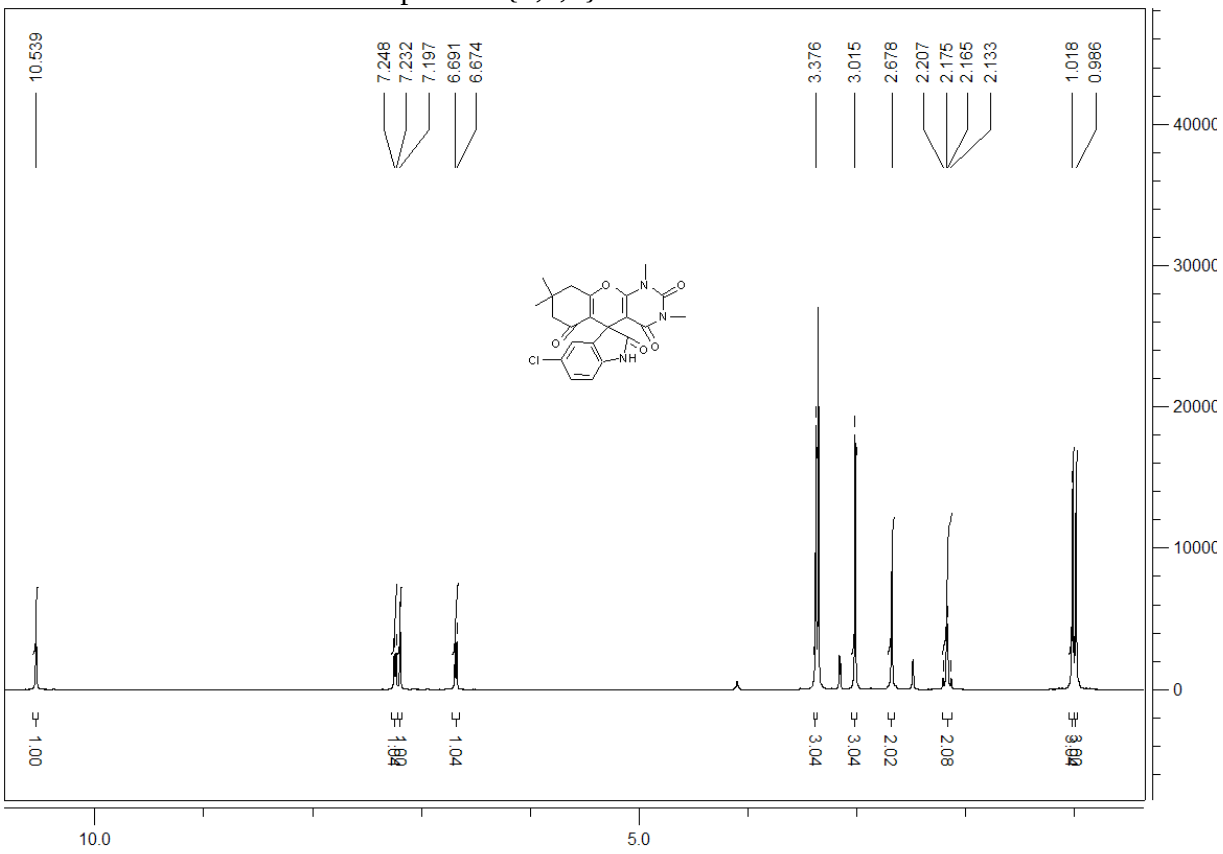


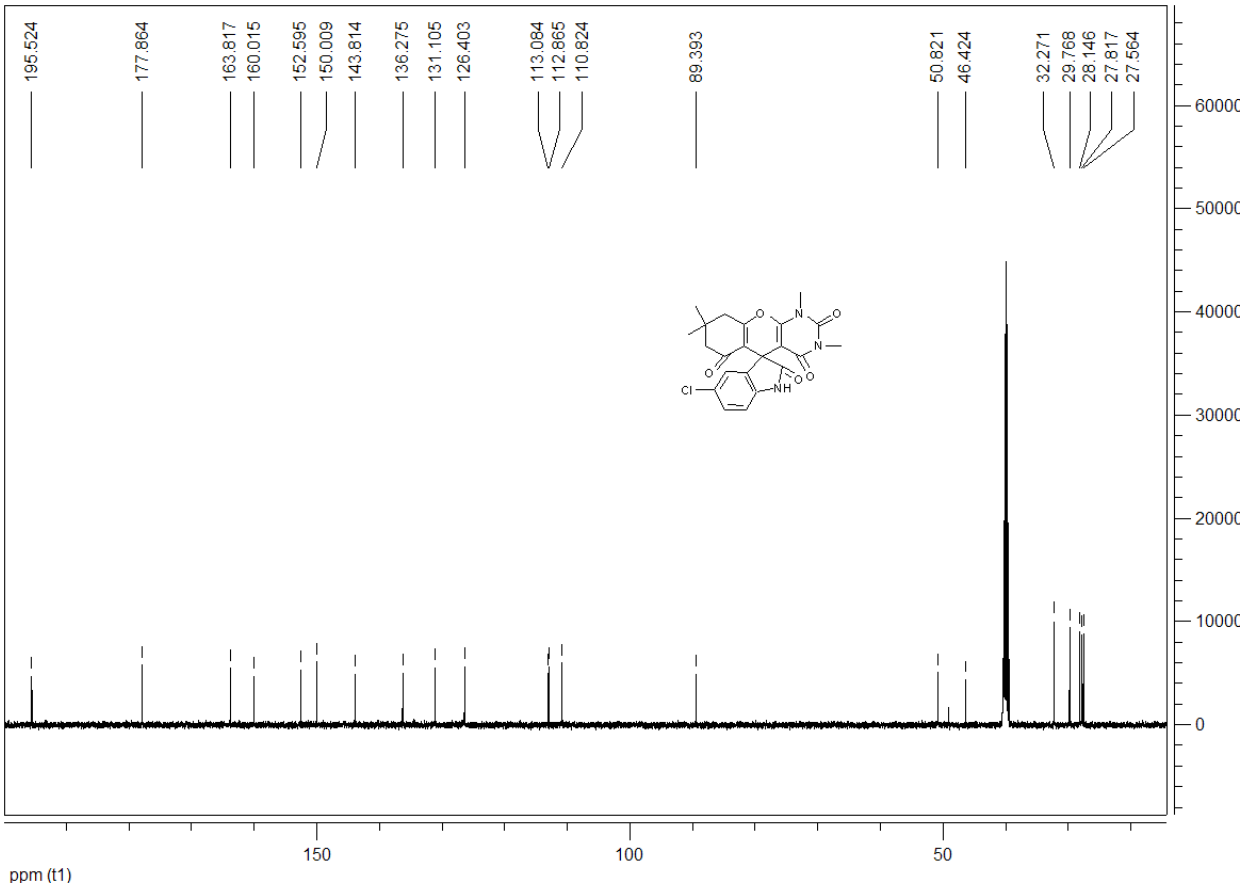



$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,2,4*}

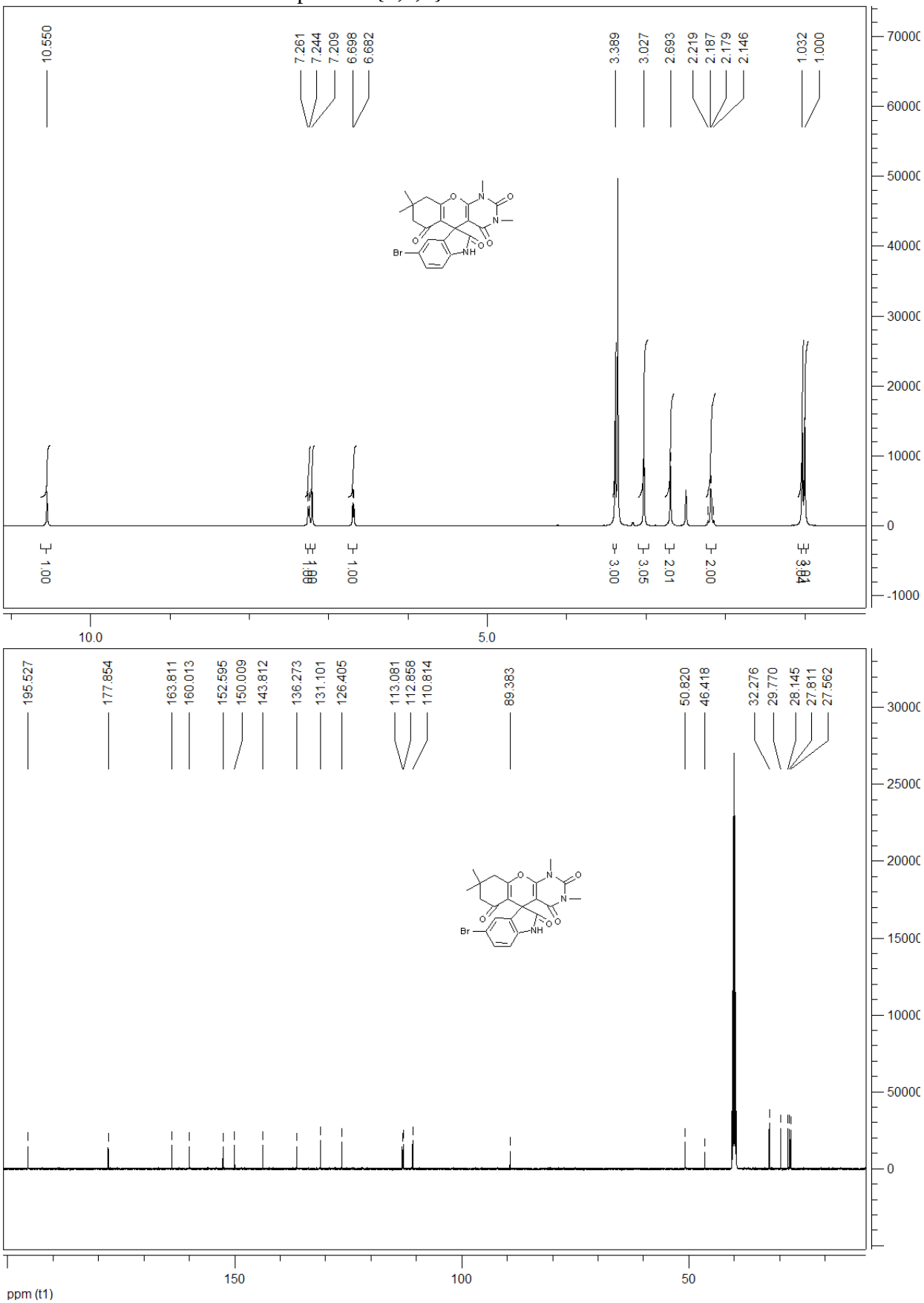


$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,2,5*}

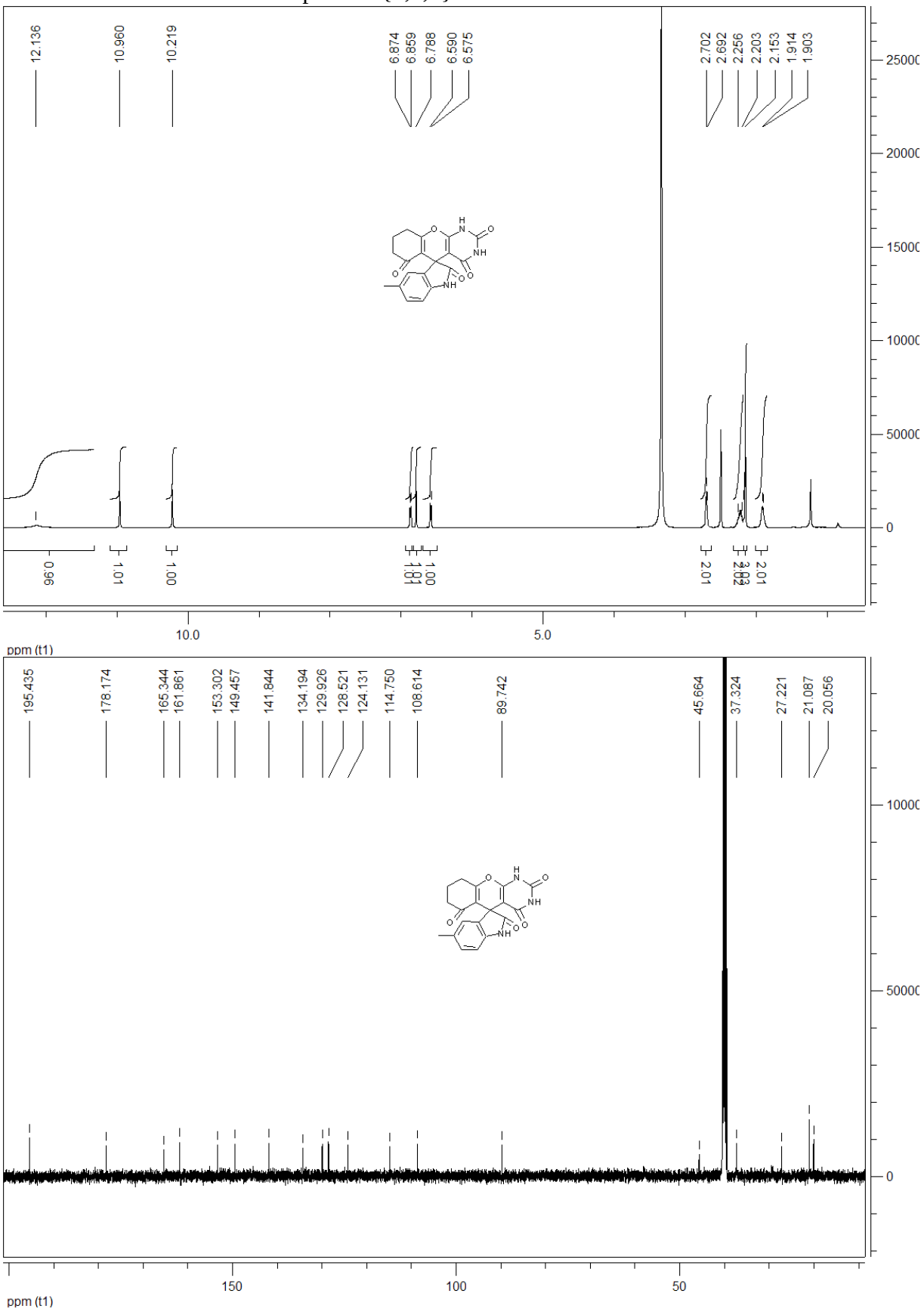


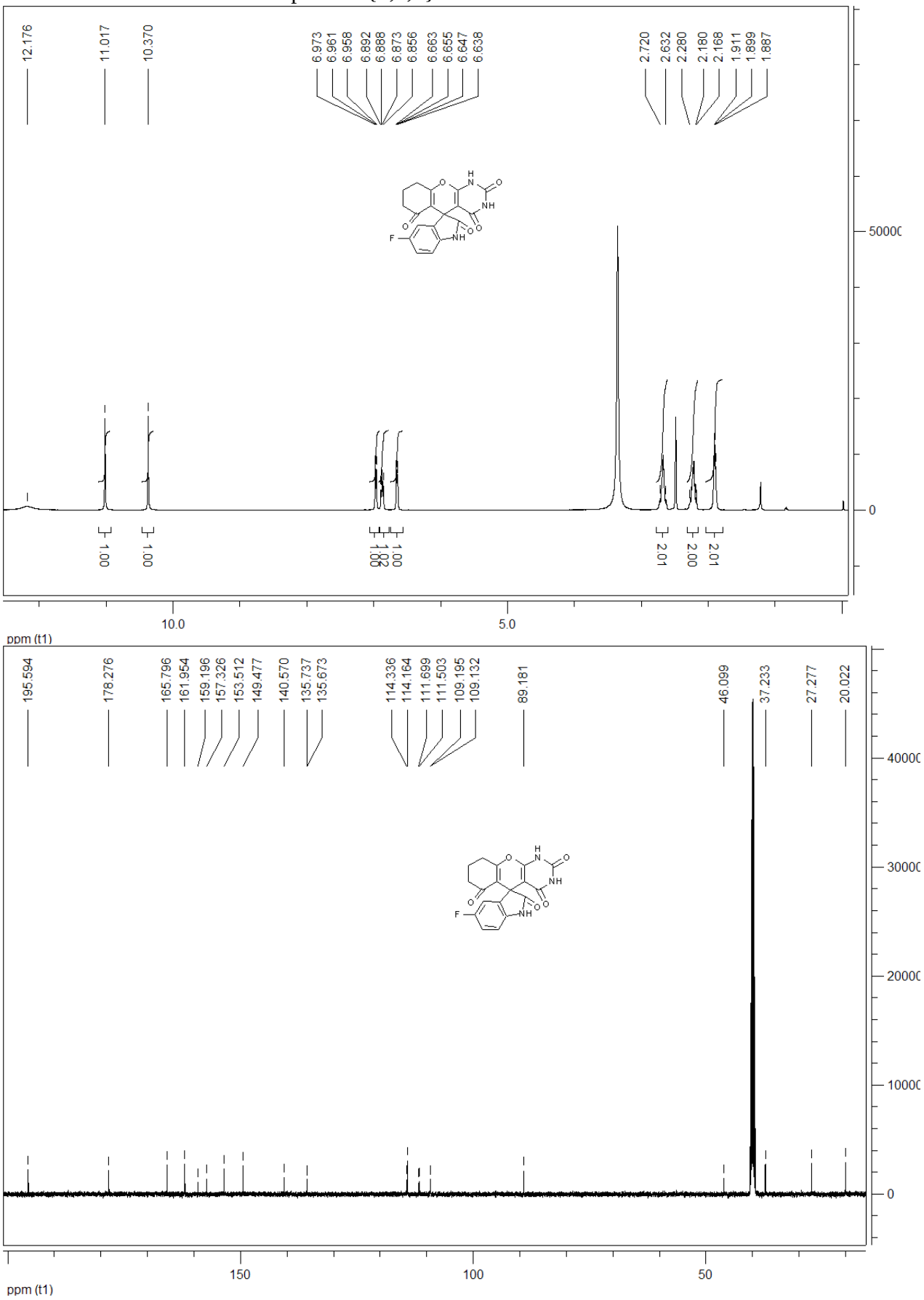
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,2,6*}



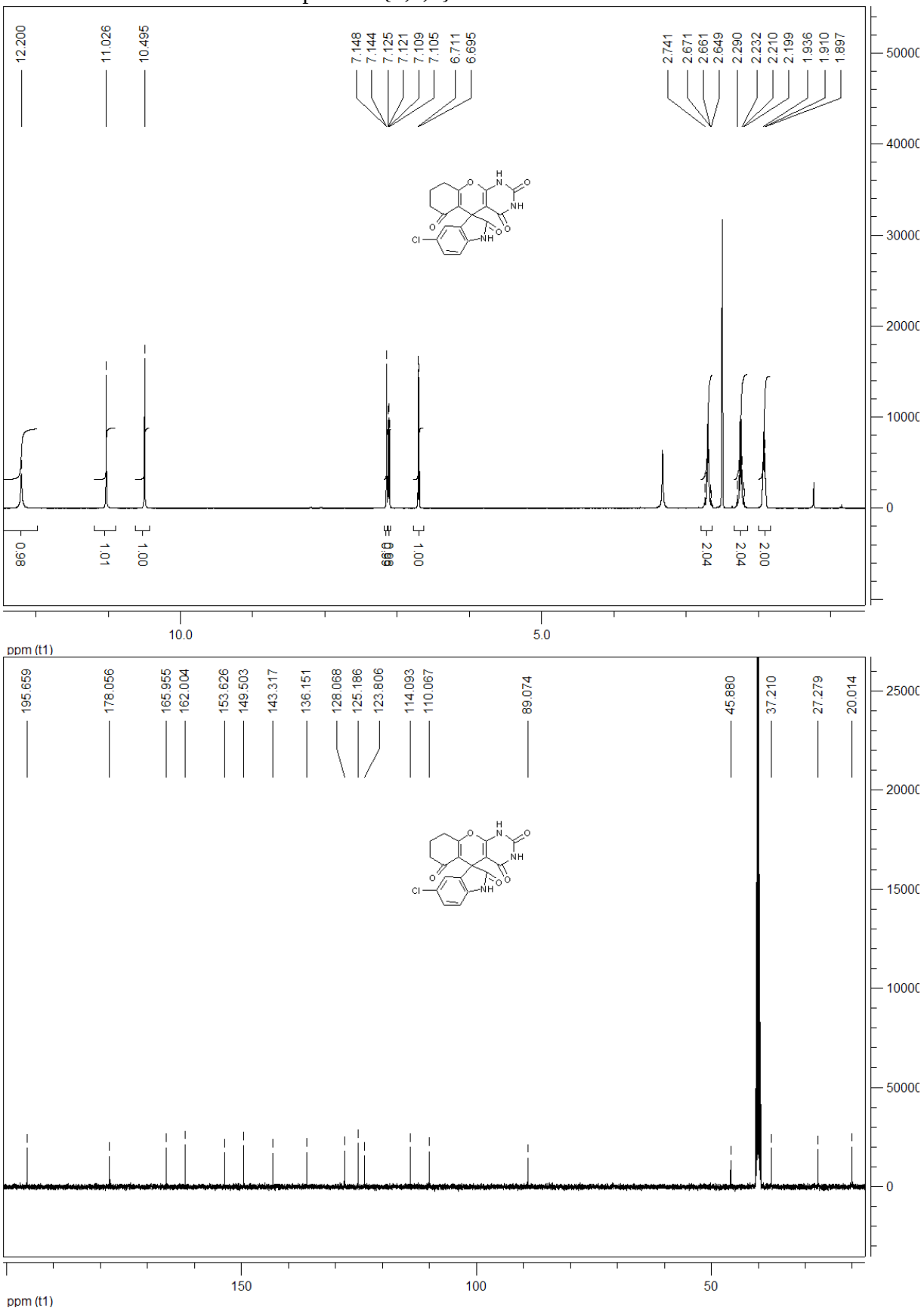
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,1,2*}



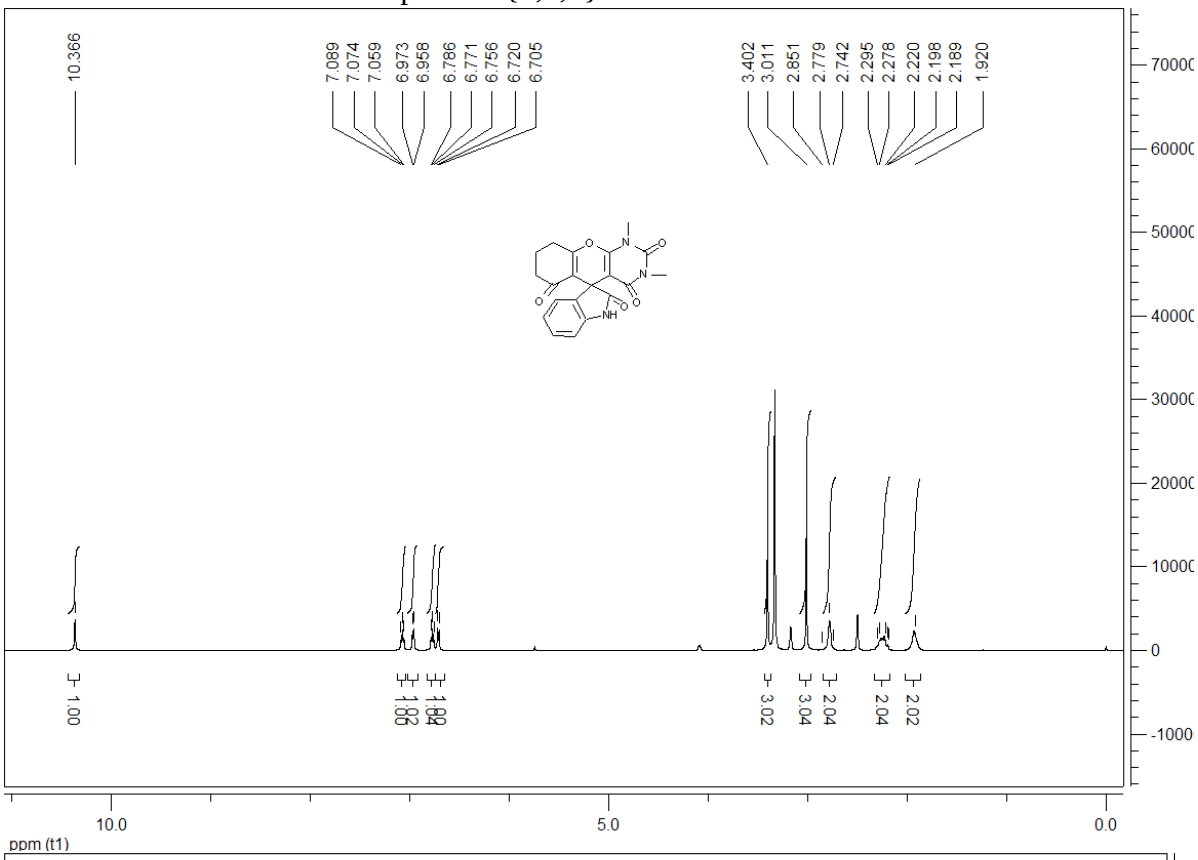
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,1,4*}

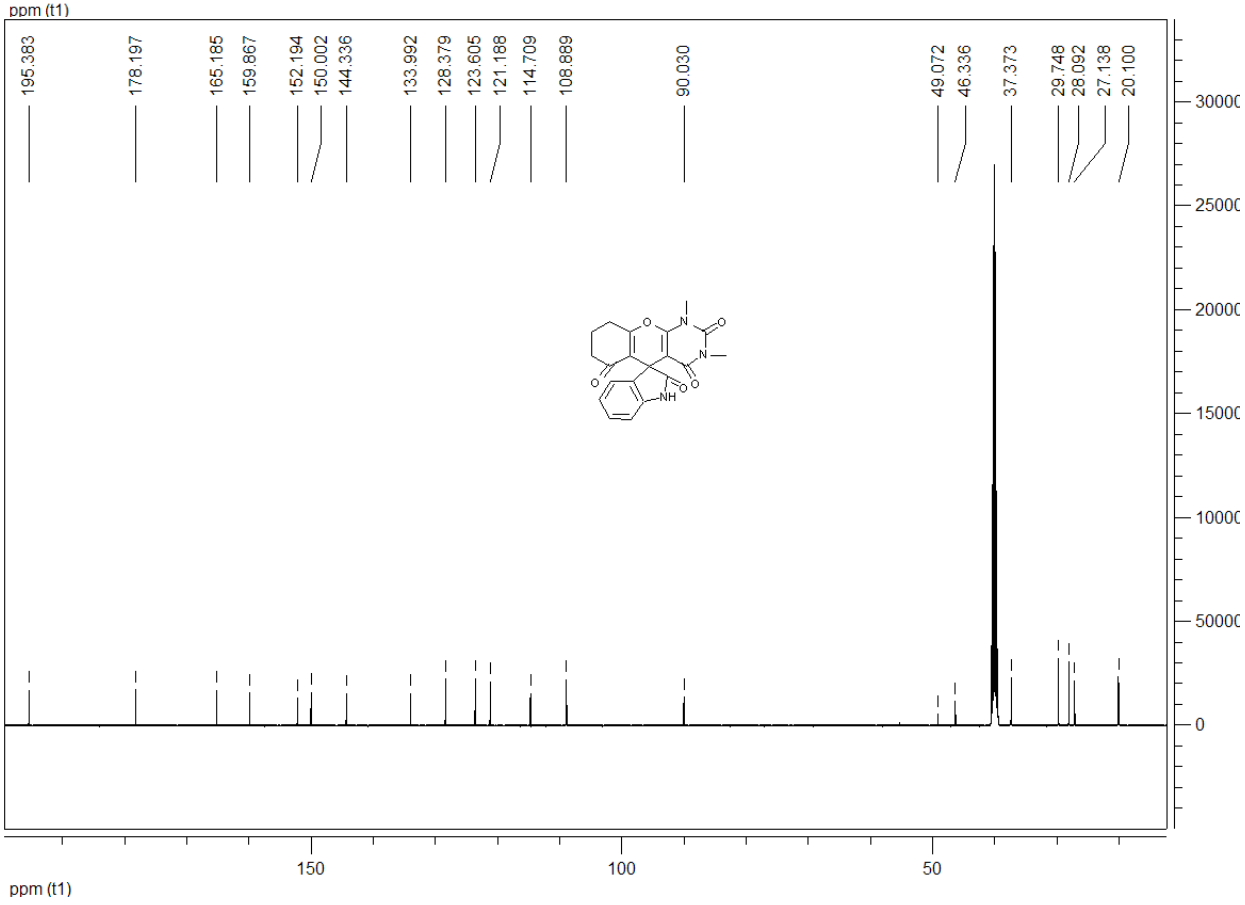


$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,1,5*}

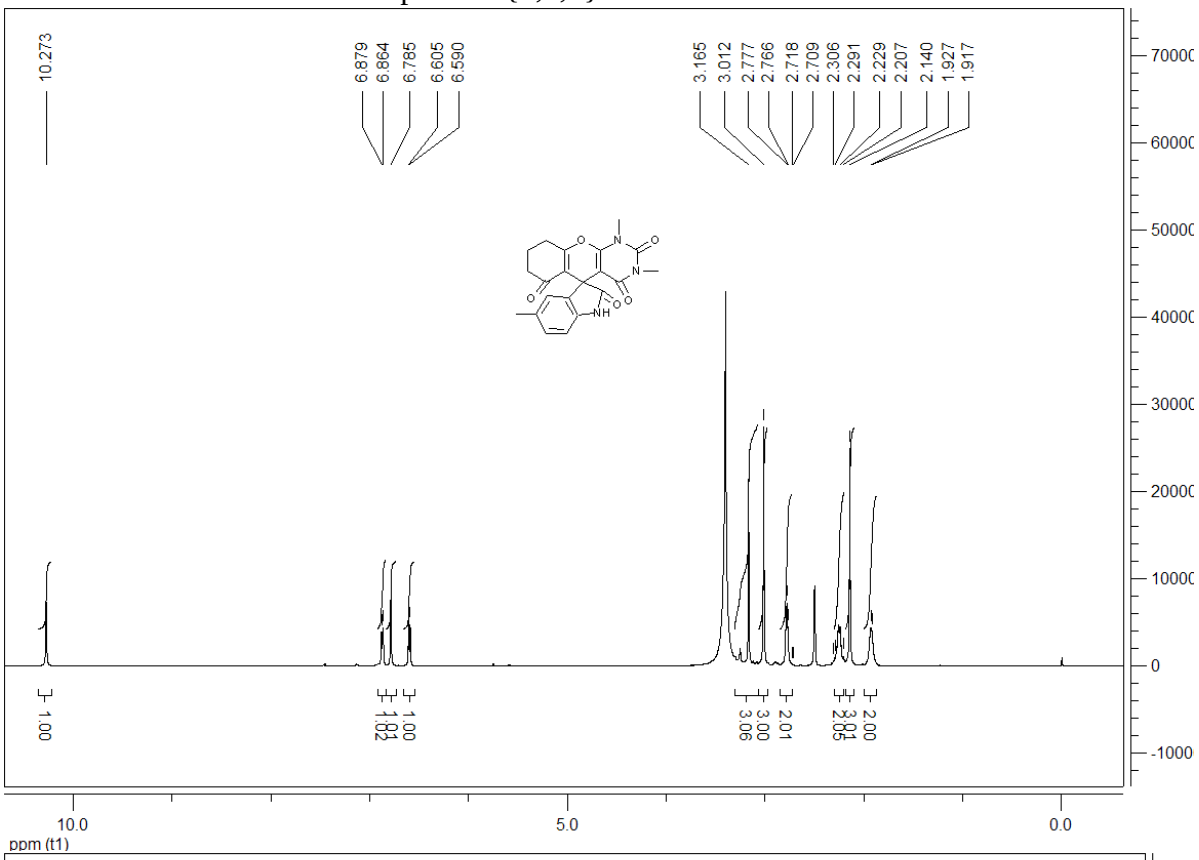


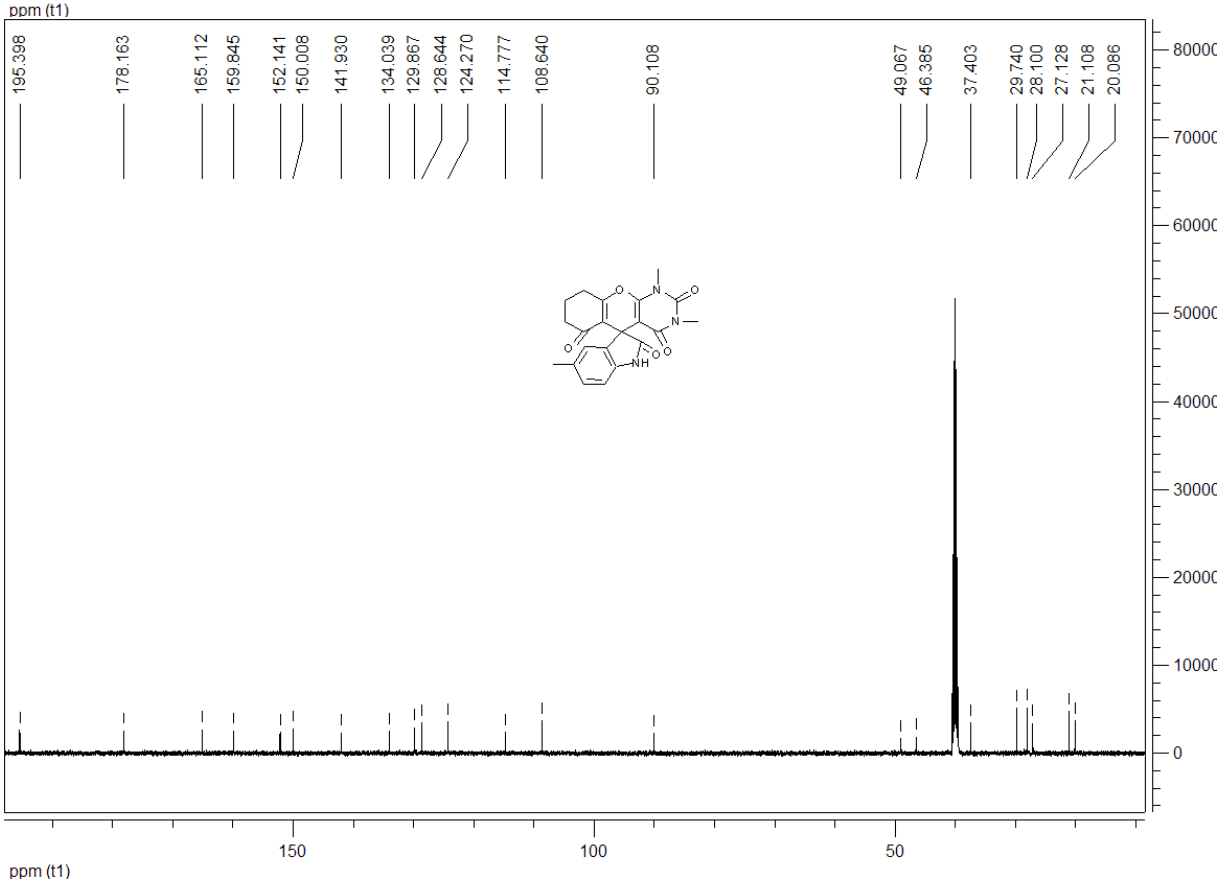
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,2,1*}



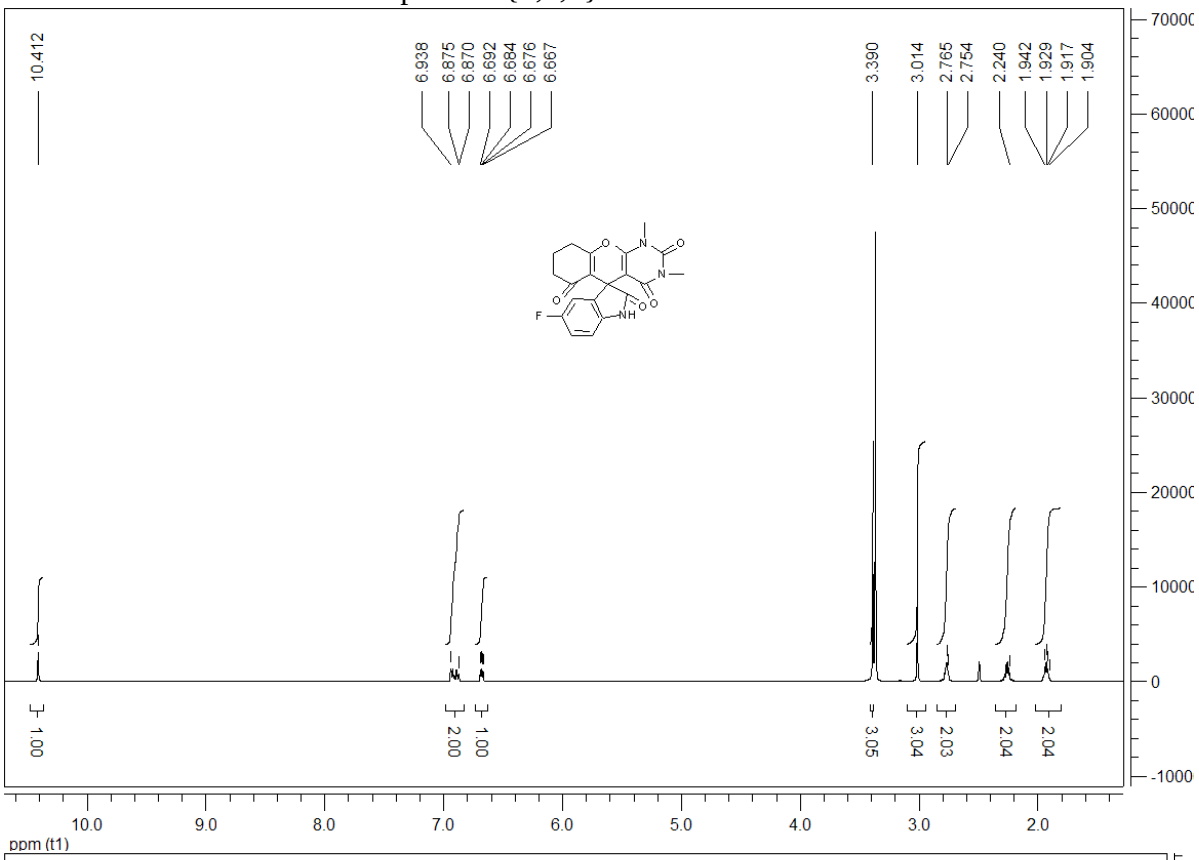



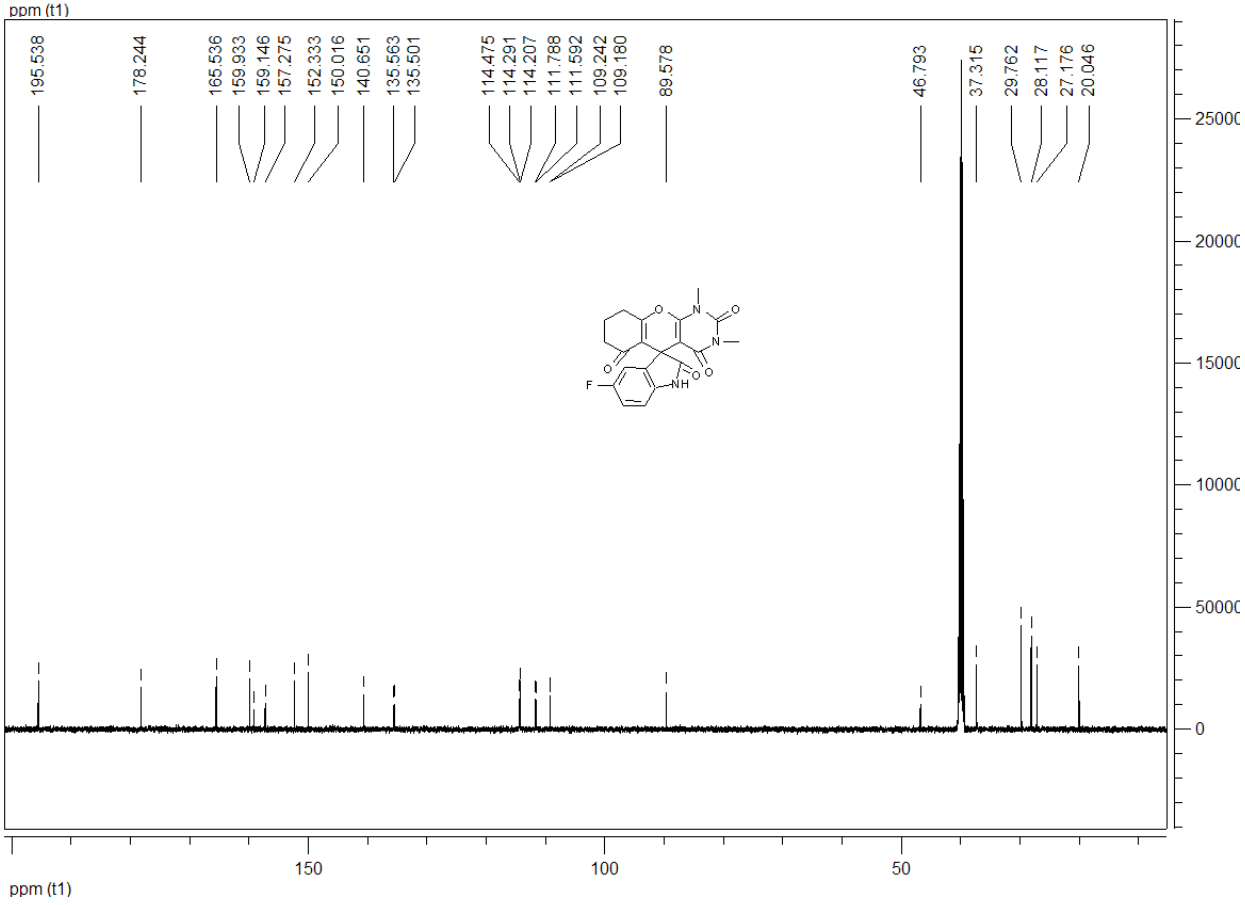
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,2,2*}



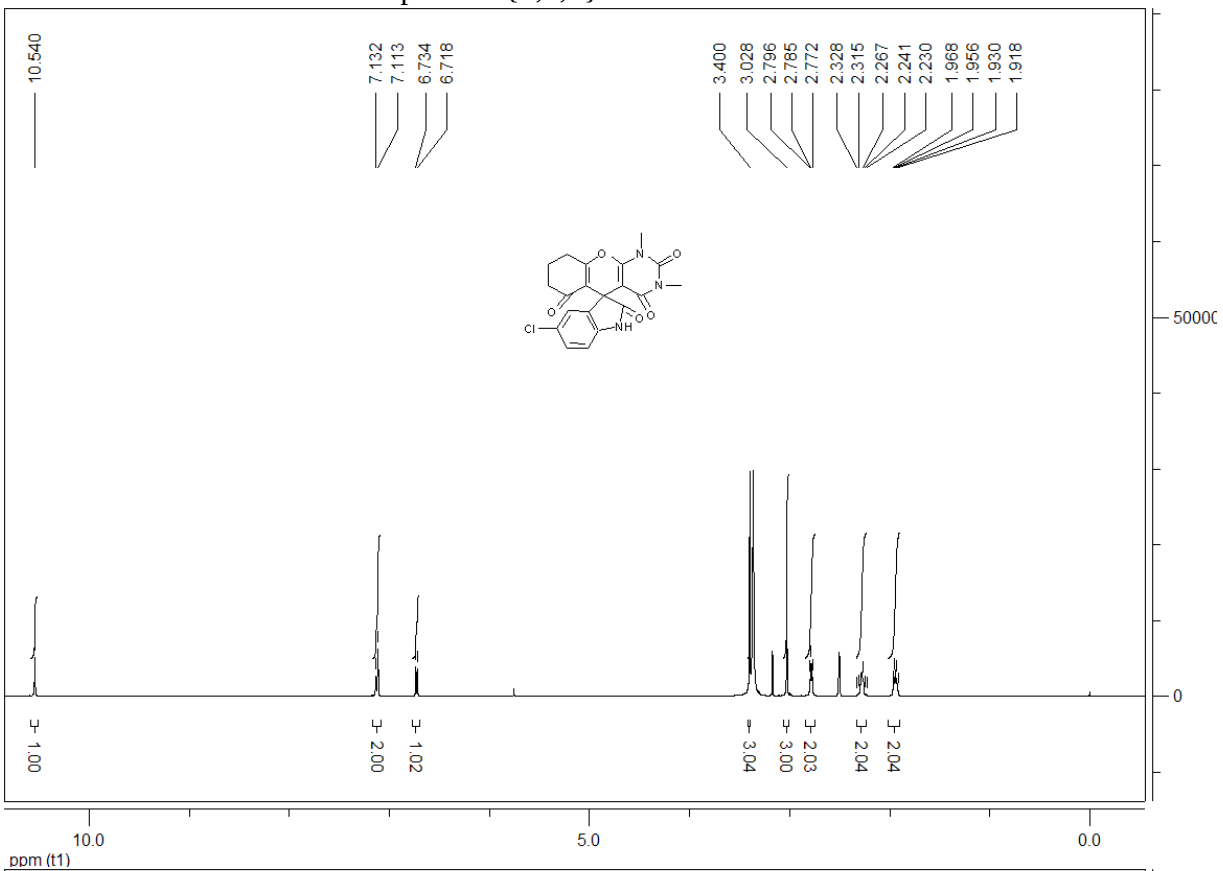



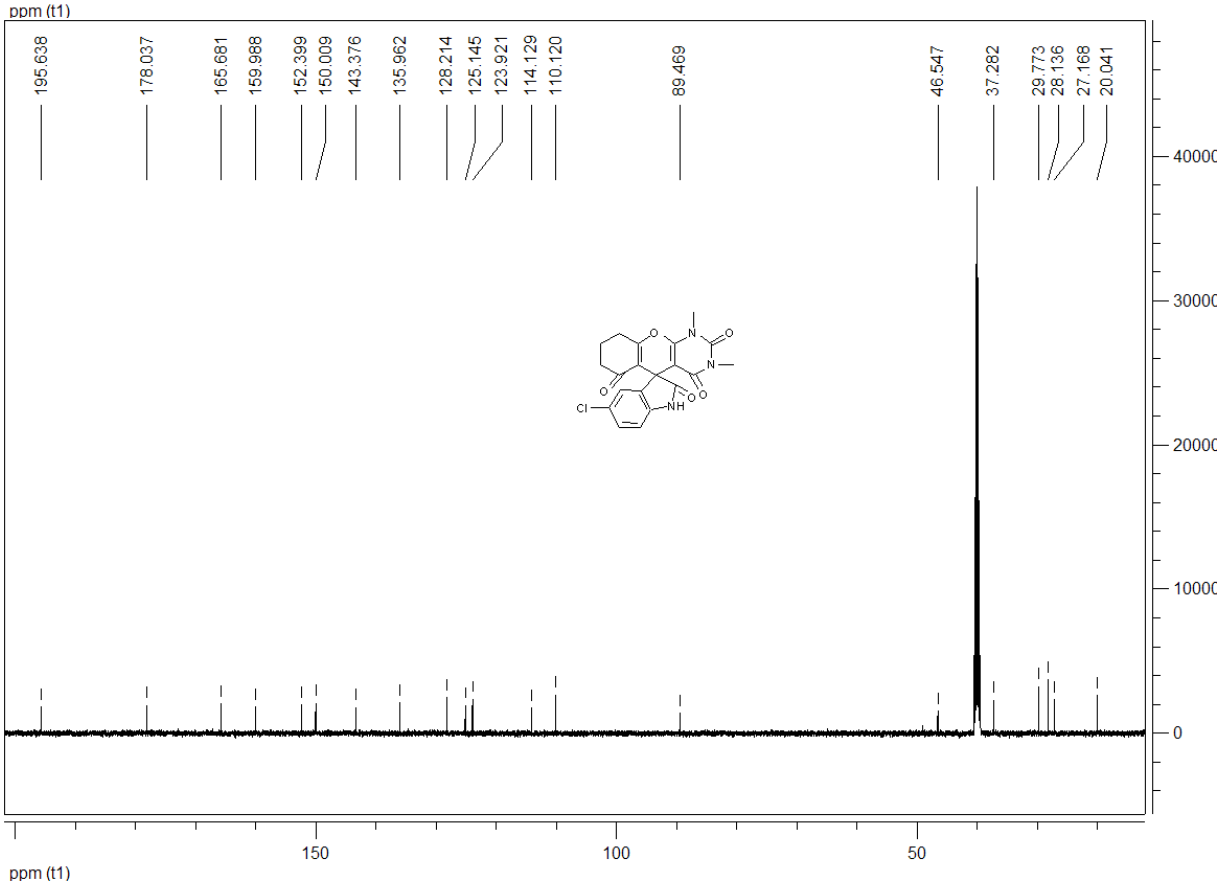
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,2,4*}



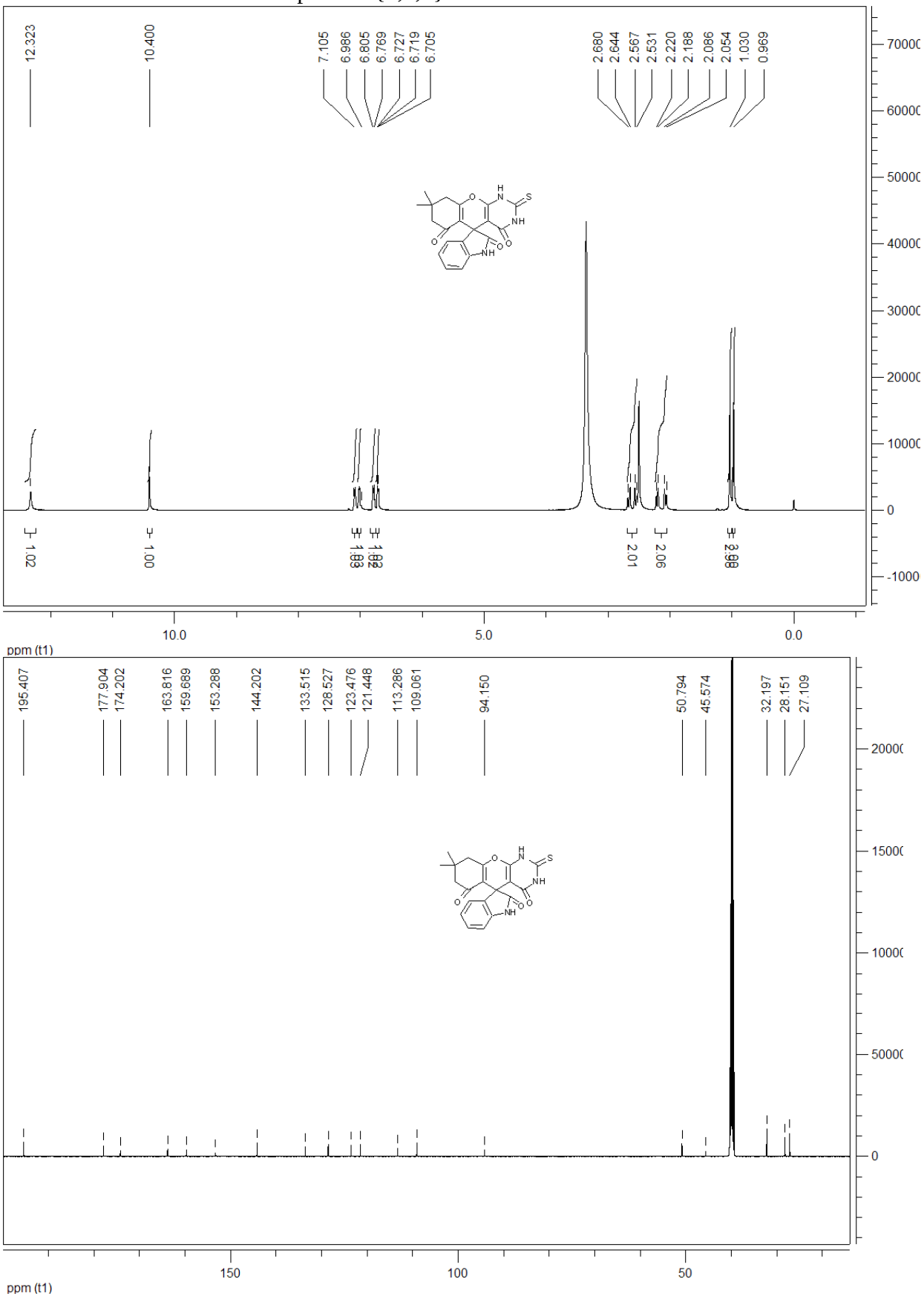



$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,2,5*}

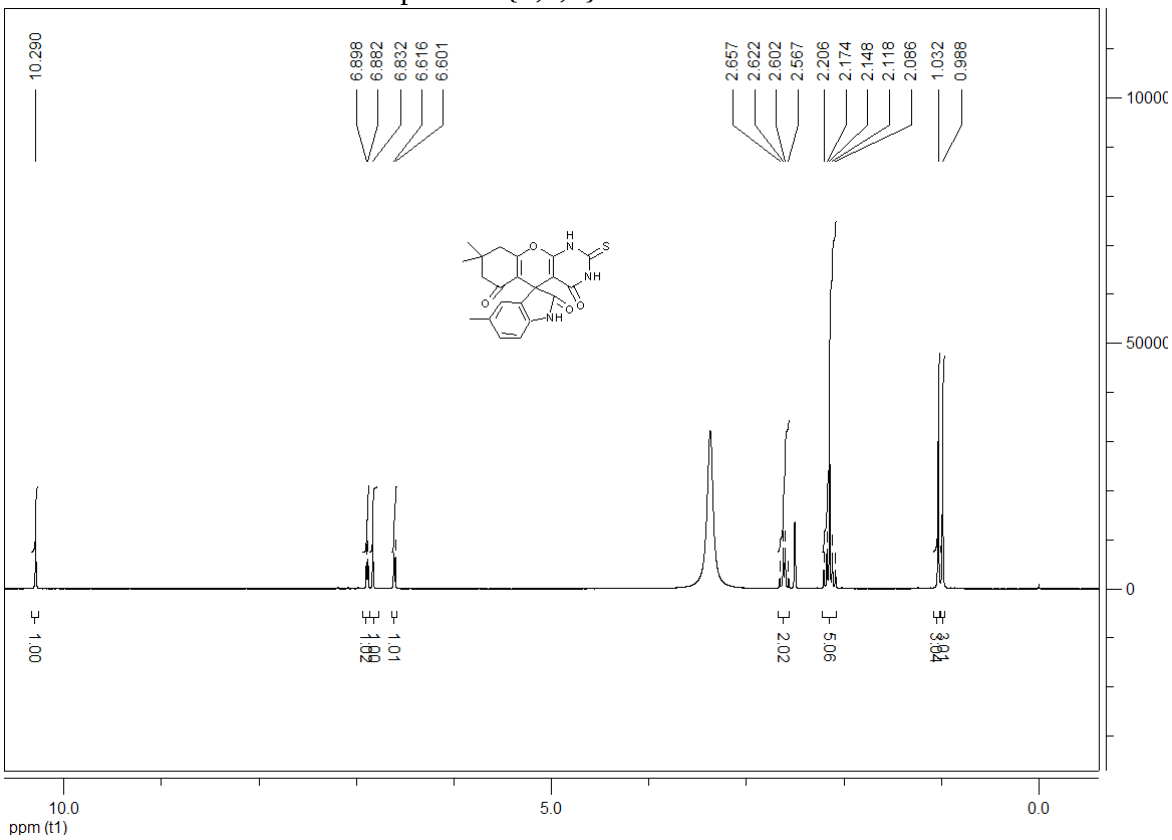


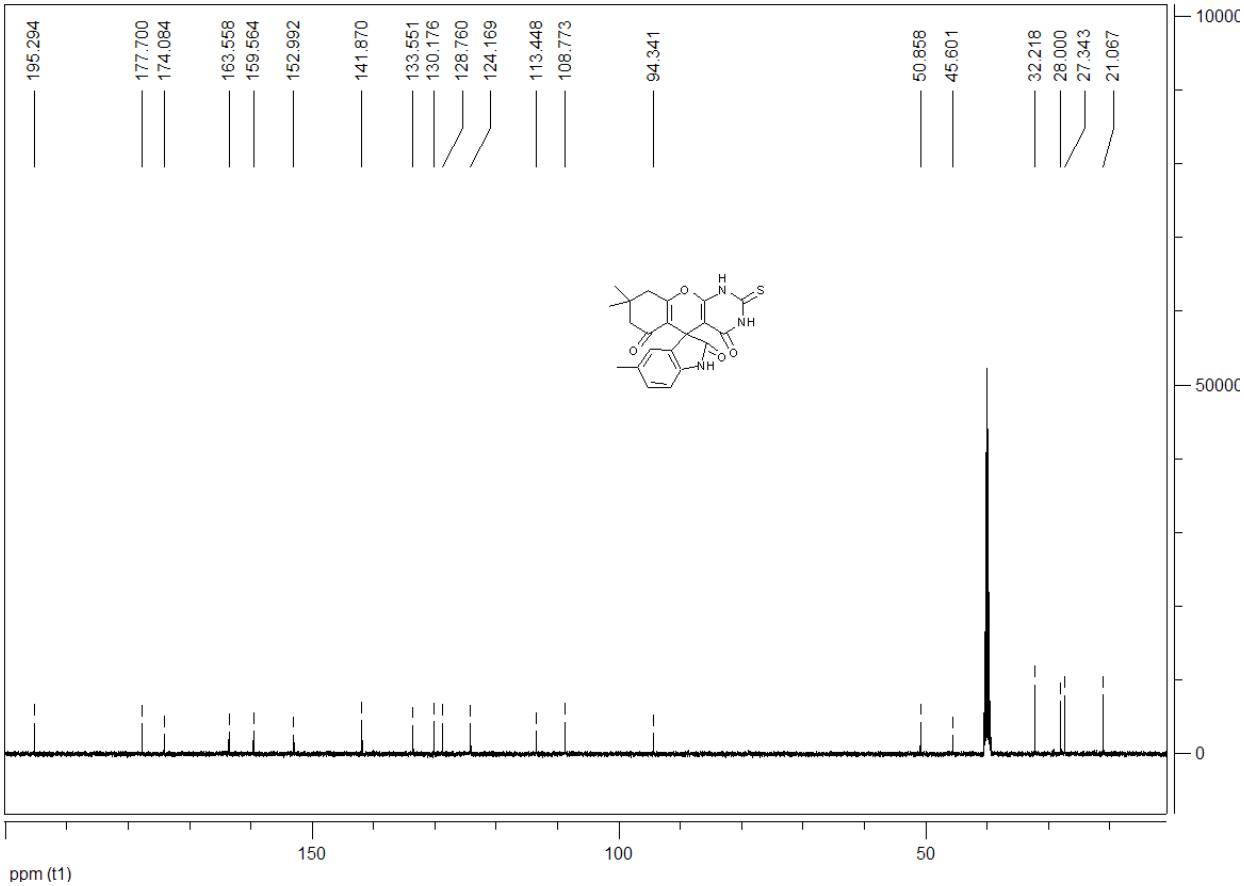



$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,3,1*}

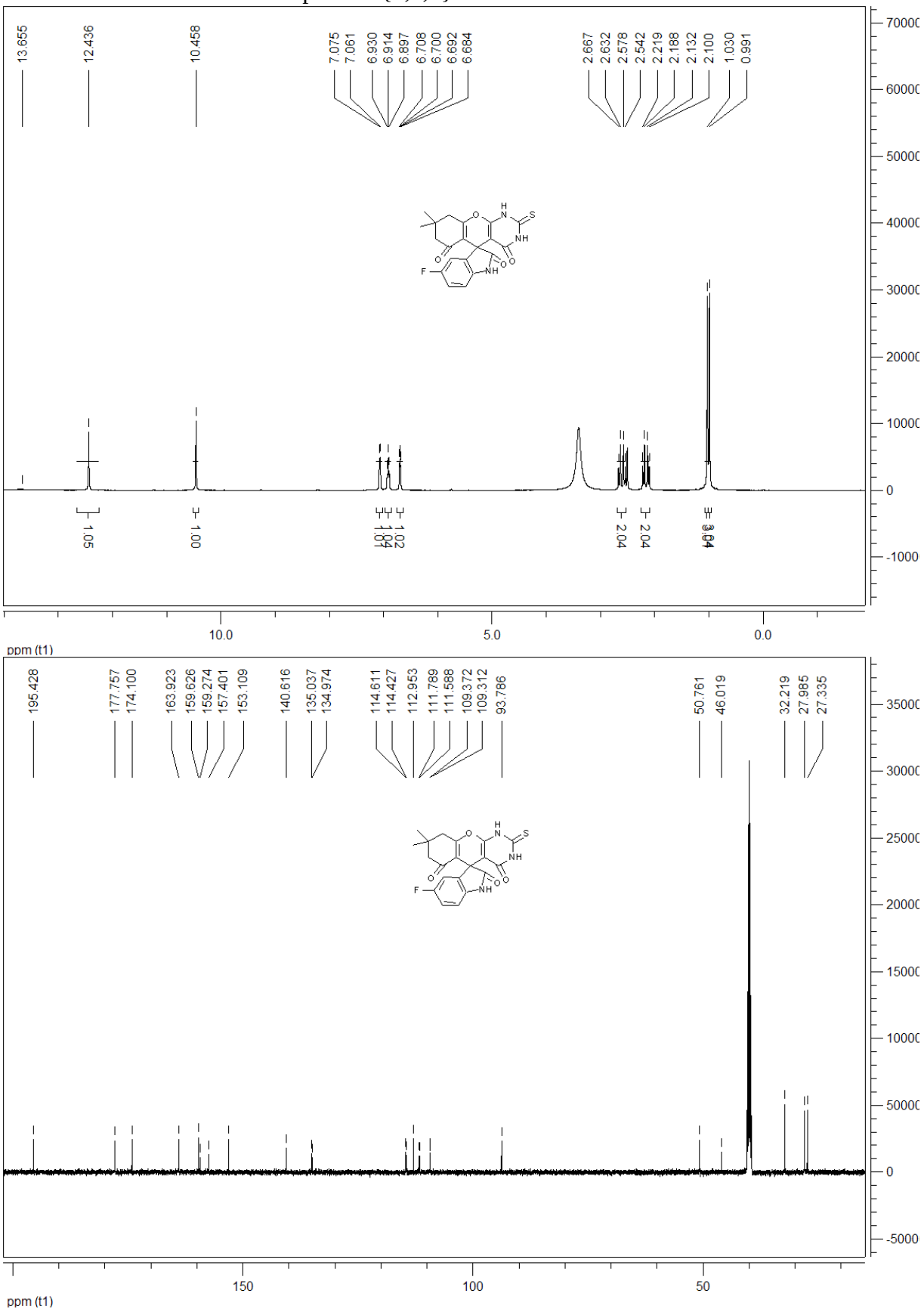


$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,3,2*}

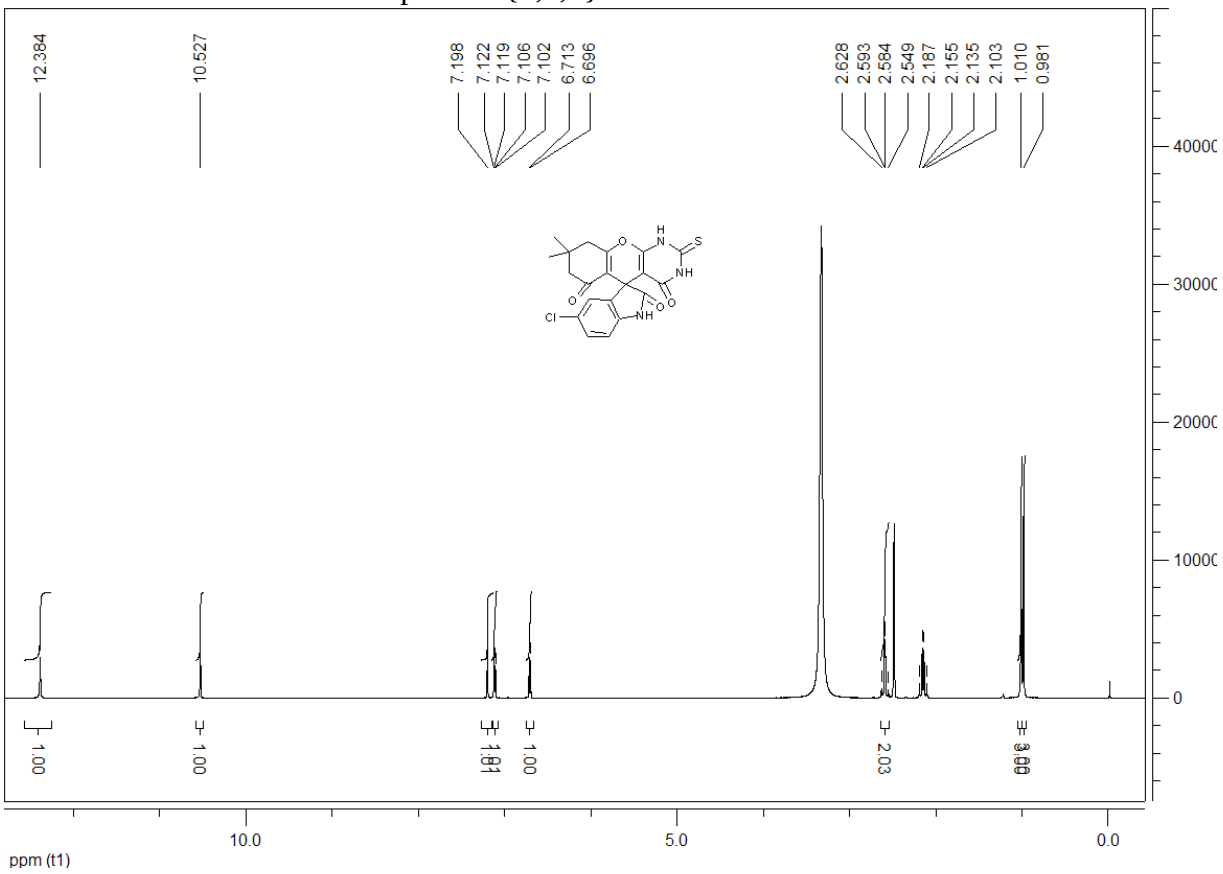


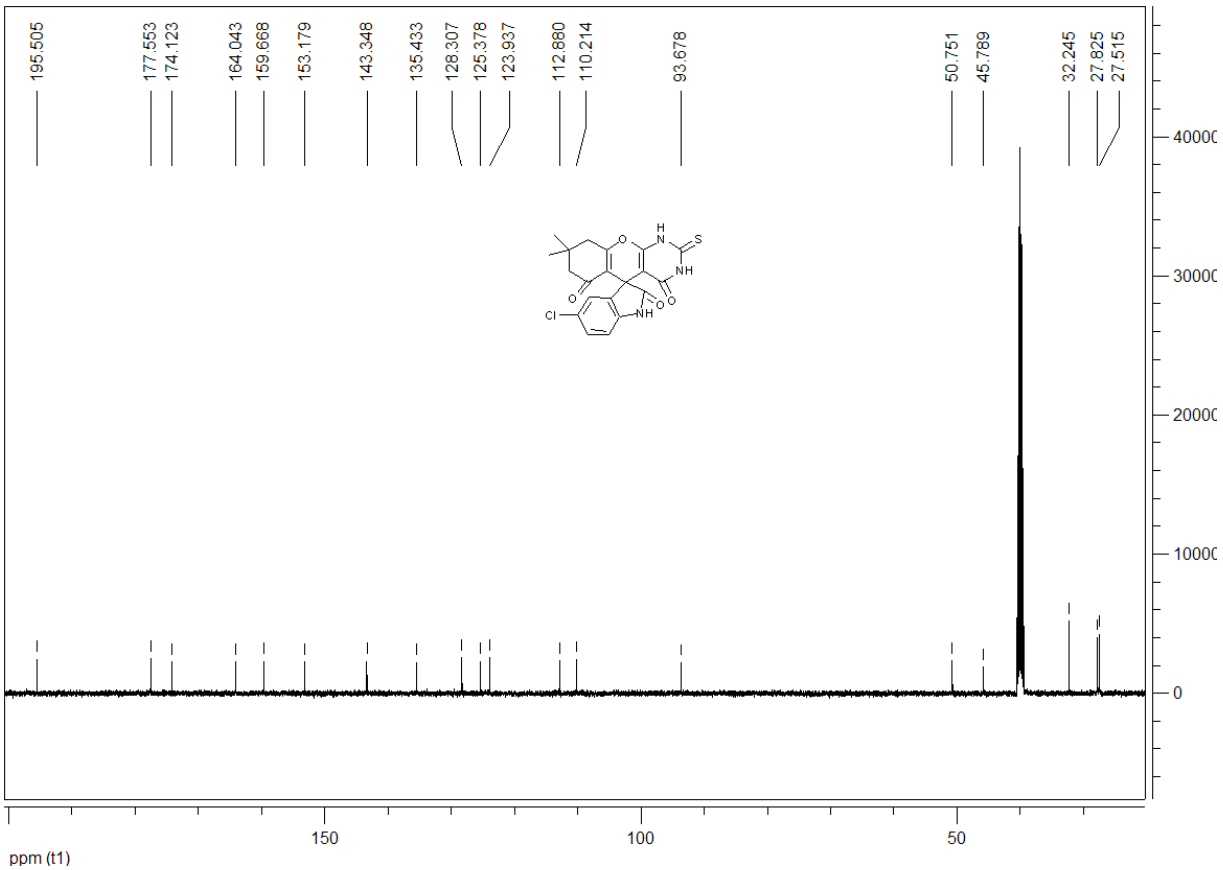



$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,3,4*}

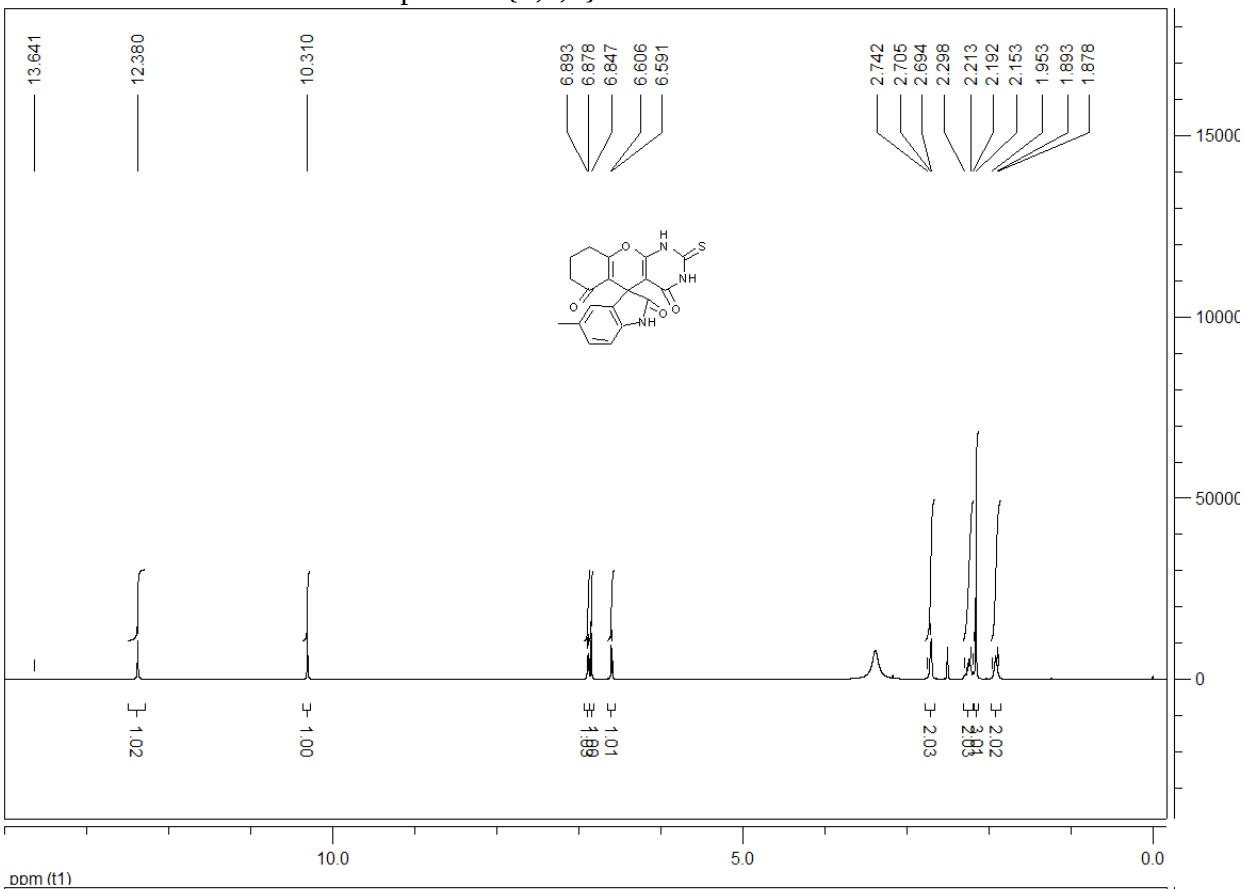


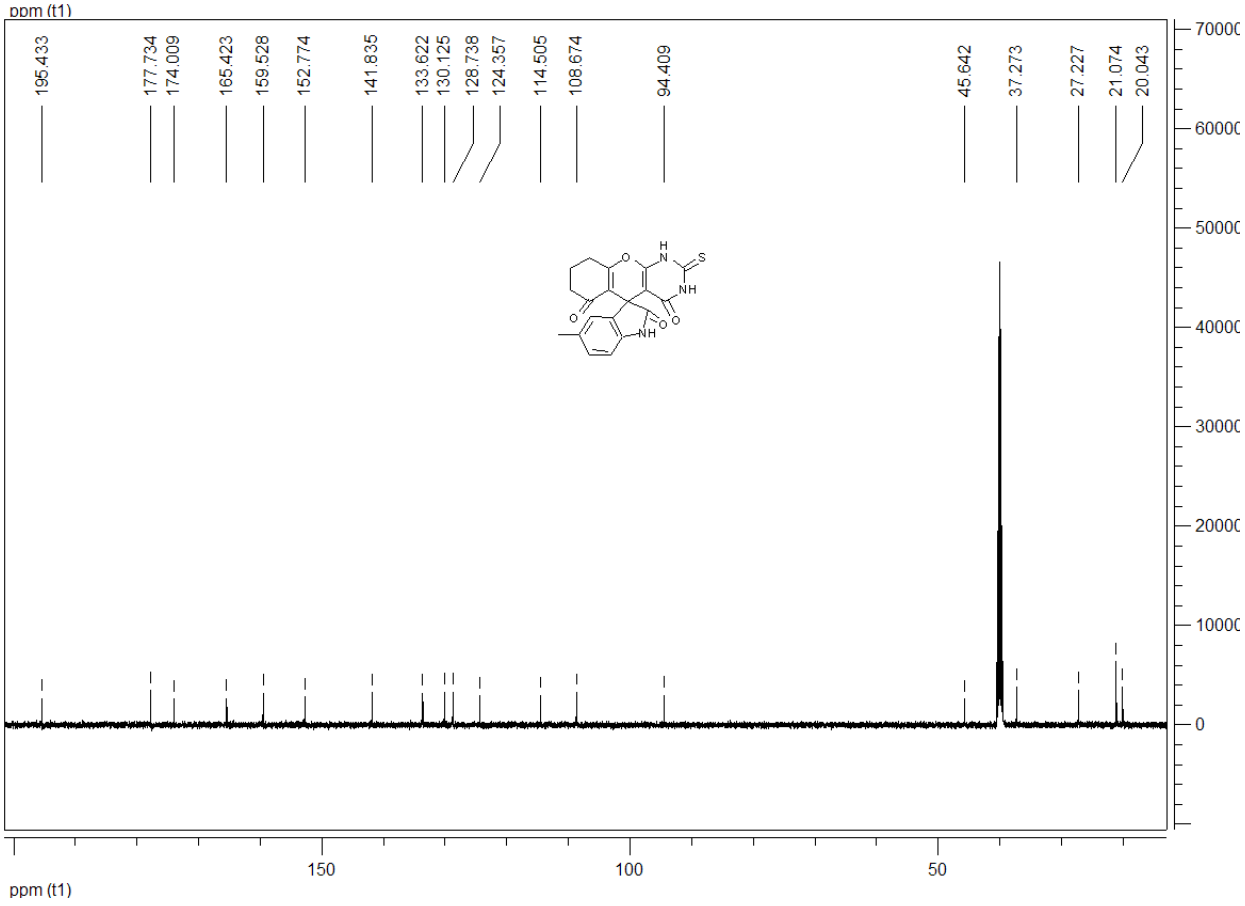
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*1,3,5*}



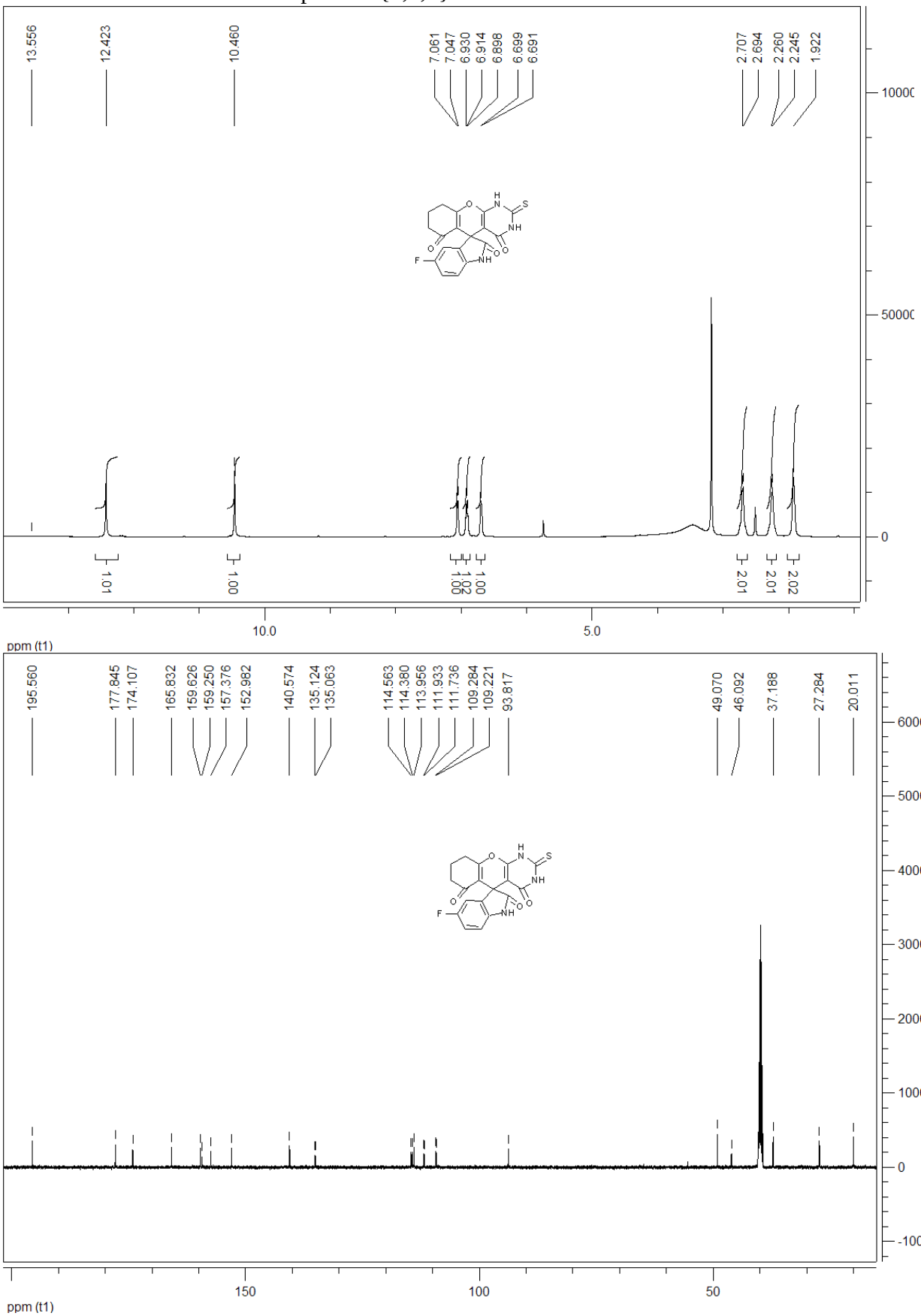



$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,3,2*}

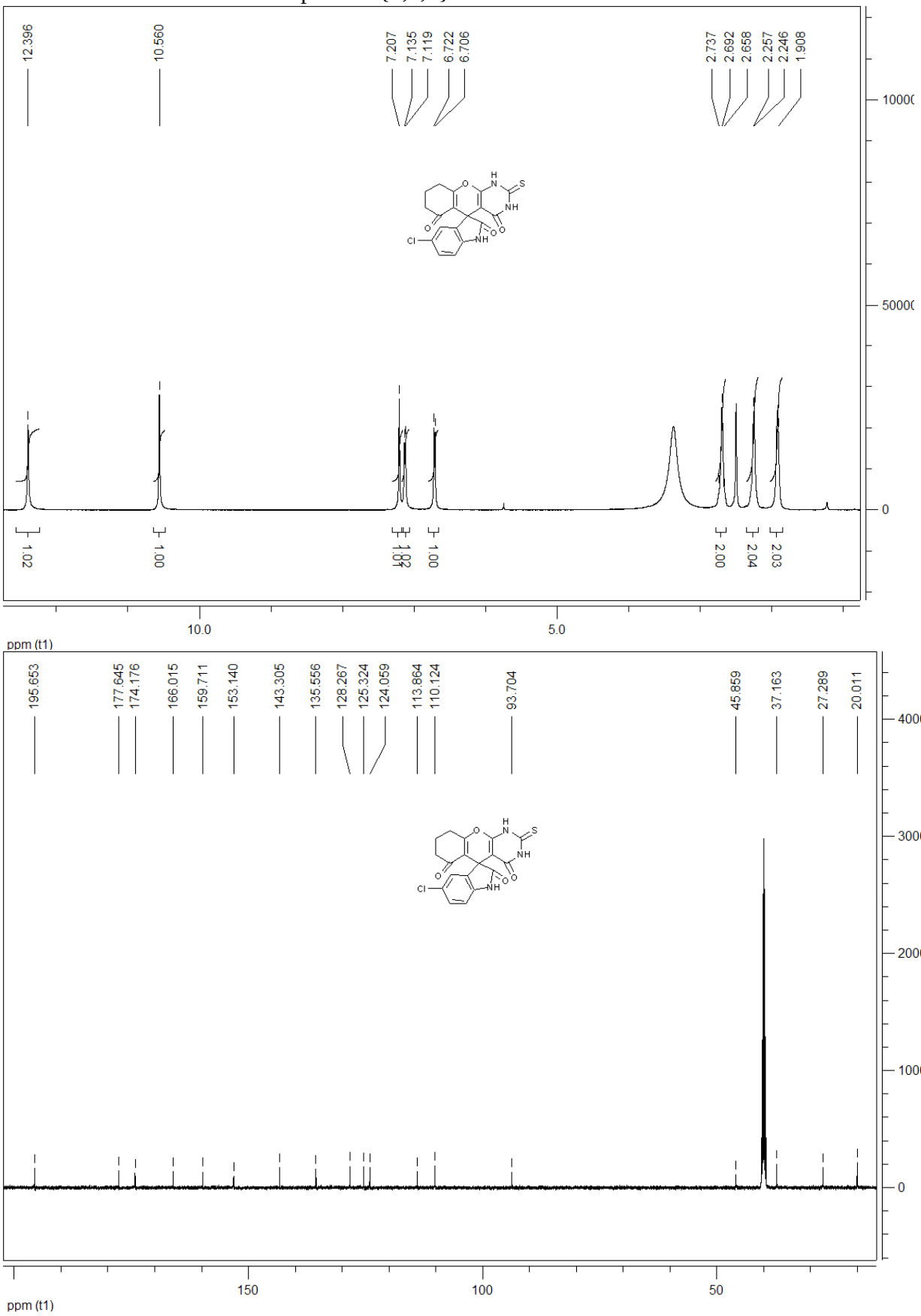


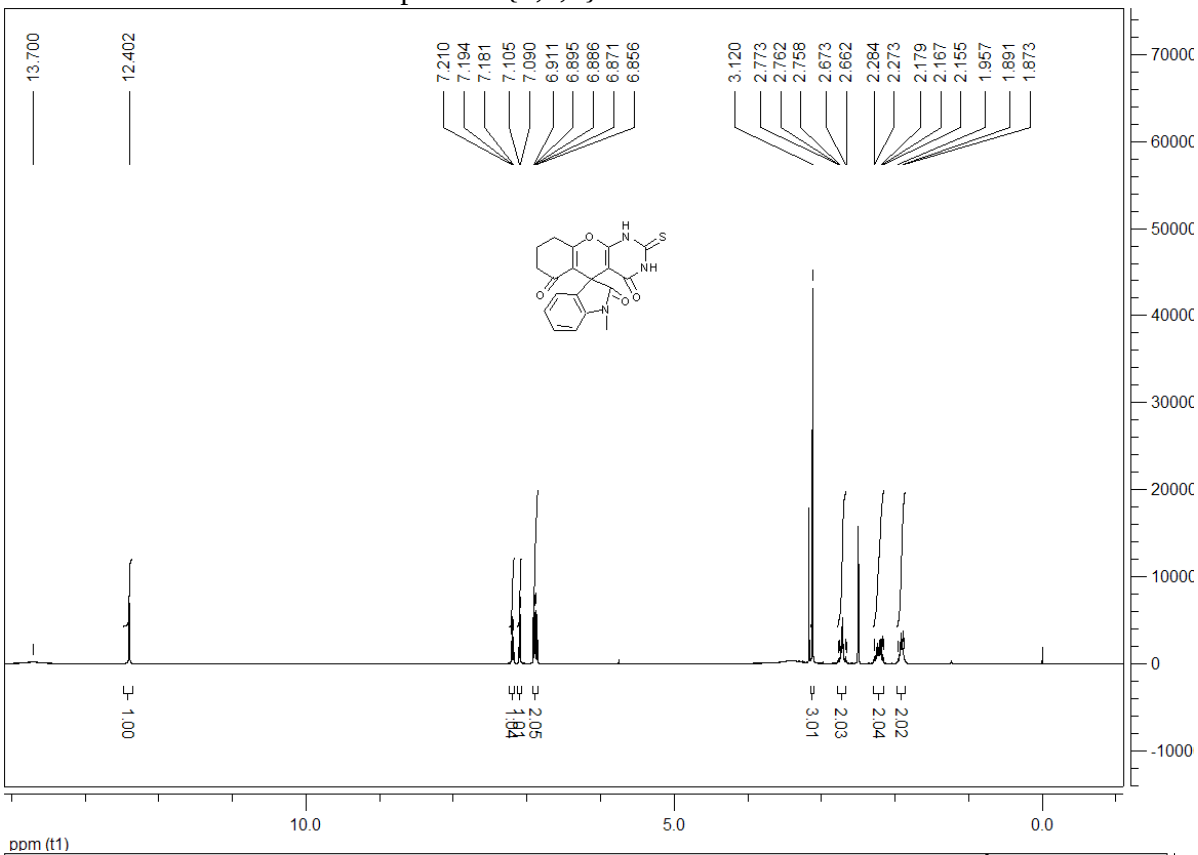
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,3,4*}

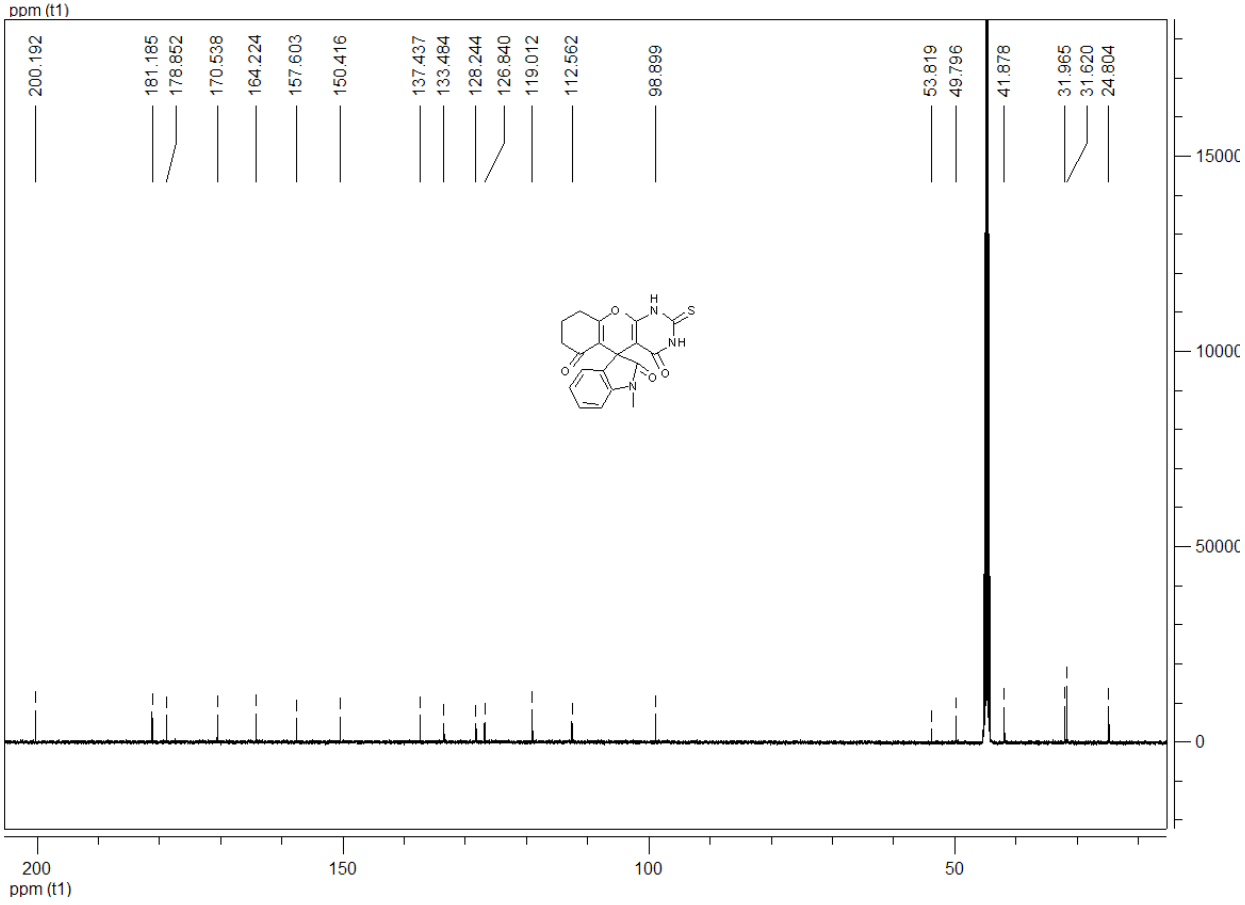


$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,3,5*}

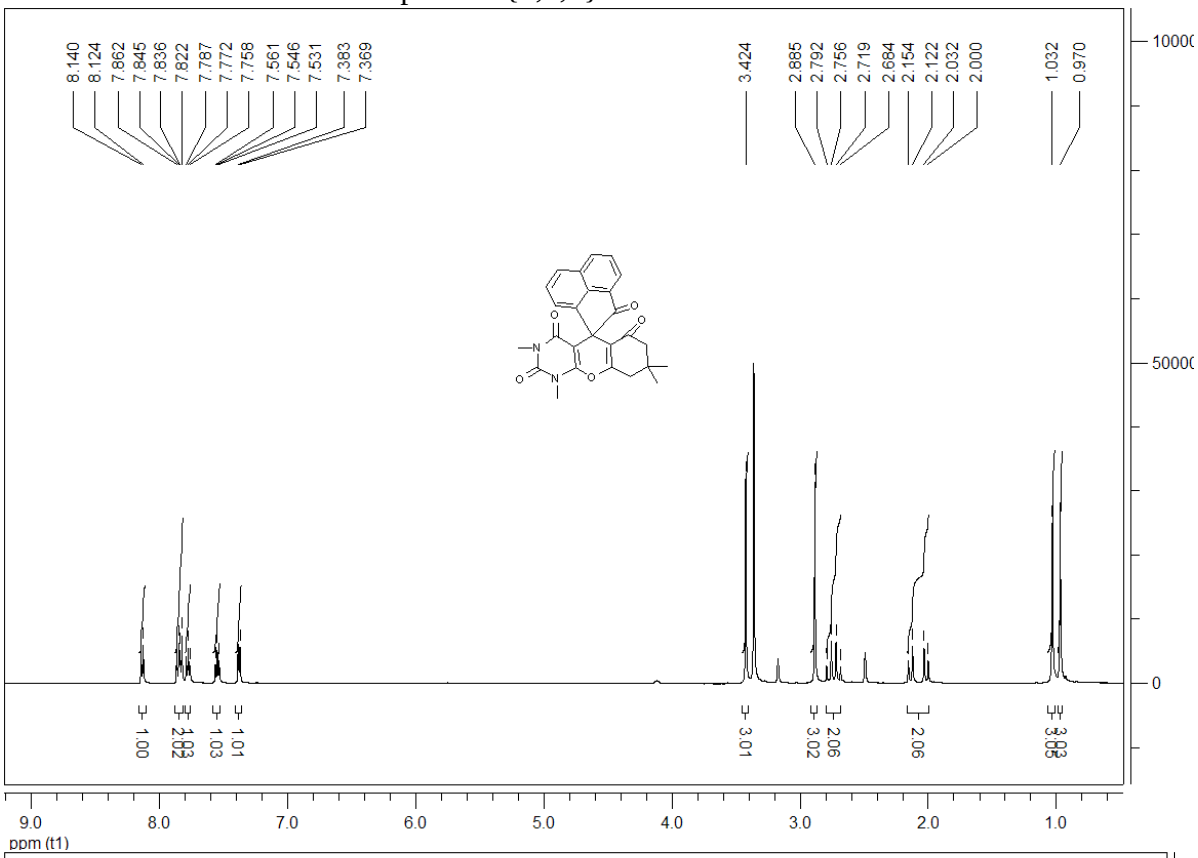


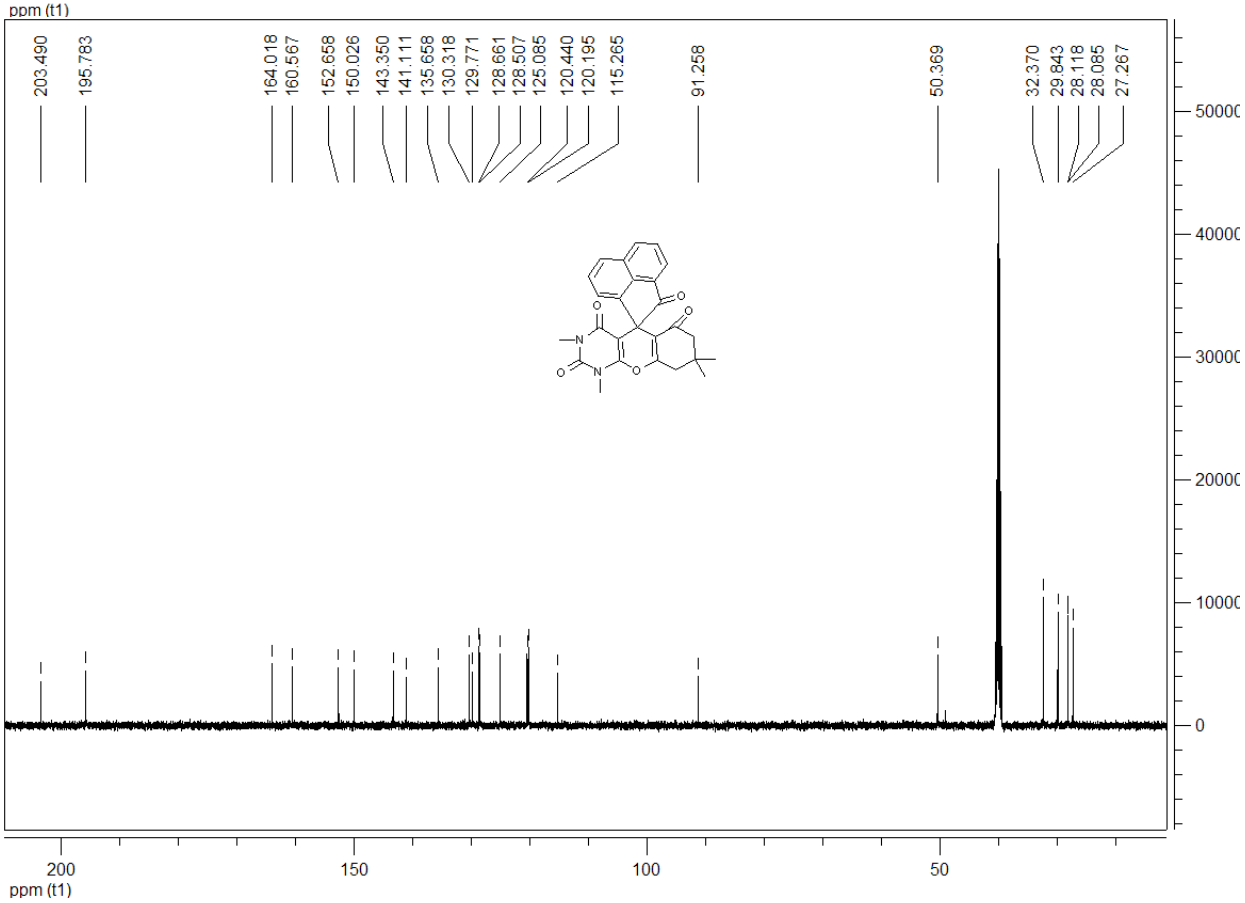
$^{1}$H NMR and $^{13}$C NMR of compound **4**{*2,3,8*}



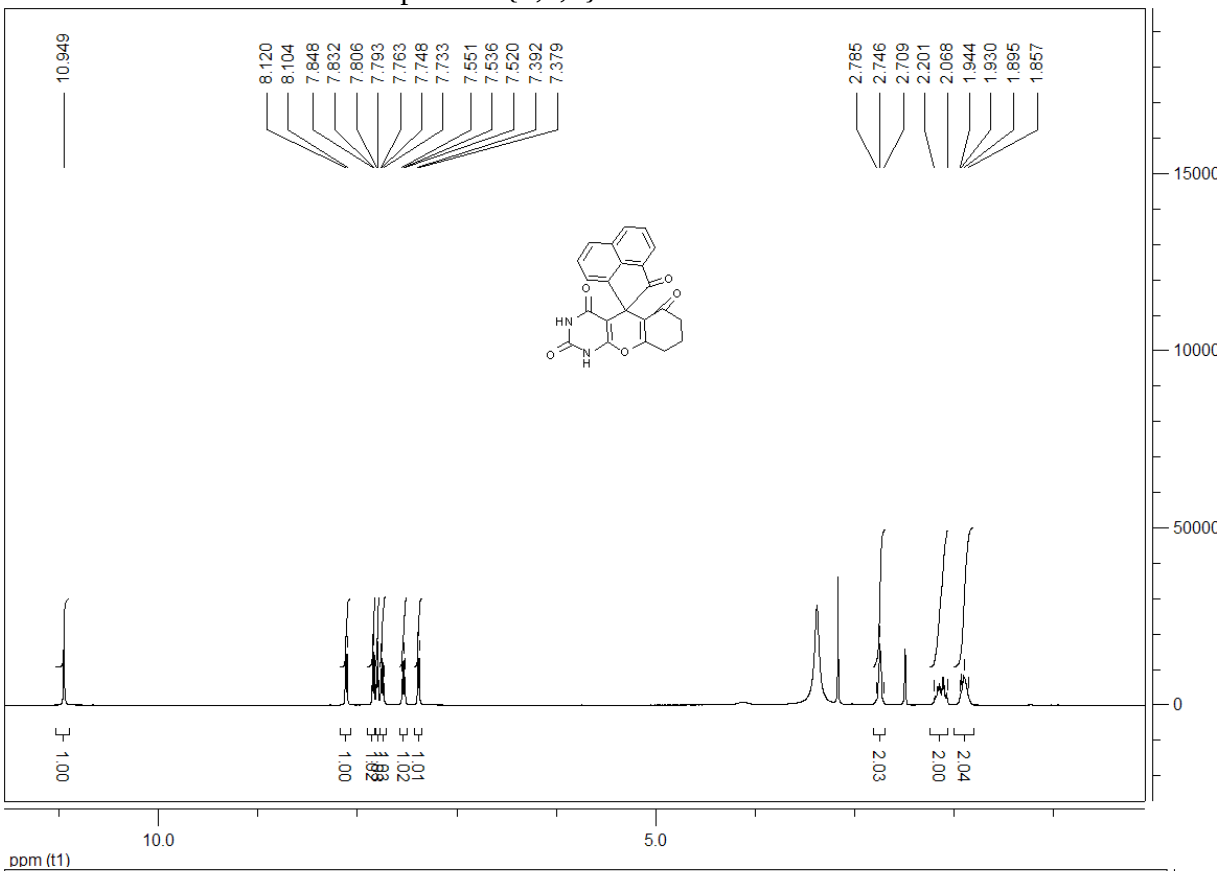



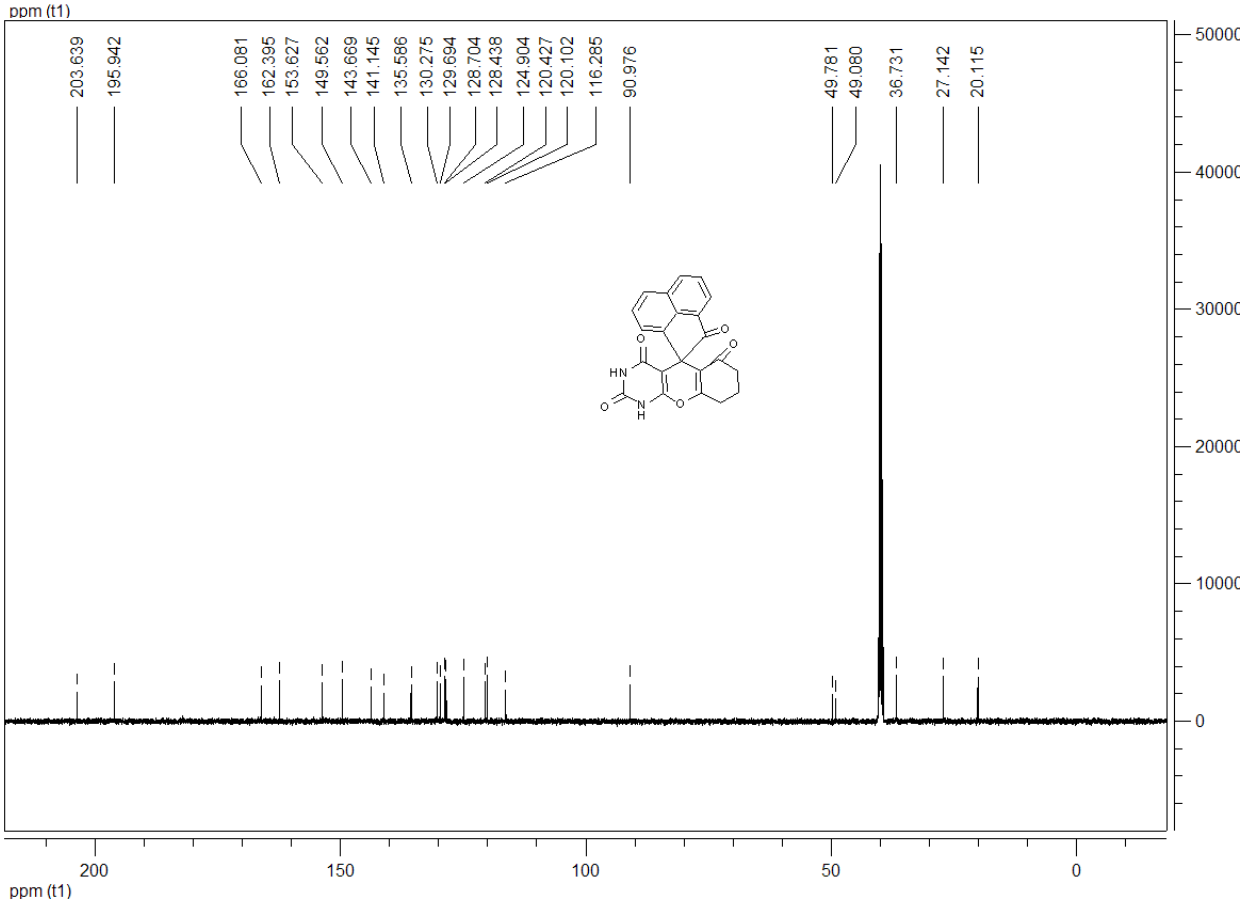
$^{1}$H NMR and $^{13}$C NMR of compound **6**{*1,2,5*}



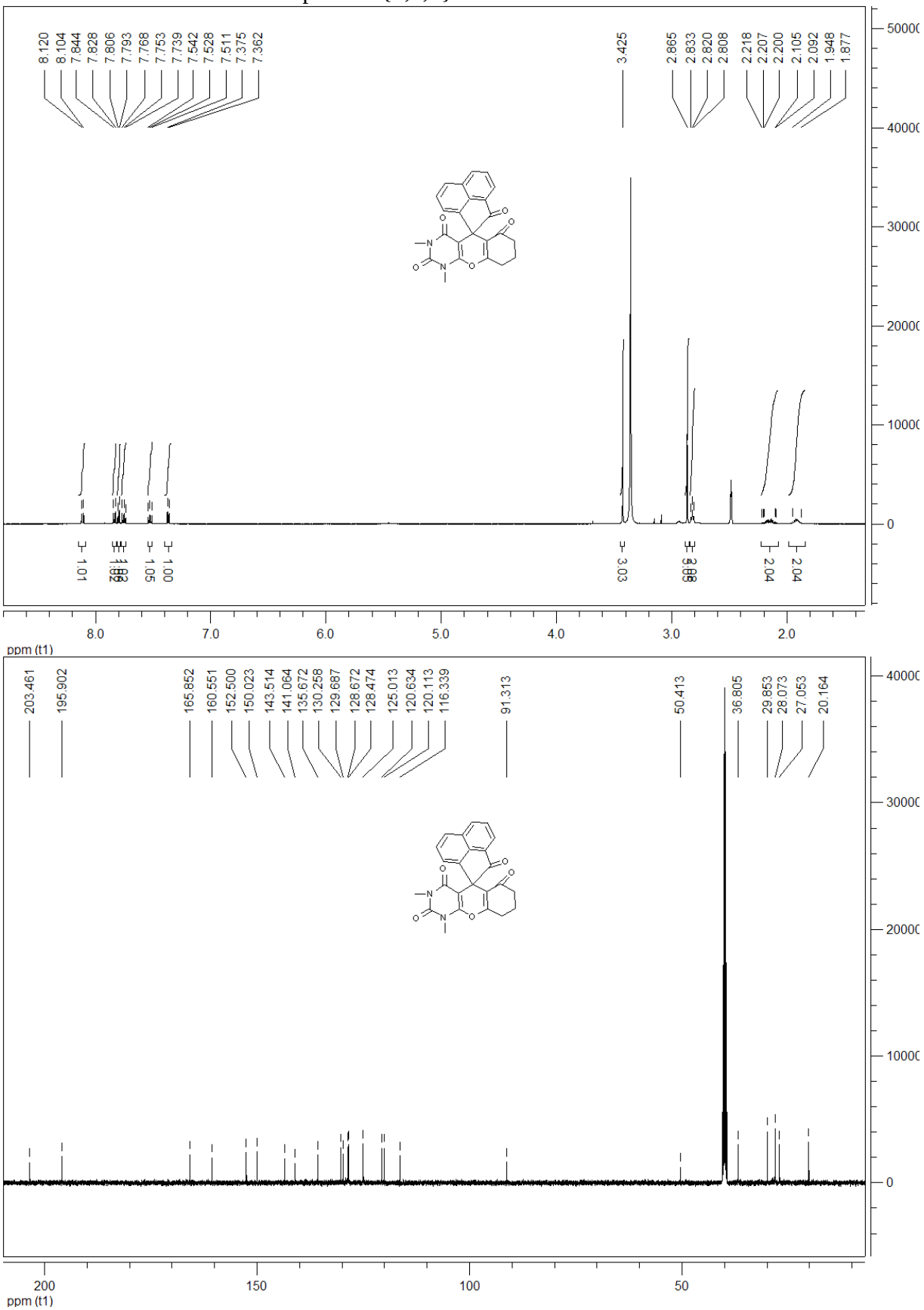



$^{1}$H NMR and $^{13}$C NMR of compound **6**{*2,1,5*}



$^{1}$H NMR and $^{13}$C NMR of compound **6**{*2,2,**5***}



$^{1}$H NMR and $^{13}$C NMR of compound **6**{*2,3,5*}