Supporting Information

A convenient metal free approach towards the synthesis of dihydropyrimidones mediated by achiral nicotinic acid without solvent

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Received 17 April 2021; accepted (revised) 9 September 2021

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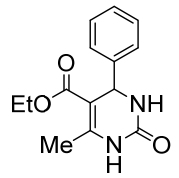
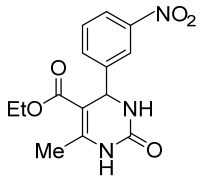
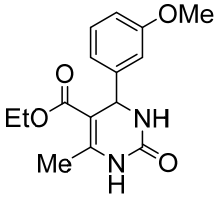
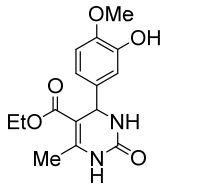
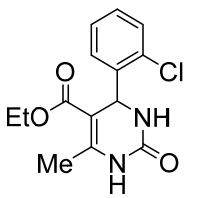
General Methods and Experimental Procedures:

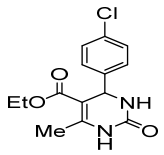
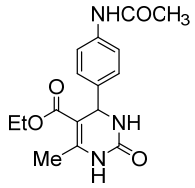
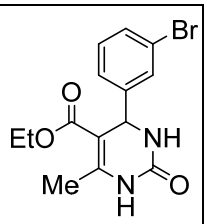
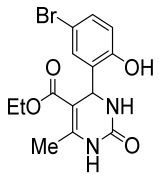
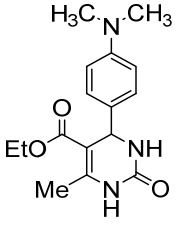
The melting points were determined on a capillary melting point apparatus and were matched with the literature values. Infrared spectra were recorded using KBr pellets for solids and neat for liquids on FT-IR 8400 Perkin-Elmer 883 grating spectrometers.^{1}H NMR spectra were taken on AC-Bruker 400 MHz spectrometer in DMSO and contained TMS as internal standard. All *J* values are given in Hz, chemical shifts in δ-units, and the necessary spectra were given as supportingfile. Progress of the reactions were monitored by TLC.

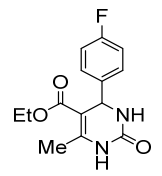
General Procedure for the Synthesis of Dihydropyrimidinone / thione Derivatives;

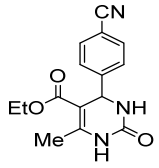
Pure aldehyde (1.0 mmol), ethyl acetoacetate (1.0 mmol), urea/thio-urea (1.2 mmol), and nicotinic acid (5-10 mol percent) were thoroughly mixed and stirred at room temperature for 30 minutes with an effective $CaCl_2$ guard tube protecting the reaction vessel. After 30 minutes of stirring at room temperature, there were no discernible changes found in the TLC.The reaction mixture was then steadily heated to 100□105^{0}C, and the reaction progress was detected after a few minutes by the appearance of solid over the reaction mixture. Stirring was continued at about 1hour till the completion of the reaction. After that, crushed ice was added to the reaction mixture, scratched, and the product was filtered.The isolated product is sufficiently pure, as shown by the ^{1}H NMR spectroscopy. To obtain analytic grade sample, the isolated product was recrystallized from ethanol.

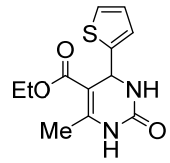
1. Spectral data of products are given below:

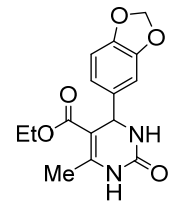
Ethyl-6-methyl-4-phenyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4a): The product was prepared following the general procedure given at page 1. Isolated yield 82%, m.p. found 202-204^{0}C, (lit. 204-206^{0}C) [16];**IR (KBr)**: 3242, 3117, 2980, 1726, 1701, 1647, 1462 cm^{-1}; **^{1}H NMR(200MHz, d_6-DMSO)** δ 9.45 (s, 1H, N<u>H</u>), 7.99 (s, 1H,Ar-<u>H</u>), 7.57-7.55 (m, 5H, Ar-<u>H</u>), 5.40 (brs, 1H, N<u>H</u>), 4.24 (q, *J*= 3.6Hz, 2H, C<u>H</u>$_2$), 2.75 (s, 1H, -C<u>H</u>), 2.50 (s, 3H, C<u>H</u>$_3$), 1.34 (t, *J*= 4.5Hz, 3H, -C<u>H</u>$_3$); **^{13}C-NMR (100 MHz, DMSO-d6**): δ 14.5, 18.2, 54.4, 59.7, 99.8, 126.7, 127.8, 128.9, 145.3, 148.8, 152.6, 165.8.	
Ethyl-6-methyl-2-oxo-4-(m-nitralyl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4b): The product was prepared by following the general procedure at page 1. Isolated yield 80%, Off-white solid, m.p. found 226-227^{0}C, (lit. 227-228^{0}C) [33];**^{1}H NMR (400 MHz**, ***d*$_6$-DMSO)**: δ 9.36 (s, 1H, N-<u>H</u>), 8.14 (d, 1H, *J*= 2Hz, Ar-<u>H</u>), 8.08 (s, 1H, Ar-<u>H</u>), 7.90 (s, 1H, N-<u>H</u>), 7.72-7.76(m, 1H, Ar-<u>H</u>), 5.30 (s, 1H,-C<u>H</u>), 3.99-3.98 (m, 2H,-C<u>H</u>$_2$), 2.27 (s, 3H, -C<u>H</u>$_3$), 1.09 (t, 3H, *J*= 7Hz, -C<u>H</u>$_3$);**^{13}C-NMR(100MHz, *d*$_6$-DMSO):** δ 14.4, 18.1, 51.9, 59.5, 98.3, 128.2, 129.3, 129.5, 129.8, 132.2, 142.2, 149.7, 151.8, 165.4 ppm.	
Ethyl4-(3-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4c): The product was prepared by following the general procedure at page 1. Isolated yield (83%), White powder, m.p., found 206-208^{0}C, (lit. 207-208^{0}C) [16]; **^{1}H NMR(400MHz, *d*$_6$-DMSO),** δ 9.17 (s, 1H, N-<u>H</u>), 7.72 (s, 1H, N-<u>H</u>), 7.24 (t, 1H, *J*= 7Hz, Ar-<u>H</u>), 6.83-6.79 (m, 3H, Ar-<u>H</u>), 5.12 (s, 1H, -C<u>H</u>), 4.0 (q, 2H, *J*= 7Hz, -C<u>H</u>$_2$), 3.72 (s, 3H, -OC<u>H</u>$_3$), 2.25 (s, 3H, -C<u>H</u>$_3$), 1.14 (t, 3H, *J*= 7Hz, -C<u>H</u>$_3$); **^{13}C NMR (100MHz, *d*$_6$-DMSO):** δ 14.5, 18.2, 54.2, 55.4, 59.7, 99.6, 112.6, 112.8, 118.7, 130.0, 146.7, 148.8, 152.7, 159.6, 165.8 ppm.	
Ethyl-4-(3-hydroxy-4-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4d): The product was prepared by following the general procedure at page 1. Isolated yield (85%), Pale yellow solid, m.p., found 184-185^{0}C;**^{1}H NMR(400MHz, *d*$_6$-DMSO),** □ δ 9.12 (s, 1H, N-<u>H</u>), 8.93 (s, 1H, Ar-<u>H</u>), 7.62 (s, 1H, N-<u>H</u>), 6.83 (d, 1H, *J*= 8Hz, Ar-<u>H</u>), 6.70 (s, 1H, Ar-<u>H</u>), 6.62 (d, 1H, *J*= 10Hz, Ar-<u>H</u>), 5.04 (s, 1H, -C<u>H</u>), 4.01 (q, 2H, *J*= 7Hz, -C<u>H</u>$_2$), 3.72 (s, 3H, -OC<u>H</u>$_3$), 2.24 (s, 3H, -C<u>H</u>$_3$), 1.12 (t, 3H, *J*= 7Hz, -C<u>H</u>$_3$);**^{13}C NMR (100MHz, *d*$_6$-DMSO),** δ 14.6, 18.2, 53.9, 56.1, 59.6, 100.2, 112.5, 114.1, 117.4, 138.0, 146.7, 147.3, 148.1, 152.7, 165.9 ppm.	
Ethyl-4-(2-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4e): The product was prepared by following the general procedure at page 1. Isolated yield (85%), White powder, m.p. found 221-223^{0}C, (lit.222-224^{0}C) [33];**^{1}H NMR(400MHz, *d*$_6$-DMSO),** δ 9.25 (s, 1H, N-<u>H</u>), 7.70 (s, 1H, N-<u>H</u>), 7.40 (d, 1H , *J*= 4Hz, Ar-<u>H</u>), 7.29 (s, 2H, Ar-<u>H</u>), 5.65 (s, 1H, -C<u>H</u>), 3.89 (q, 2H, *J*= 7Hz, -C<u>H</u>$_2$), 2.30 (s, 3H, -C<u>H</u>$_3$), 1.00 (t, 3H, *J*= 7Hz, -C<u>H</u>$_3$);**^{13}C NMR (100MHz, *d*$_6$-DMSO**), δ 14.4, 18.1, 51.9, 59.5, 98.3, 128.2, 129.3, 129.5, 129.8, 132.2, 142.2, 149.7, 151.8, 165.4 ppm. HRMS Calcd. for $C_{14}H_{15}N_2O_2Cl$: 294.7745, found: 294.7742.	

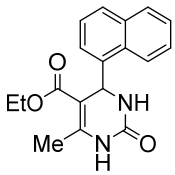
Ethyl-4-(4-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4f): The product was prepared by following the general procedure at page 1. Isolated yield (85%), m.p. found 234-236^{0}C, (lit. 233-234^{0}C) [42];**IR (KBr):** 3238, 3097, 1707, 1651, 1570, 1429 cm^{-1}; **^{1}H NMR(200MHz, d_6-DMSO)**; δ 9.53(s, 1H, NH), 8.05(s, 2H, Ar-H), 7.71-7.47(m, 2H, Ar-H), 5.39(brs, 1H, NH), 4.24(q, *J*= 3.5Hz, 2H, CH_2), 2.76(s, 1H, -CH), 2.50(s, 3H, CH_3), 1.35(t, *J*= 4.1Hz, 3H, -CH_3).	
Ethyl-4-(4-Acetamidophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4g): The product was prepared by following the general procedure at page 1. Isolated Yield (85%), Pale yellow powder, m.p. found 280-282^{0}C (new);**^{1}H-NMR (400MHz,d_6-DMSO)**, δ 9.91 (s, 1H, N-H), 9.14 (s, 1H, N-H), 7.66 (s, 1H, NH), 7.50 (d, 2H, *J*= 8Hz, Ar-H), 7.14 (d, 2H, *J*= 8Hz, Ar-H), 5.09 (s, 1H, -CH), 4.00-3.95 (q, 2H, *J*= 7Hz, -CH_2), 2.24 (s, 3H, -CH_3), 2.00(s, 3H, -CH_3), 1.01 (t, 3H, *J*= 7Hz, -CH_3);**^{13}C NMR (100MHz, d_6-DMSO),** δ 14.6, 18.2, 24.4, 54.0, 59.6, 99.8, 119.5, 127.0, 138.8, 140.0, 148.6, 152.6, 165.8, 168.7 ppm. HRMS Calcd. For $C_{16}H_{19}N_3O_4$: 317.3442, found: 317.3445.	
Ethyl-4-(3-bromophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4h): The product was prepared by following the general procedure at page 1. Isolated yield (83%), White powder, m.p. found 183-185^{0}C, (lit.185-186^{0}C) [33]; **^{1}H NMR(400MHz, d_6-DMSO),** δ 9.27 (s, 1H, N-H), 7.80 (s, 1H, N-H),7.45 (d, 1H, *J*= 6Hz, Ar-H), 7.40 (s, 1H, Ar-H), 7.30 (t, 1H, *J*= 7Hz, Ar-H), 7.24 (d, 1H, *J*= 1Hz, Ar-H), 5.16 (s, 1H, -CH), 4.04-3.96 (m, 2H, -CH_2), 2.24 (s, 3H, -CH_3), 1.10 (t, 3H, *J*=7Hz, -CH_3); **^{13}C NMR (100MHz, d_6-DMSO),** δ = 14.5, 18.3, 54.0, 59.8, 99.1, 122.0, 125.7, 129.6, 130.6, 131.3, 147.9, 149.4, 152.4, 165.6 ppm.	
Ethyl 4-(5-bromo-2-hydroxyphenyl)-6-methyl-2-oxo-1,2,3,4 tetrahydropyrimidine-5-carboxylate(4i): The product was prepared by following the general procedure at page 1. Isolated yield (83%), Gray colour solid, mp. found290-291^{0}C, (lit. 292-293^{0}C) [17]; **^{1}H NMR (400MHz, d_6-DMSO)**, δ 9.96 (s, 1H, N-H), 9.15 (s, 1H, Ar-H), 7.24 (s, 1H, N-H), 7.04 (s, 1H, Ar-H), 6.76 (d, 1H, *J*= 8Hz, Ar-H), 5.40 (s, 1H, -CH), 3.94 (q, 2H, *J*= 4Hz, -CH_2), 2.26 (s, 3H,-CH_3), 1.05 (t, 3H, *J*=7Hz,-CH_3);**^{13}C NMR (100MHz, d_6-DMSO),** δ 14.4, 18.2, 50.0, 59.5, 97.6, 110.1, 118.2, 130.5, 131.3, 133.0, 149.4, 152.5, 154.7, 165.8 ppm.	
Ethyl-4-(4-(dimethylamino)-phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4j): The product was prepared by following the general procedure at page 1. Isolated yield (85%), Yellow solid, m.p. found 256-258^{0}C, lit. 257-258^{0}C) [16];**^{1}H NMR (400MHz, d_6-DMSO)**, δ 9.08 (s, 1H, N-H), 7.58 (s, 1H, N-H), 7.04 (d, 2H, *J*= 4Hz, Ar-H), 6.65 (d, 2H, *J*= 2Hz, Ar-H), 5.05 (s, 1H, -CH), 4.01 (q, 2H, *J*= 5Hz, -CH_2), 2.84(s, 6H, -CH_3), 2.23(s, 3H, -CH_3),1.10 (s, 3H, *J*= 7Hz, -CH_3);**^{13}C NMR (100MHz, d_6-DMSO**), δ 14.6, 18.2, 53.8, 59.6, 100.4, 112.7, 127.4, 133.1, 148.0, 150.2, 152.8, 165.9 ppm.	

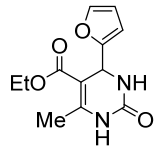
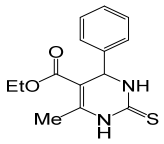
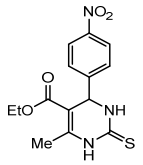
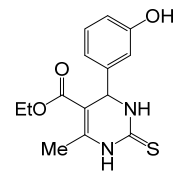
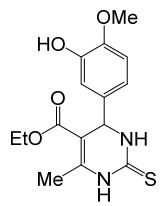
Ethyl-4-(4-fluorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4k):The product was prepared by following the general procedure at page 1. Isolated yield (82%), Pale yellow solid, m.p. found 181-183^{0}C, (lit. 182-184^{0}C) [16];**^{1}H NMR (400MHz, d_6-DMSO)**, δ 9.20 (s, 1H, N-H), 7.75 (s, 1H, N-H), 7.28(t, 2H, *J*= 2Hz, Ar-H), 7.26-7.12 (m, 2H, Ar-H), 5.16 (s, 1H, -CH), 4.01 (q, 2H, *J*= 5Hz, -CH_2), 2.26 (s, 3H, -CH_3), 1.08(t, 3H, *J*= 8Hz, -CH_3);**^{13}C NMR (100MHz, d_6-DMSO)**, δ 14.5, 18.2, 53.8, 99.6, 115.5, 128.6, 141.5, 148.9, 152.5, 160.5, 163.0, 165.7 ppm.	

Ethyl-4-(4-cyanophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4l): The product was prepared by following the general procedure at page 1. Isolated Yield (82%), White solid, m.p. found 233-235^{0}C(new); **^{1}H NMR (400MHz, d_6-DMSO**): δ 9.31 (s, 1H, N-H), 7.84 (s, 1H, N-H), 7.81 (d, 2H, *J*= 8Hz, Ar-H), 7.45 (d, 2H, *J*= 8Hz, Ar-H), 5.23 (s, 1H, -CH), 3.99 (q, 2H, *J*= 6Hz, -CH_2), 2.26 (s, 3H, -CH_3), 1.08 (t, 3H, *J*= 8Hz, -CH_3); **^{13}CNMR(100MHz, d_6-DMSO**), δ 14.4, 18.3, 54.3, 59.8, 98.7, 110.6, 119.3, 127.6, 132.54, 133.0, 149.7, 150.5, 152.3, 158.2, 165.6 ppm.	

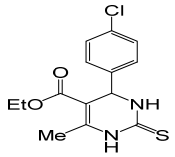
Ethyl-6-methyl-2-oxo-4-(thiophen-2-yl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4m): The product was prepared by following the general procedure at page 1. Isolated yield (84%), Pale yellow solid, m.p. found 214-217^{0}C, (lit. 215-217^{0}C) [21]; **^{1}H NMR (400MHz,d_6-DMSO)**, δ 9.31 (s, 1H , N-H), 7.90 (s, 1H, N-H), 7.35 (d, 1H, *J*= 3.8Hz, Ar-H), 6.95-6.89 (m, 2H, Ar-H), 5.4 (s, 1H, -CH), 4.01 (q, 2H, *J*= 7Hz, -CH_2), 2.24 (s, 3H, -CH_3), 1.10 (t, 3H, *J*= 7Hz, -CH_3);**^{13}C NMR (100MHz, d_6-DMSO**), δ 14.6, 18.1, 49.8, 59.8, 100.3, 123.9, 125.0, 127.1, 149.1, 149.2, 152.7, 165.	

Ethyl-4-(benzo[d][1,3]dioxol-5-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4n): The product was prepared by following the general procedure at page 1. Isolated yield (85%), Pinkish white, m.p. found 245-247^{0}C(new);**^{1}H NMR (400MHz, d_6-DMSO)**, δ 9.16 (s, 1H, N-H), 7.67 (s, 1H, N-H), 6.84 (d, 1H, *J*= 8.0 Hz, Ar-H), 6.74 (s, 1H, Ar-H), 6.68 (d, 1H, *J*= 8.0 Hz, Ar-H), (s, 1H, Ar-H), 5.97 (s, 2H, -CH_2), 5.07 (s, 1H, -CH), 3.99 (q, 2H, *J*= 7Hz, -CH_2), 2.24 (s, 3H, -CH_3), 1.10 (t, 3H, *J*= 7Hz, -CH_3);**^{13}C NMR (100MHz,d_6-DMSO),** δ 14.5, 18.2, 54.1, 59.7, 99.8, 101.4, 107.1, 108.5, 119.8, 139.3, 146.8, 147.7, 148.7, 152.5, 165.8 ppm.	

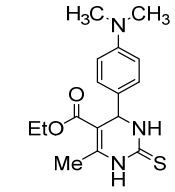
Ethyl-6-methyl-4-(naphthalen-1-yl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4o): The product was prepared by following the general procedure at page 1. Isolated Yield (82%), mp. found 248-250^{0}C; **^{1}H NMR (400MHz, d_6-DMSO),** δ 9.27(s, 1H, N-H), 8.31(d, 1H, *J*= 8Hz, Ar-H), 7.97-7.93(m, 1H, Ar-H), 7.88-7.84(m, 1H, Ar-H), 7.76(s, 1H, N-H), 7.63-7.50(m, 4H, Ar-H), 7.47(s, 1H, Ar-H), 5.46(s, 1H, -CH), 3.82(m, 2H, -CH_2), 2.37(s, 1H, -CH_3), 0.80(t, 3H, *J*= 7Hz, -CH_2-CH_3); **^{13}C NMR (100MHz, d_6-DMSO),** δ 14,3, 18.2, 50.3, 59.5, 99.6, 123.9,124.6, 126.1, 126.5, 126.7, 128.4, 128.9, 130.6, 133.9, 140.8, 149.2, 152.2, 157.8, 165.7.	

Ethyl-4-(furan-2-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate 4(p): The product was prepared by following the general procedure at page 1. Isolated yield (80%), m.p. found 202-204^{0}C, (lit. 203-204^{0}C)[16];**IR (KBr):** 3354, 1693, 1645 cm^{-1}; **^{1}H NMR(200MHz, d_6-DMSO);** δ 9.25 (s, 1H, NH), 7.68 (s, 1H, NH), 6.28 (s, 1H, furan ring), 6.15 (s, 1H, furan ring), 5.20 (s, 1H, Furan ring), 4.00 (q, *J*= 6.6Hz, 2H, CH_2), 2.48 (s, 1H, -CH), 2.20 (s, 3H, CH_3) 1.34 (t, *J*= 4.5Hz, 3H, -CH_3). HRMS calcd. for $C_{12}H_{14}O_4N_2$, 250.2548, found 250.2546.	
Ethyl-6-methyl-4-phenyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4q): The product was prepared by following the general procedure at page 1. Yield (80%), m.p. found 205-208 ^{0}C, (lit. 208-209^{0}C) [44];**IR (KBr**): 3325, 3177, 2980, 1670, 1196 cm^{-1}**; ^{1}H NMR (200MHz, d_6-DMSO):** δ, 8.71 (s, 1H, NH), 7.50 (s, 1H, ArH), 7.55 (m, 4H, ArH), 5.40 (s, 1H, NH), 4.24 (q, *J*= 4Hz, 2H, CH_2), 2.75 (s, 1H, CH), 2.50 (s, 3H, CH_3), 1.34 (t, *J*= 4Hz, 3H,CH_3).	
Ethyl-6-methyl-4-(4-nitrophenyl)-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4r): The product was prepared by following the general procedure at page 1. Isolated yield (85%), m.p. found 210-212 ^{0}C, (lit. 213-21) [16];**IR (KBr):** 3105, 1707, 1600, 1522, 1043 cm^{-1}; **^{1}H NMR (300 MHz, d_6-DMSO):** δ 9.36 (s, 1H, N-H), 8.21 (d, 2H, *J*= 6Hz, Ar-H), 7.90 (s, 1H, N-H), 7.49 (d, 2H, *J*= 2Hz, Ar-H), 5.27 (s, 1H, -CH), 3.98 (q, 2H, *J*= 7 Hz, -CH_2), 2.26 (s, 3H, -CH_3), 1.09 (t, 3H, *J*= 7 Hz, -CH_3); **^{13}C NMR (75 MHz, DMSO-d_6):**δ, 14.0, 17.8, 53.6, 59.3, 98.1, 123.8, 127.6, 146.6, 149.3, 151.7, 151.9, 154.9, 165.0.	
Ethyl-4-(3-hydroxyphenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4s): The product was prepared by following the general procedure at page 1. Isolated yield **(**82%), Solid Crystalline white; m.p. found 210-212^{0}C, (lit. 212-214^{0}C) [16];**IR (KBr):** 3529, 3310, 1694 cm^{-1}; **^{1}H NMR (200 MHz, d_6-DMSO):**δ, 8.30 (s, 1H, N-H), 7.85 (s, 1H, N-H), 7.63 (s, 1H, Ar-H), 6.84 (d, *J*= 8Hz, 1H, Ar-H), 6.51 (d, *J*= 3Hz, 1H, Ar-H), 5.00 (s, 1H, -OH), 4.02 (q, *J*= 4Hz, 2H, -CH_2), 2.50 (s, 1H, -CH), 2.22 (s, 3H, -CH_3), 1.15 (t, *J*= 5Hz, 3H, -CH_3).	
Ethyl-4-(3-hydroxy-4-methoxyphenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4t): The product was prepared by following the general procedure at page 1. Isolated yield **(**82%), m.p found 188-191 ^{0}C(new), **IR (KBr):** 3529, 3310, 1694, 1194 cm^{-1}; **^{1}H NMR (200MHz, d_6-DMSO);**δ, 8.70 (s, 1H, N-H), 8.21 (s, 1H, N-H), 7.63 (s, 1H, Ar-H), 6.84 (d, *J*= 8Hz, 1H, Ar-H), 6.51 (d, *J*= 3.6Hz, 1H, Ar-H), 5.00 (s, 1H, -OH), 4.02 (q, *J*= 4Hz, 2H, -CH_2), 3.77 (s, 3H, -CH_3), 2.50 (s, 1H, -CH), 2.22 (s, 3H, -CH_3), 1.15 (t, *J*= 5Hz, 3H, -CH_3); HRMS: calcd. for $C_{15}H_{18}O_4SN_2$, 322.3791; found 322.3794	

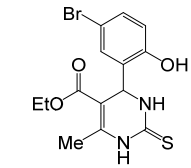
Ethyl-4-(4-chlorophenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4u): The product was prepared by following the general procedure at page 1. Isolated yield **(**85%), m.p. found 219-220 ^{0}C, (lit. 220-222^{0}C) [16]; **IR (KBr):** 3240, 3109, 1694, 1192 cm^{-1}; **^{1}H NMR (200MHz, d_6-DMSO)**δ, 8.70 (s, 1H, N-H), 7.21 (s, 2H, Ar-H), 7.15 (d, *J*= 8Hz, 1H, Ar-H), 6.88 (d, *J*= 8Hz, 1H, Ar-H), 5.08 (s, 1H, N-H), 3.98 (q, *J*= 7Hz, 2H, -CH_2), 2.50 (s, 1H, -CH), 2.23 (s, 3H, -CH_3) 1.09 (t, *J*= 7Hz, 3H, -CH_3



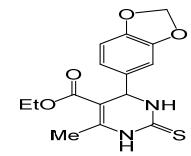
Ethyl-4-(4-(dimethylamino)-phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4v): The product was prepared by following the general procedure at page 1. Isolated yield (82%), Yellow solid, m.p. found 251-253^{0}C (new), **^{1}H NMR (400MHz, d_6-DMSO)**, δ 9.74 (s, 1H, N-H), 7.93 (s, 1H, N-H), 7.73 (d, 2H, *J*= 4Hz, Ar-H), 7.13 (d, 2H, *J*= 2Hz, Ar-H), 5.28 (s, 1H, -CH), 4.08 (q, 2H, *J*= 5Hz, -CH_2), 3.06(s, 6H, -CH_3), 2.33(s, 3H, -CH_3), 1.20 (s, 3H, *J*= 7Hz, -CH_3). **^{13}C NMR (100MHz, d_6-DMSO**), δ 14.3, 18.4, 40.4, 55.9, 60.4, 103.5, 111.1, 112.5, 127.8, 130.4, 132.1, 142.0, 150.5, 152.8, 165.5, 174.4 ppm. HRMS Calcd. for $C_{16}H_{21}N_3O_2S$: 319.4252, found: 319.4250.



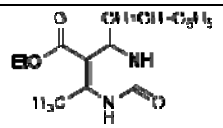
Ethyl-4-(5-bromo-2-hydroxyphenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4w): The product was prepared by following the general procedure at page 1. Isolated yield **(**85%) m.p. 290-292 ^{0}C(new); **IR(KBr)**: 3649, 1701, 1192 cm^{-1}; **^{1}H NMR (200MHz, D_6-DMSO):**δ, 9.10 (s, 1H, N-H), 8.80 (s, 1H, N-H), 7.41-7.29 (m, 1H, Ar-H), 7.19 (s, 1H, Ar-H), 6.85 (d, *J*= 8Hz, 1H, Ar-H), 5.41 (brs, 1H, -OH), 3.95 (q, *J*= 7Hz, 2H, -CH_2), 2.50 (s, 1H,-CH), 2.30 (s, 3H, -CH_3), 1.00 (t, *J*= 7Hz, 3H, -CH_3)

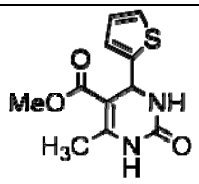


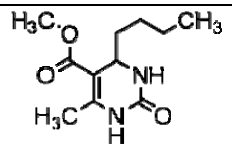
Ethyl-4-(benzo[d][1,3]dioxol-5-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4x): The product was prepared by following the general procedure at page 1. Isolated yield **(**85%), Solid; m.p. found 253-255^{0}C; **IR (KBr):** 3360, 1690, 1196 cm^{-1}; **^{1}H NMR (200 MHz, d_6-DMSO):**δ, 8.58 (s, 1H, N-H), 7.90 (s, 1H, N-H), 7.50 (s, 1H, Ar-H), 6.79-6.66 (m, 2H, Ar-H), 5.92 (s, 2H, -CH_2), 5.29 (s, 1H, -CH), 4.14-4.04 (m, 2H, -CH_2), 2.35 (s, 3H, -CH_3), 1.17 (t, 3H, *J*= 8Hz, -CH_3).

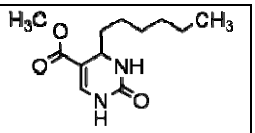


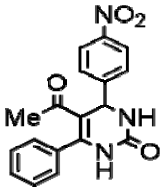
5-(Ethoxycarbonyl)-6-methyl-4-styryl-3, 4-dihydropyrimidin-2(1H)-one(4y): The product was prepared by following the general procedure at page 1. Isolated yield (81%) white solid, m.p.(175~178) lit. 176-178 [33];**^{1}H-NMR (400 MHz, DMSO-d6):** δ 1.18-1.22 (t, 3H, -OCH_2CH_3), 2.20 (s, 3H, -CH3), 4.064.11 (q, 2H, -OCH_2CH_3), 4.74 (d, 1H, -CH), 6.16-6.22 (dd,1H, -CH=C–H), 6.34-6.38 (d,1H, HC=CH) 7.23-7.41 (m, 5H, Ar-H), 7.55 (bs, 1H, -NH), 9.11 (bs, 1H, -NH)
^{13}C-NMR (100 MHz, DMSO-d6): δ 14.7, 18.2, 52.3, 59.7, 98.3, 126.8, 128.0, 128.6, 129.1, 130.4, 136.3, 149.0, 153.0, 165.7.



6-Methyl-2-oxo-4-thiophen-2-yl-1,2,3,4-tetrahydropyrimidine-5-carboxylic acid methyl ester (4z) Yield (86%) white solid; m.p. 157–159 □; **^{1}H NMR (400 MHz, DMSO d6):** δ (ppm), 9.33(s, 1H, NH), 7.94 (s, 1H, NH), 7.36 (d, 1H, J = 8.0 Hz, thiophene-SCH), 6.92 (m, 2H, thiophene-CH), 5.41 (s, 1H, CH), 3.64 (s, 3H, OCH_3), 2.25 (s, 3H, CH_3); **^{13}C NMR (100 MHz, DMSO d6)** δ (ppm), 165.31, 152.5, 150.3, 141.4, 127.0, 126.0, 124.6, 110.51, 58.5, 52.0, 19.33; **IR (KBr)** cm^{-1}: 3311 (NH), 3120 (NH), 1705 (CO), 1653 (CO); MS m/z 253 (M+1); Anal. Calc. for C11H12N2O3S: C, 52.37; H, 4.79; N, 11.10; found: C, 52.35; H, 4.80; N, 11.00.	

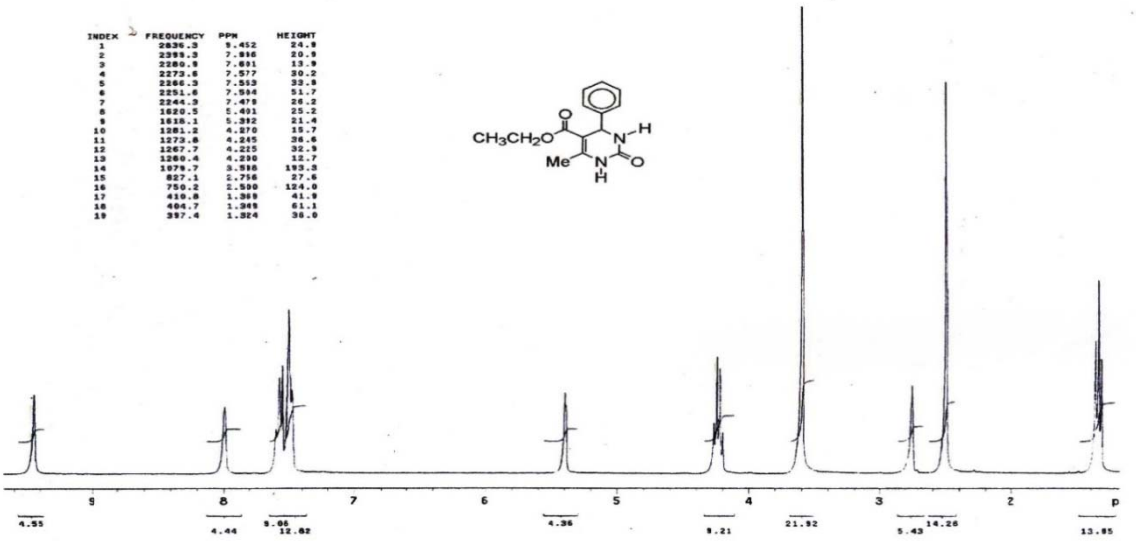
Methyl 4-butyl-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4ab). White solid, yield (80%); m.p. 171-173□,lit.172-175□ [43]; **^{1}H NMR(600 MHz, CDCl3)**: δ (ppm) 8.45 (s, 1H), 6.01 (s, 1H), 4.28 (dt, J = 7.8Hz, 3.6Hz, 1H), 3.71 (s, 3H), 2.31 (s, 3H), 1.58-1.48 (m, 2H), 1.38-1.24 (m, 4H), 0.88 (t, J = 6.6 Hz, 3H); **^{13}C NMR(150 MHz, CDCl3)**: δ (ppm) 166.4, 154.7, 147.0, 101.28, 51.5, 51.1, 36.7, 26.4, 22.4, 18.5, 13.9.	

Methyl 4-hexyl-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4ac): White solid, yield (82%); m.p. 148-150 □(lit,146-149) [43];**^{1}H NMR(600 MHz, CDCl3):** δ (ppm) 8.15 (s, 1H), 5.8 (s, 1H), 4.28 (dt, J = 7.8Hz, 3.6Hz, 1H), 3.71 (s, 3H), 2.28 (s, 3H), 1.59-1.47 (m, 2H), 1.39-1.33 (m, 1H), 1.29-1.25 (m, 7H), 0.83 (t, J = 6.6 Hz, 3H); **^{13}C NMR(150 MHz, CDCl3)**: δ (ppm) 166.33, 154.5, 146.90, 101.4, 51.6, 51.1, 36.9, 31.7, 29.0, 24.3, 22.6, 18.6, 14.1	

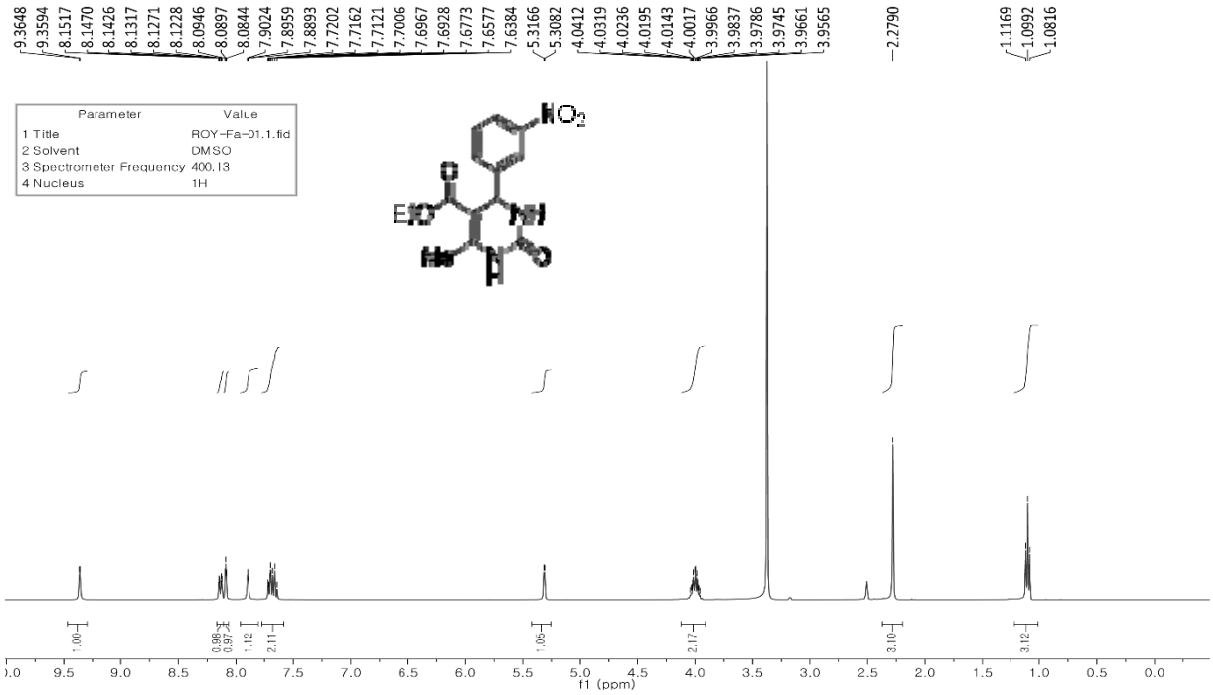
5-Acetyl-4-(4-nitrophenyl)-6-phenyl-3,4-dihydropyrimidin-2(1H)one(4ad) Yield:(84%);Yellow solid,m.p.253–256°C; **^{1}H NMR (500 MHz, DMSO**): δ 9.12 (s, 1H), 7.61 (s, 1H), 7.03 (d, J = 8.6 Hz, 2H), 6.66 (d, J = 8.5 Hz, 2H), 5.03 (s, 1H), 3.97 (dd, J=7.0,3.4Hz,2H),2.85(s,6H),2.23(s,3H),1.11(t,J=7.1Hz,3H); **^{13}C NMR (100 MHz, DMSO):** δ 165.94, 152.77, 150.20, 148.05, 133.12,127.35,112.69,100.32,59.57,53.75,40.68,18.20,14.61.	

Copies of NMR (^{1}H &^{13}C) spectra of products:

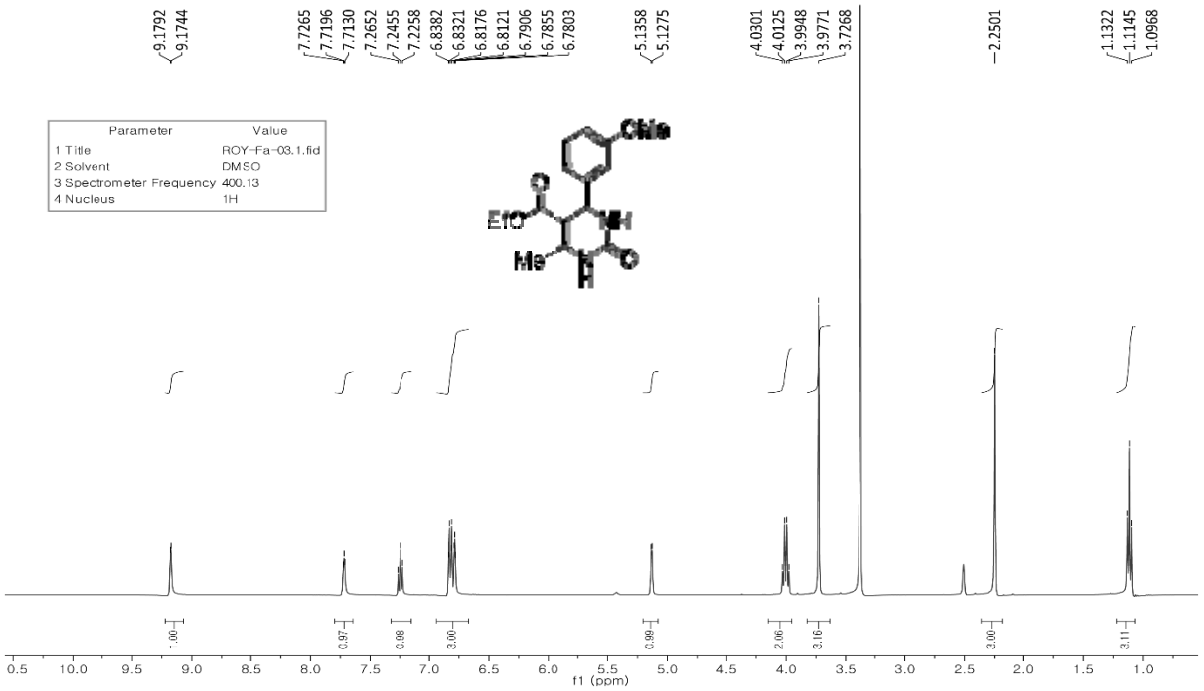
^{1}H NMR: Ethyl 6-methyl-4-phenyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4a)



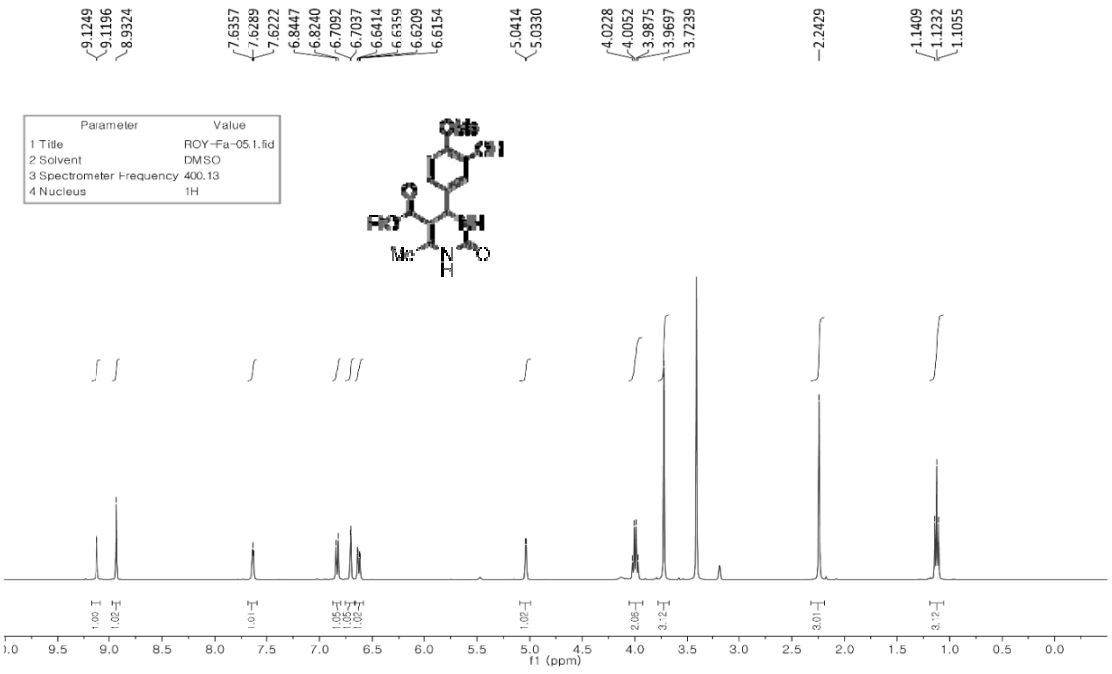
^{1}H NMR: Ethyl 6-methyl-2-oxo-4-(m-nitrayl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4b)



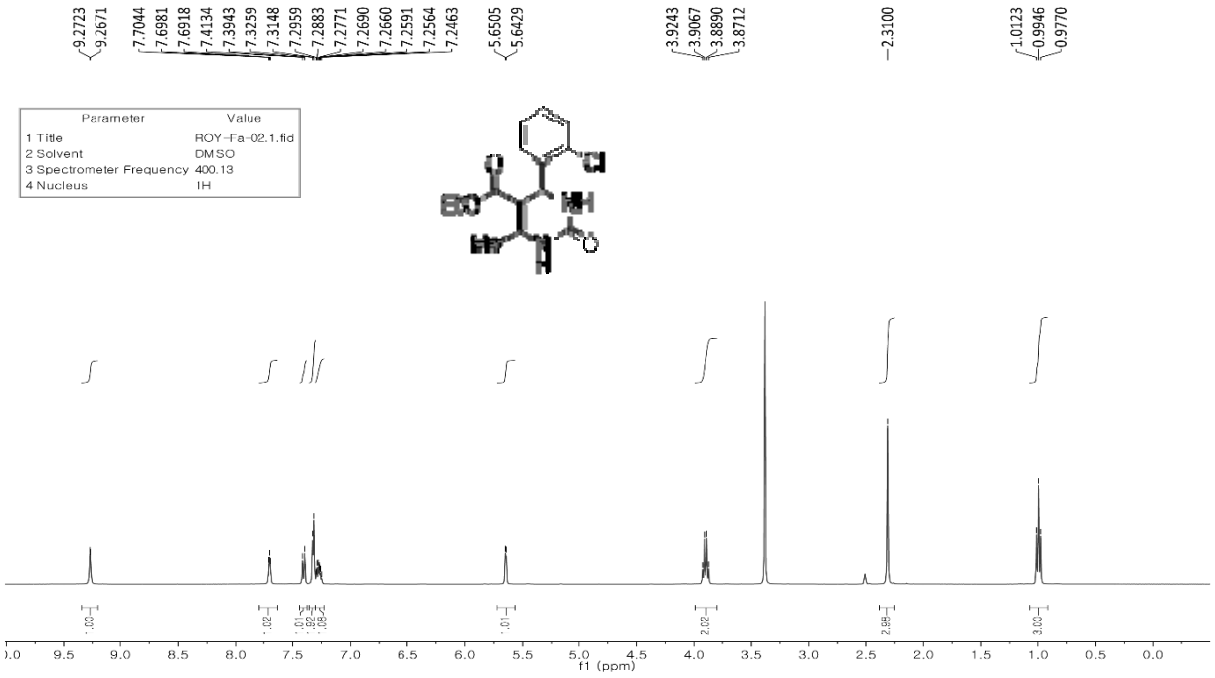
^{1}H NMR: Ethyl4-(3-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4c)



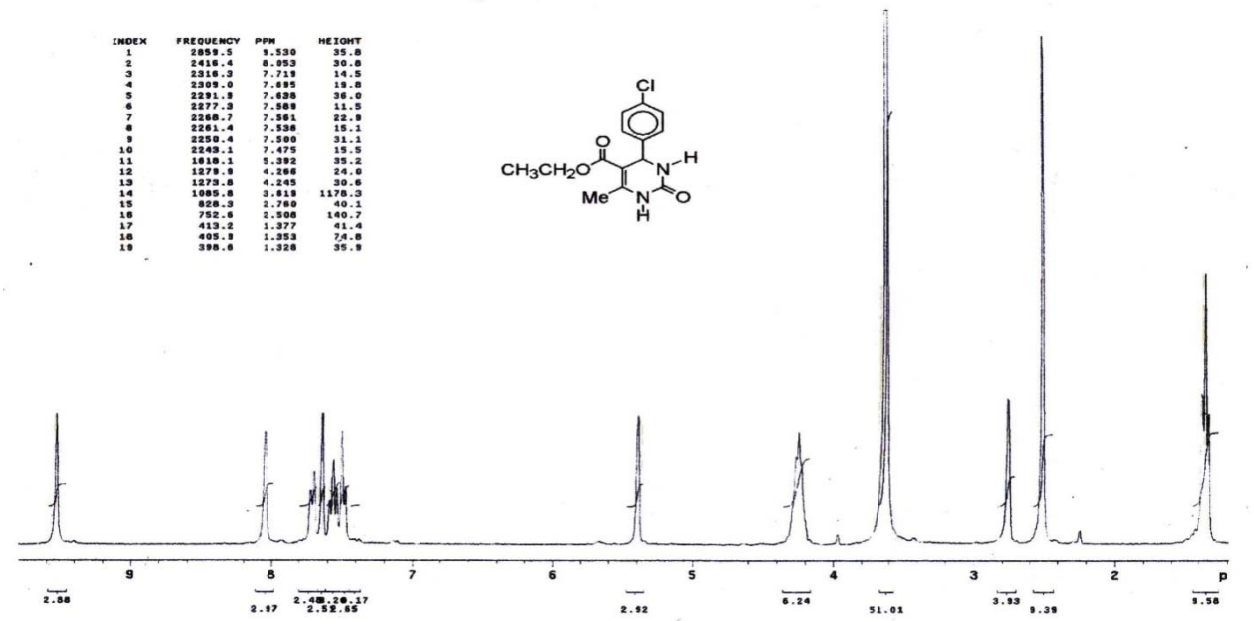
^{1}HNMR: Ethyl-4-(3-hydroxy-4-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4d)



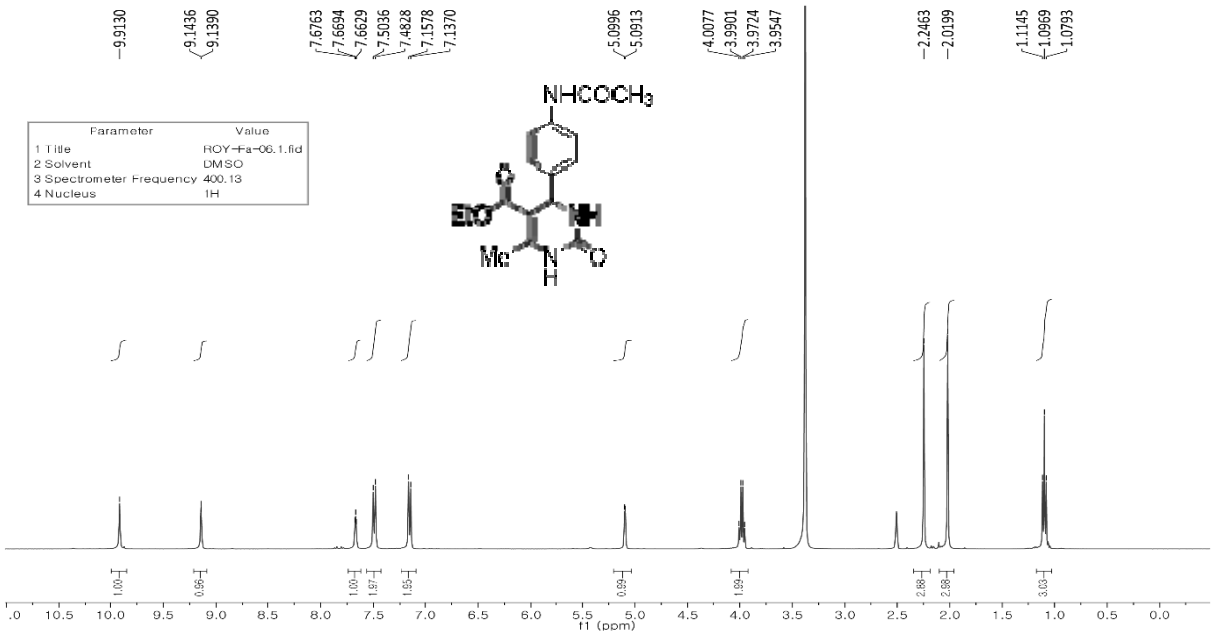
^{1}H NMR: Ethyl-4-(2-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4e)



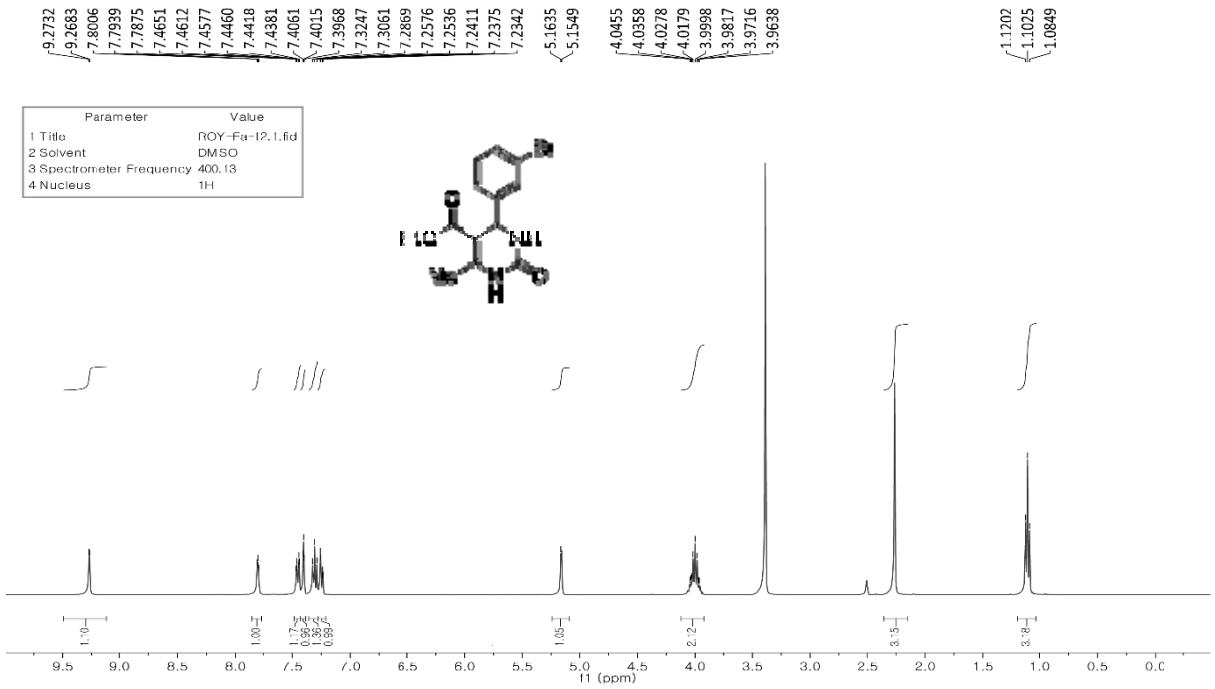
^{1}H NMR: Ethyl-4-(4-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4f)



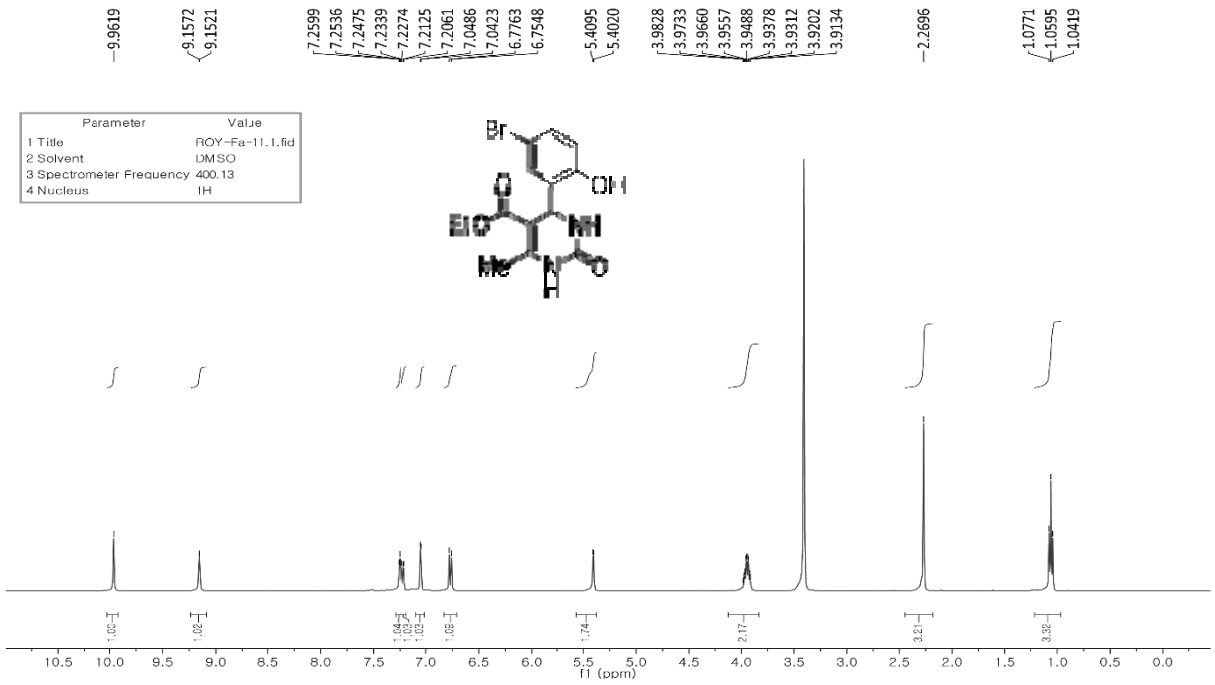
^{1}H NMR: Ethyl-4-(4-Acetamidophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4g)



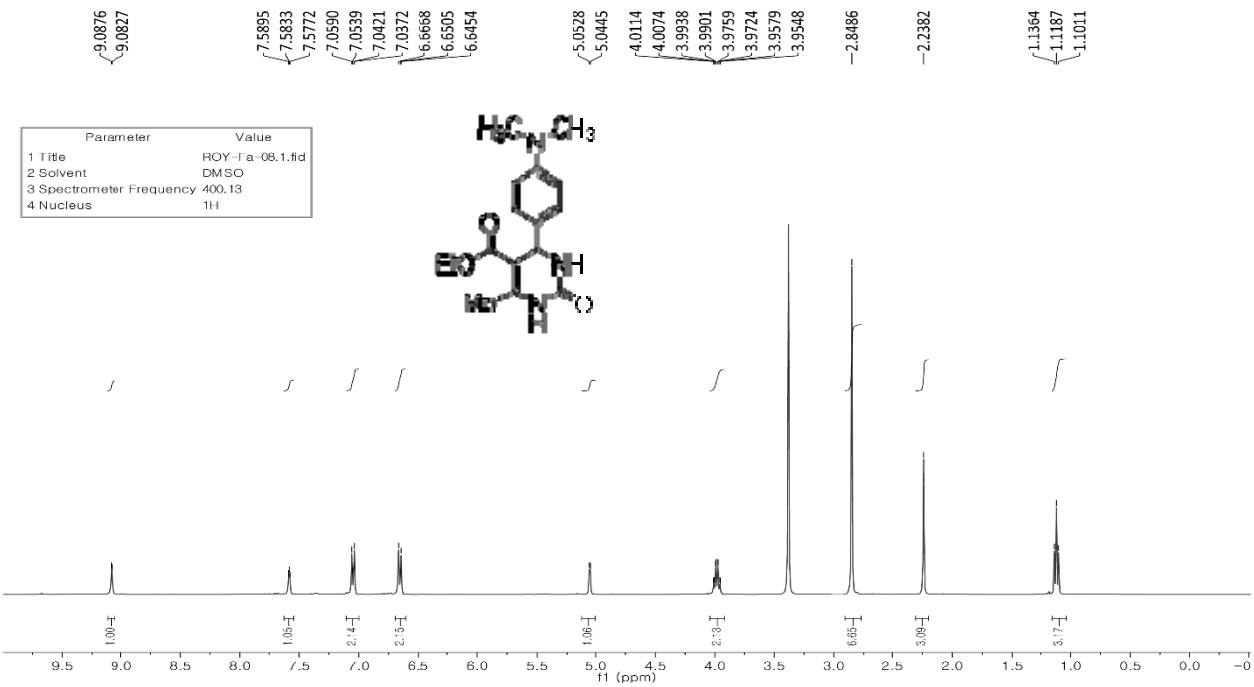
^{1}H NMR: Ethyl-4-(3-bromophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4h)



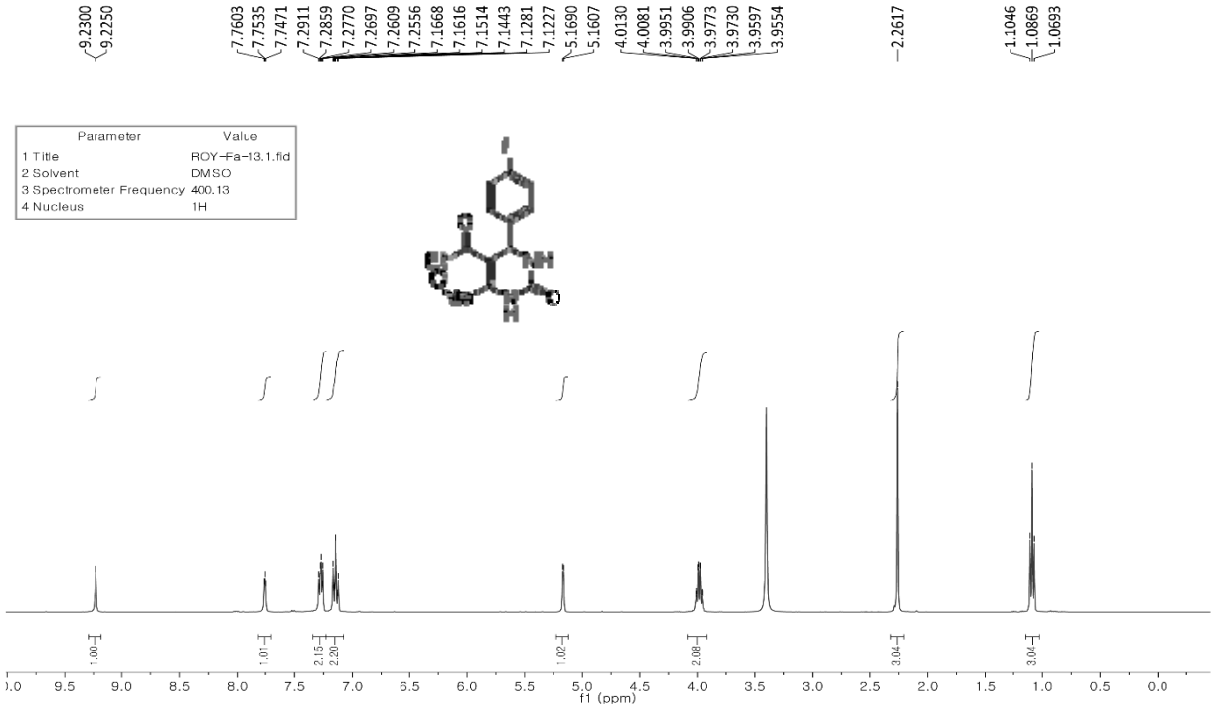
[1]HNMR: Ethyl 4-(5-bromo-2-hydroxyphenyl)-6-methyl-2-oxo-1,2,3,4 tetrahydropyrimidine-5-carboxylate(4i).



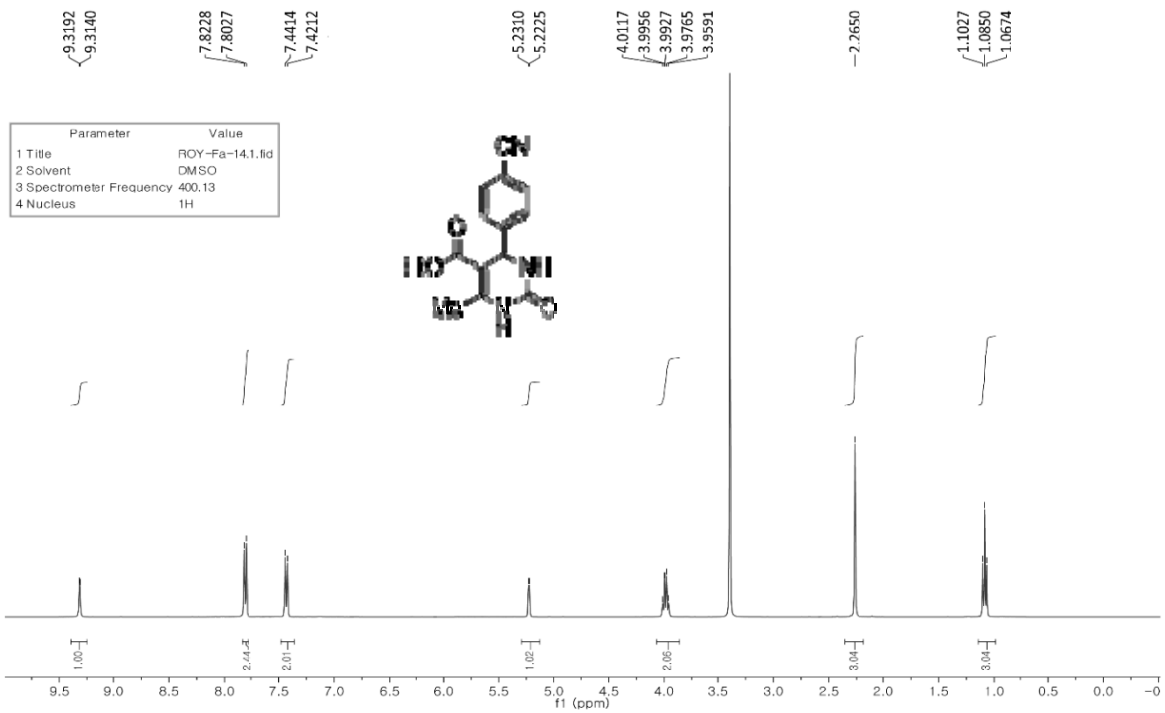
[1]H NMR: Ethyl 4-(4-(dimethylamino)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4j).



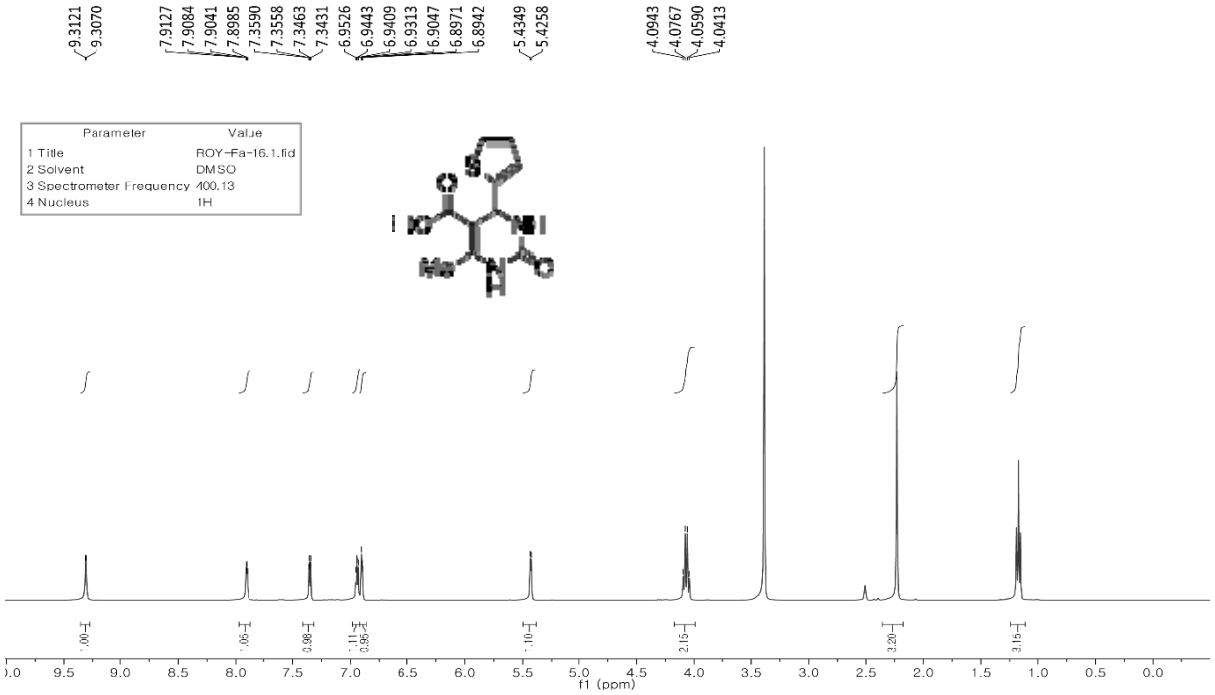
^{1}H NMR: Ethyl 4-(4-fluorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4k).



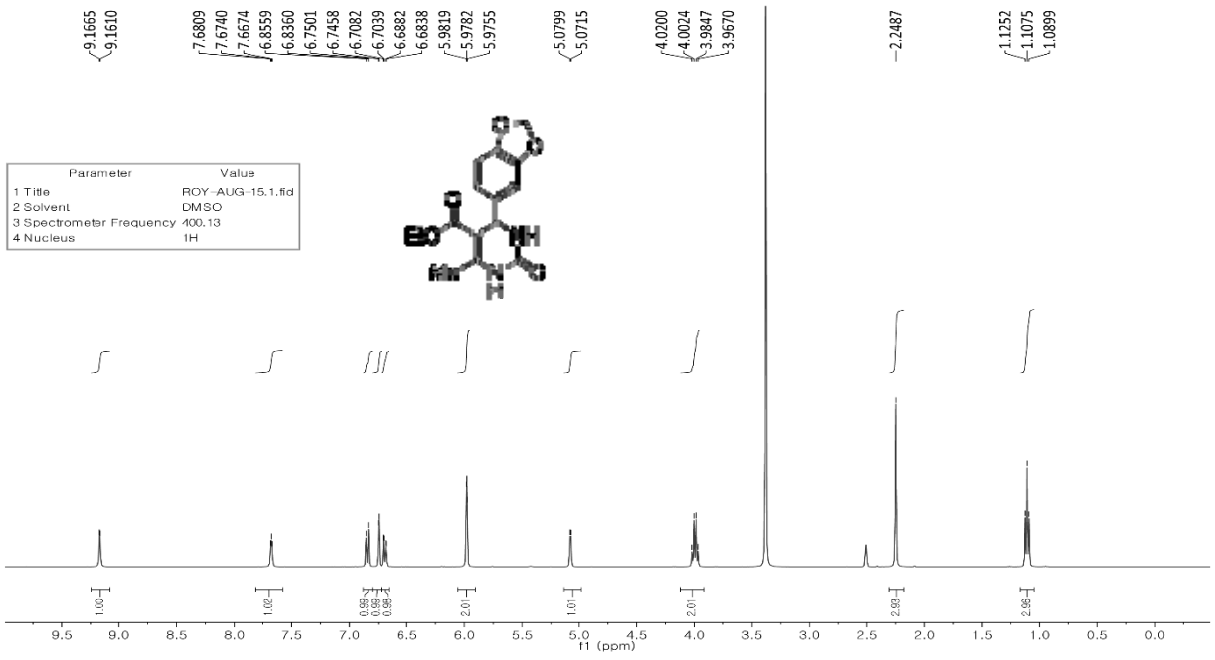
^{1}H NMR: Ethyl 4-(4-cyanophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4l)



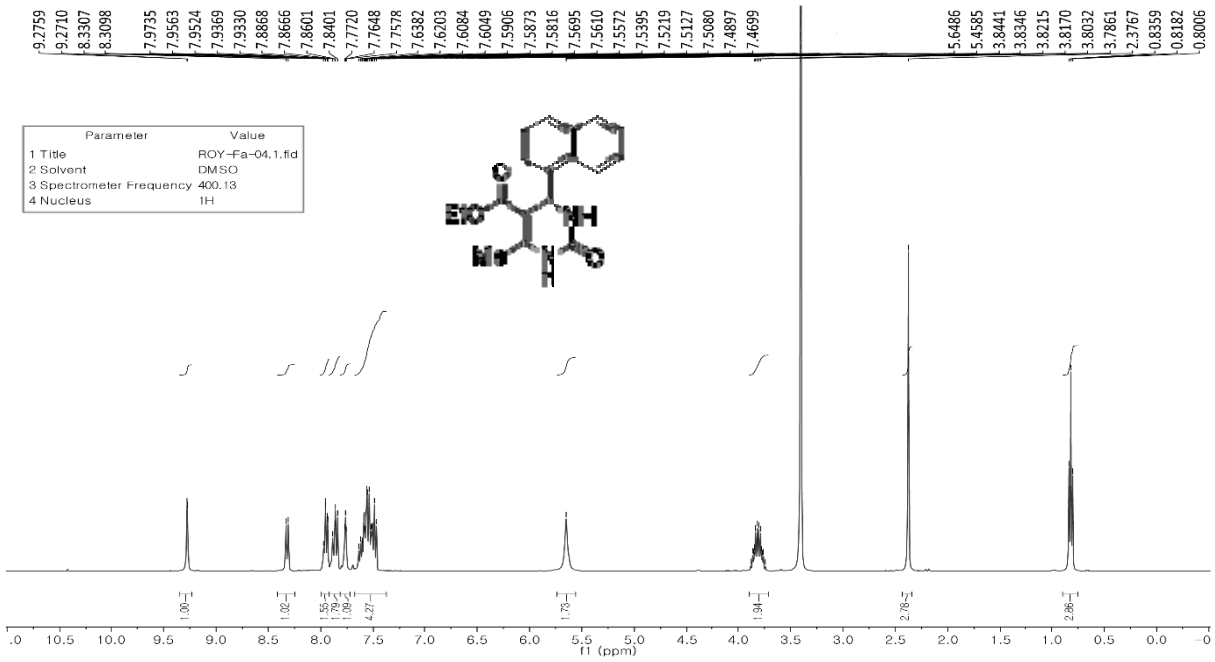
^{1}H NMR: Ethyl 6-methyl-2-oxo-4-(thiophen-2-yl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4m)



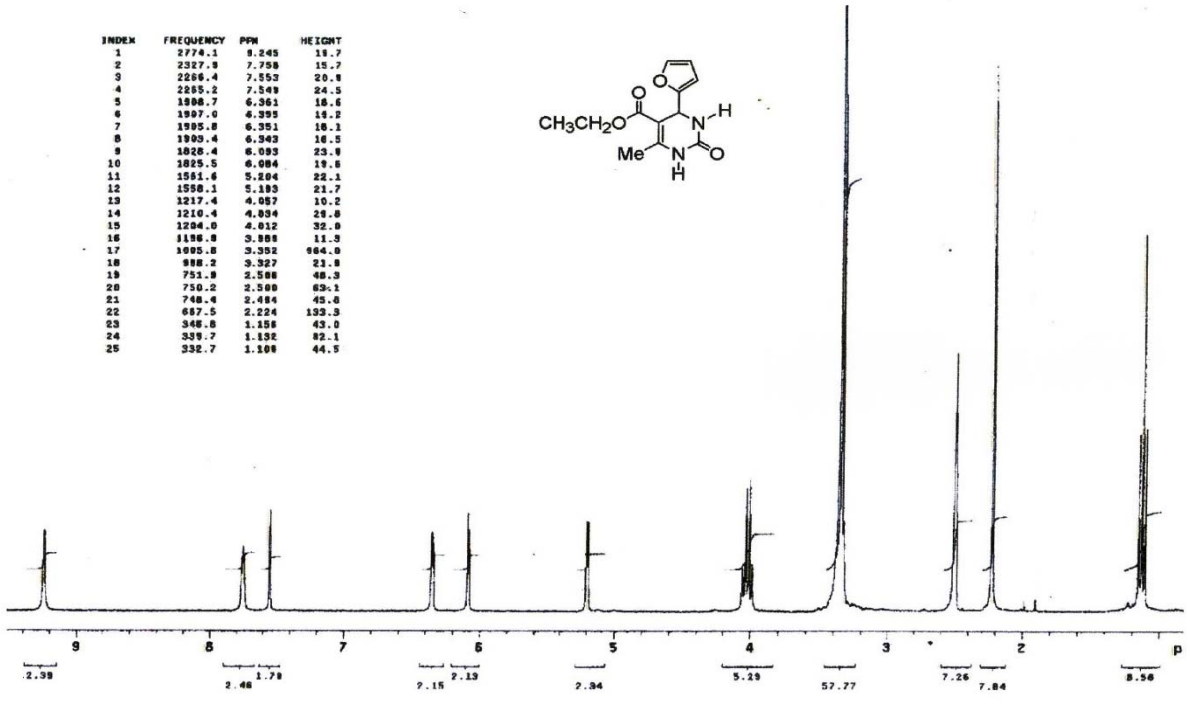
^{1}H NMR: Ethyl 4-(benzo[d][1,3]dioxol-5-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4n).



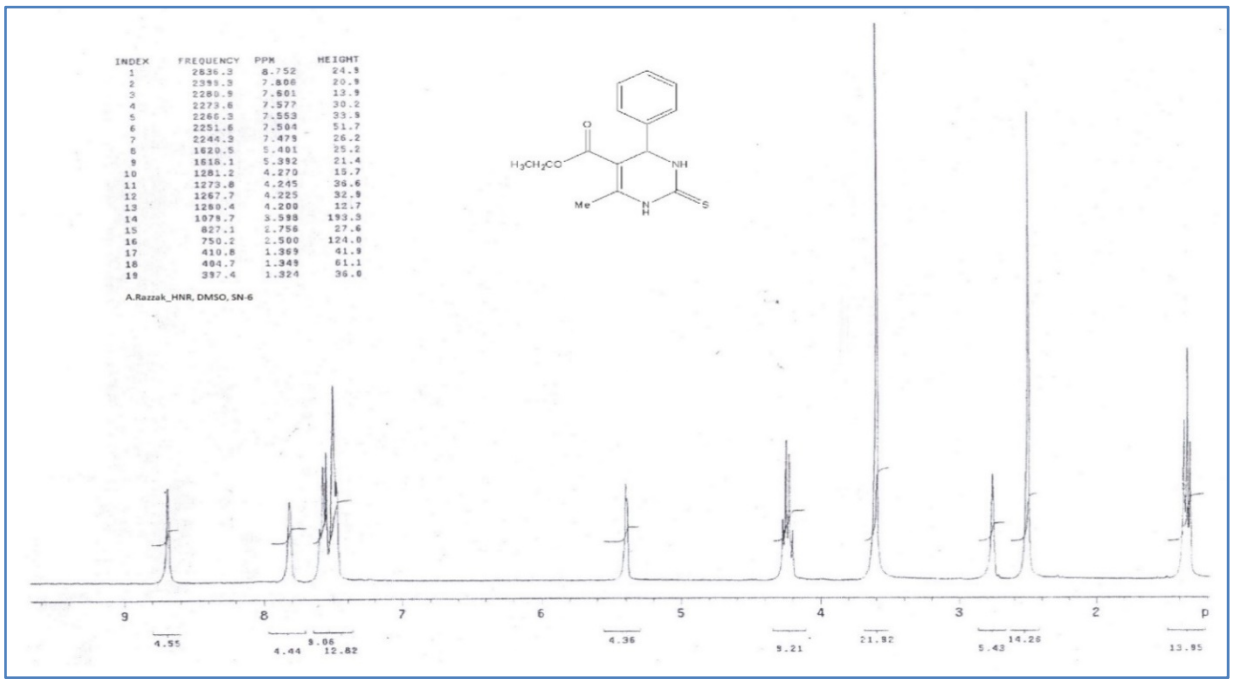
^{1}H NMR: Ethyl 6-methyl-4-(naphthalen-1-yl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4o)



^{1}H NMR: Ethyl 4-(furan-2-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate 4(p)

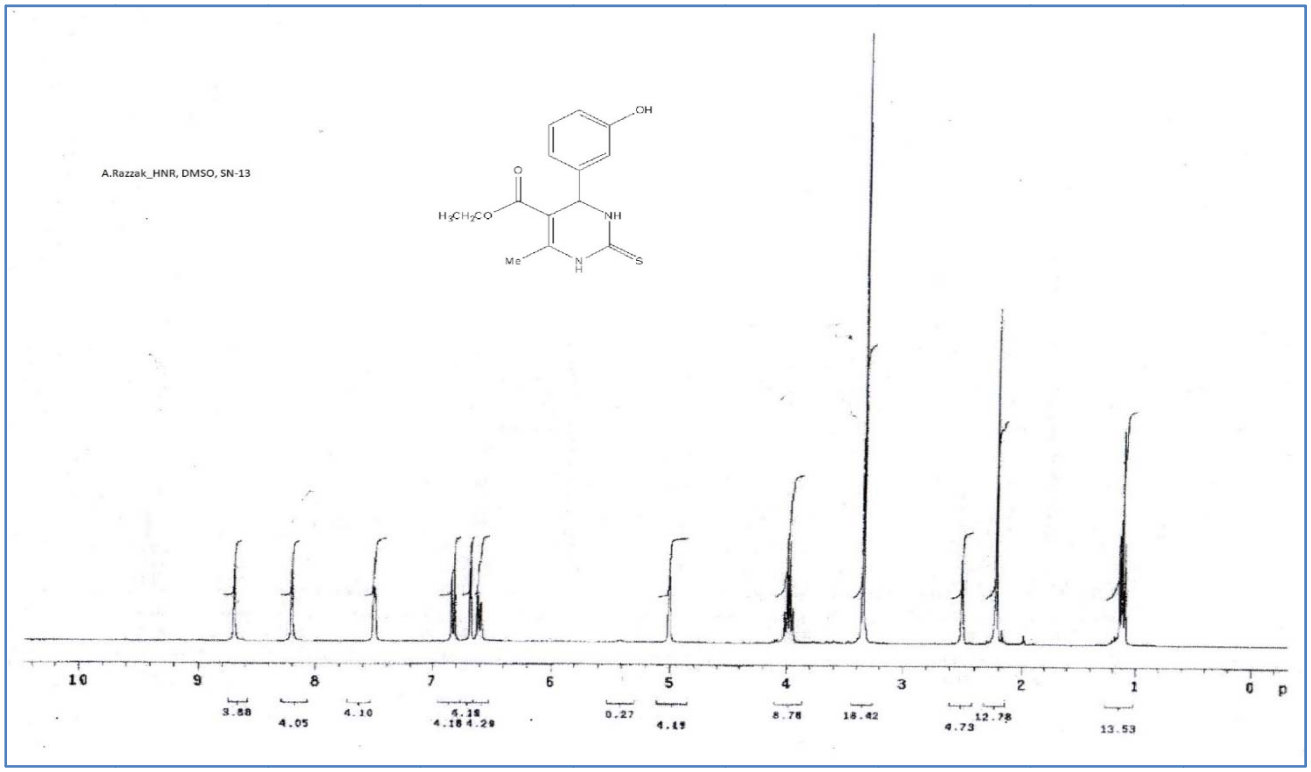


^{1}H NMR: Ethyl 6-methyl-4-phenyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4q).

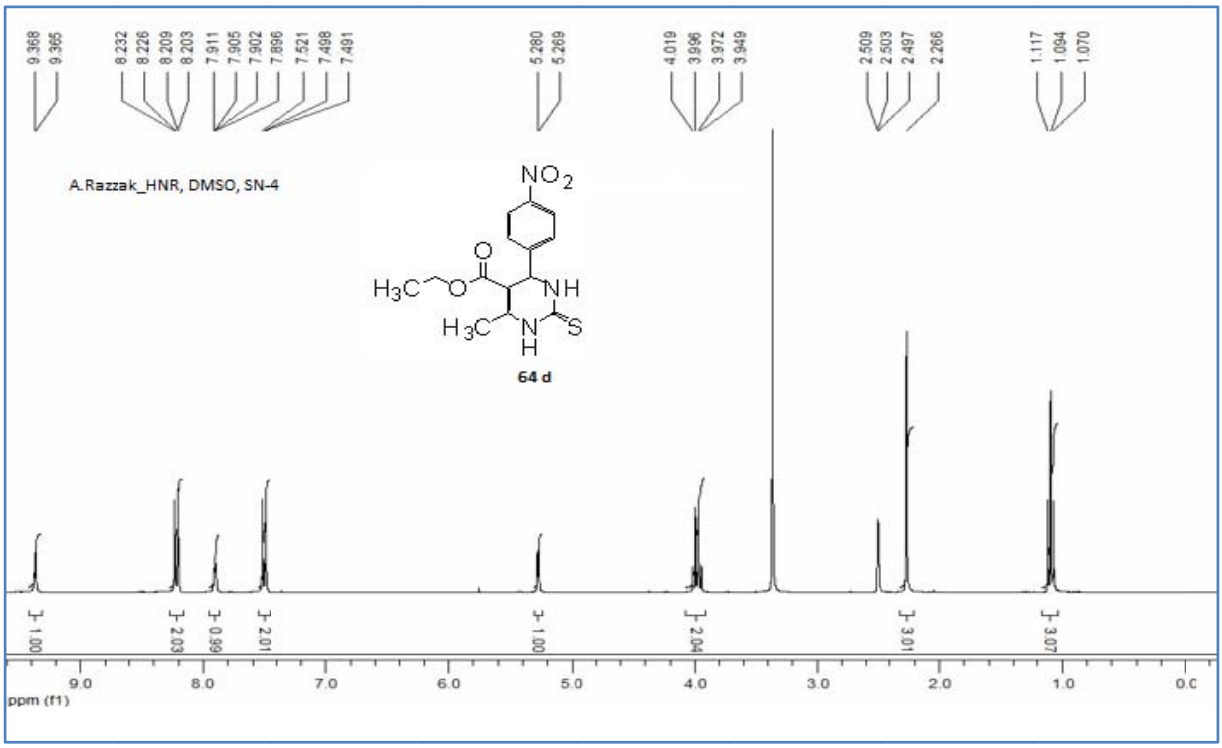


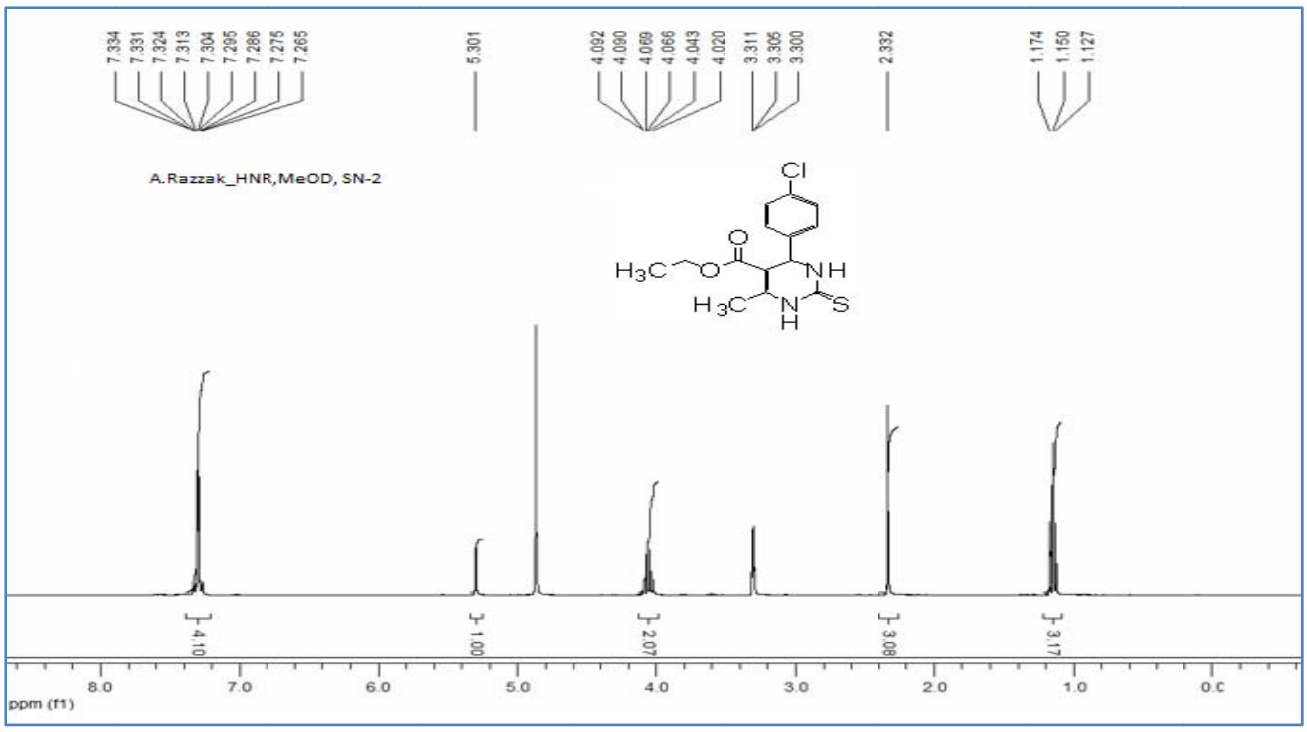
^{1}H NMR: Ethyl 6-methyl-4-(4-nitrophenyl)-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4r)

^{1}H NMR: Ethyl 4-(3-hydroxyphenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4s)

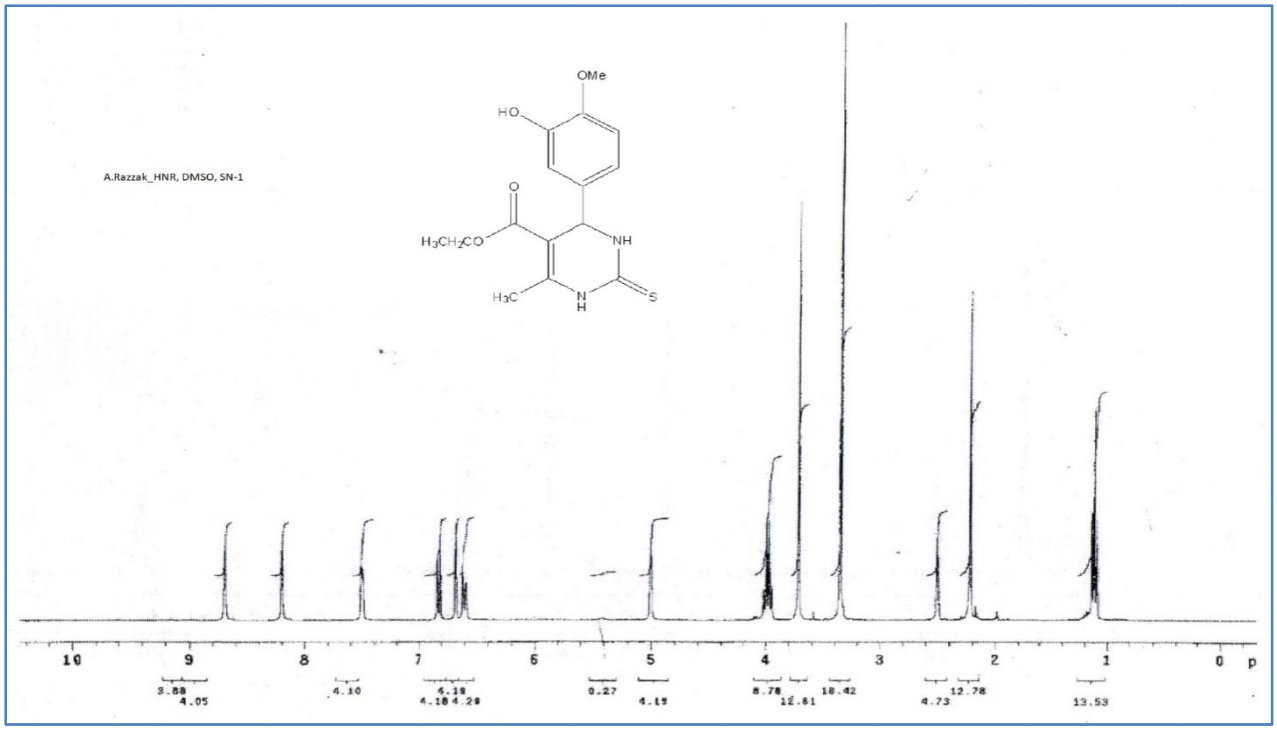


^{1}H NMR: Ethyl 4-(4-chlorophenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4u)

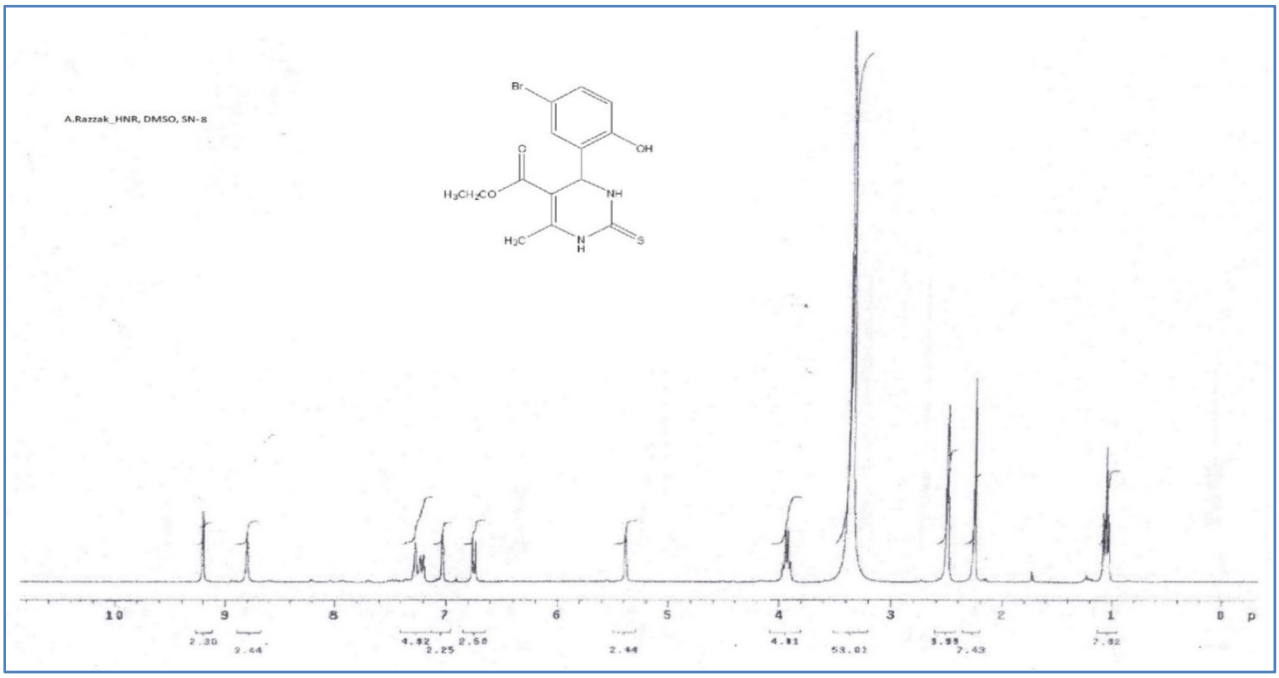




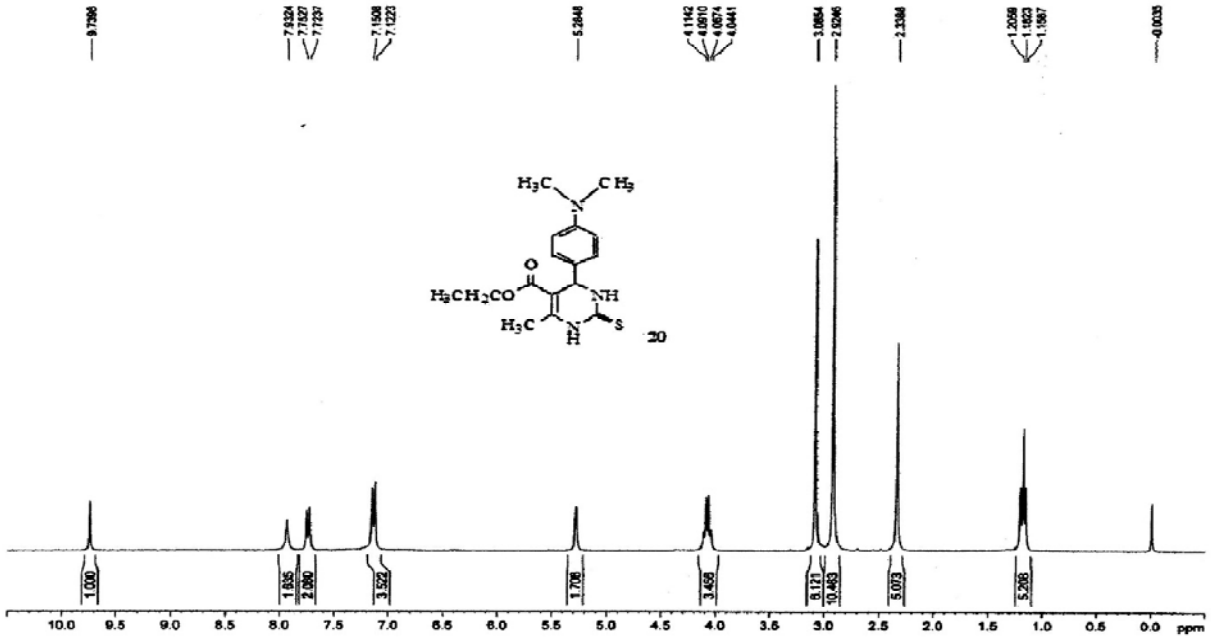
^{1}H NMR: Ethyl-4-(3-hydroxy-4-methoxyphenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4t)



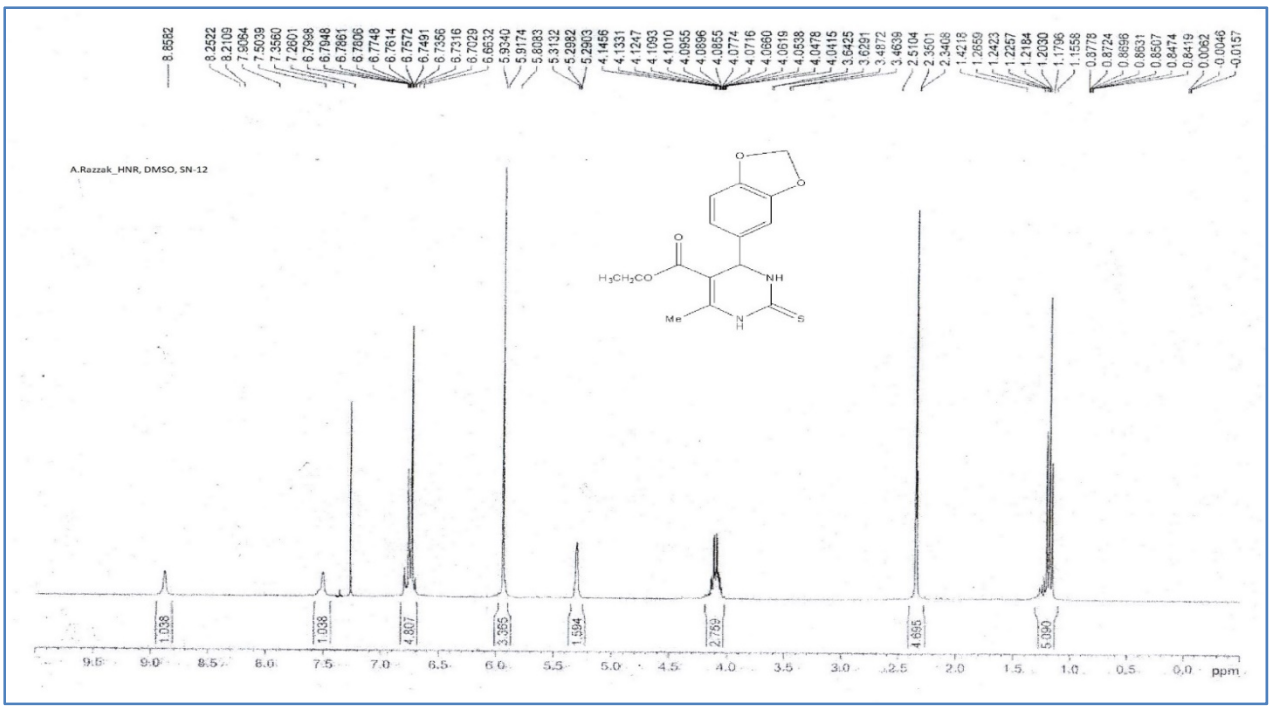
^{1}H NMR: Ethyl-4-(5-bromo-2-hydroxyphenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4w)



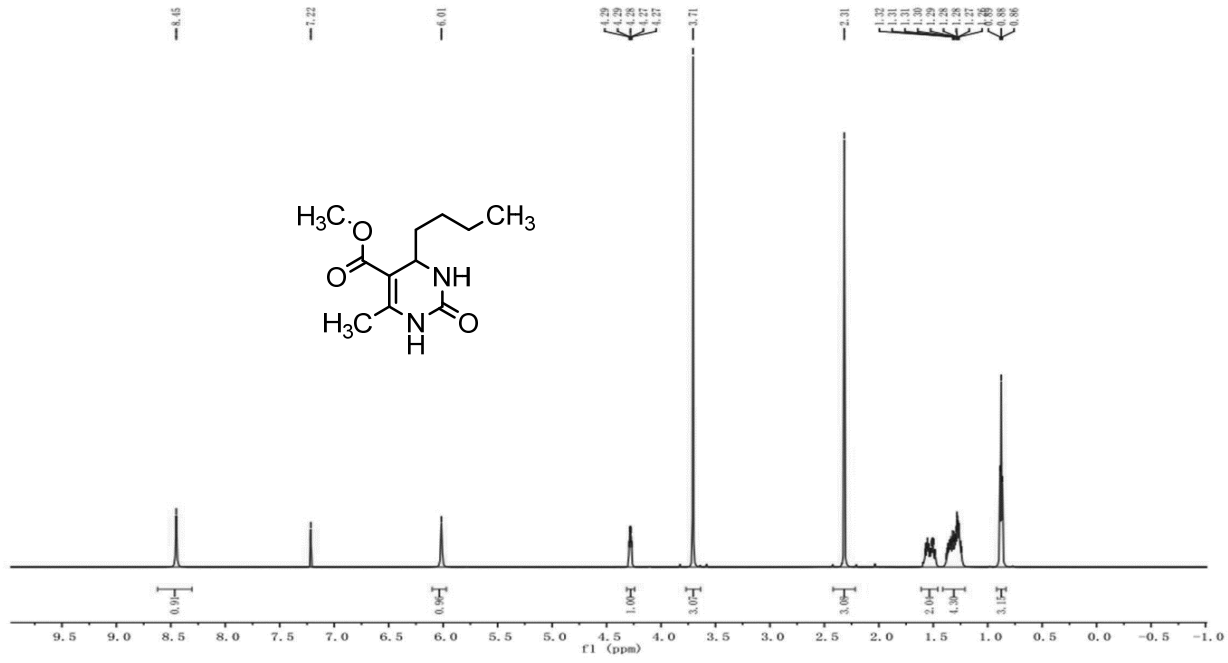
^{1}H NMR: Ethyl 4-(4-(dimethylamino)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4v)



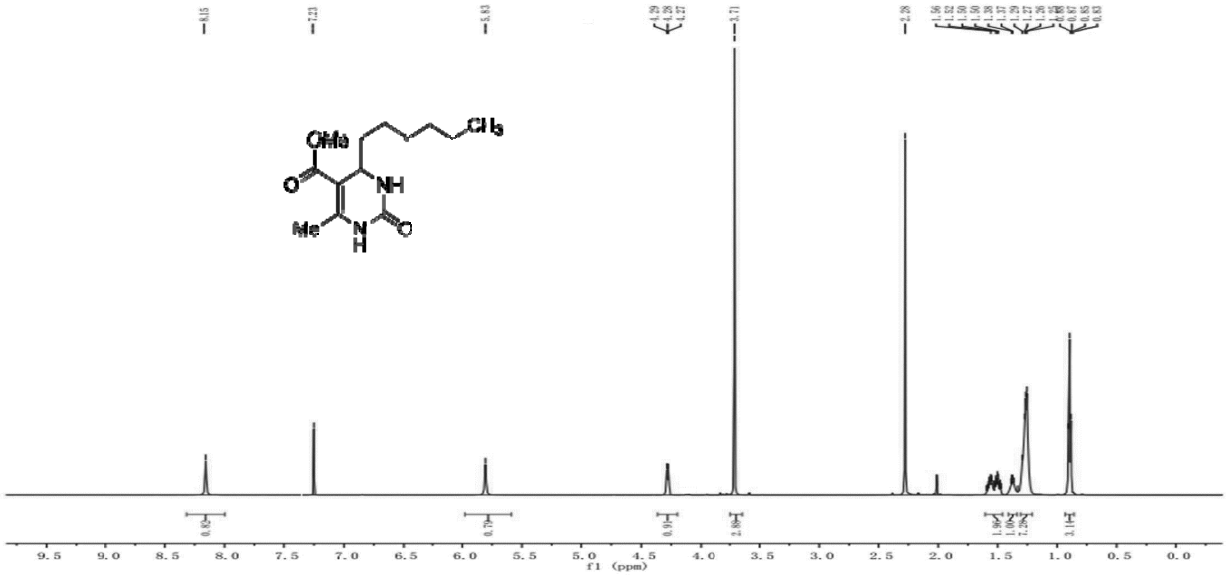
^{1}H NMR: Ethyl 4-(benzo[d][1,3]dioxol-5-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4x).



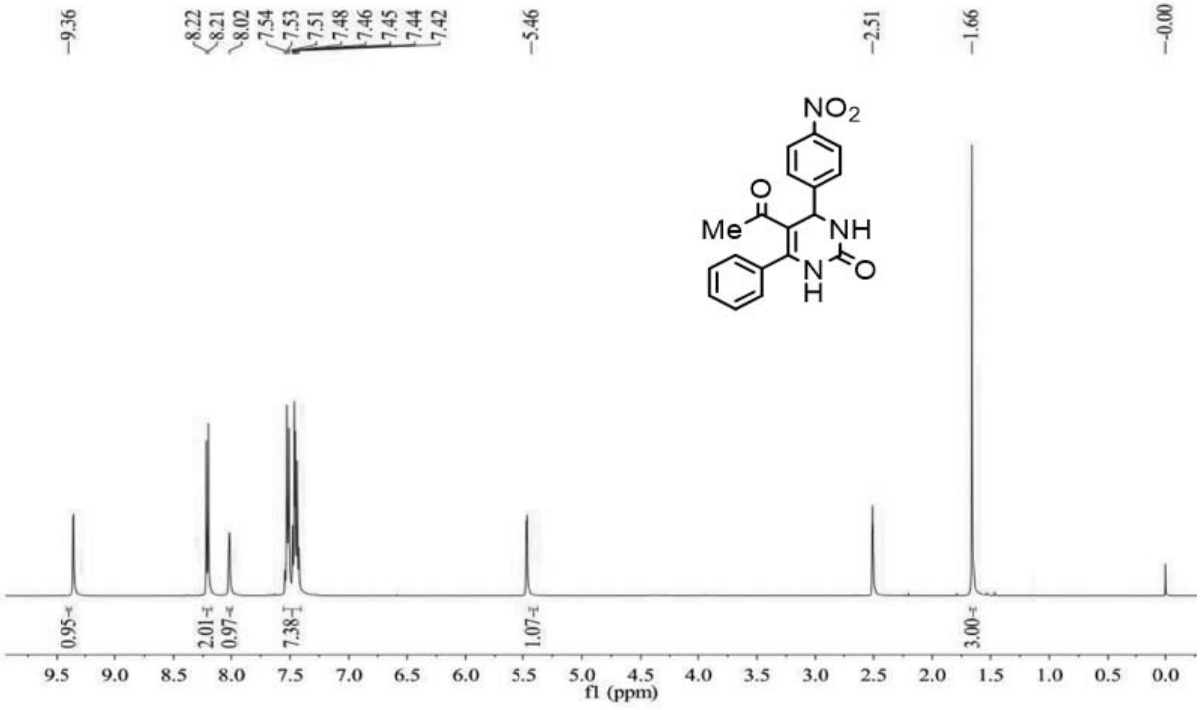
[1]H NMR:Methyl 4-butyl-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4ab)



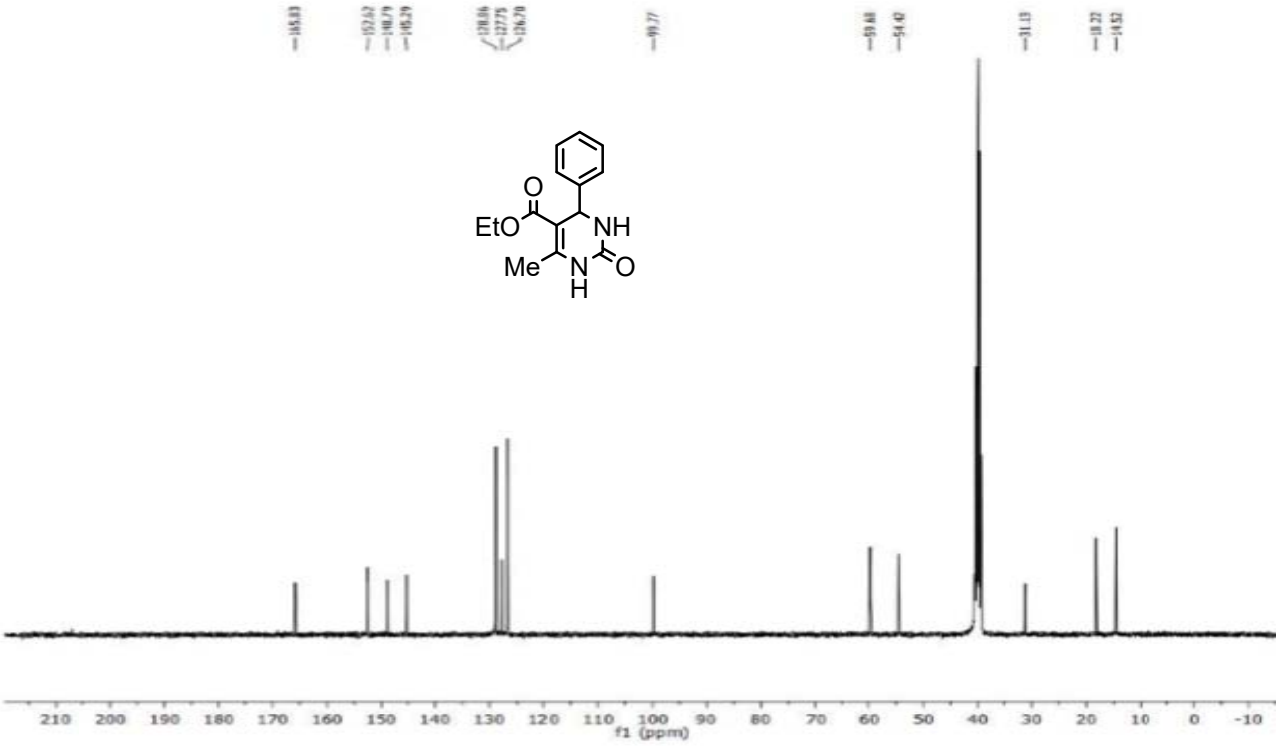
[1]H NMR: Methyl 4-hexyl-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4ac)



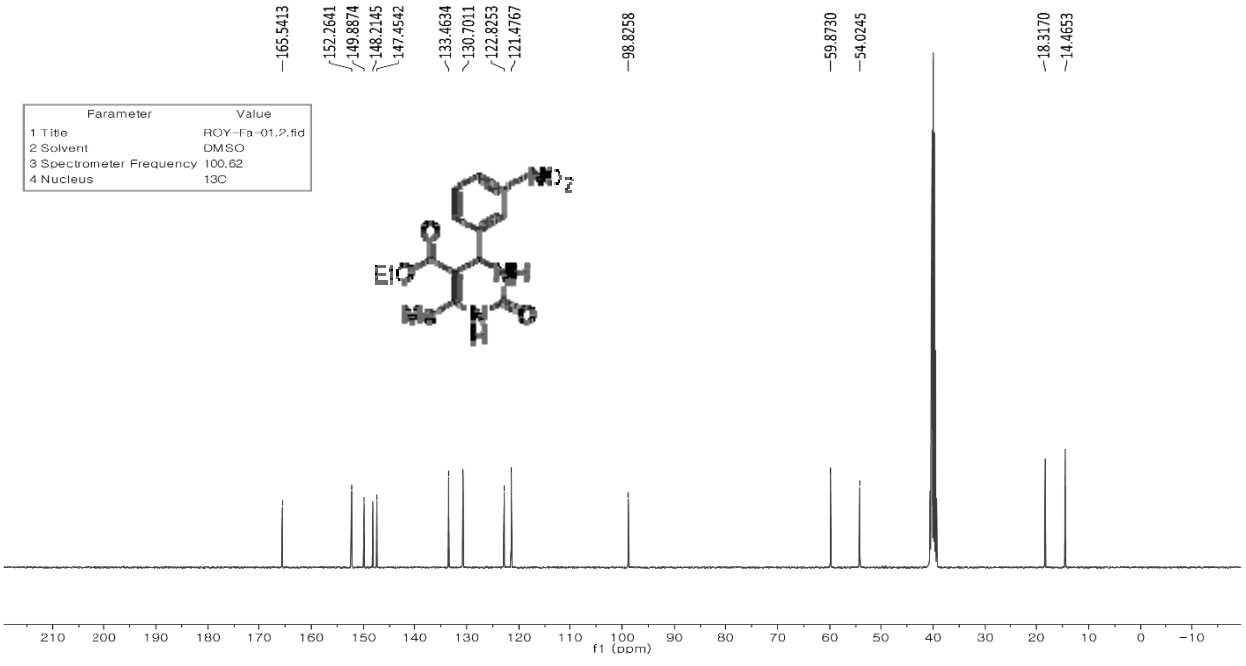
[1]H NMR: Ethyl 6-methyl-4-(4-nitrophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine5-carboxylate(4ad)



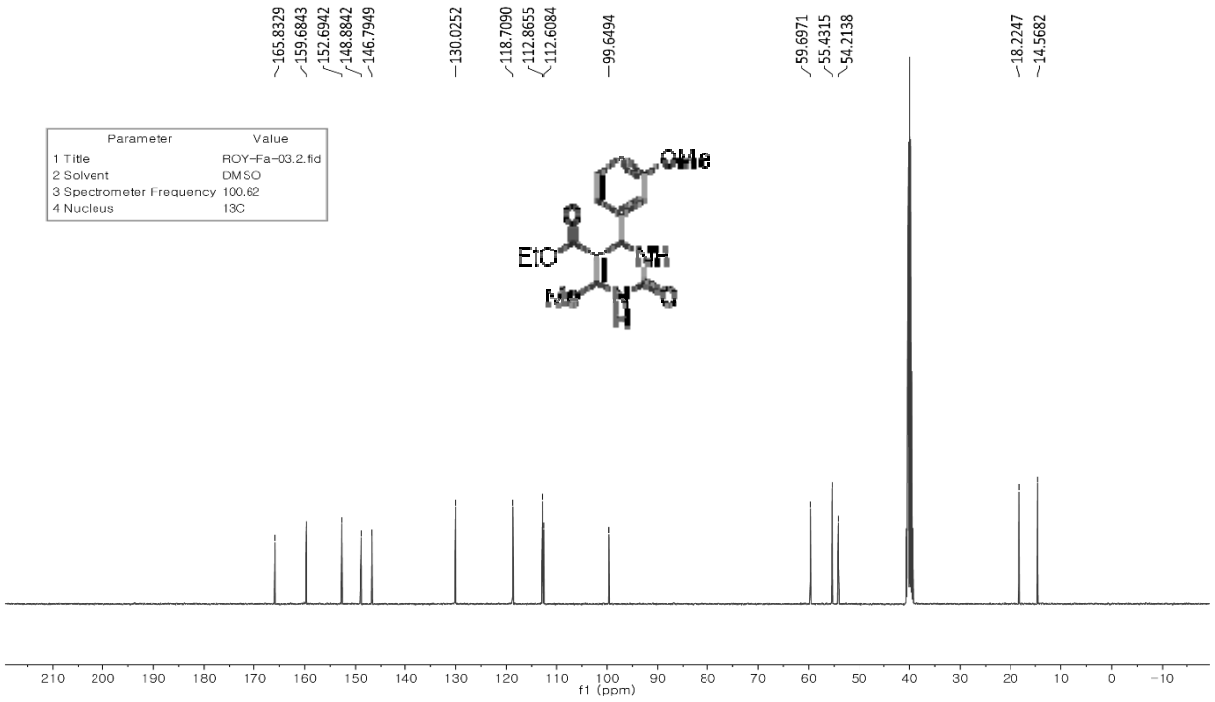
^{13}C NMR: Ethyl 6-methyl-4-phenyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4a)



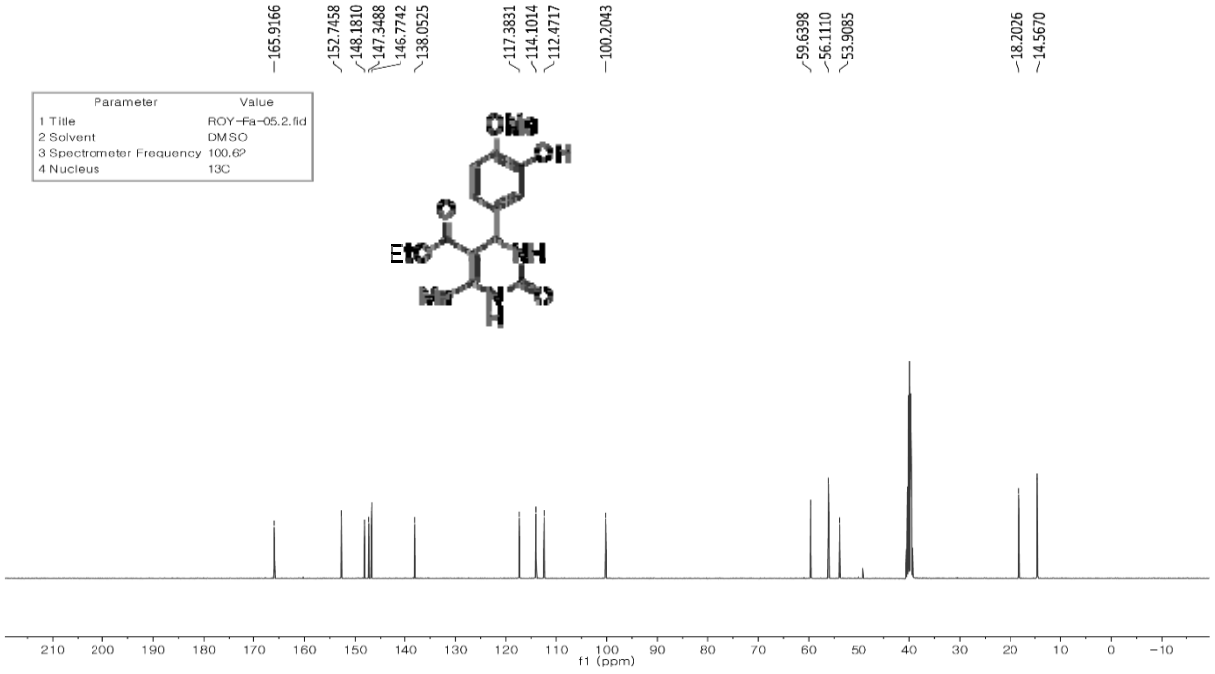
^{13}C NMR: Ethyl 6-methyl-2-oxo-4-(m-nitrayl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4b)



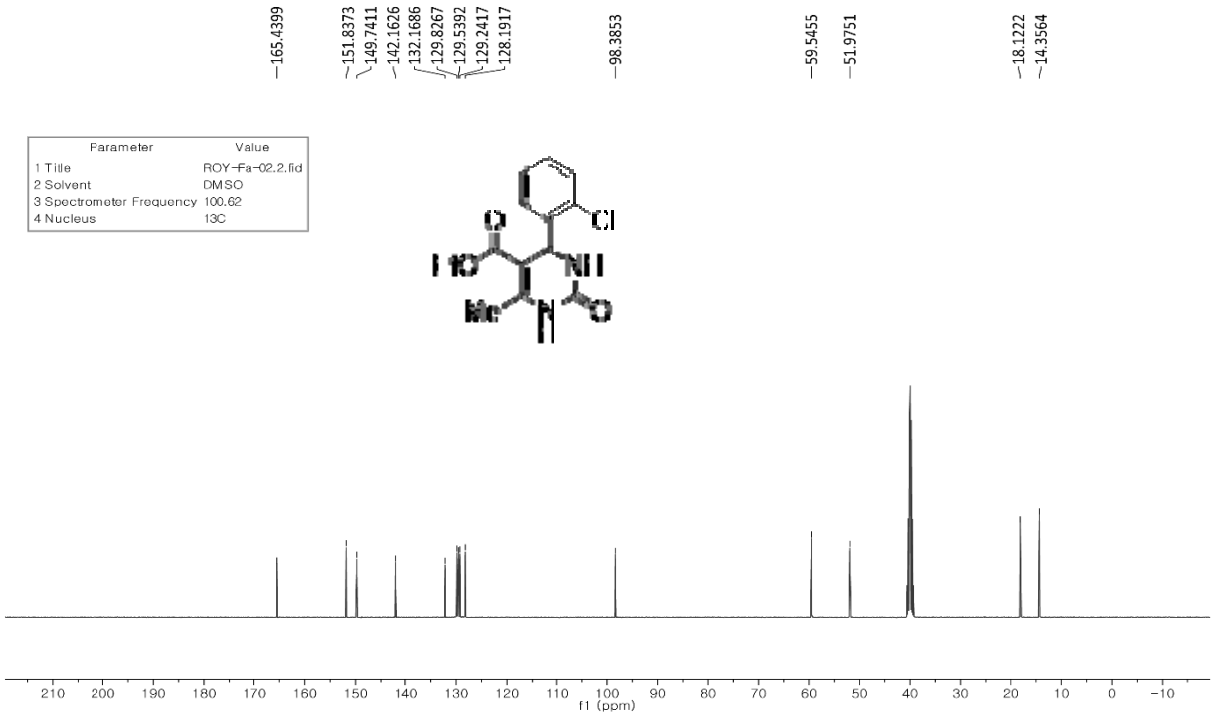
^{13}C NMR: Ethyl-4-(3-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4c)



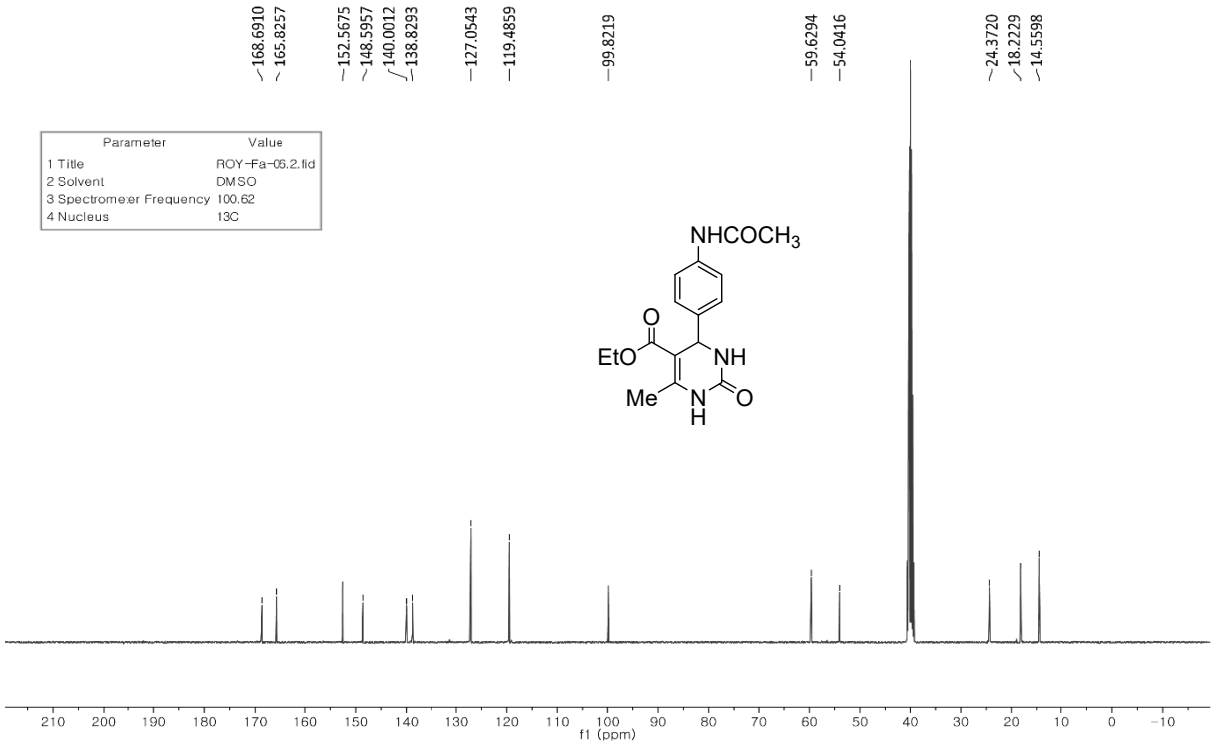
^{13}CNMR: Ethyl-4-(3-hydroxy-4-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4d)



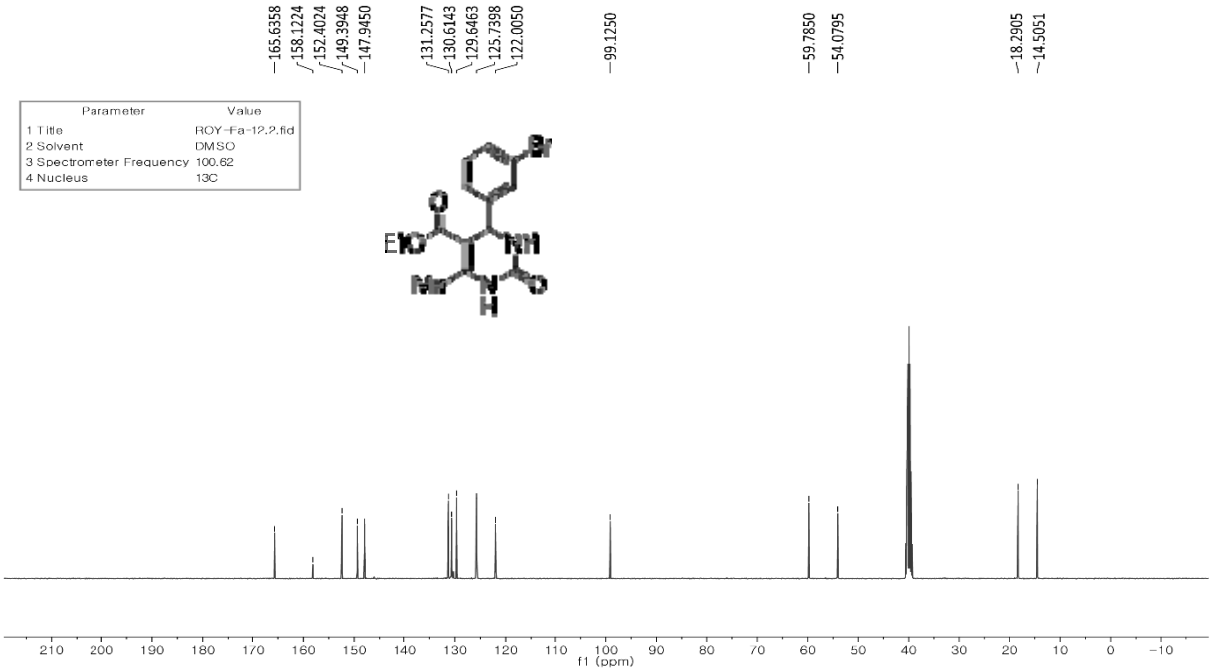
^{13}C NMR: Ethyl-4-(2-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4e)



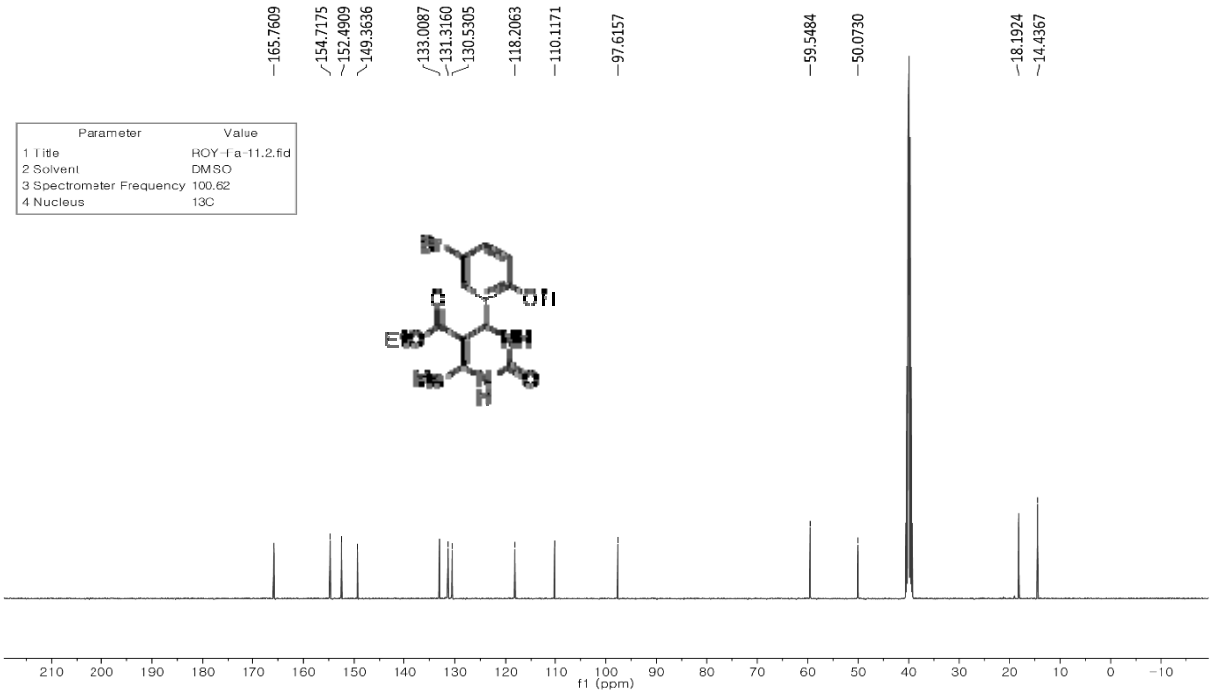
^{13}C NMR: Ethyl-4-(4-Acetamidophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4g).



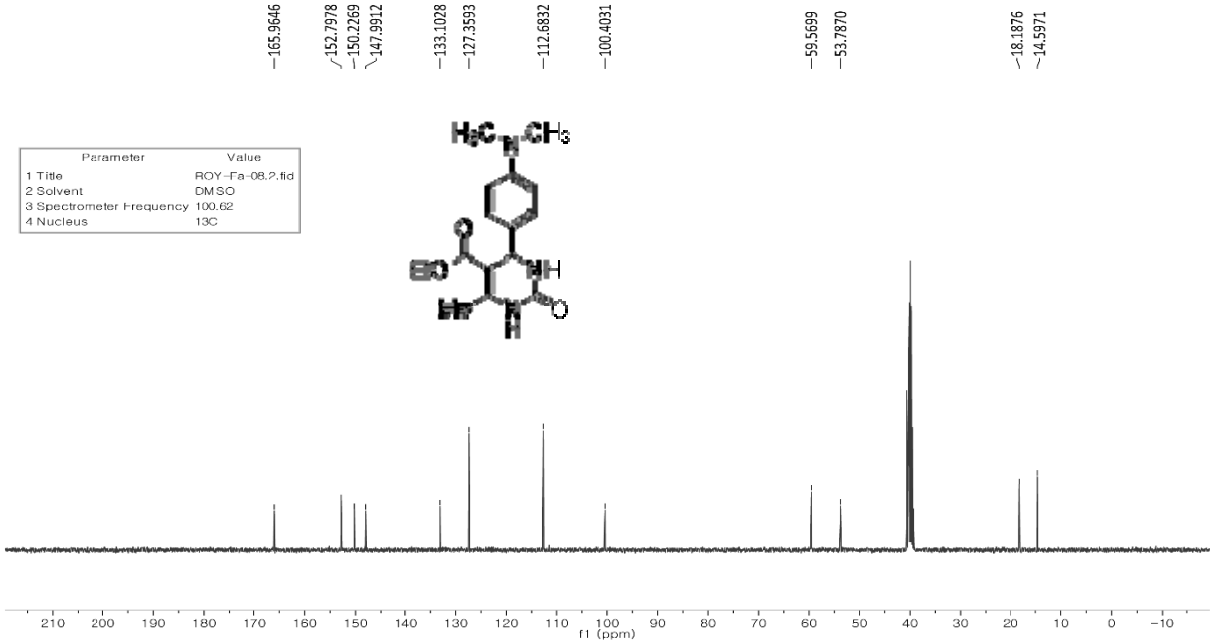
^{13}C NMR: Ethyl-4-(3-bromophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4h).



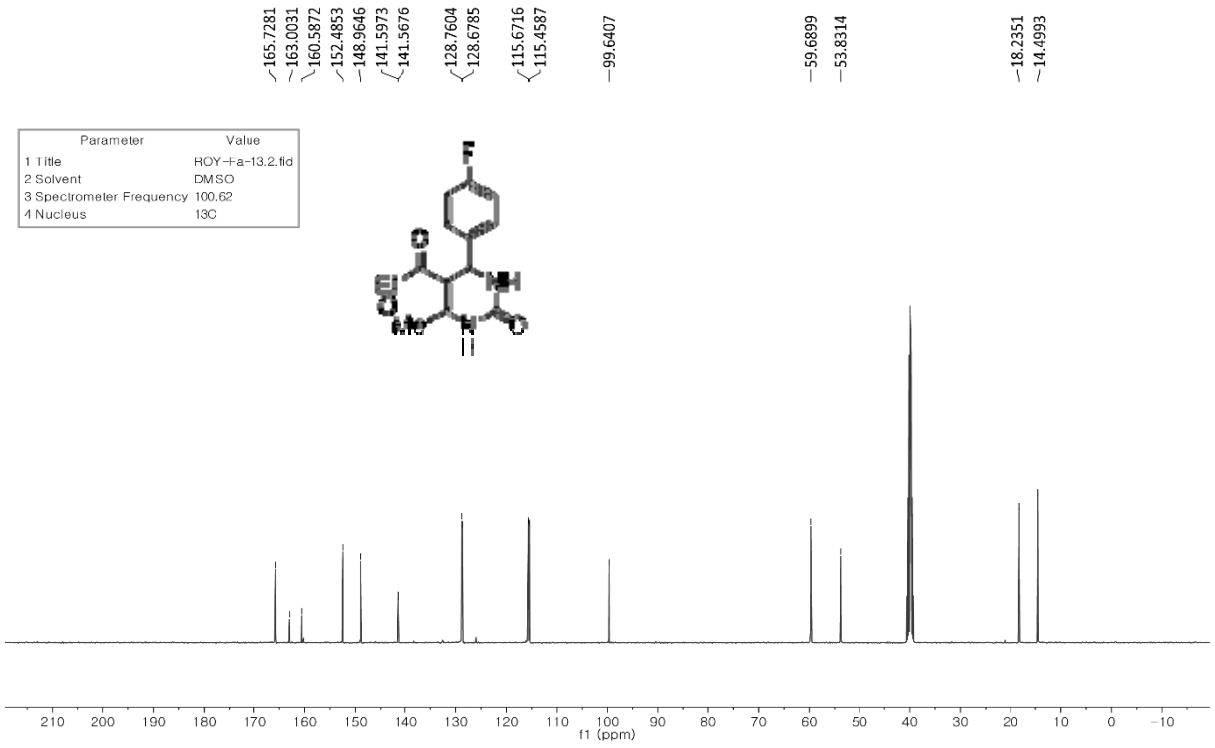
^{13}C NMR: Ethyl 4-(5-bromo-2-hydroxyphenyl)-6-methyl-2-oxo-1,2,3,4 tetrahydropyrimidine-5-carboxylate(4i).



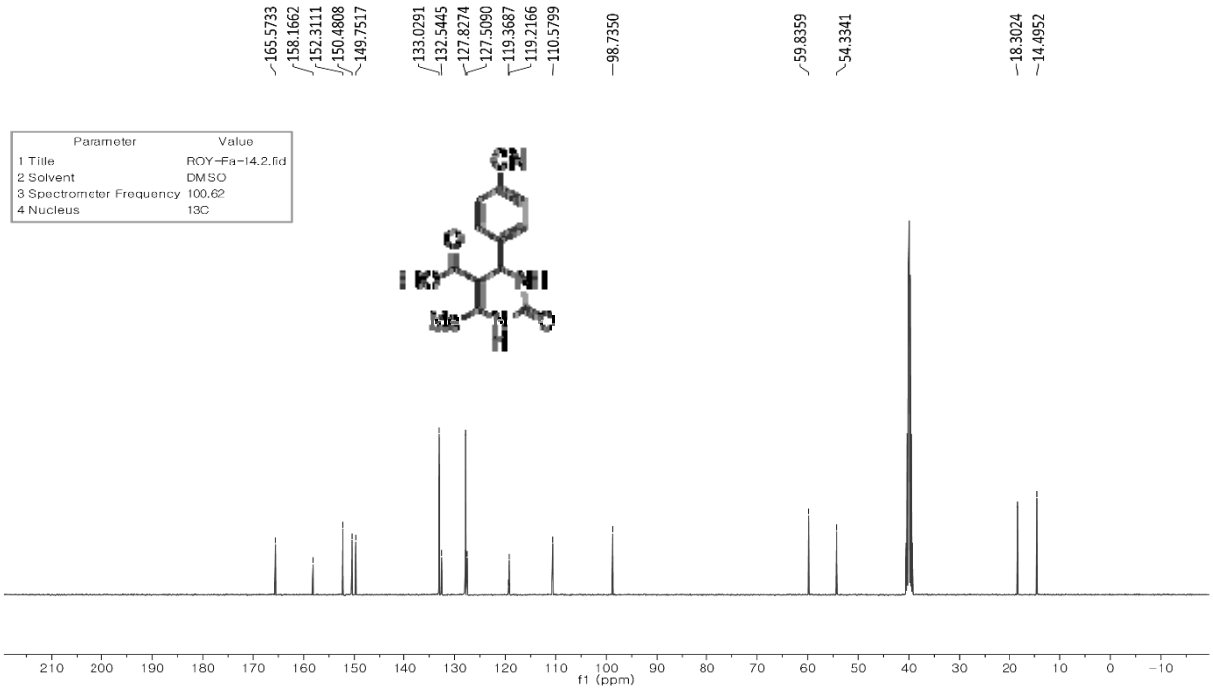
^{13}C NMR: Ethyl 4-(4-(dimethylamino)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4j).



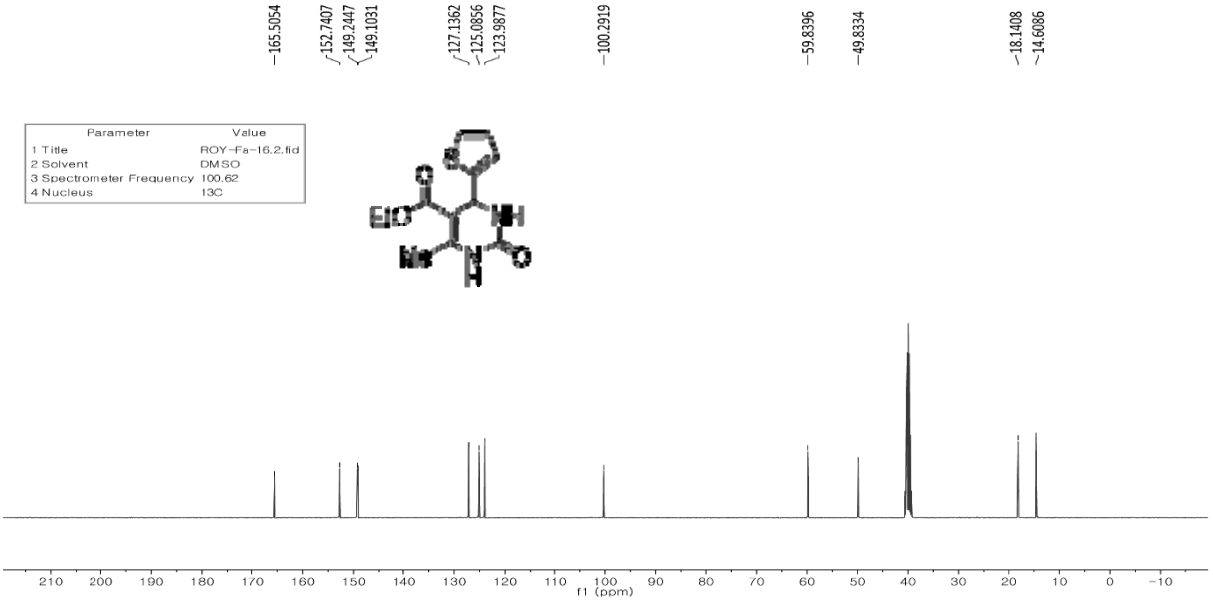
^{13}C NMR: Ethyl 4-(4-fluorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4k)



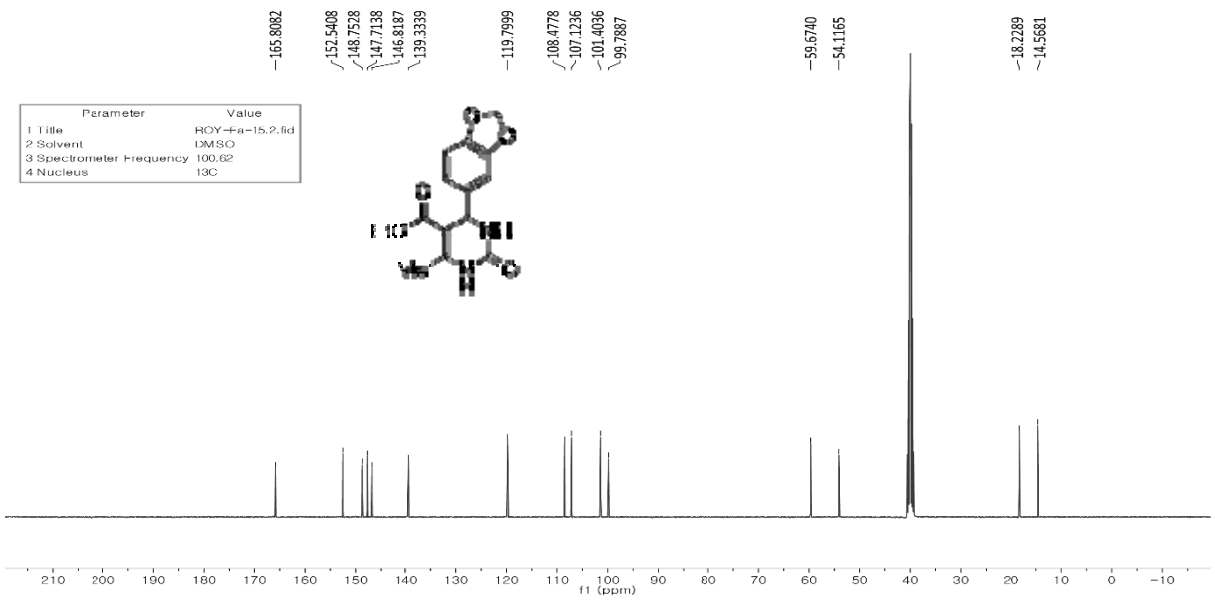
^{13}C NMR: Ethyl 4-(4-cyanophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4l).



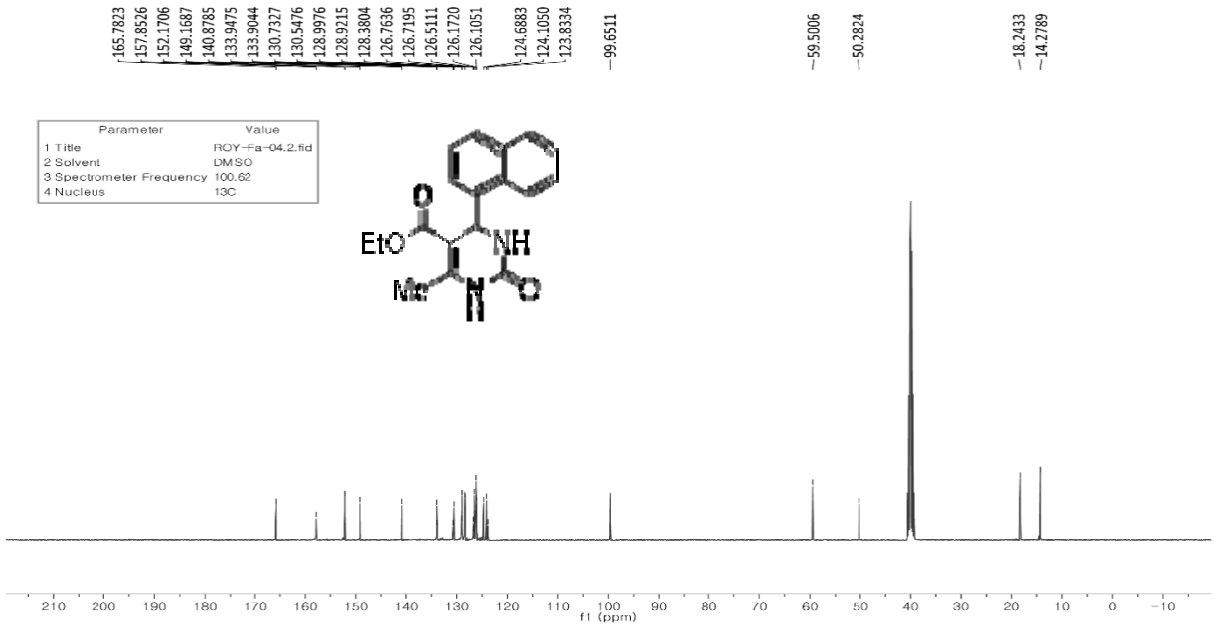
^{13}C NMR: Ethyl 6-methyl-2-oxo-4-(thiophen-2-yl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4m).



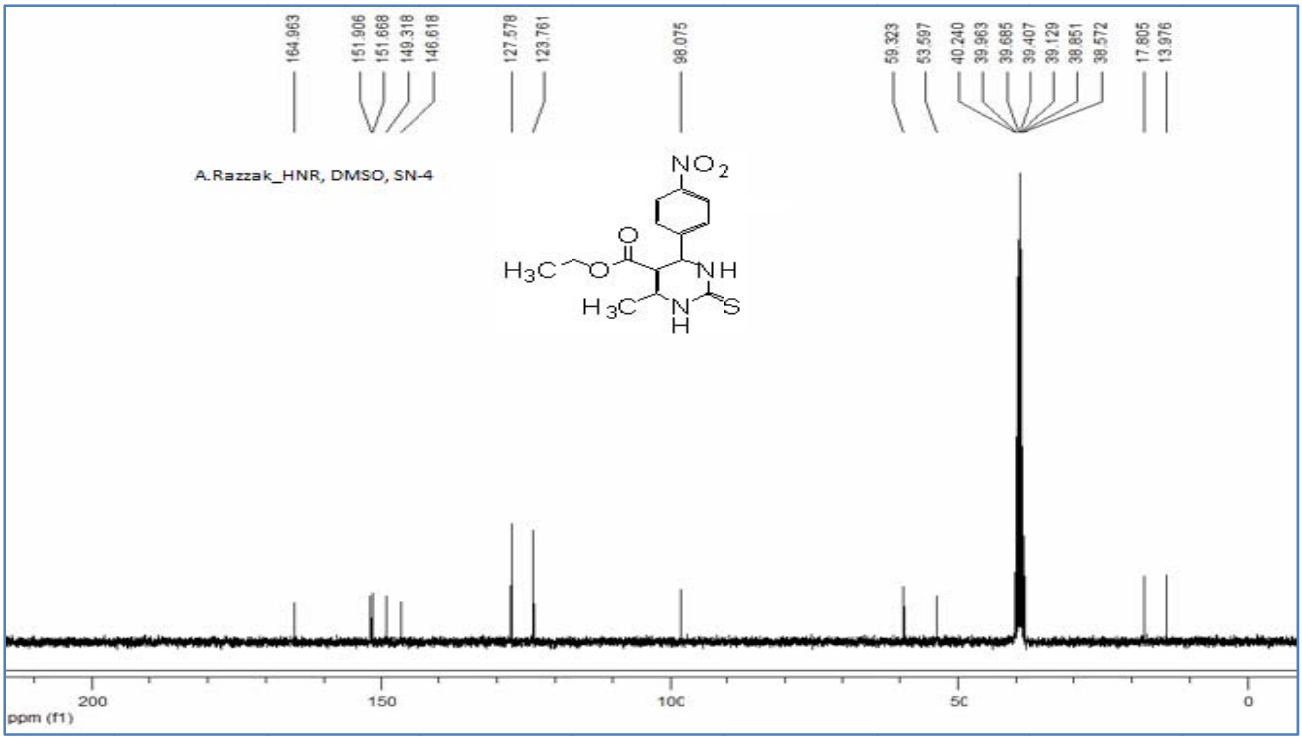
^{13}C NMR: Ethyl 4-(benzo[d][1,3]dioxol-5-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4n).



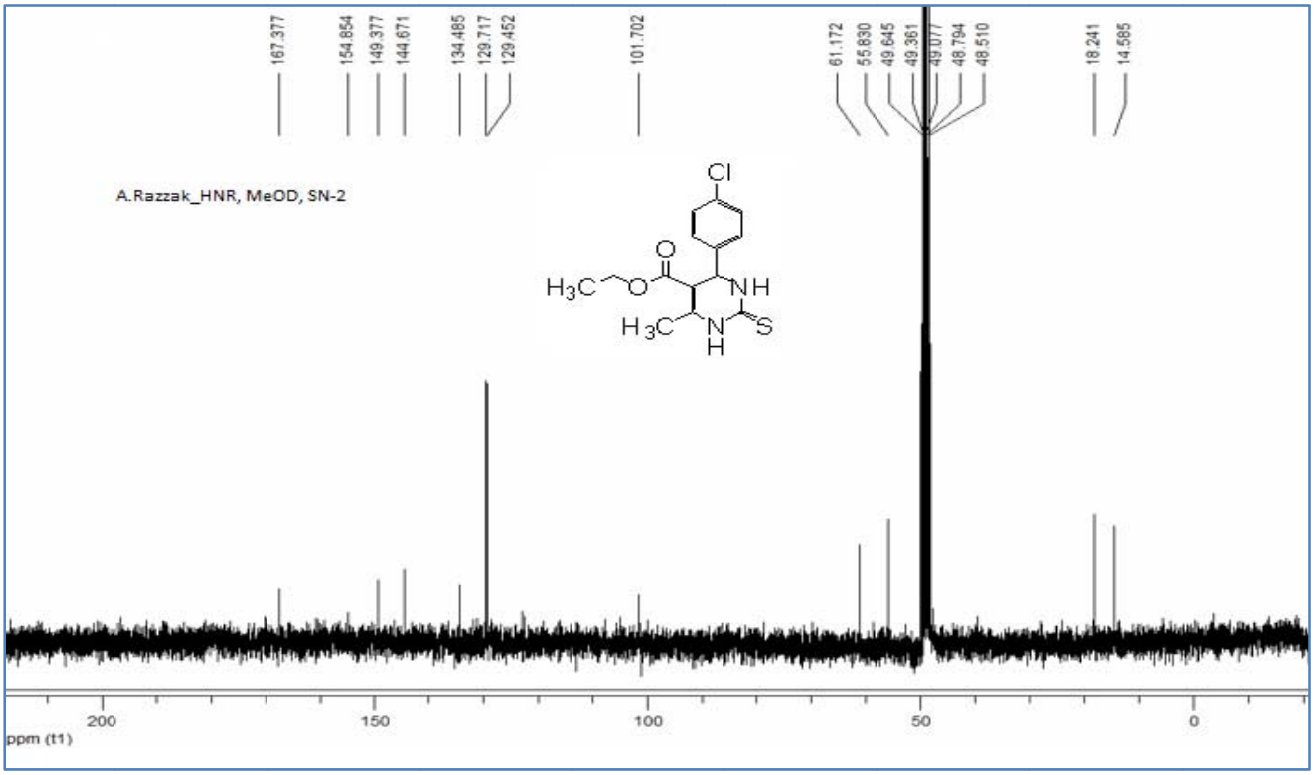
^{13}C NMR: Ethyl 6-methyl-4-(naphthalen-1-yl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4o).



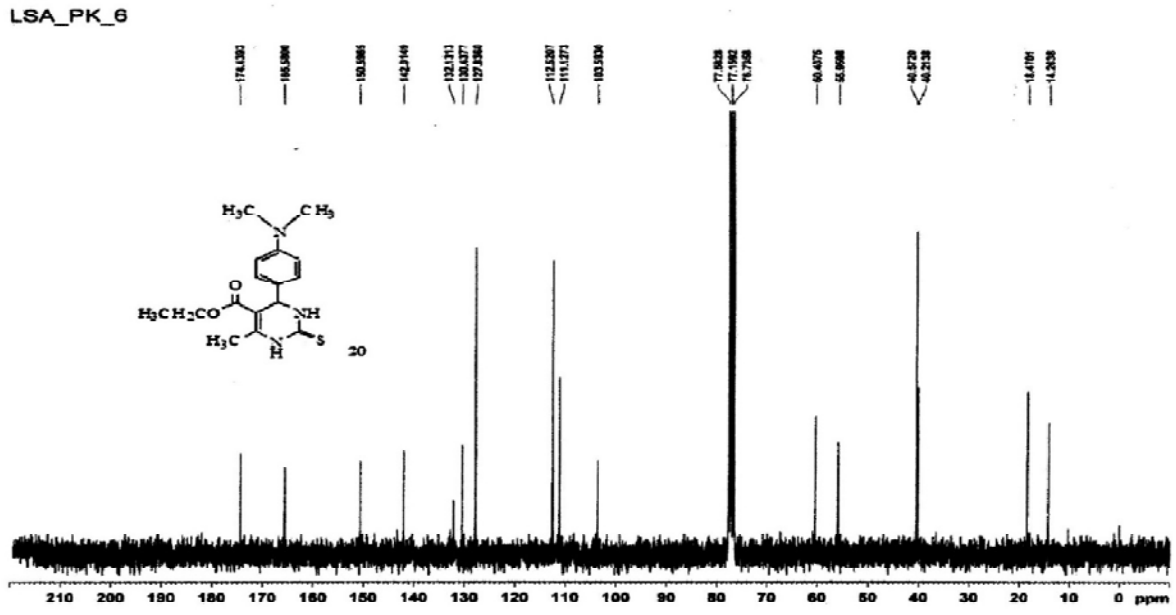
^{13}C NMR: Ethyl 6-methyl-4-(4-nitrophenyl)-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4r).



^{13}C NMR: Ethyl 4-(4-chlorophenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4u).



^{13}C NMR: Ethyl 4-(4-(dimethylamino)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4v).



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