

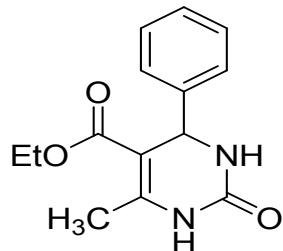
## **Supporting Information**

### **NanocrystallineCdS thin film as heterogenous, recyclable, catalyst for effective synthesis of Dihydropyrimidinones and a new class of carbazolyldihydropyrimidinones via an improved Biginelli protocol**

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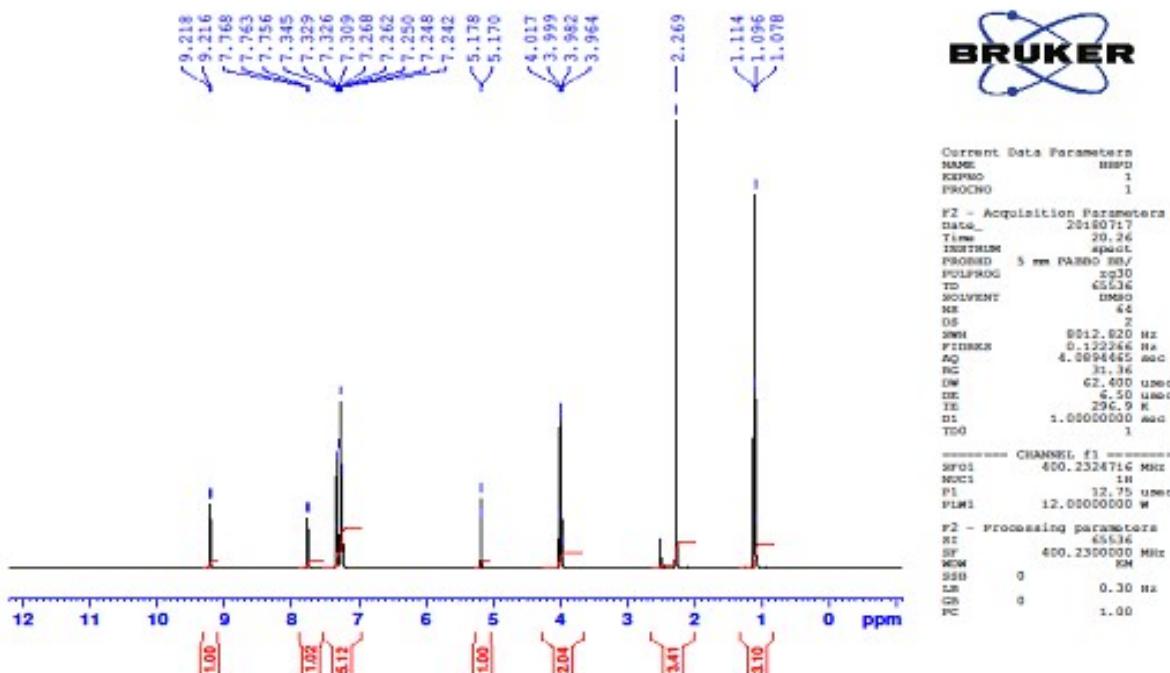
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#### **Ethyl-6-methyl-2-oxo-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4a)**

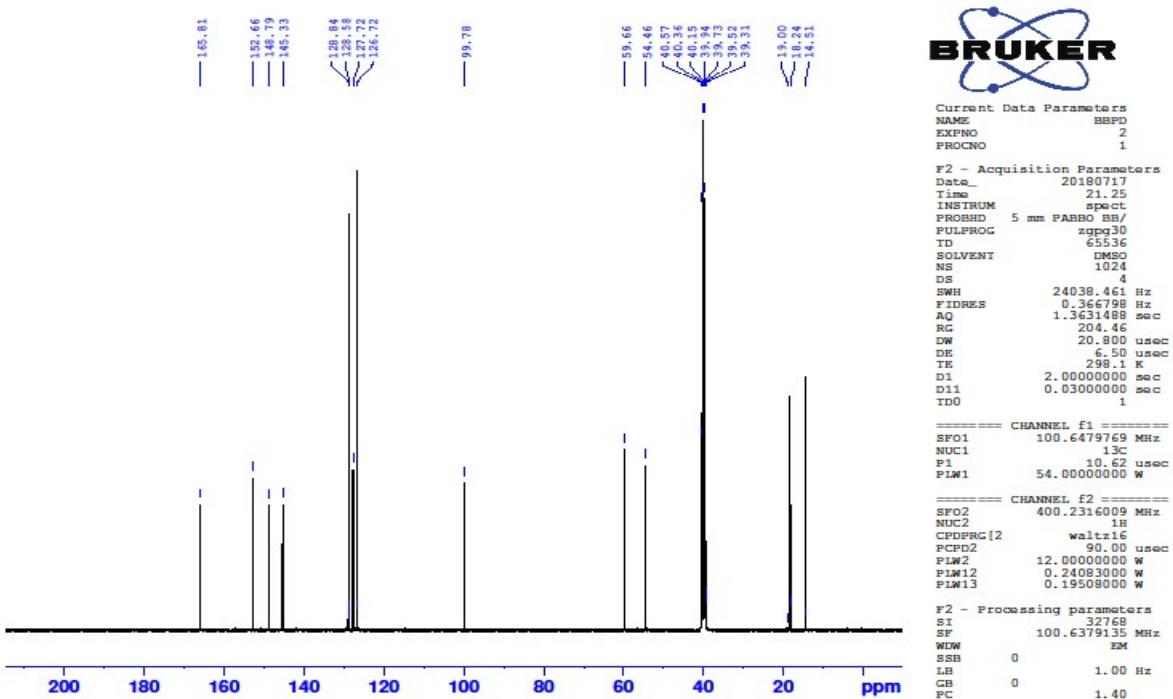


**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3639, 2967, 1896, 1608, 1223; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz)  $\delta$ : 9.21 (brs, 1H, NH), 7.76 (brs, 1H, NH), 7.34-7.24 (m, 5H), 5.17 (s, 1H), 4.01-3.96 (q, 2H), 2.26 (s, 3H), 1.11-1.07 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6)  $\delta$ : 165.8, 152.6, 148.7, 145.3, 128.8, 128.5, 127.7, 126.7, 99.7, 59.6, 54.4, 18.2, 14.5; **MS**: m/z: 260 ( $M^+$ ); **Anal.** Calcd. for C<sub>14</sub>H<sub>16</sub>N<sub>2</sub>O<sub>3</sub>: C, 64.60; H, 6.20; N, 10.76; **Found**: C, 64.47; H, 6.14; N, 10.59.

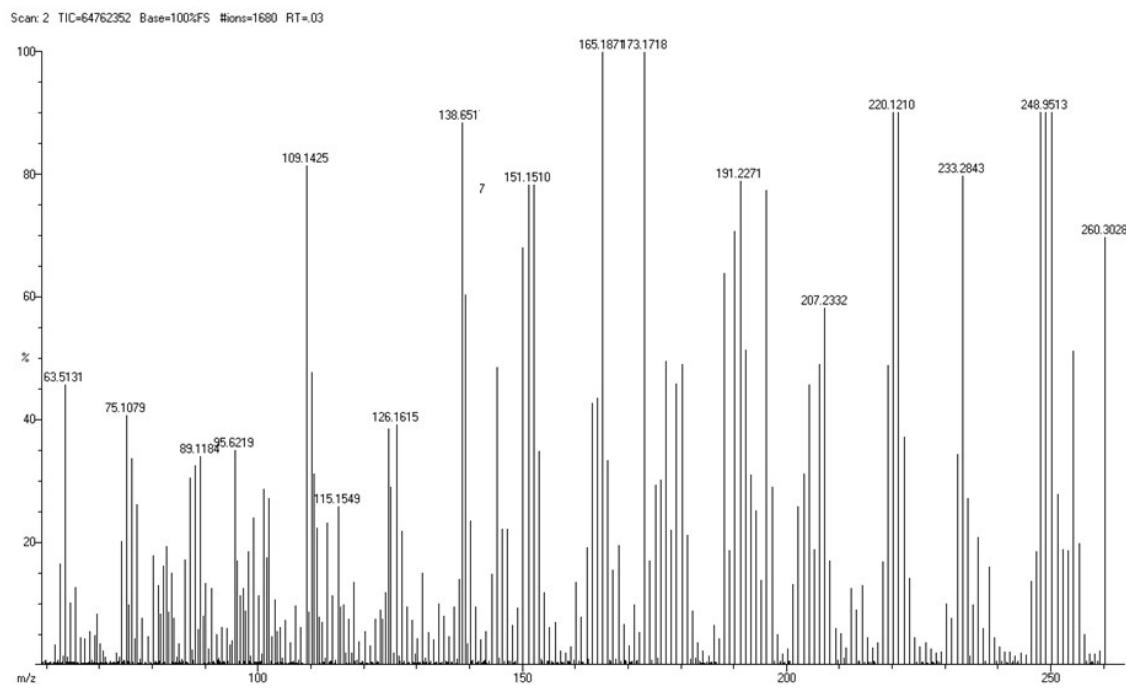
**<sup>1</sup>H-NMR Spectrum of Ethyl-6-methyl-2-oxo-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4a)**



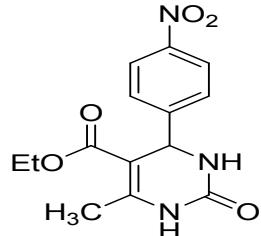
**<sup>13</sup>C-NMR Spectrum of Ethyl-6-methyl-2-oxo-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4a)**



**Mass Spectrum of Ethyl-6-methyl-2-oxo-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate  
(4a)**

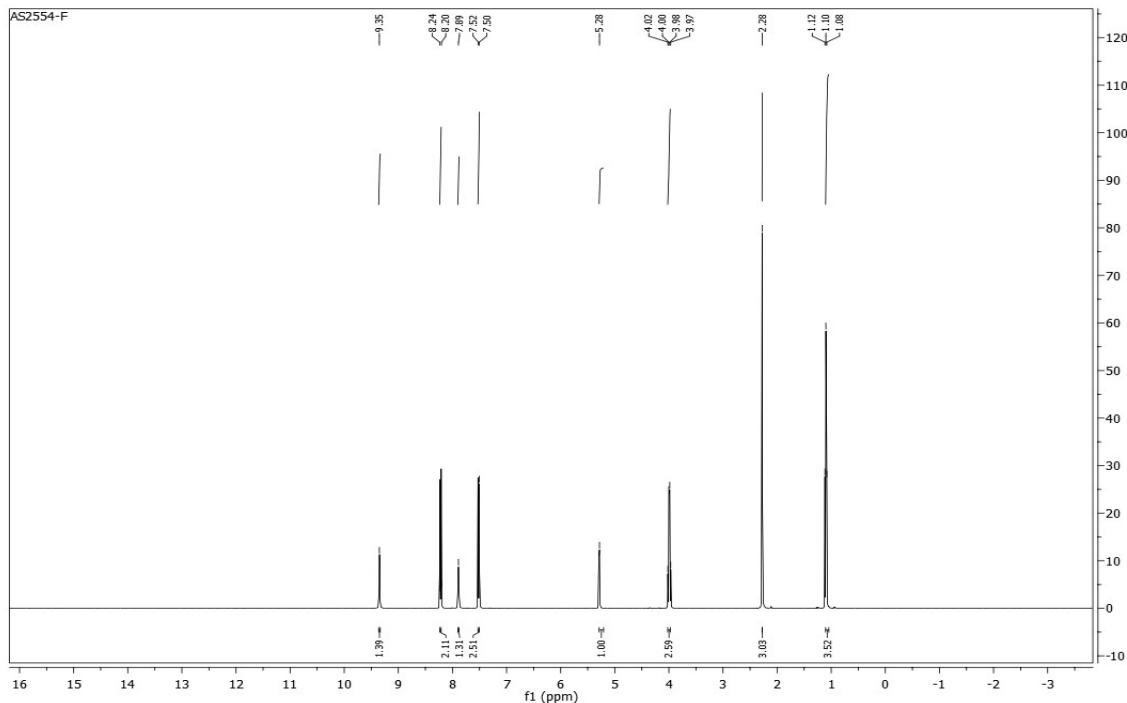


**Ethyl 6-methyl-4-(4-nitrophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4b)**

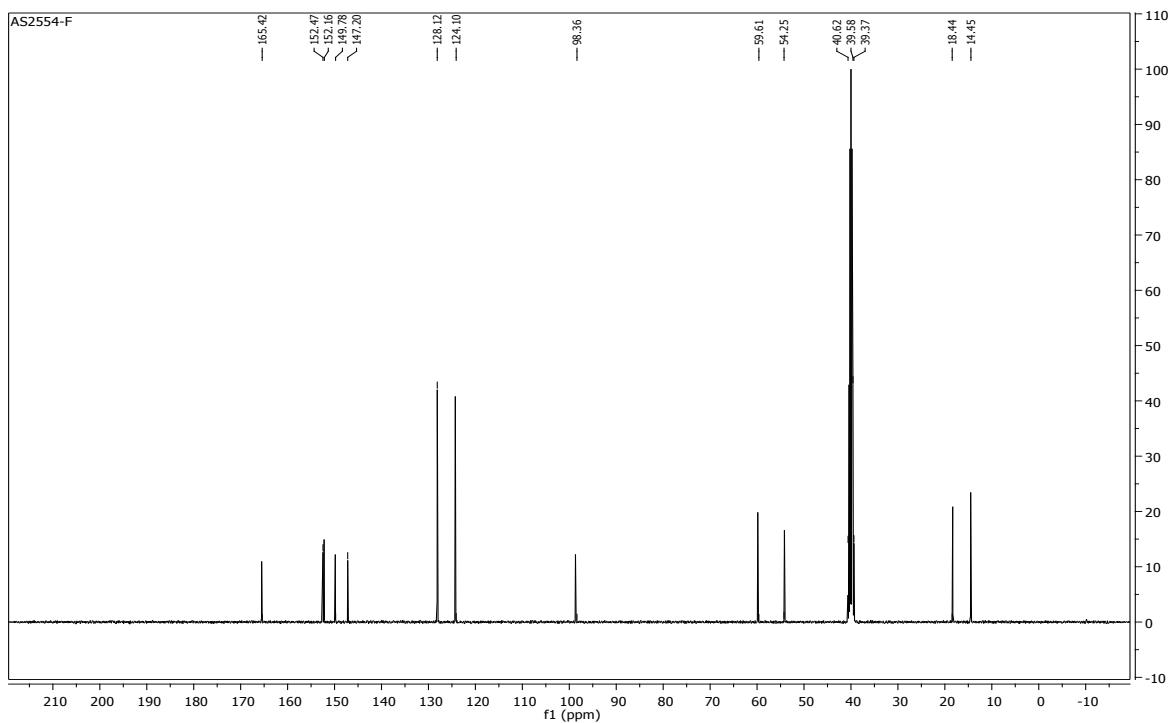


**FT- IR (KBr)**  $\nu_{\text{max}}$   $\text{cm}^{-1}$ : 3435, 2986, 1746, 1635;  **$^1\text{H NMR}$**  (DMSO-d6, 400 MHz)  $\delta$ : 9.35 (brs, 1H, NH), 8.24 (d, 2H), 7.89 (brs, 1H, NH), 7.52 (d, 2H), 5.28 (s, 1H), 4.02 (q, 2H), 2.28 (s, 3H), 1.12 (t, 3H);  **$^{13}\text{C NMR}$**  (100MHz, DMSO-d6)  $\delta$ : 165.4, 152.4, 152.1, 149.7, 147.2, 128.1, 124.1, 98.3, 69.6, 54.2, 18.4, 14.4; **MS:** m/z: 305 ( $\text{M}^+$ ); **Anal. Calcd. for**  $\text{C}_{14}\text{H}_{15}\text{N}_3\text{O}_5$ : C, 55.08; H, 4.97; N, 13.76; **Found:** C, 55.01; H, 4.84; N, 13.61.

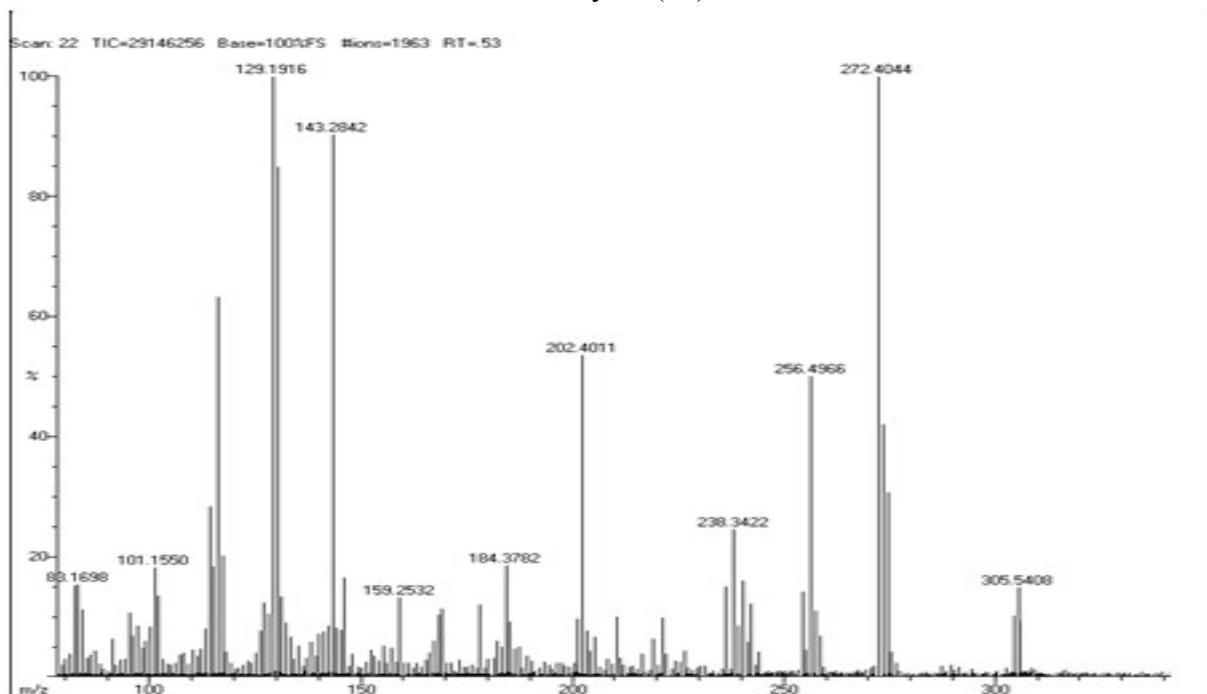
**<sup>1</sup>H-NMR Spectrum of Ethyl 6-methyl-4-(4-nitrophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4b)**



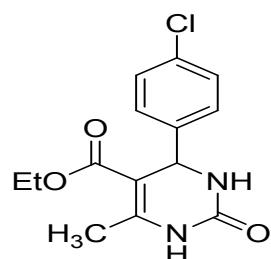
**<sup>13</sup>C-NMR Spectrum of Ethyl 6-methyl-4-(4-nitrophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4b)**



**Mass Spectrum** of Ethyl 6-methyl-4-(4-nitrophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate(4b)

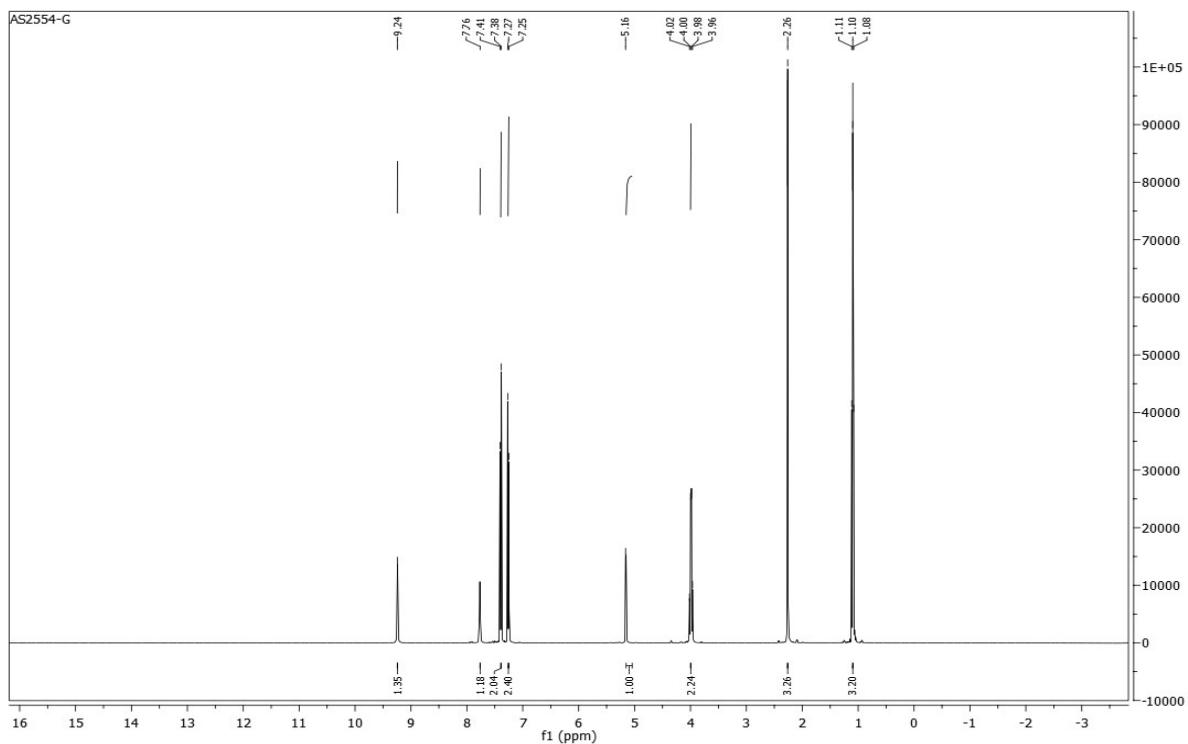


**Ethyl -4-(4-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4c)**

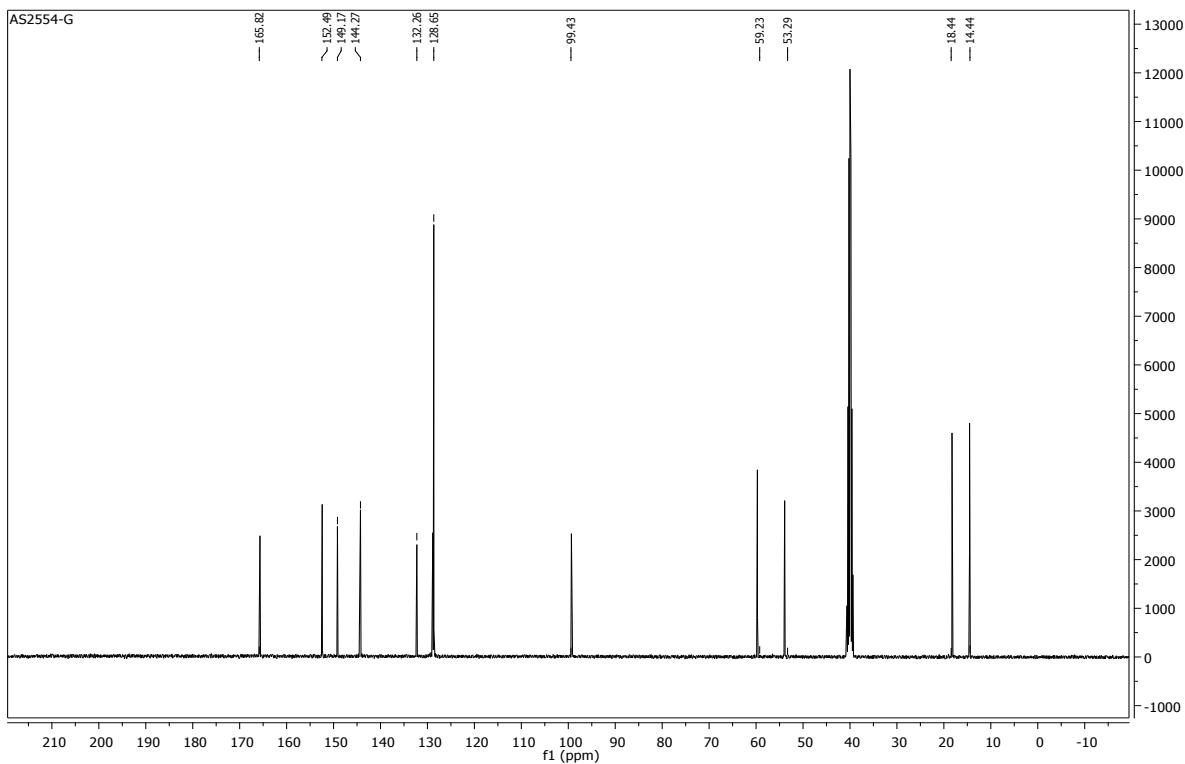


**FT- IR (KBr)**  $\nu_{\text{max}}$  cm<sup>-1</sup>: 3428, 2967, 1921, 1117, 758; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz)  $\delta$ : 9.24 (brs, 1H, NH), 7.76 (brs, 1H, NH), 7.41 (d, 2H), 7.27 (d, 2H), 5.16 (s, 1H), 4.02-3.96 (q, 2H), 2.26 (s, 3H), 1.11 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6)  $\delta$ : 165.8, 152.4, 149.1, 144.2, 132.2, 128.6, 99.4, 69.2, 53.2, 18.4, 14.4; **MS**: m/z: 294 ( $M^+$ ); **Anal. Calcd. for** C<sub>14</sub>H<sub>15</sub>ClN<sub>2</sub>O<sub>3</sub>: C, 55.05; H, 5.13; N, 9.50; **Found**: C, 55.01; H, 5.10; N, 9.45.

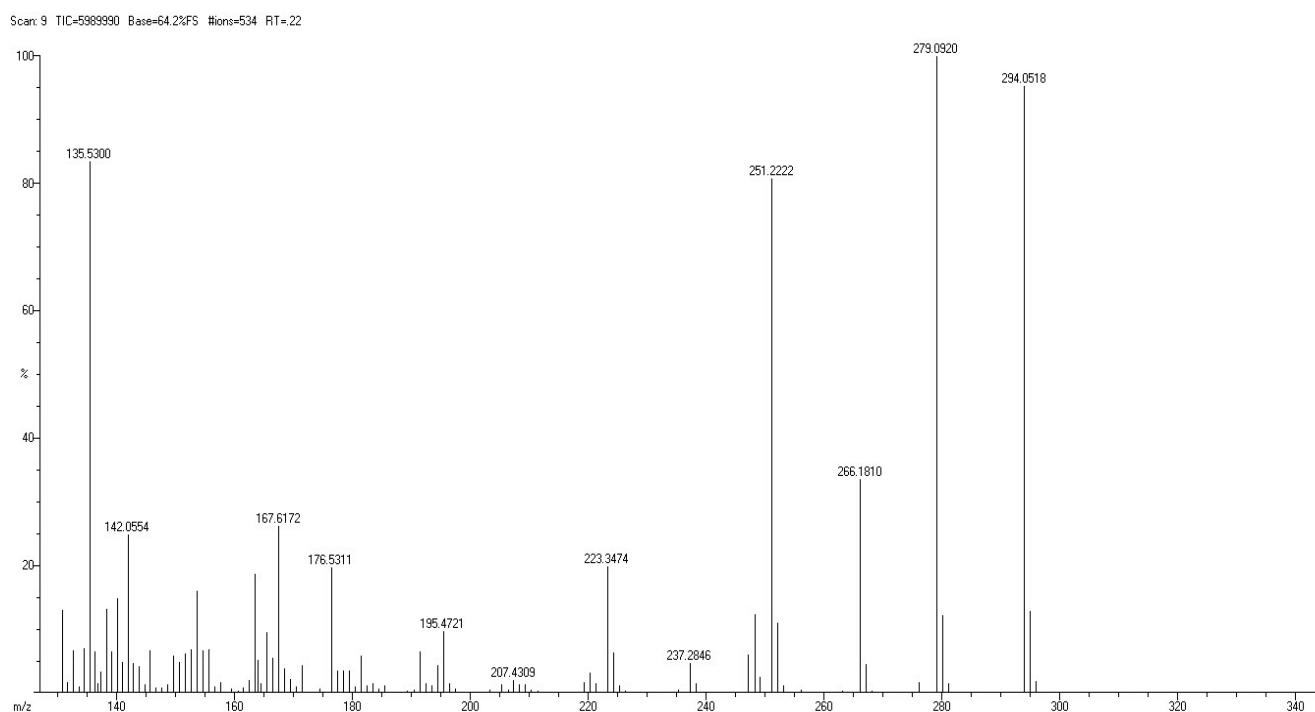
**<sup>1</sup>H-NMR Spectrum of Ethyl -4-(4-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4c)**



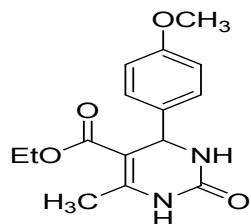
**<sup>13</sup>C-NMR Spectrum of Ethyl -4-(4-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4c)**



**Mass Spectrum of Ethyl -4-(4-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4c)**

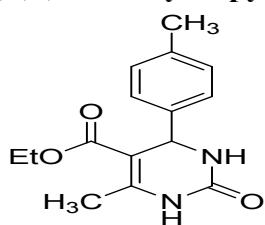


**Ethyl -4-(4-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4d)**



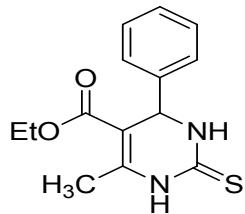
**FT- IR (KBr)** V<sub>max</sub> cm<sup>-1</sup>: 3533, 2969, 1724, 1638; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 9.27 (brs, 1H, NH), 7.72 (brs, 1H, NH), 7.21 (d, 2H), 6.87 (d, 2H), 5.15 (s, 1H), 4.24 (q, 2H), 3.79 (s, 3H, OCH<sub>3</sub>), 2.30 (s, 3H), 1.23 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 167.7, 158.6, 150.1, 147.3, 135.6, 125.7, 114.1, 106.8, 60.7, 55.2, 18.5, 14.3; **MS**: m/z: 290 (M<sup>+</sup>); **Anal. Calcd.** for C<sub>15</sub>H<sub>18</sub>N<sub>2</sub>O<sub>4</sub>: C, 62.06; H, 6.25; N, 9.65; **Found**: C, 63.02; H, 6.19; N, 9.59.

**Ethyl-6-methyl-2-oxo-4-(p-tolyl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4e)**



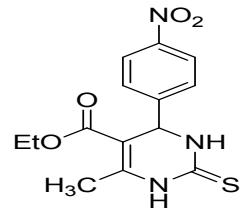
**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3477, 2971, 1729, 1640; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 9.22 (brs, 1H, NH), 7.71 (brs, 1H, NH), 7.11 (m, 4H), 5.19 (s, 1H), 4.30 (q, 2H), 2.34 (s, 3H, CH<sub>3</sub>), 2.32 (s, 3H), 1.30 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 167.5, 150.6, 147.3, 140.3, 136.4, 128.8, 126.8, 106.0, 61.0, 55.7, 21.3, 18.3, 14.5; **MS**: m/z: 274 (M<sup>+</sup>); **Anal.** **Calcd.** for C<sub>15</sub>H<sub>18</sub>N<sub>2</sub>O<sub>3</sub>: C, 65.68; H, 6.61; N, 10.21; **Found**: C, 65.55; H, 6.54; N, 10.16.

### Ethyl-6-methyl-4-phenyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (6a)



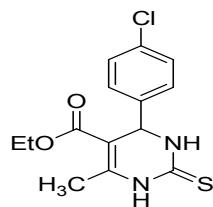
**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3340, 2979, 1823, 1597, 1122; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 9.41 (brs, 1H, NH), 7.83 (brs, 1H, NH), 7.36-7.24 (m, 5H), 5.16 (s, 1H), 4.20 (q, 2H), 2.26 (s, 3H), 1.19 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 174.1, 167.2, 160.3, 143.3, 128.5, 126.9, 126.7, 104.2, 60.6, 54.3, 17.9, 14.2; **MS**: m/z: 276 (M<sup>+</sup>); **Anal. Calcd.** for C<sub>14</sub>H<sub>16</sub>N<sub>2</sub>O<sub>2</sub>S: C, 60.85; H, 5.84; N, 10.14; **Found**: C, 60.77; H, 5.75; N, 10.09.

### Ethyl-6-methyl-4-(4-nitrophenyl)-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (6b)



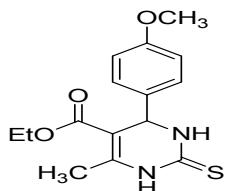
**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3398, 2943, 1722, 1640, 1541, 1533; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 9.36 (brs, 1H, NH), 7.85 (brs, 1H, NH), 7.16 (d, 2H), 7.49 (d, 2H), 5.20 (s, 1H), 4.26 (q, 2H), 2.29 (s, 3H), 1.29 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 174.5, 167.6, 160.2, 149.4, 145.9, 128.3, 125.1, 104.6, 61.2, 58.6, 18.5, 14.5; **MS**: m/z: 321 (M<sup>+</sup>); **Anal. Calcd.** for C<sub>14</sub>H<sub>15</sub>N<sub>3</sub>O<sub>4</sub>S: C, 52.33; H, 4.70; N, 13.08; **Found**: C, 52.27; H, 4.64; N, 13.03.

**Ethyl-4-(4-chlorophenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (6c)**



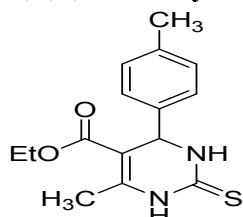
**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3439, 2939, 1723, 1630, 1549, 1543, 761; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 9.42 (brs, 1H, NH), 7.88 (brs, 1H, NH), 7.37-7.34 (m, 4H), 5.22 (s, 1H), 4.31 (q, 2H), 2.24 (s, 3H), 1.32 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 174.7, 167.3, 160.5, 141.4, 132.3, 128.6, 126.1, 104.1, 61.3, 58.9, 18.1, 14.0; **MS**: m/z: 310 (M<sup>+</sup>); **Anal. Calcd.** for C<sub>14</sub>H<sub>15</sub>ClN<sub>2</sub>O<sub>2</sub>S: C, 54.10; H, 4.86; N, 9.01; **Found**: C, 54.04; H, 4.74; N, 9.00.

**Ethyl-4-(4-methoxyphenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (6d)**



**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3341, 2945, 1725, 1636, 1575, 1541; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 9.45 (brs, 1H, NH), 7.85 (brs, 1H, NH), 7.12 (d, 2H), 6.87 (d, 2H), 5.27 (s, 1H), 4.30 (q, 2H), 3.99 (s, 3H, OCH<sub>3</sub>), 2.28 (s, 3H), 1.18 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 174.4, 167.5, 160.6, 158.6, 135.6, 125.7, 114.3, 104.4, 61.4, 55.7, 55.4, 18.6, 14.7; **MS**: m/z: 306 (M<sup>+</sup>); **Anal. Calcd.** for C<sub>15</sub>H<sub>18</sub>N<sub>2</sub>O<sub>3</sub>S: C, 58.80; H, 5.92; N, 9.14; **Found**: C, 58.71; H, 5.84; N, 9.08.

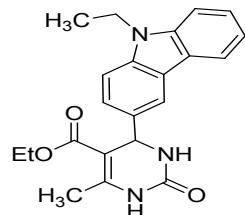
**Ethyl-6-methyl-2-thioxo-4-(*p*-tolyl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate (6e)**



**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3469, 2956, 1729, 1631, 1578, 1547; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 9.36 (brs, 1H, NH), 7.76 (brs, 1H, NH), 7.12 (d, 2H), 6.87 (d, 2H), 5.27 (s, 1H), 4.30 (q, 2H), 3.85 (s, 3H, OCH<sub>3</sub>), 2.28 (s, 3H), 1.38 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ:

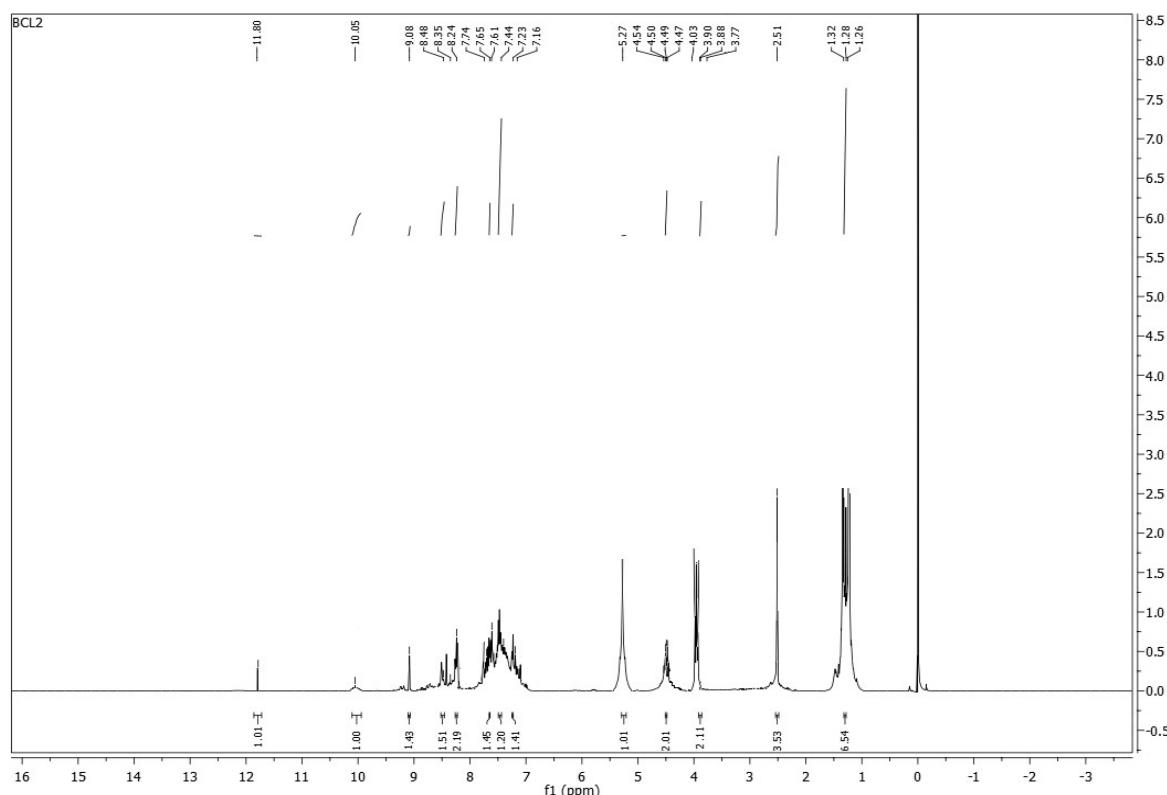
174.4, 167.5, 160.6, 158.6, 135.6, 125.7, 114.3, 104.4, 60.9, 56.7, 55.8, 18.6, 14.7; **MS**: m/z: 290 ( $M^+$ ); **Anal. Calcd.** for  $C_{15}H_{18}N_2O_2S$ : C, 62.04; H, 6.25; N, 9.65; **Found**: C, 62.00; H, 6.16; N, 9.59.

**Ethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (7a)**

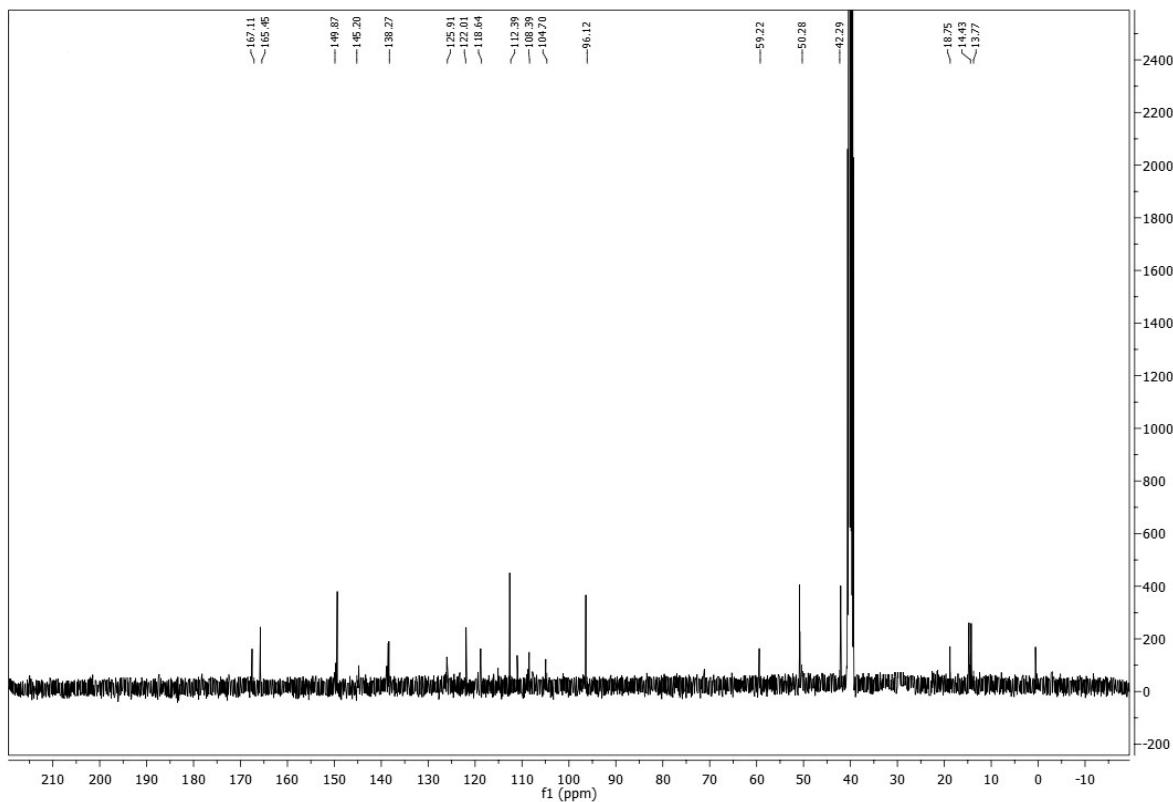


**FT- IR (KBr)**  $V_{max}$  cm<sup>-1</sup>: 3455, 2973, 1833, 1739, 1681, 1544; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 11.80 (brs, 1H, NH), 10.05 (brs, 1H, NH), 9.08 (s, 1H), 8.48 (d, 1H), 8.24 (d, 2H), 7.74 (d, 1H), 7.44 (d, 1H), 7.23 (s, 1H), 5.27 (s, 1H), 4.54 (q, 2H), 4.03 (q, 2H), 2.51 (s, 3H), 1.32 (t, 6H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 167.1, 165.4, 149.8, 145.2, 138.2, 125.9, 122.0, 118.6, 112.3, 108.3, 104.7, 96.1, 59.2, 50.2, 42.2, 18.7, 14.4, 13.7; **MS**: m/z: 377 ( $M^+$ ); **Anal. Calcd.** for  $C_{22}H_{23}N_3O_3$ : C, 70.01; H, 6.14; N, 11.13; **Found**: C, 70.00; H, 6.11; N, 11.05.

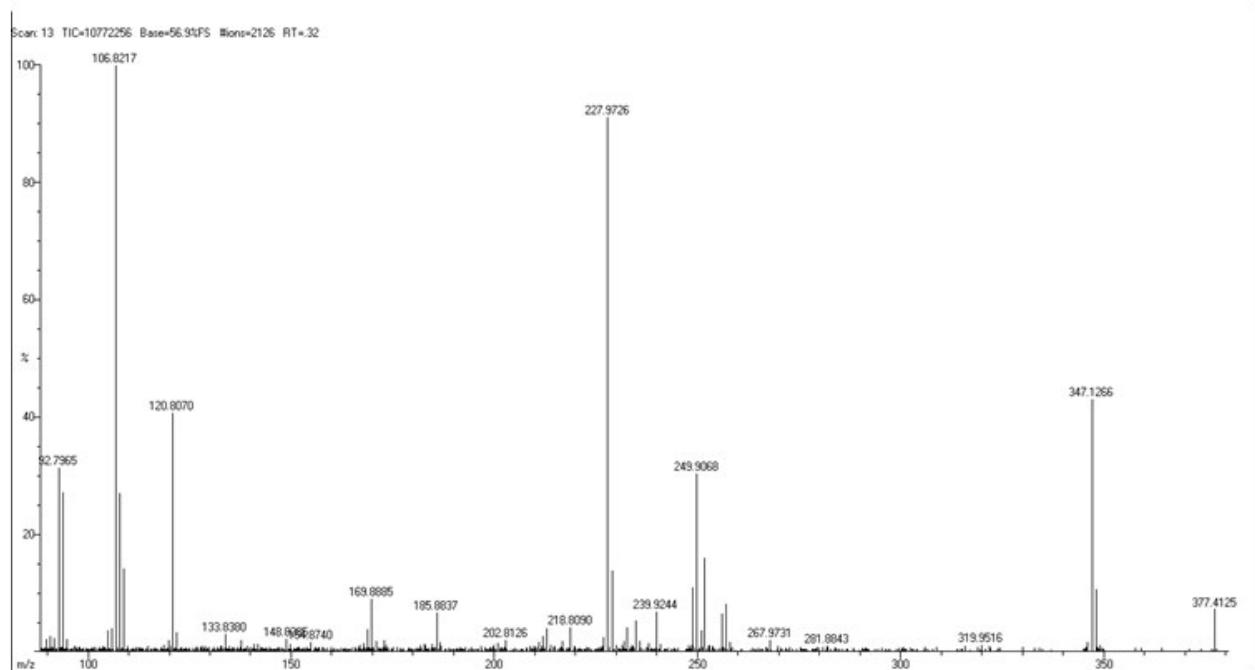
**<sup>1</sup>H-NMR Spectrum of Ethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (7a)**



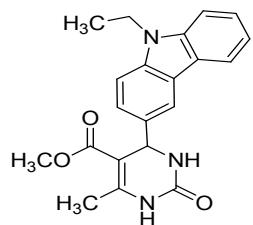
**C<sup>13</sup>-NMR Spectrum of Ethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (7a)**



**Mass Spectrum of Ethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (7a)**

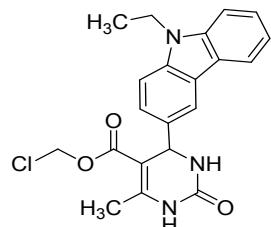


**Methyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (7b)**



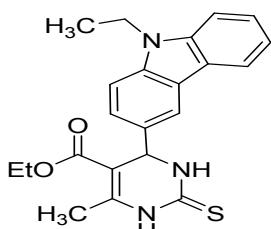
**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3572, 2967, 1823, 1740, 1657, 1534; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 11.47 (brs, 1H, NH), 10.09 (brs, 1H, NH), 9.11 (s, 1H), 8.39 (d, 1H), 8.21 (d, 1H), 7.65 (d, 1H), 7.44 (d, 2H), 7.31 (s, 1H), 5.19 (s, 1H), 4.55 (q, 2H), 3.76 (s, 3H), 2.24 (s, 3H), 1.35 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 167.7, 165.2, 149.8, 145.6, 138.4, 125.5, 122.3, 118.7, 112.3, 108.9, 104.5, 96.6, 59.5, 51.9, 42.4, 18.6, 14.5, 13.2; **MS**: m/z: 363 (M<sup>+</sup>); **Anal. Calcd. for** C<sub>21</sub>H<sub>21</sub>N<sub>3</sub>O<sub>3</sub>: C, 69.41; H, 5.82; N, 11.56; **Found**: C, 69.34; H, 5.76; N, 11.50.

**Chloromethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (7c)**



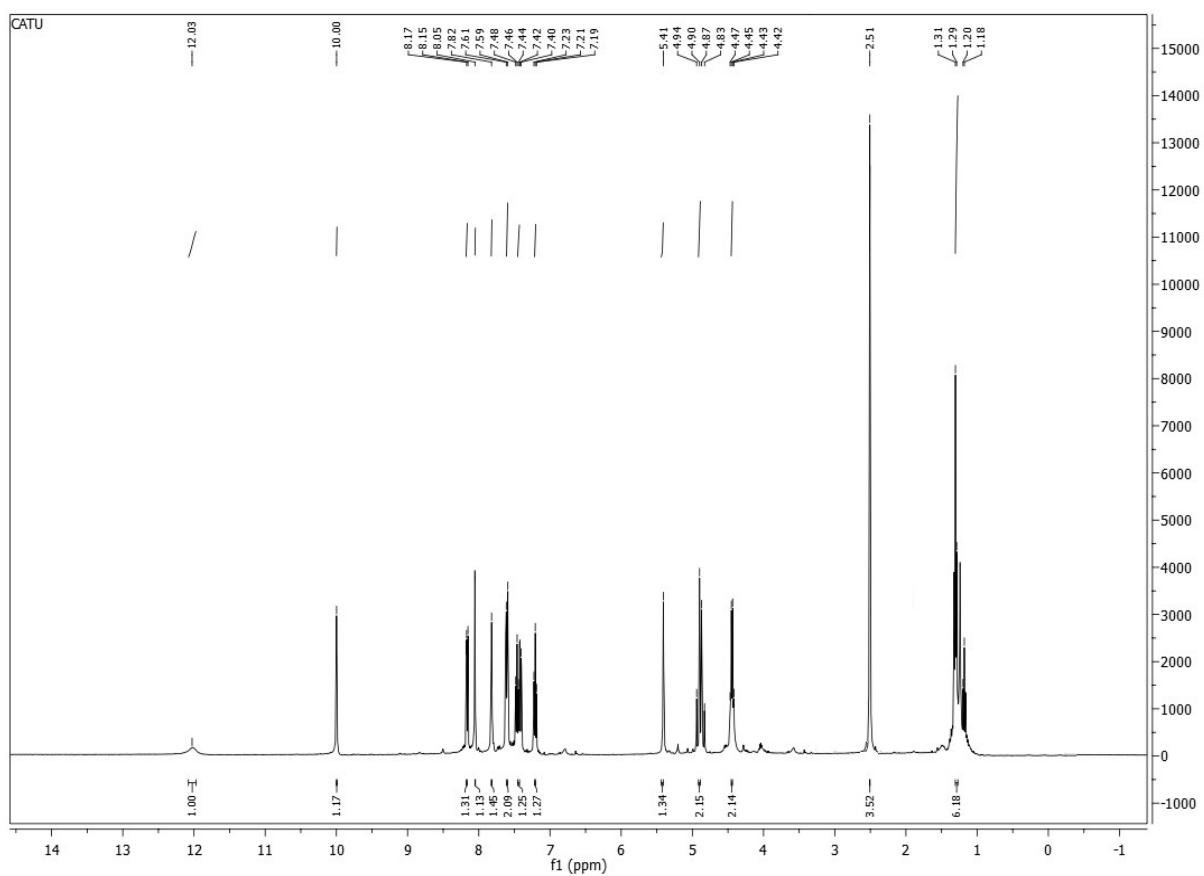
**FT- IR (KBr)**  $V_{\max}$  cm<sup>-1</sup>: 3477, 2979, 1825, 1738, 1670, 1551; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 11.34 (brs, 1H, NH), 10.13 (brs, 1H, NH), 9.14 (d, 1H), 8.33 (s, 1H), 7.64 (d, 1H), 7.50 (d, 1H), 7.47 (d, 2H), 7.39 (s, 1H), 6.24 (s, 2H), 5.18 (s, 1H), 4.56 (q, 2H), 2.29 (s, 3H), 1.30 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 167.5, 165.7, 149.2, 145.7, 138.2, 125.9, 122.6, 118.7, 112.5, 108.3, 104.5, 94.4, 68.8, 53.5, 40.4, 17.3, 14.3, 13.7; **MS**: m/z: 397 (M<sup>+</sup>); **Anal. Calcd. for** C<sub>21</sub>H<sub>20</sub>ClN<sub>3</sub>O<sub>3</sub>: C, 63.40; H, 5.07; N, 10.56; **Found**: C, 63.32; H, 5.01; N, 11.51.

**Ethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (8a)**

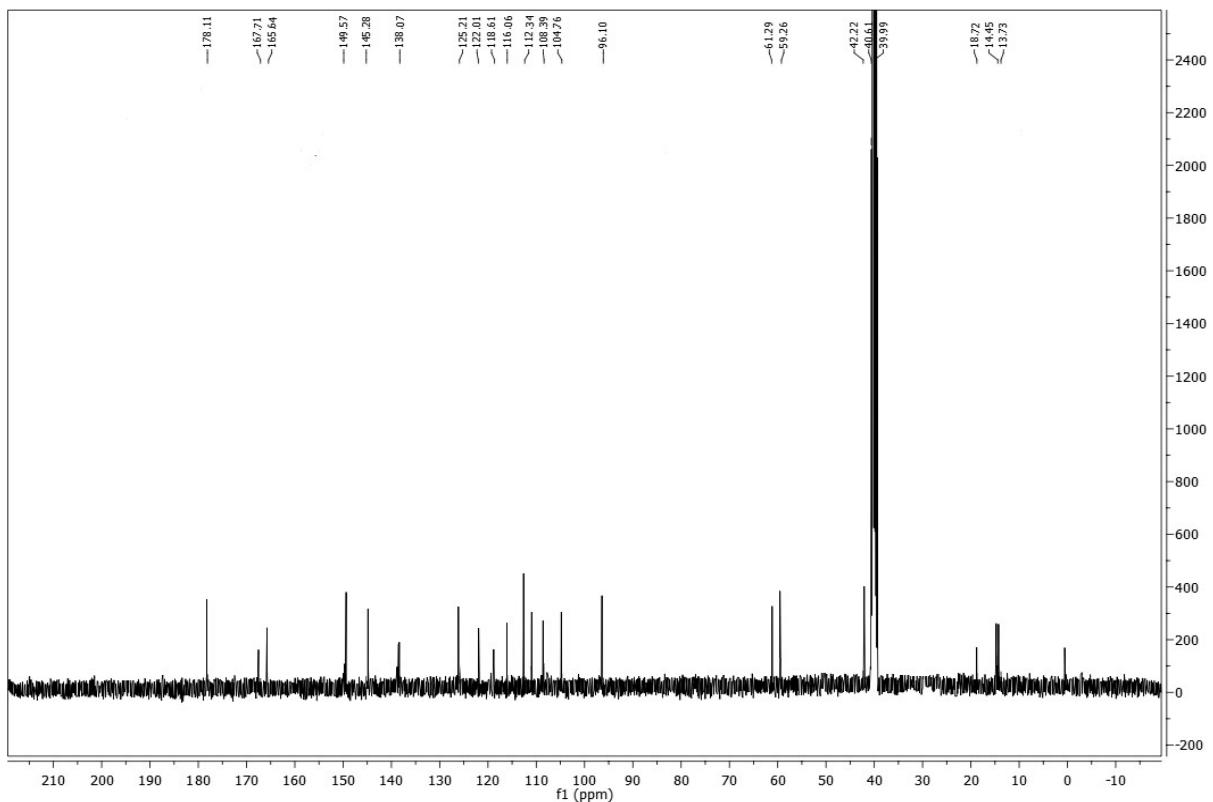


**FT- IR (KBr)**  $\nu_{\text{max}}$   $\text{cm}^{-1}$ : 3560, 2949, 1825, 1739, 1670, 1545;  **$^1\text{H NMR}$**  (DMSO-d6, 400 MHz)  $\delta$ : 12.03 (brs, 1H, NH), 10.00 (brs, 1H, NH), 8.17 (d, 1H), 8.05 (d, 1H), 7.61 (s, 1H), 7.59 (d, 2H), 7.48 (d, 1H), 7.40 (s, 1H), 5.41 (s, 1H), 4.94 (q, 2H), 4.44 (q, 2H), 2.51 (s, 3H), 1.31 (t, 6H);  **$^{13}\text{C NMR}$**  (100MHz, DMSO-d6)  $\delta$ : 178.1, 167.7, 165.6, 149.5, 144.2, 139.0, 125.2, 122.0, 118.6, 116.0, 112.3, 108.3, 104.7, 96.1, 61.2, 59.2, 42.2, 18.7, 14.4, 13.7; **MS**: m/z: 393 ( $\text{M}^+$ ); **Anal. Calcd.** for  $\text{C}_{22}\text{H}_{23}\text{N}_3\text{O}_2\text{S}$ : C, 67.15; H, 5.89; N, 10.68; **Found**: C, 67.10; H, 5.76; N, 10.59.

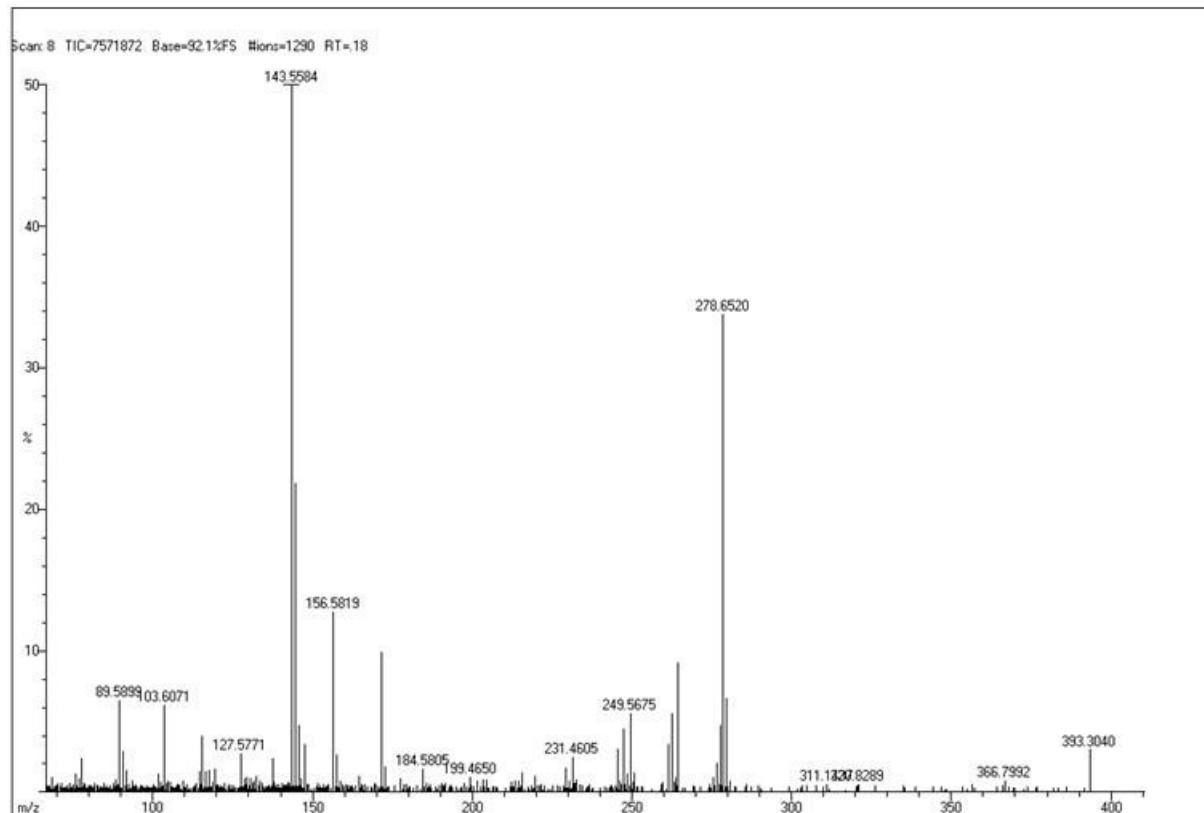
**$^1\text{H-NMR Spectrum}$  of Ethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (8a)**



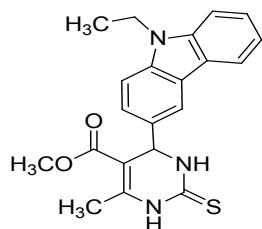
**C<sup>13</sup>-NMR Spectrum of Ethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (8a)**



**Mass Spectrum of Ethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (8a)**

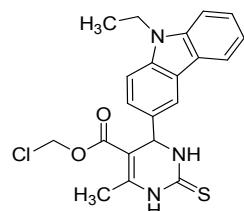


**Methyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (8b)**



**FT- IR (KBr)** V<sub>max</sub> cm<sup>-1</sup>: 3449, 2985, 1843, 1750, 1543, 1122; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 13.71 (brs, 1H, NH), 10.07 (brs, 1H, NH), 8.12 (s, 1H), 7.89 (s, 1H), 7.59-7.39 (m, 5H), 5.19 (s, 1H), 4.52 (q, 2H), 3.79 (s, 3H), 2.23 (s, 3H), 1.31 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 174.5, 167.1, 165.5, 149.6, 145.6, 138.4, 125.7, 122.8, 118.1, 116.8, 112.1, 108.4, 104.1, 96.7, 61.7, 58.9, 42.7, 18.5, 14.8, 13.1; **MS**: m/z: 379 (M<sup>+</sup>); **Anal. Calcd. for** C<sub>21</sub>H<sub>21</sub>N<sub>3</sub>O<sub>2</sub>S: C, 66.47; H, 5.58; N, 11.07; **Found**: C, 66.40; H, 5.46; N, 11.00.

**Chloromethyl-4-(9-ethyl-9H-carbazol-3-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (8c)**



**FT- IR (KBr)** V<sub>max</sub> cm<sup>-1</sup>: 3536, 2988, 1849, 1740, 1549, 1117; **<sup>1</sup>H NMR** (DMSO-d6, 400 MHz) δ: 13.69 (brs, 1H, NH), 10.11 (brs, 1H, NH), 8.25 (s, 1H), 7.92 (s, 1H), 7.61-7.37 (m, 5H), 6.24 (s, 2H), 5.22 (s, 1H), 4.50 (q, 2H), 2.30 (s, 3H), 1.35 (t, 3H); **<sup>13</sup>C NMR** (100MHz, DMSO-d6) δ: 174.3, 167.4, 165.1, 149.9, 145.1, 138.0, 125.3, 122.5, 118.4, 116.3, 112.5, 108.6, 104.3, 96.3, 61.5, 58.7, 42.5, 18.6, 14.2, 13.2; **MS**: m/z: 413 (M<sup>+</sup>); **Anal. Calcd. for** C<sub>21</sub>H<sub>20</sub>ClN<sub>3</sub>O<sub>2</sub>S: C, 60.94; H, 4.87; N, 10.15; **Found**: C, 60.80; H, 4.76; N, 10.09.