

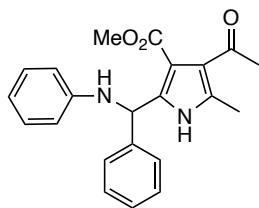
# SYNTHESIS OF POLYSUBSTITUTED 5,6-DIHYDROPYRROLO[3,4-*b*]PYRROL-4(1*H*)-ONES FROM 2-[ARYL(AZIDO)METHYL]-1*H*-PYRROLE-3-CARBOXYLATES VIA A CONCISE THREE-STEP SEQUENCE

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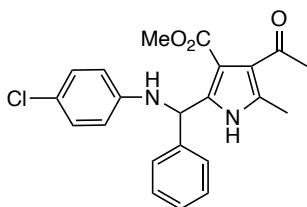
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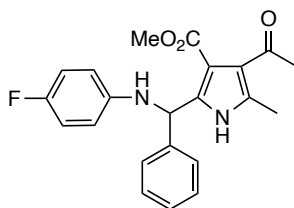
## Substitution Products for 2a–2f



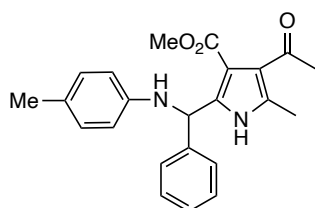
**Methyl 4-Acetyl-5-methyl-2-(phenyl(phenylamino)methyl)-1H-pyrrole-3-carboxylate.** Yellow solids; Mp 172.0–173.0 °C; IR (KBr) : 3357, 3269, 1680, 1658, 1603, 1514, 1448, 1355, 1319, 1210, 1083  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  8.85 (br, 1H), 7.26–7.37 (m, 5H) , 7.17 (dd,  $J = 7.5, 8.0$  Hz, 2H), 6.83 (t,  $J = 6.9$  Hz, 1H), 6.60 (d,  $J = 8.0$  Hz, 2H), 5.95 (s, 1H), 3.68 (s, 3H), 2.45 (s, 3H), 2.32 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.8, 164.7, 147.1, 141.1, 137.5, 131.2, 129.3, 128.9, 128.1, 127.3, 124.3, 119.2, 113.8, 110.4, 57.2, 51.0, 31.1, 12.7.



**Methyl 4-Acetyl-2-(((4-chlorophenyl)amino)(phenyl)methyl)-5-methyl-1H-pyrrole-3-carboxylate.** Pale yellow solids; Mp 153.0–154.0 °C; IR (KBr) : 3374, 3288, 1701, 1643, 1597, 1496, 1443, 1293, 1199, 1092  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  8.93 (s, 1H) , 7.25–7.35 (m, 5H), 7.10 (d,  $J = 8.6$  Hz, 2H), 6.51 (d,  $J = 8.6$  Hz, 2H), 5.93 (s, 1H), 3.68 (s, 3H), 2.41 (s, 3H), 2.28 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  196.2, 164.6, 146.8, 142.1, 136.4, 134.5, 129.9, 129.3, 129.2, 128.8, 127.6, 124.3, 116.5, 116.1, 60.7, 31.9, 31.1, 11.6.

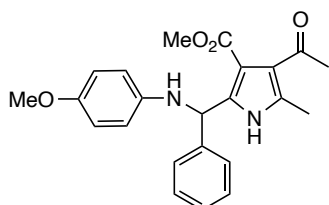


**Methyl 4-Acetyl-2-(((4-fluorophenyl)amino)(phenyl)methyl)-5-methyl-1H-pyrrole-3-carboxylate.** Yellow solids; Mp 159.0–160.0 °C; IR (KBr) : 3382, 3292, 1698, 1650, 1511, 1443, 1342, 1294, 1198, 1083  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  9.0 (s, 1H) , 7.22–7.32 (m, 5H) , 6.83 (t,  $J = 8.6$  Hz, 2H), 6.51 (dd,  $J = 4.0, 9.2$  Hz, 2H), 5.88 (s, 1H), 3.65 (s, 3H) , 2.38 (s, 3H), 2.25 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  196.3, 164.8, 156.8 ( $J = 212$  Hz), 143.4, 141.0, 134.6, 137.2, 131.1, 129.0, 128.2, 127.2, 124.4, 115.9 ( $J = 22.8$  Hz), 114.8 ( $J = 7.2$  Hz), 110.5, 57.6, 51.1, 31.1, 12.7.

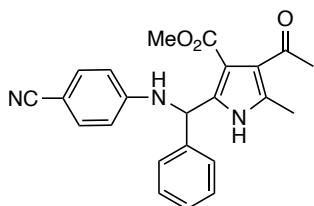


**Methyl 4-Acetyl-5-methyl-2-(phenyl(*p*-tolylamino)methyl)-1H-pyrrole-3-**

carboxylate. Yellow solids; Mp 156.0–157.0 °C; IR (KBr) : 3363, 3322, 1710, 1651, 1520, 1442, 1354, 1301, 1200, 1079  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  9.0 (s, 1H), 7.25-7.33 (m, 5H), 6.98 (d,  $J$  = 8.0 Hz, 2H), 6.50 (d,  $J$  = 8.0 Hz, 2H), 5.90 (s, 1H), 2.23 (s, 3H), 3.66 (s, 3H), 2.43 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  196.4, 164.8, 146.8, 141.7, 135.1, 135.0, 134.7, 129.3, 129.1, 129.0, 127.7, 123.9, 116.4, 61.0, 31.9, 31.1, 20.8, 11.6 .

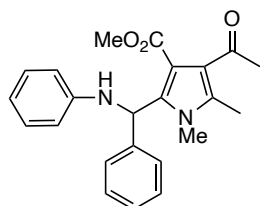


Methyl 4-Acetyl-2-(((4-methoxyphenyl)amino)(phenyl)methyl)-5-methyl-1H-pyrrole-3-carboxylate. Yellow solids; Mp 157.0–158.0 °C; IR (KBr) : 3353, 3299, 1706, 1654, 1513, 1442, 1355, 1290, 1243, 1204, 1035  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  8.98 (s, 1H), 7.26-7.35 (m, 5H), 6.76 (d,  $J$  = 8.6 Hz, 2H), 6.55 (d,  $J$  = 8.6 Hz, 2H), 5.86 (s, 1H), 3.74 (s, 3H), 3.69 (s, 3H), 2.44 (s, 3H), 2.32 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.9, 164.9, 153.3, 141.5, 141.4, 137.9, 131.2, 129.1, 129.0, 128.2, 127.4, 124.5, 115.2, 115.0, 110.4, 58.1, 55.8, 51.2, 31.7, 31.3, 22.8, 12.9.



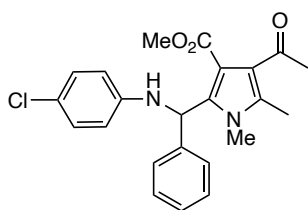
Methyl 4-Acetyl-2-(((4-cyanophenyl)amino)(phenyl)methyl)-5-methyl-1H-pyrrole-3-carboxylate. Yellow solids; Mp 190.0–191.0 °C; IR (KBr) : 3377, 3245, 2224, 1687, 1651, 1605, 1526, 1439, 1343, 1211, 1179, 1096  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  8.43 (s, 1H), 7.44 (d,  $J$  = 8.6 Hz, 2H), 7.28-7.39 (m, 5H), 6.60 (d,  $J$  = 8.6 Hz, 2H), 6.08 (s, 1H), 3.71 (s, 3H), 2.44 (s, 3H), 2.30 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.9, 165.0, 150.3, 139.8, 135.9, 133.9, 131.9, 129.3, 128.6, 127.2, 124.2, 120.2, 113.5, 111.5, 100.6, 55.6, 51.5.

### Methylation products for 2a–2f

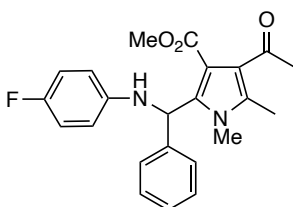


Methyl 4-Acetyl-1,5-dimethyl-2-(phenyl(phenylamino)methyl)-1H-pyrrole-3-carboxylate. Pale yellow solid; Mp 146.0–147.0 °C; IR (KBr) : 3352, 1704, 1667, 1602, 1537, 1496, 1435, 1410, 1206, 1186, 1169  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.20-7.32 (m, 5H), 7.17 (dd,  $J$  = 7.5, 8.6 Hz, 2H), 6.75 (t,  $J$  = 7.5 Hz, 2H), 6.66 (d,  $J$  = 8.0 Hz, 1H), 6.29 (d,  $J$  = 5.7 Hz, 1H), 4.57 (d,  $J$  = 6.0 Hz, 1H), 3.41 (s, 3H), 3.67 (s, 3H), 2.35 (s, 3H),

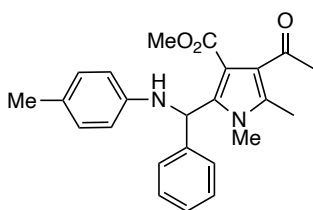
2.30 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ )  $\delta$  198.0, 166.1, 147.1, 139.8, 136.0, 133.4, 129.3, 128.7, 127.4, 126.9, 122.9, 118.4, 113.5, 113.1, 53.6, 51.5, 31.7, 30.8, 11.1.



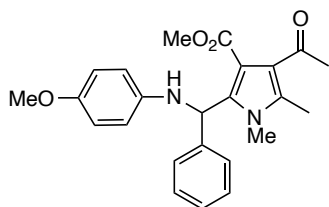
**Methyl 4-Acetyl-2-(((4-chlorophenyl)amino)(phenyl)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** Pale yellow solid; Mp 203.0–204.0 °C; IR (KBr) : 3366, 1706, 1654, 1596, 1534, 1491, 1447, 1363, 1315, 1164, 1027  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.10 (d,  $J$  = 8.0 Hz, 2H), 7.26-7.34 (m, 5H), 6.58 (d,  $J$  = 8.0 Hz, 2H), 6.24 (d,  $J$  = 5.7 Hz, 1H), 4.66 (d,  $J$  = 5.7 Hz, 1H), 3.38 (s, 3H), 3.67 (s, 3H), 2.34 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.0, 166.1, 145.6, 139.4, 135.6, 133.4, 129.1, 128.8, 127.6, 126.9, 123.0, 114.6, 113.1, 53.8, 51.6, 31.7, 30.9, 11.1.



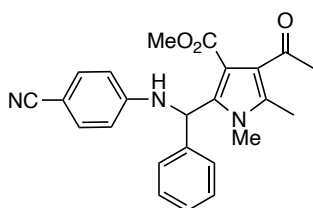
**Methyl 4-Acetyl-2-(((4-fluorophenyl)amino)(phenyl)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** Yellow solid; Mp 170.0–171.0 °C; IR (KBr) : 3367, 1707, 1649, 1507, 1441, 1411, 1304, 1211, 1163, 1026  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.28-7.33 (m, 5H), 6.87 (t,  $J$  = 8.6 Hz, 2H), 6.59 (dd,  $J$  = 4.6, 9.2 Hz, 2H), 6.21 (d,  $J$  = 5.7 Hz, 1H), 4.49 (d,  $J$  = 5.7 Hz, 1H), 3.68 (s, 3H), 3.39 (s, 3H), 2.35 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.0, 166.1, 156.3 ( $J$  = 236 Hz), 143.4, 139.6, 135.8, 133.4, 128.7, 127.5, 126.9, 122.9, 115.7 ( $J$  = 21.5 Hz), 114.5 ( $J$  = 7.2 Hz), 54.3, 51.6, 31.7, 30.9, 11.1.



**Methyl 4-Acetyl-1,5-dimethyl-2-(phenyl(*p*-tolylamino)methyl)-1H-pyrrole-3-carboxylate.** Pale yellow solid; Mp 202.0–203.0 °C; IR (KBr) : 3363, 1704, 1655, 1615, 1518, 1446, 1298, 1195, 1164, 1030  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.25-7.32 (m, 5H), 6.97 (d,  $J$  = 8.6 Hz, 2H), 6.57 (d,  $J$  = 8.6 Hz, 2H), 6.25 (s, 1H), 3.66 (s, 3H), 3.39 (s, 3H), 2.34 (s, 3H), 2.29 (s, 3H), 2.22 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.0, 166.1, 144.8, 139.9, 136.1, 133.4, 129.8, 128.6, 127.3, 126.9, 122.8, 113.6, 113.0, 53.9, 51.5, 31.7, 30.8, 20.4, 11.1.

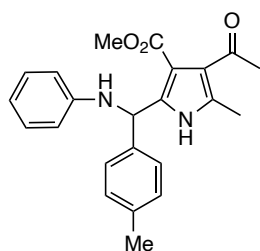


**Methyl 4-Acetyl-2-(((4-methoxyphenyl)amino)(phenyl)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** White solid; Mp 144.0–145.0 °C; IR (KBr) : 3378, 1709, 1649, 1511, 1438, 1347, 1293, 1169, 1123, 1038  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.30-7.34 (m, 5H), 6.75 (d,  $J = 8.0$  Hz, 2H), 6.63 (d,  $J = 8.0$  Hz, 2H), 6.21 (s, 1H), 3.73 (s, 3H), 3.69 (s, 3H), 3.40 (s, 3H), 2.35 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.2, 166.3, 152.8, 141.4, 140.2, 136.4, 133.6, 128.8, 127.5, 127.1, 123.0, 115.1, 115.0, 113.1, 55.8, 54.7, 51.7, 31.9, 31.0, 11.3.

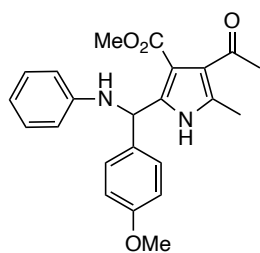


**Methyl 4-Acetyl-2-(((4-cyanophenyl)amino)(phenyl)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** Pale yellow solid; Mp 188.0–189.0 °C; IR (KBr) : 3340, 2214, 1701, 1656, 1605, 1525, 1344, 1210, 1171  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.24-7.43 (m, 7H), 2.29 (s, 3H), 6.67 (d,  $J = 8.6$  Hz, 2H), 6.33 (d,  $J = 6.9$  Hz, 1H), 5.38 (d,  $J = 6.9$  Hz, 1H), 3.69 (s, 3H), 3.39 (s, 3H), 2.35 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  197.9, 166.1, 150.2, 138.5, 134.8, 133.8, 133.3, 128.9, 127.9, 126.7, 123.2, 120.1, 113.1, 112.9, 100.0, 52.8, 51.7, 31.7, 30.9, 11.1.

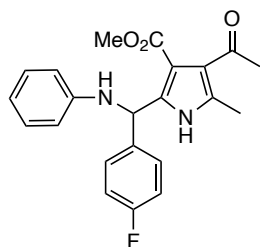
### Substitution Products for 3a–3h



**Methyl 4-Acetyl-5-methyl-2-((phenylamino)(*p*-tolyl)methyl)-1H-pyrrole-3-carboxylate.** Yellow liquid; IR (KBr) : 3346, 3281, 1707, 1603, 1525, 1514, 1081  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  8.99 (s, 1H), 7.18-7.13 (m, 6H), 6.80 (t,  $J = 7.4$  Hz, 1H), 6.58 (dd,  $J = 8.6, 1.1$  Hz, 2H), 5.91 (s, 1H), 4.07 (s, 1H), 3.68 (s, 3H), 2.43 (s, 3H), 2.33 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  199.0, 164.8, 147.3, 138.4, 138.0, 137.9, 131.2, 129.7, 129.5, 127.3, 124.5, 119.3, 113.9, 110.3, 57.1, 51.2, 31.2, 21.2, 12.8.

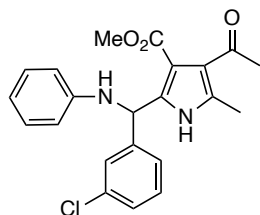


**Methyl 4-Acetyl-2-((4-methoxyphenyl)(phenylamino)methyl)-5-methyl-1H-pyrrole-3-carboxylate.** Yellow liquid; IR (KBr) : 3381, 1710, 1604, 1511, 1416, 1250, 1176, 1082  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  8.93 (s, 1H), 7.16-7.19 (m, 4H), 6.86 (dd,  $J = 6.9, 2.3$  Hz, 2H), 6.81 (t,  $J = 7.2$  Hz, 1H), 6.59 (d,  $J = 7.4$ , 2H), 5.89 (s, 1H), 4.04 (s, 1H), 3.79 (s, 3H), 3.69 (s, 1H), 2.44 (s, 3H), 2.30 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.9, 164.8, 159.4, 147.3, 138.0, 133.5, 131.1, 129.5, 128.6, 124.5, 119.3, 114.4, 113.9, 110.2, 56.9, 55.4, 51.2, 31.3, 12.8.

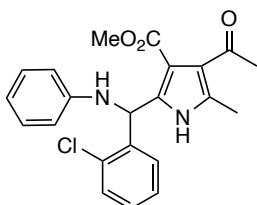


**Methyl 4-Acetyl-2-((4-fluorophenyl)(phenylamino)methyl)-5-methyl-1H-pyrrole-3-carboxylate.** Yellow solids; Mp 173.5–1174.5  $^\circ\text{C}$ ; IR (KBr) : 3358, 3284, 3016, 2947, 1604, 1437, 1353, 1320, 1229, 1207, 1183, 1162, 1084  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  8.88 (s, 1H), 7.24-7.26 (m, 2H), 7.19 (t,  $J = 7.7$  Hz, 2H), 7.05-7.01 (m, 2H), 6.83 (t,  $J = 7.4$  Hz, 1H), 6.59 (dd,  $J = 8.6, 1.1$  Hz, 2H), 5.94 (s, 1H), 4.02 (s, 1H), 3.69 (s, 3H), 2.44 (s, 3H), 2.31 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.9, 164.7, 162.5 ( $J = 247$  Hz), 147.0, 141.8, 137.4, 137.1, 131.1, 129.6, 129.2 ( $J = 7.2$  Hz), 124.6, 119.6, 116.0 ( $J = 21.6$ Hz), 114.0, 110.4, 56.8, 51.2, 31.3, 12.8.

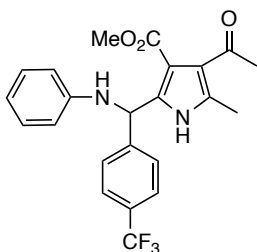
**Methyl 4-Acetyl-2-((4-chlorophenyl)(phenylamino)methyl)-5-methyl-1H-pyrrole-3-carboxylate.** Pale yellow solids; Mp 64.5–64.0  $^\circ\text{C}$ ; IR (KBr) : 3370, 3250, 3021, 2949, 2368, 2352, 1709, 1603, 1490, 1438, 1205  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  9.12 (s, 1H), 7.30 (dd,  $J = 6.6, 2.0$  Hz, 2H), 7.23-7.21 (m, 2H), 7.17 (t,  $J = 7.7$  Hz, 2H), 6.82 (d,  $J = 7.4$  Hz, 1H), 6.58 (d,  $J = 7.4$  Hz, 2H), 5.95 (d,  $J = 1.7$  Hz, 1H), 4.13 (d,  $J = 1.4$  Hz, 1H), 3.68 (s, 3H), 2.42 (s, 3H), 2.27 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  199.0, 164.8, 147.0, 139.7, 137.2, 134.0, 131.5, 129.5, 129.2, 128.8, 124.4, 119.5, 114.0, 110.6, 56.5, 51.3, 31.3, 12.8.



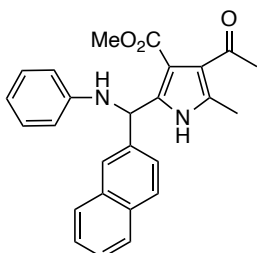
**Methyl 4-Acetyl-2-((3-chlorophenyl)(phenylamino)methyl)-5-methyl-1H-pyrrole-3-carboxylate.** White solids; Mp 62.0–63.0 °C; IR (KBr) : 3370, 3263, 2951, 2359, 2348, 1714, 1603, 1504, 1435, 1414, 1207, 1081 cm<sup>-1</sup>; <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) : δ 8.93 (s, 1H), 7.29-7.26 (m, 3H), 7.20-7.17 (m, 3H, Ph), 6.83 (t, *J* = 7.7 Hz, 1H), 6.59 (d, *J* = 8.6 Hz, 2H), 5.94 (s, 1H), 4.07 (s, 1H), 3.70 (s, 3H), 2.44 (s, 3H), 2.30 (s, 3H); <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>) : δ 198.9, 164.6, 146.9, 143.1, 136.7, 134.9, 131.4, 130.5, 130.4, 129.7, 129.6, 128.5, 127.6, 127.5, 125.8, 125.7, 124.6, 119.6, 114.1, 114.0, 57.0, 56.9, 51.4, 51.3, 31.3, 12.8.



**Methyl 4-Acetyl-2-((2-chlorophenyl)(phenylamino)methyl)-5-methyl-1H-pyrrole-3-carboxylate.** Dark yellow liquid; IR (KBr) : 3410, 3353, 3283, 3235, 3010, 2950, 1669, 1659, 1636, 1444, 1182 cm<sup>-1</sup>; <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) : δ 9.12 (s, 1H), 7.43 (dd, *J* = 7.7, 1.4 Hz, 1H), 7.23-7.26 (m, 1H), 7.20-7.16 (m, 3H), 6.94 (dd, *J* = 8.0, 1.7 Hz, 1H), 6.82 (t, *J* = 7.4 Hz, 1H), 6.61 (d, *J* = 8.0 Hz, 2H), 6.29 (s, 1H), 4.03 (s, 1H), 3.58 (s, 3H), 2.46 (s, 3H), 2.31 (s, 3H); <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>) : δ 198.9, 164.6, 147.4, 138.9, 136.4, 133.9, 131.6, 130.0, 129.6, 129.5, 128.6, 127.5, 124.7, 119.5, 113.8, 110.8, 54.9, 51.3, 31.3, 12.9.

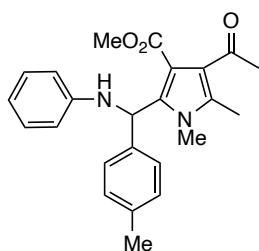


**Methyl 4-Acetyl-5-methyl-2-((phenylamino)(4-(trifluoromethyl)phenyl)methyl)-1H-pyrrole-3-carboxylate.** Yellow liquid; IR (KBr) : 3366, 3249, 1711, 1604, 1416, 1326, 1124, 1068 cm<sup>-1</sup>; <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) : δ = 9.11 (s, 1H), 7.60 (d, *J* = 8.6 Hz, 2H), 7.43 (d, *J* = 8.0 Hz, 2H), 7.20-7.16 (m, 2H), 6.83 (t, *J* = 7.2 Hz, 1H), 6.60 (d, *J* = 8.0 Hz, 2H), 6.05 (s, 1H), 4.15 (s, 1H), 3.69 (s, 3H), 2.42 (s, 3H), 2.28 (s, 3H); <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>) : δ 199.0, 164.7, 146.8, 145.0, 136.7, 131.5, 130.6, 130.3, 129.6, 127.9, 126.1, 126.0, 124.5, 119.7, 114.0, 110.8, 56.7, 51.3, 31.3, 12.8.

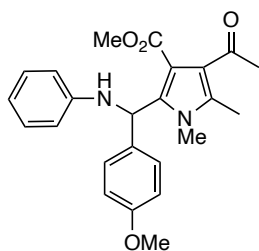


**Methyl 4-Acetyl-5-methyl-2-(naphthalen-2-yl(phenylamino)methyl)-1H-pyrrole-3-carboxylate.** Yellow liquid; IR (KBr) : 3366, 3241, 3052, 1715, 1602, 1505, 1200, 1081  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  9.07 (s, 1H), 7.83-7.77 (m, 3H), 7.70 (s, 1H), 7.47 (q,  $J$  = 3.1 Hz, 2H), 7.42-7.40 (m, 1H), 7.18 (t,  $J$  = 7.7 Hz, 1H), 6.81 (t,  $J$  = 7.2 Hz, 1H), 6.62 (d,  $J$  = 8.0 Hz, 2H), 6.14 (s, 1H), 4.21 (s, 1H), 3.64 (s, 3H), 2.42 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  199.0, 164.8, 147.3, 138.5, 137.4, 133.5, 133.1, 131.5, 129.5, 129.0, 128.2, 127.8, 126.6, 126.5, 126.2, 125.4, 124.5, 119.3, 114.0, 110.7, 57.4, 51.2, 31.3, 12.8.

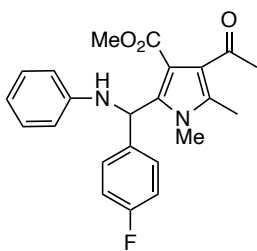
### Methylation products for 3a–3h



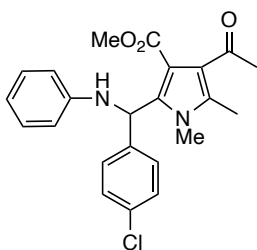
**Methyl 4-Acetyl-1,5-dimethyl-2-((phenylamino)(*p*-tolyl)methyl)-1H-pyrrole-3-carboxylate.** White solid; Mp 177.5–178.5  $^{\circ}\text{C}$ ; IR (KBr) : 3354, 3022, 2947, 2368, 2349, 1705, 1667, 1602, 1509, 1208, 1168  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.25-7.12 (m, 6H), 6.74 (d,  $J$  = 7.4 Hz, 1H), 6.65 (d,  $J$  = 8.6 Hz, 2H), 6.25 (d,  $J$  = 5.2 Hz, 1H), 4.56 (s, 1H), 3.67 (s, 3H), 3.40 (s, 3H), 2.34-2.29 (m, 6H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.1, 166.3, 147.3, 137.3, 136.9, 136.2, 133.5, 129.5, 129.4, 129.2, 127.8, 127.0, 122.9, 118.3, 113.5, 113.1, 53.5, 51.6, 31.8, 31.0, 21.1, 11.2.



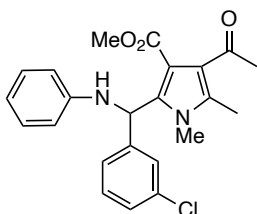
**Methyl 4-Acetyl-2-((4-methoxyphenyl)(phenylamino)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** White solid; Mp 177.5–178.5  $^{\circ}\text{C}$ ; IR (KBr) : 3400, 3103, 3055, 2999, 2952, 2901, 1603, 1247, 1166  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.26-7.23 (m, 2H), 7.16 (t,  $J$  = 7.2 Hz, 2H), 6.85 (d,  $J$  = 8.6 Hz, 2H), 6.74 (t,  $J$  = 7.4 Hz, 1H), 6.64 (d,  $J$  = 8.0 Hz, 2H), 6.20 (d,  $J$  = 4.6 Hz, 1H), 4.54 (d,  $J$  = 4.6 Hz, 1H), 3.79 (s, 3H), 3.67 (s, 3H), 3.40 (s, 3H), 2.34 (s, 3H), 2.30 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.0, 166.4, 159.0, 147.2, 136.1, 133.6, 131.9, 129.4, 128.3, 122.9, 118.3, 114.1, 113.5, 113.1, 55.4, 53.3, 51.7, 31.7, 30.9, 11.3.



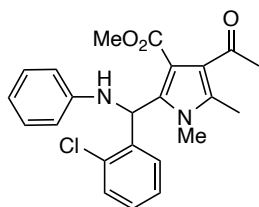
**Methyl 4-Acetyl-2-((4-fluorophenyl)(phenylamino)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** Pale yellow solid; Mp 180.0–180.5 °C; IR (KBr) : 3358, 2949, 1706, 1666, 1602, 1537, 1504, 1437, 1411, 1311, 1260, 1218, 1188, 1170  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.31 (dd,  $J = 8.2, 5.6$  Hz, 2H), 7.17 (t,  $J = 8.0$  Hz, 2H), 7.00 (t,  $J = 8.6$  Hz, 2H), 6.75 (t,  $J = 7.4$  Hz, 1H), 6.64 (d,  $J = 8.0$  Hz, 2H), 6.24 (d,  $J = 5.7$  Hz, 1H), 4.64 (d,  $J = 6.3$  Hz, 1H), 3.67 (s, 3H), 3.40 (s, 3H), 2.34 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.0, 166.2, 162.1 ( $J = 246$  Hz), 147.0, 135.9, 135.6, 133.5, 129.4, 128.8 ( $J = 7.2$  Hz), 123.0, 118.6, 115.6 ( $J = 21.6$  Hz), 113.6, 113.1, 53.1, 51.7, 31.8, 31.0, 11.2.



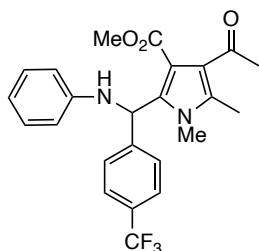
**Methyl 4-Acetyl-2-((4-chlorophenyl)(phenylamino)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** Pale yellow solid; Mp 190.0–191.0 °C; IR (KBr) : 3355, 1706, 1665, 1603, 1537, 1516, 1497, 1436, 1410, 1310, 1210, 1189, 1168  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.28-7.26 (m, 4H), 7.17 (t,  $J = 7.4$  Hz, 2H), 6.76 (t,  $J = 6.9$  Hz, 1H), 6.63 (d,  $J = 8.6$  Hz, 2H), 6.25 (d,  $J = 5.7$  Hz, 1H), 4.63 (d,  $J = 6.3$  Hz, 1H), 3.67 (s, 3H), 3.40 (s, 3H), 2.34 (s, 3H), 2.28 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.1, 166.1, 147.0, 138.5, 135.8, 133.5, 133.3, 129.5, 128.9, 128.5, 123.1, 118.7, 113.6, 113.1, 53.1, 51.7, 31.8, 31.0, 11.2.



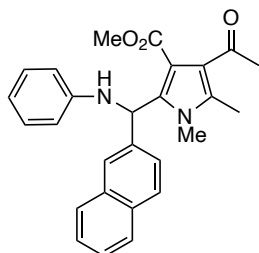
**Methyl 4-Acetyl-2-((3-chlorophenyl)(phenylamino)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** White solid; Mp 152.0–153.0 °C; IR (KBr) : 3358, 3058, 3006, 2952, 2360, 2350, 1709, 1657, 1601, 1306, 1210  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.38 (s, 1H), 7.25-7.22 (m, 3H), 7.16 (t,  $J = 7.2$  Hz, 2H), 6.76 (d,  $J = 6.9$  Hz, 1H), 6.64 (d,  $J = 8.6$  Hz, 2H), 6.28 (d,  $J = 5.7$  Hz, 1H), 4.72 (d,  $J = 5.7$  Hz, 1H), 3.68 (s, 3H), 3.39 (s, 3H), 2.34 (s, 3H), 2.26 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.2, 166.0, 147.0, 142.3, 135.6, 134.7, 133.7, 130.0, 129.5, 127.8, 127.2, 125.3, 123.1, 118.7, 113.6, 113.1, 53.2, 51.7, 31.9, 31.0, 11.



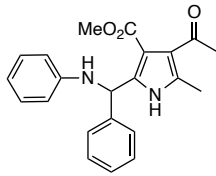
**Methyl 4-Acetyl-2-((2-chlorophenyl)(phenylamino)methyl)-1,5-dimethyl-1H-pyrrole-3-carboxylate.** White solid; Mp 132.0–133.0 °C; IR (KBr) : 3334, 2946, 2349, 1727, 1651, 1603, 1516, 1439, 1285  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.48 (t,  $J$  = 4.9 Hz, 1H), 7.37 (t,  $J$  = 4.6 Hz, 1H), 7.22 (td,  $J$  = 3.4, 1.9 Hz, 2H), 7.15 (dd,  $J$  = 8.7, 7.3 Hz, 2H), 6.74 (d,  $J$  = 7.4 Hz, 1H), 6.55 (d,  $J$  = 7.4 Hz, 2H), 6.05 (d,  $J$  = 6.9 Hz, 1H), 4.62 (d,  $J$  = 7.4 Hz, 1H), 3.51 (s, 3H), 3.46 (s, 3H), 2.42 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  195.7, 167.3, 146.4, 136.2, 134.1, 133.7, 132.7, 129.9, 129.8, 129.4, 129.2, 126.7, 121.6, 118.4, 115.0, 113.1, 51.9, 51.8, 31.0, 30.3, 11.8.



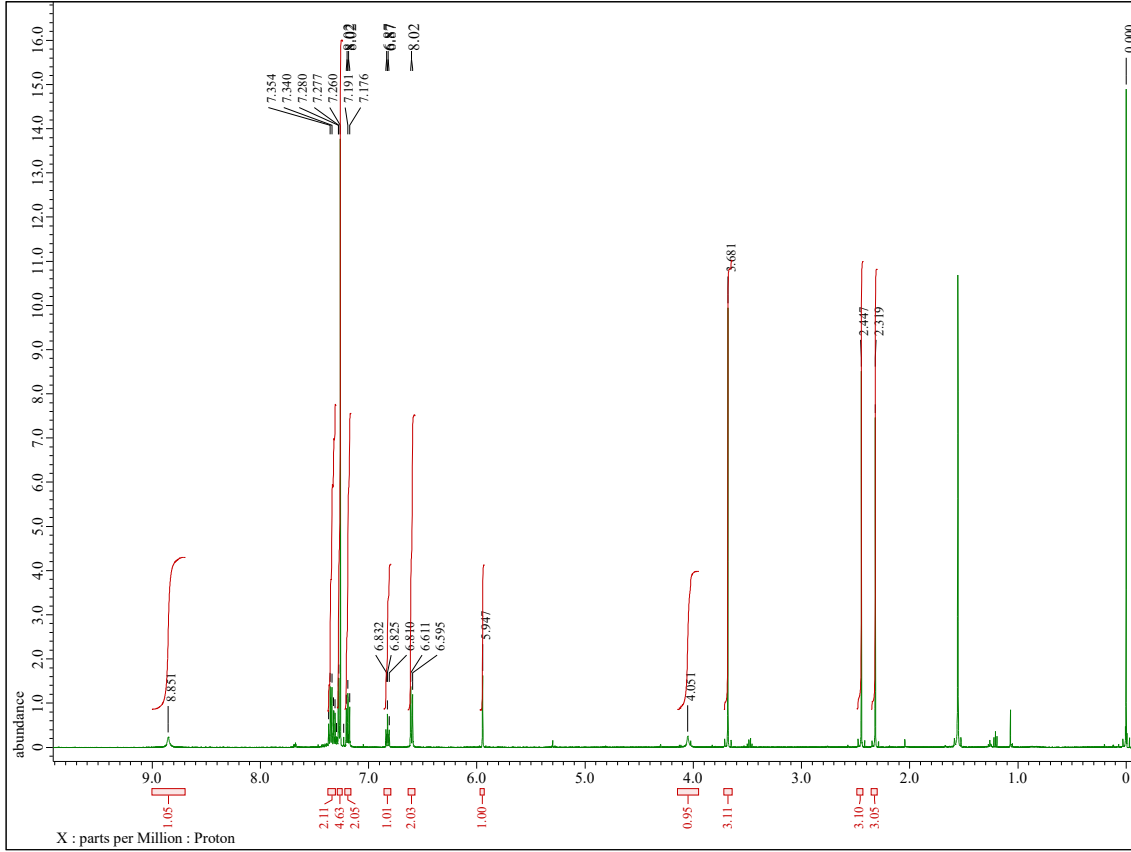
**Methyl 4-Acetyl-1,5-dimethyl-2-((phenylamino)(4-(trifluoromethyl)phenyl)methyl)-1H-pyrrole-3-carboxylate.** White solid; Mp 178.0–179.0 °C; IR (KBr) : 3361, 2947, 2368, 2350, 1711, 1663, 1604, 1327, 1119  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.57 (d,  $J$  = 8.0 Hz, 2H), 7.48 (d,  $J$  = 8.6 Hz, 2H), 7.18 (dd,  $J$  = 8.4, 7.6 Hz, 2H), 6.78 (t,  $J$  = 7.2 Hz, 1H), 6.64 (d,  $J$  = 8.0 Hz, 2H), 6.34 (d,  $J$  = 5.7 Hz, 1H), 4.71 (d,  $J$  = 6.3 Hz, 1H), 3.66 (s, 3H), 3.41 (s, 3H), 2.35 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.1, 166.0, 146.9, 144.1, 135.6, 133.5, 129.9, 129.6, 129.5, 127.5, 125.6, 125.6, 125.2, 123.3, 118.9, 113.7, 113.1, 53.3, 51.7, 31.9, 31.0, 11.2.



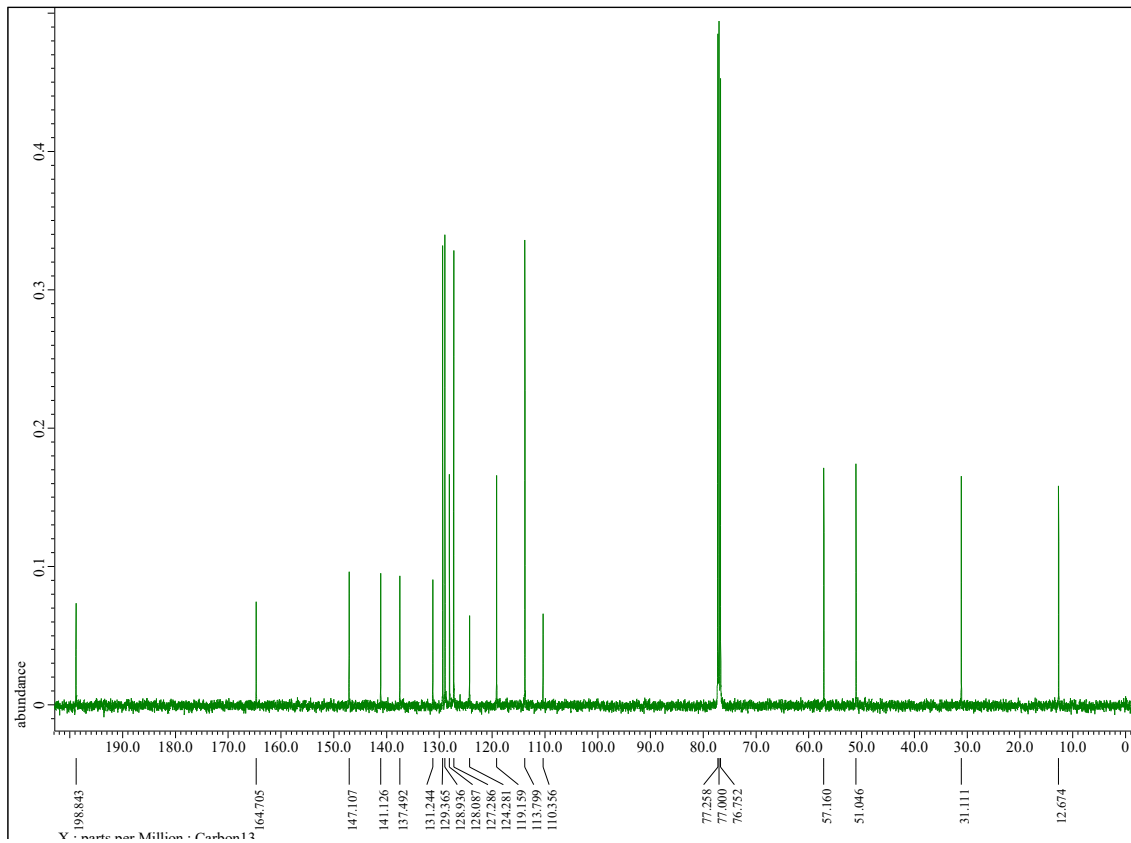
**methyl 4-acetyl-1,5-dimethyl-2-((naphthalen-2-yl)(phenylamino)methyl)-1H-pyrrole-3-carboxylate.** White solid; Mp 158.0–159.0 °C; IR (KBr) : 3361, 3053, 2952, 1712, 1653, 1601, 1534, 1515, 1437, 1411, 1301, 1206, 1172  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ ) :  $\delta$  7.80 (d,  $J$  = 13.2 Hz, 4H), 7.47–7.44 (m, 3H), 7.17 (t,  $J$  = 7.2 Hz, 2H), 6.76 (s, 1H), 6.70 (d,  $J$  = 7.4 Hz, 2H), 6.46 (d,  $J$  = 5.2 Hz, 1H), 4.67 (d,  $J$  = 5.2 Hz, 1H), 3.64 (s, 3H), 3.40 (s, 3H), 2.36 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (125 MHz,  $\text{CDCl}_3$ ) :  $\delta$  198.2, 166.2, 147.3, 137.3, 135.9, 133.7, 133.3, 132.8, 129.5, 128.7, 128.2, 127.7, 126.4, 126.2, 125.6, 125.3, 123.1, 118.5, 113.7, 113.3, 53.9, 51.7, 31.9, 31.0, 11.3.

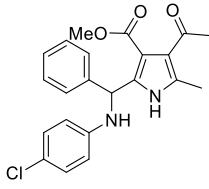


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

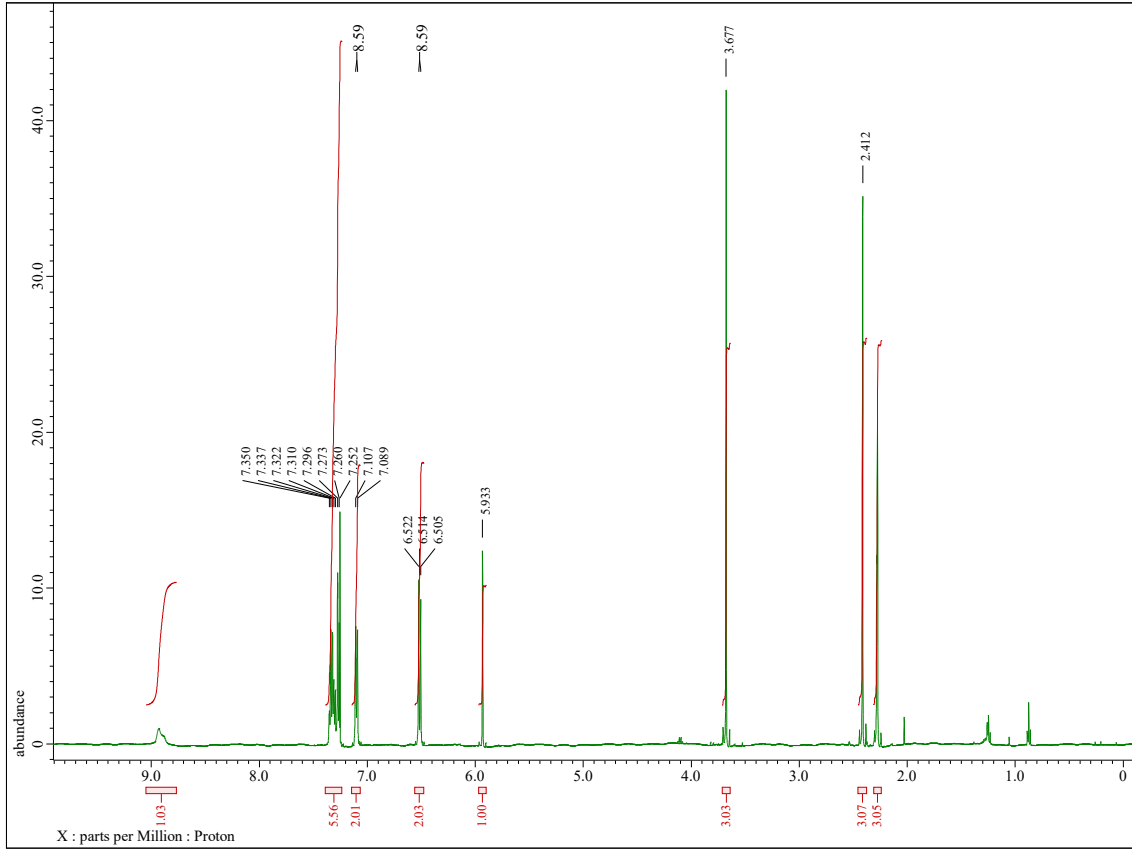


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

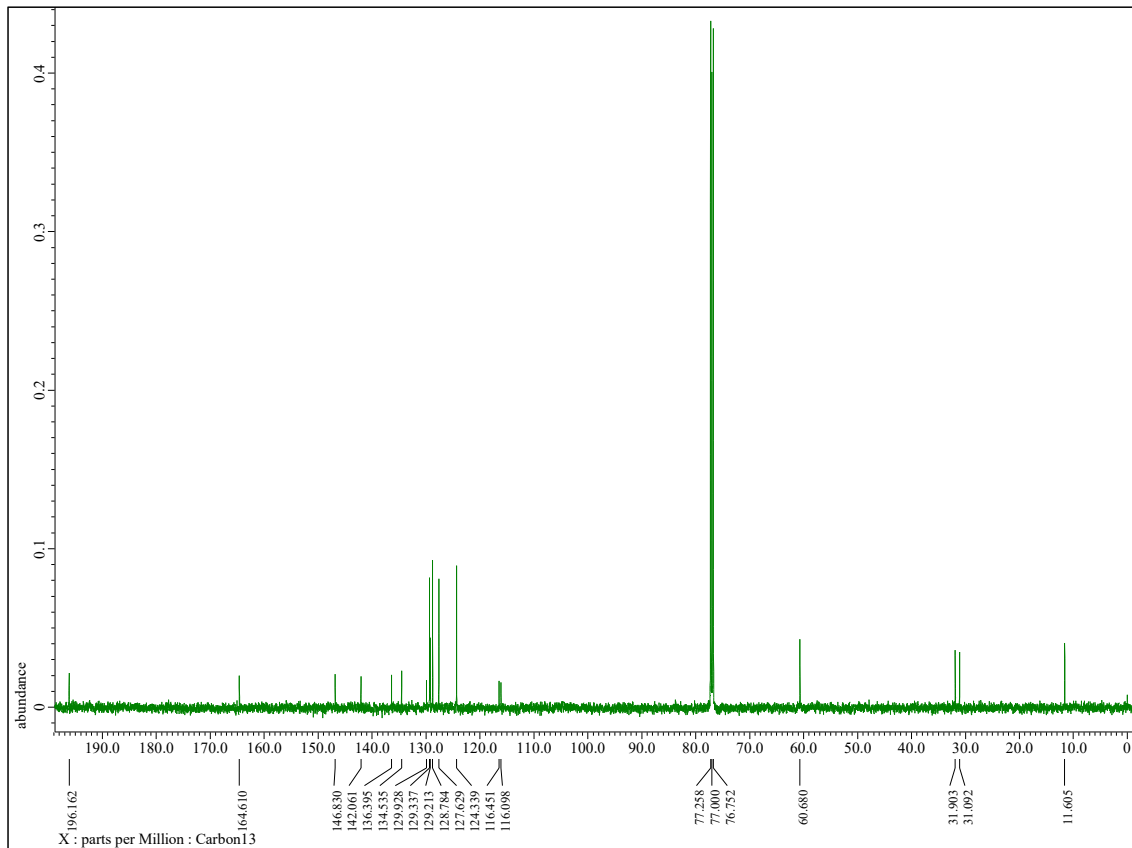


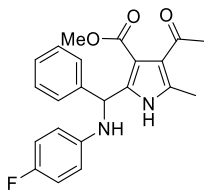


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

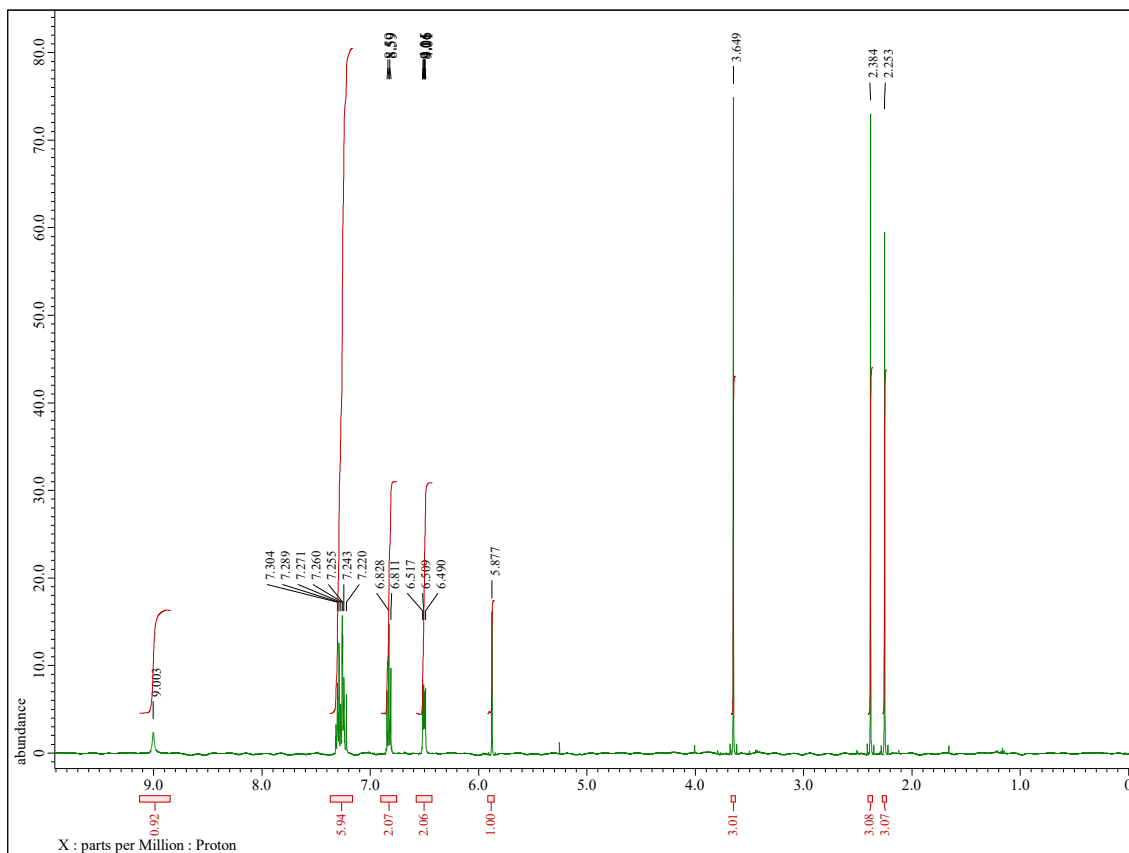


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

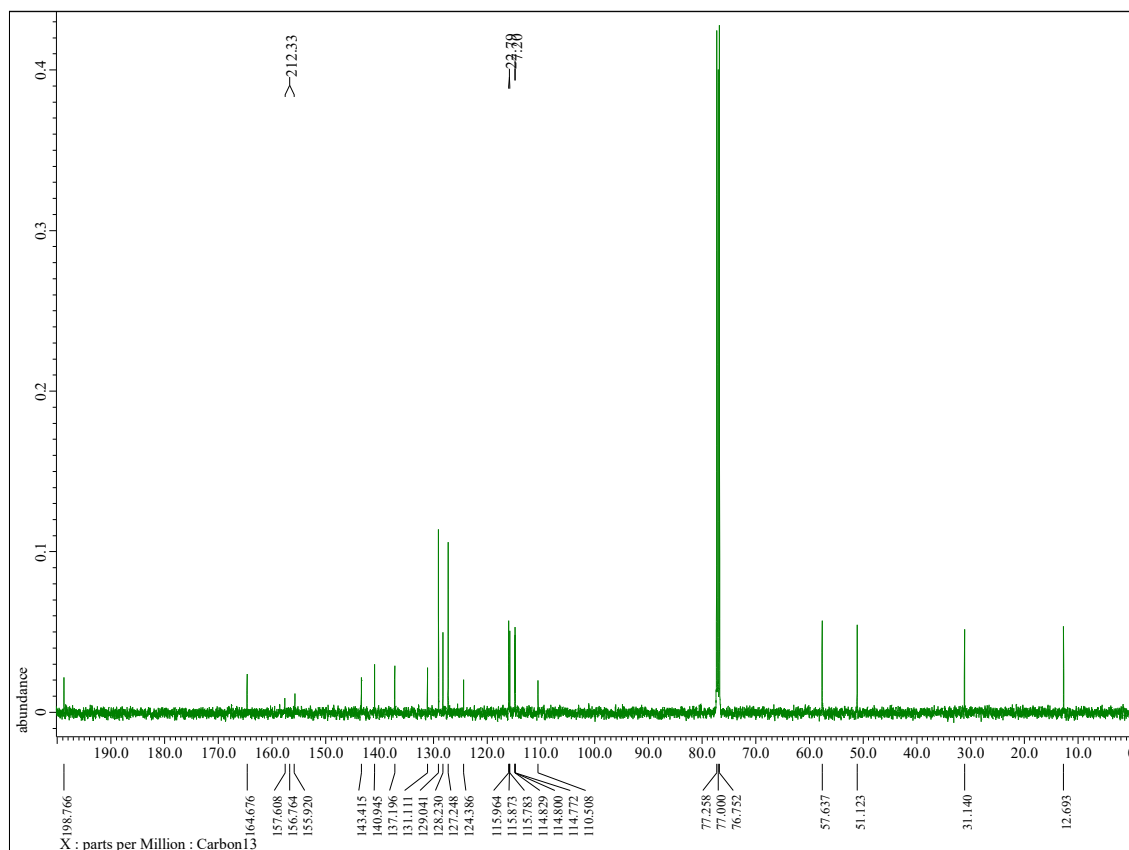


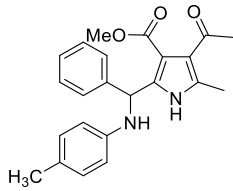


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

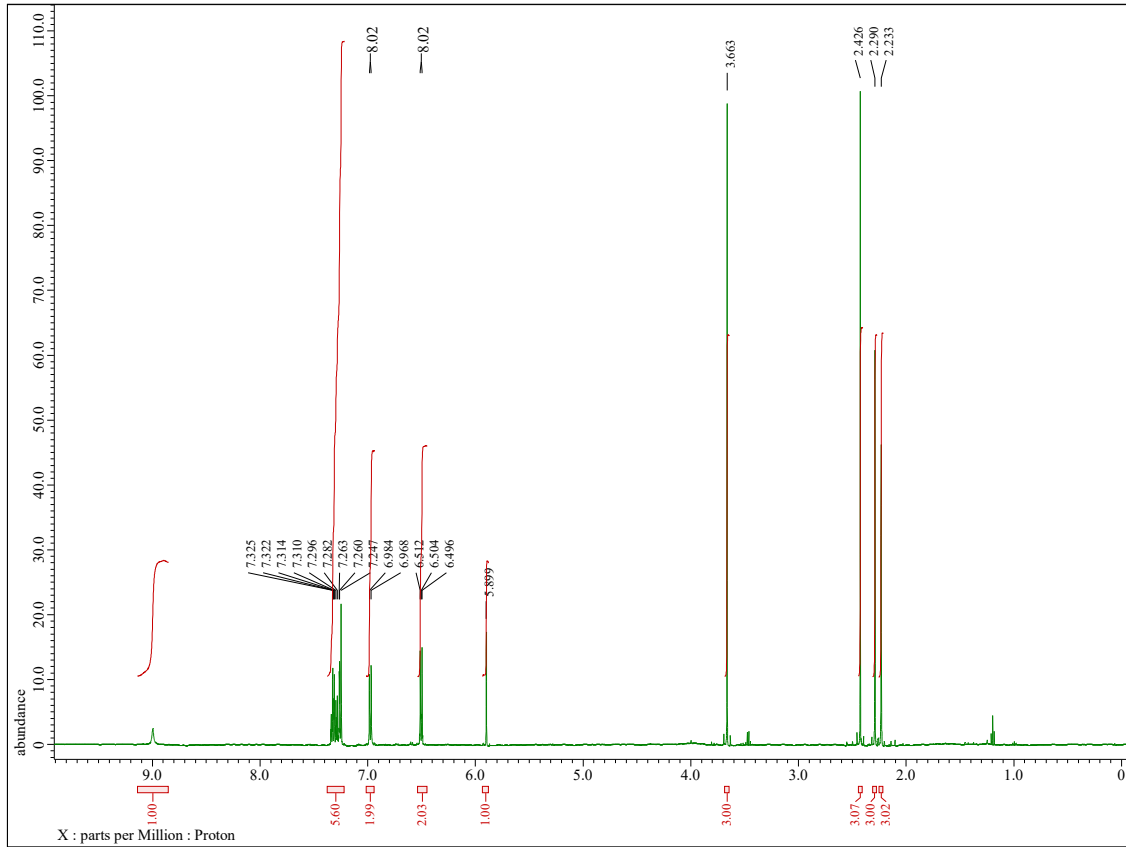


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

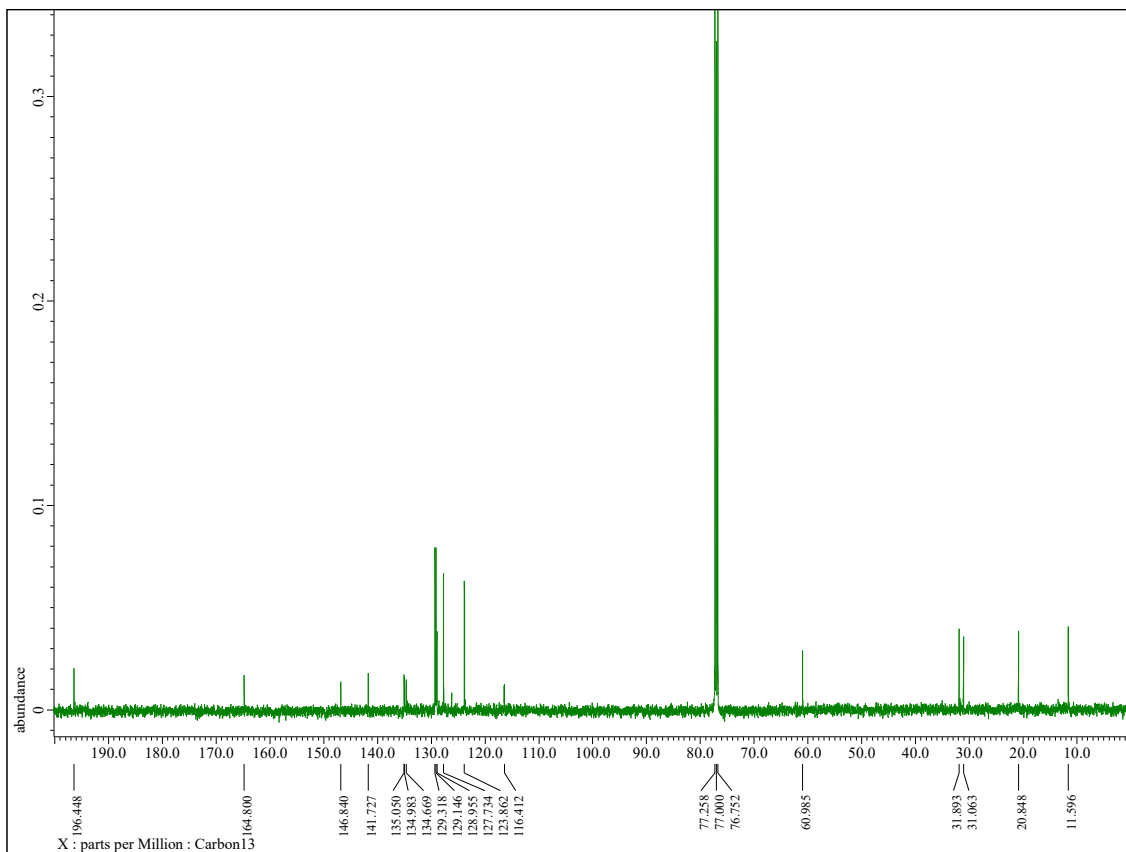


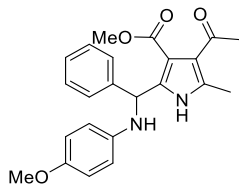


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

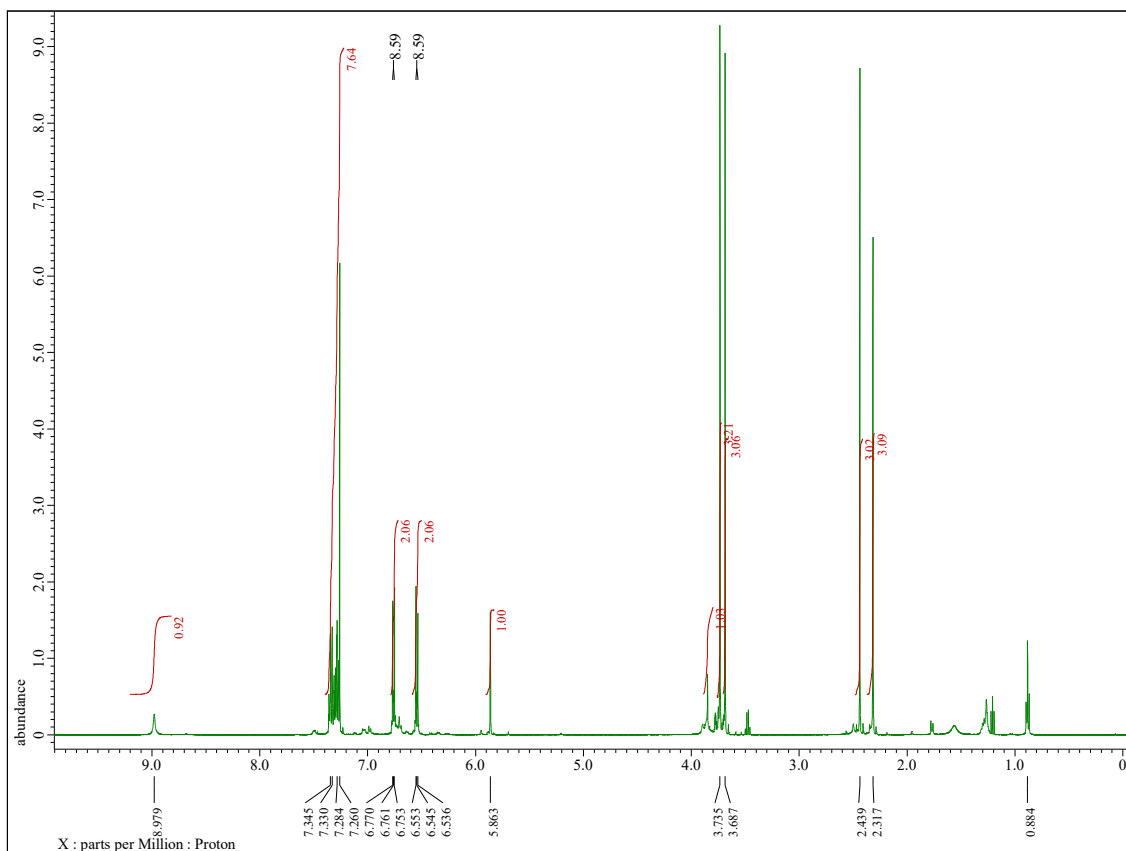


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

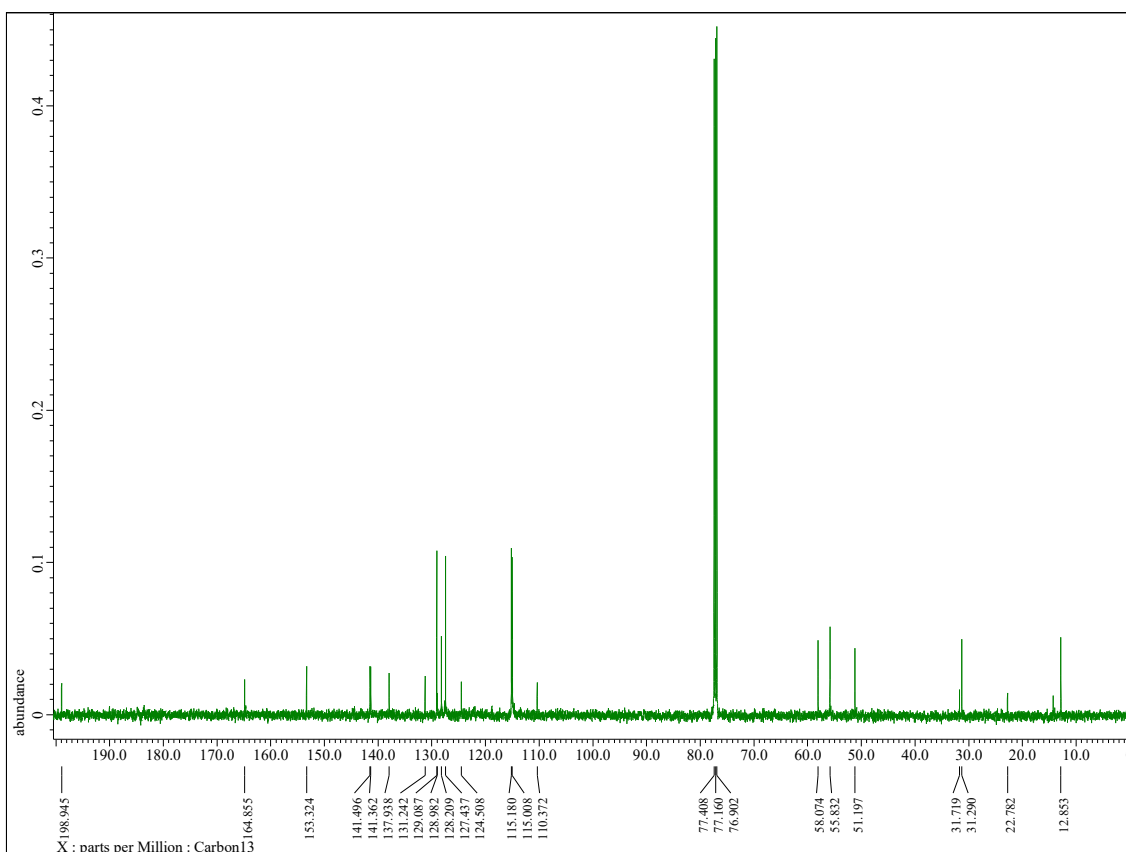


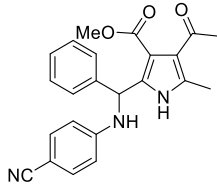


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

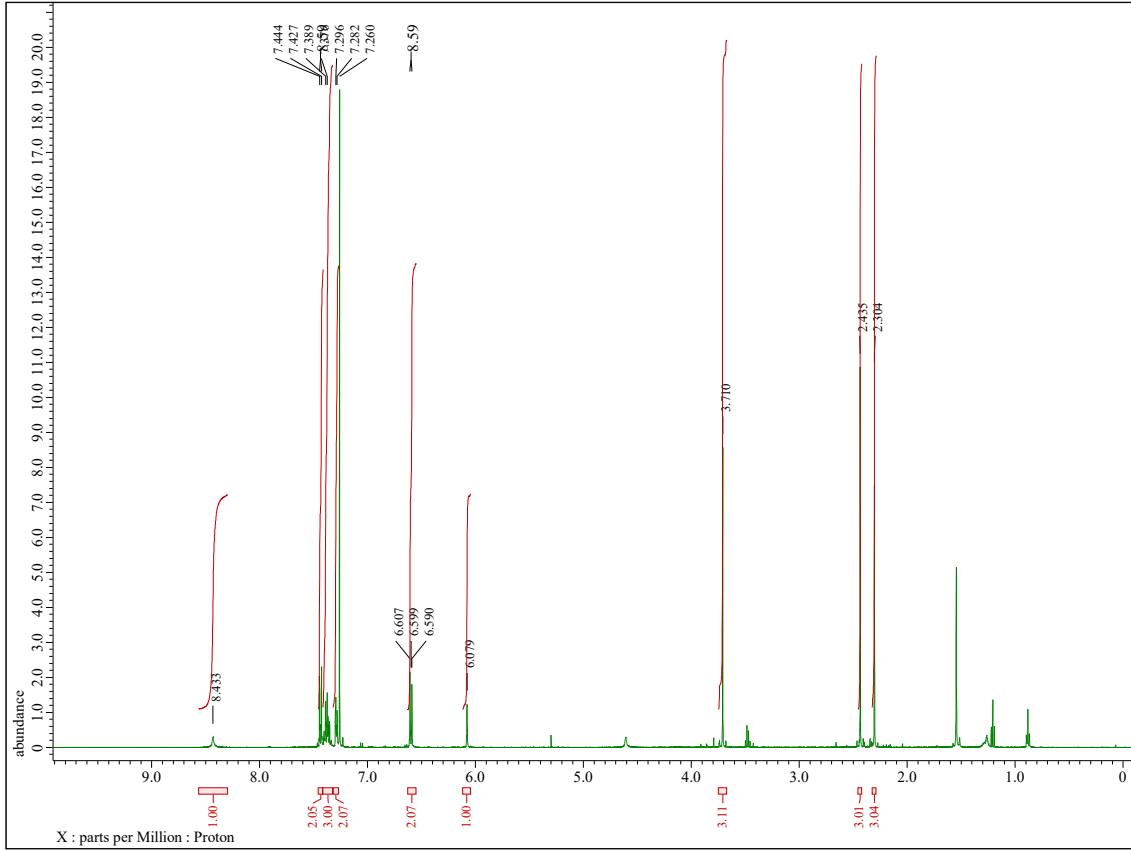


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

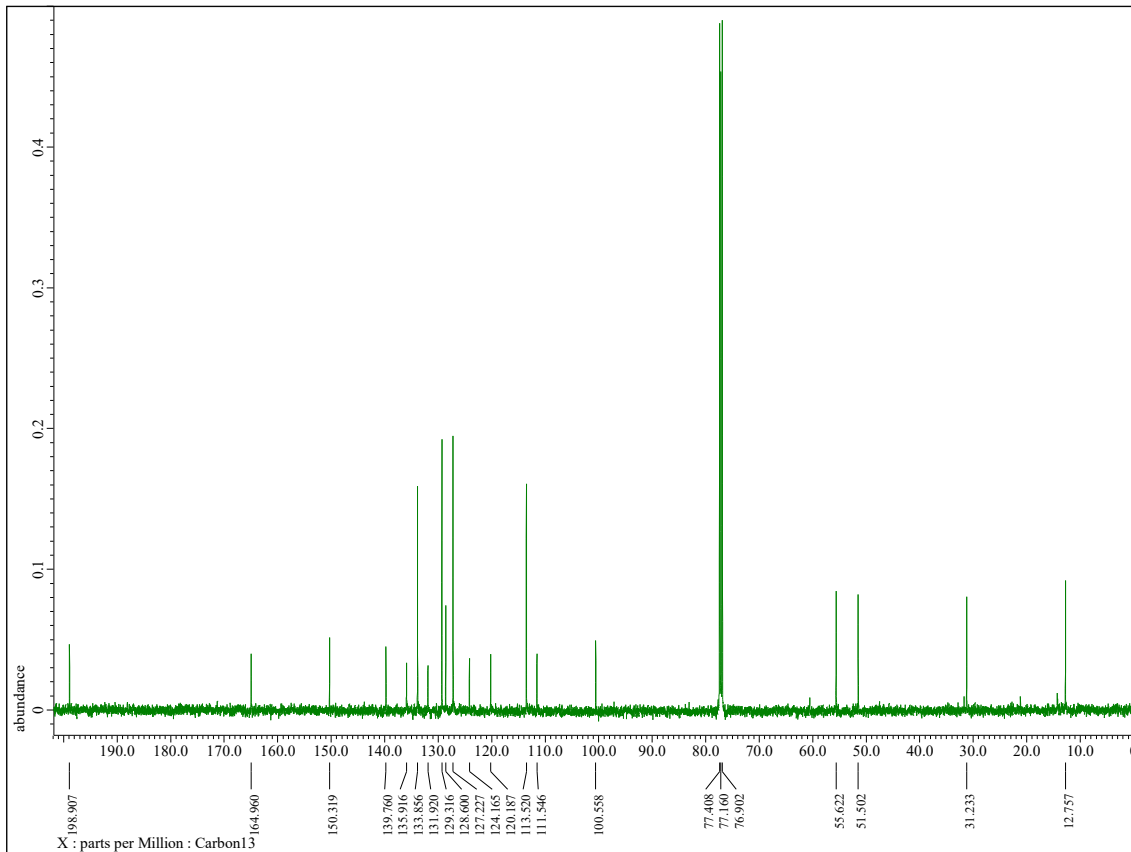


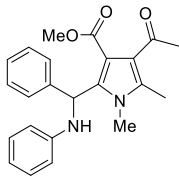


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

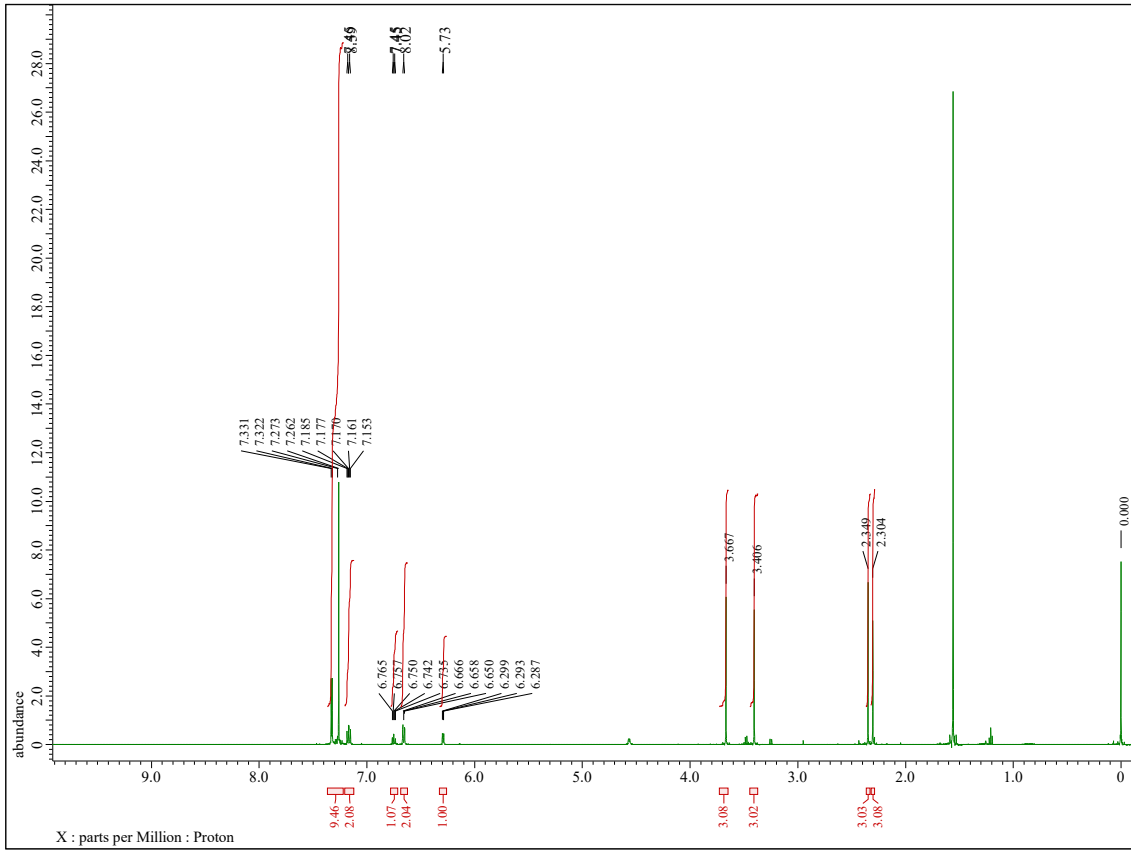


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

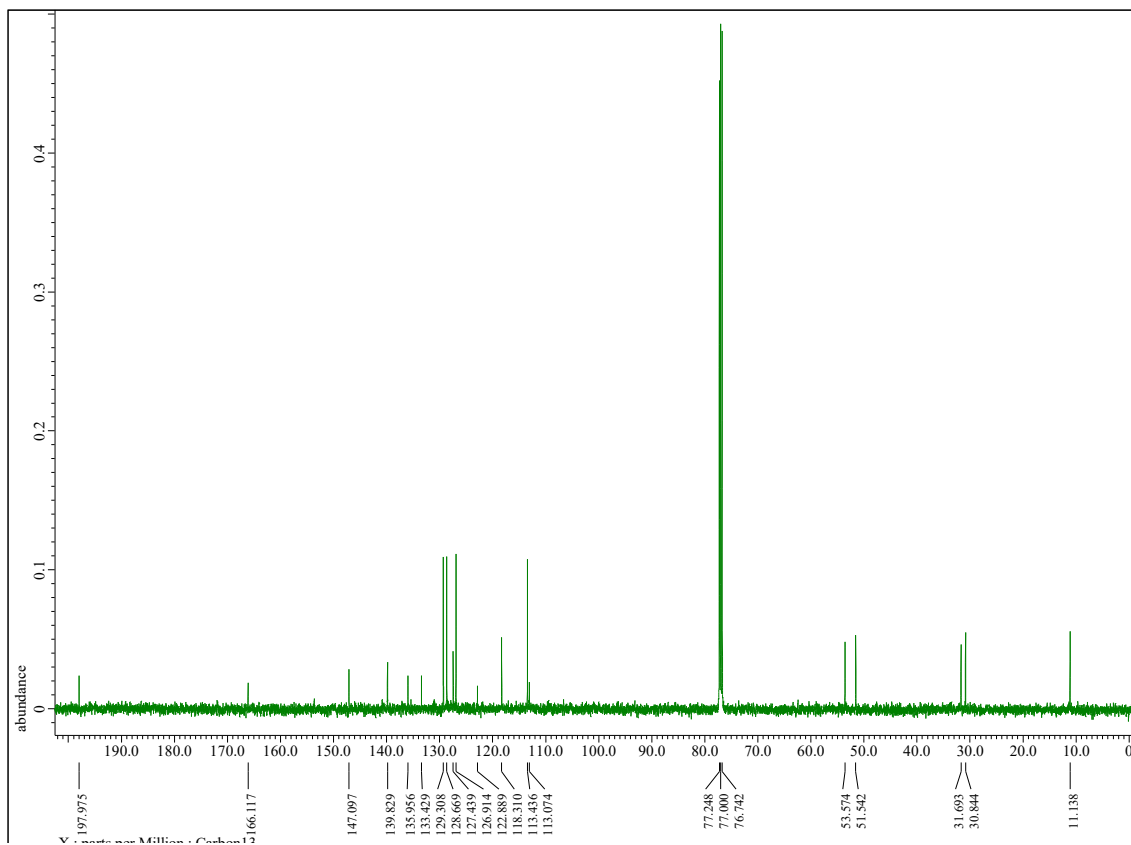


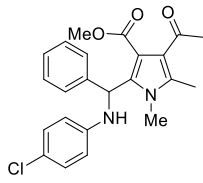


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

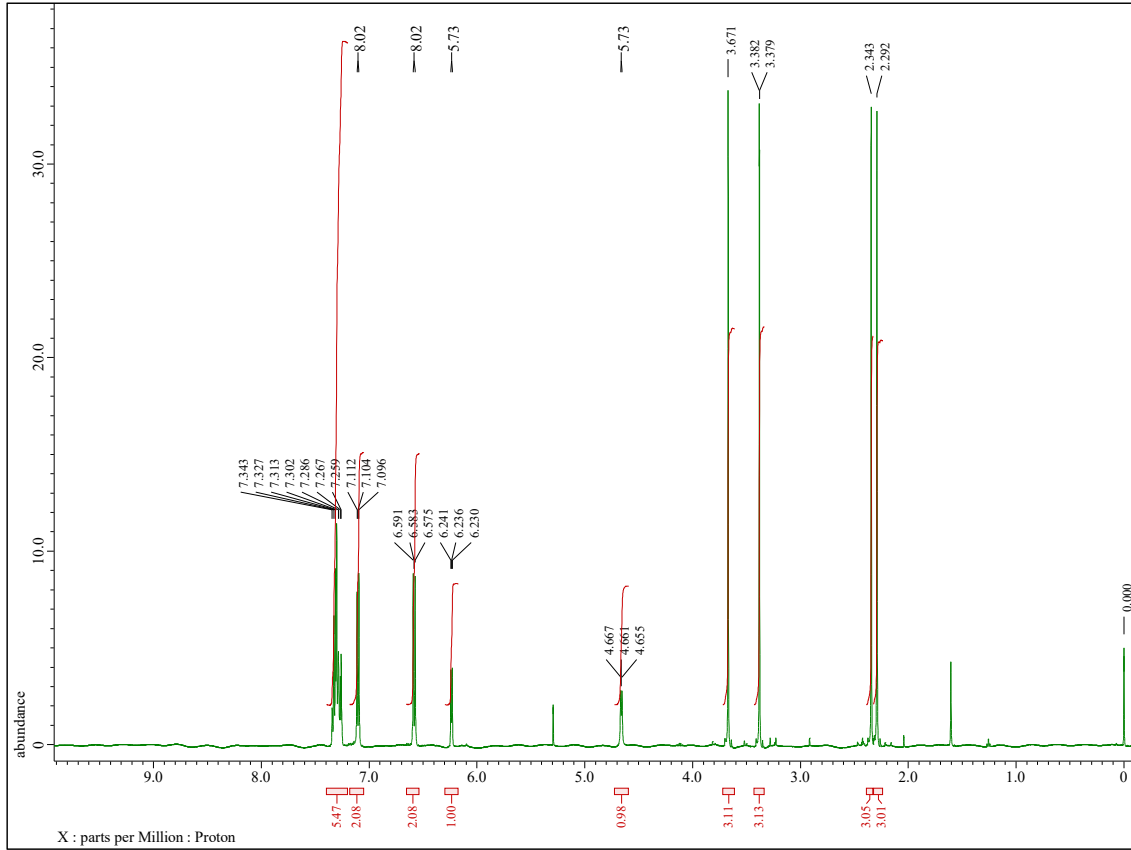


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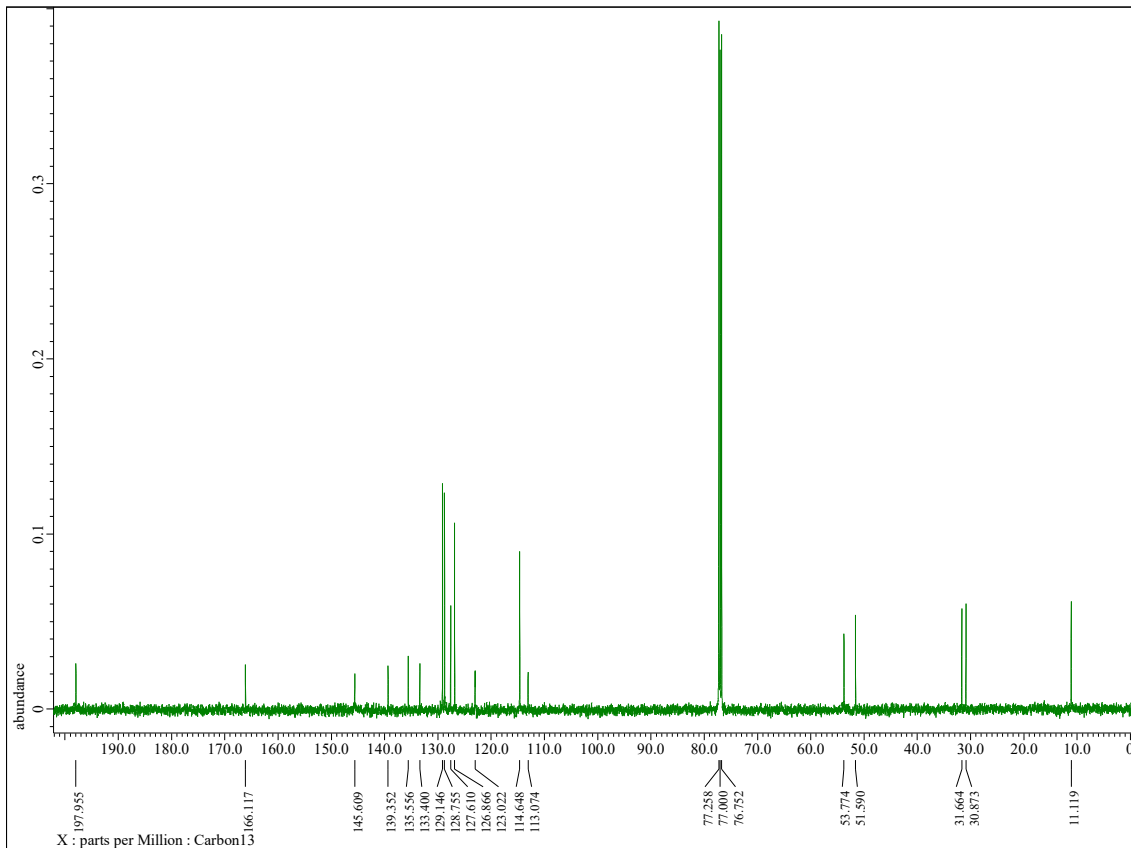


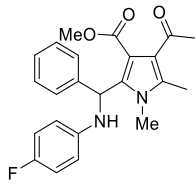


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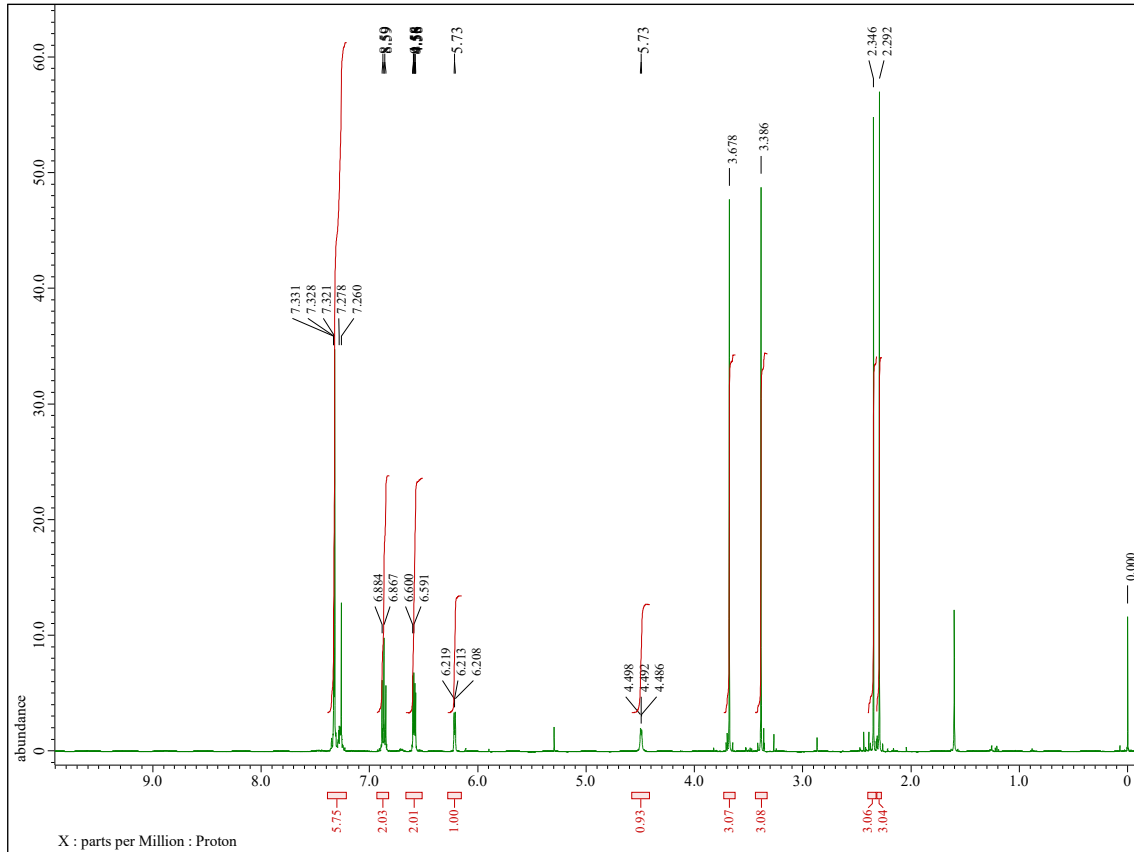


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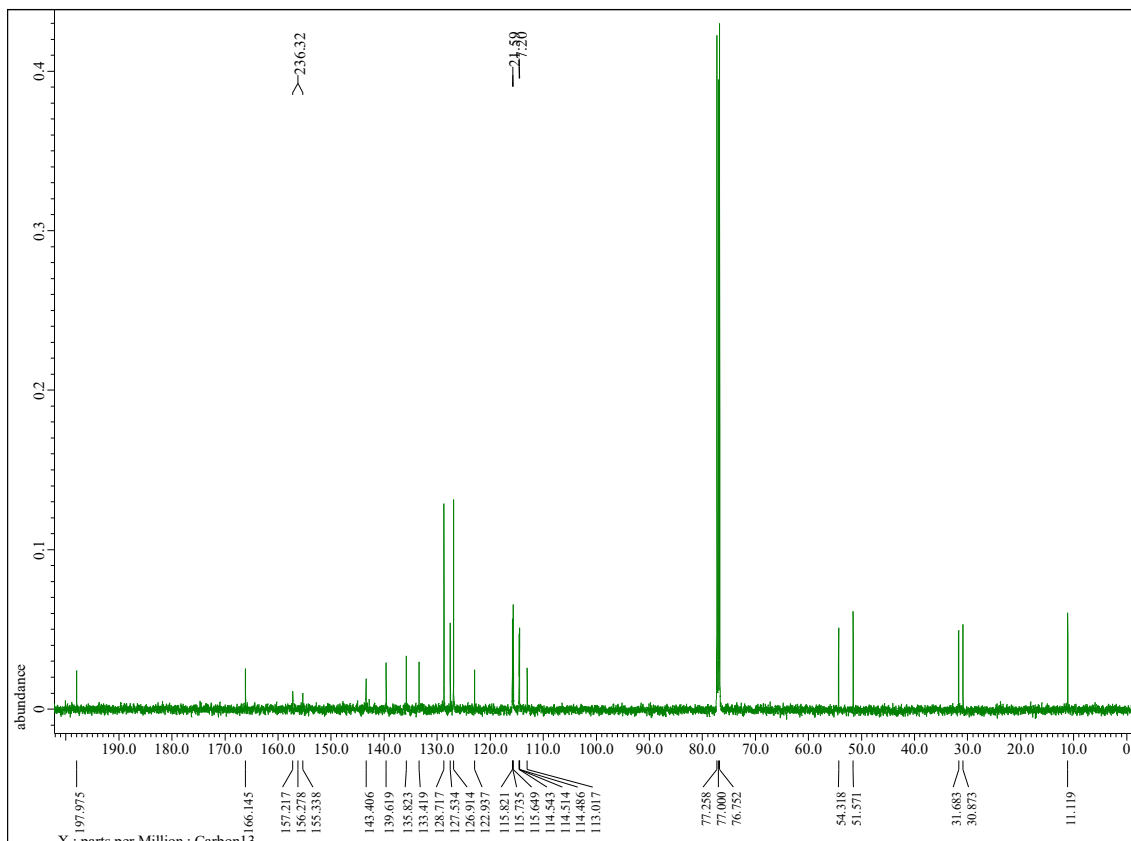


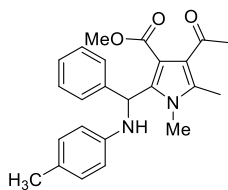


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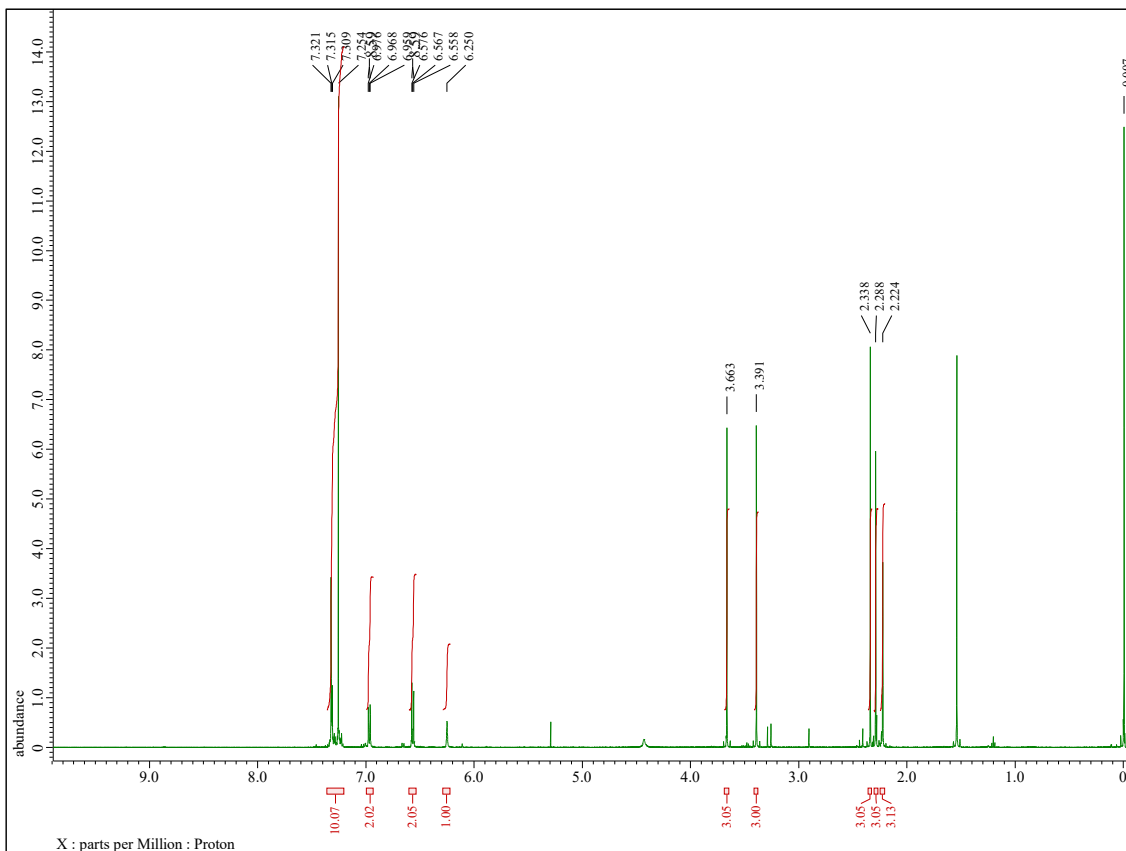


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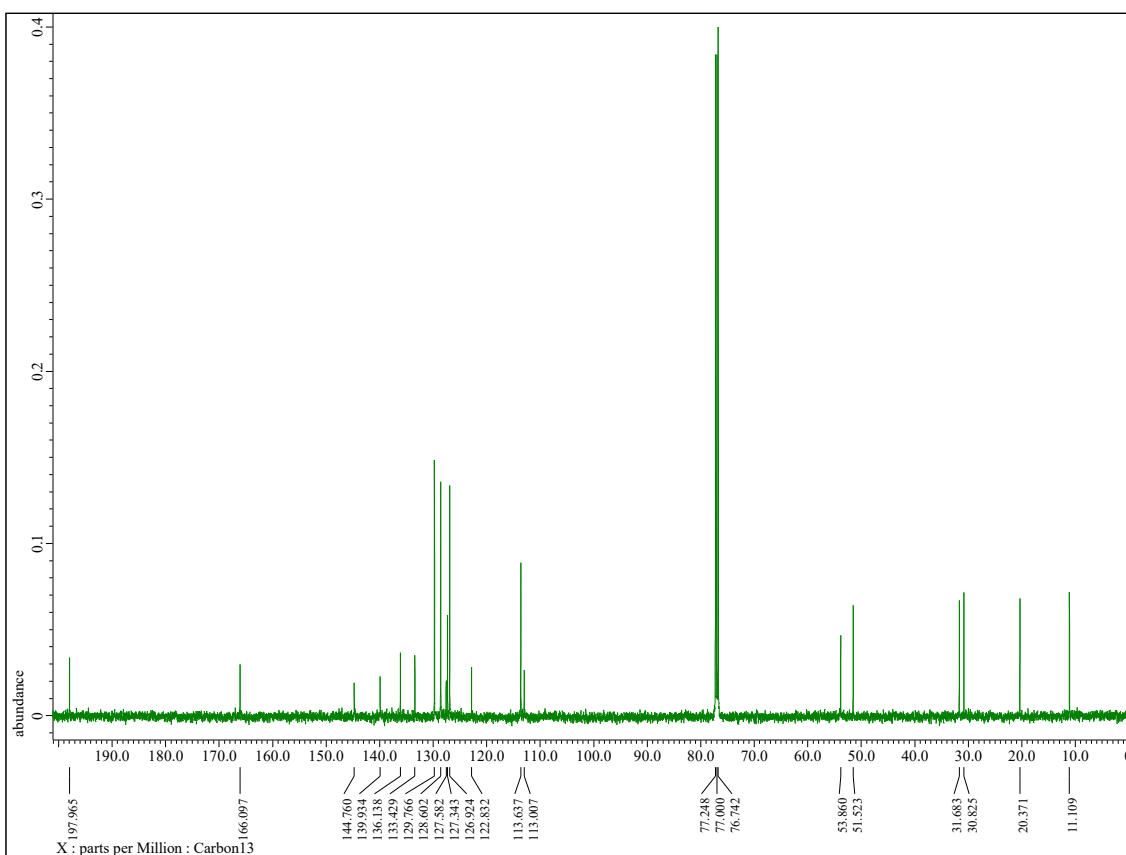


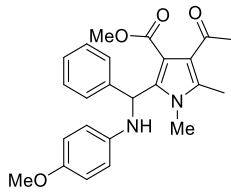


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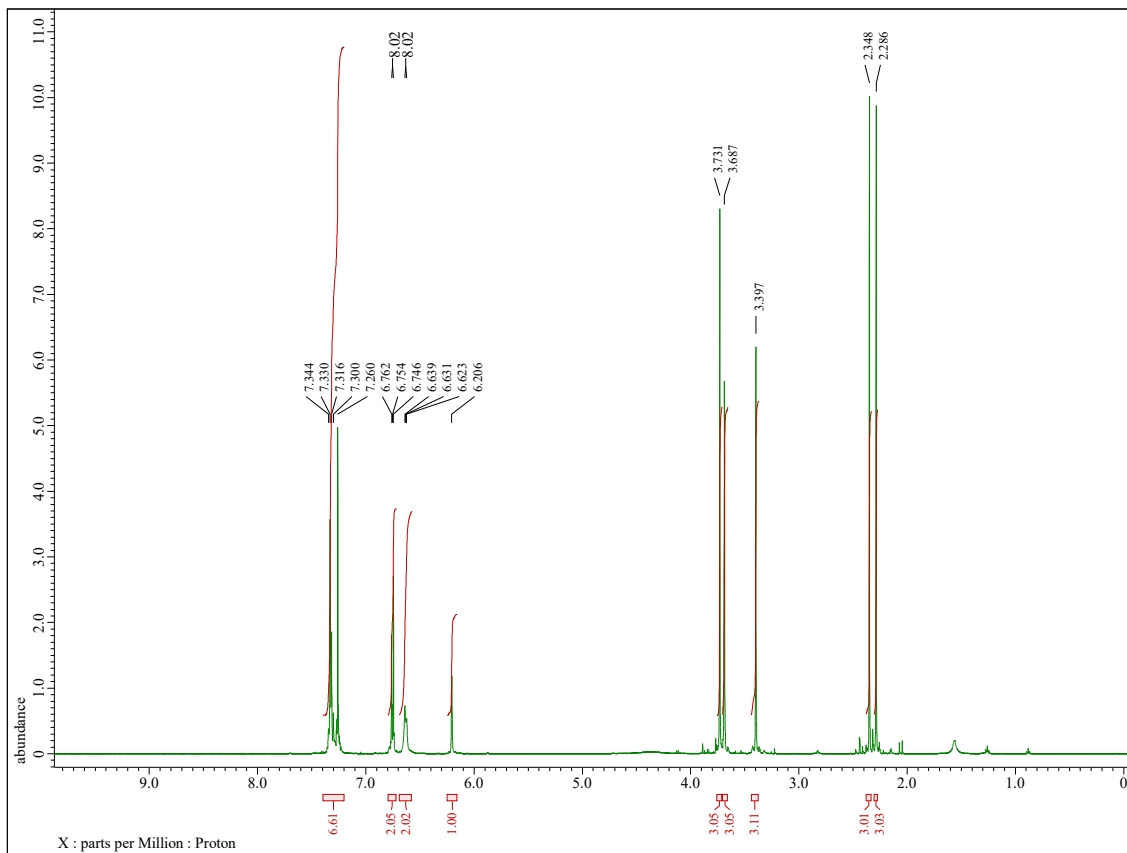


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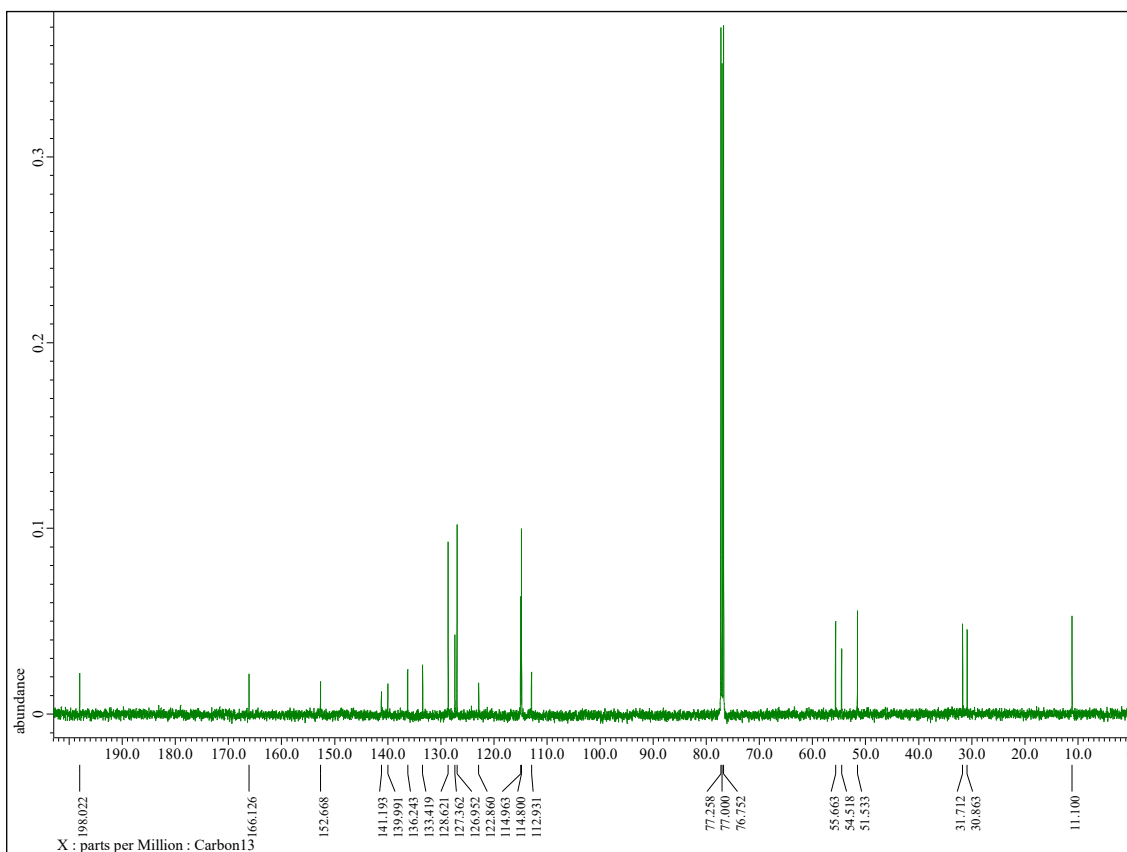


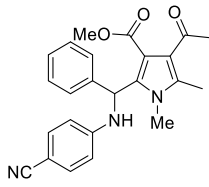


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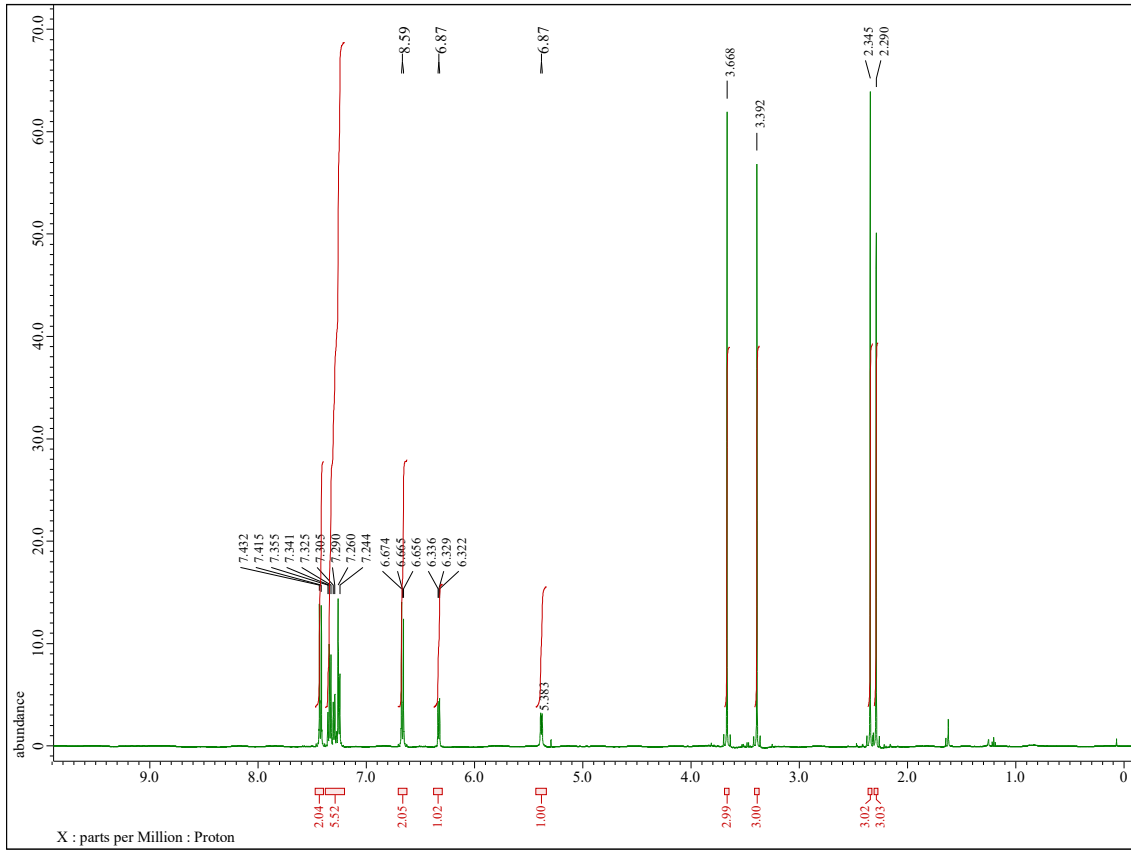


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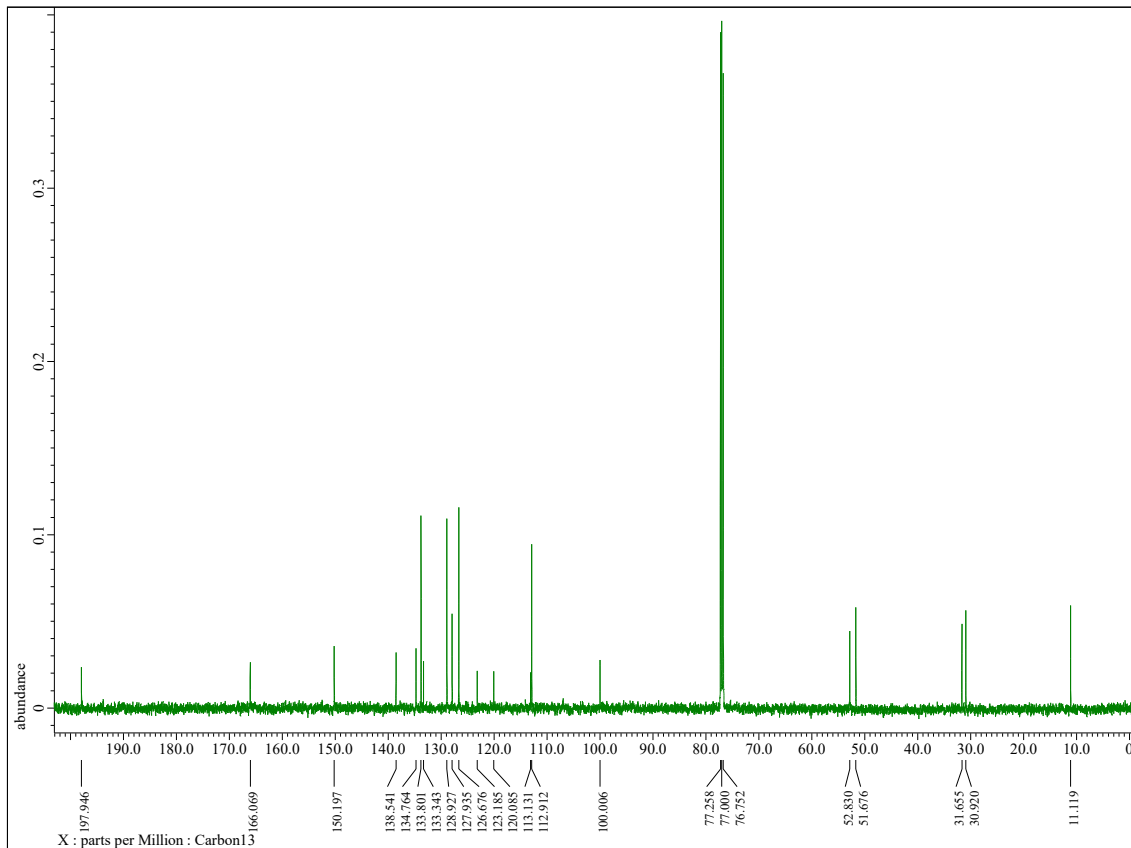


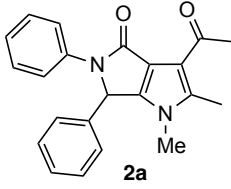


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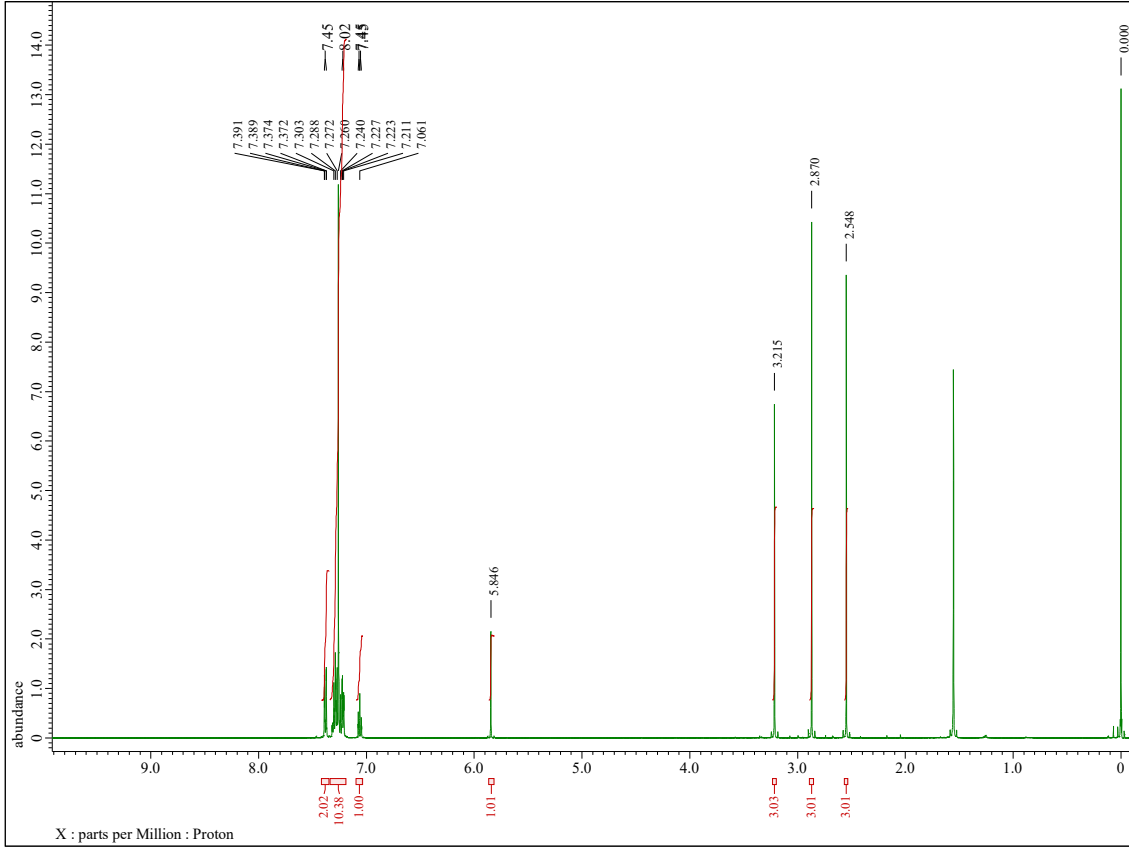


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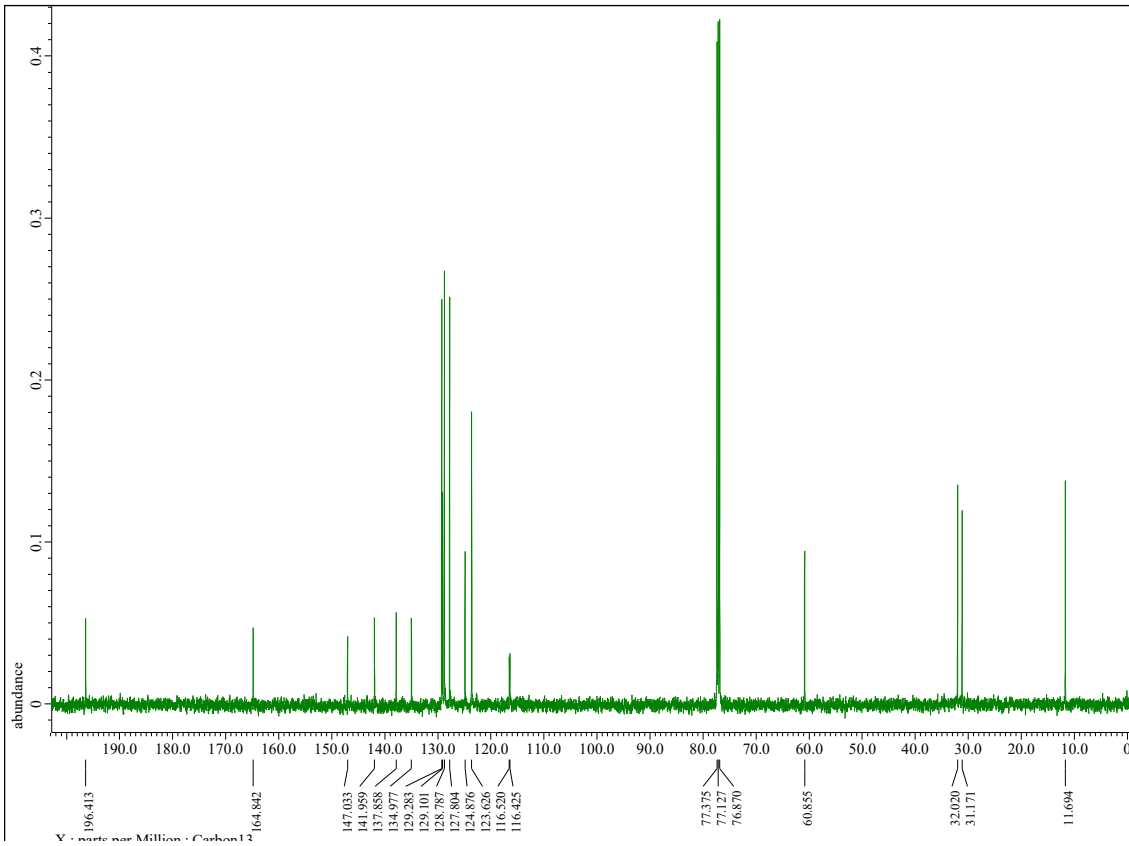


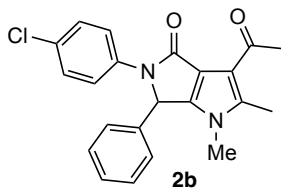


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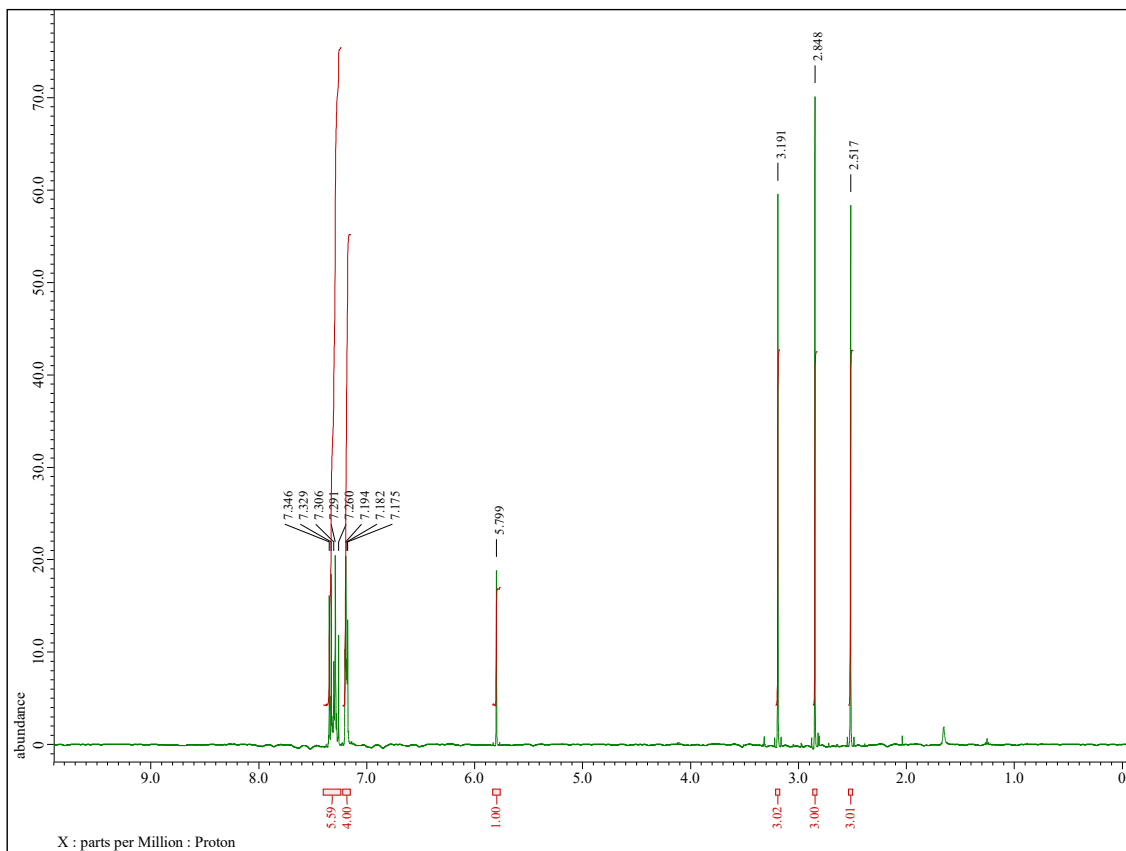


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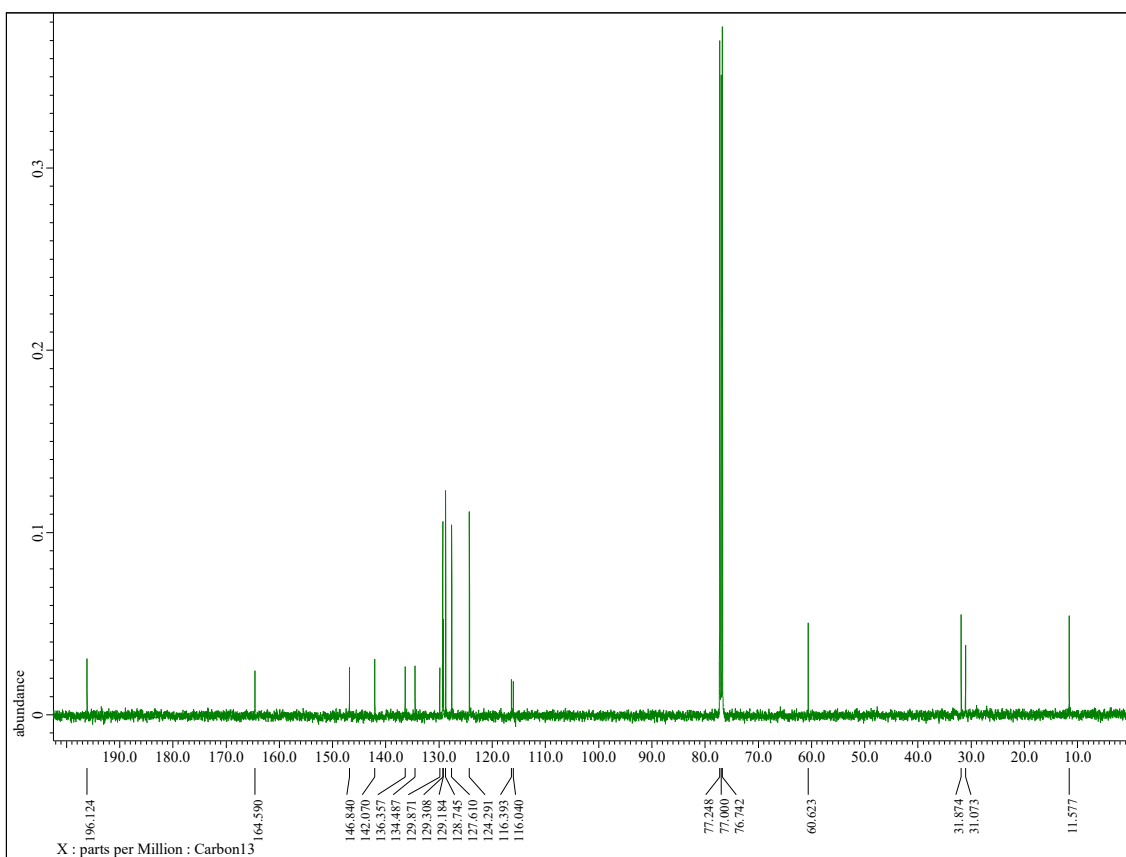


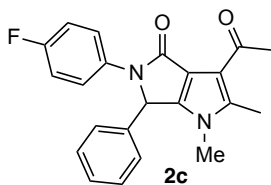


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

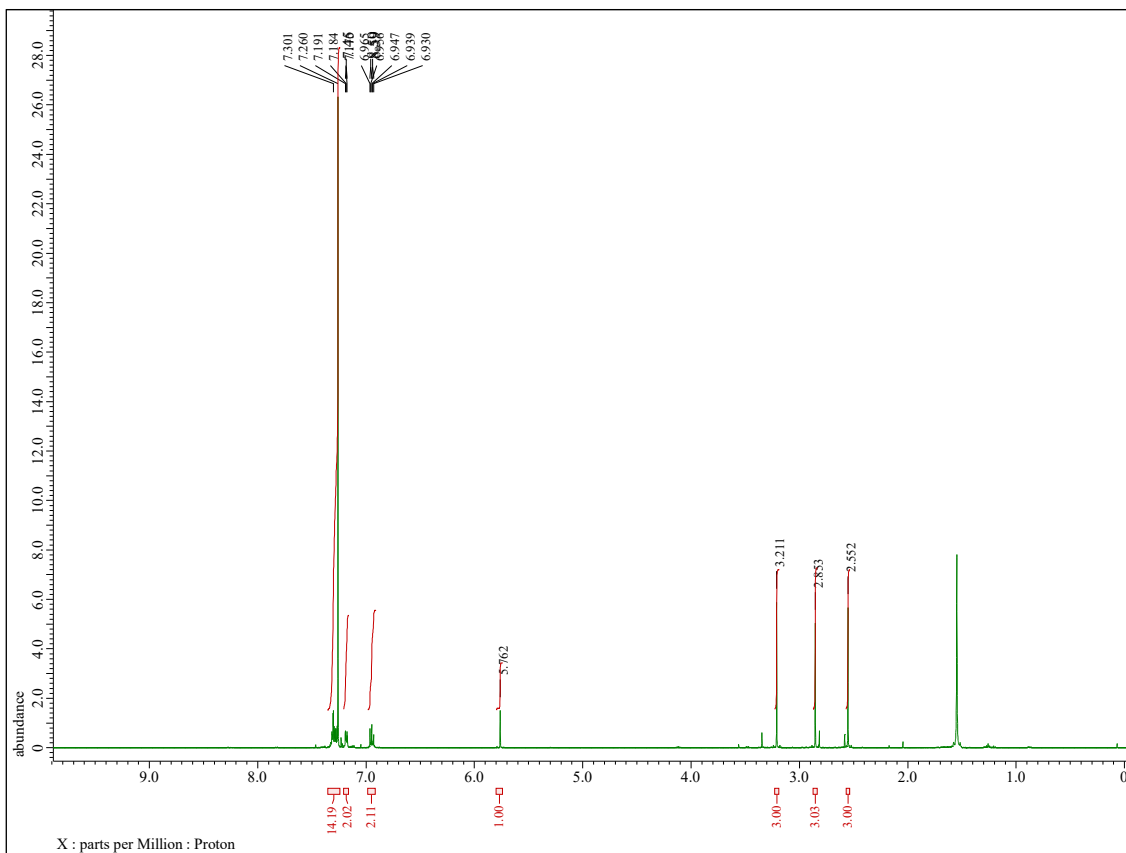


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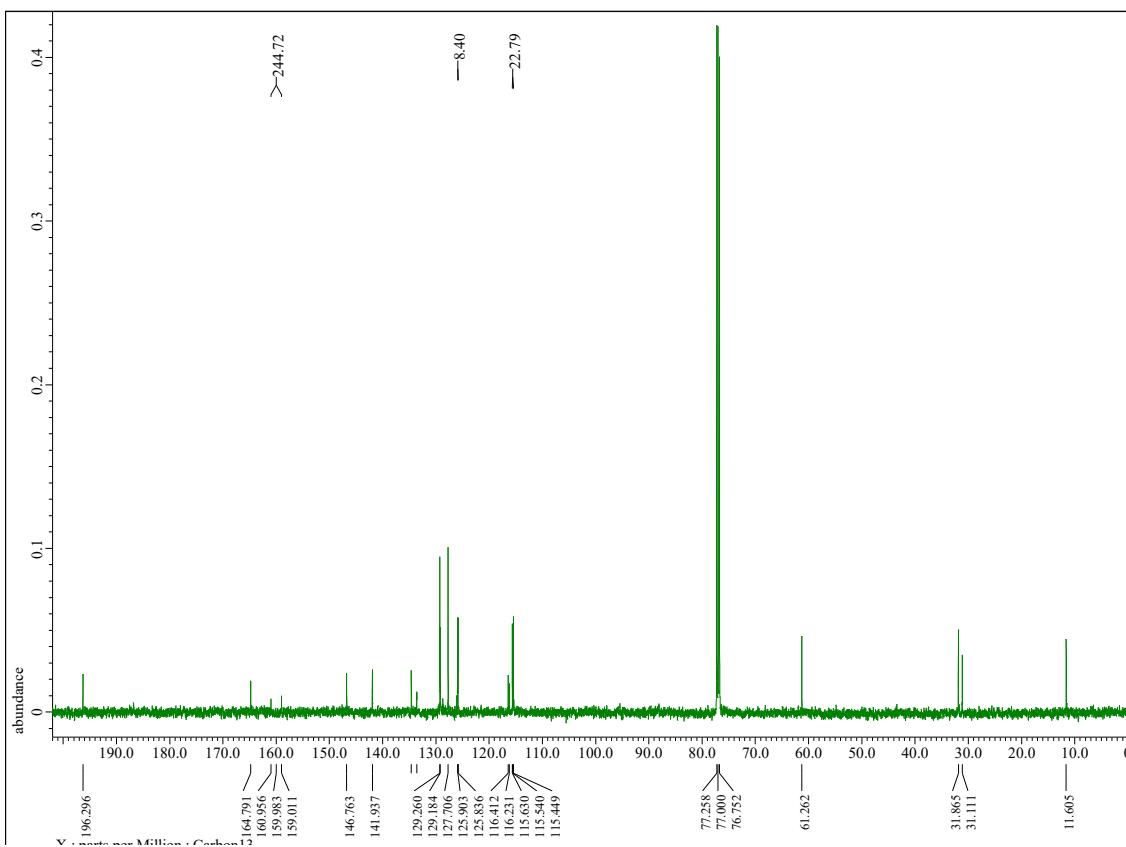


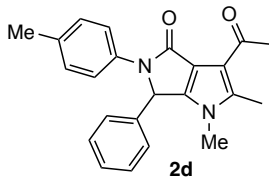


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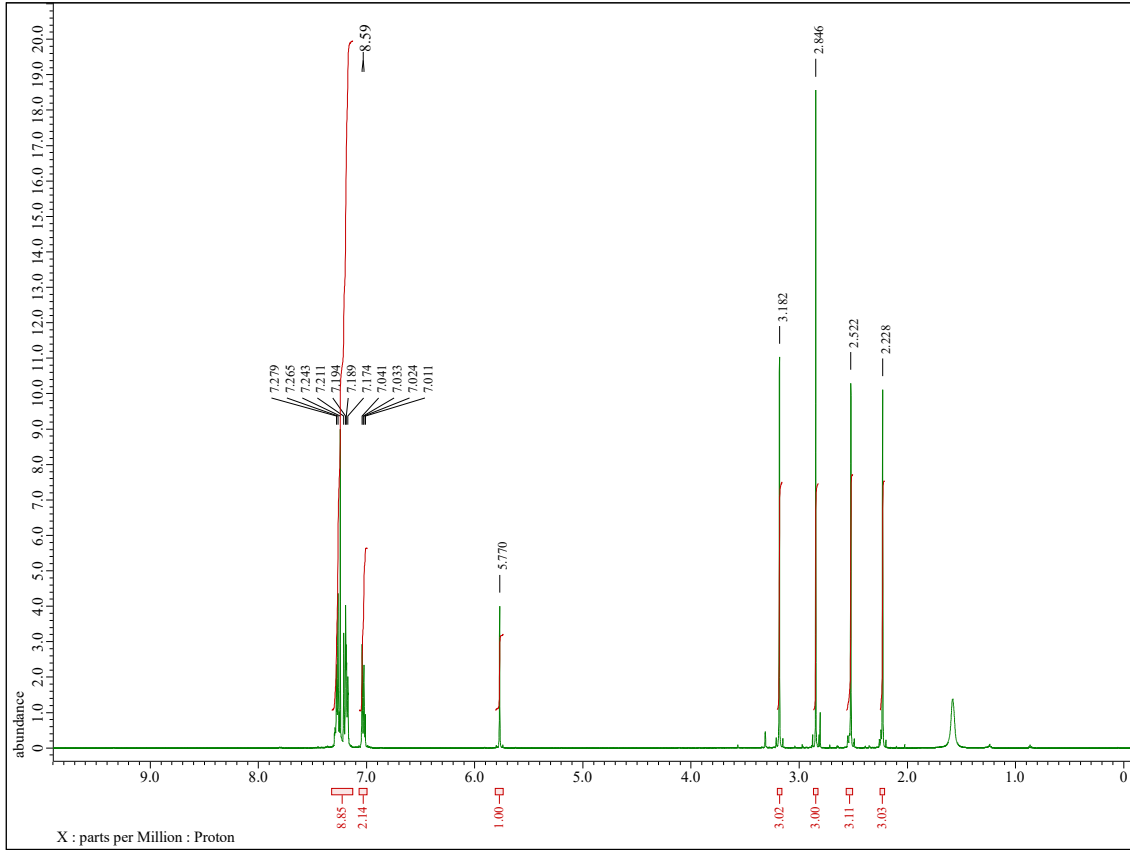


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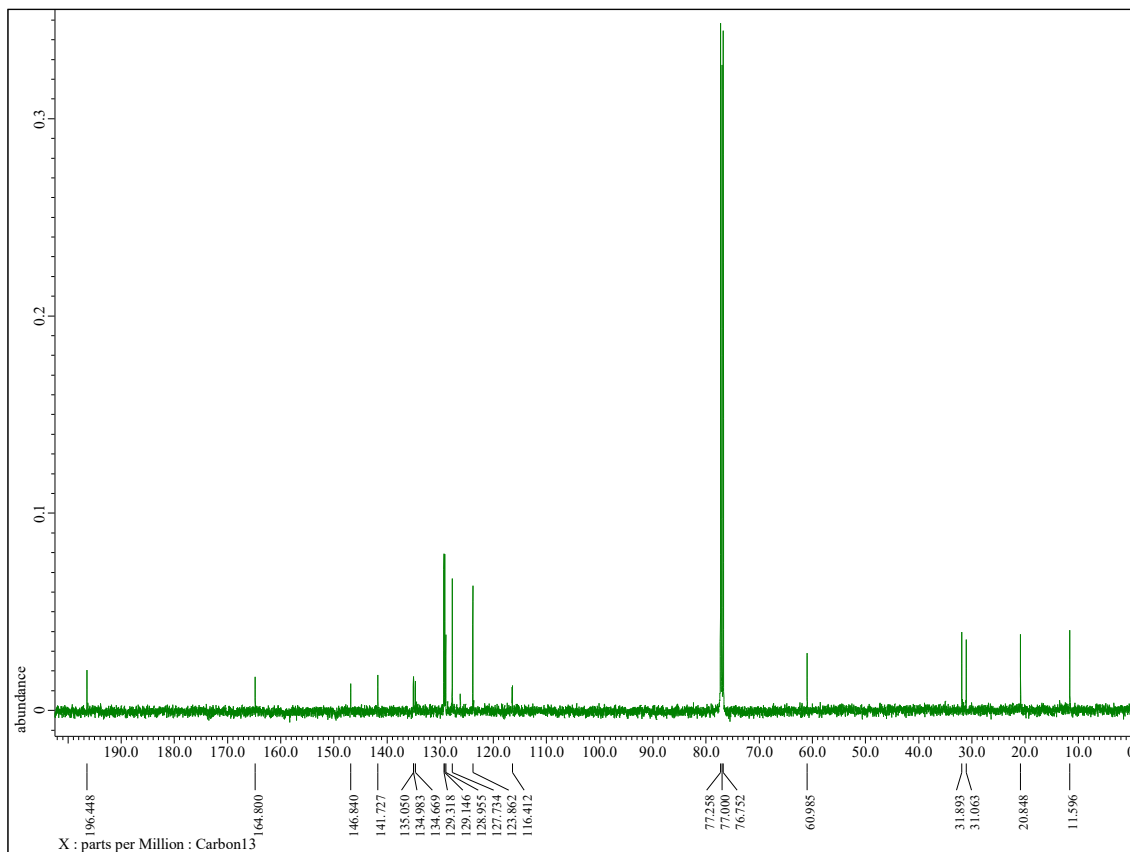


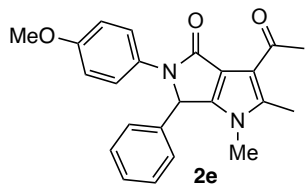


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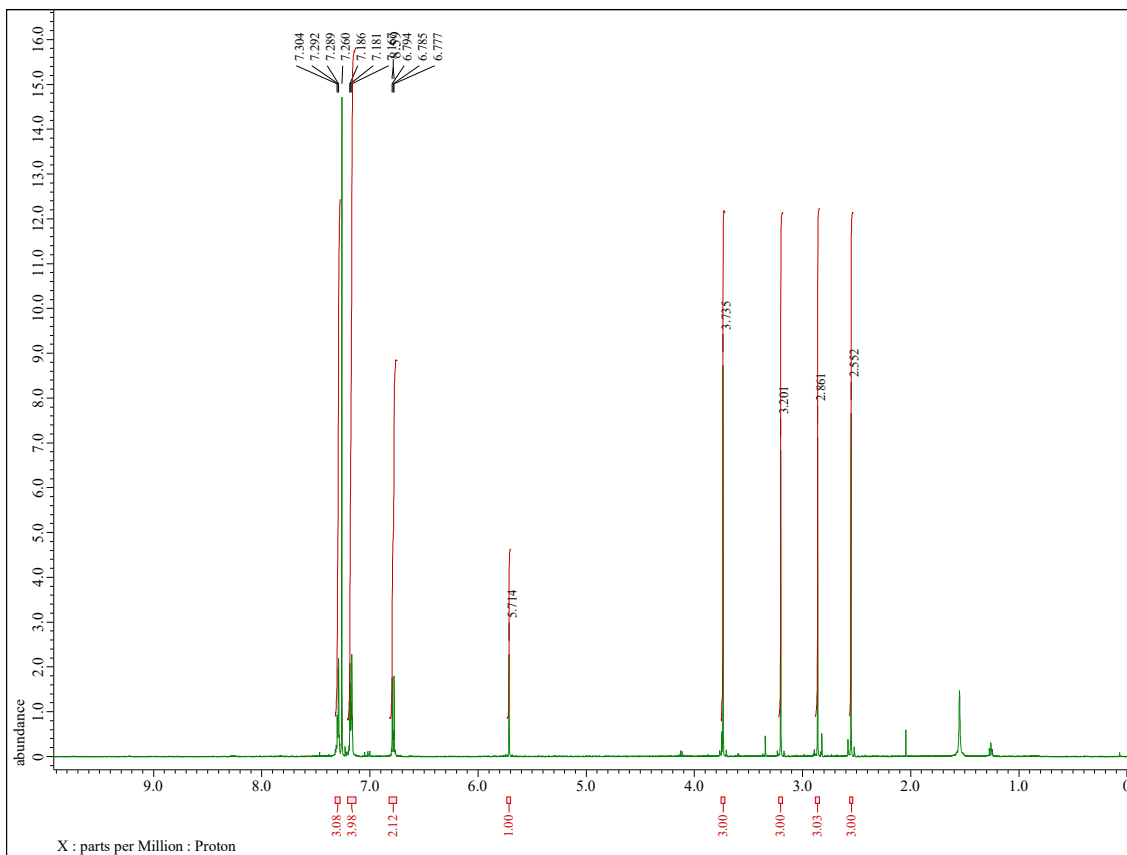


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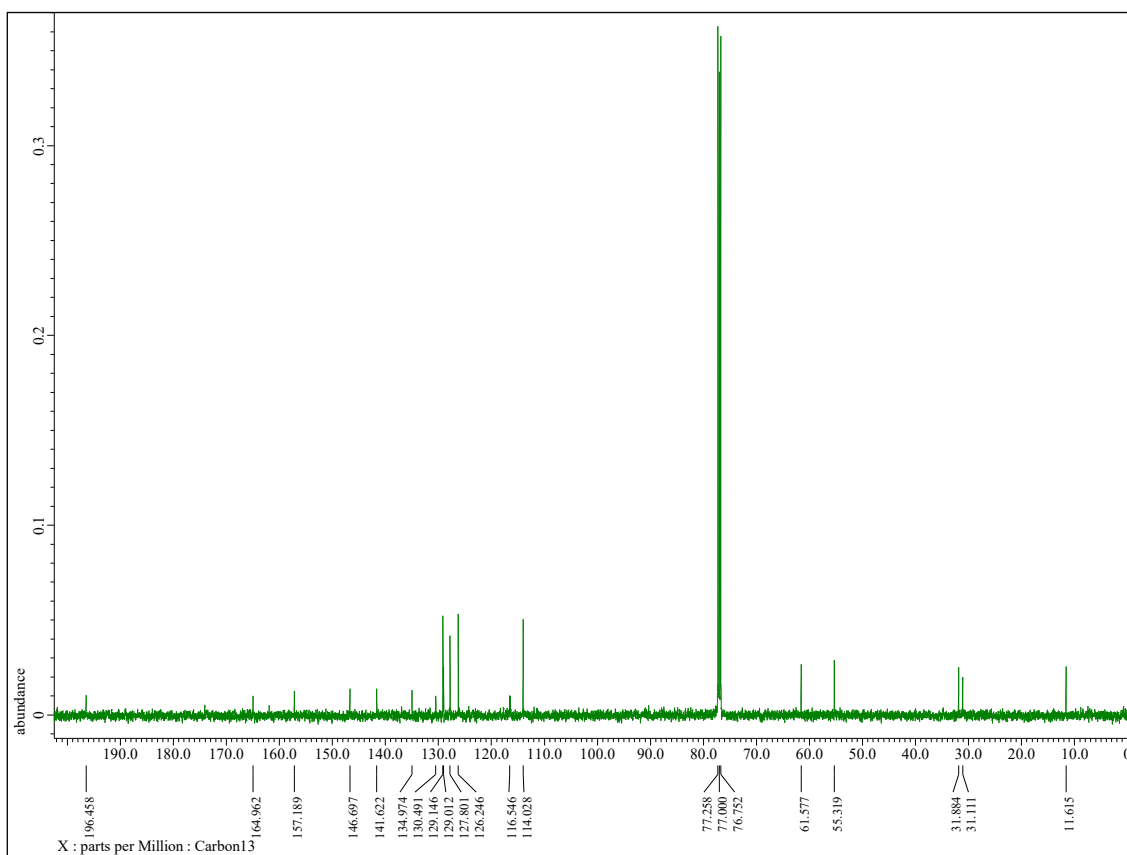


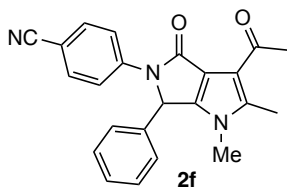


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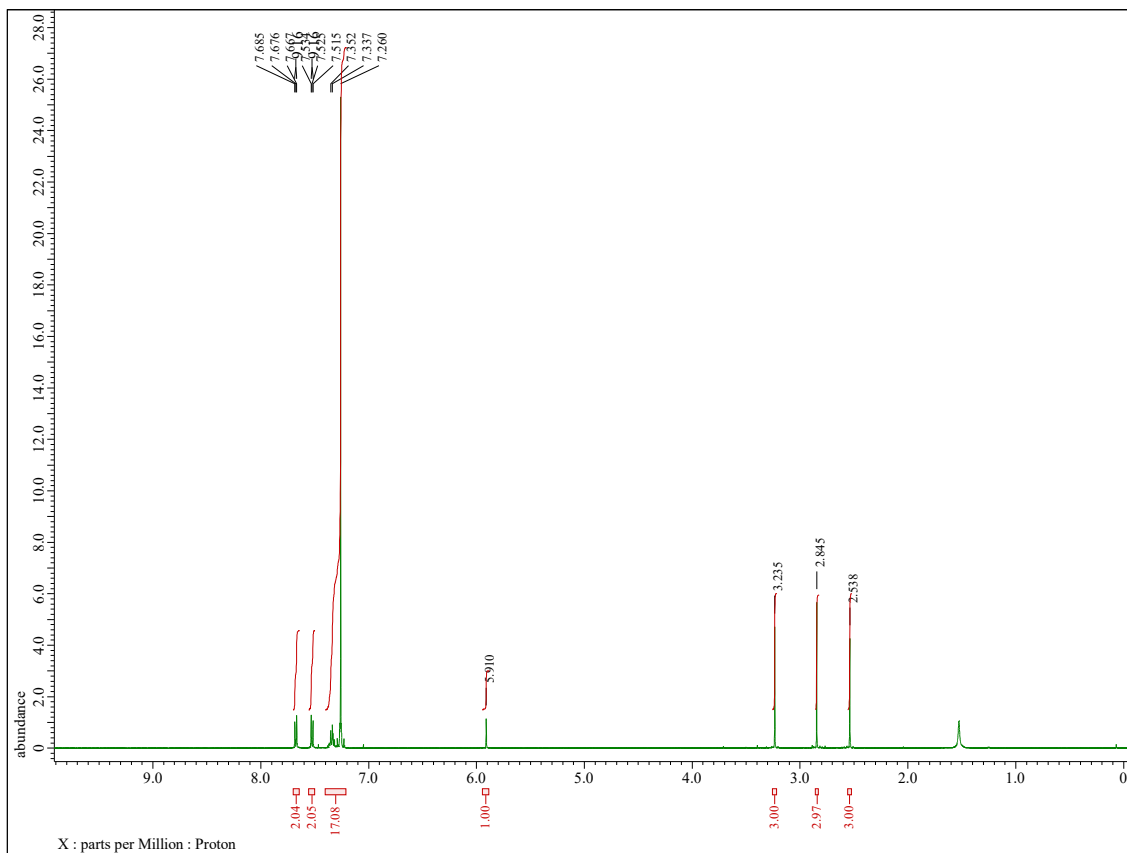


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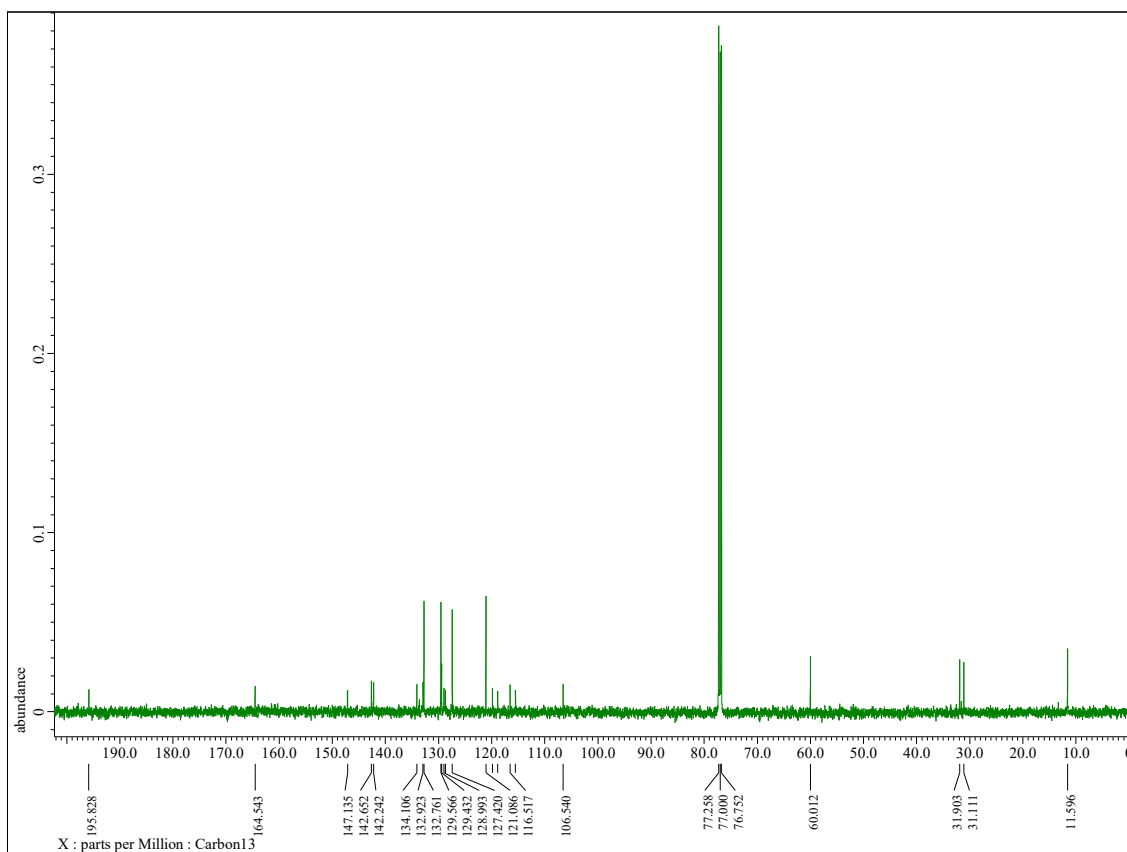


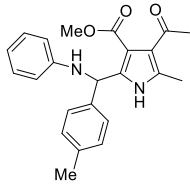


500 MHz <sup>1</sup>HNMR in CDCl<sub>3</sub>

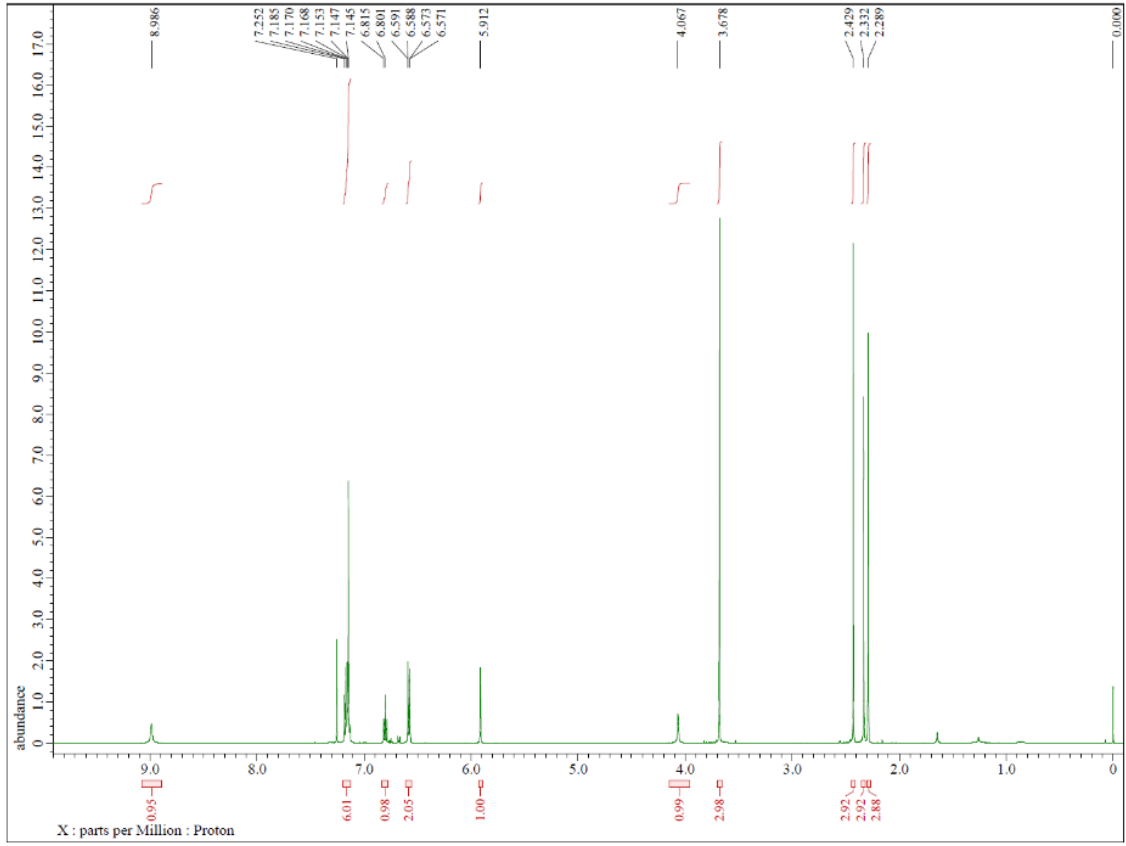


125 MHz <sup>13</sup>CNMR in CDCl<sub>3</sub>

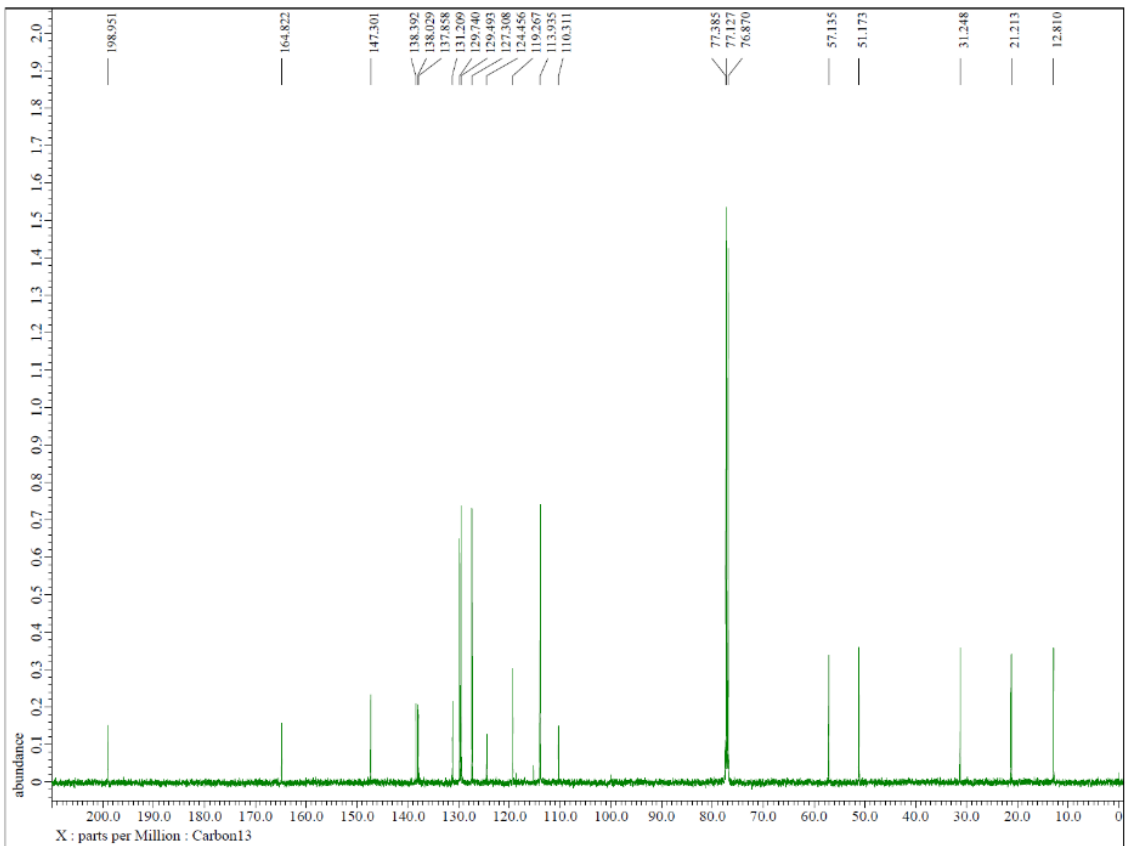


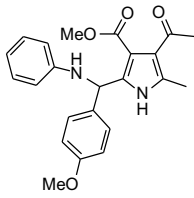


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

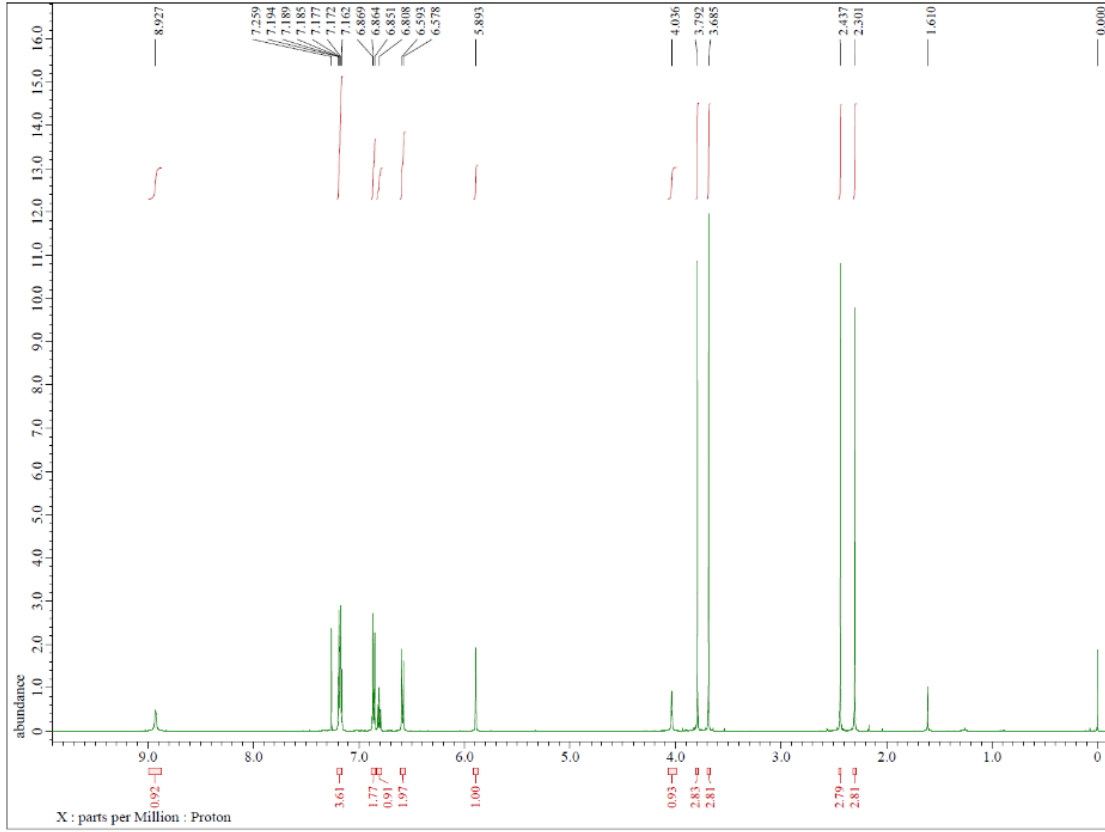


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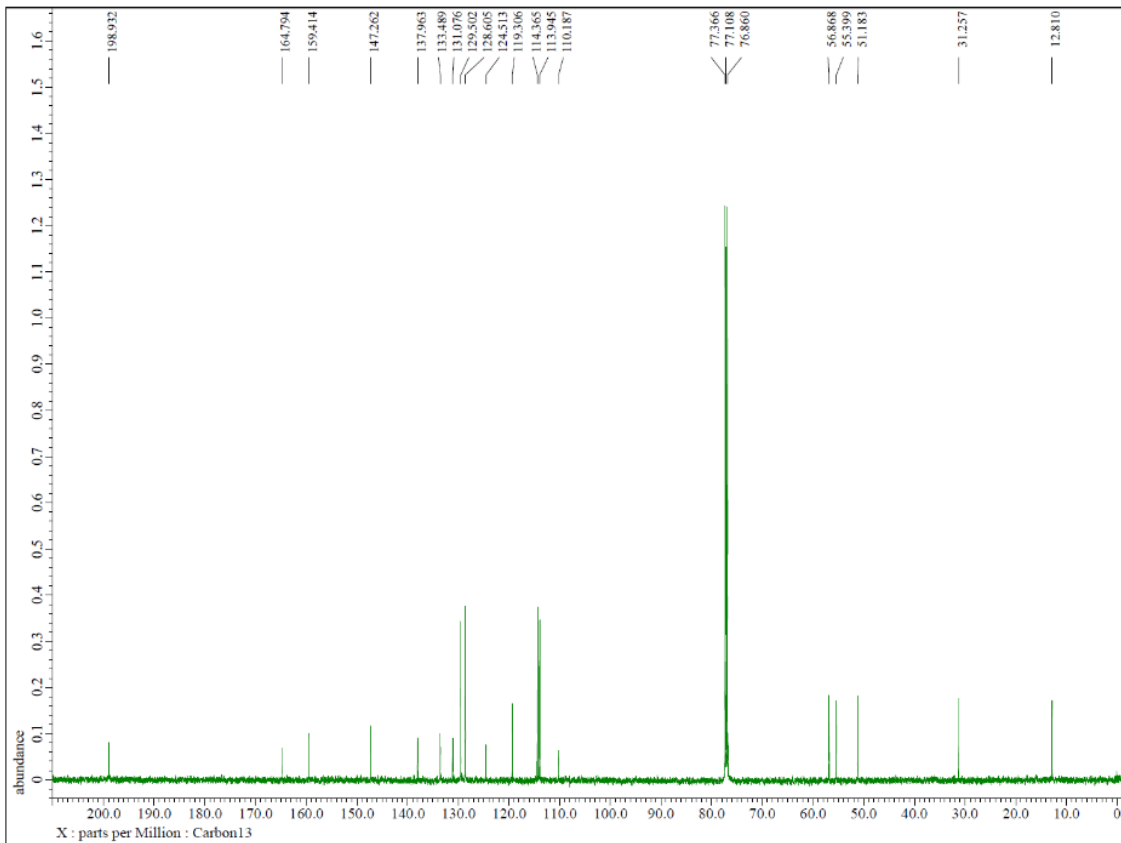


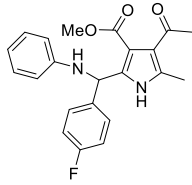


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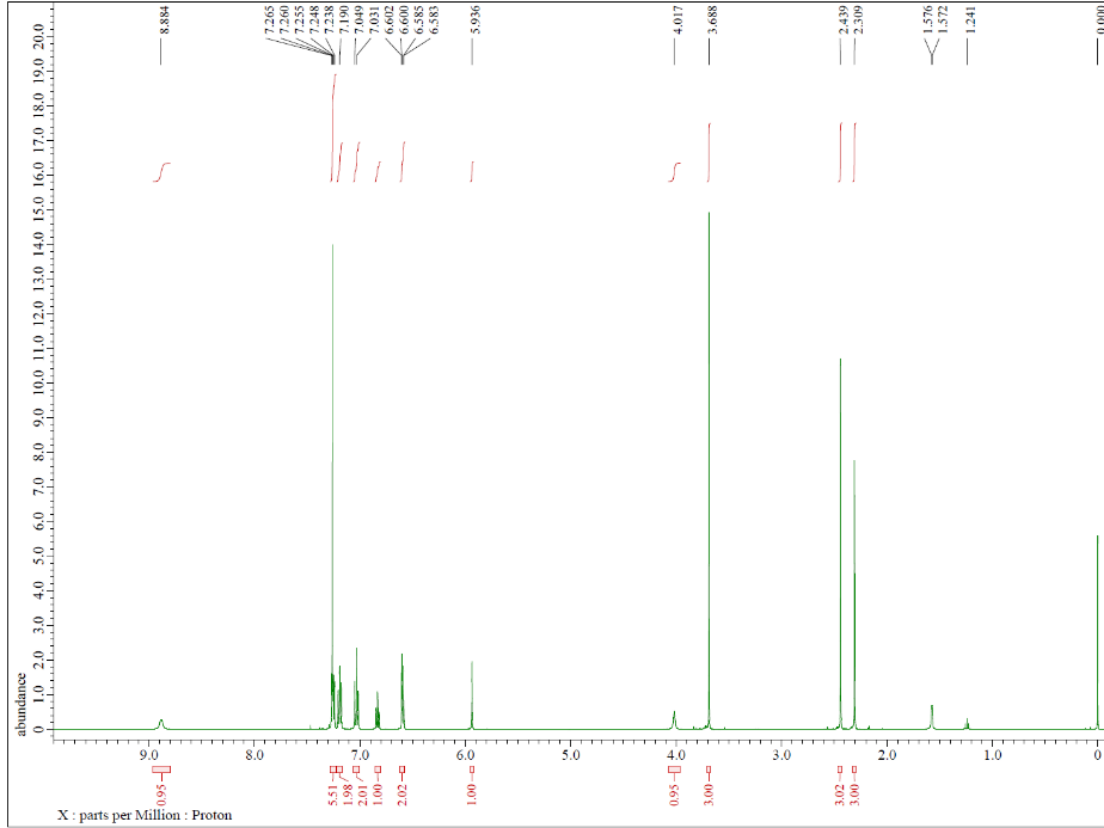


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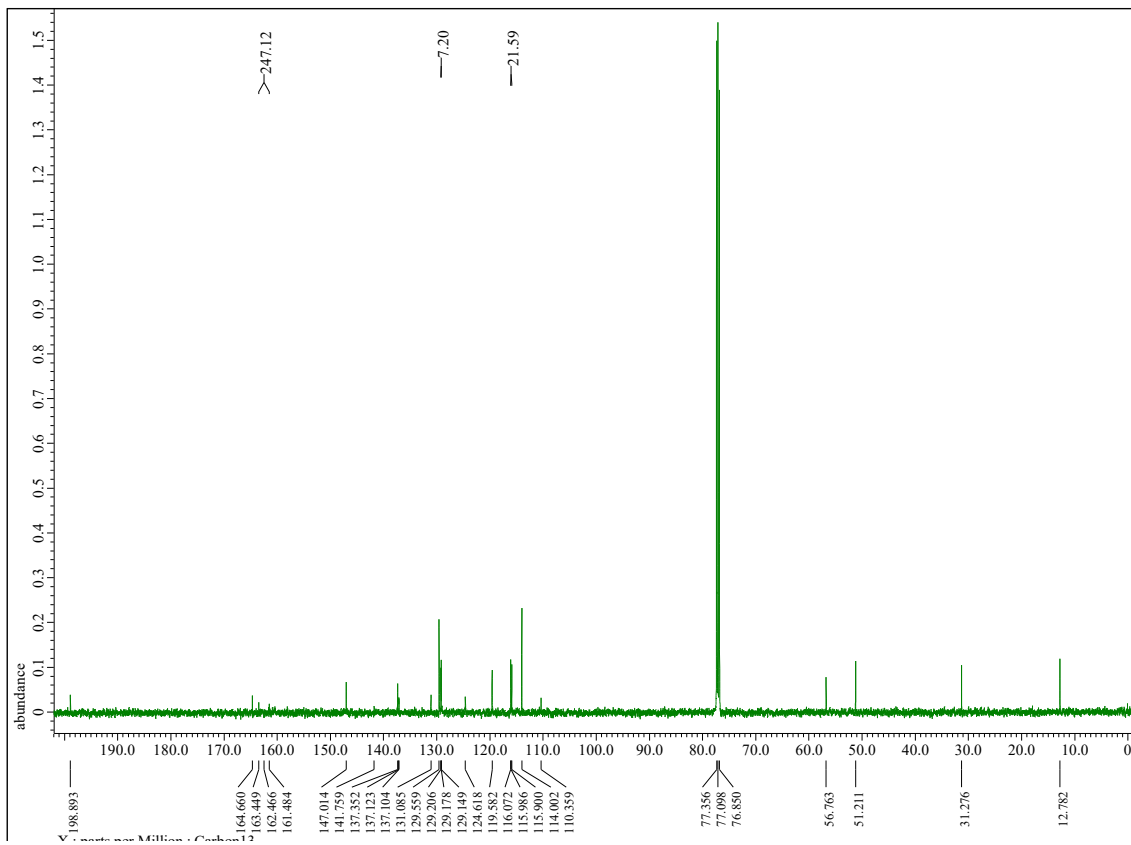


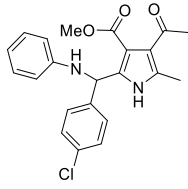


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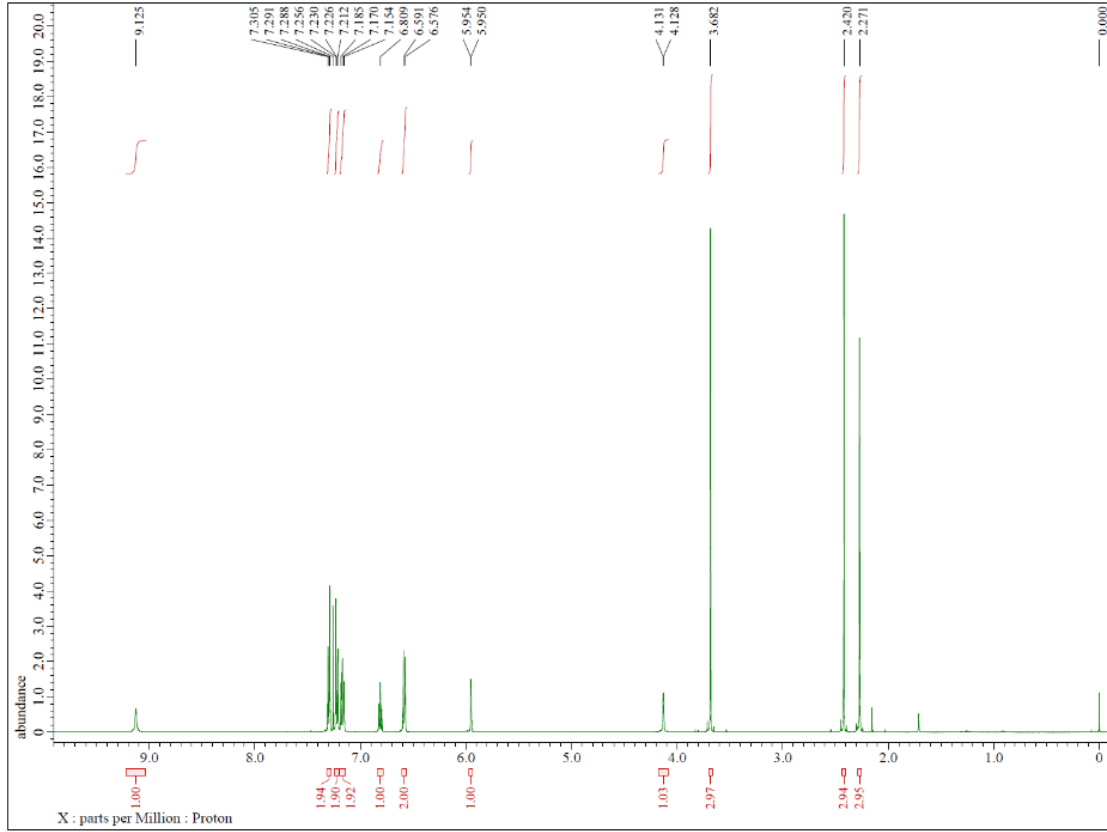


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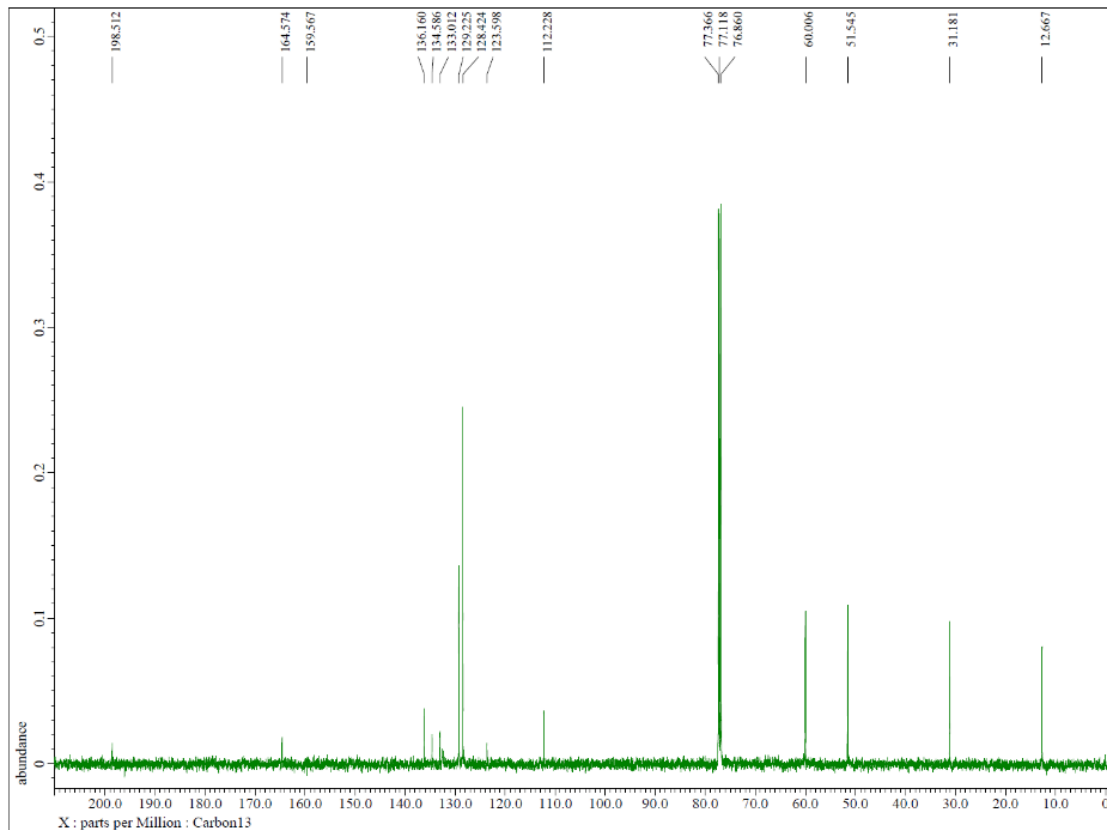


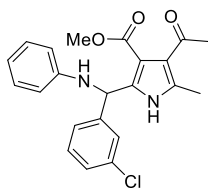


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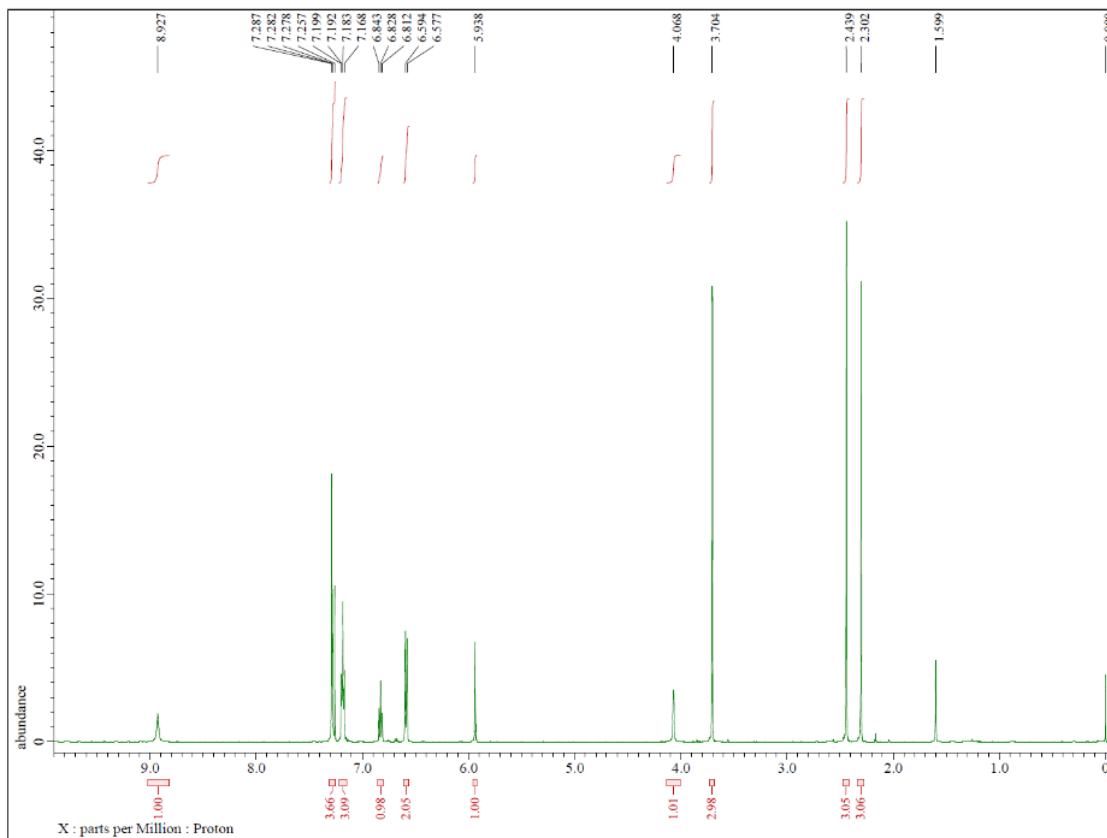


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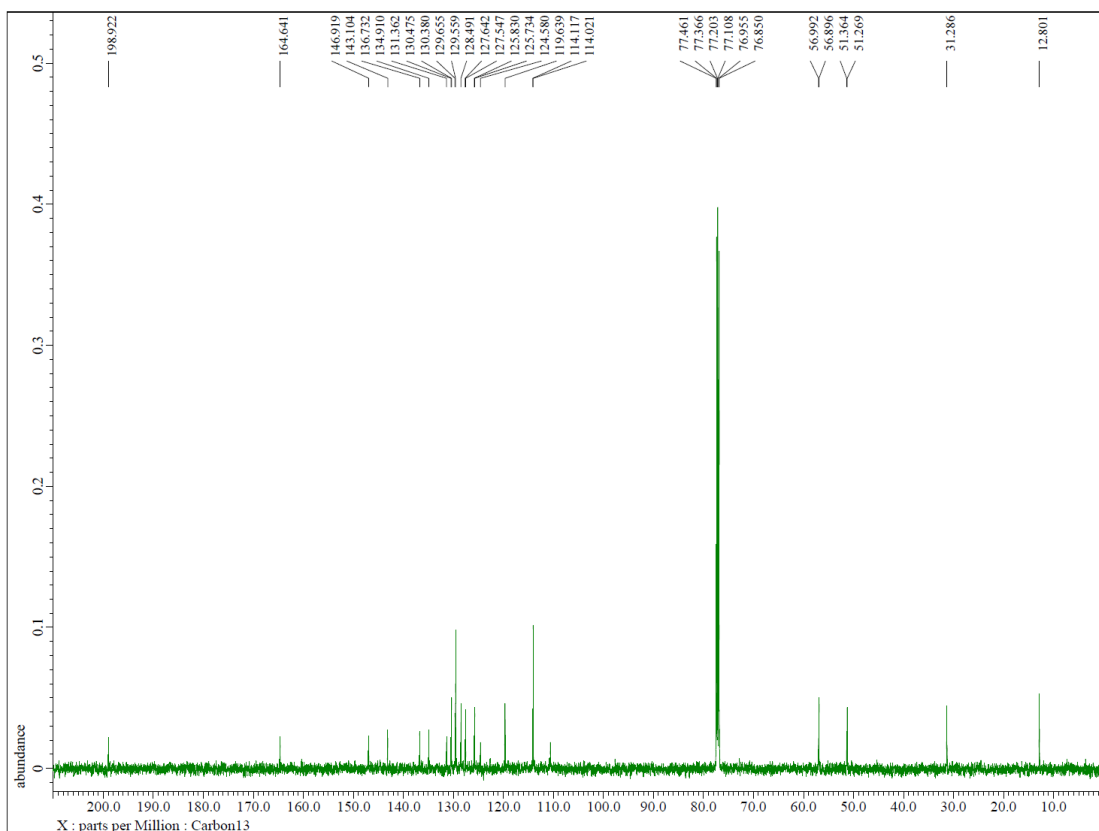


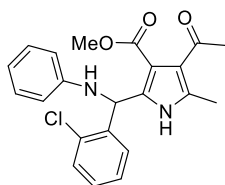


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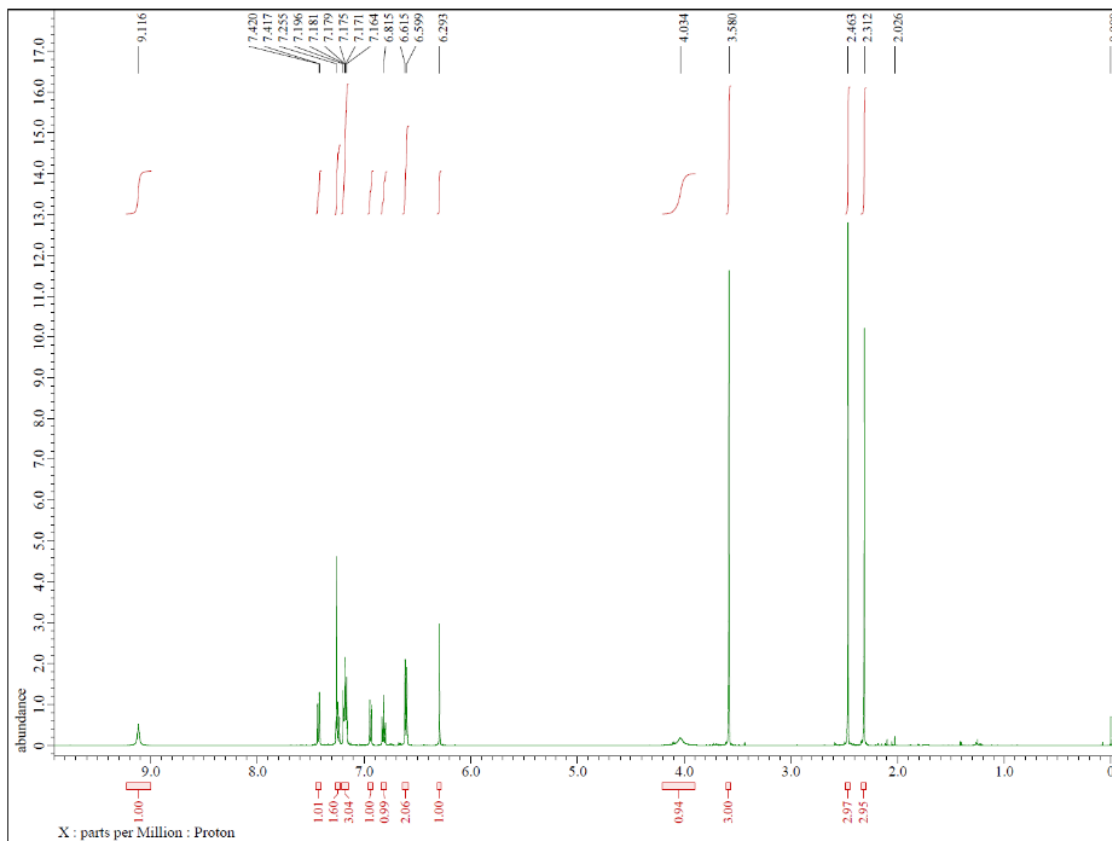


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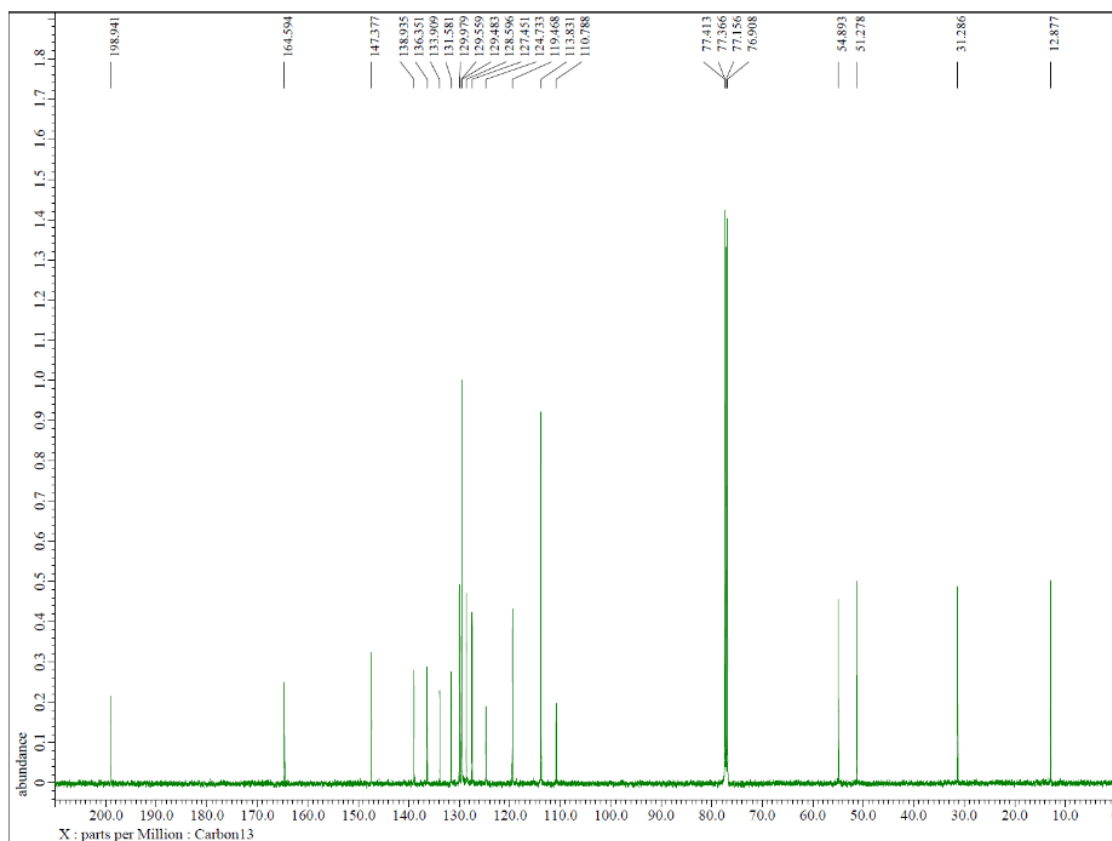


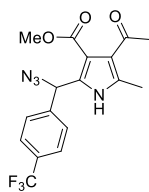


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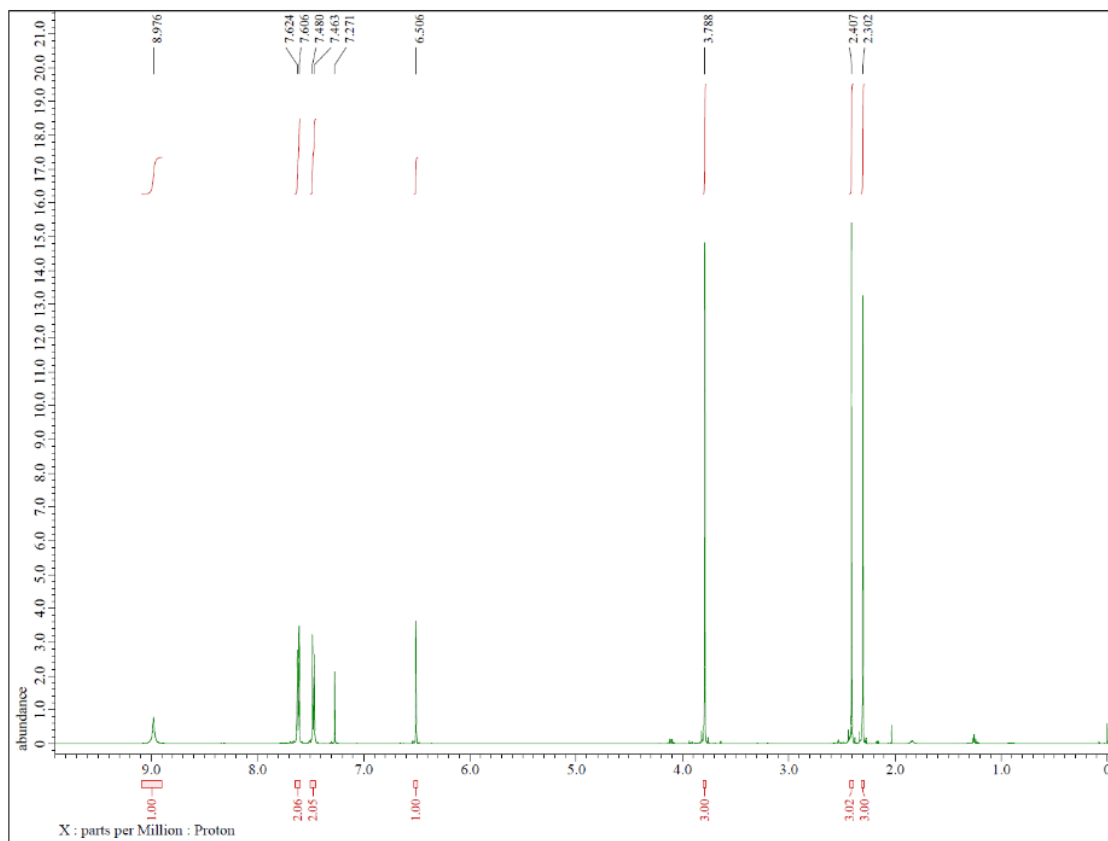


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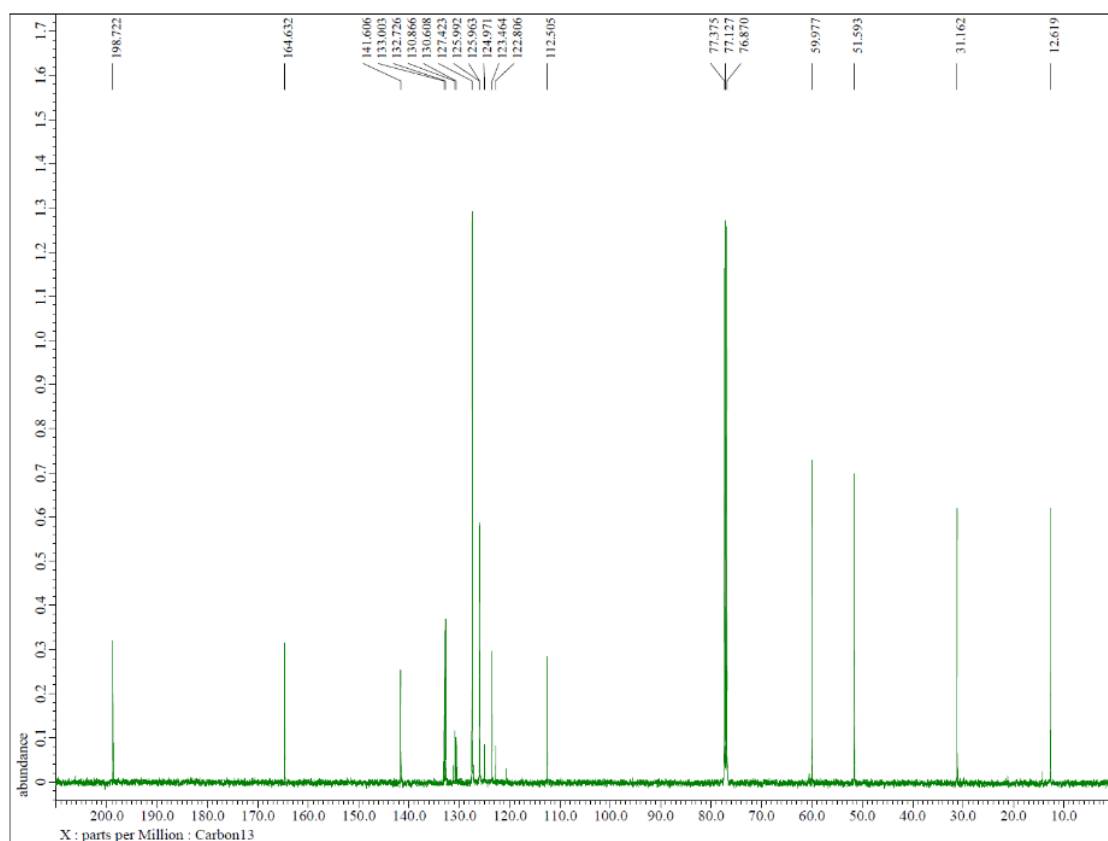


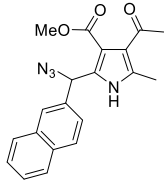


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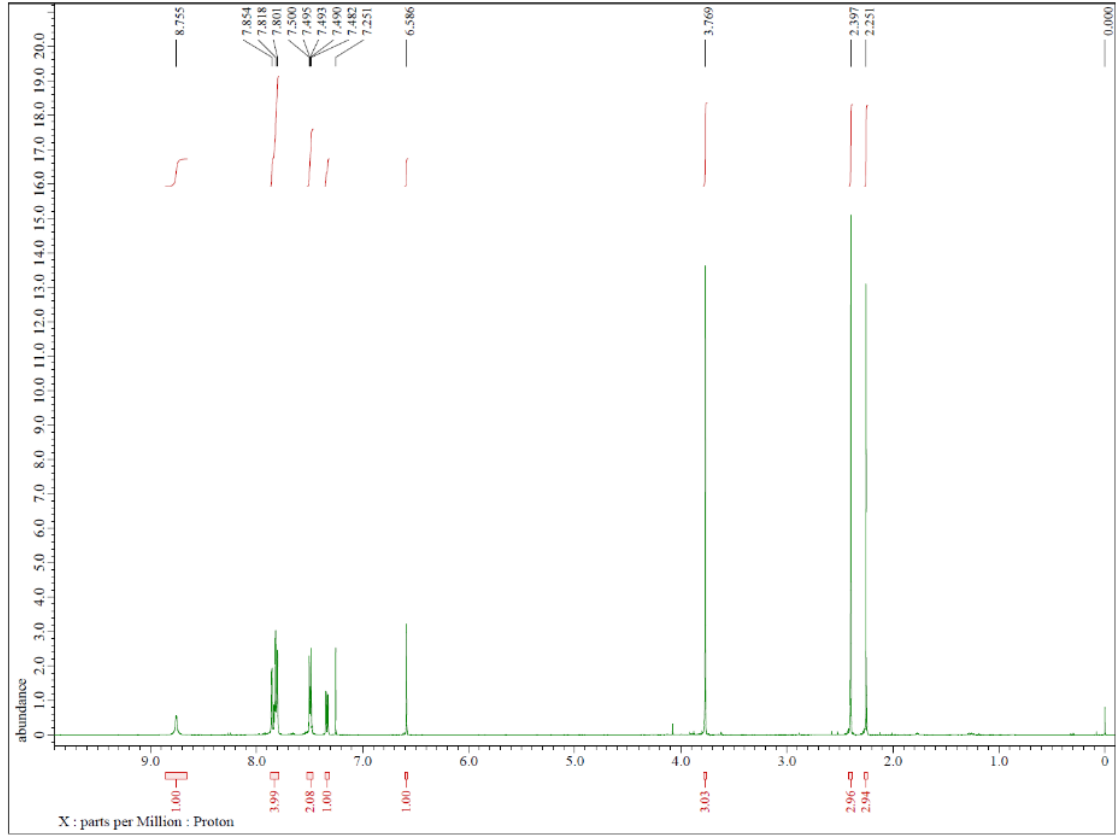


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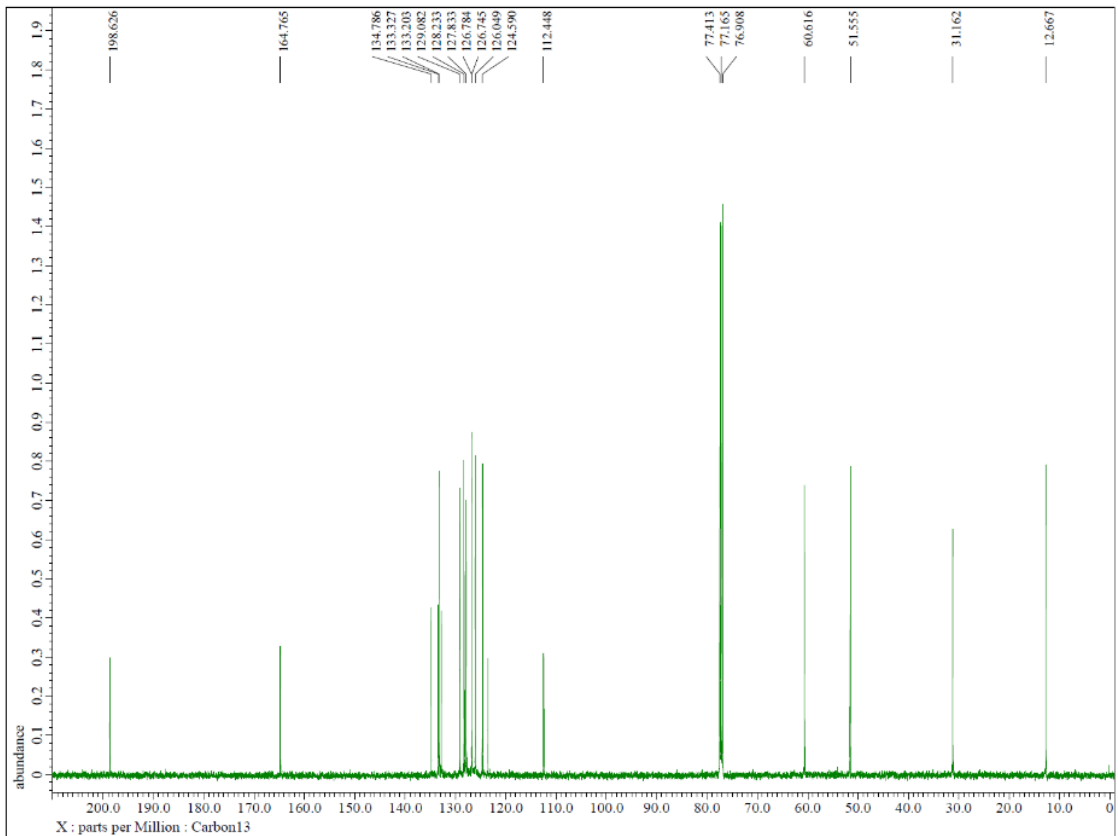


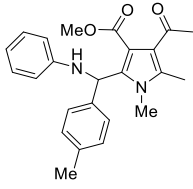


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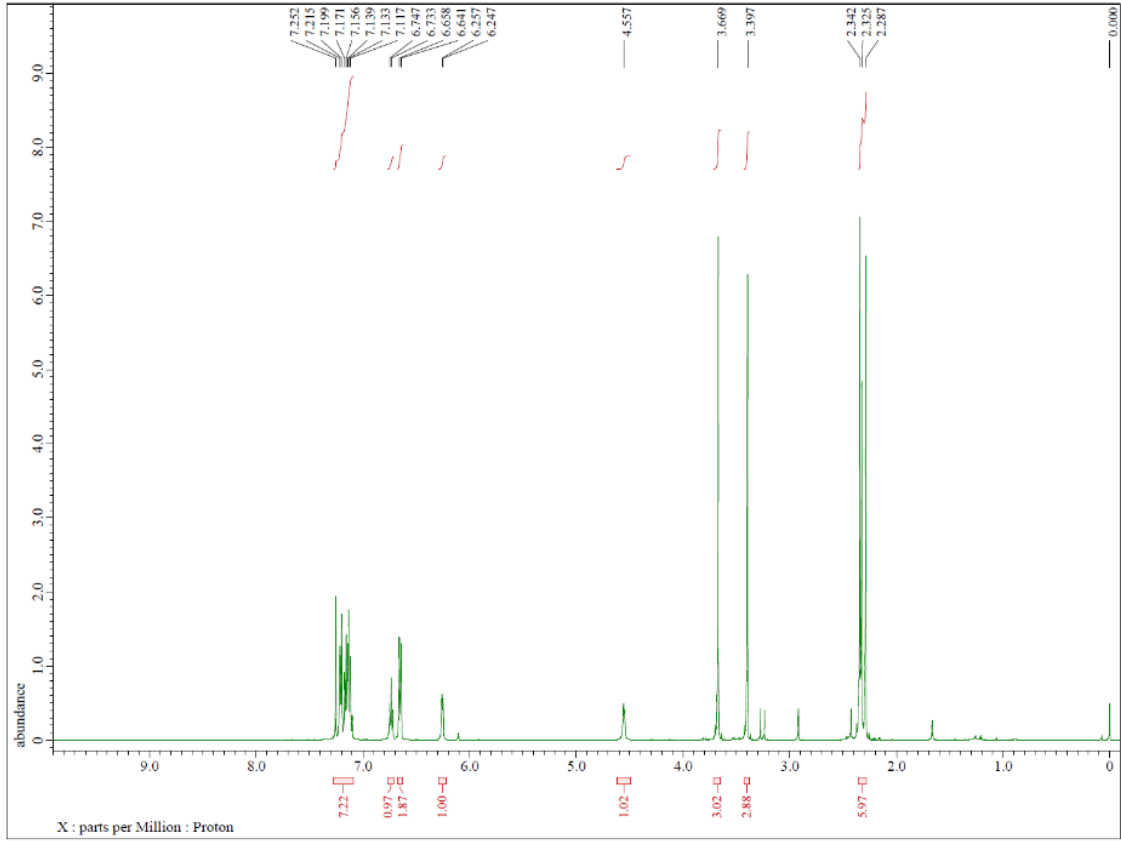


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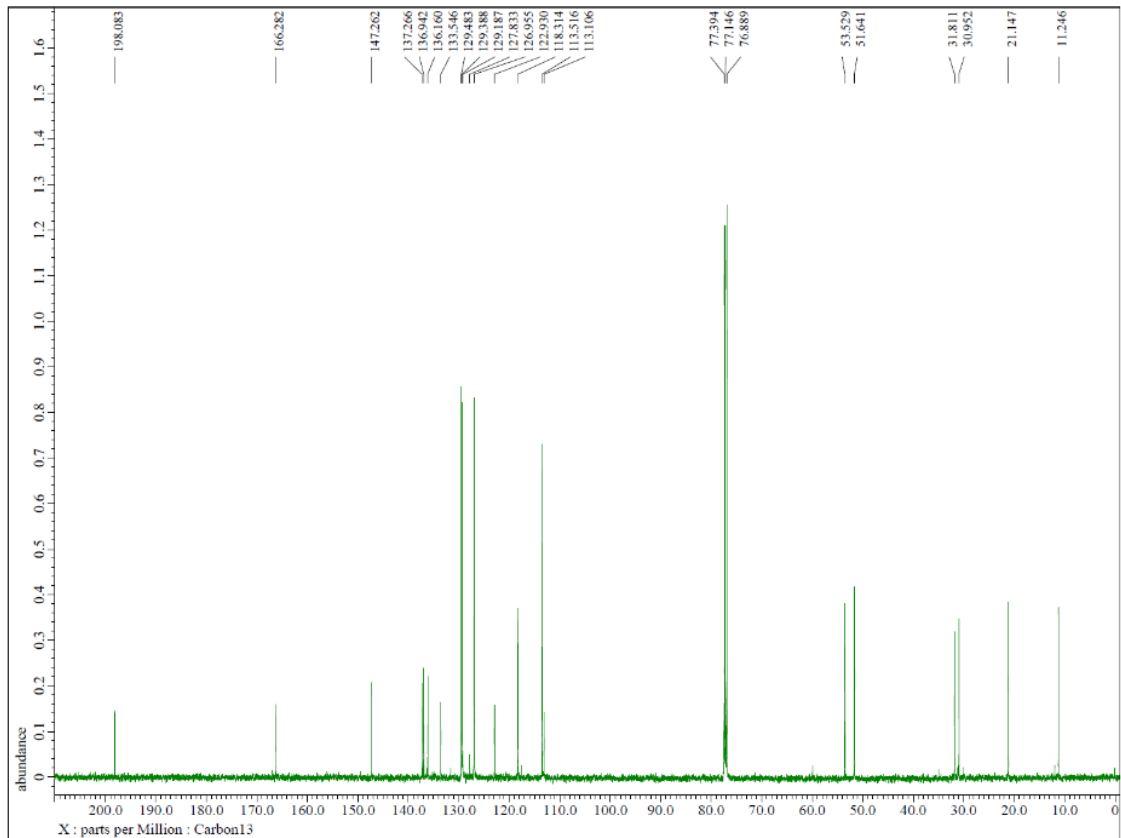


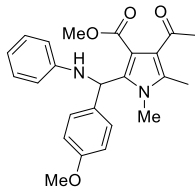


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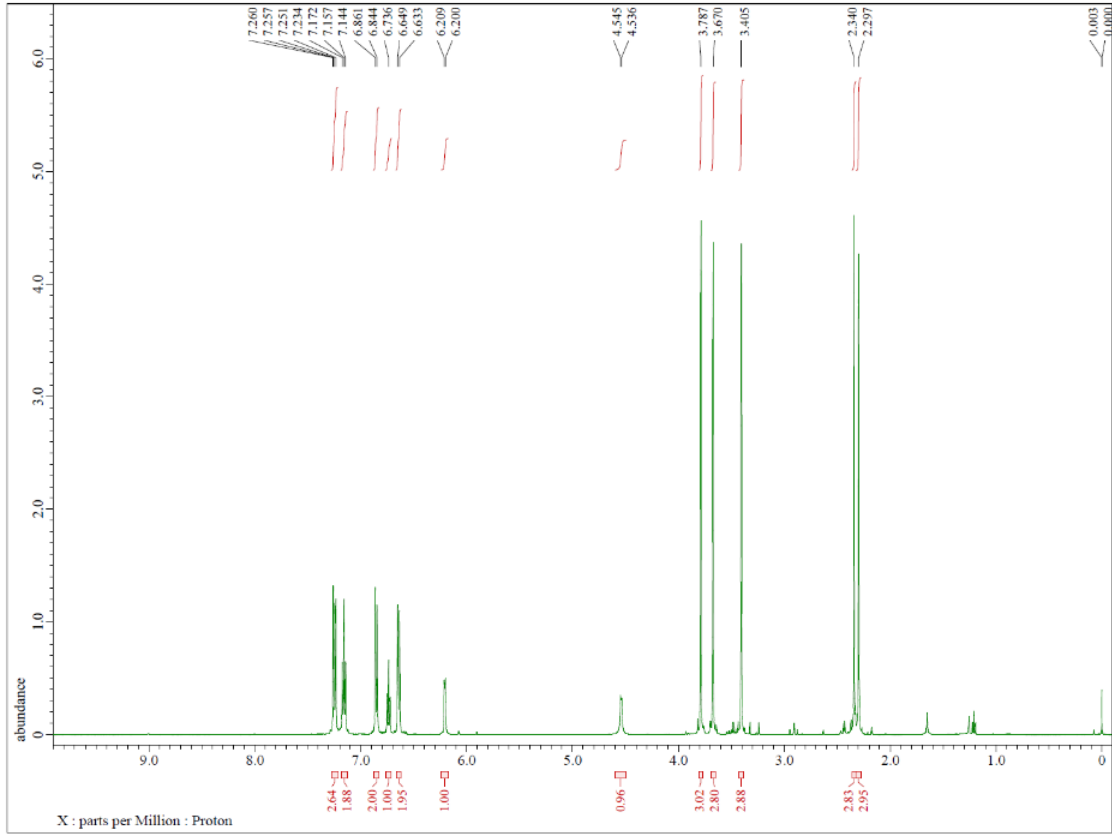


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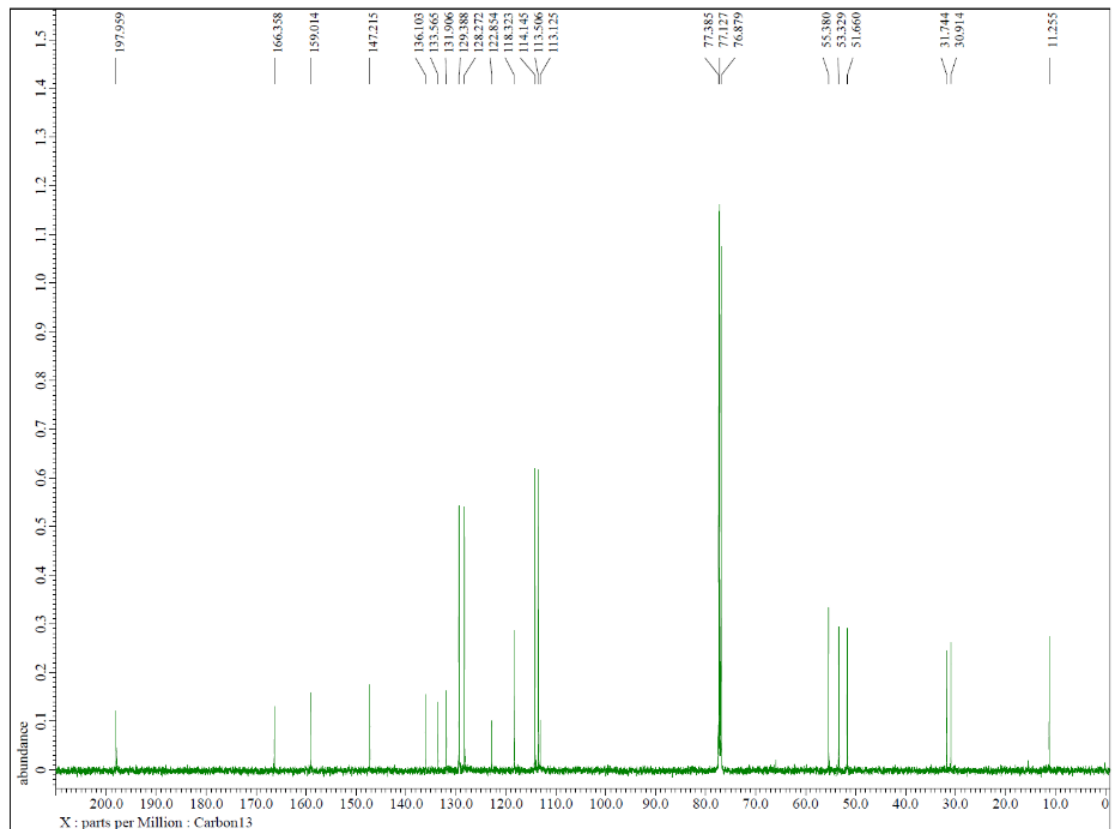


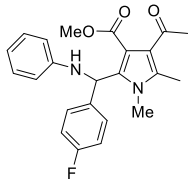


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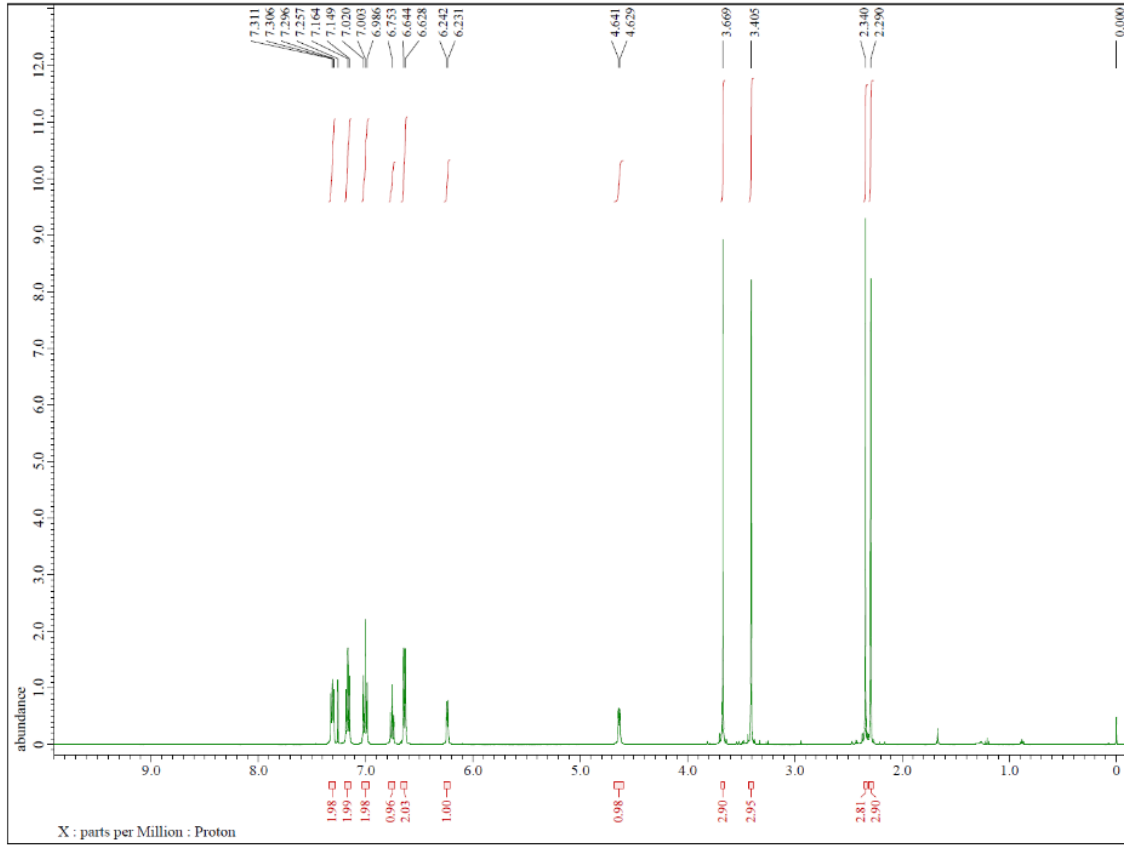


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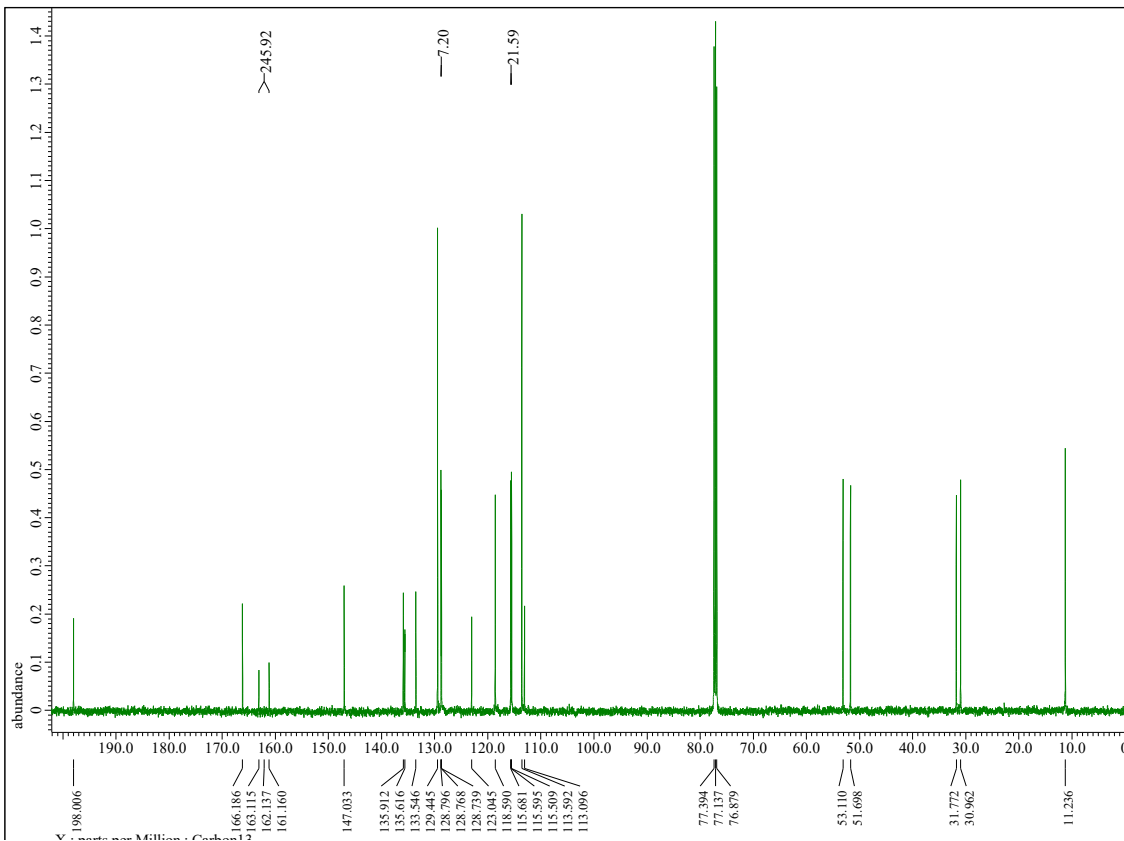


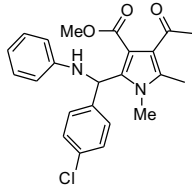


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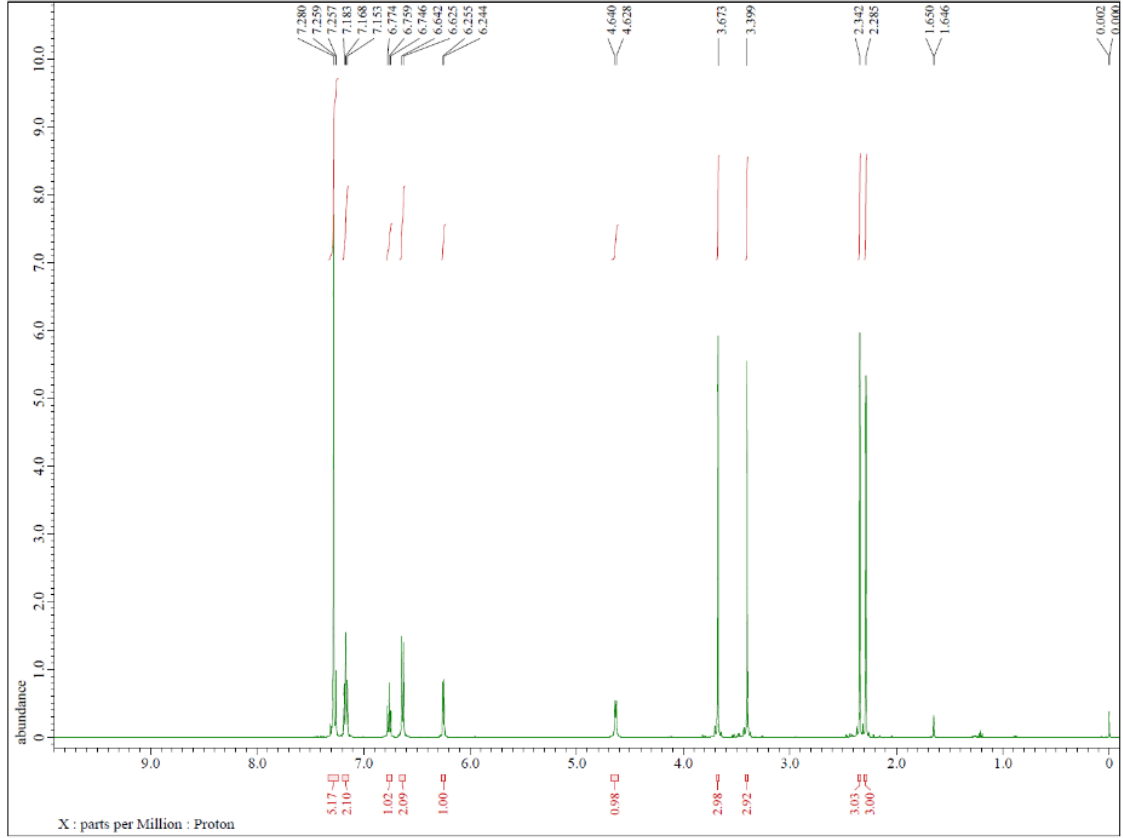


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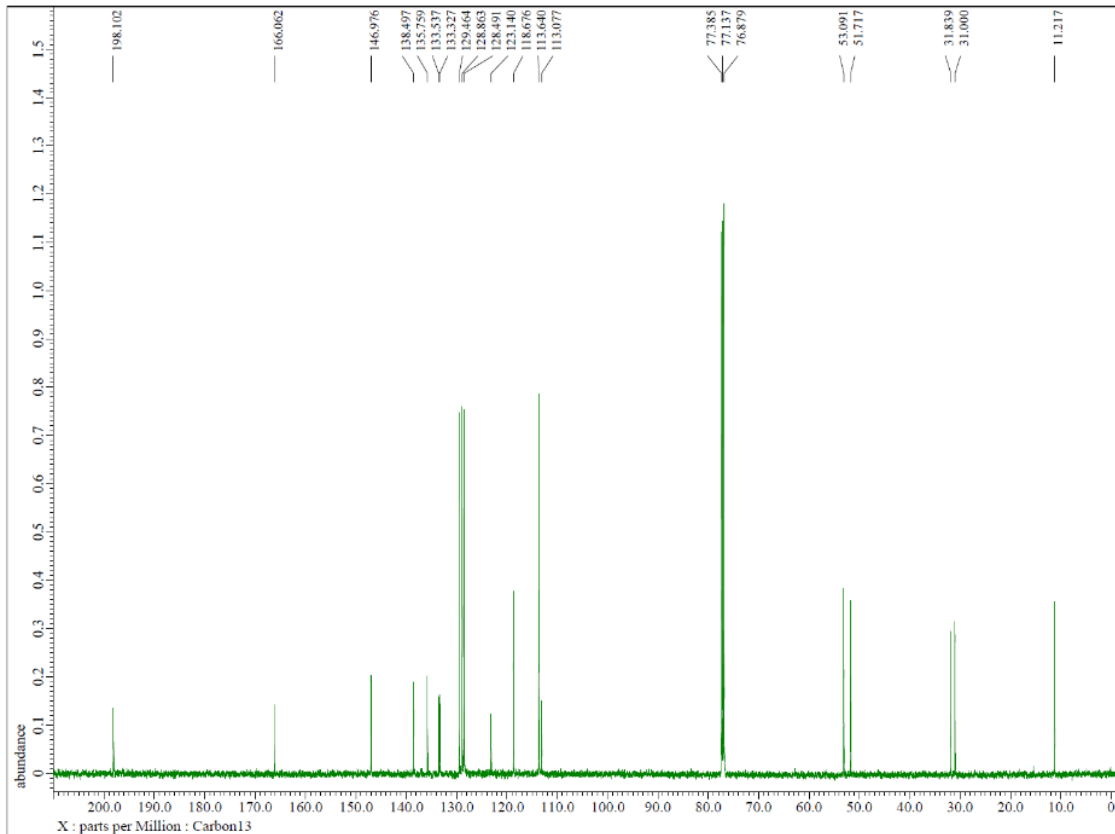


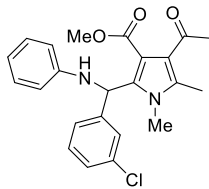


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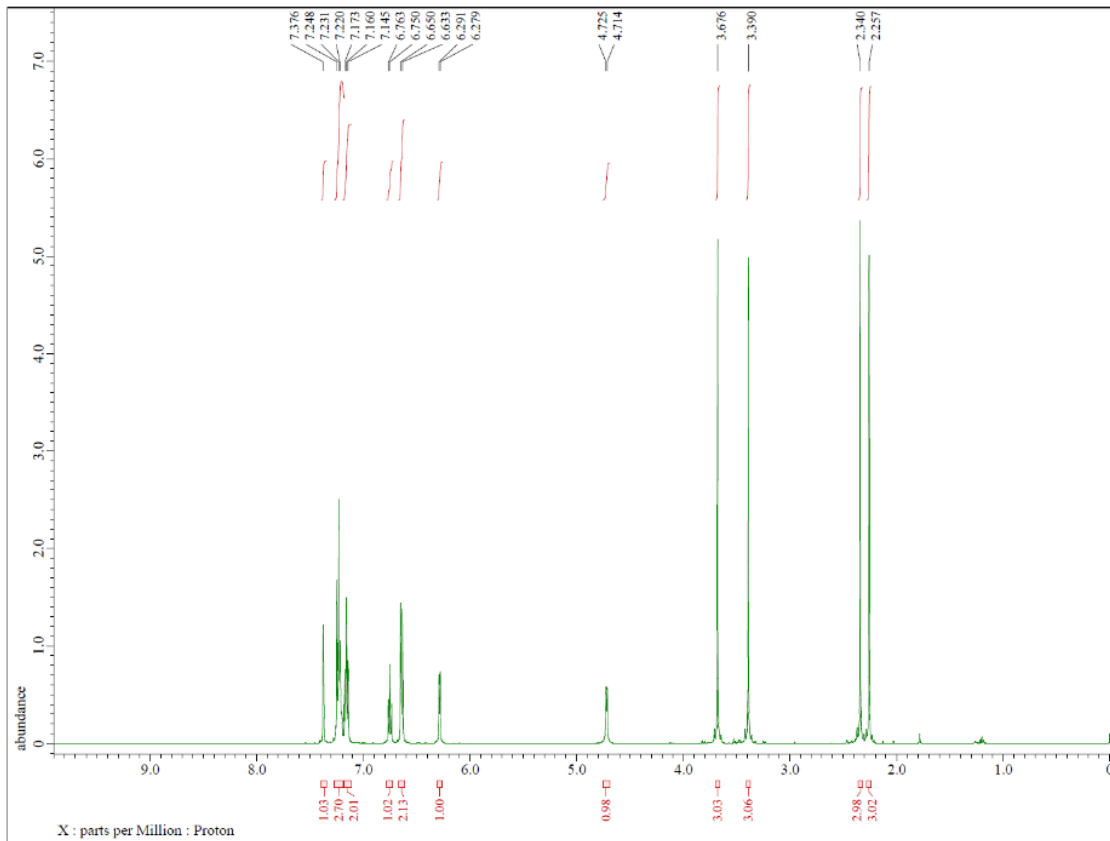


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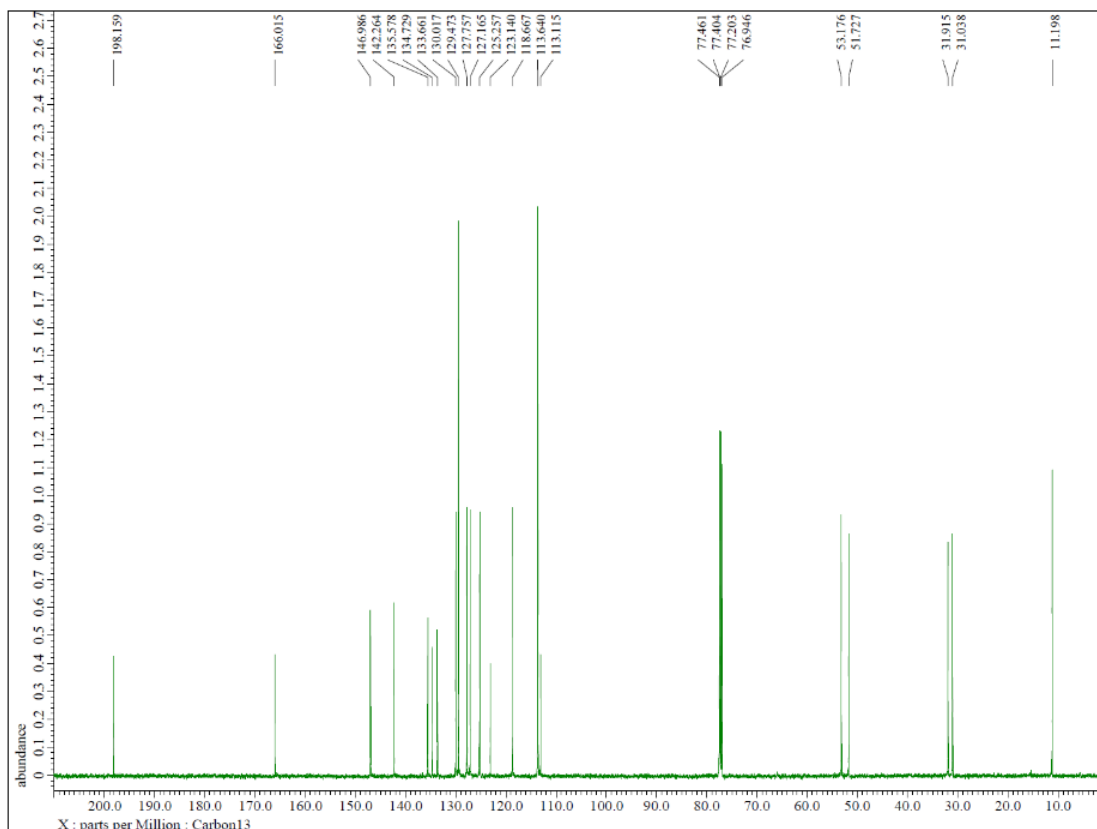


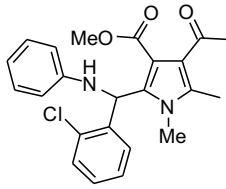


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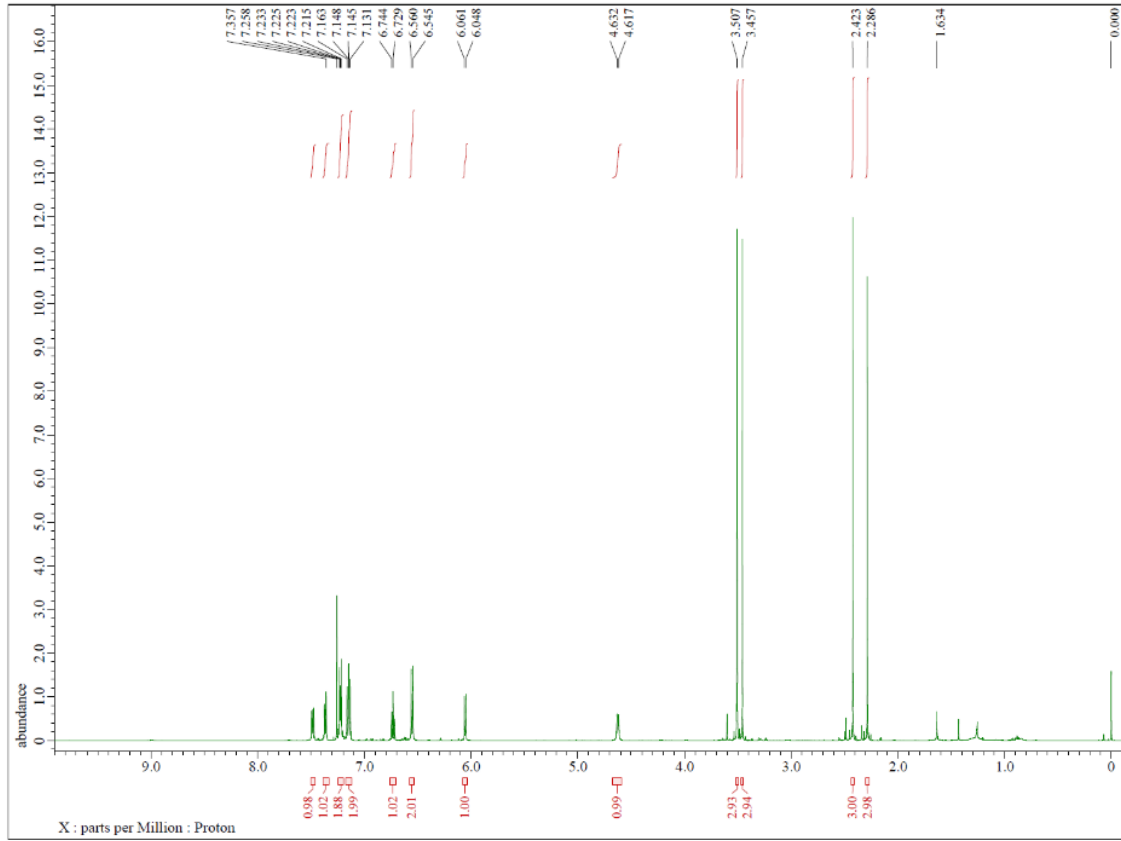


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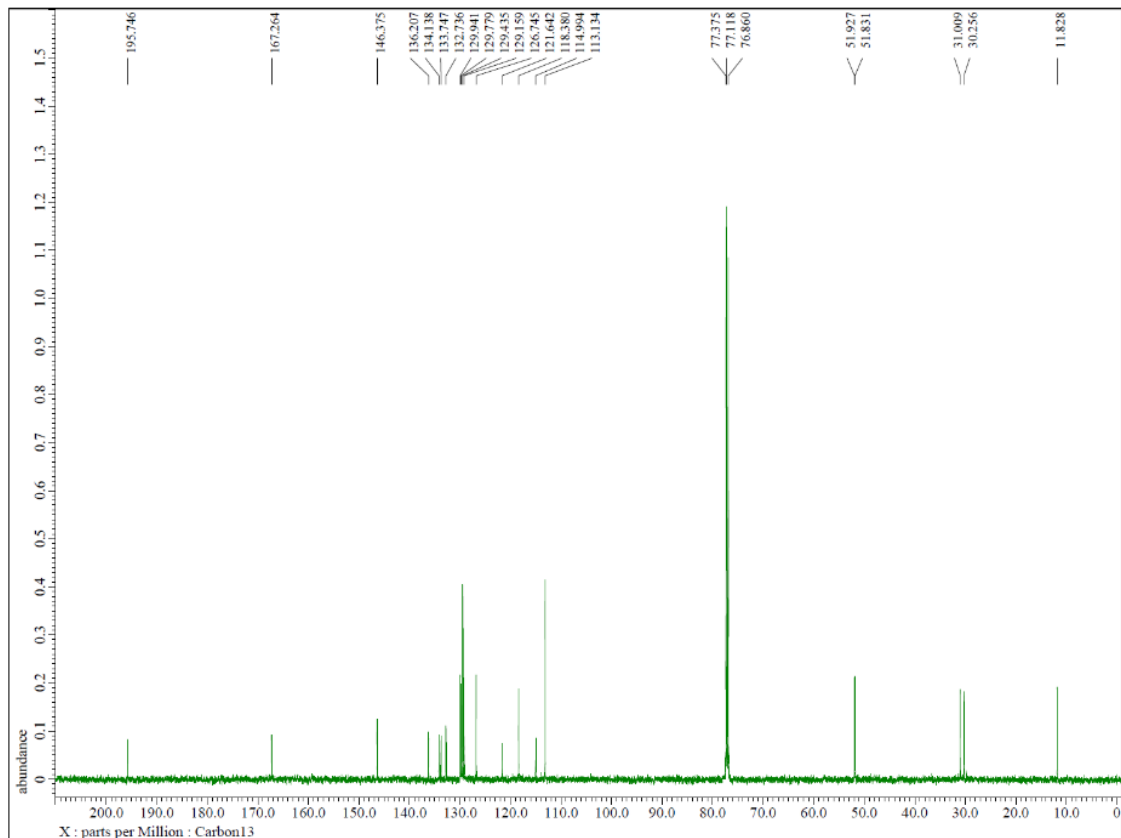


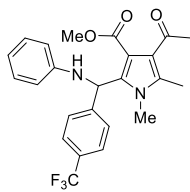


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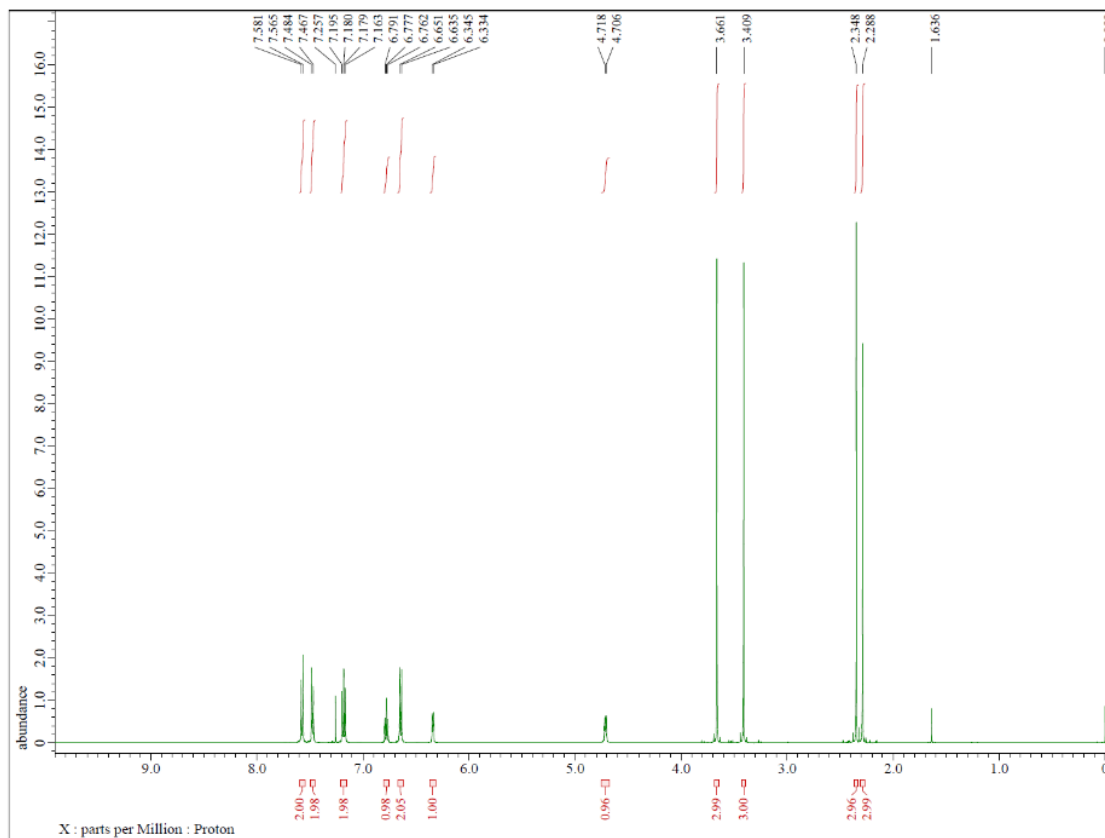


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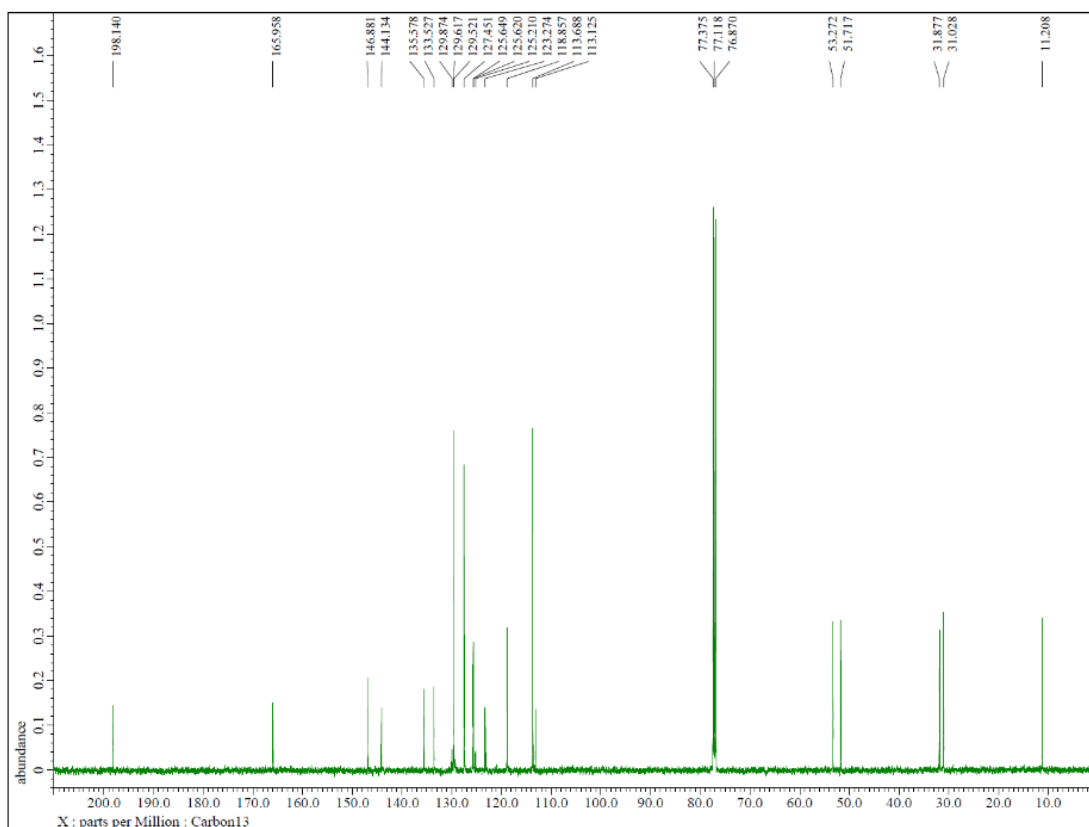


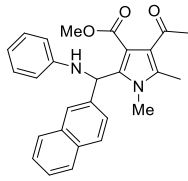


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

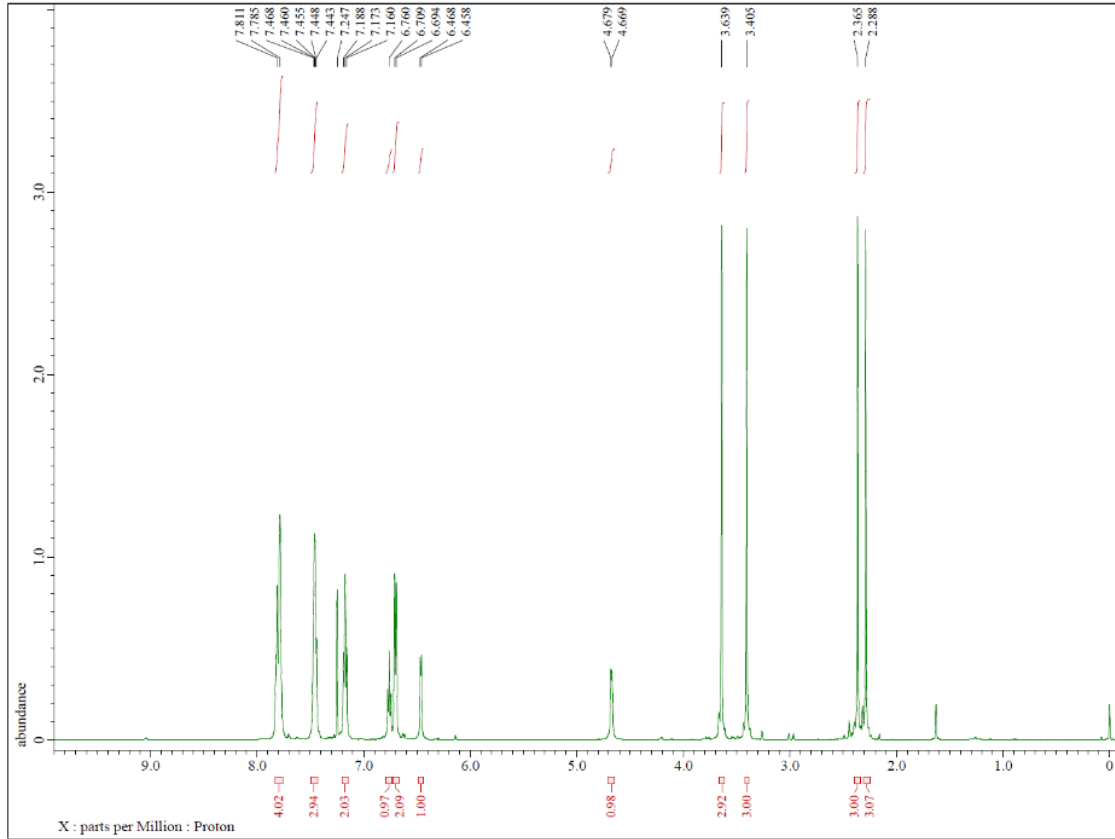


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

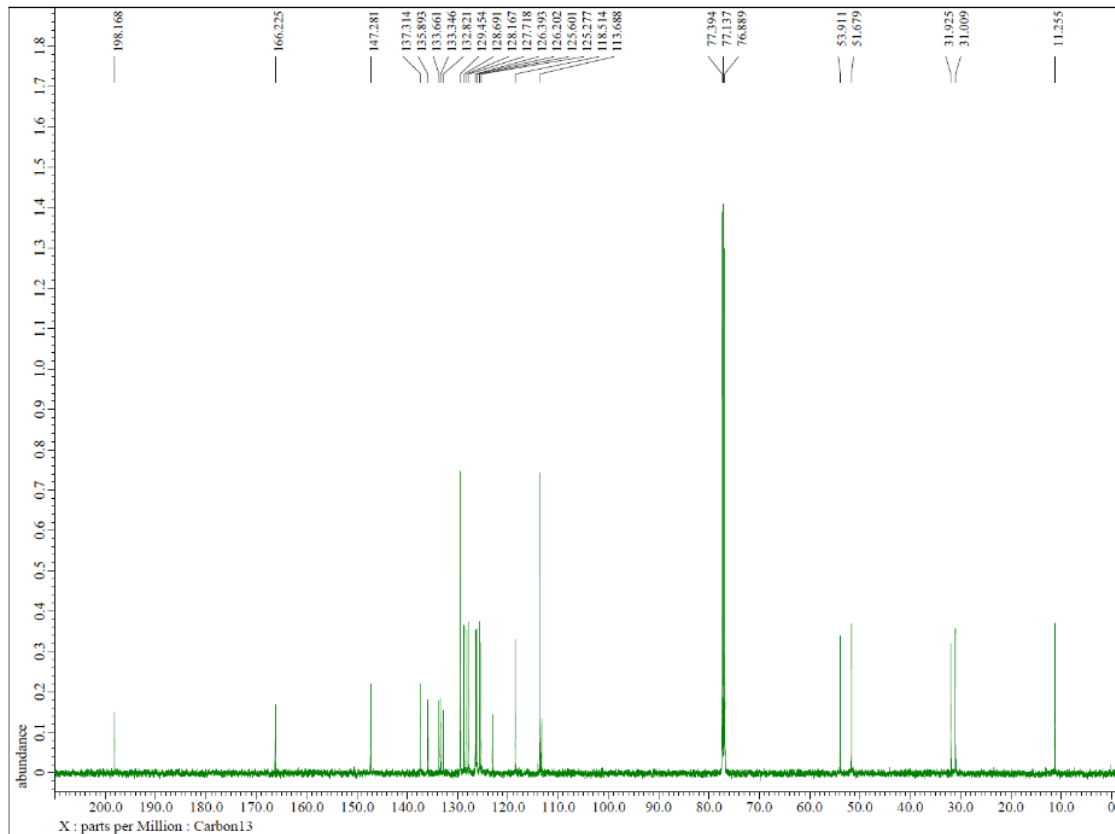


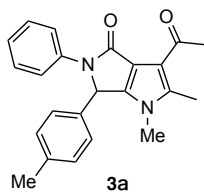


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

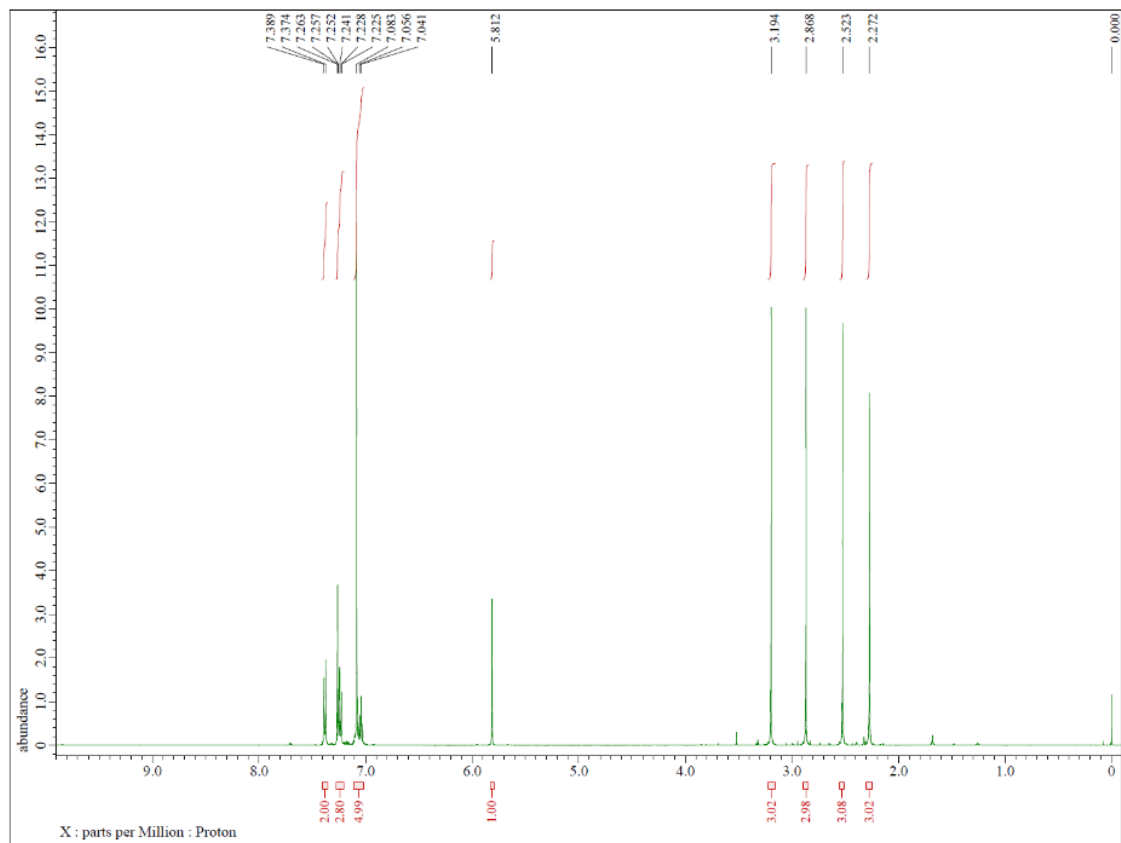


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

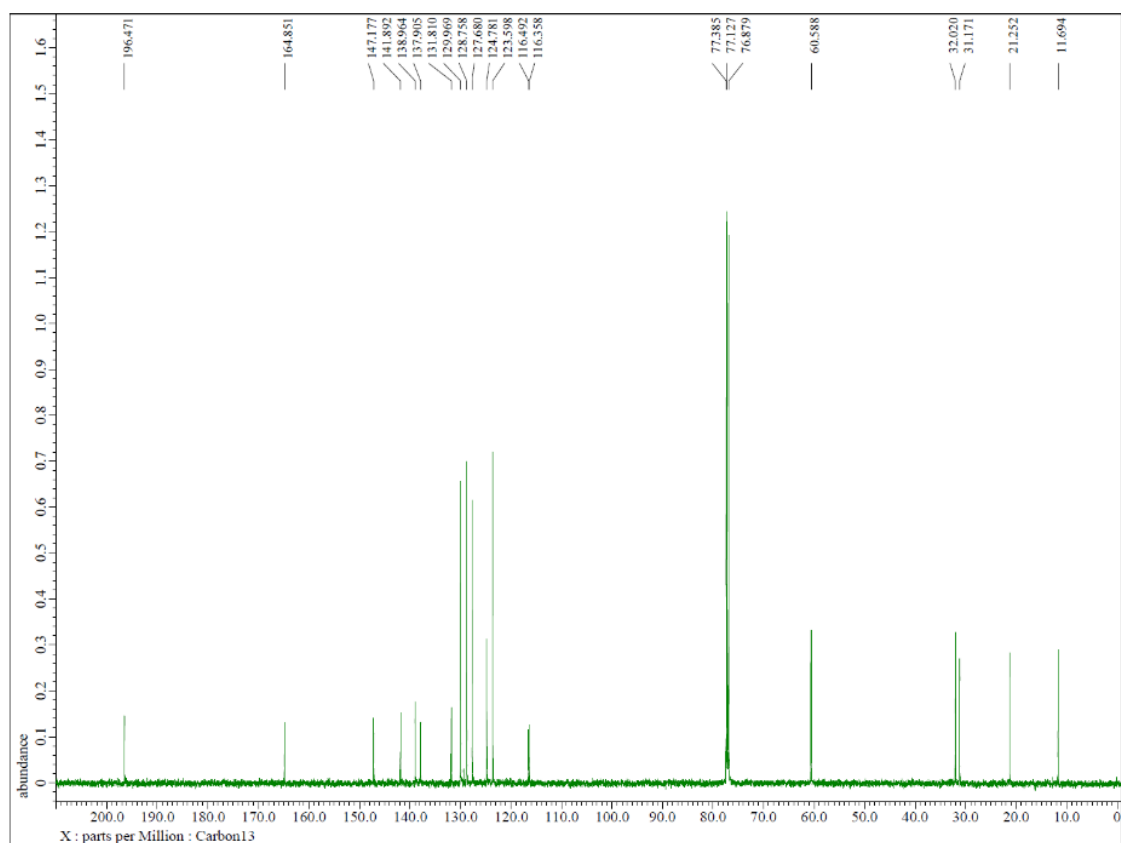


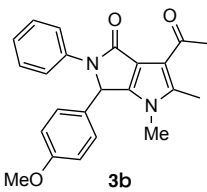


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

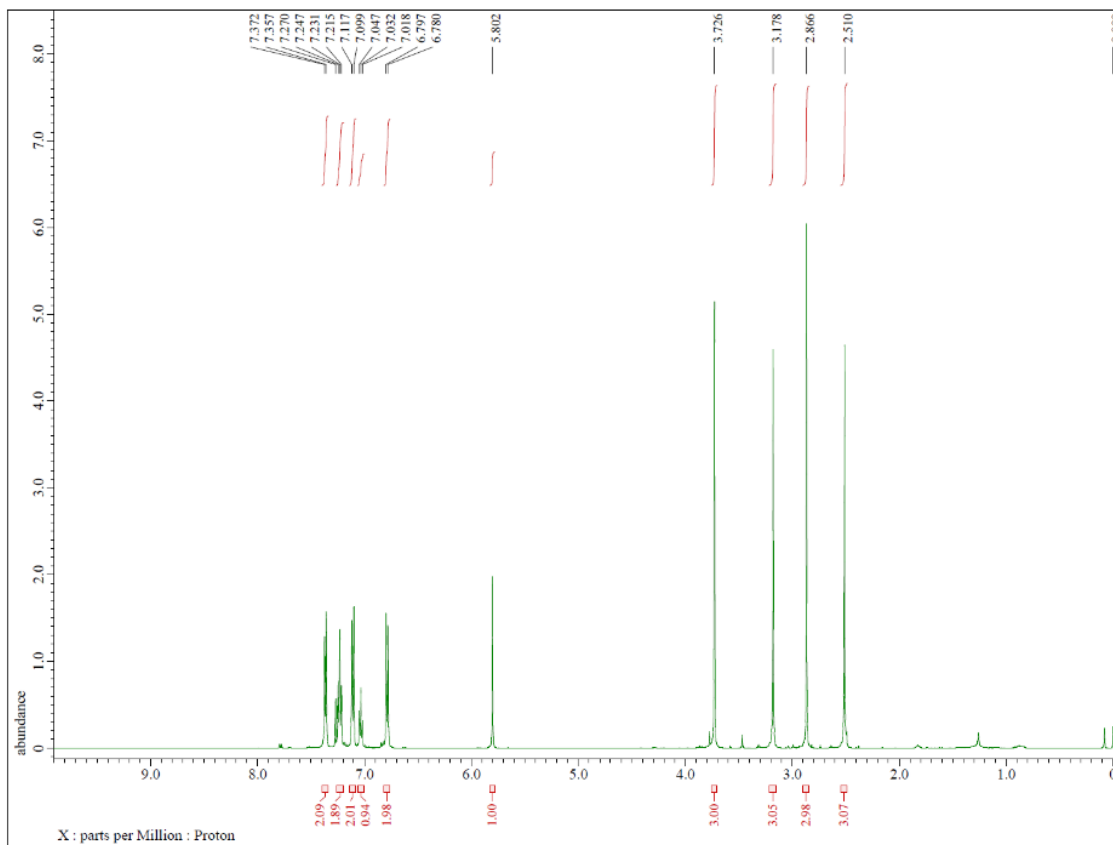


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

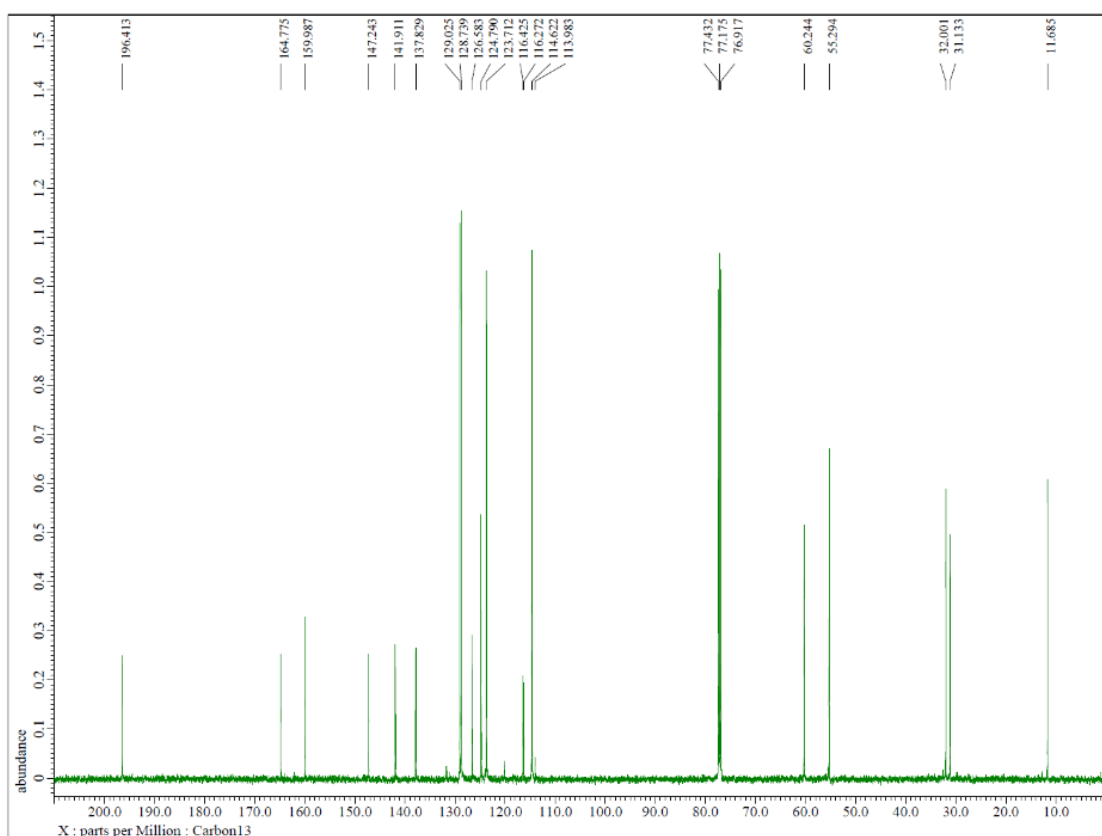


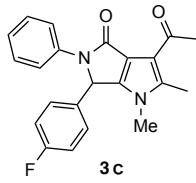


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

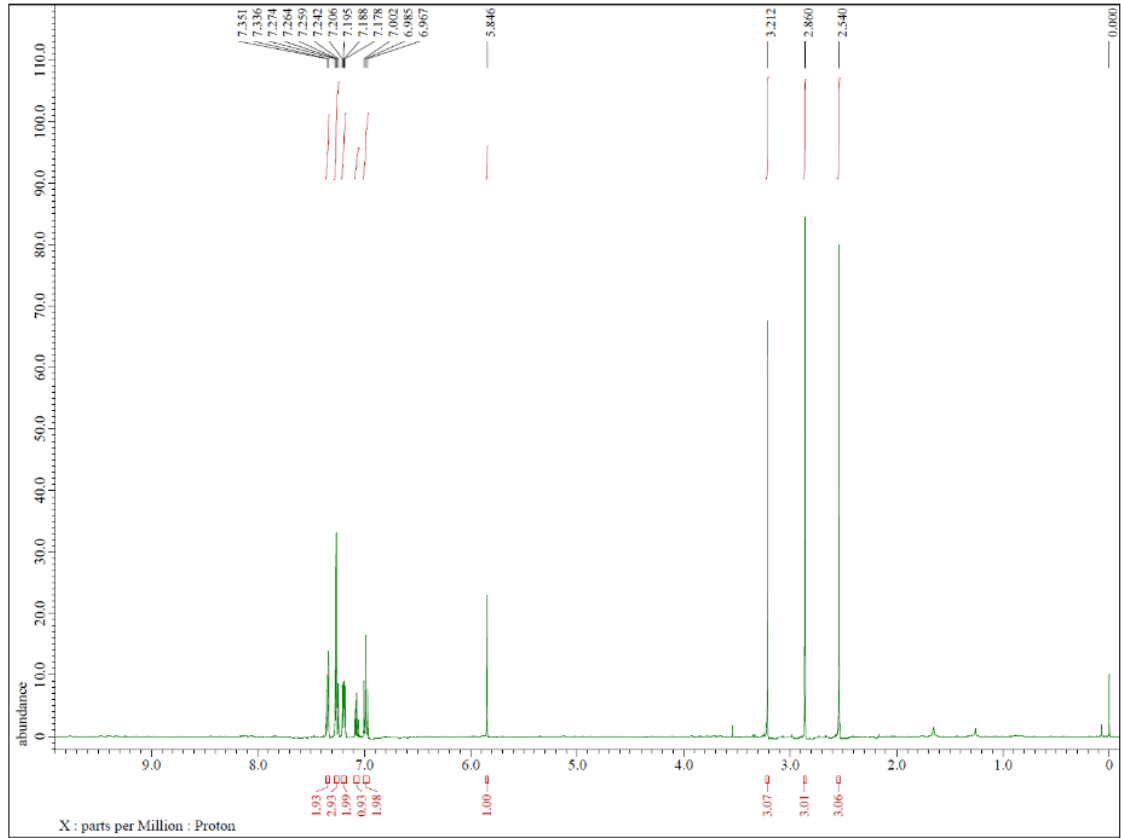


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

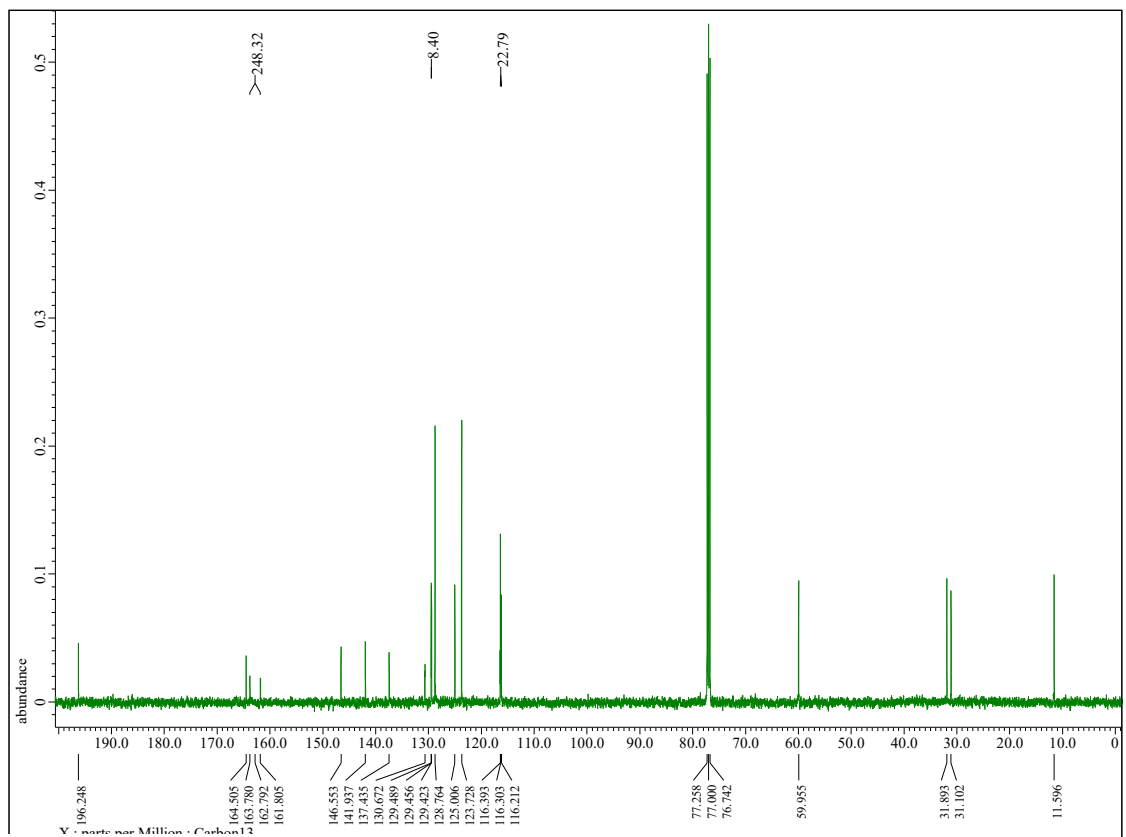


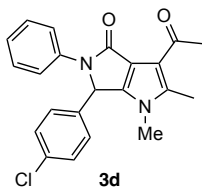


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

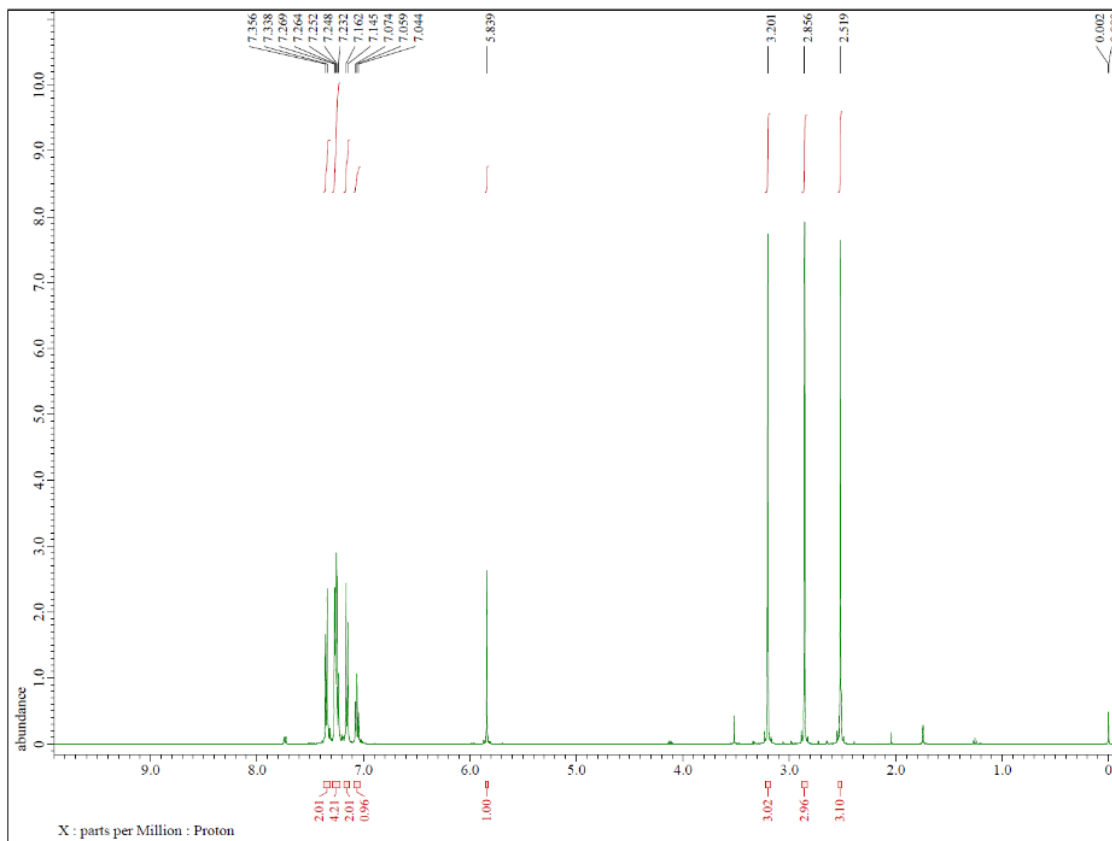


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

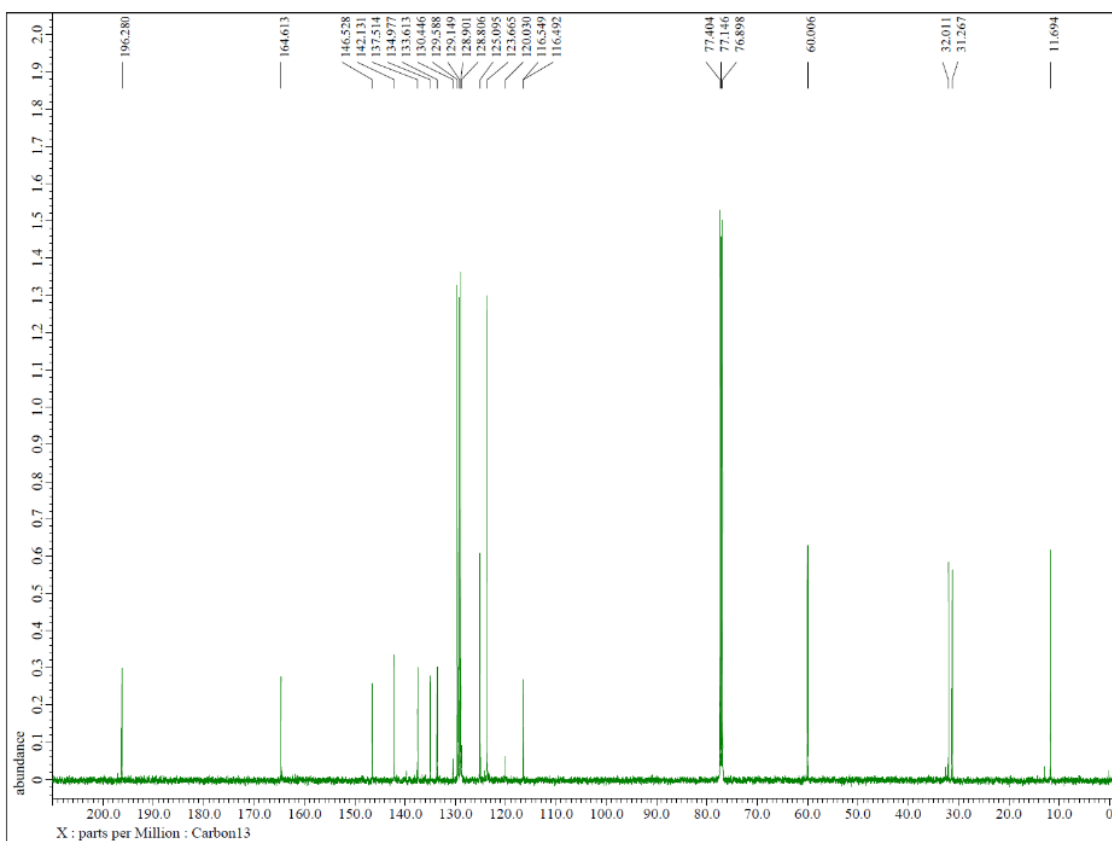


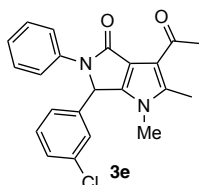


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

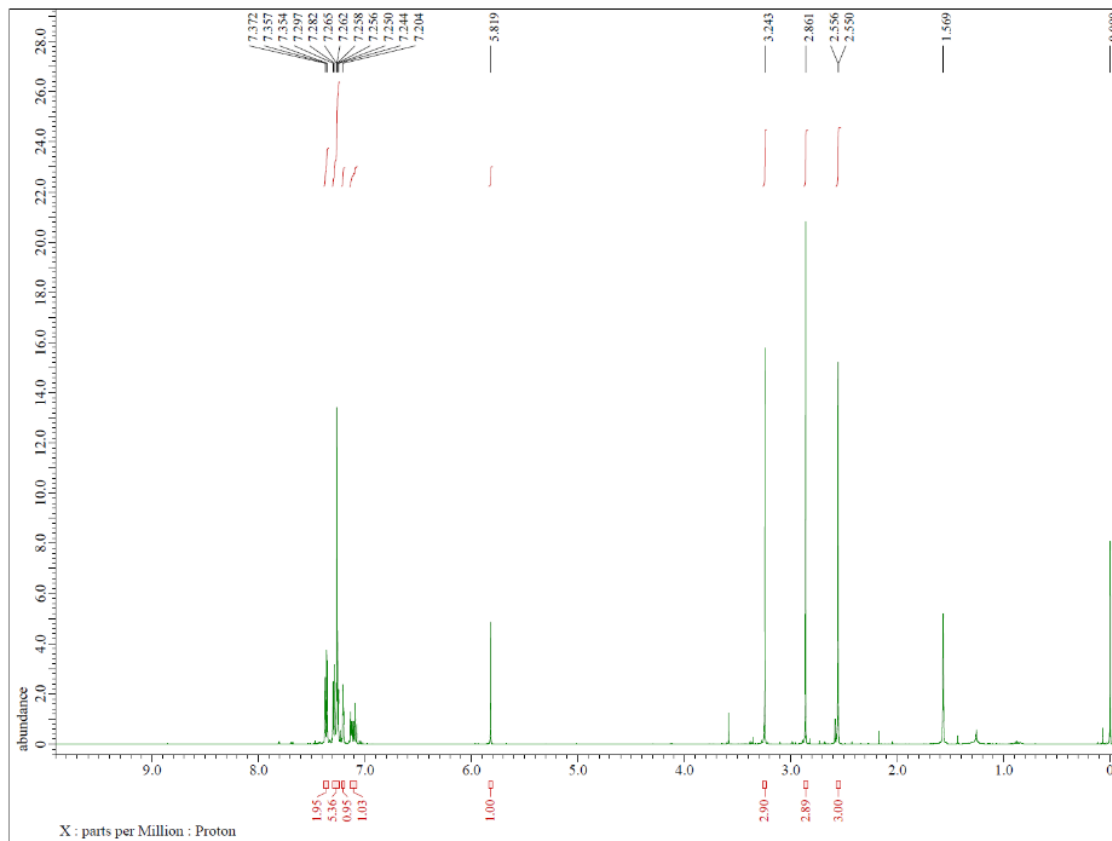


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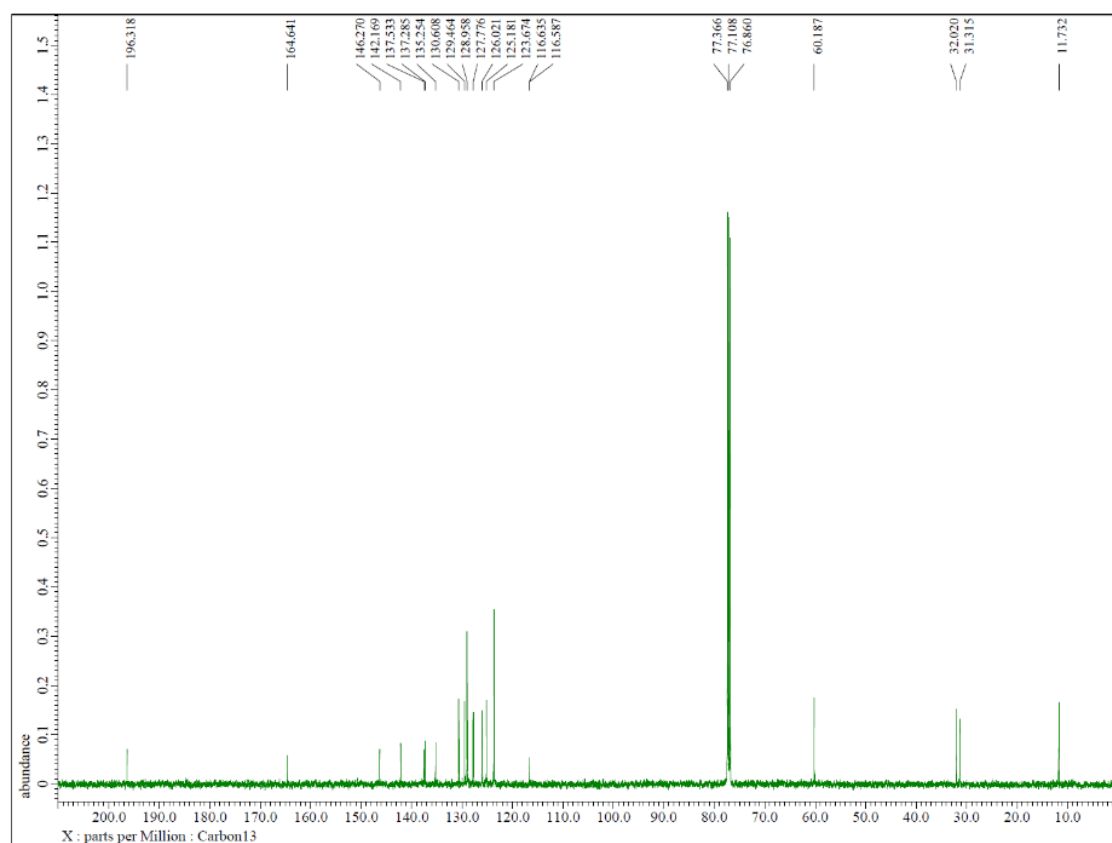


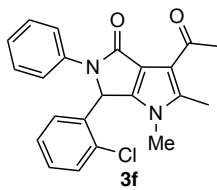


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

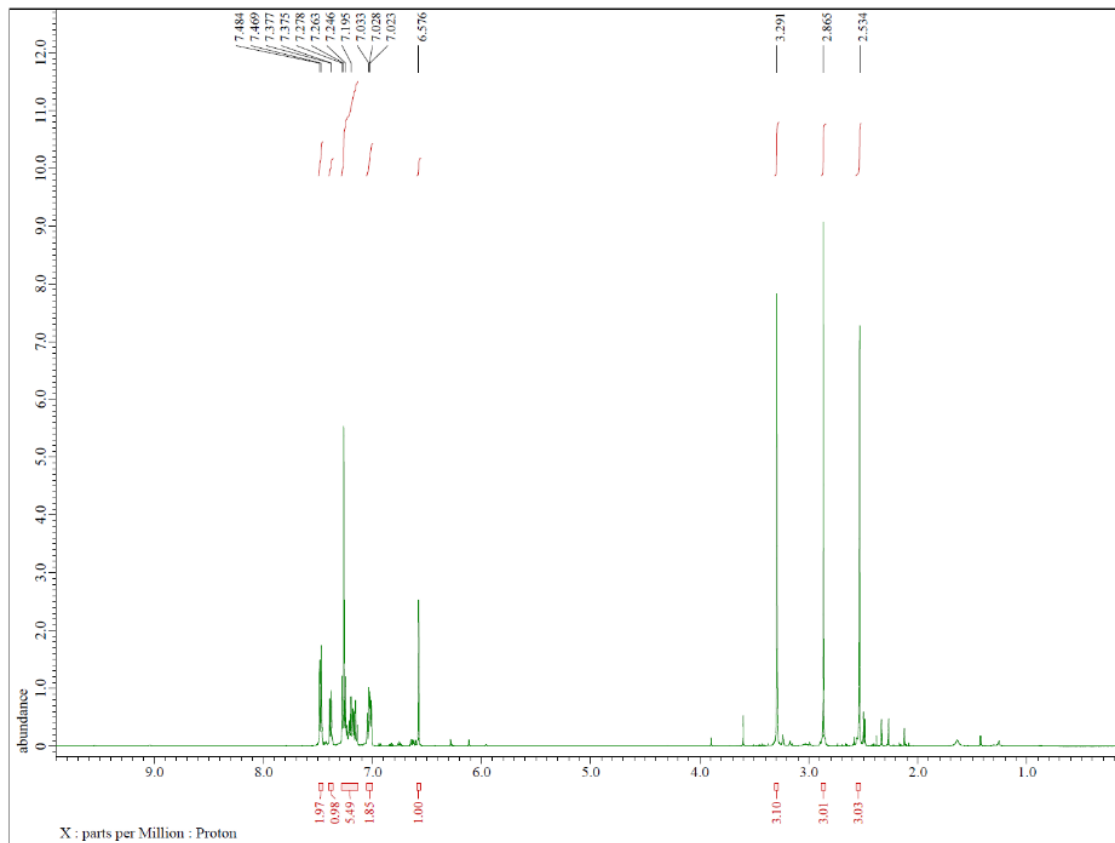


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

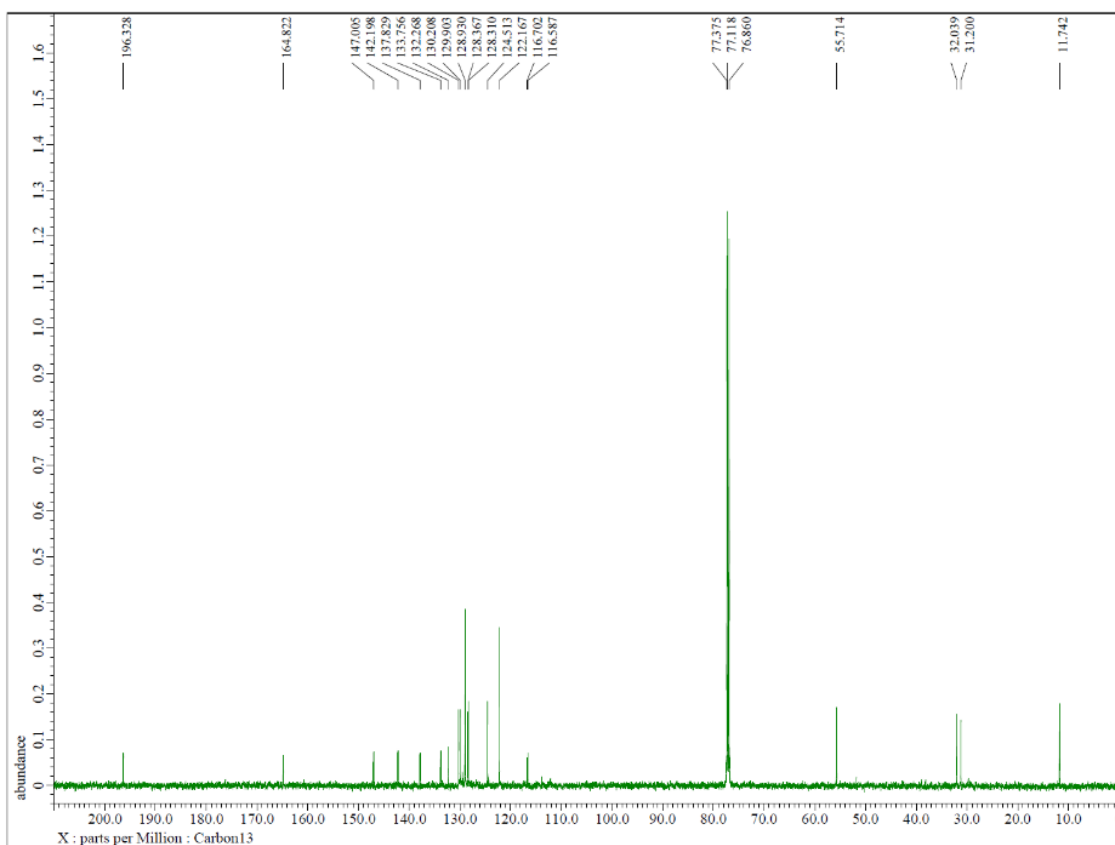


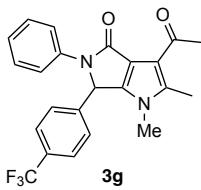


500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$

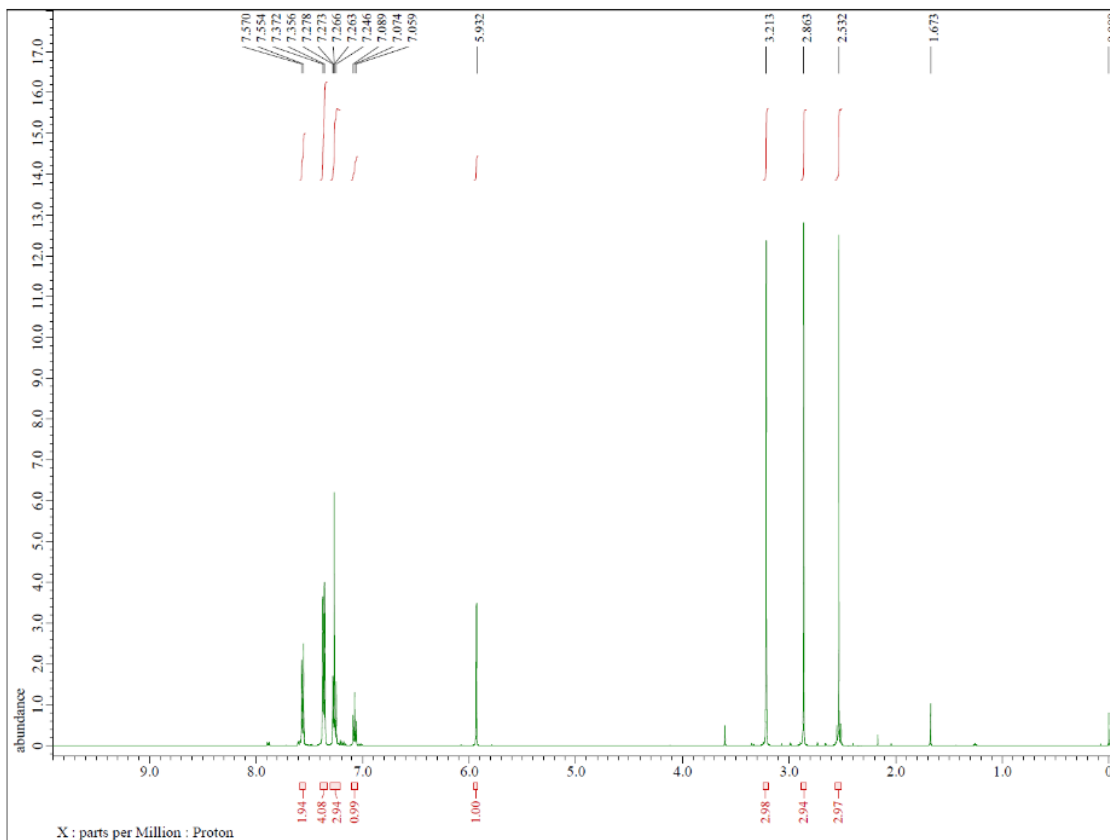


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

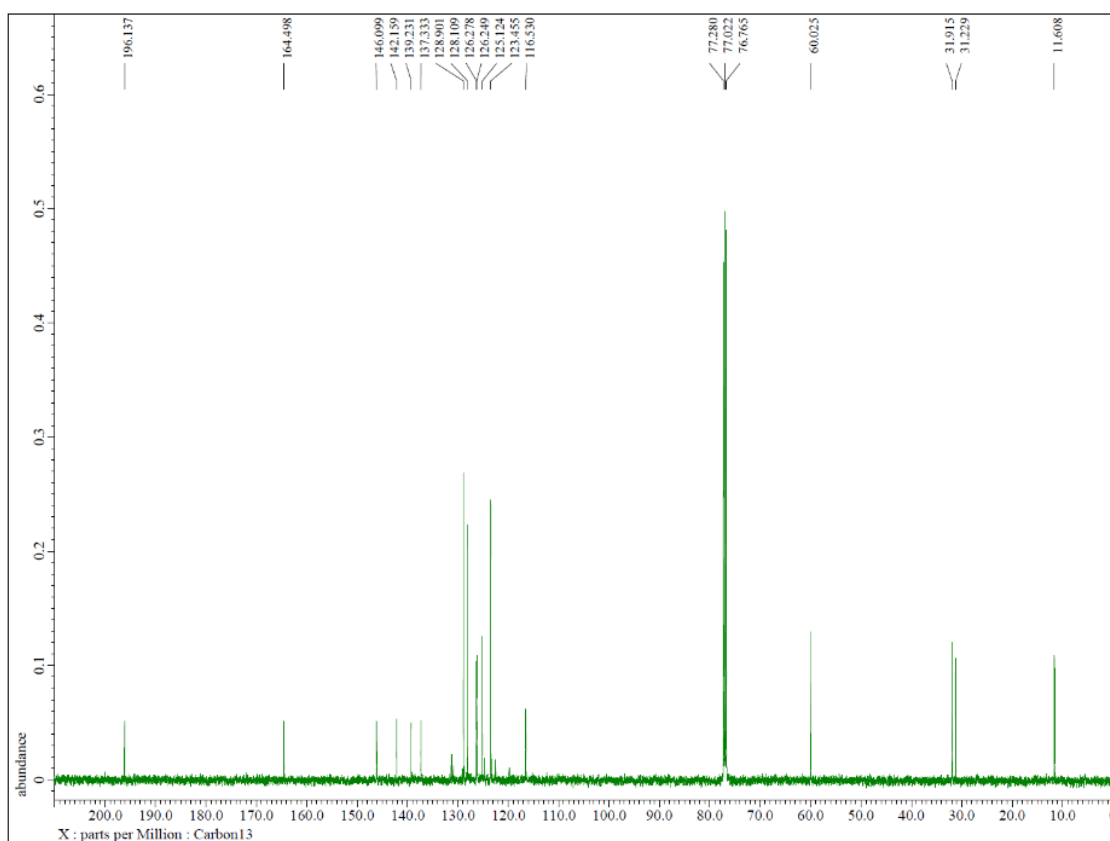


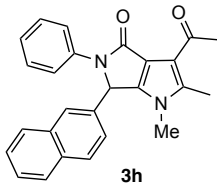


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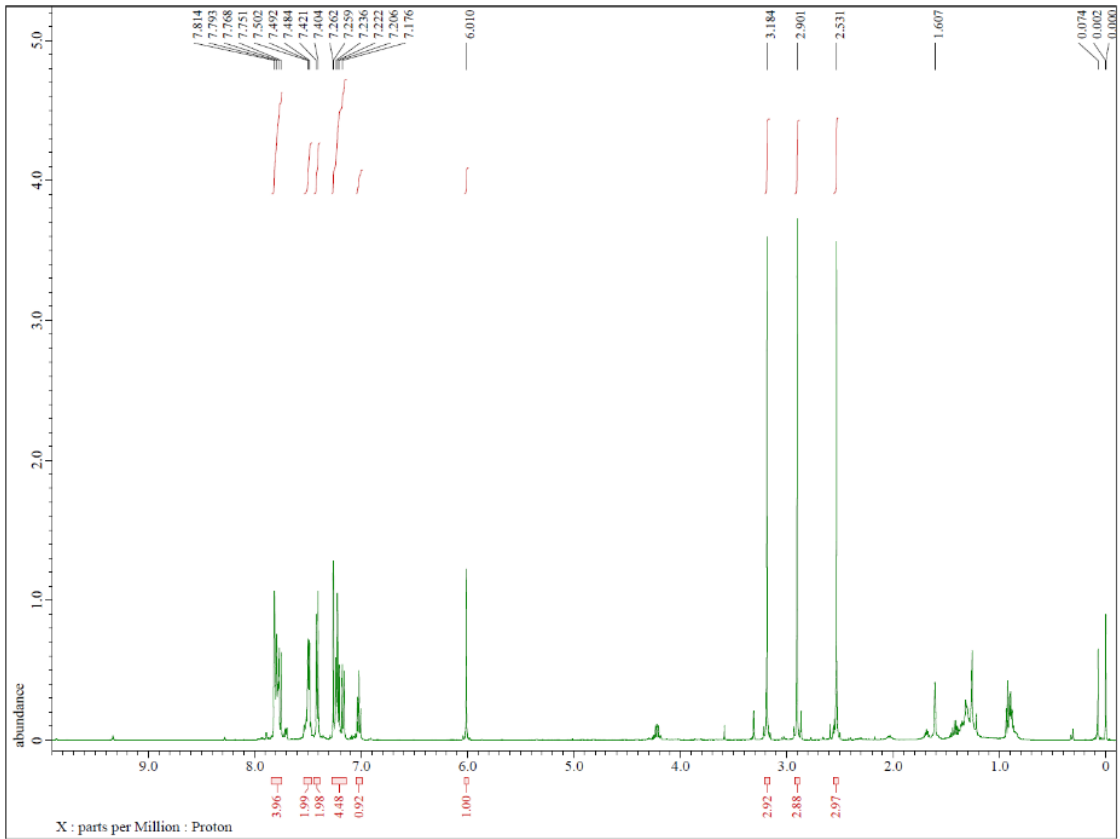


125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$





500 MHz  $^1\text{H}$ NMR in  $\text{CDCl}_3$



125 MHz  $^{13}\text{C}$ NMR in  $\text{CDCl}_3$

