

Three-component synthesis of polysubstituted pyrrole core containing heterocyclic scaffolds over magnetically separable nanocrystalline copper ferrite

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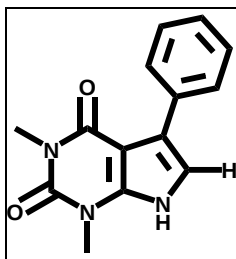
ardchem@caluniv.ac.in, ardas66@rediffmail.com (A R Das)

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1. Materials and Methods

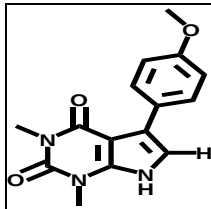
^1H -NMR and ^{13}C -NMR spectral analysis were carried out on Bruker-Advance Digital 300 MHz and 75 MHz instruments; tetramethylsilane (TMS) was used as internal standard. Infrared spectra were recorded in KBr pellets in reflection mode on a Perkin Elmer RX-1 FTIR spectrophotometer. Melting points were determined on a K f ler Block apparatus. Merck aluminum-blocked silica gel plates coated with silica gel G were used for analytical TLC and monitored under UV light and also by exposure to iodine vapor. Synthetic grade chemicals from Sigma-Aldrich, Spectrochem and E-Merck were used for carrying out the organic reactions. All the solvents used in the reaction were distilled and dried properly.

1,3-Dimethyl-5-phenyl-1,7-dihydro-pyrrolo[2,3-d]pyrimidine-2,4-dione (3a):



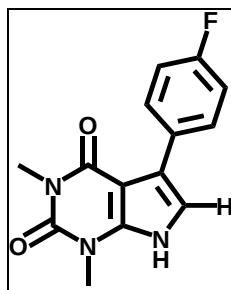
Yield: 89%; Characteristic: Pink powder; Mp: >300 °C; IR (KBr): 3131, 1731, 1610 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 3.17 (3H, s), 3.39 (3H, s), 7.02 (1H, s), 7.16-7.27 (3H, m), 7.74 (2H, d, J=6.9 Hz), 11.84 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 28.2, 30.7, 96.1, 115.6, 122.1, 126.5, 128.2, 128.5, 134.2, 140.7, 150.9, 158.9; Anal. calcd for C₁₄H₁₃N₃O₂: C 65.87, H 5.13, N 16.46 %. Found: C 65.90, H 5.15, N 16.42 %.

5-(4-Methoxy-phenyl)-1,3-dimethyl-1,7-dihydro-pyrrolo[2,3-d]pyrimidine-2,4-dione (3b):



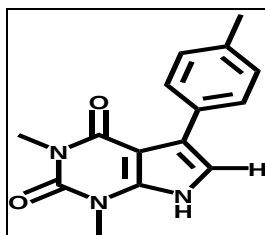
Yield: 92%; Characteristic: Pink powder; Mp: >300 °C; IR (KBr): 3131, 1723, 1609 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 3.01 (3H, s), 3.23 (3H, s), 3.61 (3H, s), 6.74 (1H, s), 6.81 (2H, d, J= 8.1 Hz), 7.83(2H, d, J=8.1 Hz), 11.71 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 27.7, 30.5, 55.4, 95.9, 113.9, 121.8, 126.5, 129.2, 132.7, 140.1, 151.1, 158.5; Anal. calcd for C₁₅H₁₅N₃O₃: C 63.15, H 5.30, N 14.73 %. Found: C 63.13, H 5.34, N 14.70 %.

5-(4-Fluoro-phenyl)-1,3-dimethyl-1,7-dihydro-pyrrolo[2,3-d]pyrimidine-2,4-dione (3c):



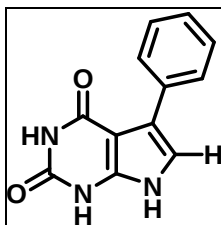
Yield: 88%; Characteristic: Pink powder; Mp: >300 °C; IR (KBr): 3132, 1723, 1621 cm^{-1} ; ^1H NMR (300 MHz, DMSO-d_6): δ 3.05 (3H, s), 3.27 (3H, s), 6.84 (1H, s), 6.92 (2H, d, J = 8.1 Hz), 7.43(2H, d, J =8.1 Hz), 11.77 (1H, s); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.1, 29.9, 96.3, 114.6, 121.0, 128.1, 129.5, 136.2, 150.6, 159.4, 162.6; Anal. calcd for $\text{C}_{14}\text{H}_{12}\text{FN}_3\text{O}_2$: C 61.53, H 4.43, N 15.38 %. Found: C 61.51, H 4.40, N 15.40 %.

1,3-Dimethyl-5-p-tolyl-1,7-dihydro-pyrrolo[2,3-d]pyrimidine-2,4-dione (3d):



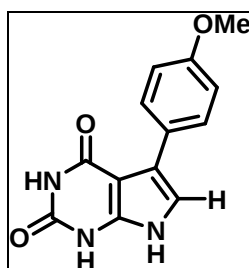
Yield: 92%; Characteristic: Pink powder; Mp: >300 °C; IR (KBr): 3140, 1731, 1600 cm^{-1} ; ^1H NMR (300 MHz, DMSO-d_6): δ 2.21 (3H, s), 3.17 (3H, s), 3.38 (3H, s), 7.05 (1H, s), 7.22 (2H, d, J = 6.9 Hz), 7.63(2H, d, J =6.9 Hz), 11.94 (1H, s); ^{13}C NMR (75 MHz, DMSO-d_6): δ 21.0, 27.7, 30.4, 96.0, 114.7, 127.9, 128.4, 128.8, 128.9, 129.0, 135.9, 140.1, 151.1, 158.6; Anal. calcd for $\text{C}_{15}\text{H}_{15}\text{N}_3\text{O}_2$: C 66.90, H 5.61, N 15.60 %. Found: C 66.88, H 5.59, N 15.63 %.

5-Phenyl-1,7-dihydro-pyrrolo[2,3-d]pyrimidine-2,4-dione(3e):



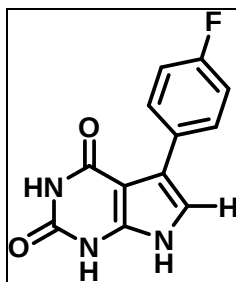
Yield: 91%, Characteristic: Pink powder, Mp: 250 °C (dec), IR (KBr): 3131, 1738, 1663, cm^{-1} ; ^1H NMR (300 MHz, DMSO-d_6): δ 6.84 (1H, s) 7.15-7.78 (5H, m), 10.46 (1H, s), 11.24 (1H, s), 11.49 (1H, s); ^{13}C NMR (75 MHz, DMSO-d_6): δ 96.2, 115.2, 121.4, 126.3, 128.2, 128.3, 134.3, 140.9, 151.1, 160.5; Anal. calcd for $\text{C}_{12}\text{H}_9\text{N}_3\text{O}_2$: C 63.43, H 3.99, N 18.49 %. Found: C 63.41, H 3.96, N 18.51 %; HRMS Calcd for $\text{C}_{12}\text{H}_9\text{N}_3\text{O}_2$ ($[\text{M}+\text{H}]^+$) 228.0774 found : 228.0771

5-(4-Methoxy-phenyl)-1,7-dihydro-pyrrolo[2,3-d]pyrimidine-2,4-dione (3f):



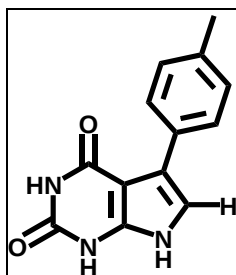
Yield: 91%, Characteristic: Pink powder, Mp: 270 °C (dec), IR (KBr): 3135, 1736, 1663, 1601 cm^{-1} ; ^1H NMR (300 MHz, DMSO-d_6): δ 3.70 (3H, s), 6.74 (1H, s), 6.83 (2H, d, $J=8.4$ Hz), 7.70 (2H, d, $J=8.4$ Hz), 10.42 (1H, s), 11.14 (1H, s), 11.43 (1H, s); ^{13}C NMR (75 MHz, DMSO-d_6): δ 56.7, 96.1, 113.3, 114.1, 117.8, 120.7, 128.9, 142.6, 150.9, 161.9; Anal. calcd for $\text{C}_{13}\text{H}_{11}\text{N}_3\text{O}_3$: C 60.70, H 4.31, N 16.33 %. Found: C 60.73, H 4.30, N 16.35 %; HRMS Calcd for $\text{C}_{13}\text{H}_{11}\text{N}_3\text{O}_3$ ($[\text{M}+\text{H}]^+$) 258.0879 found : 228.0870

5-(4-Fluoro-phenyl)-1,7-dihydro-pyrrolo[2,3-d]pyrimidine-2,4-dione (3g):



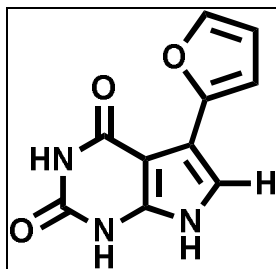
Yield: 86%, Characteristic: Pink powder, Mp: 281 °C (dec), IR (KBr): 3138, 1734, 1604 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 6.85 (1H, s), 7.09 (2H, d, J=7.5Hz), 7.82 (2H, d, J=7.5Hz), 10.48 (1H, s), 11.26 (1H, s), 11.50 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 95.7, 114.5, 114.7, 119.8, 129.5, 129.6, 130.4, 140.5, 150.7, 160.1; Anal. calcd for C₁₂H₈FN₃O₂: C 58.78, H 3.29, N 17.14 %. Found: C 58.80, H 3.27, N 17.18 %; HRMS Calcd for C₁₂H₈FN₃O₂ ([M+H]⁺) 246.068 found : 246.0678

5-p-Tolyl-1,7-dihydro-pyrrolo[2,3-d]pyrimidine-2,4-dione (3h):



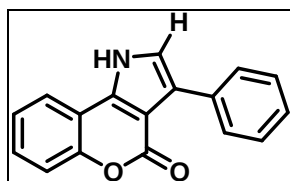
Yield: 88 %; Characteristic: Pink powder; Mp: >300 °C; IR (KBr): 3130, 1727, 1620 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 2.25 (3H, s), 6.85 (1H, s), 7.06 (2H, d, J=7.5 Hz), 7.35 (2H, d, J=7.5 Hz), 10.42 (1H, s), 11.21 (1H, s), 11.46 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 20.5, 96.2, 113.8, 117.6, 127.4, 128.4, 141.9, 152.3, 161.9; Anal. calcd for C₁₃H₁₁N₃O₂: C 64.72, H 4.60, N 17.42 %. Found: C 64.71, H 4.63, N 17.40 %.

5-(furan-2-yl)-1H-pyrrolo[2,3-d]pyrimidine-2,4(3H,7H)-dione (3i):



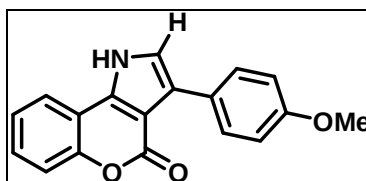
Yield: 78 %; Characteristic: Pink powder; Mp: >300 °C; IR (KBr): 3139, 1720, 1611 cm^{-1} ; ^1H NMR (300 MHz, DMSO-d_6): δ 5.67 (1H, s), 5.86 (1H, d, $J = 2.7$ Hz), 6.13 (1H, t, $J = 2.4$ Hz), 6.85 (1H, s), 10.48 (1H, s), 11.13 (1H, s), 11.71 (1H, s); ^{13}C NMR (75 MHz, DMSO-d_6): δ 96.0, 104.1, 109.6, 113.2, 114.4, 140.5, 150.6, 158.3, 160.2; Anal. calcd for $\text{C}_{10}\text{H}_7\text{N}_3\text{O}_3$: C, 55.30; H, 3.25; N, 19.35 %. Found: C, 55.35; H, 3.22; N, 19.38 %. HRMS of $[\text{C}_{10}\text{H}_7\text{N}_3\text{O}_3 + \text{H}^+]$: Calcd: 218.0560 found: 218.0569.

3-Phenyl-1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4a):



Yield: 89%, Characteristic: Grey amorphous solid, Mp: 295 °C (dec), IR (KBr): 3109, 1679, 1574, 1462 cm^{-1} ; ^1H NMR (300 MHz, DMSO-d_6): δ 7.21-7.44 (6H, m), 7.55 (1H, s), 7.73 (2H, d, $J = 7.5$ Hz), 8.05 (1H, d, $J = 7.5$ Hz), 12.83 (1H, s); ^{13}C NMR (75 MHz, DMSO-d_6): δ 104.6, 113.7, 116.7, 121.3, 122.5, 124.1, 124.5, 126.6, 128.0, 128.6, 128.9, 133.3, 136.5, 151.2, 157.9, 161.9; Anal. calcd for $\text{C}_{17}\text{H}_{11}\text{NO}_2$: C 78.15, H 4.24, N 5.36 %. Found: C 78.14, H 4.29, N 5.38 %. HRMS Calcd for $\text{C}_{17}\text{H}_{11}\text{NO}_2$ ($[\text{M}+\text{H}]^+$) 262.0869 found : 262.0861

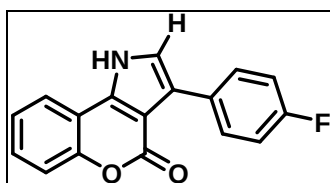
3-(4-Methoxy-phenyl)-1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4b):



Yield: 95%, Characteristic: Grey amorphous solid, Mp: >300 °C, IR (KBr): 3219, 1685 cm⁻¹;

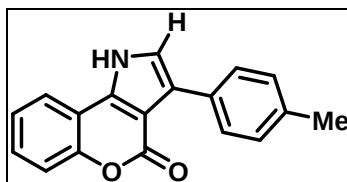
¹H NMR (300 MHz, DMSO-d₆): δ 3.74 (3H, s), 6.92 (2H, d, J= 7.8Hz), 7.32-7.46 (4H, m), 7.66 (2H, d, J= 7.8Hz), 8.03 (1H, d, J= 7.2Hz), 12.74 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 55.6, 104.9, 113.9, 114.2, 117.0, 121.7, 122.0, 124.5, 124.6, 126.2, 129.2, 130.1, 136.6, 142.5, 151.6, 158.4, 158.6, 162.3; Anal. calcd for C₁₈H₁₃NO₃: C 74.22, H 4.50, N 4.81 %. Found: C 74.26, H 4.48, N 4.78 %. HRMS Calcd for C₁₈H₁₃NO₃ ([M+H]⁺) 292.0974 found : 292.0970

3-(4-Fluoro-phenyl)-1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4c):



Yield: 85%; Characteristic: Grey amorphous solid; Mp: >300 °C; IR (KBr): 3113, 1678 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 7.13-7.37 (5H, m), 7.52 (1H, s), 7.75 (2H, s), 8.02 (1H, d, J= 6.9 Hz), 12.79 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 104.6, 113.8, 114.7, 114.9, 116.7, 121.4, 122.4, 123.4, 124.2, 128.9, 129.8, 130.4, 130.5, 136.5, 151.2, 158.0, 159.7, 162.9; Anal. calcd for C₁₇H₁₀FNO₂: C 73.11, H 3.61, N 5.02 %. Found: C 73.18, H 3.64 N 5.00 %. HRMS Calcd for C₁₇H₁₀FNO₂ ([M+H]⁺) 280.0775 found : 280.0778

3-*p*-Tolyl-1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4d):

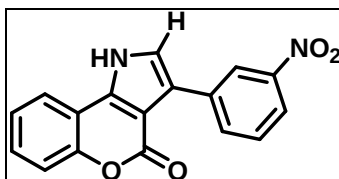


Yield: 87%, Characteristic: Yellow amorphous solid, Mp: >300 °C, IR (KBr): 3185, 1690 cm⁻¹;

¹H NMR (300 MHz, DMSO-d₆): δ 2.28 (3H, s), 7.15 (2H, d, J=7.8Hz), 7.30-7.45 (3H, m), 7.49 (1H, s), 7.62 (2H, d, J= 7.5 Hz), 8.03 (1H, d, J= 7.5 Hz), 12.76 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 20.8, 104.6, 113.8, 116.7, 121.3, 122.0, 124.1, 124.5, 128.5, 128.8, 130.5, 135.6, 136.3, 151.2, 157.9; Anal.

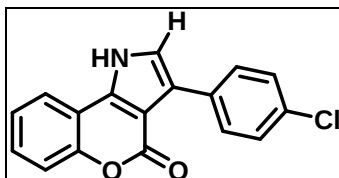
calcd for $C_{18}H_{13}NO_2$: C 78.53, H 4.76, N 5.09 %. Found: C 78.59, H 4.73, N 5.05 %. ; HRMS Calcd for $C_{18}H_{13}NO_2$ ($[M+H]^+$) 276.1025 found : 276.1019

5. 3-(3-Nitro-phenyl) -1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4e):



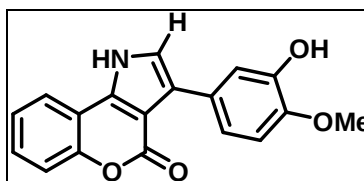
Yield: 80%, Characteristic: Yellow amorphous solid, Mp: 290 °C (dec), IR (KBr): 3117, 1678 cm^{-1} ; 1H NMR (300 MHz, DMSO- d_6): δ 7.36-7.64 (4H, m), 7.83 (1H, s), 8.05-8.22 (3H, m), 8.69 (1H, s), 13.03 (1H, s); ^{13}C NMR (75 MHz, DMSO- d_6): δ 113.5, 116.8, 117.8, 121.2, 121.9, 122.7, 123.9, 124.3, 129.5, 134.7, 135.0, 137.1, 147.9, 151.2, 158.0, 161.9; Anal. calcd for $C_{17}H_{10}N_2O_4$: C 66.67, H 3.29, N 9.15 %. Found: C 66.63, H 3.30, N 9.19 %; HRMS Calcd for $C_{17}H_{10}N_2O_4$ ($[M+H]^+$) 307.0720 found : 307.0728

3-(4-Chloro-phenyl) -1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4f):



Yield: 85%, Characteristic: Yellow amorphous solid, Mp: 296 °C (dec), IR (KBr): 3114, 1684 cm^{-1} ; 1H NMR (300 MHz, DMSO- d_6): δ 7.31-7.41 (5H, m), 7.60 (1H, s), 7.77 (2H, d, J = 8.1Hz), 8.03 (1H, d, J = 7.5 Hz), 12.86 (1H, s); ^{13}C NMR (75 MHz, DMSO- d_6): δ 104.5, 113.6, 116.7, 121.3, 122.8, 122.9, 124.1, 127.9, 129.0, 130.1, 131.1, 132.2, 136.7, 151.2, 157.9; Anal. Calcd for $C_{17}H_{10}ClNO_2$: C 69.05, H 3.41, N 4.74%. Found: C 69.02, H 3.48, N 4.77%; HRMS Calcd for $C_{17}H_{10}ClNO_2$ ($[M+H]^+$) 296.0479 found : 296.0471

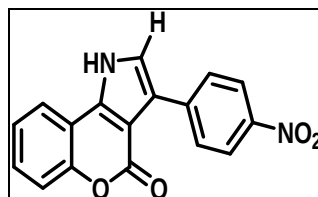
3-(3-Hydroxy-4-methoxy-phenyl)-1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4g):



Yield: 84%, Characteristic: Yellow amorphous solid, Mp: >300 °C, IR (KBr): 3134, 1657 cm⁻¹;

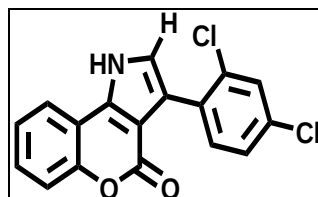
¹H NMR (300 MHz, DMSO-d₆): δ 3.75 (3H, s), 6.89 (1H, d, J=8.1), 7.11-7.55 (6H, m), 8.03 (1H, d, J=7.5Hz), 8.85 (1H, s), 12.71 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 56.2, 104.7, 112.4, 114.2, 116.7, 117.0, 120.0, 121.7, 122.1, 124.5, 125.0, 126.7, 129.2, 136.5, 146.4, 147.2, 151.5, 158.3, 162.3; Anal. calcd for C₁₈H₁₃NO₄: C 70.35, H 4.26, N 4.56 %. Found: C 70.38, H 4.28, N 4.59 %; HRMS Calcd for C₁₈H₁₃NO₄ ([M+H]⁺) 308.0924 found : 308.0928

3-(4-Nitro-phenyl)-1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4h):



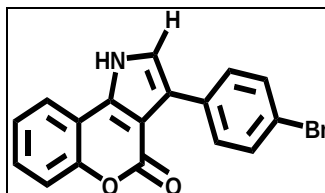
Yield: 85%, Characteristic: Yellow amorphous solid, Mp: 290 °C (dec), IR (KBr): 3150, 1682 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 7.30-7.37 (4H, m) 7.73 (1H, s), 8.03-8.15 (4H, m), 12.98 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 104.7, 113.8, 116.8, 118.8, 121.2, 121.4, 121.9, 122.7, 123.9, 124.9, 129.3, 129.5, 134.7, 135.0, 137.1, 147.9, 151.5, 157.7, 161.7; Anal. calcd for C₁₇H₁₀N₂O₄: C 66.67, H 3.29, N 9.15 %. Found: C 66.63, H 3.30, N 9.19 %; HRMS Calcd for C₁₇H₁₀N₂O₄ ([M+H]⁺) 307.072 found : 307.0719.

3-(2,4-Dichloro-phenyl)-1H-5-oxa-1-aza-cyclopenta[a]naphthalene-4-one (4i):



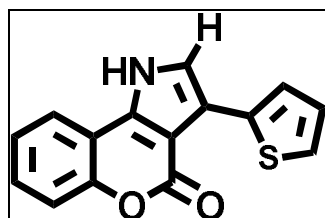
Yield: 90%; Characteristic: White amorphous solid; Mp: 292 °C (dec); IR (KBr): 3219, 1683 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 7.31-7.49 (7H, m), 7.97 (1H, s), 12.80 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 106.9, 114.1, 117.3, 120.8, 121.8, 124.6, 127.9, 129.5, 129.8, 131.5, 132.1, 135.6, 151.7, 157.7; Anal. calcd for C₁₇H₉Cl₂NO₂: C 61.84, H 2.75, N 4.24 %. Found: C 61.80, H 2.72, N 4.27 %.

3-(4-Bromo-phenyl)-1H-5-oxa-1-aza-cyclopenta[a]naphthalen-4-one (4j):

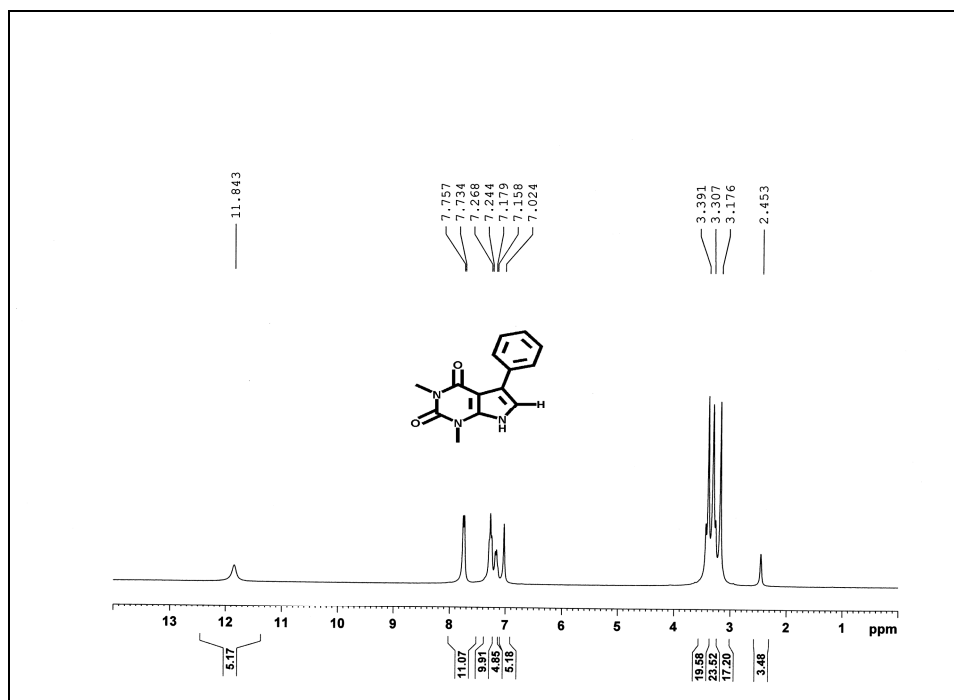


Yield: 87%; Characteristic: White amorphous solid; Mp: 290 °C (dec); IR (KBr): 3220, 1681 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 7.33-7.72 (8H, m), 8.03 (1H, d, J = 6.6 Hz), 12.87 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 104.5, 113.7, 116.7, 119.8, 121.4, 122.9, 123.1, 124.2, 129.1, 130.9, 132.6, 136.7, 151.2, 157.9; Anal. calcd for C₁₇H₁₀BrNO₂: C 60.02, H 2.96, N 4.12 %. Found: C 60.00, H 2.98, N 4.10 %.

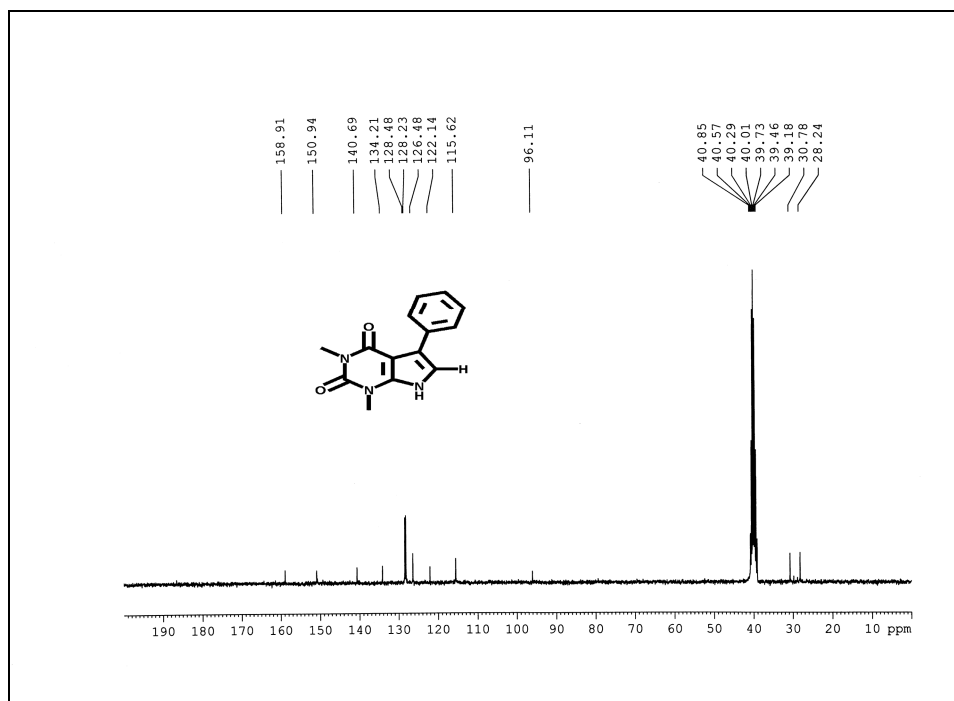
3-(thiophen-2-yl)chromeno[4,3-b]pyrrol-4(1H)-one (4k):



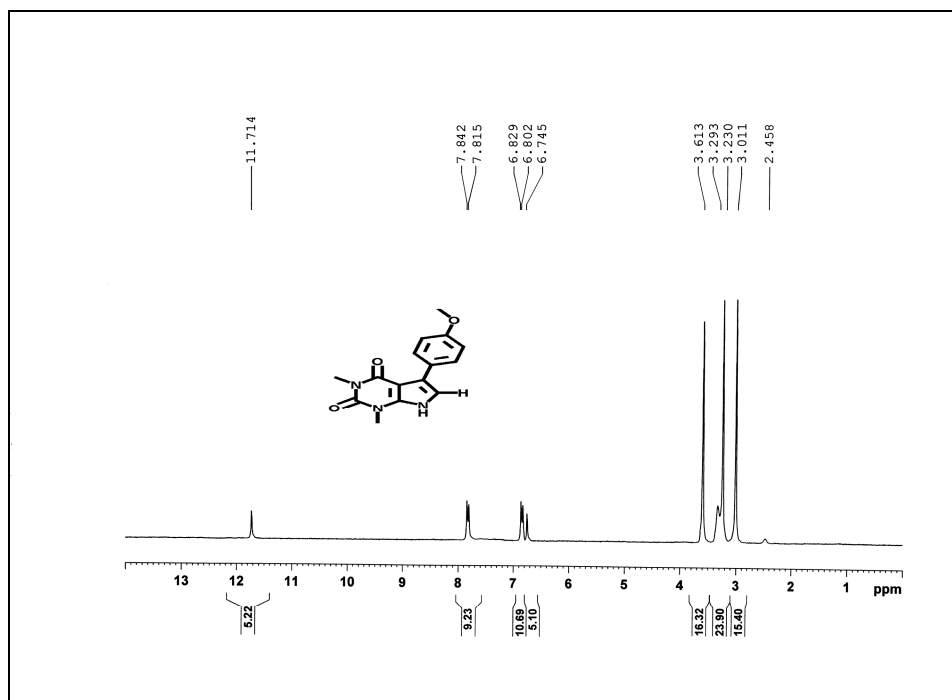
Yield: 81%; Characteristic: Brown amorphous solid; Mp: 282 °C (dec); IR (KBr): 3215, 1680 cm⁻¹; ¹H NMR (300 MHz, DMSO-d₆): δ 7.04 (1H, s), 7.30-7.47 (4H, m), 7.61 (1H, s), 7.71 (1H, s), 8.05 (1H, d, J = 7.5 Hz), 12.89 (1H, s); ¹³C NMR (75 MHz, DMSO-d₆): δ 104.5, 113.9, 117.1, 118.0, 121.8, 122.3, 124.6, 124.7, 126.6, 127.9, 129.5, 135.5, 136.8, 151.6, 158.2. Anal. calcd for C₁₅H₉NO₂S : C 67.40, H 3.39, N 5.24%. Found: C 67.42, H 3.32, N 5.26%. HRMS of [C₁₅H₉NO₂S + H⁺]: Calcd: 268.0427 found: 268.0420.



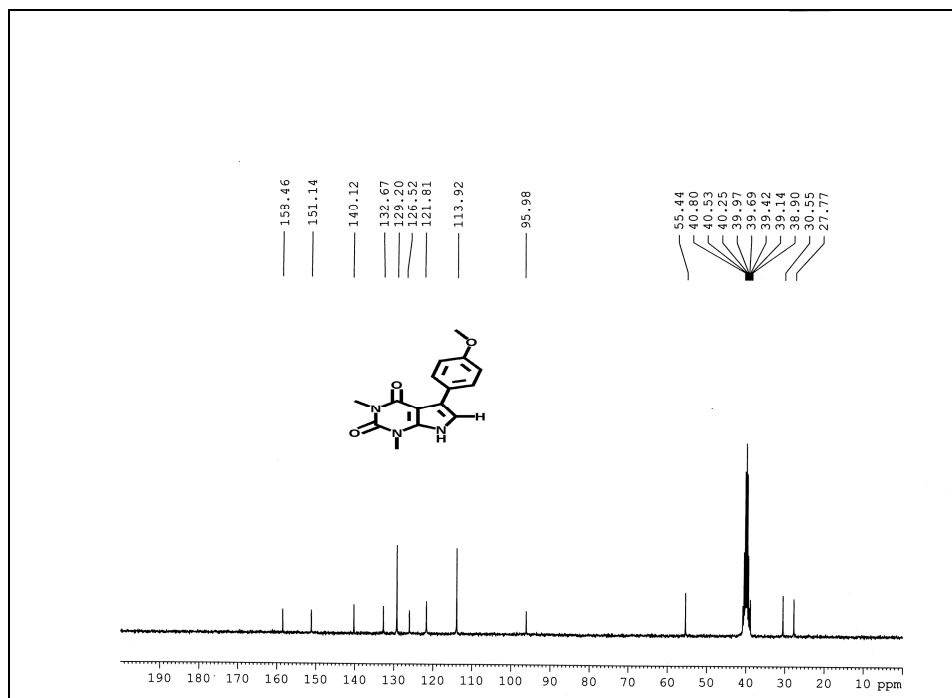
¹H NMR spectrum of the product (3a)



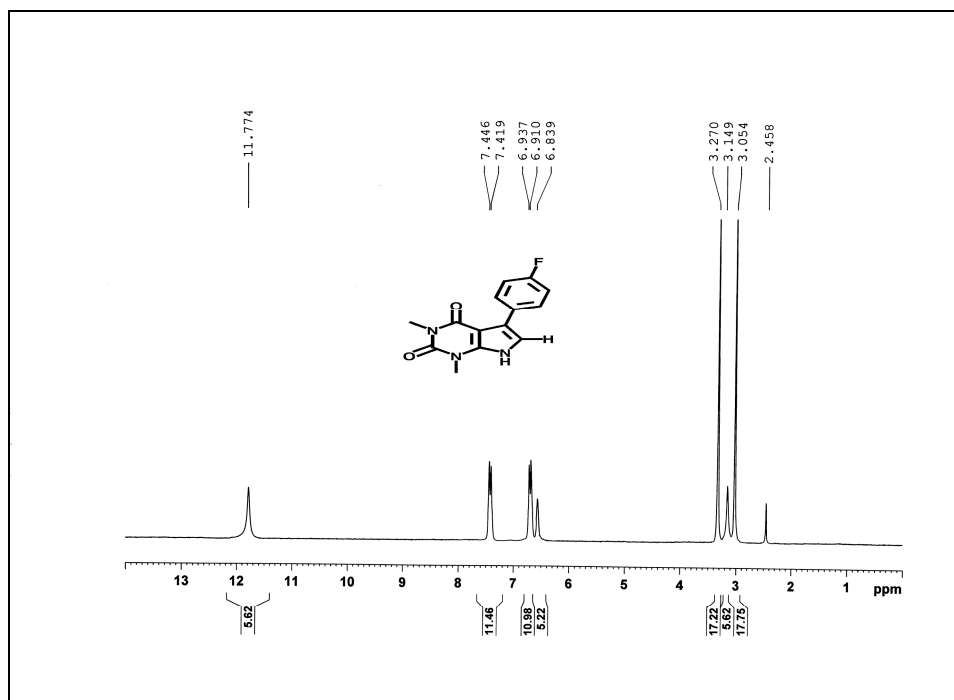
¹³C NMR spectrum of the product (3a)



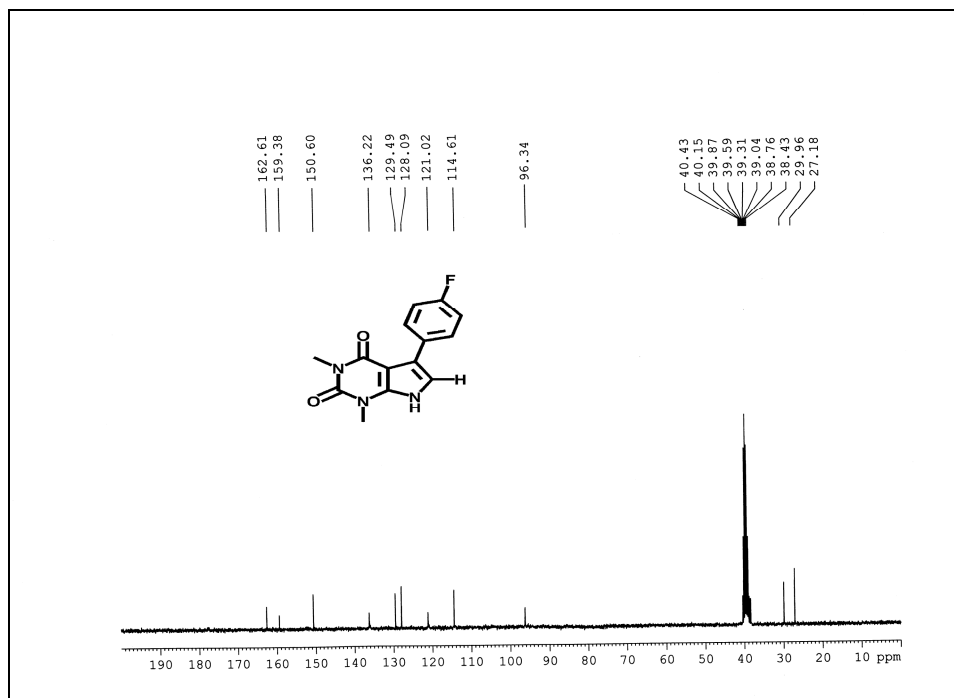
¹H NMR spectrum of the product (3b)



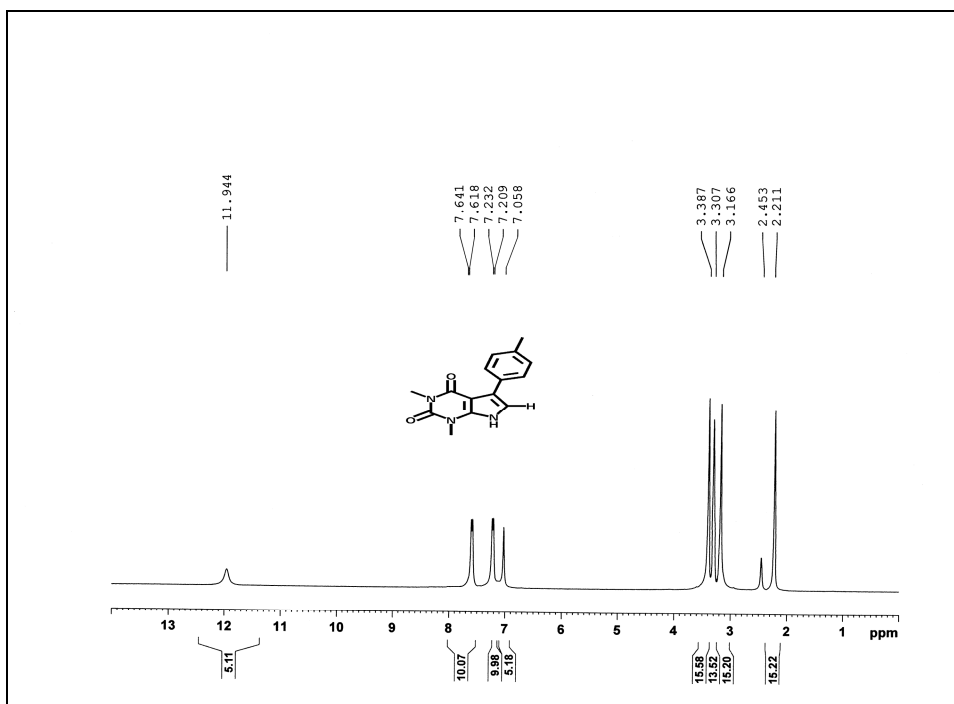
¹³C NMR spectrum of the product (3b)



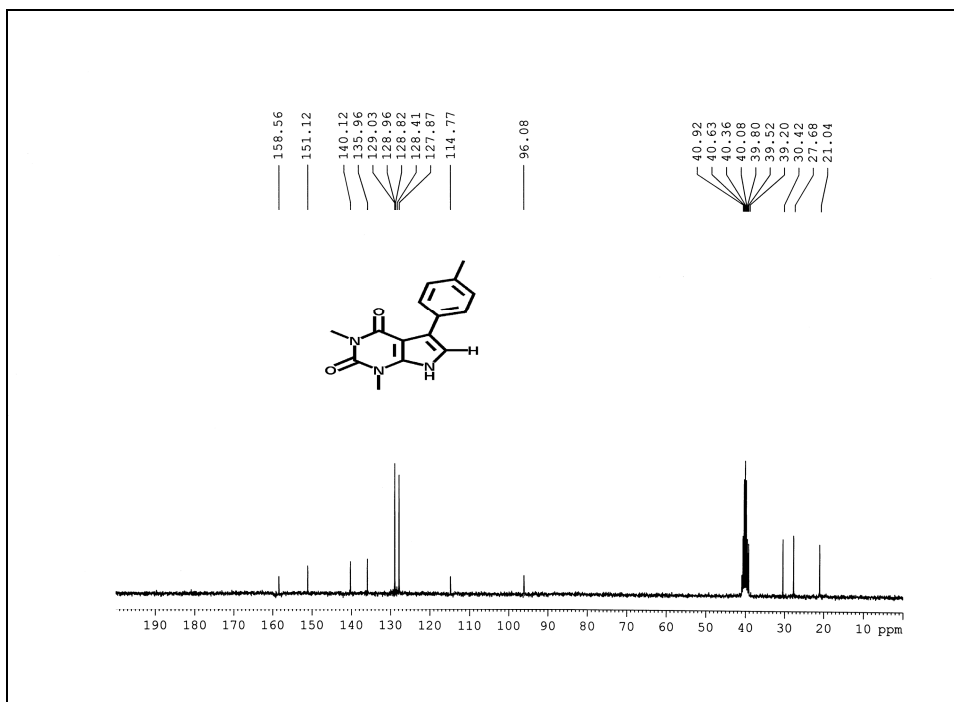
¹H NMR spectrum of the product (3c)



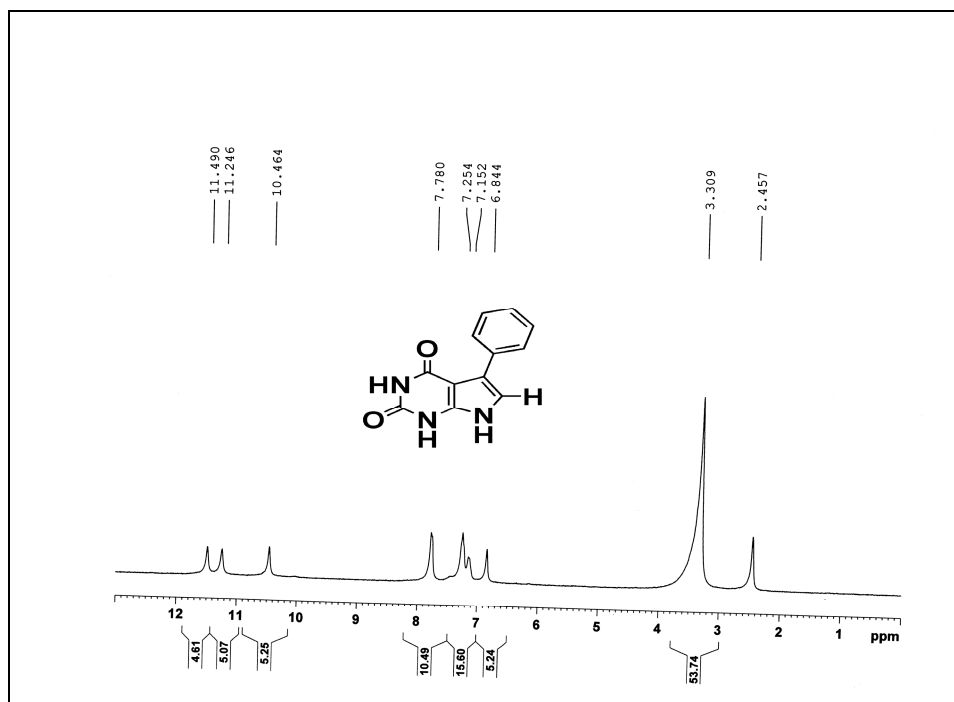
¹³C NMR spectrum of the product (3c)



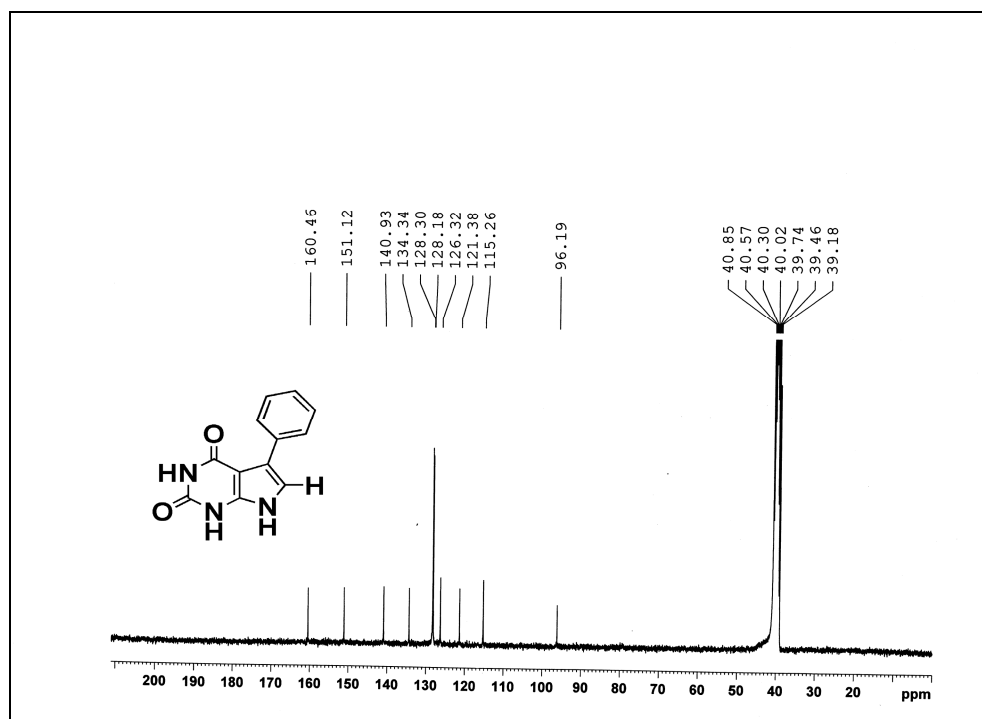
¹H NMR spectrum of the product (3d)



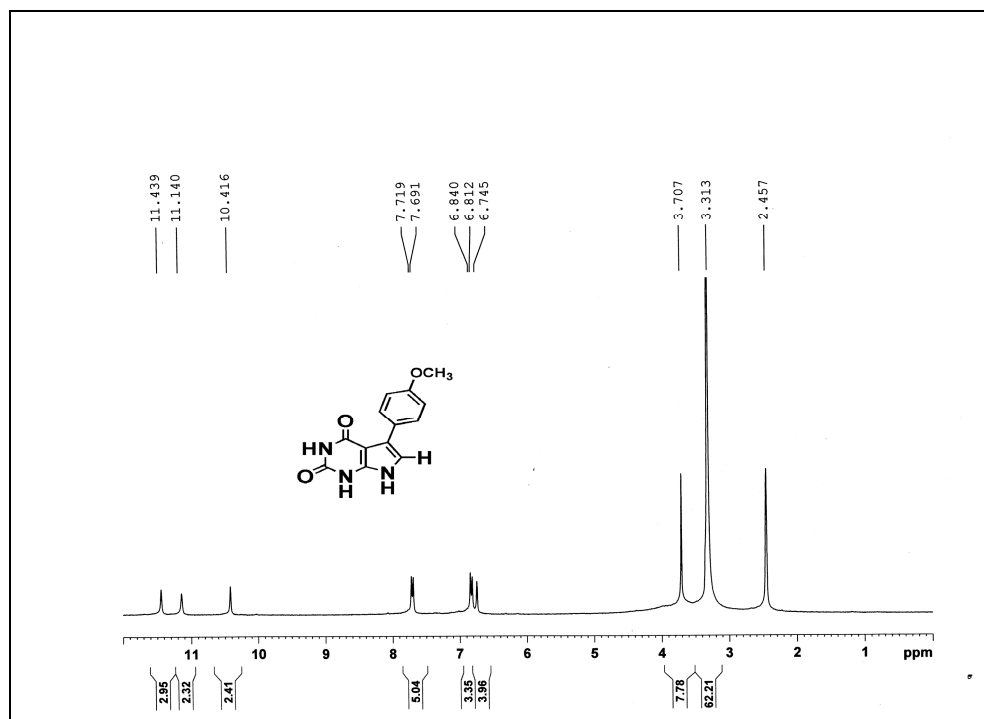
¹³C NMR spectrum of the product (3d)



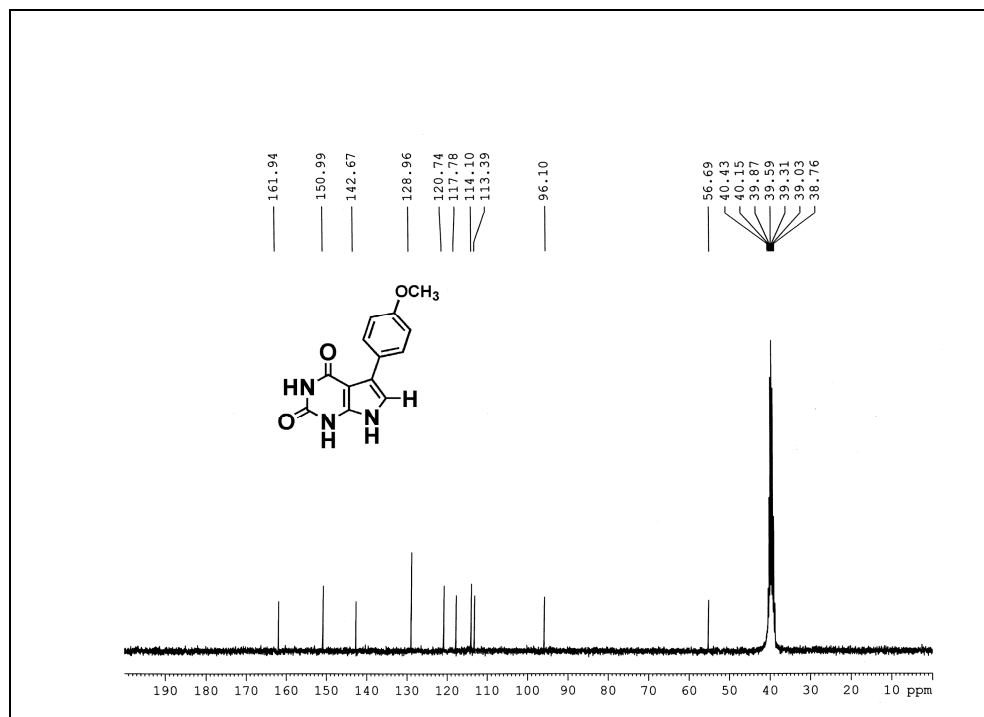
¹H NMR spectrum of the product (3e)



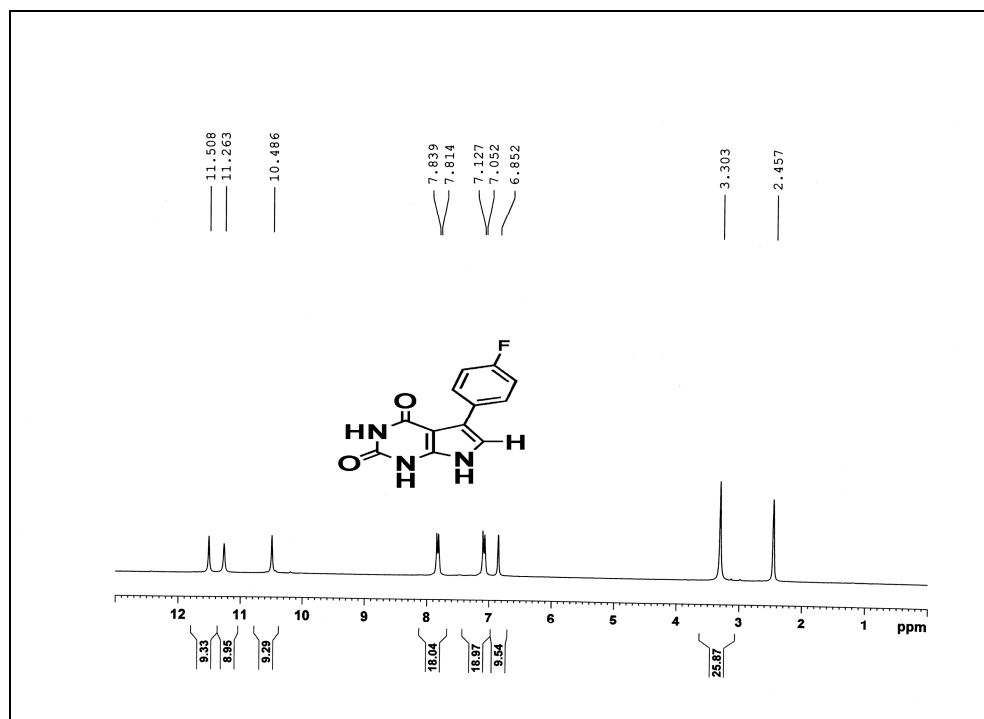
¹³C NMR spectrum of the product (3e)



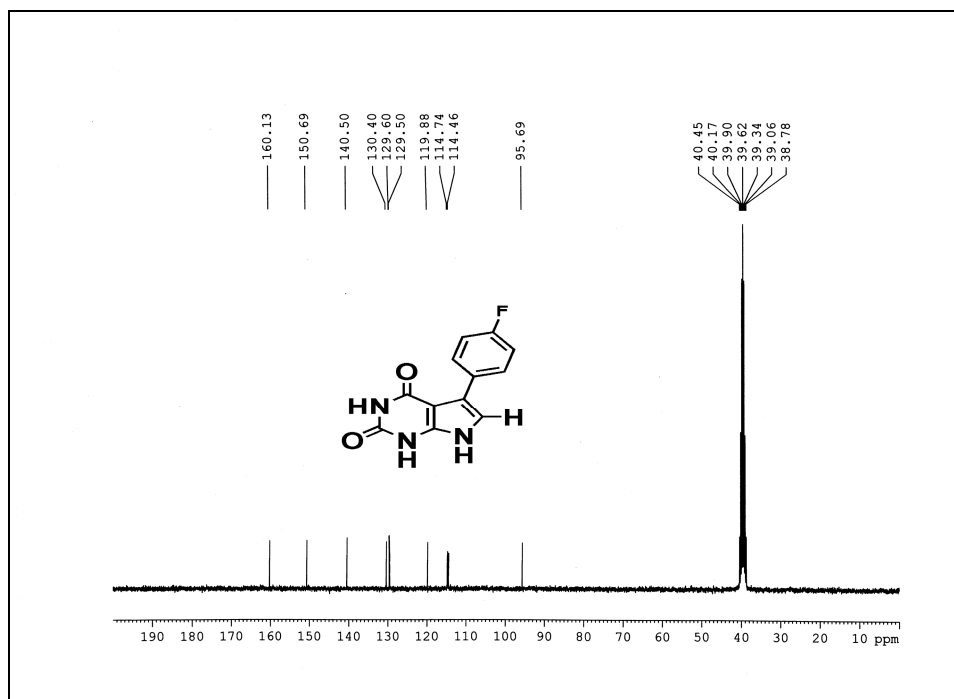
¹H NMR spectrum of the product (3f)



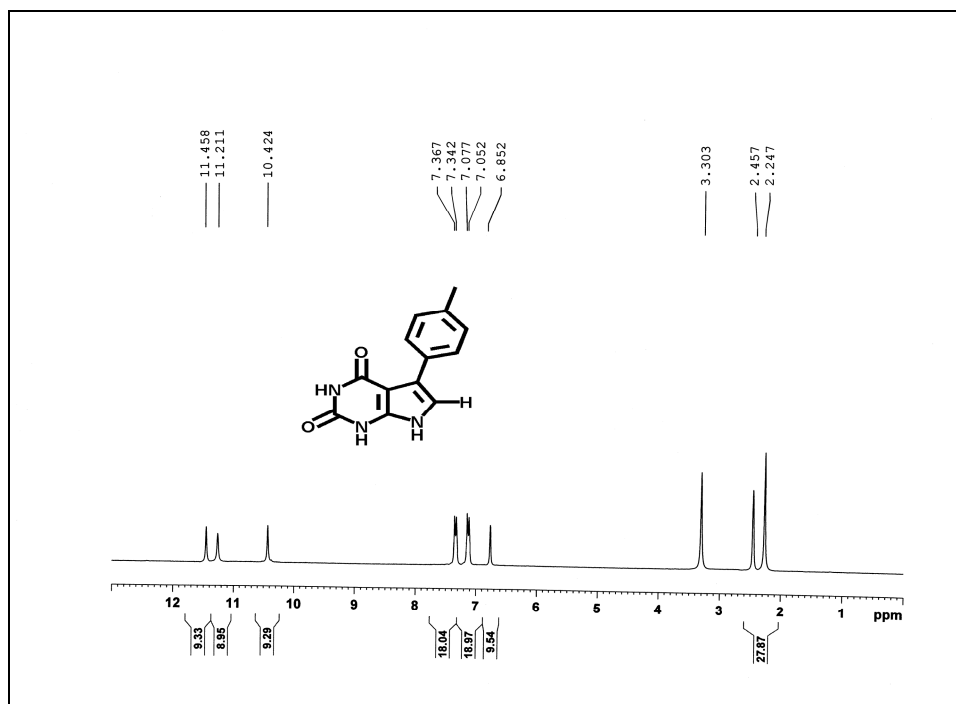
¹³C NMR spectrum of the product (3f)



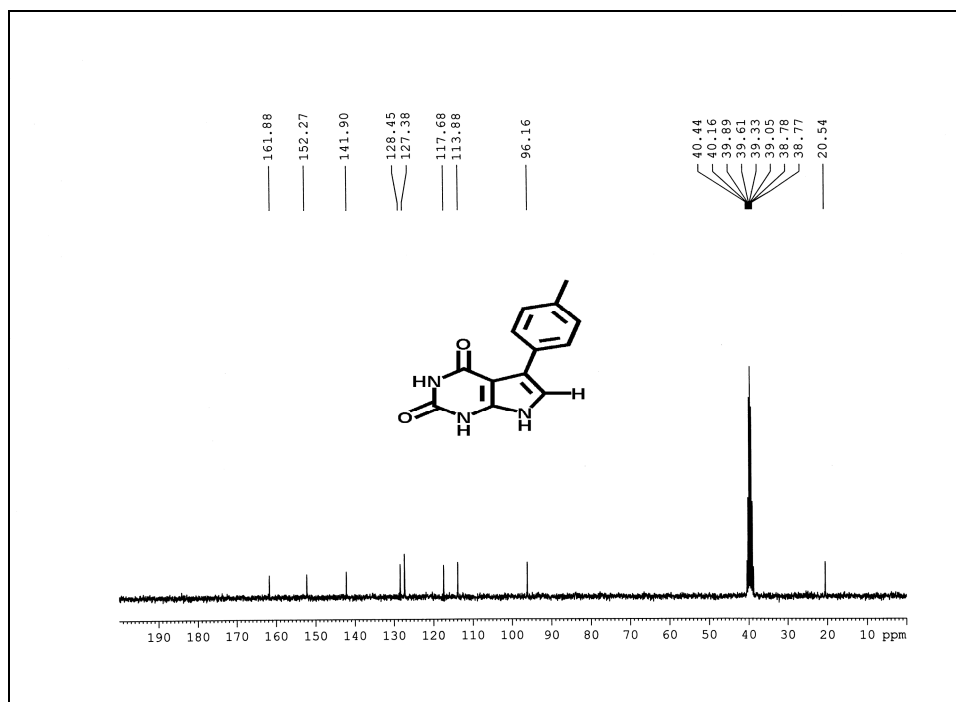
¹H NMR spectrum of the product (3g)



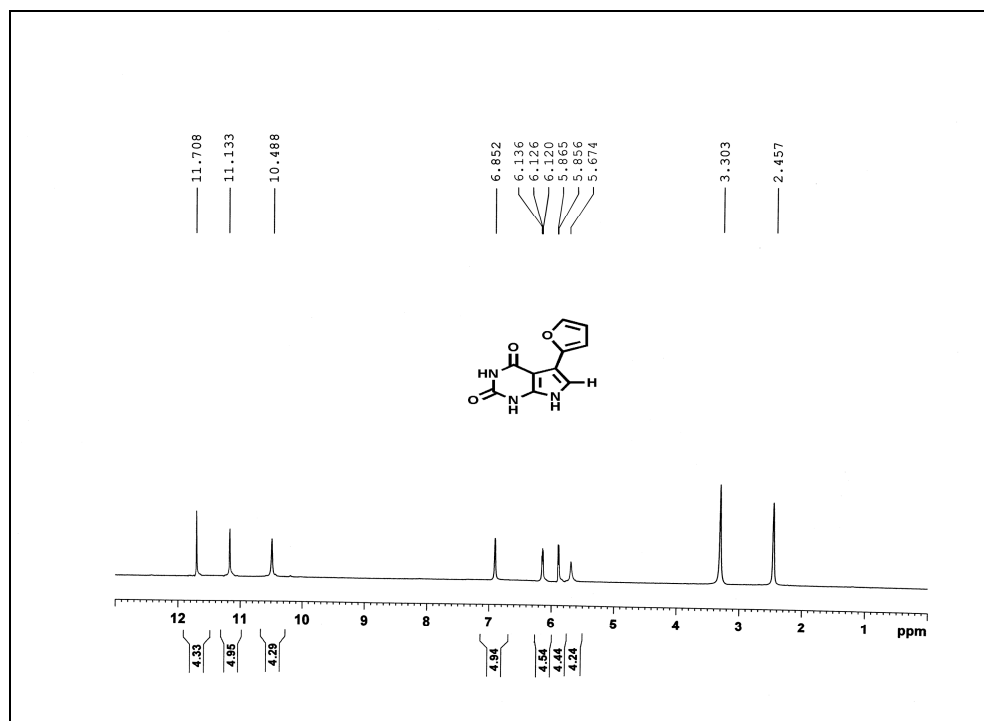
¹³C NMR spectrum of the product (3g)



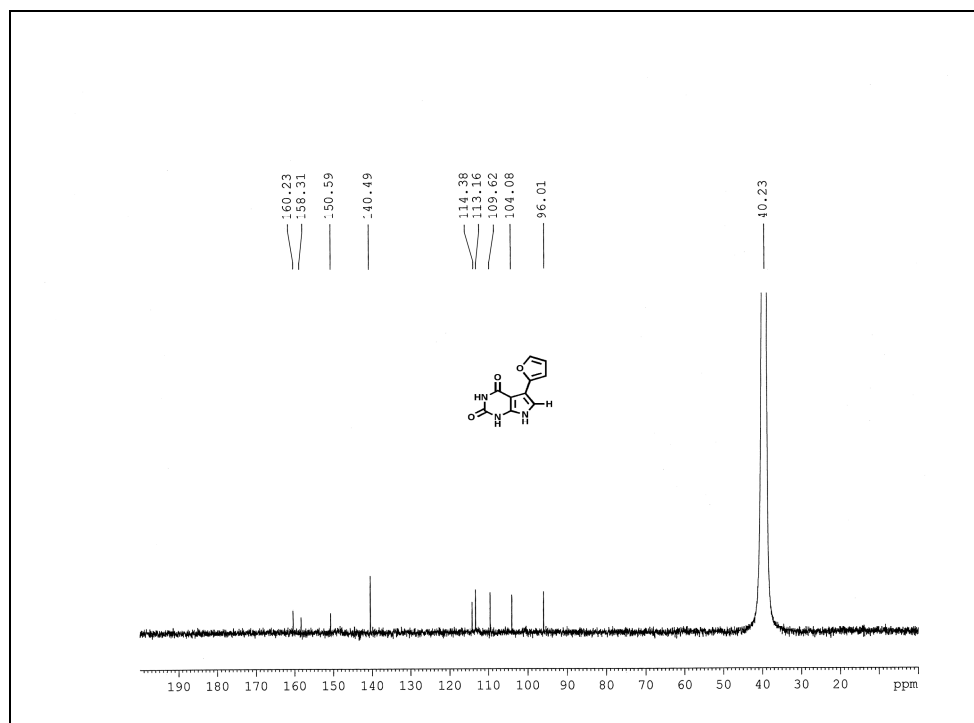
¹H NMR spectrum of the product (3h)



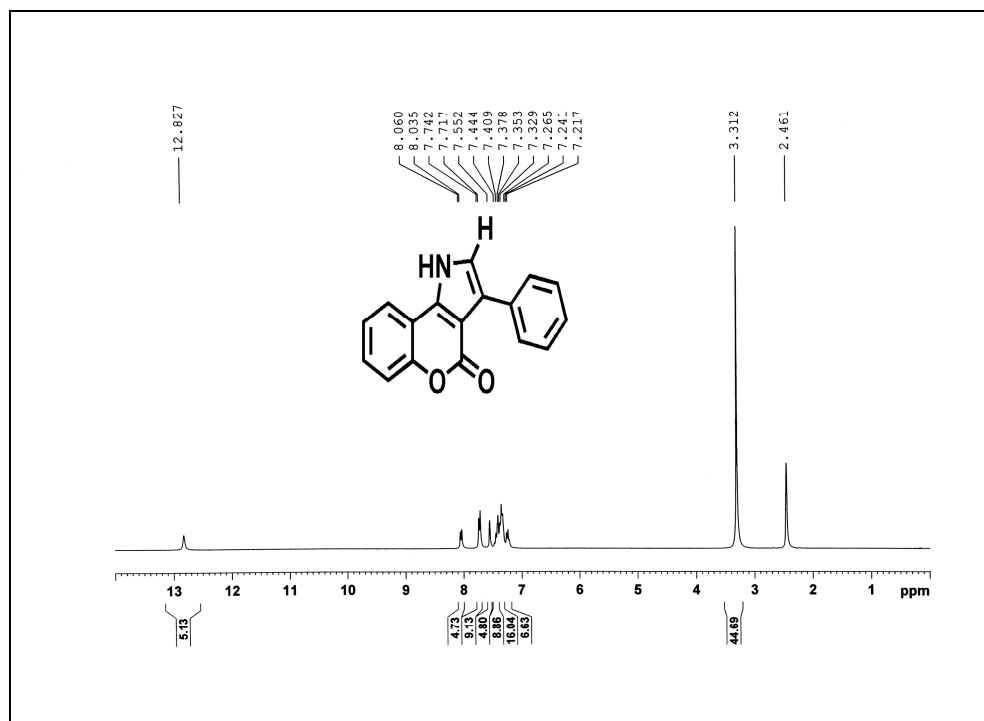
¹³C NMR spectrum of the product (3h)



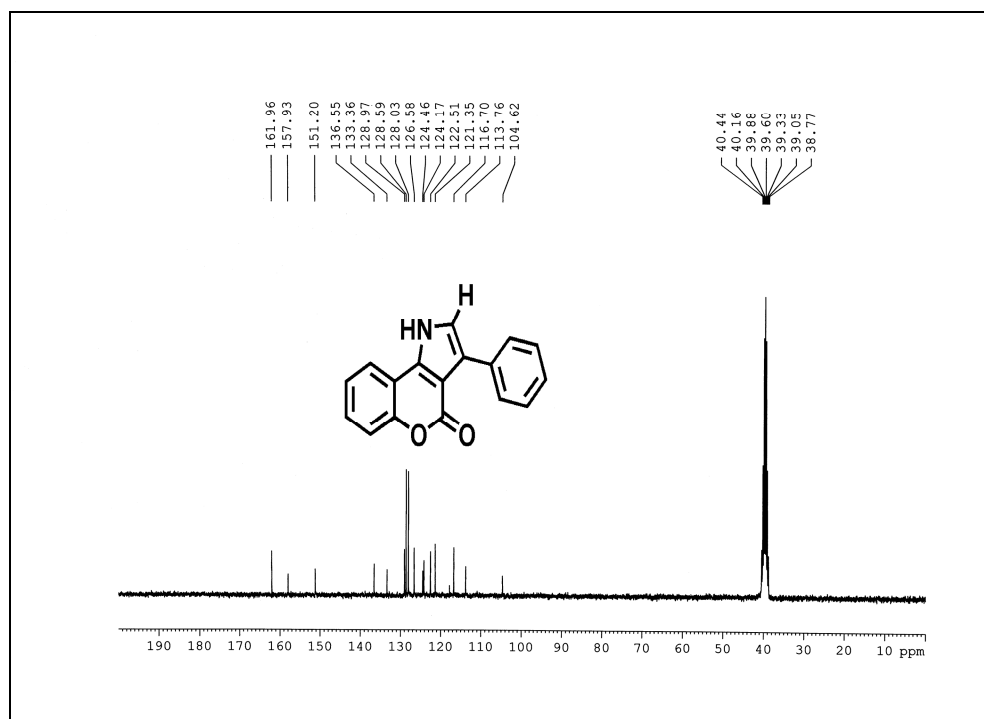
¹H NMR spectrum of the product (3i)



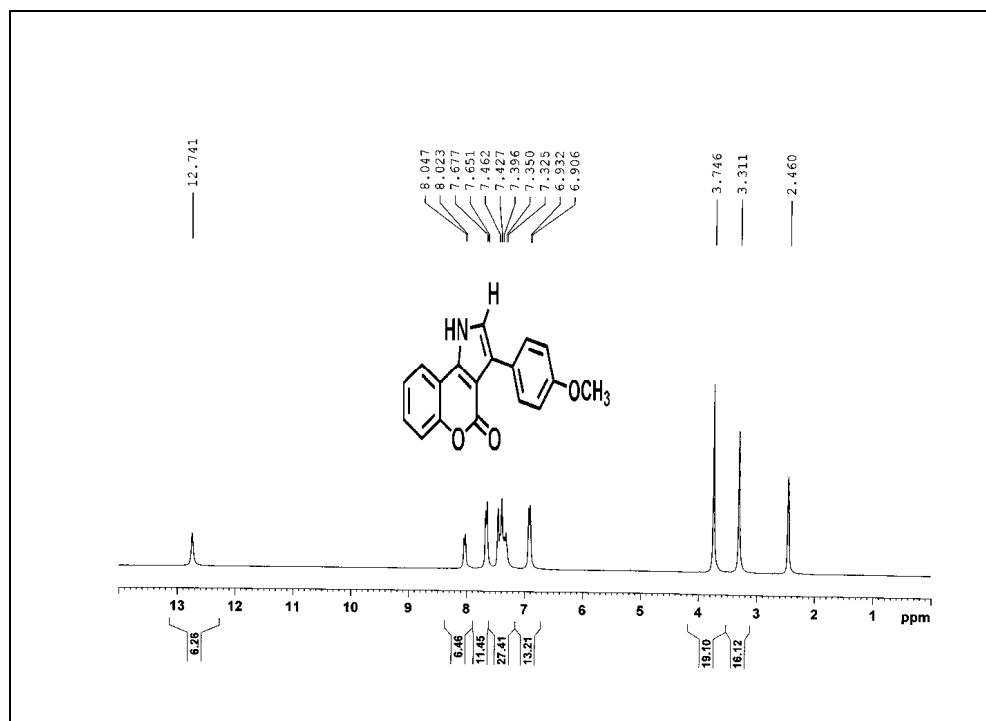
¹³C NMR spectrum of the product (3i)



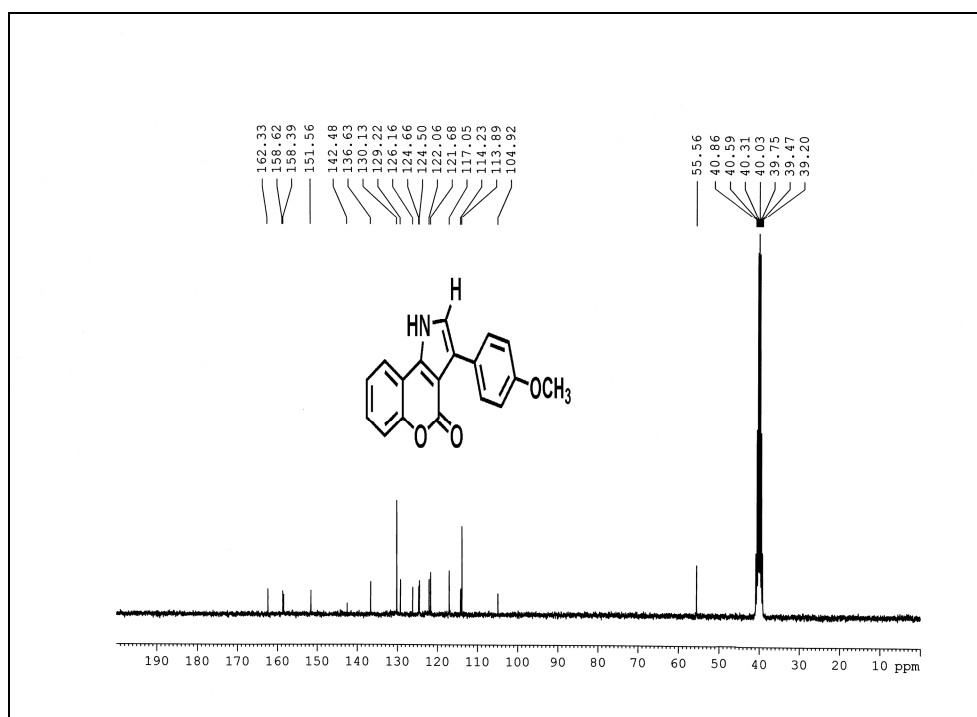
¹H NMR spectrum of the product (4a)



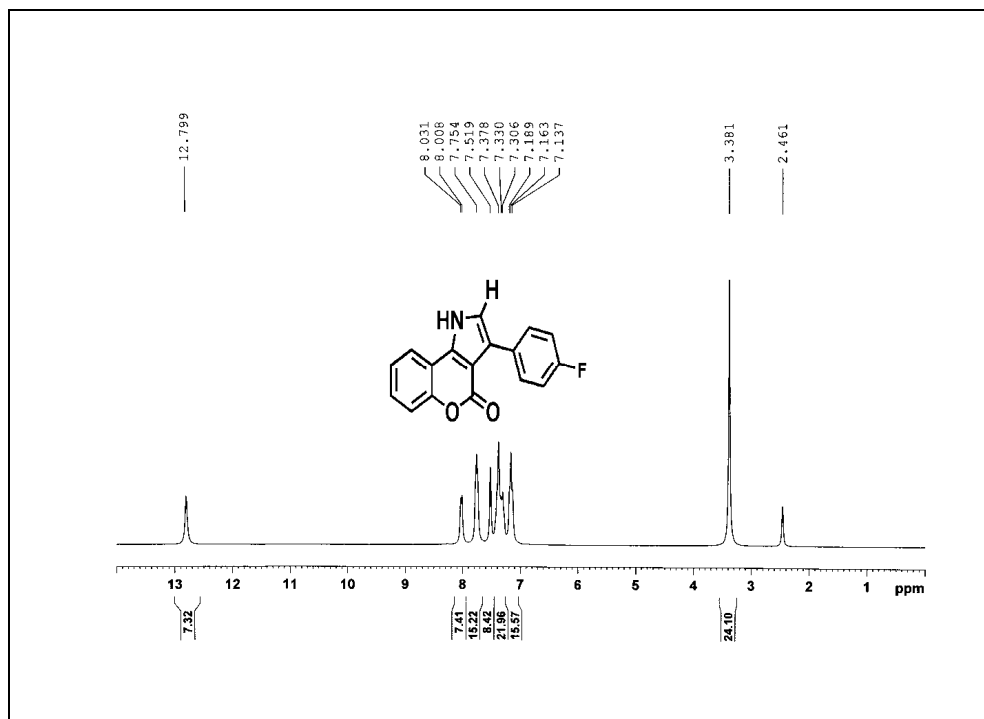
¹³C NMR spectrum of the product (4a)



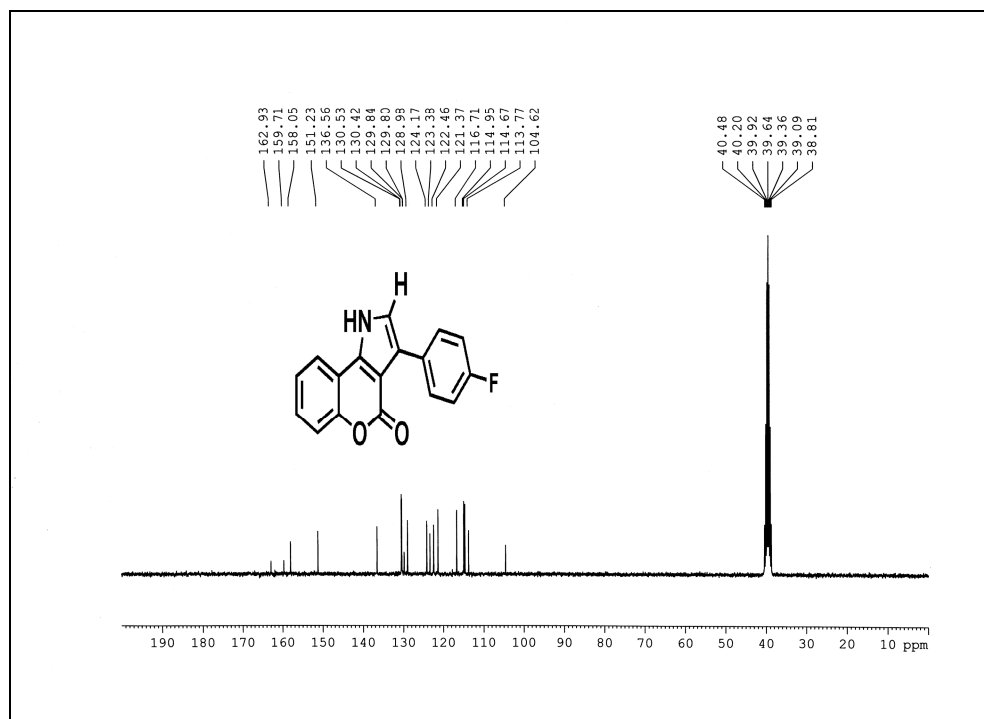
¹H NMR spectrum of the product (4b)



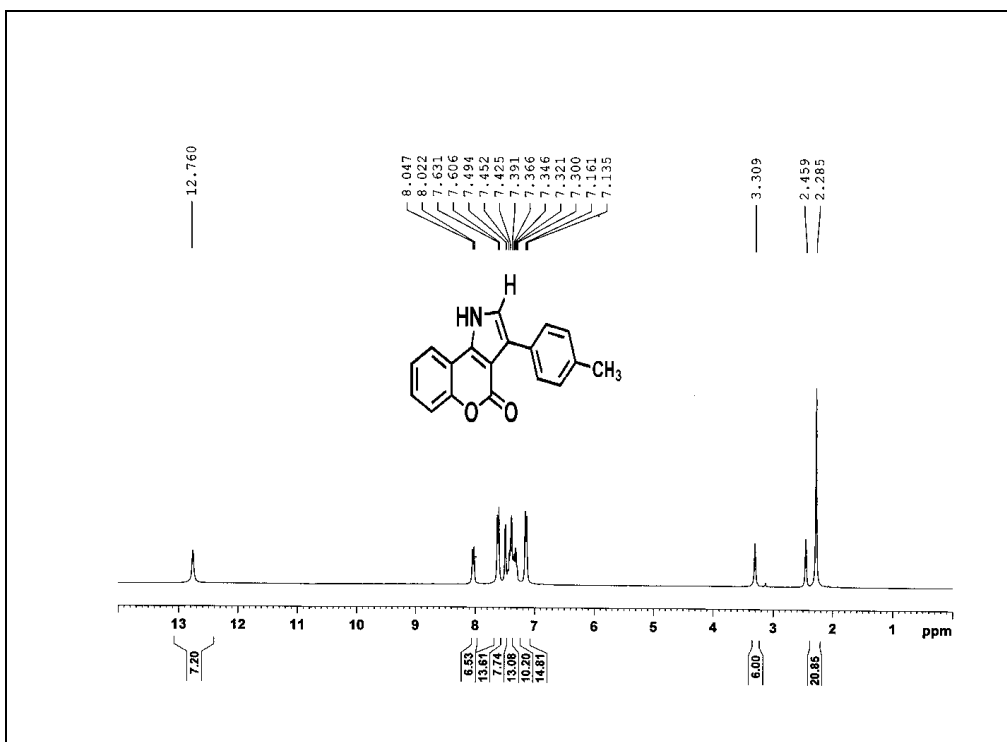
¹³C NMR spectrum of the product (4b)



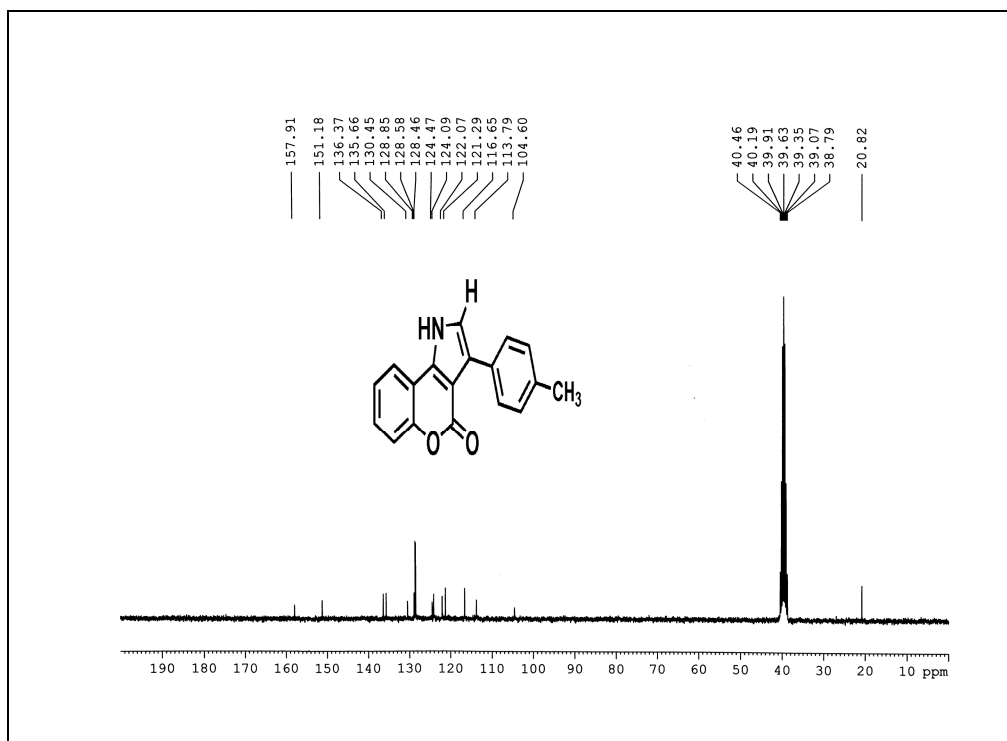
¹H NMR spectrum of the product (4c)



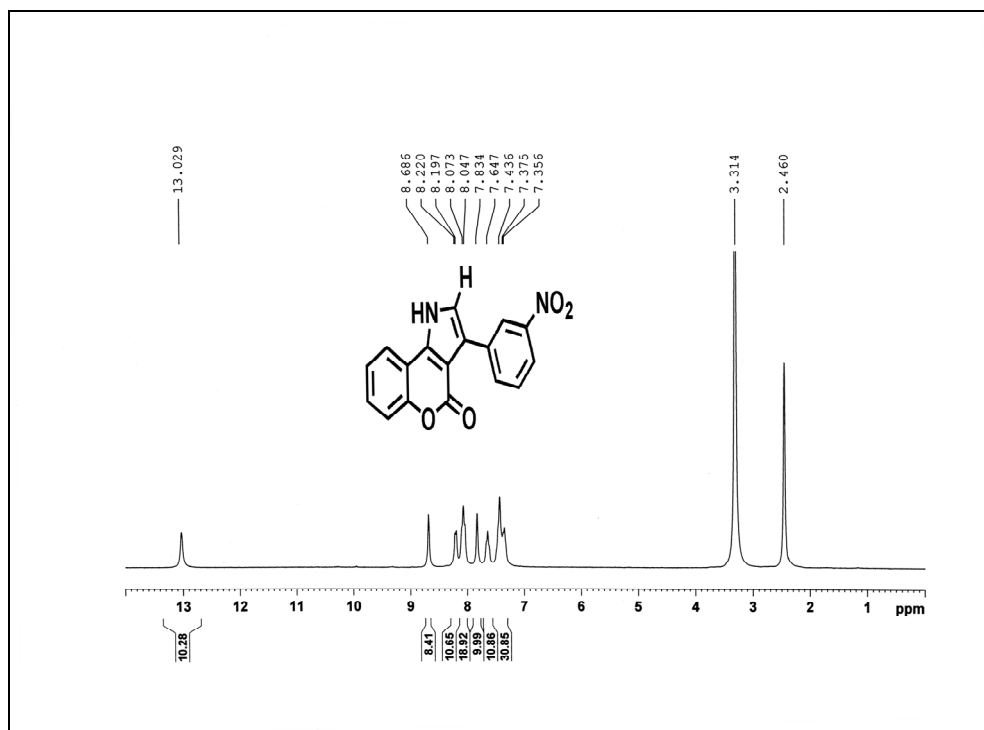
¹³C NMR spectrum of the product (4c)



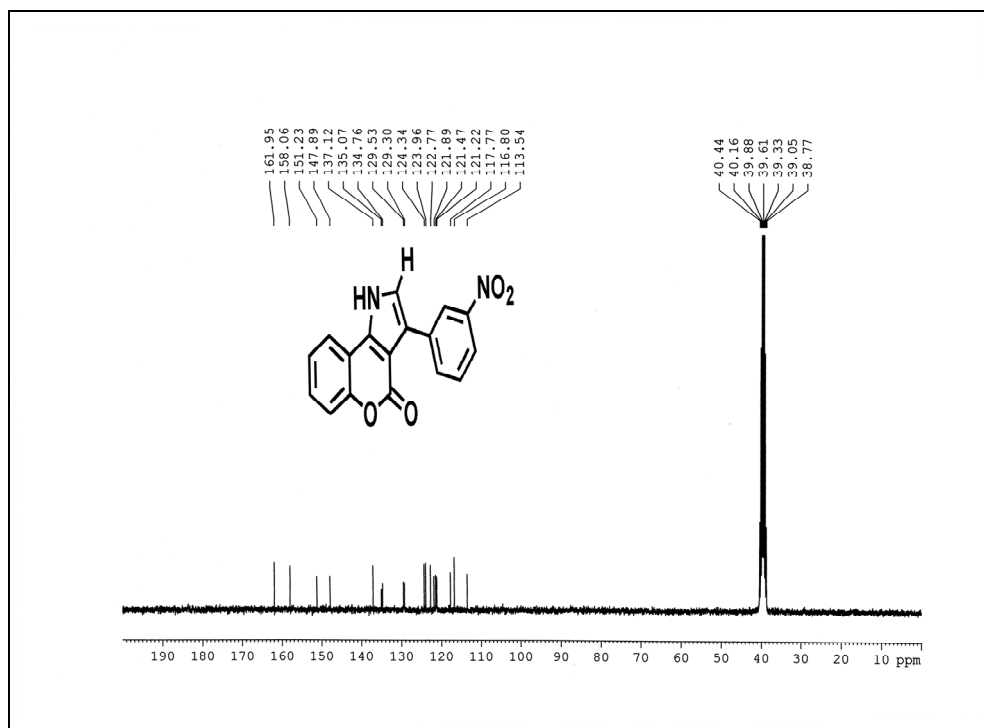
¹H NMR spectrum of the product (4d)



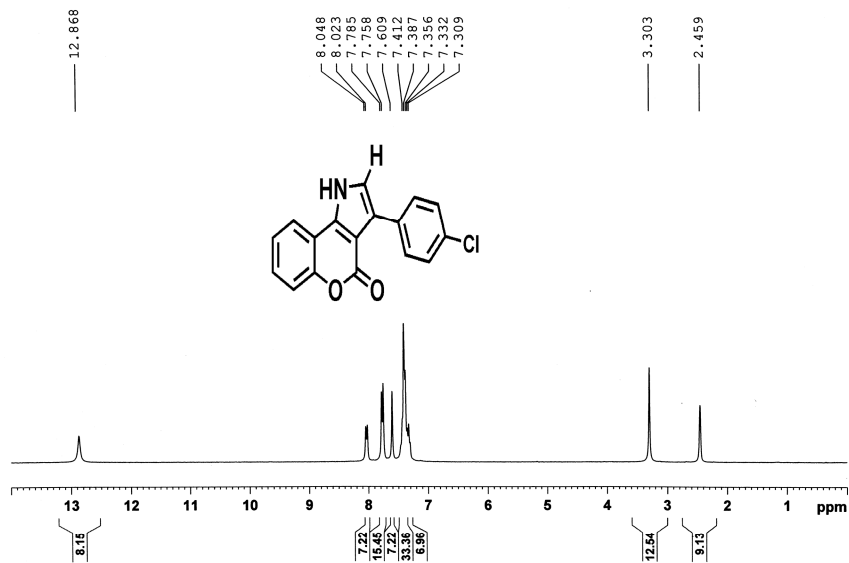
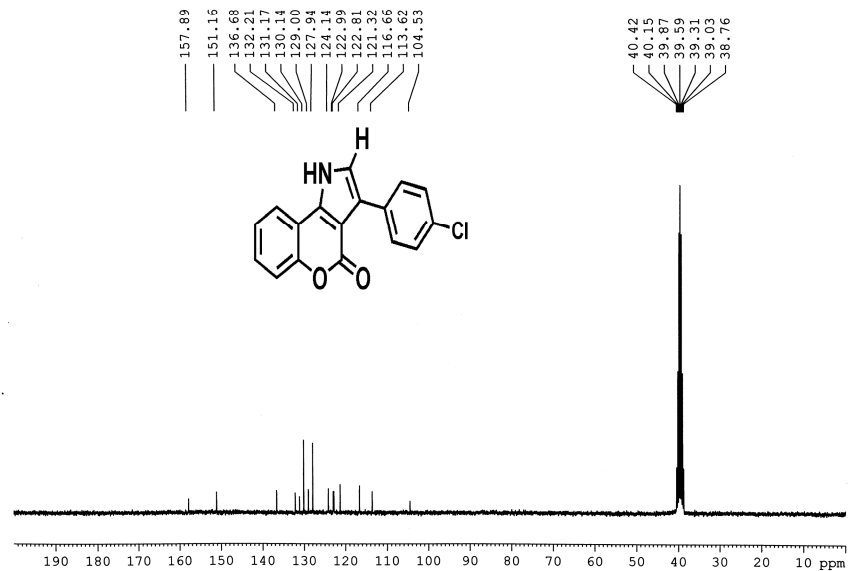
¹³C NMR spectrum of the product (4d)

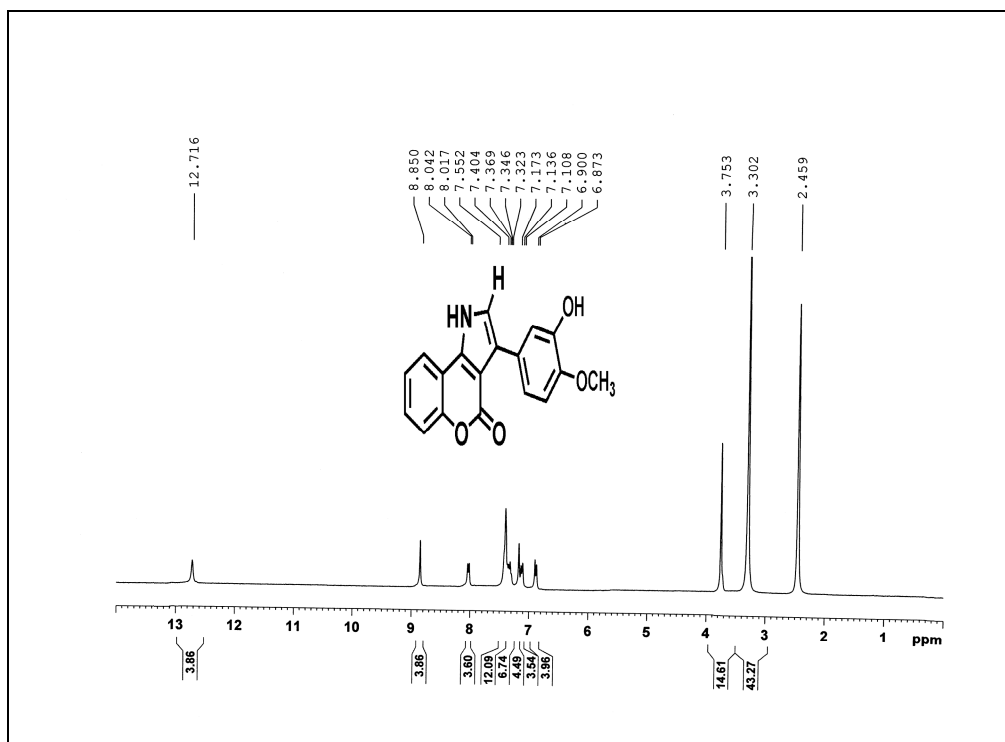


¹H NMR spectrum of the product (4e)

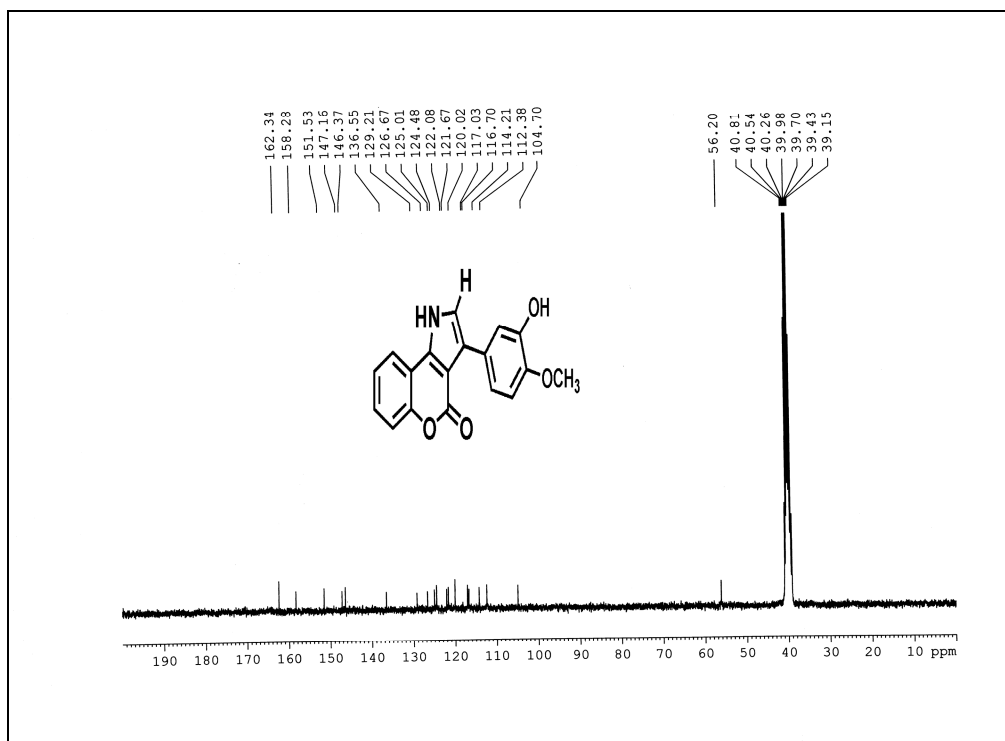


¹³C NMR spectrum of the product (4e)

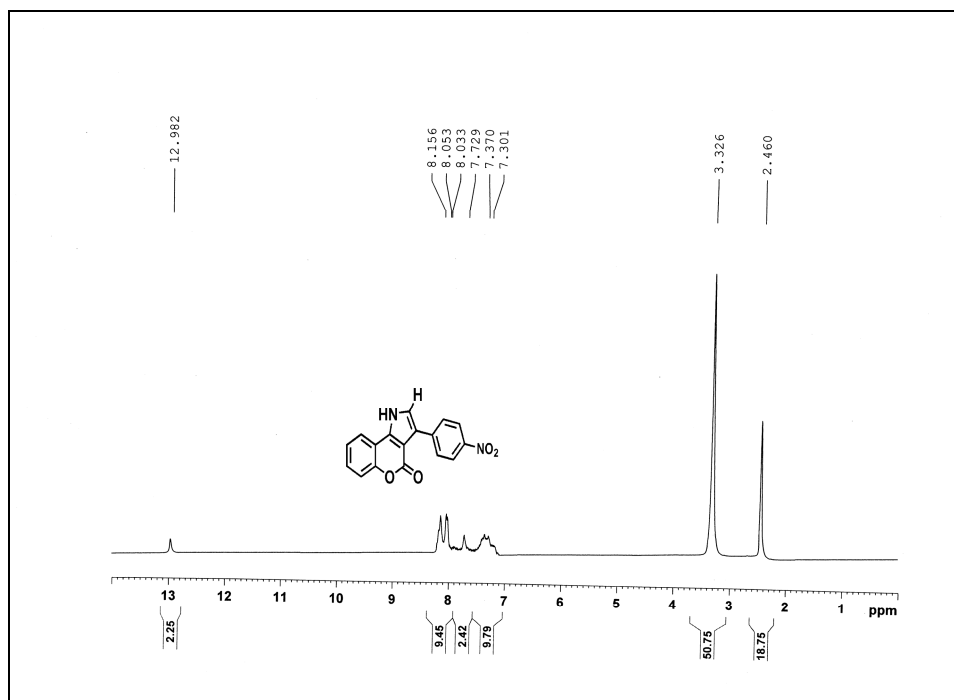
¹H NMR spectrum of the product (4f)¹³C NMR spectrum of the product (4f)



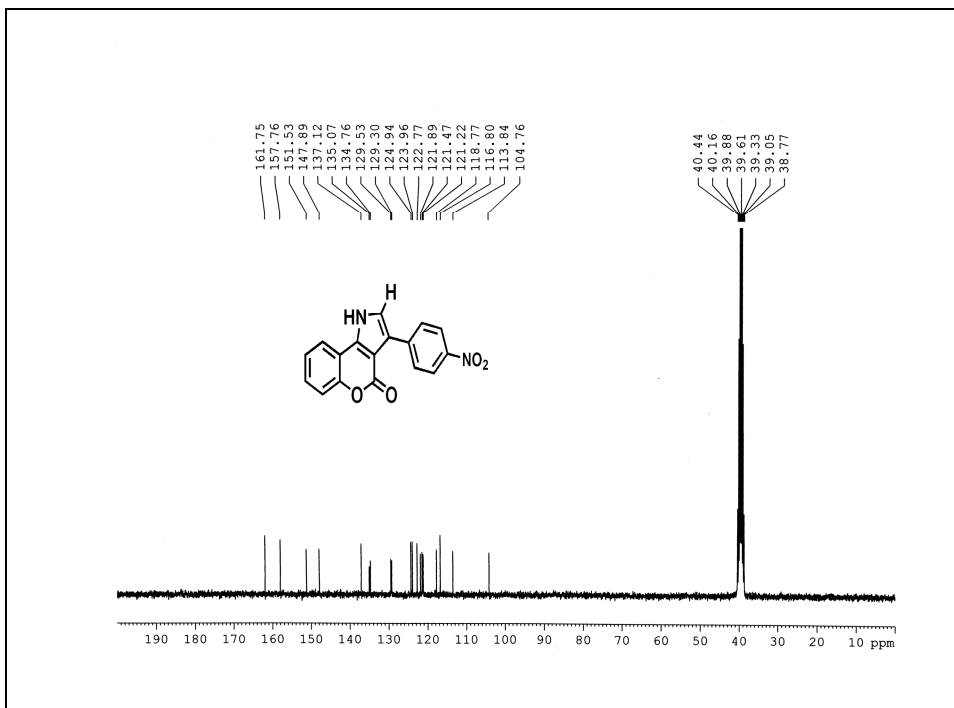
¹H NMR spectrum of the product (4g)



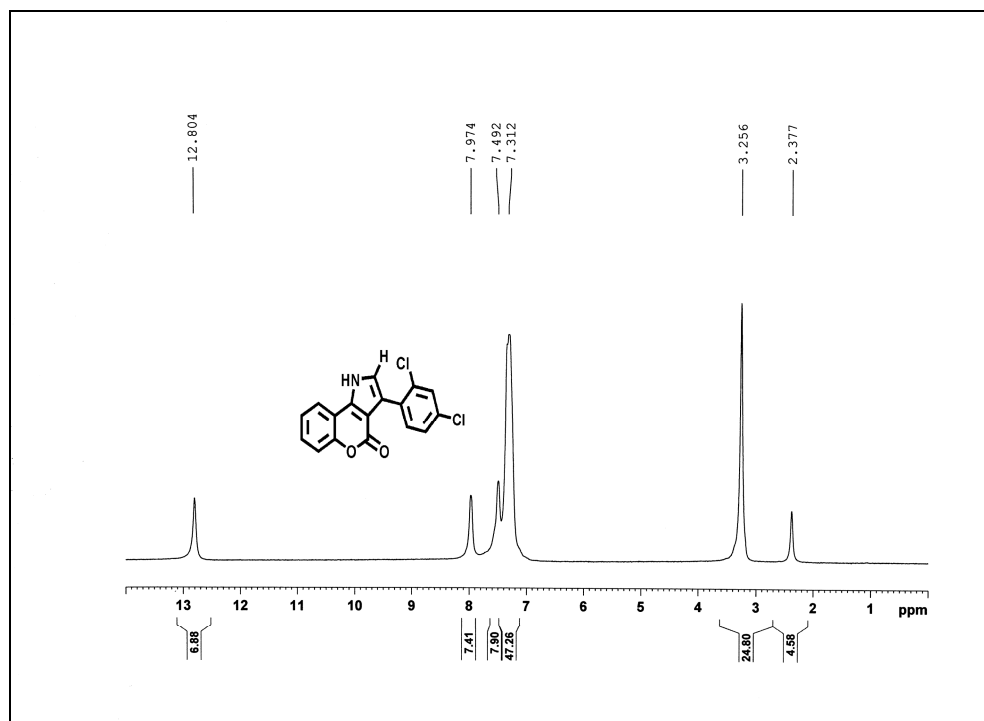
¹³C NMR spectrum of the product (4g)



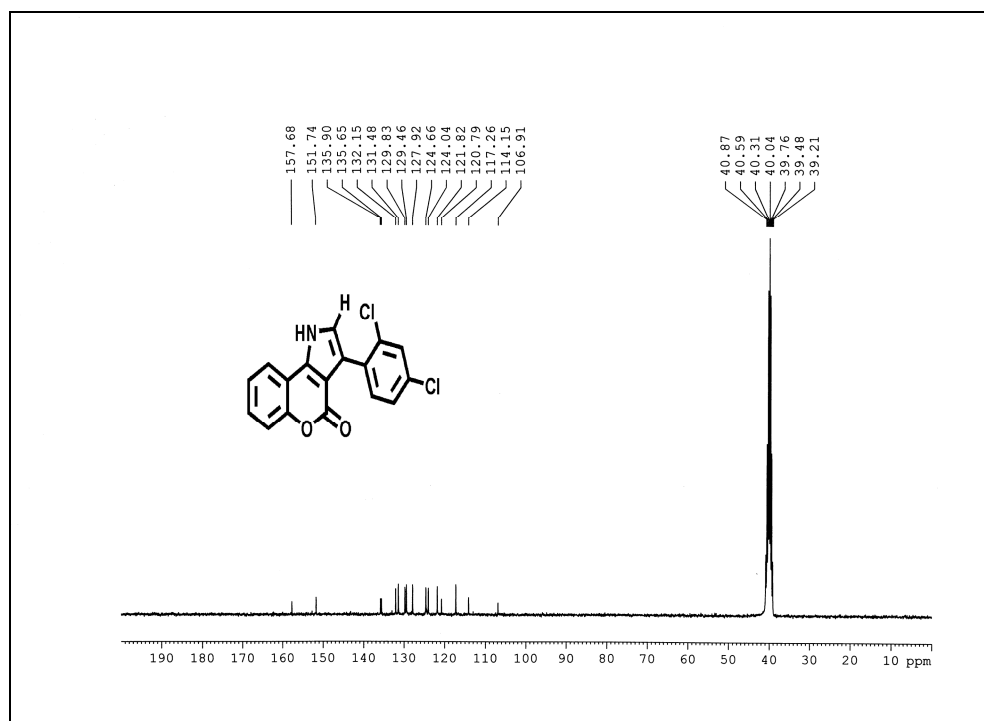
¹H NMR spectrum of the product (4h)



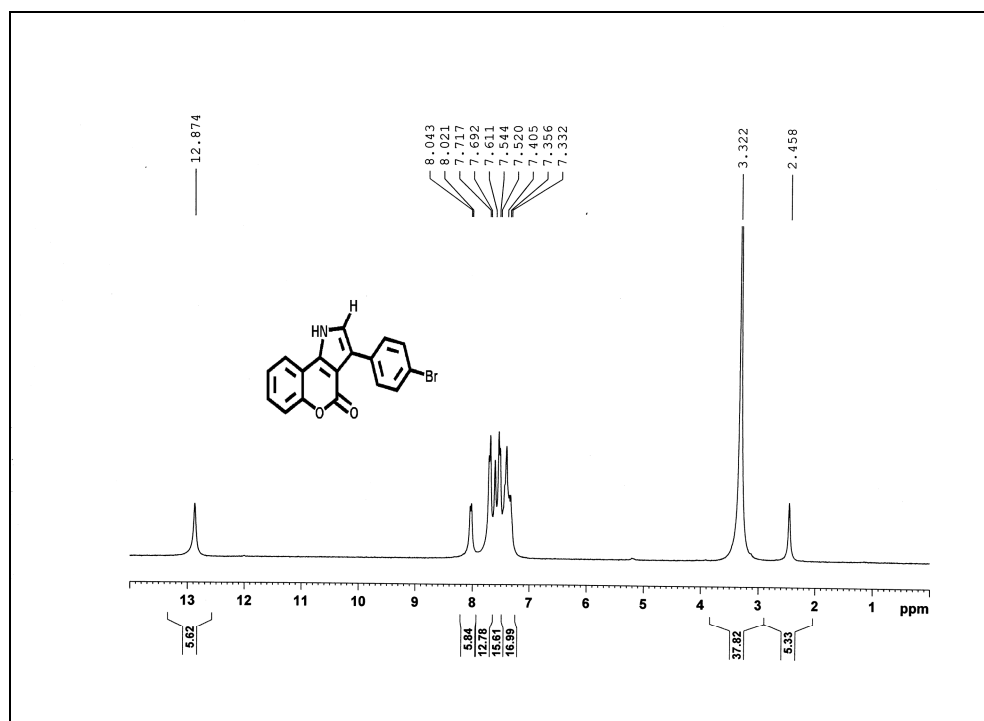
¹³C NMR spectrum of the product (4h)



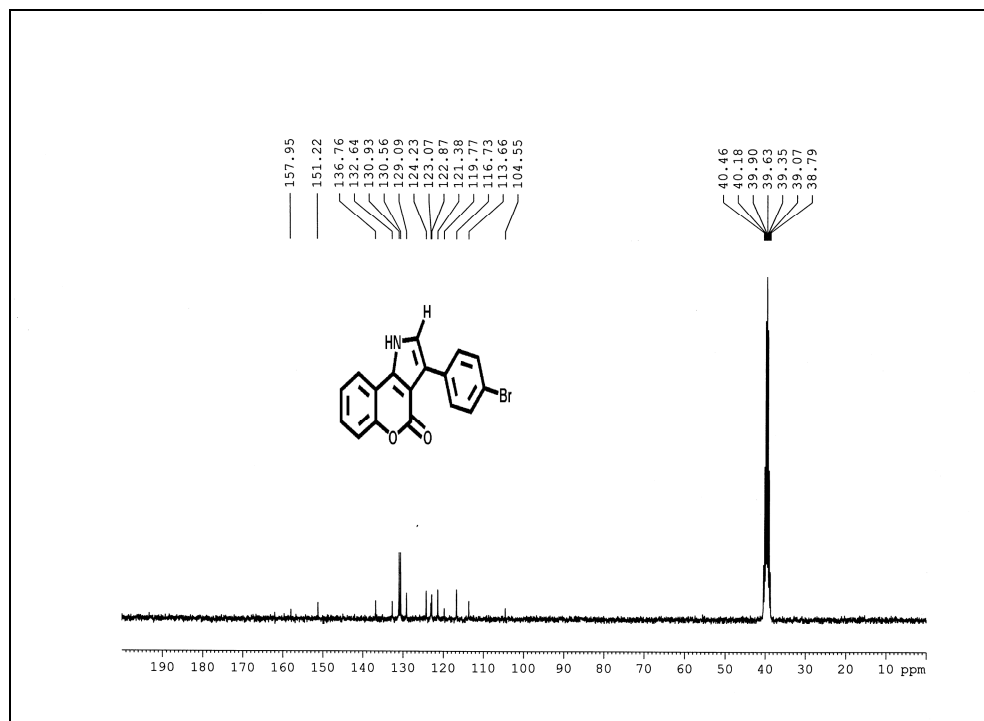
¹H NMR spectrum of the product (4i)



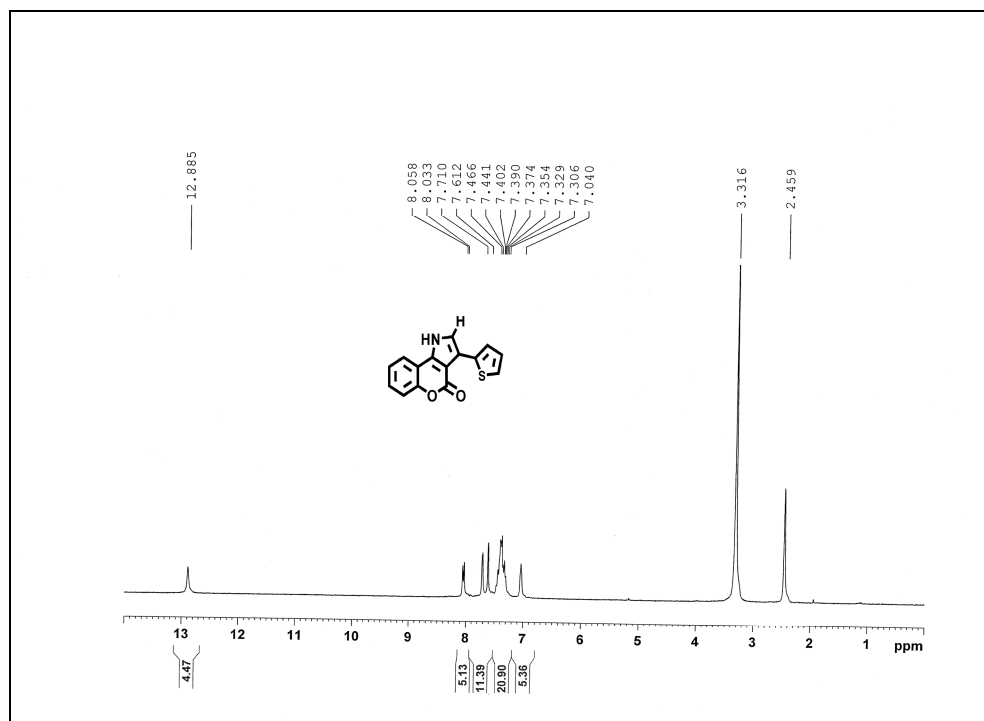
¹³C NMR spectrum of the product (4i)



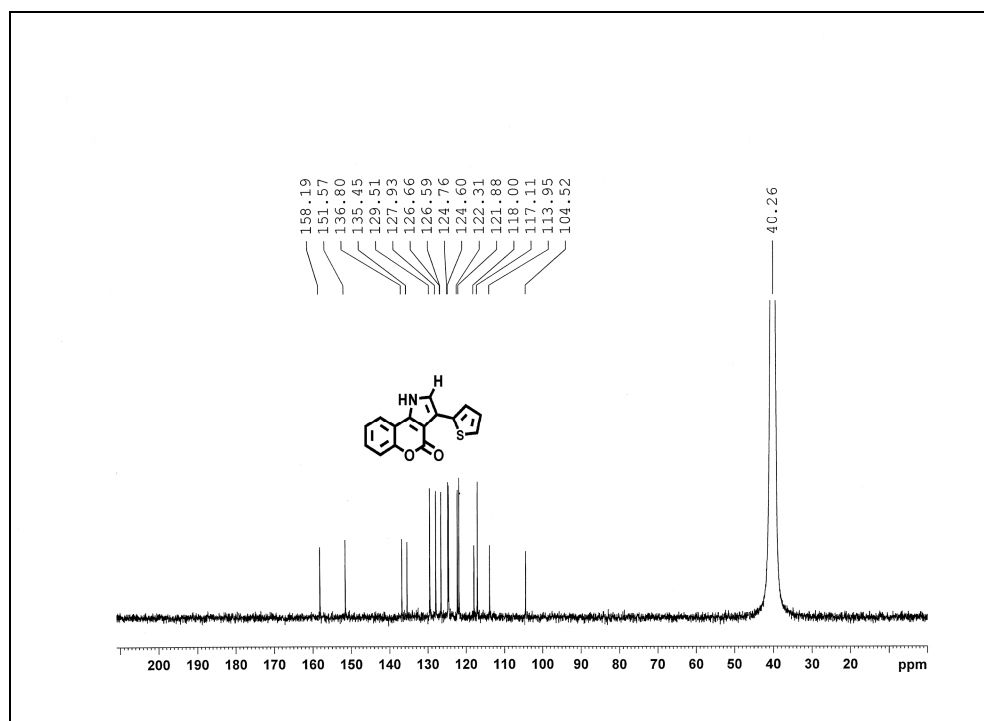
¹H NMR spectrum of the product (4j)



¹³C NMR spectrum of the product (4j)



¹H NMR spectrum of the product (4k)



¹³C NMR spectrum of the product (4k)