Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

Tetrazol-Cu (I) immobilized on nickel ferrite catalyzed green synthesis of indenopyridopyrimidine derivatives in aqueous media

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Table of Contents

A. Experimental section

A typical procedure for the synthesis of 5-(4-chlorophenyl)-1H-indeno[2',1':5,6]pyrido [2,3-d]pyrimidine-2,4,6(3H)-trione

A mixture of 4-chlorobenzaldehyde (1 mmol), 1,3-indanedione (1 mmol), barbituric acid (1 mmol), and ammonium acetate (1.3 mmol) was stirred in water at 95 °C for the appropriate time. The progress of the reaction is monitored by TLC. After completion of the reaction, the corresponding solid product was obtained through simple filtering, and recrystallized from hot ethanol affording the highly pure desired product. The spectral information of various products, including IR, ^{1}H NMR, ^{13}C NMR and C.H.N. analyses are given below.

The IR, ^{1}H NMR and ^{13}C NMR spectra of compounds:

5-(4-chlorophenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione(1a)

White powder; M.P: 258 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1413, 1558 (C=C, Ar), 1634, 1693 (C=O), 3145 (C–H, sp2 stretch), 3265, 3444 (NH). ^{1}H NMR (DMSO-d$_6$, 400 MHz) δ (ppm): 7.45-7.66 (d, 2H, *J* = 8.0 Hz), 7.51-7.52 (m, 2 H), 7.67-7.75 (d, 2H), 7.80-7.82 (d, 2H, *J* = 8.0 Hz), 9.95 (s, 1H). ^{13}C NMR (100 MHz, DMSO-d$_6$) δ (ppm): 123.52, 125.91, 127.94, 128.68, 130.23, 132.23, 135.48, 139.16, 149.25, 162.75, 165.13, 170.00, 79.00; Anal. Calcd for $C_{20}H_{10}N_3O_3Cl$: C, 64; H, 2.67; N, 11.2%; Found: C, 63.91; H, 2.70; N, 11.23%.

5-(4-methoxyphenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (2a)

Yellow powder; M.P: 245 °C, decompose, M.P(Lit)[41] 248 °C;; IR (KBr) (ν_{max}/cm^{-1}): 1443, 1529 (C=C, Ar), 1665, 1750 (C=O), 2916 (C–H, sp3), 3243(C–H, sp2 stretch), 3439, 3585 (NH). ^{1}H NMR (DMSO-d$_6$, 400 MHz) δ (ppm): 7.02-7.04 (d, 2H, *J* = 8.0 Hz), 7.48-7.49 (m, 2 H), 7.58-7.60 (d, 2H, *J* = 8.4 Hz), 7.80-7.81 (m, 2 H). Anal. Calcd for $C_{21}H_{13}N_3O_4$: C, 67.92; H, 3.50; N, 11.32%; Found: C, 67.91; H, 3.47; N, 11.35%.

5-(3-methoxyphenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (3a)

Cream powder; M.P: 231 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1421, 1539 (C=C, Ar), 1635, 1711 (C=O), 2922(C–H, sp3), 3179 (C–H, sp2 stretch), 3342, 3467 (NH). ^{1}H NMR (DMSO-d$_6$, 400 MHz) δ (ppm): 7.04-7.06 (d, 2H), 7.23 (s, 1H), 7.33-7.35 (m, 2 H), 7.51-7.52 (m, 2H), 7.68-7.69 (d, 2 H). ^{13}C NMR (100 MHz, DMSO-d$_6$) δ (ppm): 61.13, 111.67, 112.22, 118.79, 123.57, 125.91, 127.79, 129.18, 130.20, 135.46, 149.57, 161.71, 169.76, 176.16, 179.48.

5-(3-chlorophenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione(4a)

Yellow powder; M.P: 258 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1415, 1501 (C=C, Ar), 1645, 1715, 1736 (C=O), 3241 (C–H, sp2 stretch), 3342, 3515 (NH). ^{1}H NMR (DMSO-

d_6, 400 MHz) δ (ppm): 7.07-7.09 (d, 2 H), 7.28 (s, 1H), 7.44-7.45 (m, 2H), 7.51-7.58 (m, 2H), 7.66-7.68 (d, 2 H). Anal. Calcd for $C_{20}H_{10}N_3O_3Cl$: C, 63.93; H, 2.68; N, 11.18%; Found: C, 65.12; H, 2.53; N, 10.73%.

5-(2-fluorophenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione(5a)

Brown powder; M.P: 293 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1463, 1511 (C=C, Ar), 1608, 1722 (C=O), 3192 (C–H, sp2 stretch), 3418 (NH). ^{1}H NMR (DMSO-d_6, 400 MHz) δ (ppm): 7.42-7.43 (d, 2 H), 7.51-7.52 (m, 2H), 7.78-7.88 (m, 2H), 8.01-8.02 (d, 2H). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 123.52, 125.93, 127.56, 128.62, 130.26, 131.83, 135.53, 139.14, 149.23, 165.37, 174.18, 176.16. Anal. Calcd for $C_{20}H_{10}N_3O_3F$: C, 66.85; H, 2.79; N, 11.70%; Found: C, 66.90; H, 2.77; N, 11.66%.

5-(o-tolyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (6a)

Cream powder; M.P: 282 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1478, 1593 (C=C, Ar), 1609, 1667, 1710 (C=O, 2922 (C–H, sp3), 3189 (C–H, sp2 stretch), 3313, 3413 (NH). ^{1}H NMR (DMSO-d_6, 400 MHz) δ(ppm): 2.877 (s, 3H) , 7.07-7.08 (d, 2 H), 7.41-743 (m, 2 H), 7.51-7.52 (m, 2 H), 7.73-7.74 (d, 2 H). Anal. Calcd for $C_{21}H_{13}N_3O_3$: C, 70.99; H, 3.66; N, 11.83%; Found: C, 70.92; H, 3.69; N, 11.87%.

5-(2,4-dichlorophenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (7a)

Brown powder; M.P: 285 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1423, 1512 (C=C, Ar), 1654, 1712, 1728 (C=O), 3241 (C–H, sp2 stretch), 3341, 3415 (NH). ^{1}H NMR (DMSO-d_6, 400 MHz) δ (ppm): 6.95 (s, 1H), 7.33-7.35 (d, 2 H), 7.44-7.46 (m, 2 H), 7.62-7.64 (d, 2 H). Anal. Calcd for $C_{20}H_9N_3O_3Cl_2$: C, 58.68; H, 2.20; N, 10.27%; Found: C, 58.65; H, 2.17; N, 10.30%.

5-(3-nitrophenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (8a)

White powder; M.P: 279 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1418, 1561 (C=C, Ar), 1369, 1512 (N=O), 1619, 1711 (C=O), 2909 (C–H, sp3), 3158 (C–H, sp2 stretch), 3509 (NH). ^{1}H NMR (DMSO-d_6, 400 MHz) δ (ppm): 7.13-7.16 (m, 2 H), 7.33- 7.35 (d, 2 H), 7.46-7.47 (d, 2 H), 7.50-7.55 (m, 2 H), 7.66 (s, 1H). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 121.27, 123.46, 125.86, 127.76, 129.46, 130.24, 134.23, 135.36, 147.63, 149.46, 168.93, 170.57, 177.77. Anal. Calcd for $C_{20}H_{10}N_4O_5$: C, 62.18; H, 2.59; N, 18.13%; Found: C, 62.11; H, 2.57; N, 18.17%.

5-(2-chlorophenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (9a)

Yellow powder; M.P: 228 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1429, 1541 (C=C, Ar), 1619, 1668 (C=O), 3219 (C–H, sp2 stretch), 3336, 3412 (NH). ^{1}H NMR (DMSO-d_6, 400 MHz) δ (ppm): 7.07-7.09 (d, 2 H), 7.31-7.33 (m, 2 H), 7.50-7.52 (m, 2 H), 7.62-7.64 (d, 2 H). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 123.52, 126.23, 127.63, 128.65, 130.23, 131.83, 135.19, 139.11, 149.20, 168.77, 174.16, 176.44.

5-(4-bromophenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (10a)

Cream powder; M.P: 242 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1447, 1559 (C=C, Ar), 1681, 1719 (C=O), 3072 (C–H, sp2 stretch), 3235, 3527 (NH). ^{1}H NMR (DMSO-d$_6$, 400 MHz) δ (ppm): 7.36-7.38 (d, 2 H, *J*= 8.0 Hz), 7.53-7.55 (m, 2 H), 7.67-7.69 (d, 2 H, *J*= 8.0 Hz), 7.724 (d, 2 H). ^{13}C NMR (100 MHz, DMSO-d$_6$) δ (ppm): 120.62, 123.50, 125.88, 126.75, 127.81, 130.18, 135.46, 139.13, 141.65, 147.57, 149.23, 168.63, 174.27, 176.64. Anal. Calcd for $C_{20}H_{10}N_3O_3Br$: C, 57.14; H, 2.38; N, 10%; Found: C, 57.11; H, 2.41; N, 10.03%.

5-(5-methylfuran-2-yl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (11a)

Red brown powder; M.P: 339 °C, decompose; IR (KBr) (ν_{max}/cm^{-1}): 1423, 1531 (C=C, Ar), 1631, 1733 (C=O), 2931 (C–H, sp3), 3130 (C–H, sp2 stretch), 3242, 3425 (NH). ^{1}H NMR (DMSO-d$_6$, 400 MHz) δ(ppm): 2.73 (s, 3H), 7.45-7.47 (d, 2 H), 7.67-7.70 (m, 2 H), 7.84-7.87 (d, 2 H). Anal. Calcd for $C_{19}H_{11}N_3O_4$: C, 66.08; H, 3.19; N, 12.17%; Found: C, 66.03; H, 3.11; N, 12.21%.

5-(4-nitrophenyl)-1H-indeno[2',1':5,6]pyrido[2,3-d]pyrimidine-2,4,6(3H)-trione (12a)

Dark brown powder; M.P: 343 °C, decompose, M.P (Lit)41 345 °C; IR (KBr) (ν_{max}/cm^{-1}): 1433, 1531 (C=C, Ar), 1317, 1553 (N=O), 1621, 1739 (C=O), 3205 (C–H, sp2stretch), 3242, 3428 (NH). ^{1}H NMR (DMSO-d$_6$, 400 MHz) δ (ppm): 7.754-7.775 (d, 2 H, *J*= 8.0 Hz), 7.85-7.86 (m, 2 H), 8.18-8.20 (m, 2 H), 8.37-8.40 (d, 2 H, *J*= 8.4 Hz).

Spectra of IR, ^{1}H NMR and ^{13}C NMR

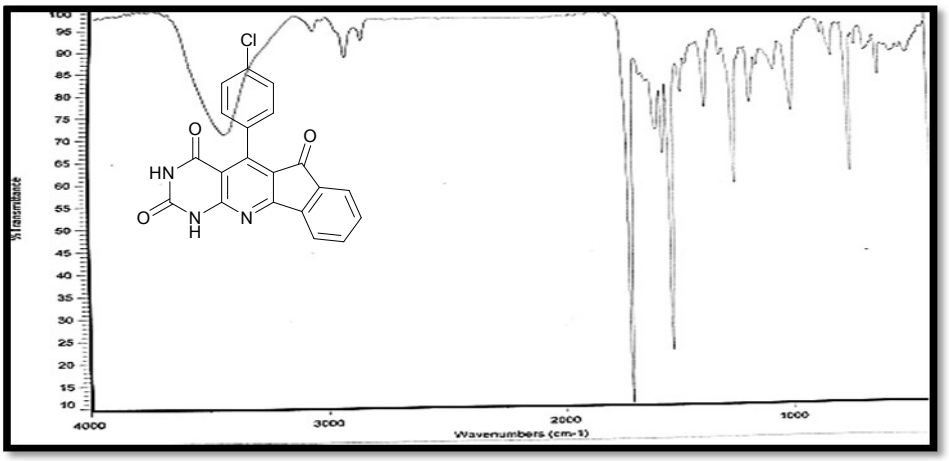

Figure 1s: IR spectrum of 1a

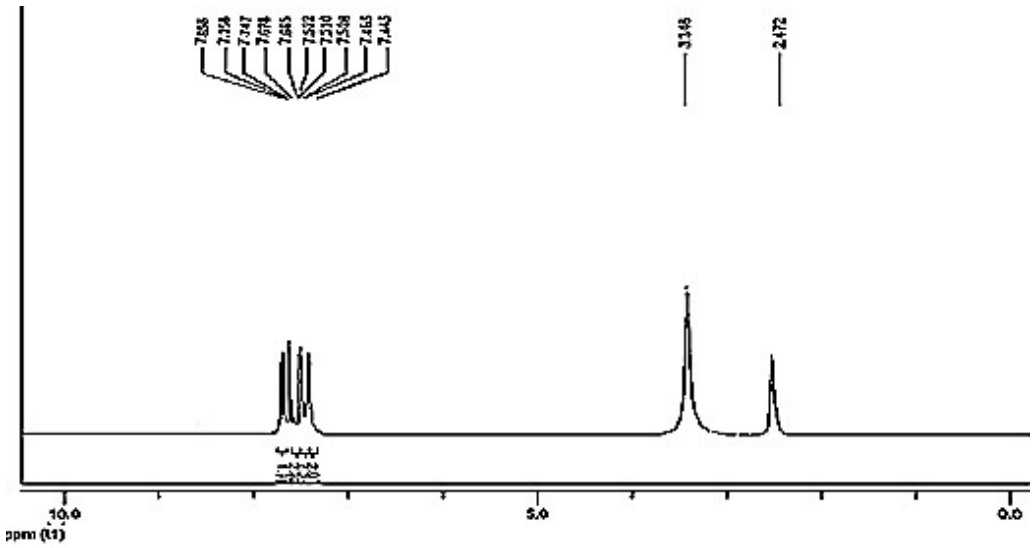

Figure 2s: ^{1}H NMR spectrum of 1a

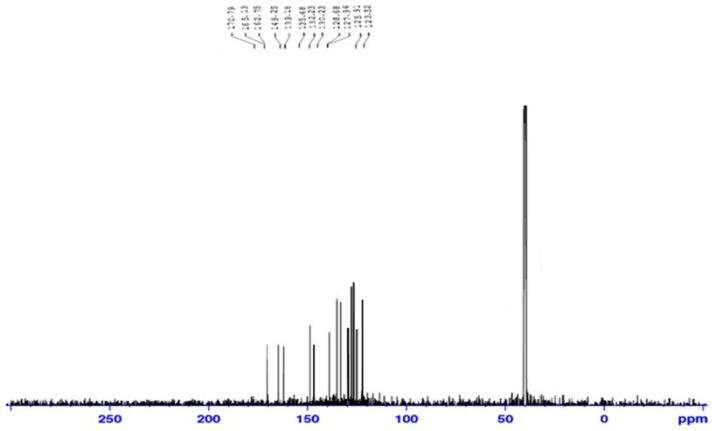

Figure 3s: ^{13}C NMR spectrum of 1a

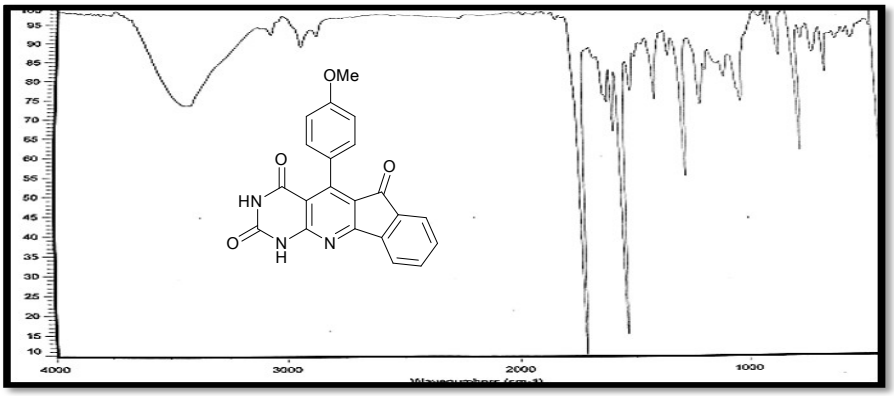

Figure 4s: IR spectrum of 2a

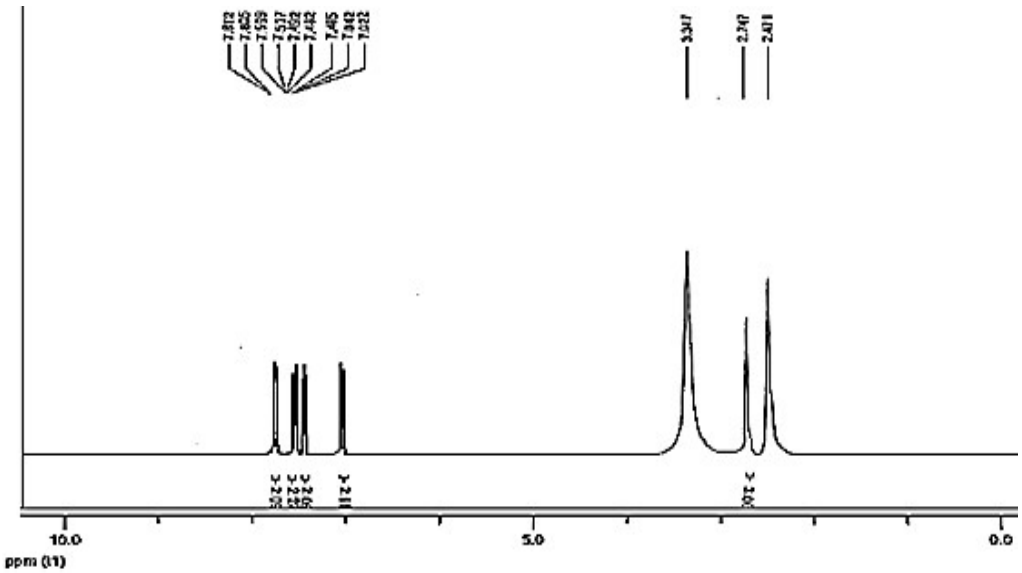

Figure 5s: ^{1}H NMR spectrum of 2a

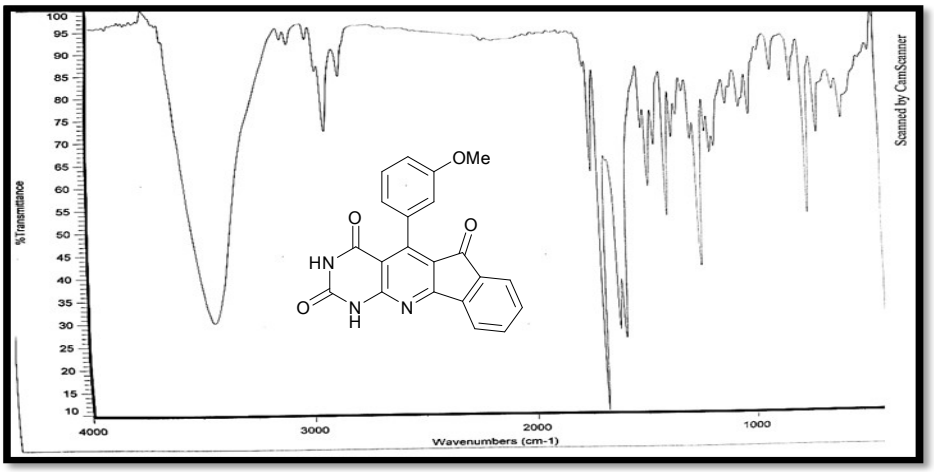

Figure 6s: IR spectrum of 3a

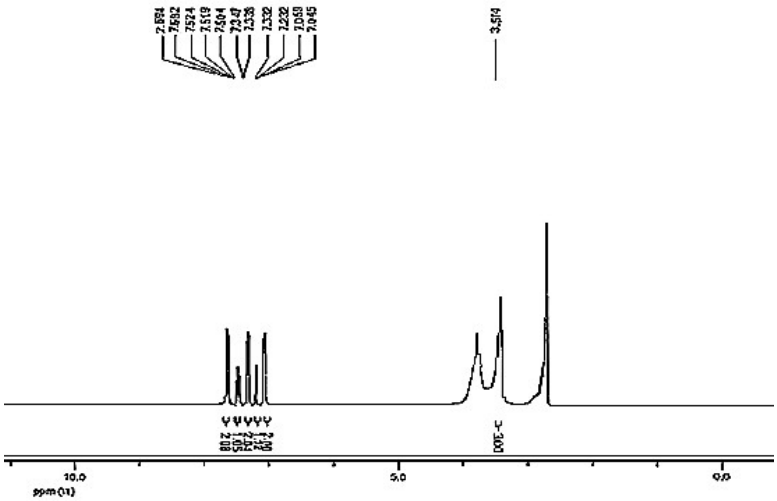

Figure 7s: ^{1}H NMR spectrum of 3a

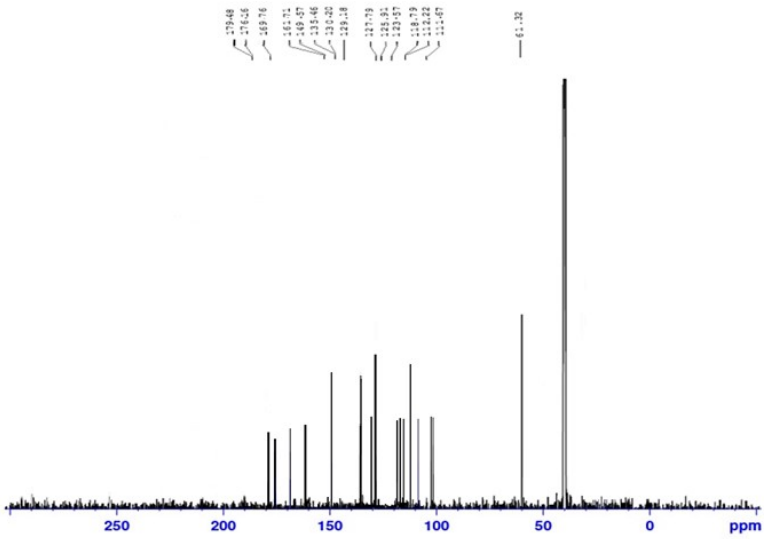

Figure 8s: ^{13}C NMR spectrum of 3a

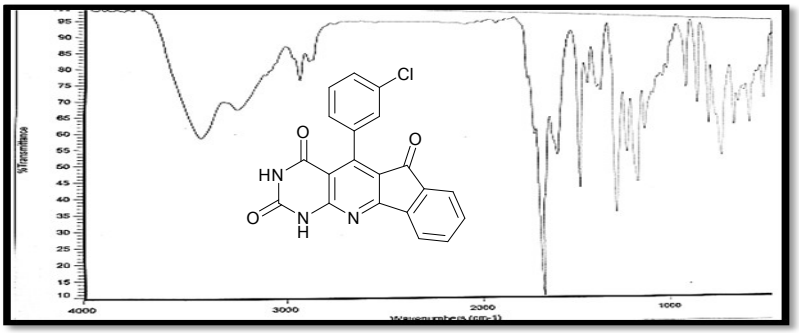

Figure 9s: IR spectrum of 4a

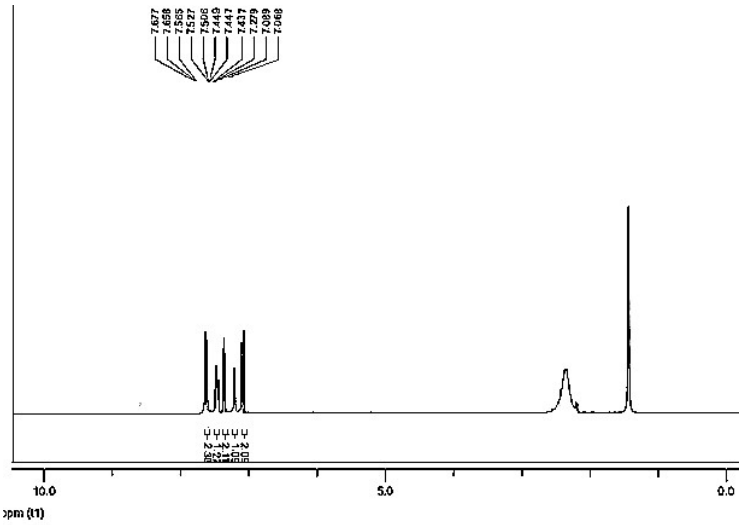

Figure 10s: ^{1}H NMR spectrum of 4a

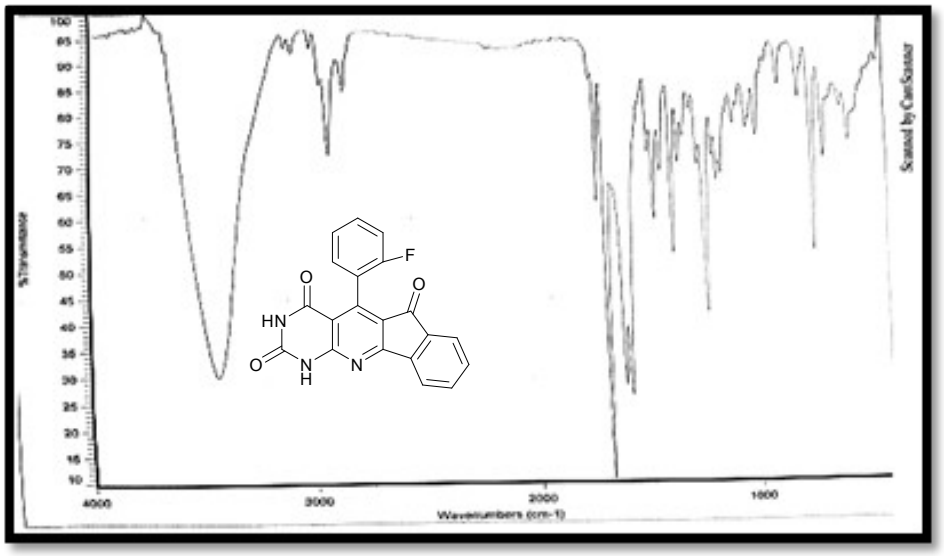

Figure 11s: IR spectrum of 5a

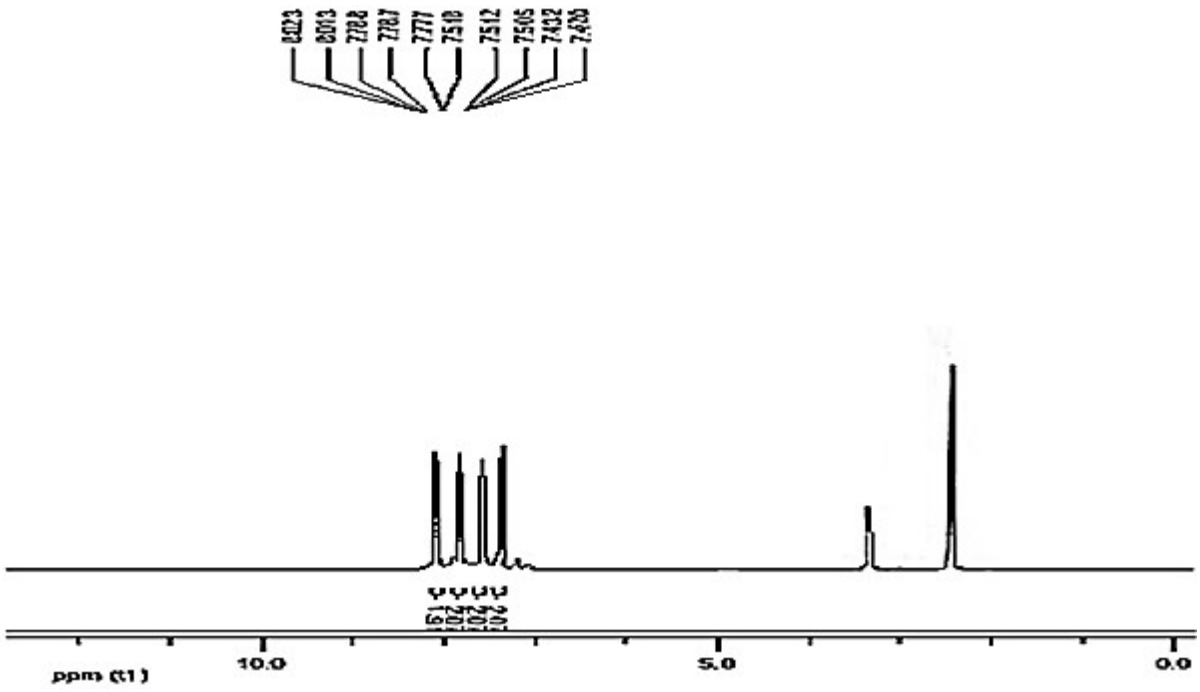

Figure 12s: ^{1}H NMR spectrum of 5a

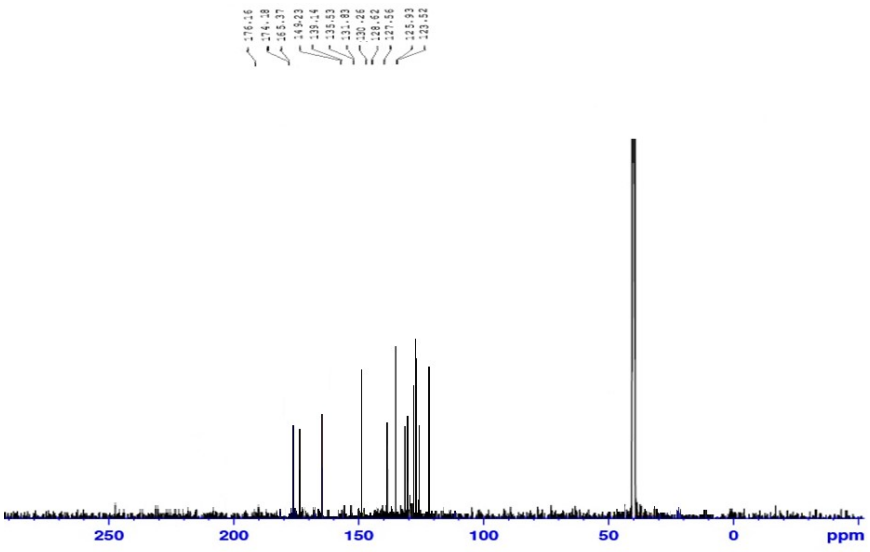

Figure 13s: ^{13}C NMR spectrum of 5a

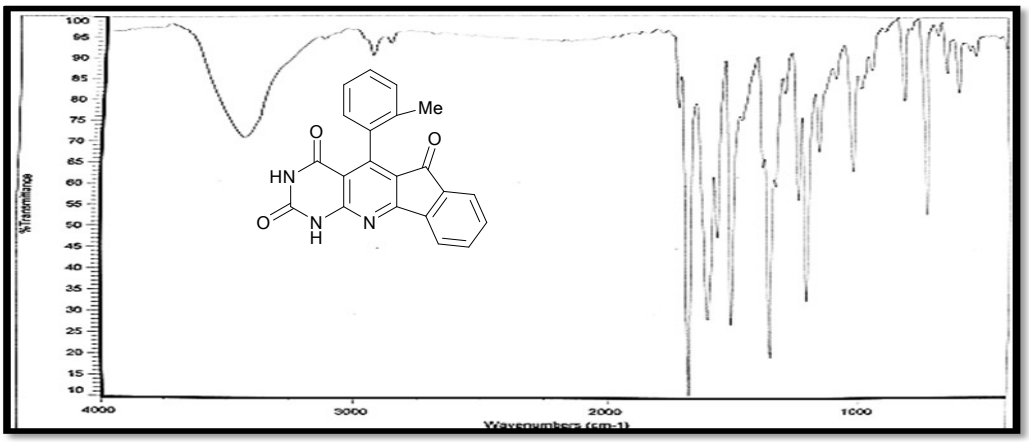

Figure 14s: IR spectrum of 6a

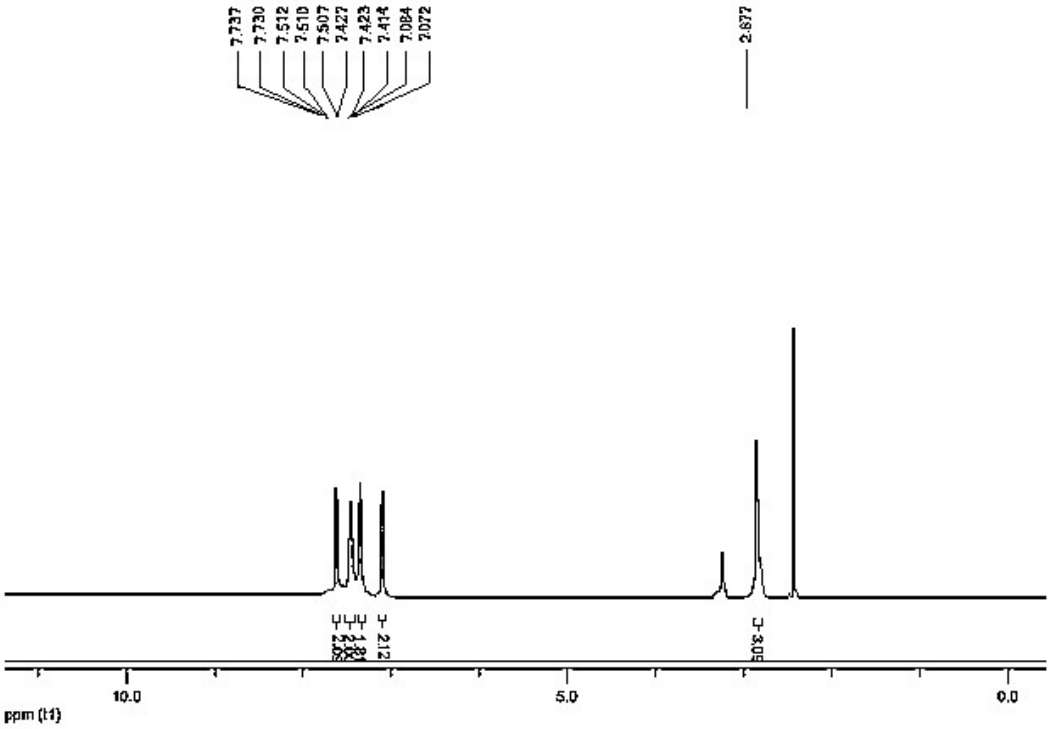

Figure 15s: ^{1}H NMR spectrum of 6a

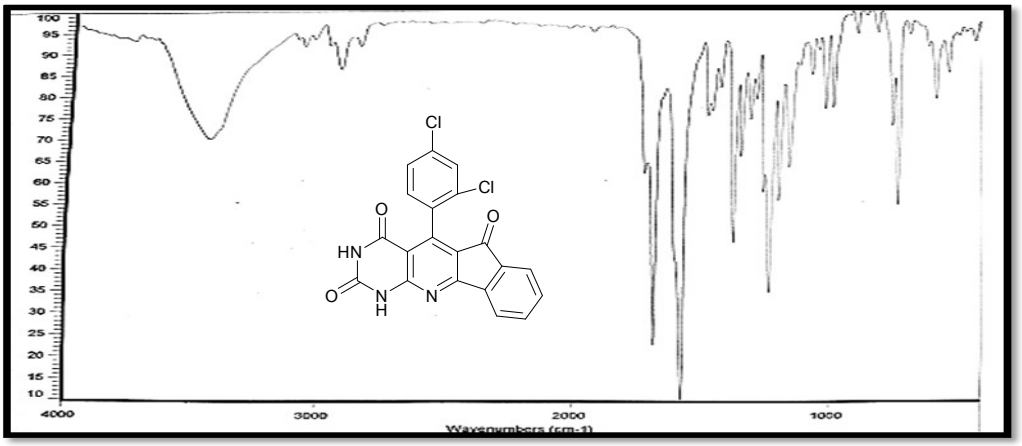

Figure 16s: IR spectrum of 7a

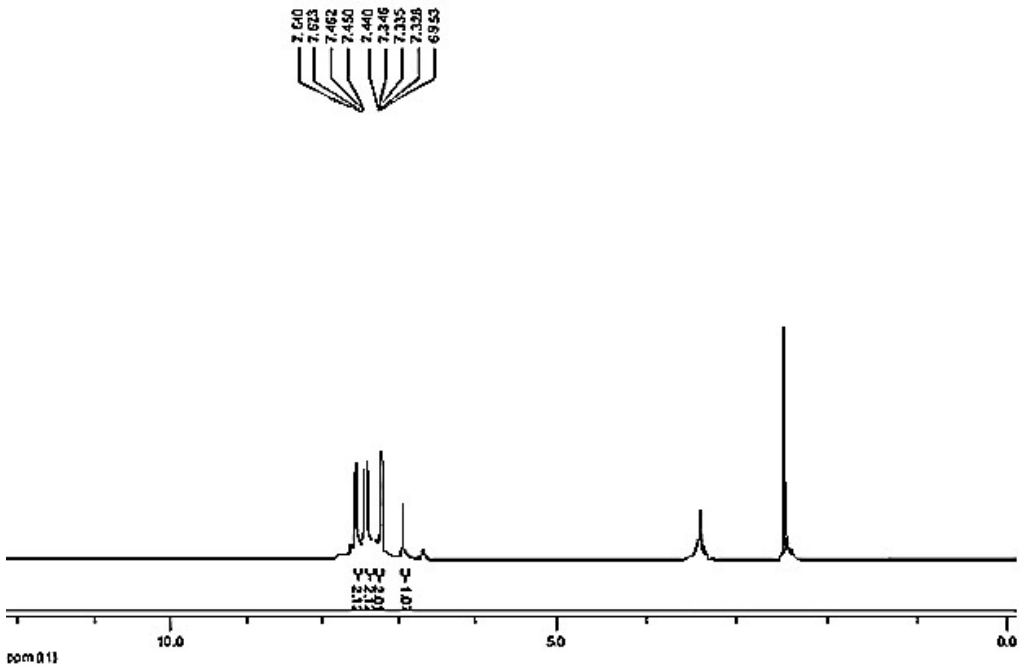

Figure 17s: ^{1}H NMR spectrum of 7a

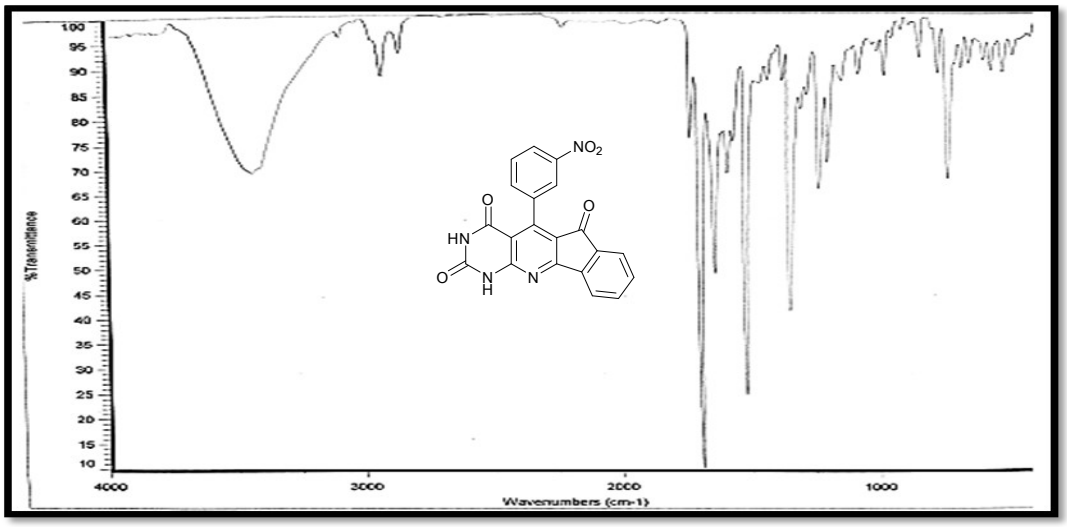

Figure 18s: IR spectrum of 8a

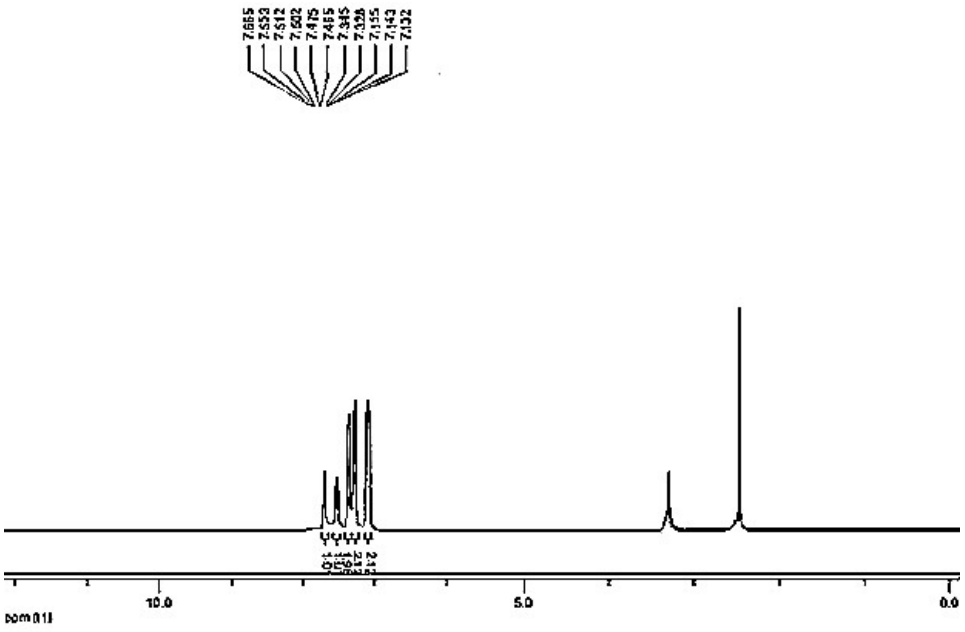

Figure 19s: ^{1}H NMR spectrum of 8a

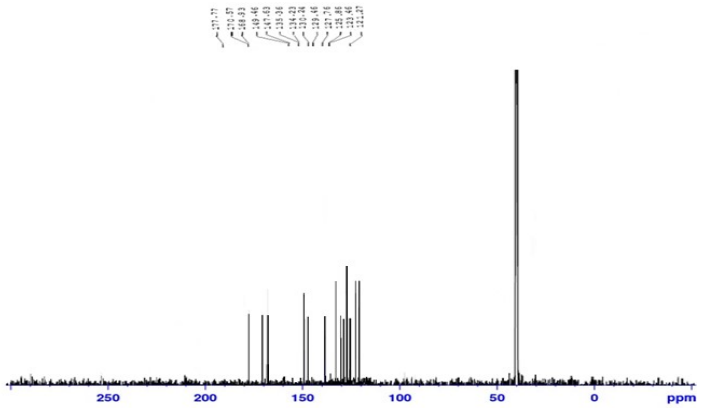

Figure 20s: **^{13}C NMR spectrum of 8a**

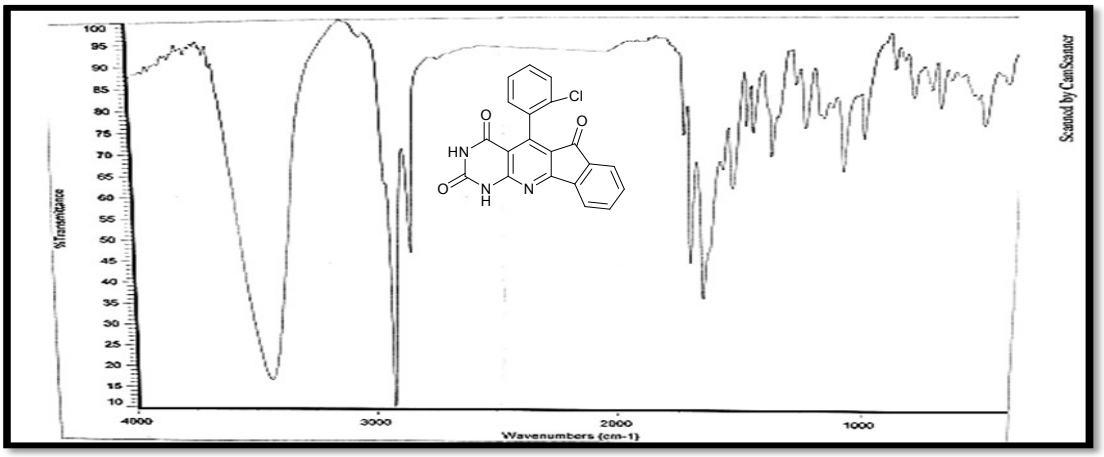

Figure 21s: IR spectrum of 9a

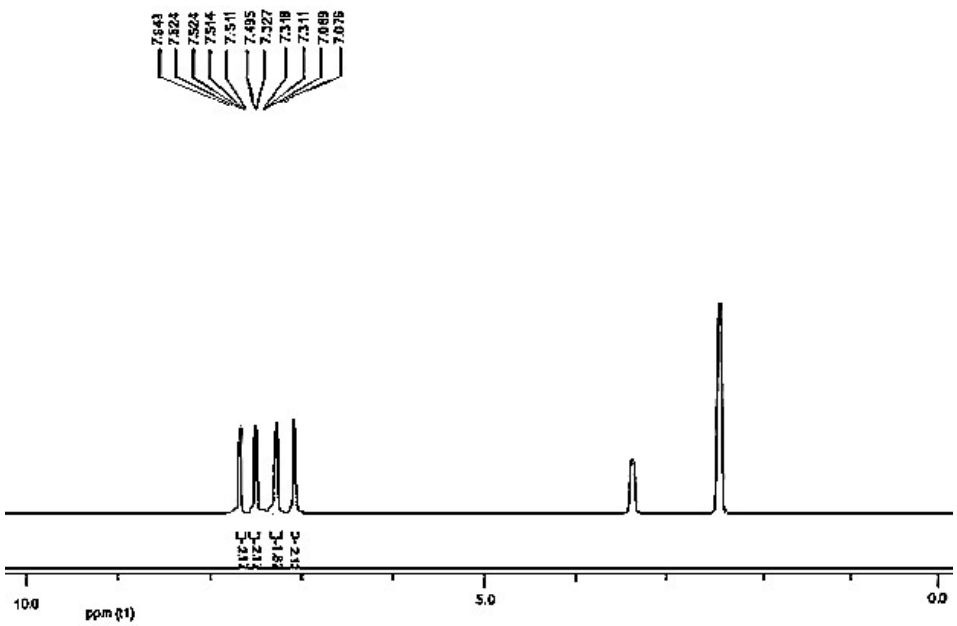

Figure 22s: ^{1}H NMR spectrum of 9a

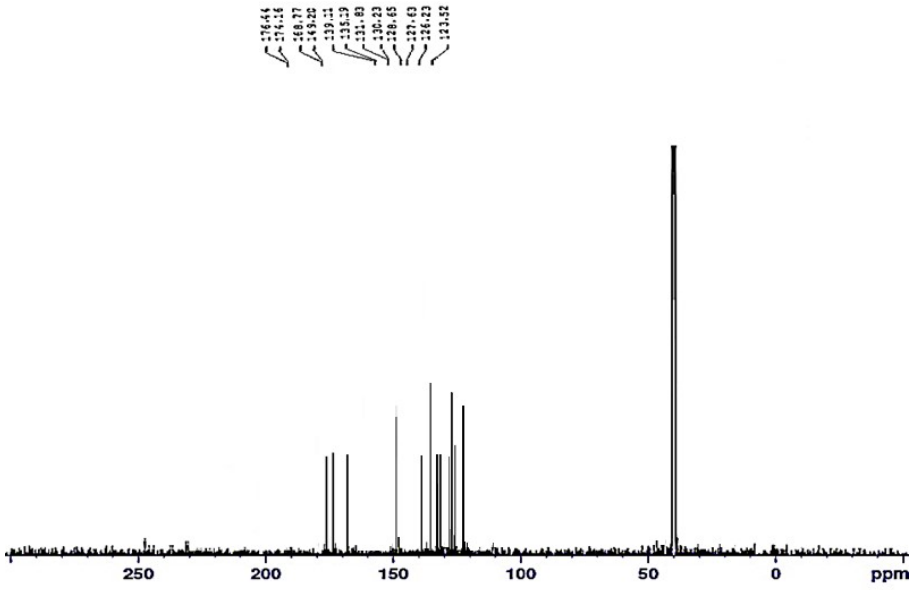

Figure 21s: **^{13}C NMR spectrum of 9a**

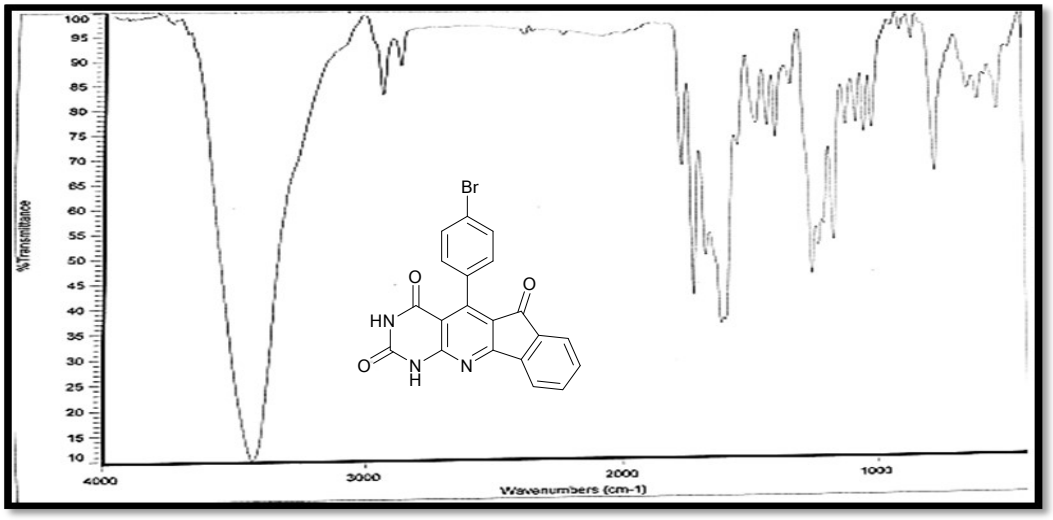

Figure 22s: IR spectrum of 10a

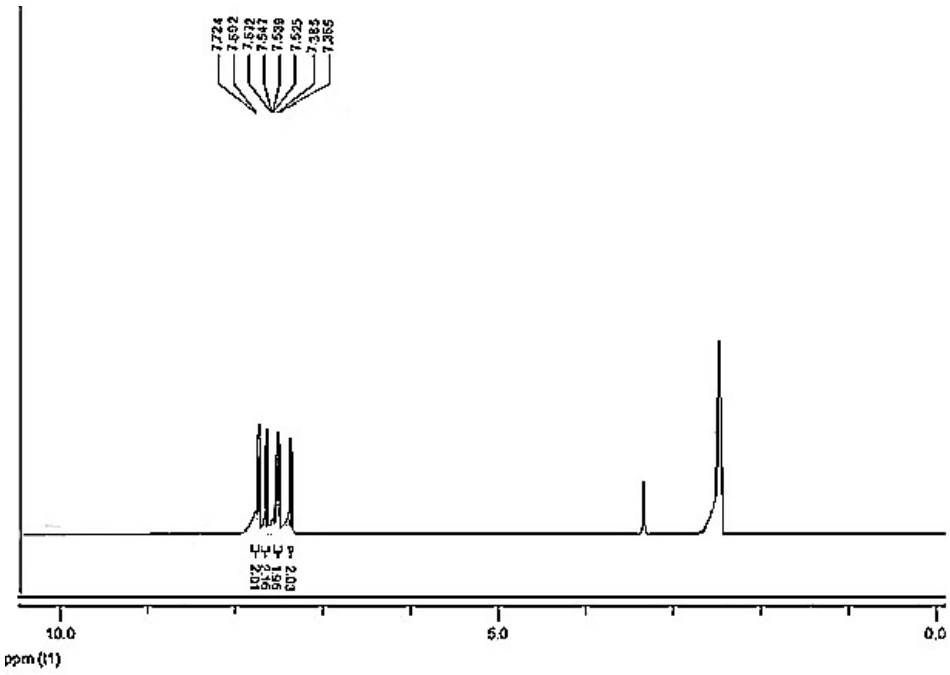

Figure 23s: ^{1}H NMR spectrum of 10a

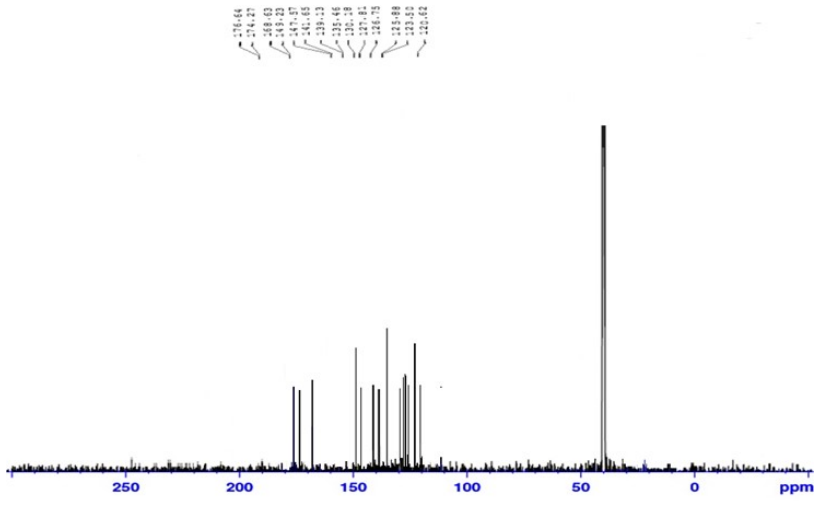

Figure 24s: **^{13}C NMR spectrum of 10a**

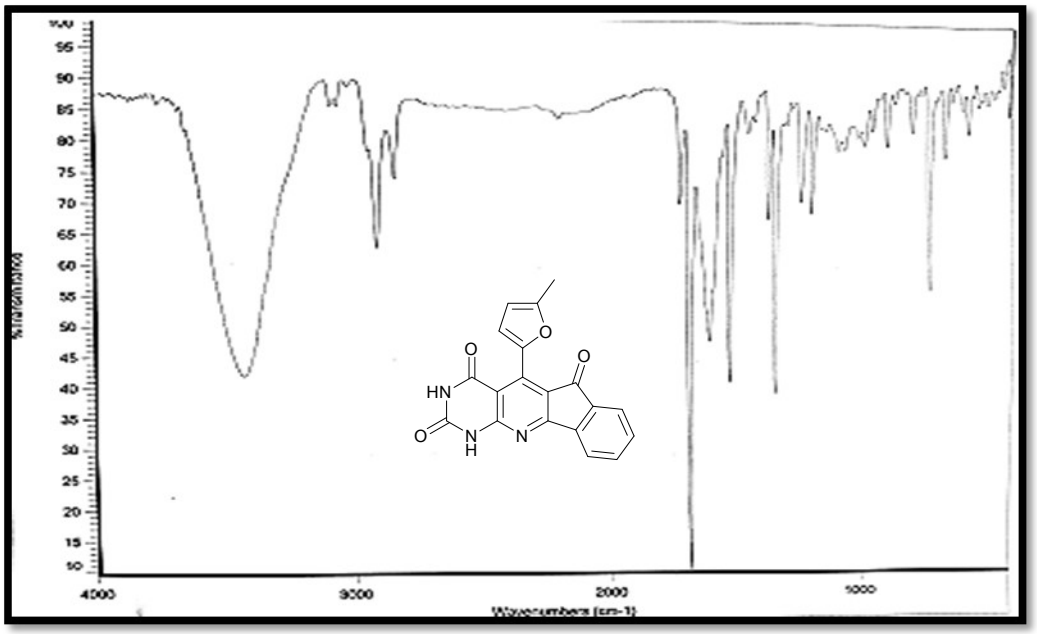

Figure 25s: IR spectrum of 11a

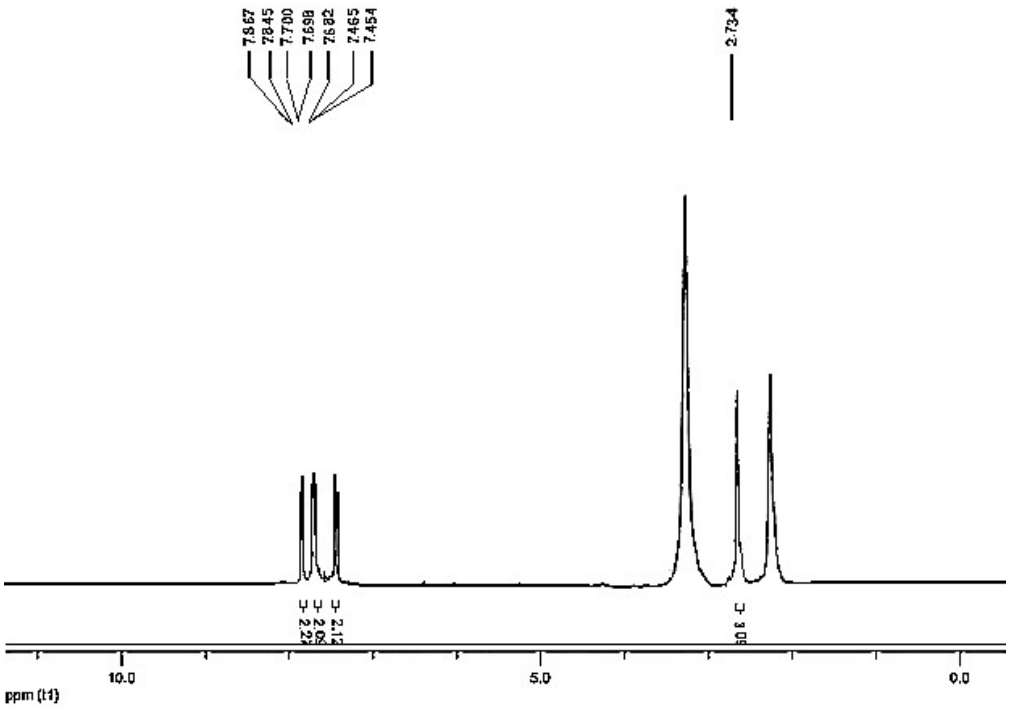

Figure 26s: ^{1}H NMR spectrum of 11a

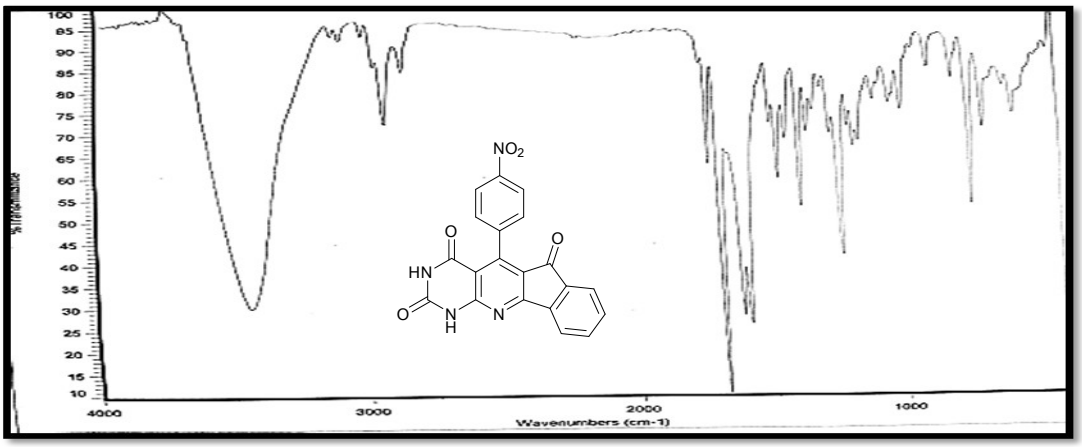

Figure 27s: IR spectrum of 12a

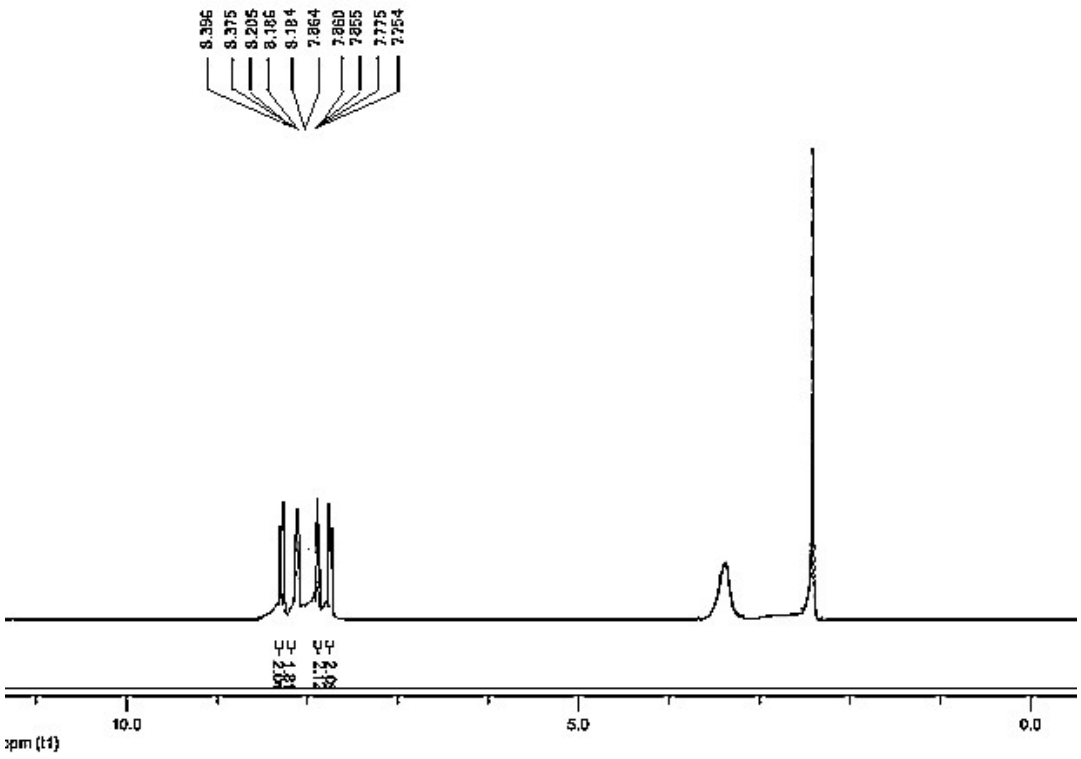

Figure 28s: ^{1}H NMR spectrum of 12a