Electronic Supplementary Information

A one-pot catalyst/external oxidant/solvent-free cascade approach to pyrimidines *via* 1,5-hydride transfer

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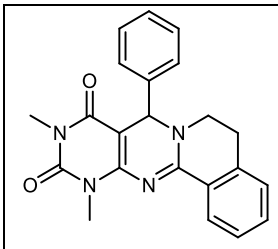
General information

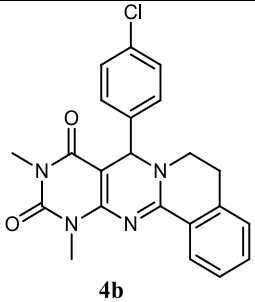
All the commercially available reagents were used as received. Melting points were determined in open capillary tubes with a Buchi-540 micro melting point apparatus and were uncorrected. I.R. spectra were recorded on a Spectrum 2, Perkin-Elmer FT-IR spectrometer. Mass spectra (ESI-HRMS) were recorded on Xevo XS Q-Tof mass spectrometer, Waters ACQUITY UHPLC. NMR spectra were recorded on a Bruker Avance III 500, 400 & 300 MHz NMR spectrometer with TMS as the internal standard at room temperature. Chemical shifts (δ) are quoted in ppm and coupling constants (*J*) are measured in Hertz (Hz). All the experiments were monitored by thin layer chromatography (TLC) on pre-coated silica gel plates (Merck) and visualized under UV lamp at 254 nm for UV active materials. Further visualization was achieved by staining with iodine vapor. Column chromatography wherever applicable, was performed on silica gel (100-200 mesh, Merck) using ethyl acetate/hexane as eluent.

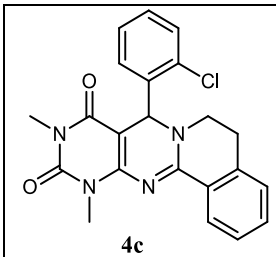
The representative procedure for the synthesis of 4a: 1,3-dimethyl-6-aminouracil (**1a**, 1 mmol, 155 mg), benzaldehyde (**2a**, 1 mmol, 106 mg) and tetrahydroisoquinoline (**3a**, 1 mmol, 133 mg) were added in a round-bottom flask. The reaction mixture was heated at 130 ^{o}C under stirring. Progress of the reaction was monitored by TLC. After 1 hour, the reaction was completed. The reaction mixture was cooled down to room temperature and added ethanol (5 mL). Stirred it for 30 minutes and filtered the solid to obtain the pure product as yellow solid.

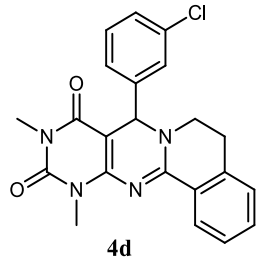


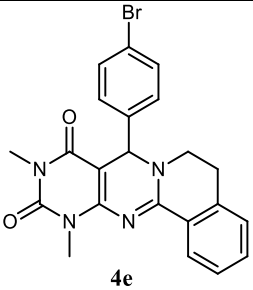
4a

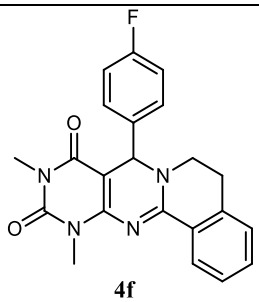
10,12-dimethyl-8-phenyl-10,12-dihydro-5H-pyrimido[4',5':4,5] pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione: Yellow solid; Yield 92 %, 342 mg; M.P.: 210-212 ^{o}C; R_f = 0.5 (hexane/EtOAc, 1:1); IR (KBr): 3037, 2925, 1689, 1645, 1582, 1456, 1083, 779, 564 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 8.27 (d, *J* = 7.3 Hz, 1H), 7.55-7.52 (m, 1H), 7.45-7.39 (m, 3H), 7.34-7.26 (m, 4H), 5.63 (s, 1H), 3.73-3.67 (m, 1H), 3.52 (s, 3H), 3.25-3.16 (m, 1H), 3.10 (s, 3H), 3.05-2.99 (m, 1H), 2.89-2.83 (m, 1H); ^{13}C NMR (125 MHz, DMSO-d_6): δ 159.8, 156.4, 151.6, 150.2, 141.9, 137.1, 132.5, 128.7, 128.3 (2C), 127.8, 127.7, 127.1, 127.0, 88.1, 60.0, 46.0, 29.0, 27.3, 26.8; HRMS (ESI) exact mass calculated for $C_{22}H_{20}N_4O_2$ $[M+H]^+$: 373.1659; found: 373.1667.

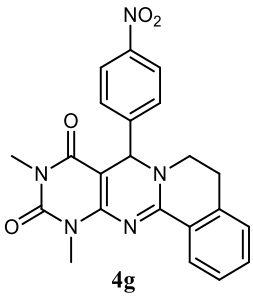
 **4b**	**8-(4-chlorophenyl)-10,12-dimethyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 94 %, 382 mg; M.P.: 268-270 ^{o}C; R_f = 0.5 (hexane/EtOAc, 1:1); IR (KBr): 3041, 2927, 1643, 1584, 1462, 1317, 1162, 1088, 778, 755 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.27 (d, *J* = 7.9 Hz, 1H), 7.45-7.40 (m, 1H), 7.36-7.32 (m, 3H), 7.22-7.19 (m, 2H), 7.13 (d, *J* = 7.2 Hz, 1H), 5.51 (s, 1H), 3.57 (s, 3H), 3.47-3.42 (m, 1H), 3.34-3.26 (m, 1H), 3.20 (s, 3H), 2.97-2.90 (m, 1H), 2.84-2.79 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.9, 156.9, 152.3, 150.5, 140.4, 136.3, 134.3, 132.6, 129.0, 128.6, 128.4, 128.3, 127.5, 127.4, 89.0, 60.9, 46.5, 29.3, 27.7 (2C); HRMS (ESI) exact mass calculated for $C_{22}H_{19}ClN_4O_2$ $[M+H]^+$: 407.1269; found: 407.1274.

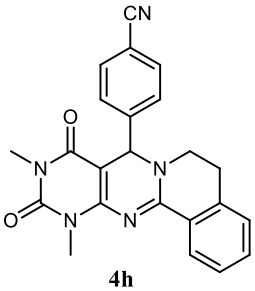
 **4c**	**8-(2-chlorophenyl)-10,12-dimethyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 90 %, 365 mg; M.P.: 204-206 ^{o}C; R_f = 0.5 (hexane/EtOAc, 1:1); IR (KBr): 3051, 2926, 2854, 1648, 1582, 1455, 1316, 1159, 1082, 779, 734 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.39 (d, *J* = 7.6 Hz, 1H), 8.20 (d, *J* = 8.5 Hz, 2H), 7.68 (d, *J* = 8.5 Hz, 2H), 7.55-7.52 (m, 1H), 7.47-7.44 (m, 1H), 7.29-7.23 (m, 1H), 5.76 (s, 1H), 3.66 (s, 3H), 3.61-3.56 (m, 1H), 3.39-3.34 (m, 1H), 3.28 (s, 3H), 3.09-3.03 (m, 1H), 2.96-2.92 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.9, 157.2, 152.2, 150.6, 148.4, 147.8, 136.2, 132.9, 128.6, 128.3, 128.0, 127.5 (2C), 127.4, 124.1, 123.3, 88.4, 61.0, 46.8, 29.3, 27.7, 27.6; HRMS (ESI) exact mass calculated for $C_{22}H_{19}ClN_4O_2$ $[M+H]^+$: 407.1269; found: 407.1276.

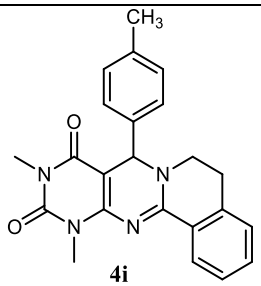
 **4d**	**8-(3-chlorophenyl)-10,12-dimethyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione):** Yellow solid; Yield 93 %, 378 mg; M.P.: 139-141 ^{o}C; R_f = 0.5 (hexane/EtOAc, 1:1); IR (KBr): 3047, 2945, 1686, 1643, 1582, 1458, 1318, 1241, 1158, 1079, 779, 738, 693 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.36-8.34 (m, 1H), 7.51-7.48 (m, 1H), 7.43-7.40 (m, 2H), 7.37-7.35 (m, 1H), 7.26-7.24 (m, 2H), 7.21 (d, *J* = 7.3 Hz, 1H), 5.58 (s, 1H), 3.65 (s, 3H), 3.55-3.50 (m, 1H), 3.40-3.34 (m, 1H), 3.29 (s, 3H), 3.05-2.99 (m, 1H), 2.92-2.87 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.9, 156.9, 152.3, 150.6, 150.4, 147.0, 136.3, 132.6, 128.8, 128.7, 128.5, 127.4, 127.3, 127.2, 125.4, 124.4, 88.8, 61.2, 46.5, 29.3, 27.7, 27.6; HRMS (ESI) exact mass calculated for $C_{22}H_{19}ClN_4O_2$ $[M+H]^+$: 407.1269; found: 407.1275.

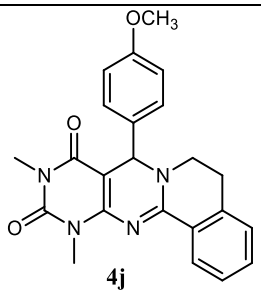
 **4e**	**8-(4-bromophenyl)-10,12-dimethyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 96 %, 433 mg; M.P.: 266-268 ^{o}C; R_f = 0.5 (hexane/EtOAc, 1:1); IR (KBr): 3068, 2926, 1643, 1461, 1244, 1162, 1071, 848, 778 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.35 (d, J = 7.8 Hz, 1H), 7.51-7.48 (m, 1H), 7.45-7.40 (m, 3H), 7.35 (d, J = 8.4 Hz, 2H), 7.20 (d, J = 7.5 Hz, 1H), 5.57 (s, 1H), 3.64 (s, 3H), 3.55-3.49 (m, 1H), 3.39-3.33 (m, 1H), 3.28 (s, 3H), 3.04-2.98 (m, 1H), 2.92-2.86 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.9, 156.9, 152.3, 150.4, 140.9, 136.3, 132.6, 131.9, 128.8, 128.6, 128.4, 127.4, 122.6, 88.9, 61.0, 46.5, 29.3, 27.7 (2C); HRMS (ESI) exact mass calculated for $C_{22}H_{19}BrN_4O_2$ $[M+H]^+$: 451.0764; found: 451.0771.

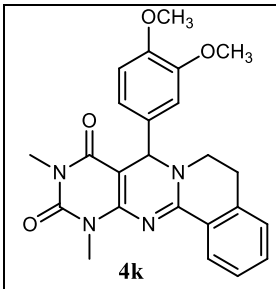
 **4f**	**8-(4-fluorophenyl)-10,12-dimethyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 95 %, 371 mg; M.P.: 270-272 ^{o}C; R_f = 0.5 (hexane/EtOAc, 1:1); IR (KBr): 3031, 2927, 1644, 1467, 1087, 798, 562 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.35 (d, J = 7.9 Hz, 1H), 7.51-7.48 (m, 1H), 7.45-7.40 (m, 3H), 7.20 (d, J = 7.5 Hz, 1H), 7.01-6.98 (m, 2H), 5.59 (s, 1H), 3.65 (s, 3H), 3.55-3.50 (m, 1H), 3.39-3.34 (m, 1H), 3.28 (s, 3H), 3.04-2.98 (m, 1H), 2.91-2.86 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 162.7 (d, $J_{C\text{-}F}$ = 247.1 Hz), 161.0, 156.8, 152.4, 150.4, 137.8 (d, $J_{C\text{-}F}$ = 2.7 Hz), 136.3, 132.5, 128.8 (d, $J_{C\text{-}F}$ = 8.2 Hz), 128.7, 128.4, 127.4, 115.7 (d, $J_{C\text{-}F}$ = 21.8 Hz), 89.2, 60.9, 46.5, 29.3, 27.7 (2C); HRMS (ESI) exact mass calculated for $C_{22}H_{19}FN_4O_2$ $[M+H]^+$: 391.1565; found: 391.1577.

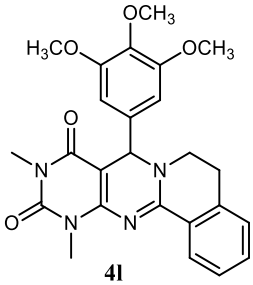
 **4g**	**10,12-dimethyl-8-(4-nitrophenyl)-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione :** Yellow solid; Yield 98 %, 409 mg; M.P.: 245-247 ^{o}C; R_f = 0.4 (hexane/EtOAc, 1:1); IR (KBr): 3061, 2923, 2860, 1641, 1525, 1458, 1311, 1246, 1161, 782, 744, 544 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.27 (d, J = 7.8 Hz, 1H), 7.47-7.45 (m, 1H), 7.41-7.37 (m, 1H), 7.33-7.30 (m, 1H), 7.29-7.27 (m, 1H), 7.18-7.12 (m, 2H), 7.09 (d, J = 7.6 Hz, 1H), 6.12 (s, 1H), 3.60 (s, 3H), 3.54-3.49 (m, 1H), 3.25-3.21 (m, 1H), 3.19 (s, 3H), 2.94-2.88 (m, 1H), 2.76-2.70 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.8, 156.5, 152.4, 151.1, 139.2, 136.4, 132.7, 132.4, 130.5, 129.7, 128.7, 128.4, 127.8, 127.3, 127.2, 88.6, 56.0, 45.7, 29.3, 27.6, 27.5; HRMS (ESI) exact mass calculated for $C_{22}H_{19}N_5O_4$ $[M+H]^+$: 418.1510; found: 418.1514.

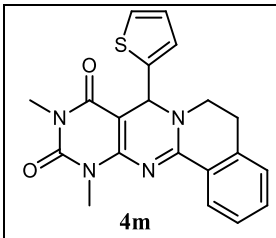
 **4h**	**4-(10,12-dimethyl-9,11-dioxo-6,8,9,10,11,12-hexahydro-5H-pyrimido[4',5':4,5]pyrimido[2,1-a]isoquinolin-8-yl)benzonitrile:** Yellow solid; Yield 96 %, 381 mg; M.P.: 266-268 ^{o}C; R_f = 0.4 (hexane/EtOAc, 1:1); IR (KBr): 3053, 2925, 2854, 2227, 1644, 1453, 1303, 1242, 1160, 1081, 782, 657 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 8.36 (d, J = 6.7 Hz, 1H), 7.62-7.60 (m, 4H), 7.52-7.44 (m, 2H), 7.27-7.22 (m, 1H), 5.68 (s, 1H), 3.64 (s, 3H), 3.55-3.53 (m, 1H), 3.34-3.32 (m, 1H), 3.28 (s, 3H), 3.06-2.90 (m, 2H); ^{13}C NMR (125 MHz, DMSO-d_6): δ 160.9, 157.2, 152.2, 150.6, 146.6, 136.2, 132.8, 132.7, 128.5, 128.4, 127.8, 127.5, 118.4, 112.3, 88.5, 61.3, 46.8, 29.3, 27.7, 27.6; HRMS (ESI) exact mass calculated for $C_{23}H_{19}N_5O_2$ $[M+H]^+$: 398.1612; found: 398.1622.

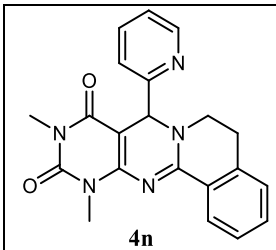
 **4i**	**10,12-dimethyl-8-(p-tolyl)-10,12-dihydro-5H-pyrimido[4',5':4,5] pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 93 %, 359 mg; M.P.: 244-246 ^{o}C; R_f = 0.5 (hexane/EtOAc, 1:1); IR (KBr): 3045, 2923, 2860, 1641, 1525, 1458, 1311, 1246, 1161, 782, 744 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.35 (d, J = 7.9 Hz, 1H), 7.50-7.46 (m, 1H), 7.42-7.39 (m, 1H), 7.35 (d, J = 8.2 Hz, 2H), 7.19 (d, J = 6.9 Hz, 1H), 7.12 (d, J = 7.8 Hz, 2H), 5.56 (s, 1H), 3.65 (s, 3H), 3.54-3.49 (m, 1H), 3.43-3.36 (m, 1H), 3.27 (s, 3H), 3.03-2.96 (m, 1H), 2.91-2.85 (m, 1H), 2.29 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.9, 156.7, 152.4, 150.4, 139.0, 138.3, 136.3, 132.4, 129.4, 128.8, 128.4, 127.3 (2C), 126.9, 89.4, 61.2, 46.4, 29.2, 27.7 (2C), 21.1; HRMS (ESI) exact mass calculated for $C_{23}H_{22}N_4O_2$ $[M+H]^+$: 387.1816; found: 387.1818.

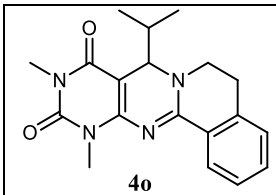
 **4j**	**8-(4-methoxyphenyl)-10,12-dimethyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 92 %, 370 mg; M.P.: 179-192 ^{o}C; R_f = 0.4 (hexane/EtOAc, 1:1); IR (KBr): 3035, 2933, 2837, 1684, 1642, 1457, 1247, 1161, 1030, 756, 745 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.35 (d, J = 7.9 Hz, 1H), 7.48-7.46 (m, 1H), 7.42-7.37 (m, 3H), 7.19 (d, J = 7.5 Hz, 1H), 6.83 (d, J = 8.7 Hz, 2H), 5.54 (s, 1H), 3.76 (s, 3H), 3.65 (s, 3H), 3.54-3.49 (m, 1H), 3.42-3.36 (m, 1H), 3.28 (s, 3H), 3.03-2.96 (m, 1H), 2.91-2.85 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 161.0, 159.6, 156.6, 152.4, 150.3, 136.3, 134.3, 132.4, 128.9, 128.4, 128.3, 127.3 (2C), 114.1, 89.5, 60.9, 55.2, 46.3, 29.3, 27.7 (2C); HRMS (ESI) exact mass calculated for $C_{23}H_{22}N_4O_3$ $[M+H]^+$: 403.1765; found: 403.1774.

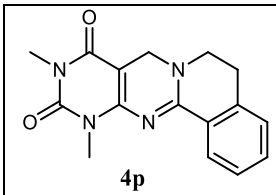
 **4k**	**8-(3,4-dimethoxyphenyl)-10,12-dimethyl-10,12-dihydro-5H-pyrimido[4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 90 %, 389 mg; M.P.: 130-132 ^{o}C; R_f = 0.3 (hexane/EtOAc, 1:1); IR (KBr): 3049, 2927, 2854, 1692, 1642, 1517, 1455, 1237, 1139, 1026, 741 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.35 (d, *J* = 7.6 Hz, 1H), 7.50-7.47 (m, 1H), 7.42-7.39 (m, 1H), 7.20 (d, *J* = 7.5 Hz, 1H), 7.01-6.99 (m, 2H), 6.79 (d, *J* = 8.7 Hz, 1H), 5.54 (s, 1H), 3.83 (s, 3H), 3.83(s, 3H), 3.65 (s, 3H), 3.56-3.51 (m, 1H), 3.44-3.38 (m, 1H), 3.30 (s, 3H), 3.03-2.97 (m, 1H), 2.92-2.87 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 161.1, 156.7, 152.4, 150.4, 149.1, 149.0, 136.3, 134.7, 132.4, 128.8, 128.3, 127.3 (2C), 119.4, 111.1, 110.2, 89.4, 61.2, 56.0, 55.8, 46.3, 29.3, 27.7; HRMS (ESI) exact mass calculated for $C_{24}H_{24}N_4O_4$ $[M+H]^+$: 433.1870; found: 433.1883.

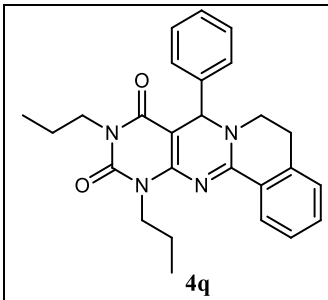
 **4l**	**10,12-dimethyl-8-(3,4,5-trimethoxyphenyl)-10,12-dihydro-5H-pyrimido[4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 90 %, 416 mg; M.P.: 181-183 ^{o}C; R_f = 0.25 (hexane/EtOAc, 1:1); IR (KBr): 3033, 2936, 2832, 1688, 1644, 1465, 1281, 1160, 1085, 1009, 736 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.31 (d, *J* = 7.9 Hz, 1H), 7.45-7.42 (m, 1H), 7.38-7.35 (m, 1H), 7.15 (d, *J* = 7.5 Hz, 1H), 7.07 (d, *J* = 8.7 Hz, 1H), 6.62 (d, *J* = 8.7 Hz, 1H), 5.90 (s, 1H), 4.01 (s, 3H), 3.87 (s, 3H), 3.81(s, 3H), 3.69 (s, 3H), 3.62-3.57 (m, 1H), 3.40-3.34 (m, 1H), 3.30 (s, 3H), 3.01-2.92 (m, 1H), 2.81-2.76 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 161.0, 156.3, 153.7, 152.5, 151.3, 141.8, 136.5, 132.1, 129.1, 128.2, 127.6, 127.2, 127.1, 123.9, 107.7, 88.3, 61.4, 60.8, 55.8, 45.5, 29.3, 27.7, 27.6; HRMS (ESI) exact mass calculated for $C_{25}H_{26}N_4O_5$ $[M+H]^+$: 463.1976; found: 463.1986.

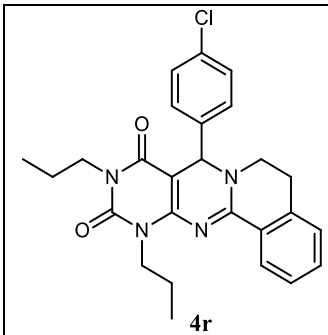
 **4m**	**10,12-dimethyl-8-(thiophen-2-yl)-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 95 %, 359 mg; M.P.: 192-194 ^{o}C; R_f = 0.4 (hexane/EtOAc, 1:1); IR (KBr): 3089, 2944, 1642, 1455, 1317, 1248, 1162, 1049, 783, 751 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.34 (d, *J* = 7.8 Hz, 1H), 7.49-7.46 (m, 1H), 7.41-7.38 (m, 1H), 7.21-7.18 (m, 2H) 7.14 (d, *J* = 3.4 Hz, 1H), 6.92-6.91 (m, 1H), 5.92 (s, 1H), 3.65 (s, 3H), 3.62-3.59 (m, 2H), 3.32 (s, 3H), 3.07-2.96 (m, 2H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.9, 156.4, 152.3, 150.2, 145.0, 136.3, 132.4, 128.8, 128.4, 127.3 (2C), 126.8, 125.6, 125.5, 89.2, 56.3, 46.6, 29.3, 27.8; HRMS (ESI) exact mass calculated for $C_{20}H_{18}N_4O_2S$ $[M+H]^+$: 379.1223; found: 379.1232.

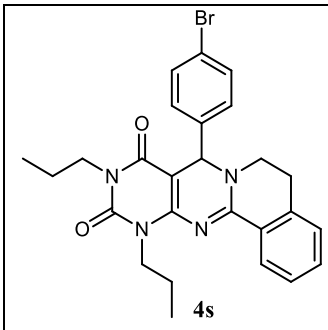
 **4n**	**10,12-dimethyl-8-(pyridin-2-yl)-10,12-dihydro-5H-pyrimido[4',5':4,5] pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 91 %, 339 mg; M.P.: 230-233 ^{o}C; R_f = 0.3 (hexane/EtOAc, 1:1); IR (KBr): 3046, 2925, 1687, 1633, 1584, 1456, 1323, 1160, 1081, 995, 766 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.53-8.52 (m, 1H), 8.38 (d, *J* = 7.8 Hz, 1H), 7.68-7.64 (m, 1H), 7.53-7.51 (m, 1H), 7.49-7.46 (m, 1H), 7.41-7.38 (m, 1H), 7.20-7.17 (m, 2H), 5.78 (s, 1H), 3.64 (s, 3H), 3.59-3.53 (m, 1H), 3.46-3.41 (m, 1H), 3.30 (s, 3H), 3.04-2.90 (m, 2H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 161.1, 159.1, 157.7, 152.4, 151.4, 149.8, 136.8, 136.6, 132.3, 128.9, 128.6, 127.2 (2C), 123.3, 122.0, 87.9, 62.2, 46.8, 29.3, 27.7, 27.6; HRMS (ESI) exact mass calculated for $C_{21}H_{19}N_5O_2$ $[M+H]^+$: 374.1612; found: 374.1612.

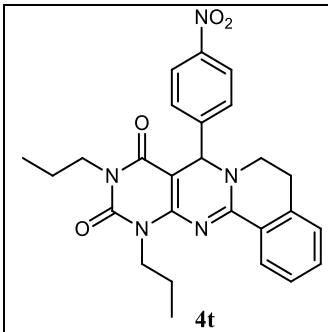
 **4o**	**8-isopropyl-10,12-dimethyl-10,12-dihydro-5H-pyrimido[4',5':4,5] pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow gummy; Yield 72 %, 243 mg; R_f = 0.5 (hexane/EtOAc, 1:1); IR (KBr): 3048, 2933, 1661, 1544, 1447, 1213, 1161, 834, 744 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.27 (d, *J* = 7.5 Hz, 1H), 7.49-7.46 (m, 1H), 7.41-7.37 (m, 1H), 7.24 (d, *J* = 7.6 Hz, 1H), 4.69 (d, *J* = 3.2 Hz, 1H), 3.69-3.65 (m, 1H), 3.61 (s, 3H), 3.38 (s, 3H), 3.18-3.11 (m, 1H), 3.02-2.96 (m, 1H), 2.93-2.84 (m, 1H), 2.11-2.06 (m, 1H), 0.98 (d, *J* = 7.0 Hz, 3H), 0.90 (d, *J* = 6.9 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 161.7, 157.7, 152.5, 152.4, 136.4, 132.2, 128.3, 127.3 (2C), 126.6, 85.8, 62.1, 48.6, 35.6, 29.3, 28.0, 18.6, 17.1; HRMS (ESI) exact mass calculated for $C_{19}H_{22}N_4O_2$ $[M+H]^+$: 339.1816; found: 339.1823.

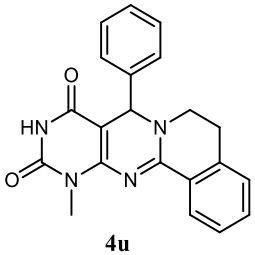
 **4p**	**10,12-dimethyl-10,12-dihydro-5H-pyrimido[4',5':4,5]pyrimido[2,1-a] isoquinoline-9,11(6H,8H)-dione:** Yellow Solid; Yield 77 %, 228 mg; M.P.: 265-269 ^{o}C; R_f = 0.3 (hexane/EtOAc, 1:1); IR (KBr): 3011, 2925, 2854, 1642, 1538, 1458, 1331, 1239, 1162, 763, 740 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.27 (d, *J* = 7.8 Hz, 1H), 7.50-7.46 (m, 1H), 7.39-7.36 (m, 1H), 7.23 (d, *J* = 7.5 Hz, 1H), 4.49 (s, 2H), 2.58 (s, 3H), 3.48-3.47 (m, 2H), 3.35 (s, 3H), 3.08-3.05 (m, 2H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 161.2, 157.9, 152.4, 151.7, 136.2, 132.4, 128.4, 128.2, 127.3, 127.2, 82.9, 47.6 (2C), 29.7, 27.7, 27.0; HRMS (ESI) exact mass calculated for $C_{16}H_{16}N_4O_2$ $[M+H]^+$: 297.1346; found: 297.1355.

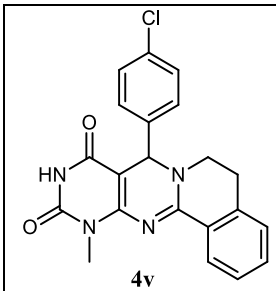
 **4q**	**8-phenyl-10,12-dipropyl-10,12-dihydro-5H-pyrimido[4',5':4,5] pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 82 %, 351 mg; M.P.: 113-115 ^{o}C; R_f = 0.8 (hexane/EtOAc, 1:1); IR (KBr): 3061, 3056, 2961, 2873, 1692, 1642, 1524, 1454, 1305, 1245, 1155, 1077, 765, 698 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.33 (d, *J* = 7.8 Hz, 1H), 7.51-7.42 (m, 4H), 7.35-7.27 (m, 3H), 7.21 (d, *J* = 7.5 Hz, 1H), 5.60 (s, 1H), 4.24 (t, *J* = 7.6 Hz, 2H), 3.84 (t, *J* = 7.2 Hz, 2H), 3.57-3.51 (m, 1H), 3.42-3.37 (m, 1H), 3.04-2.98 (m, 1H), 2.93-2.87 (m, 1H), 1.84-1.79 (m, 2H), 1.68-1.58 (m, 2H), 1.04 (t, *J* = 7.3 Hz, 3H), 0.91 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.9, 156.6, 151.8, 150.3, 142.0, 136.3, 132.3, 128.9, 128.7, 128.4, 128.3, 127.3 (2C), 127.0, 89.4, 61.4, 46.4, 43.8, 42.6, 27.7, 22.1, 21.1, 11.4, 11.3; HRMS (ESI) exact mass calculated for $C_{26}H_{28}N_4O_2$ $[M+H]^+$: 429.2285; found: 429.2395.

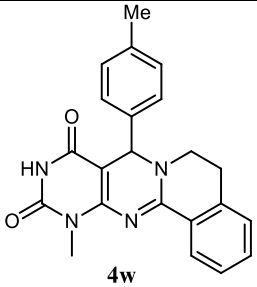
 **4r**	**8-(4-chlorophenyl)-10,12-dipropyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 84 %, 388 mg; M.P.: 96-98 ^{o}C; R_f = 0.8 (hexane/EtOAc, 1:1); IR (KBr): 3072, 3058, 2960, 1694, 1635, 1524, 1454, 1308, 1249, 1159, 1077, 764 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.23 (d, *J* = 7.8 Hz, 1H), 7.42-7.37 (m, 3H), 7.35-7.32 (m, 1H), 7.25-7.20 (m, 2H), 7.11 (d, *J* = 7.0 Hz, 1H), 5.51 (s, 1H), 4.15 (t, *J* = 7.5 Hz, 2H), 3.76-3.73 (m, 2H), 3.47-3.42 (m, 1H), 3.32-3.27 (m, 1H), 2.95-2.90 (m, 1H), 2.83-2.77 (m, 1H), 1.77-1.66 (m, 2H), 1.58-1.47 (m, 2H), 0.94 (t, *J* = 7.5 Hz, 3H), 0.81 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.9, 156.6, 151.8, 150.3, 142.0, 136.3, 132.3, 129.7, 128.9, 128.7, 128.4, 128.3, 127.3 (2C), 127.0, 89.4, 61.4, 46.4, 43.8, 42.6, 27.7, 22.1, 21.1, 11.4, 11.3; HRMS (ESI) exact mass calculated for $C_{26}H_{27}ClN_4O_2$ $[M+H]^+$: 463.1895; found: 463.1906.

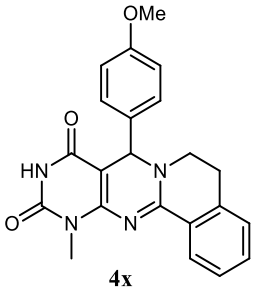
 **4s**	**8-(4-bromophenyl)-10,12-dipropyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 84 %, 425 mg; M.P.: 145-147 ^{o}C; R_f = 0.8 (hexane/EtOAc, 1:1); IR (KBr): 3068, 3041, 2941, 1664, 1582, 1441, 1311, 1240, 1155, 1071, 830, 742 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.22 (d, *J* = 7.8 Hz, 1H), 7.43-7.40 (m, 1H), 7.37-7.33 (m, 3H), 7.27-7.25 (m, 2H), 7.13 (d, *J* = 7.5 Hz, 1H), 5.48 (s, 1H), 4.14 (t, *J* = 7.3 Hz, 2H), 3.77-3.71 (m, 2H), 3.47-3.42 (m, 1H), 3.30-3.25 (m, 1H), 2.95-2.89 (m, 1H), 2.84-2.79 (m, 1H), 1.73-1.67 (m, 2H), 1.58-1.47 (m, 2H), 0.93 (t, *J* = 7.3 Hz, 3H), 0.81 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.8, 156.6, 151.7, 150.3, 141.0, 136.3, 132.4, 131.9, 128.7, 128.3, 127.4, 122.4, 89.0, 60.9, 46.4, 43.9, 42.6, 27.7, 22.1, 21.1, 11.4, 11.3; HRMS (ESI) exact mass calculated for $C_{26}H_{27}BrN_4O_2$ $[M+H]^+$: 507.1390; found: 507.1386.

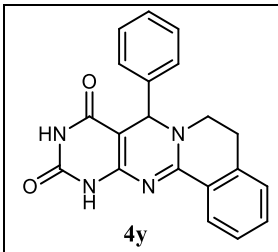
 **4t**	**8-(4-nitrophenyl)-10,12-dipropyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow gummy solid; Yield 83 %, 393 mg; R_f = 0.7 (hexane/EtOAc, 1:1); IR (KBr): 3081, 2927, 2856, 1692, 1642, 1525, 1456, 1347, 1248, 1146, 821, 742 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$): δ 8.33 (d, *J* = 7.8 Hz, 1H), 7.20 (d, *J* = 8.7 Hz, 2H), 7.67 (d, *J* = 8.7 Hz, 2H), 7.55-7.52 (m, 1H), 7.47-7.44 (m, 1H), 7.24 (d, *J* = 7.3 Hz, 1H), 5.74 (s, 1H), 4.24 (t, *J* = 7.5 Hz, 2H), 3.84-3.81 (m, 2H), 3.60-3.55 (m, 1H), 3.39-3.33 (m, 1H), 3.09-3.02 (m, 1H), 2.96-2.89 (m, 1H), 1.87-1.76 (m, 2H), 1.67-1.55 (m, 2H), 1.03 (t, *J* = 7.3 Hz, 3H), 0.90 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.8, 156.9, 151.6, 150.5, 148.6, 147.8, 136.2, 132.7, 128.5, 128.4, 127.9, 127.5 (2C), 124.1, 88.5, 61.0, 46.7, 44.0, 42.7, 27.7, 22.1, 21.1, 11.4, 11.3; HRMS (ESI) exact mass calculated for $C_{26}H_{27}N_5O_4$ $[M+H]^+$: 474.2136; found: 474.2149.

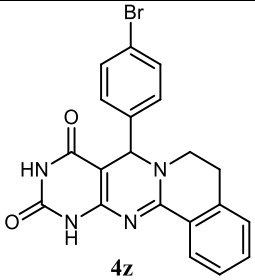
 **4u**	**12-methyl-8-phenyl-10,12-dihydro-5H-pyrimido[4',5':4,5] pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 88 %, 315 mg; M.P.: >300 °C; R_f = 0.35 (hexane/EtOAc, 2:3); IR (KBr): 3361, 3156, 3028, 2815, 1665, 1581, 1524, 1447, 1312, 1243, 1157, 757 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 10.89 (s, 1H), 8.26 (d, *J* = 7.8 Hz, 1H), 7.55-7.52 (m, 1H), 7.44-7.41 (m, 1H), 7.39-7.37 (m, 2H), 7.35-7.27 (m, 4H), 5.59 (s, 1H), 3.71-3.66 (m, 1H), 3.44 (s, 3H), 3.24-3.18 (m, 1H), 3.05-2.99 (m, 1H), 2.87-2.82 (m, 1H); ^{13}C NMR (125 MHz, DMSO-d_6): δ 160.6, 156.3, 151.6, 151.3, 142.0, 137.1, 132.5, 128.7, 128.4, 128.3, 127.8, 127.7, 127.0, 126.9, 88.6, 59.7, 46.0, 28.1, 26.8; HRMS (ESI) exact mass calculated for $C_{21}H_{18}N_4O_2$ $[M+H]^+$: 359.1503; found: 359.1493.

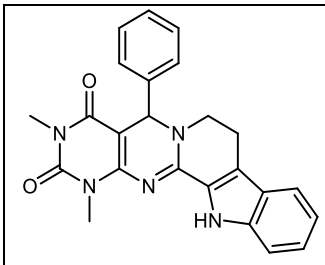
 **4v**	**8-(4-chlorophenyl)-12-methyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 90 %, 353 mg; M.P.: 249-250 °C (dec.); R_f = 0.35 (hexane/EtOAc, 2:3); IR (KBr): 3358, 3165, 3042, 2942, 2942, 1664, 1520, 1440, 1310, 1240, 1155, 1088, 843, 742 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 10.90 (bs, 1H), 8.26-8.24 (m, 1H), 7.55-7.52 (m, 1H), 7.45-7.42 (m, 1H), 7.40-7.39 (m, 4H), 7.31-7.30 (m, 1H), 5.62 (s, 1H), 3.71-3.66 (m, 1H), 3.43 (s, 3H), 3.24-3.19 (m, 1H), 3.04-2.98 (m, 1H), 2.88-2.82 (m, 1H); ^{13}C NMR (125 MHz, DMSO-d_6): δ 160.6, 156.3, 151.6, 140.9, 138.3, 137.1, 132.8, 132.6, 128.9, 128.7, 128.3, 127.9, 127.7, 127.1, 88.3, 59.0, 46.0, 28.1, 26.8; HRMS (ESI) exact mass calculated for $C_{21}H_{17}ClN_4O_2$ $[M+H]^+$: 393.1113; found: 393.1119.

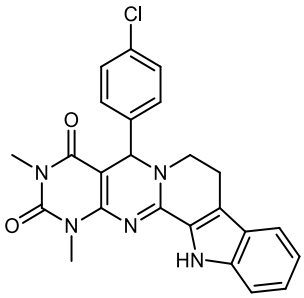
 **4w**	**12-methyl-8-(p-tolyl)-10,12-dihydro-5H-pyrimido[4',5':4,5] pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 88 %, 327 mg; M.P.: >300 ^{o}C; R_f = 0.35 (hexane/EtOAc, 2:3); IR (KBr): 3366, 3182, 3038, 2830, 1660, 1619, 1510, 1390, 1303, 1238, 1066, 773 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 10.86 (s, 1H), 8.25 (d, J = 7.0 Hz, 1H), 7.54-7.51 (m, 1H), 7.45-7.40 (m, 1H), 7.28-7.22 (m, 3H), 7.13 (d, J = 7.9 Hz, 1H), 7.01 (d, J = 7.0 Hz, 1H), 5.53 (s, 1H), 3.69-3.63 (m, 1H), 3.43 (s, 3H), 3.23-3.17 (m, 1H), 3.03-2.94 (m, 1H), 2.86-2.81 (m, 1H), 2.24 (m, 3H); ^{13}C NMR (125 MHz, DMSO-d_6): δ 160.6, 156.2, 151.5, 151.4, 139.2, 137.6, 137.0, 132.4, 128.9, 128.7, 128.4, 127.8, 127.6, 127.0, 88.7, 59.4, 45.9, 28.1, 26.8, 20.7; HRMS (ESI) exact mass calculated for $C_{22}H_{20}N_4O_2$ $[M+H]^+$: 373.1659; found: 373.1668.

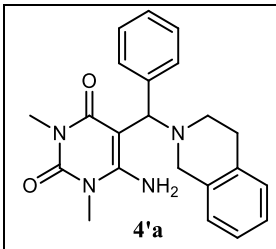
 **4x**	**8-(4-methoxyphenyl)-12-methyl-10,12-dihydro-5H-pyrimido [4',5':4,5]pyrimido[2,1-a]isoquinoline-9,11(6H,8H)-dione:** Yellow solid; Yield 87 %, 337 mg; M.P.: >300 ^{o}C; R_f = 0.3 (hexane/EtOAc, 2:3); IR (KBr): 3483, 3024, 2928, 1670, 1514, 1443, 1318, 1246, 1178, 1033, 845, 745 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 10.87 (s, 1H), 8.25 (d, J = 7.2 Hz, 1H), 7.53 (t, J = 7.5 Hz, 1H), 7.42 (t, J = 7.5 Hz, 1H), 7.31-7.28 (m, 3H), 6.89 (d, J = 7.8 Hz, 2H), 5.52 (s, 1H), 3.71 (s, 3H), 3.69-3.65 (m, 1H), 3.45 (s, 3H), 3.24-3.18 (m, 1H), 3.05-2.98 (m, 1H), 2.87-2.83 (m, 1H); ^{13}C NMR (125 MHz, DMSO-d_6): δ 160.6, 159.2, 156.1, 151.4 (2C), 137.0, 134.3, 132.4, 128.5, 128.2, 127.8, 127.7, 127.0, 113.9, 88.7, 59.1, 55.1, 45.8, 28.1, 26.8; HRMS (ESI) exact mass calculated for $C_{22}H_{20}N_4O_3$ $[M+H]^+$: 389.1608; found: 389.1615.

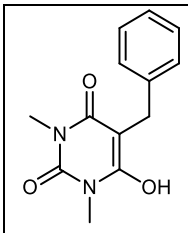
 **4y**	**8-phenyl-10,12-dihydro-5H-pyrimido[4',5':4,5]pyrimido[2,1-a] isoquinoline-9,11(6H,8H)-dione:** Off white solid; Yield 90 %, 309 mg; M.P.: >300 ^{o}C; R_f = 0.2 (hexane/EtOAc, 2:3); IR (KBr): 3415, 3176, 1709, 1627, 1516, 1456, 1392, 1237, 1162, 781, 696 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 10.91 (s, 1H), 10.6 (s, 1H), 8.17 (d, J = 7.9 Hz, 1H), 7.53-7.50 (m, 1H), 7.42-7.39 (m, 1H), 7.37-7.32 (m, 4H), 7.29-7.28 (m, 2H), 5.51(s, 1H), 3.66-3.62 (m, 1H), 3.04-2.99 (m, 2H), 2.85-2.81 (m, 1H); ^{13}C NMR (125 MHz, DMSO-d_6): δ 162.2, 156.8, 151.7, 142.2, 140.0, 137.1, 134.3, 132.4, 129.3, 128.7, 128.6, 127.7, 127.0, 126.6, 88.0, 59.6, 46.2, 32.6; HRMS (ESI) exact mass calculated for $C_{20}H_{16}N_4O_2$ $[M+H]^+$: 345.1346; found: 345.1354.

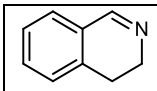
 **4z**	**8-(4-bromophenyl)-10,12-dihydro-5H-pyrimido[4',5':4,5]pyrimido [2,1-a]isoquinoline-9,11(6H,8H)-dione:** Off white solid; Yield 93 %, 392 mg; M.P.: >300 ^{o}C; R_f = 0.2 (hexane/EtOAc, 2:3); IR (KBr): 3409, 3166, 1702, 1633, 1511, 1463, 1233, 1169, 780, 693 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 10.45 (s, 1H), 10.12 (s, 1H), 8.16-8.15 (m, 1H), 7.53-7.51 (m, 1H), 7.51-7.49 (m, 1H), 7.40-7.39 (m, 1H), 7.37-7.35 (m, 1H), 7.33-7.31 (m, 1H), 5.53 (s, 1H), 3.64-3.59 (m, 1H), 3.17-3.11 (m, 1H), 3.02-2.96 (m, 1H), 2.85-2.80 (m, 1H); ^{13}C NMR (125 MHz, DMSO-d_6): δ 162.2, 156.8, 151.8, 151.2, 141.5, 137.1, 132.4, 131.6, 130.5, 129.3, 128.3, 127.7, 126.9, 121.4, 87.6, 59.1, 46.2, 26.9; HRMS (ESI) exact mass calculated for $C_{20}H_{15}BrN_4O_2$ $[M+H]^+$: 423.0451; found: 423.0460.

 **4z1**	**1,3-dimethyl-5-phenyl-5,7,8,13-tetrahydropyrimido[4'',5'':4',5'] pyrimido[1',2':1,2]pyrido[3,4-b]indole-2,4(1H,3H)-dione :** Yellow solid; Yield 63 %, 259 mg; M.P.: 107-109 ^{o}C; R_f = 0.4 (hexane/EtOAc, 1:1); IR (KBr): 3431, 3054, 2931, 1604, 1386, 1254, 1072, 750 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.95 (bs, 1H), 7.56 (d, J = 8.0 Hz, 1H), 7.50-7.45 (m, 3H), 7.37-7.31 (m, 3H), 7.30-7.28 (m, 1H), 7.19-7.15 (m, 1H), 5.59 (s, 1H), 3.74-3.67 (m, 1H), 3.62 (s, 3H), 3.53-3.46 (m, 1H), 3.28 (s, 3H), 3.05-3.01 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.9, 152.3, 150.3, 142.2, 138.0, 128.8, 128.4, 127.1, 126.7, 125.9, 125.5, 120.8, 120.2, 117.9, 112.1, 89.8, 60.2, 47.7, 29.7, 29.3, 27.8; HRMS (ESI) exact mass calculated for $C_{24}H_{21}N_5O_2$ $[M+H]^+$: 412.1768; found: 412.1772.

 **4z2**	**5-(4-chlorophenyl)-1,3-dimethyl-5,7,8,13-tetrahydropyrimido [4'',5'':4',5']pyrimido[1',2':1,2]pyrido[3,4-b]indole-2,4(1H,3H)-dione :** Yellow solid; Yield 67 %, 299 mg; M.P.: 126-127 ^{o}C; R_f = 0.4 (hexane/EtOAc, 3:2); IR (KBr): 3390, 3047, 2924, 1681, 1629, 1578, 1384, 733 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.86 (bs, 1H), 7.57 (d, J = 8.1 Hz, 1H), 7.47 (d, J = 8.3 Hz, 1H), 7.44-7.41 (m, 2H), 7.38-7.34 (m, 1H), 7.30-7.28 (m, 2H), 7.20-7.16 (m, 1H), 5.57 (s, 1H), 3.75-3.68 (m, 1H), 3.62 (s, 3H), 3.51-3.46 (m, 1H), 3.28 (s, 3H), 3.06-3.03 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.9, 152.3, 150.3, 140.8, 138.1, 134.3, 130.4, 129.0, 128.5, 126.5, 126.1, 125.5, 120.9, 120.3, 118.0, 112.1, 89.4, 59.6, 47.7, 29.7, 29.4, 27.8; HRMS (ESI) exact mass calculated for $C_{24}H_{20}ClN_5O_2$ $[M+H]^+$: 446.1378; found: 446.1386.

 **4'a**	**6-amino-5-((3,4-dihydroisoquinolin-2(1H)-yl)(phenyl)methyl)-1,3-dimethylpyrimidine-2,4(1H,3H)-dione:** White solid; Yield 45 %, 169 mg; M.P.: 140-142 ^{o}C; R_f = 0.45 (hexane/EtOAc, 1:1); IR (KBr): 3349, 3150, 2952, 2806, 1694, 1600, 1497, 1382, 1243, 1037, 935, 752, 699 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.57-7.55 (m, 2H), 7.32-7.29 (m, 2H), 7.27-7.26 (m, 1H), 7.18-7.10 (m, 3H), 6.96 (d, *J* = 7.2 Hz, 1H), 6.81 (bs, 2H), 4.92 (s, 1H), 3.66-3.64 (m, 2H), 3.36 (s, 3H), 3.28 (s, 3H), 2.92-2.91 (m, 2H), 2.88-2.74 (m, 2H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 161.9, 151.2, 150.8, 141.5, 134.5, 134.0, 128.5, 127.3, 126.8, 126.4, 125.8, 88.2, 66.8, 55.0, 49.1, 29.3, 28.5, 28.0; HRMS (ESI) exact mass calculated for $C_{22}H_{24}N_4O_2$ $[M+H]^+$: 377.1972; found: 377.1974.

 **7**	**5-benzyl-6-hydroxy-1,3-dimethylpyrimidine-2,4(1H,3H)-dione:**[1] White solid; Yield 46 %, 113 mg; ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.24-7.23 (m, 3H), 7.04-7.02 (m, 1H), 3.78 (bs, 1H), 3.47 (s, 2H), 3.13 (s, 6H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 168.3, 150.9, 135.0, 128.8, 128.6, 127.8, 50.7, 37.8, 28.1; HRMS (ESI) exact mass calculated for $C_{13}H_{14}N_2O_3$ $[M+H]^+$: 247.1077; found: 247.1080.

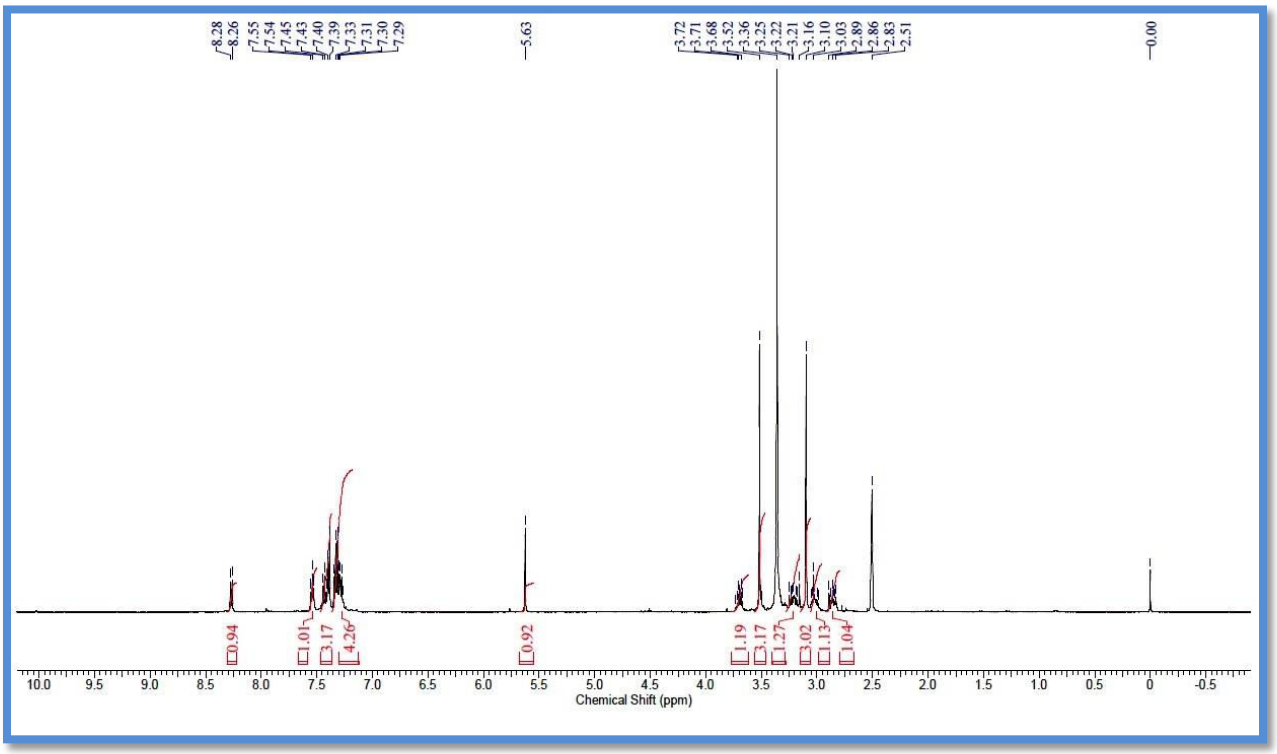
 **8**	**3,4-dihydroisoquinoline:**[2] Light yellow solid; Yield 35 %, 45 mg; ^{1}H NMR (300 MHz, $CDCl_3$): δ 8.36 (s, 1H), 7.37-7.27 (m, 3H), 7.17 (d, *J* = 6.9 Hz, 1H), 3.78 (t, *J* = 7.5 Hz, 2H), 2.76 (t, *J* = 7.2 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 160.3, 136.3, 131.0, 128.5, 127.4, 127.2, 126.9, 47.3, 24.9.

References:

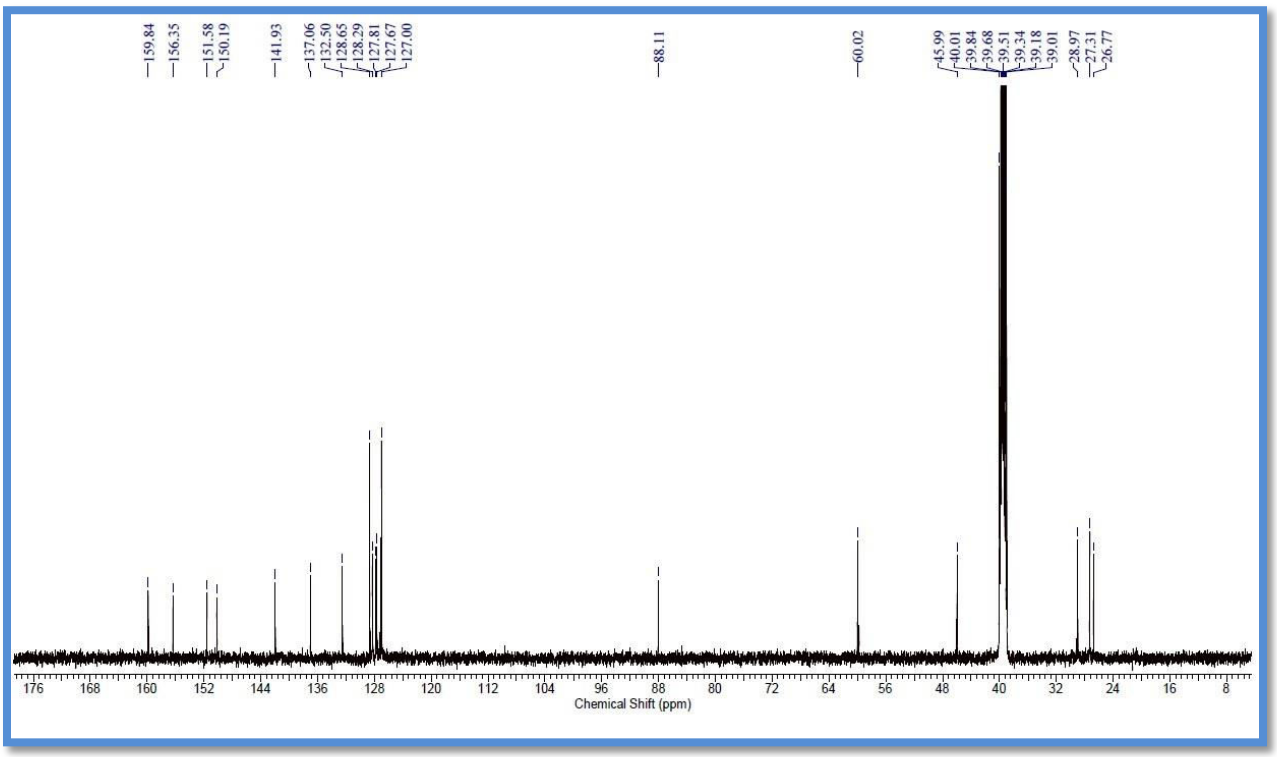
1. (a) S. E. Denmark, M. Y. S. Ibrahim, A. Ambrosi, *ACS Catal*., 2017, **7**, 613; (b) A. E. Putra, Y. Oe, T. Ohta, *Tetrahedron Lett*. 2017, **58**, 1098.

2. G. Lahm, J.-G. Deichmann, A. L. Rauen, T. Opatz, *J. Org. Chem.*, 2015, **80**, 2010.

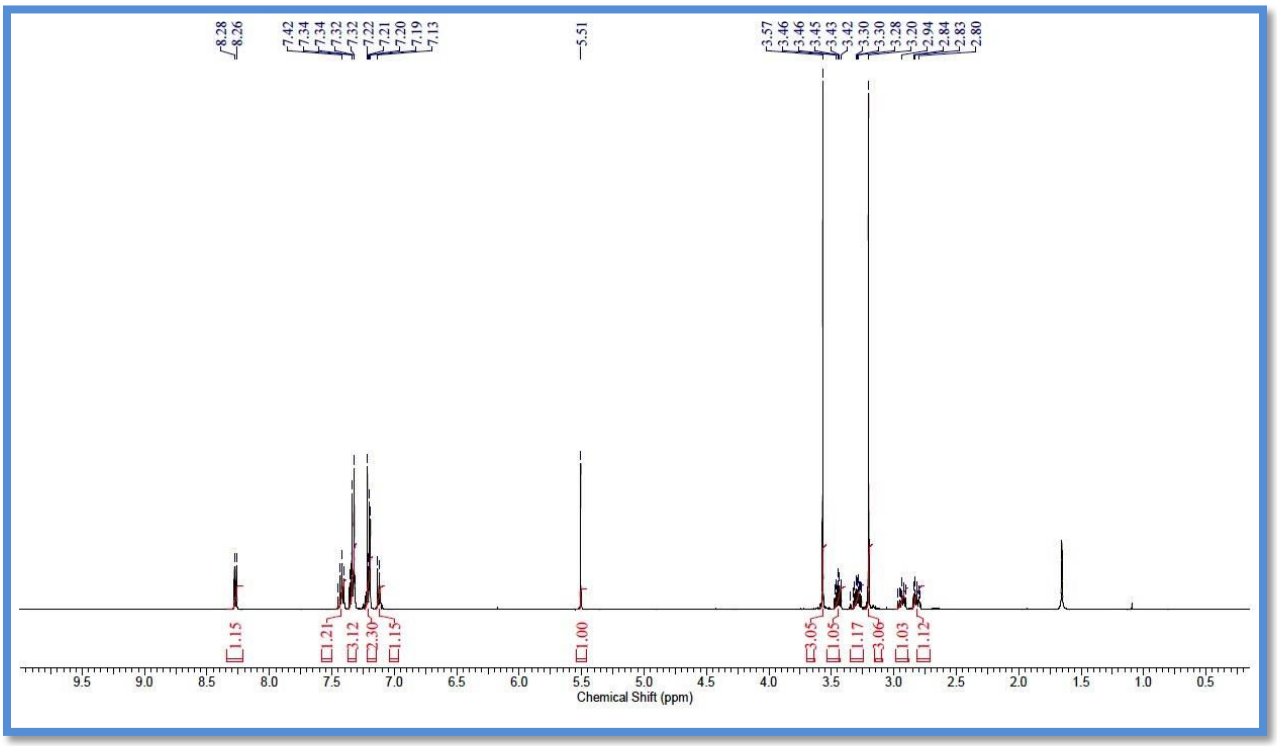
^{1}H-NMR of compound 4a



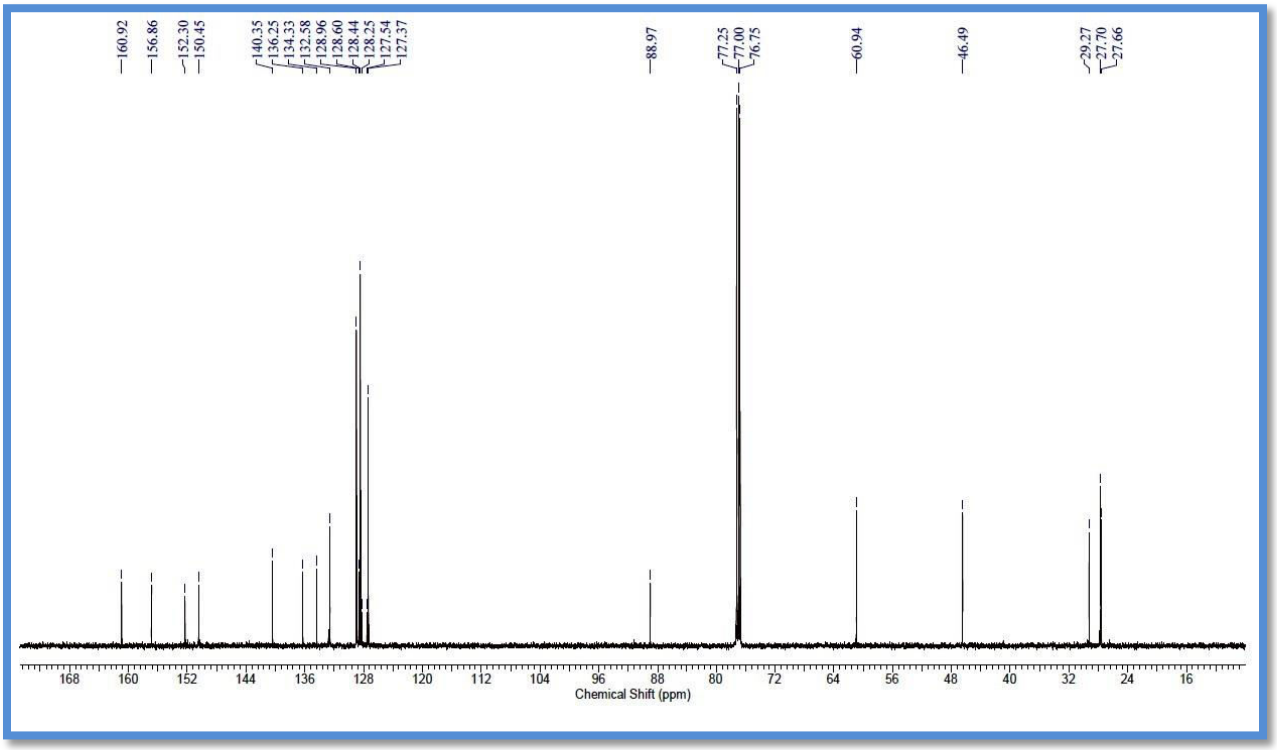
^{13}C-NMR of compound 4a



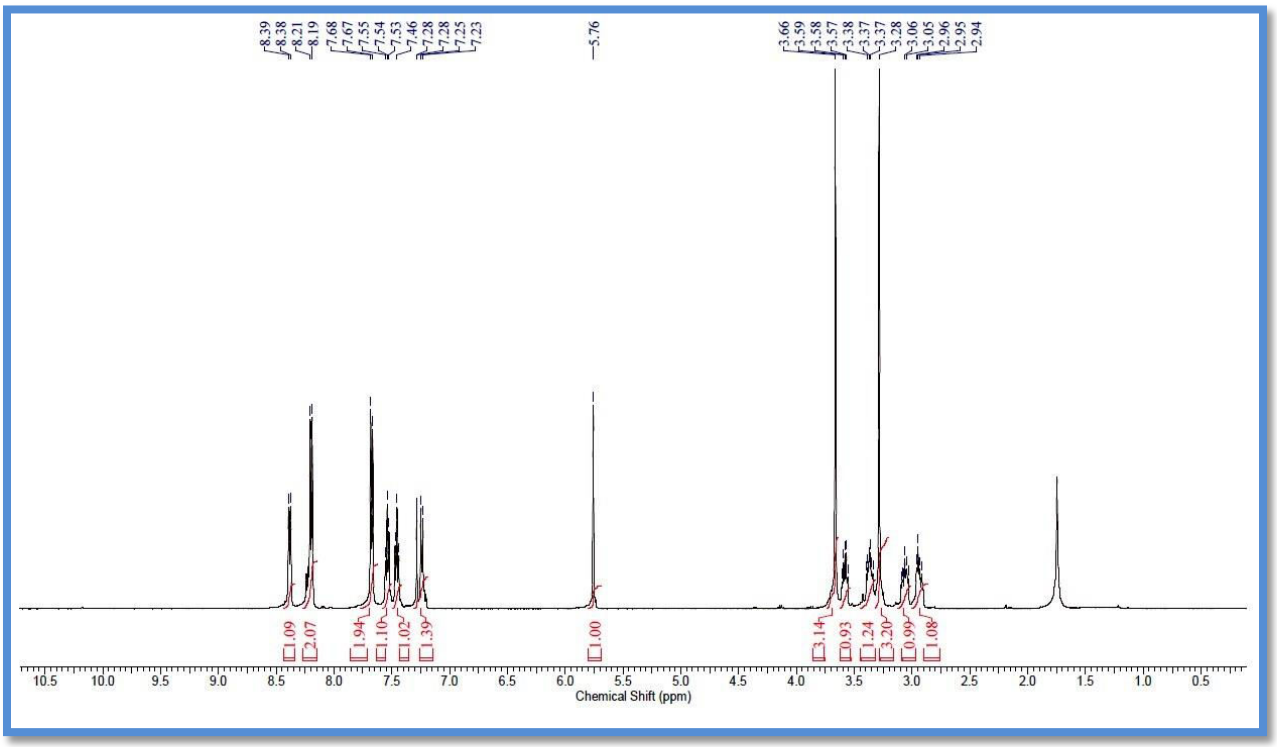
^{1}H-NMR of compound 4b



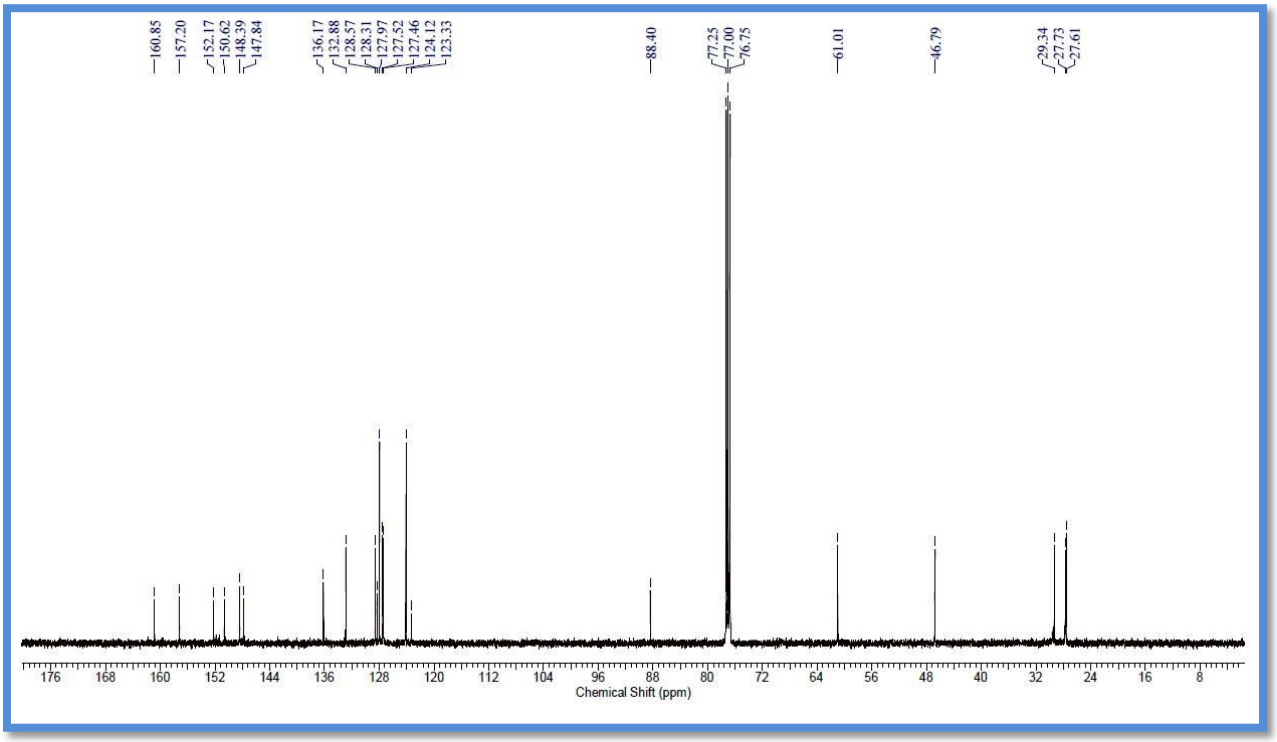
^{13}C-NMR of compound 4b



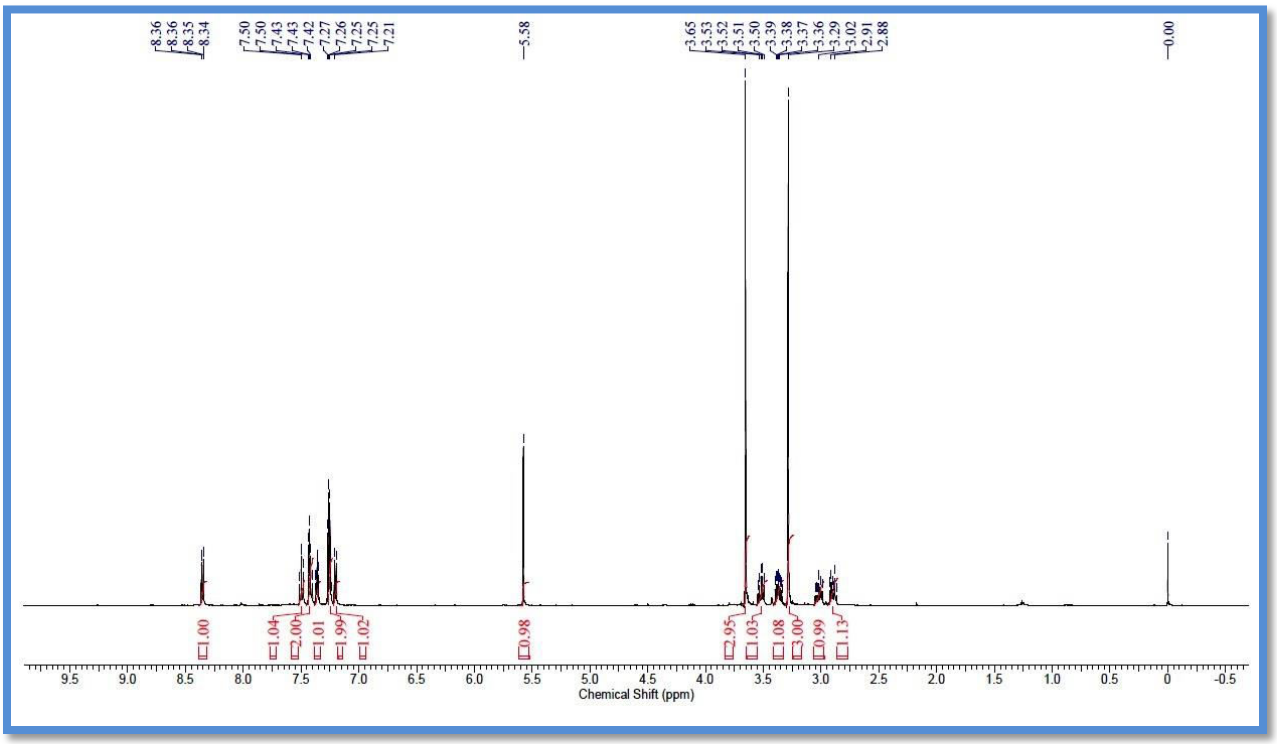
^{1}H-NMR of compound 4c



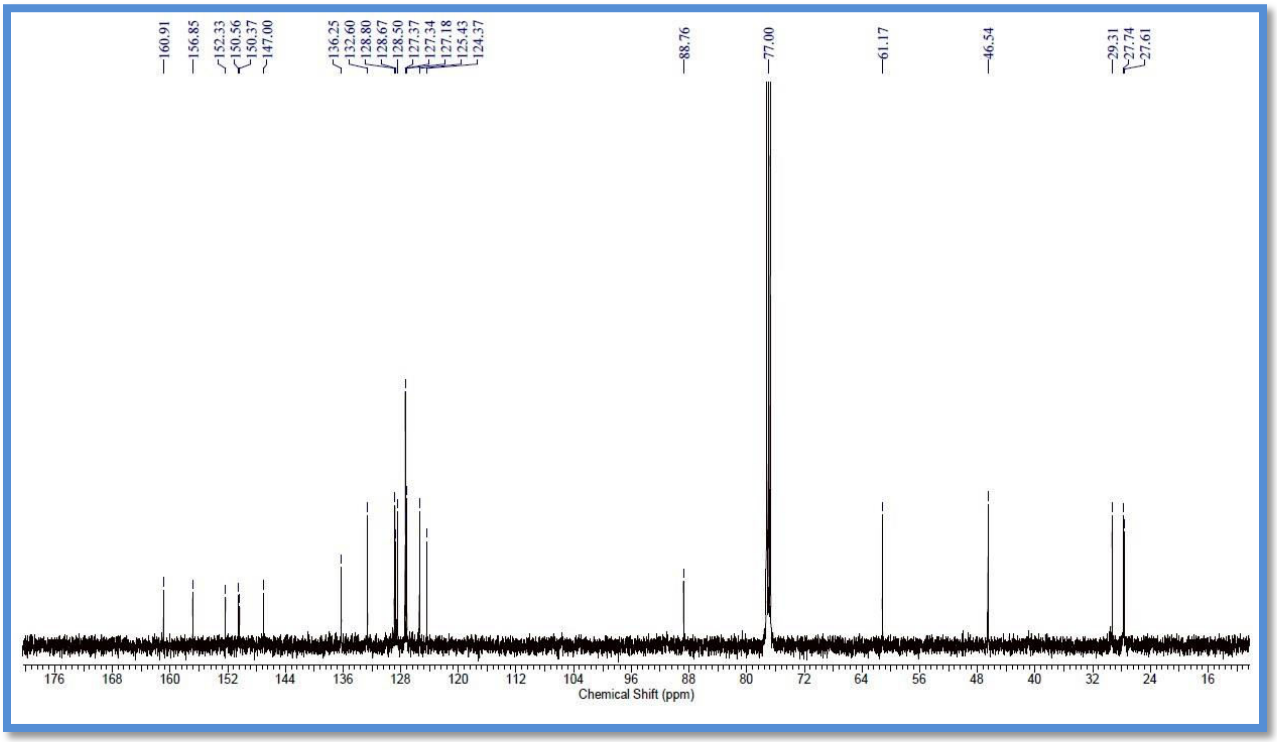
^{13}C-NMR of compound 4c



^{1}H-NMR of compound 4d



^{13}C-NMR of compound 4d



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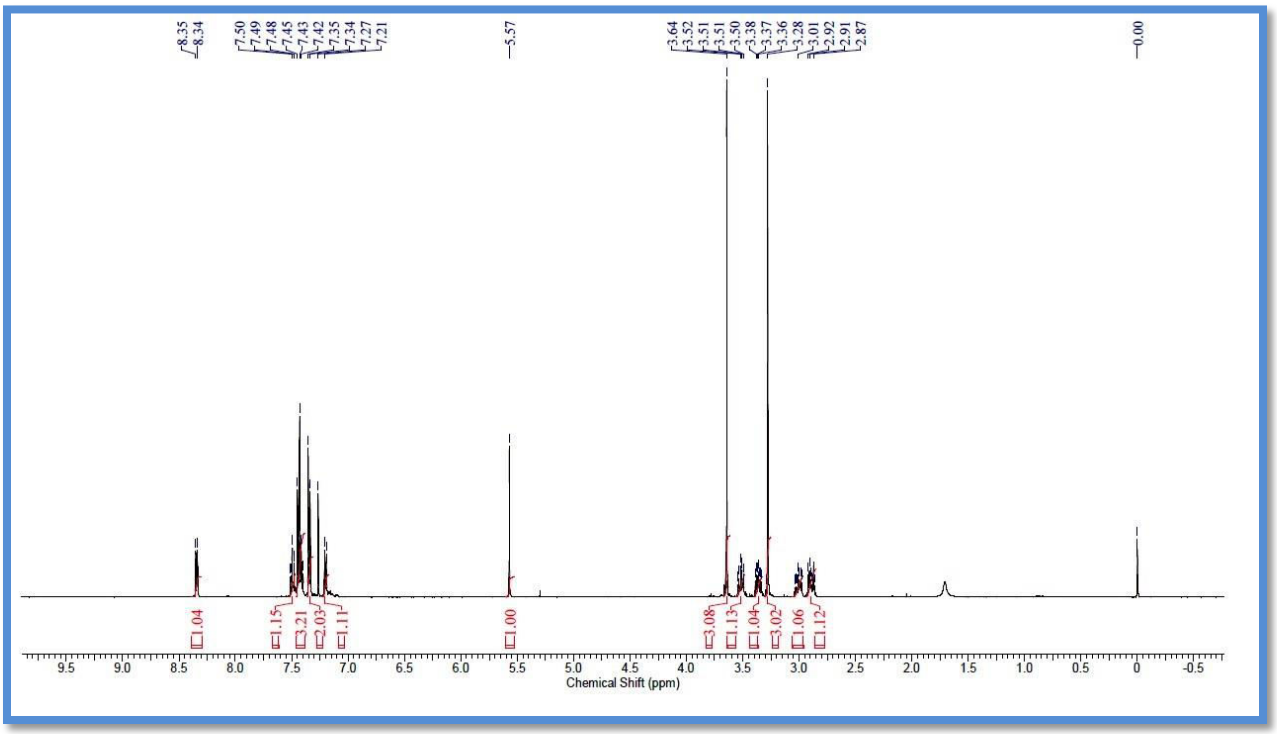

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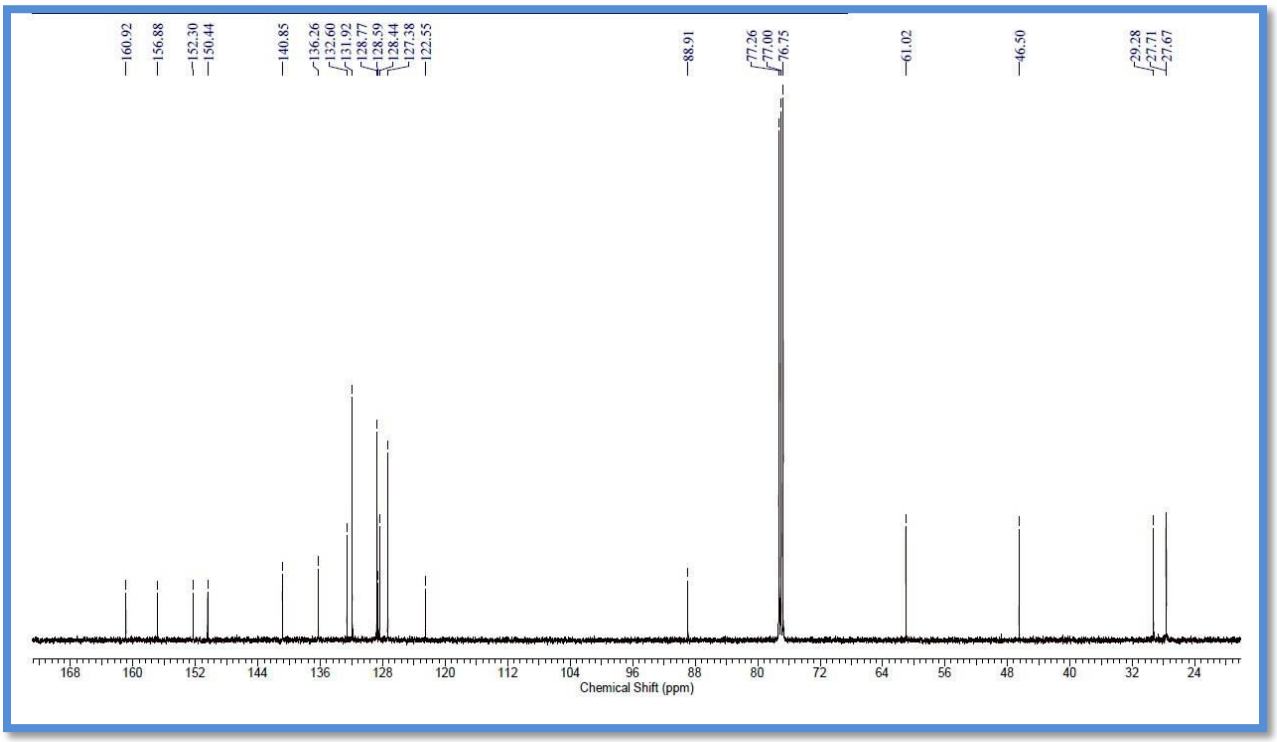
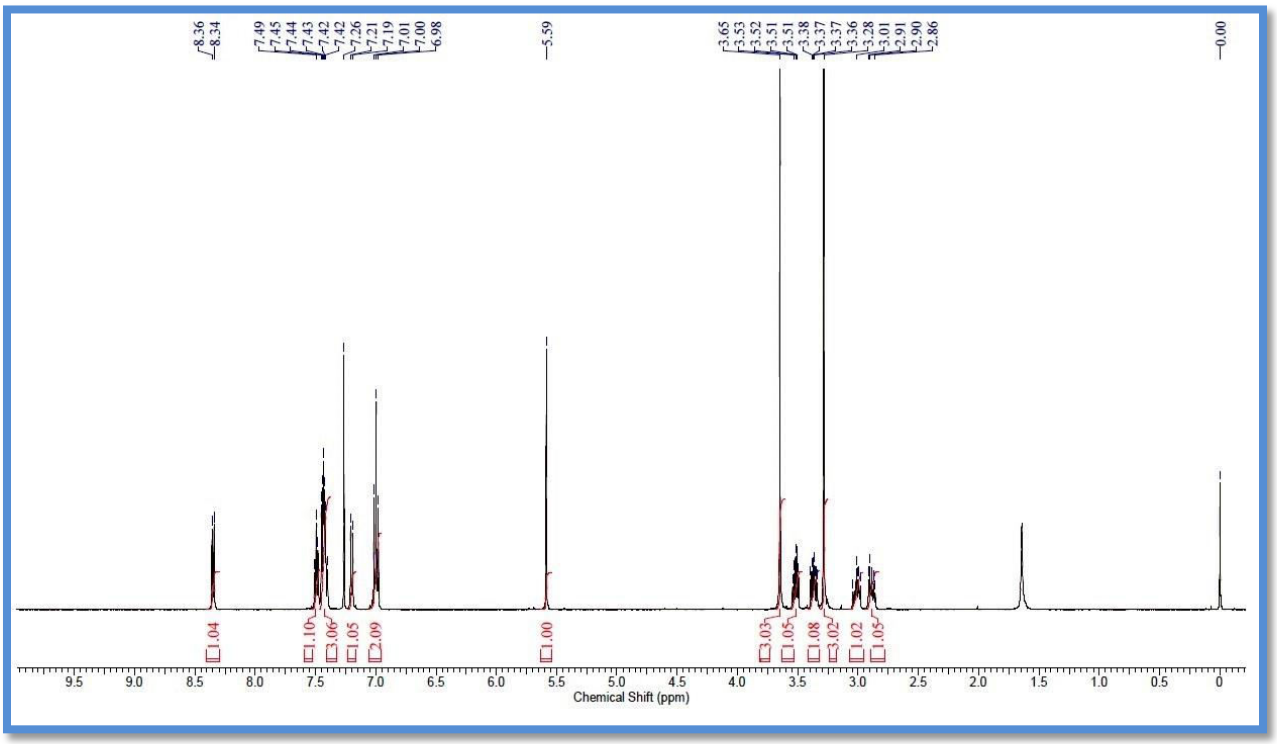
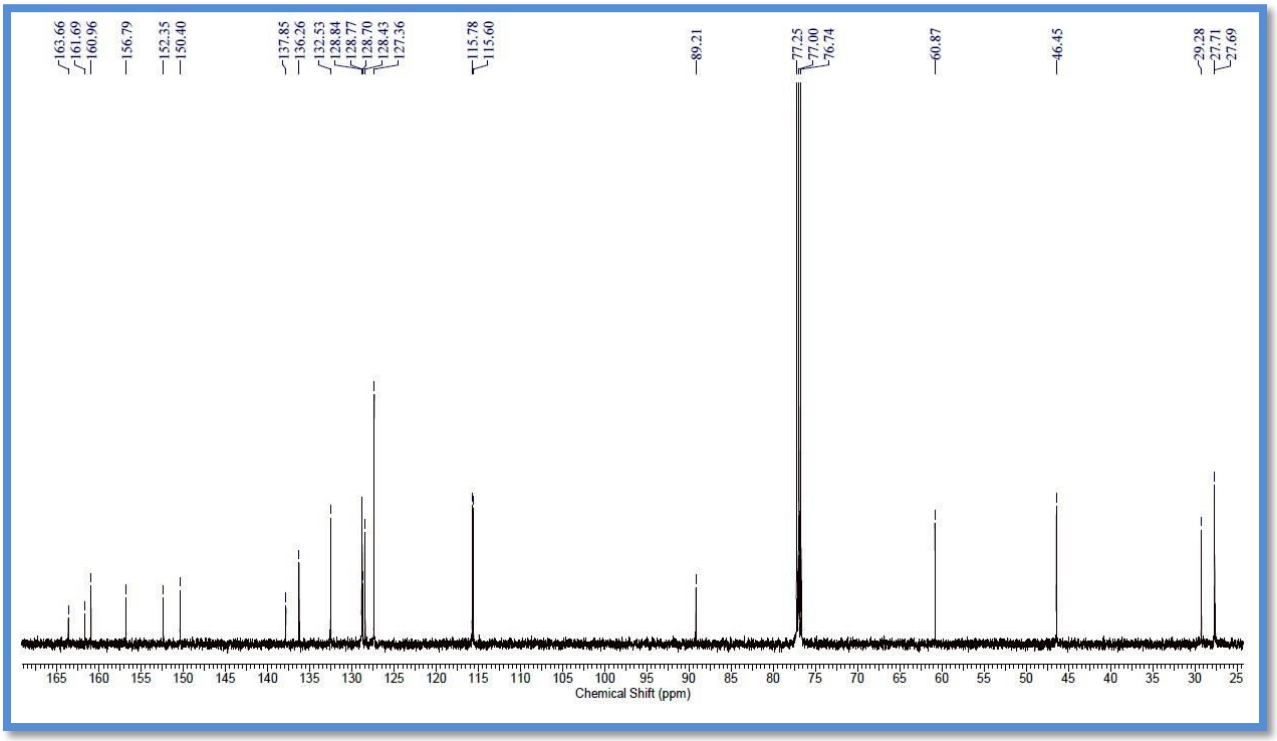


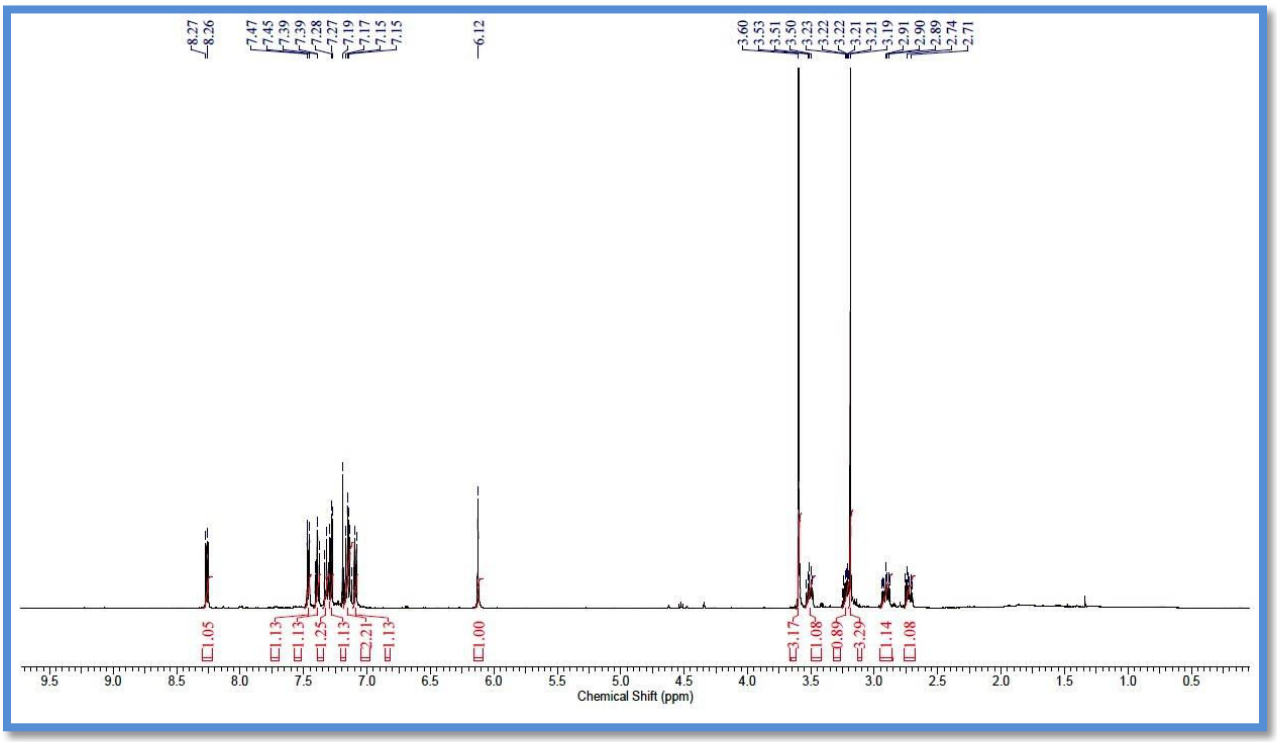
^{1}H-NMR of compound 4f



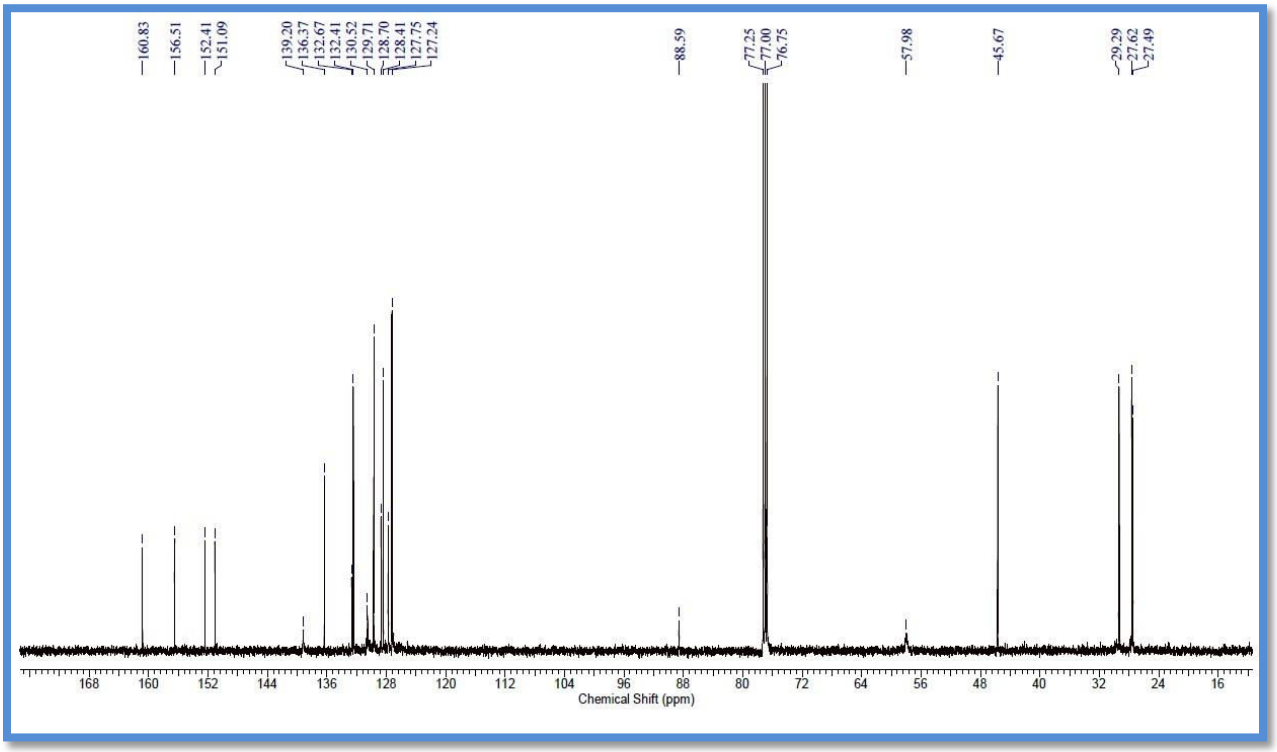
^{13}C-NMR of compound 4f



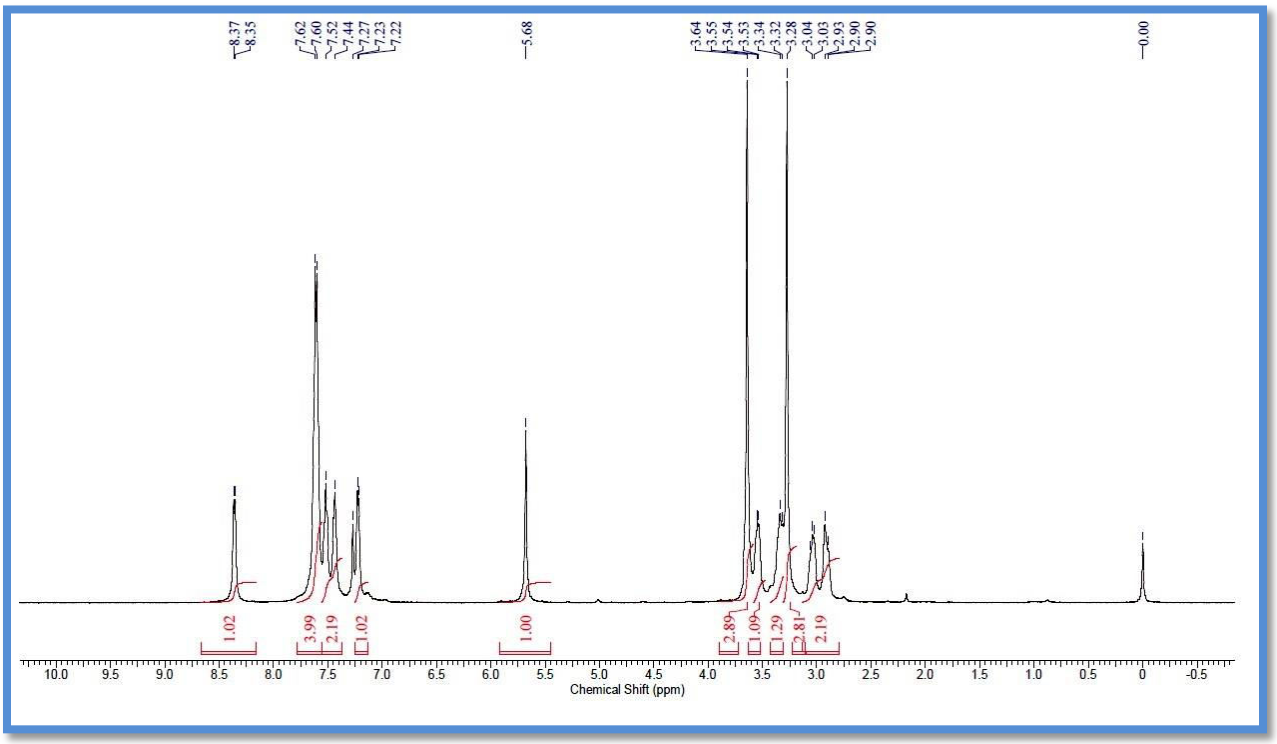
^{1}H-NMR of compound 4g



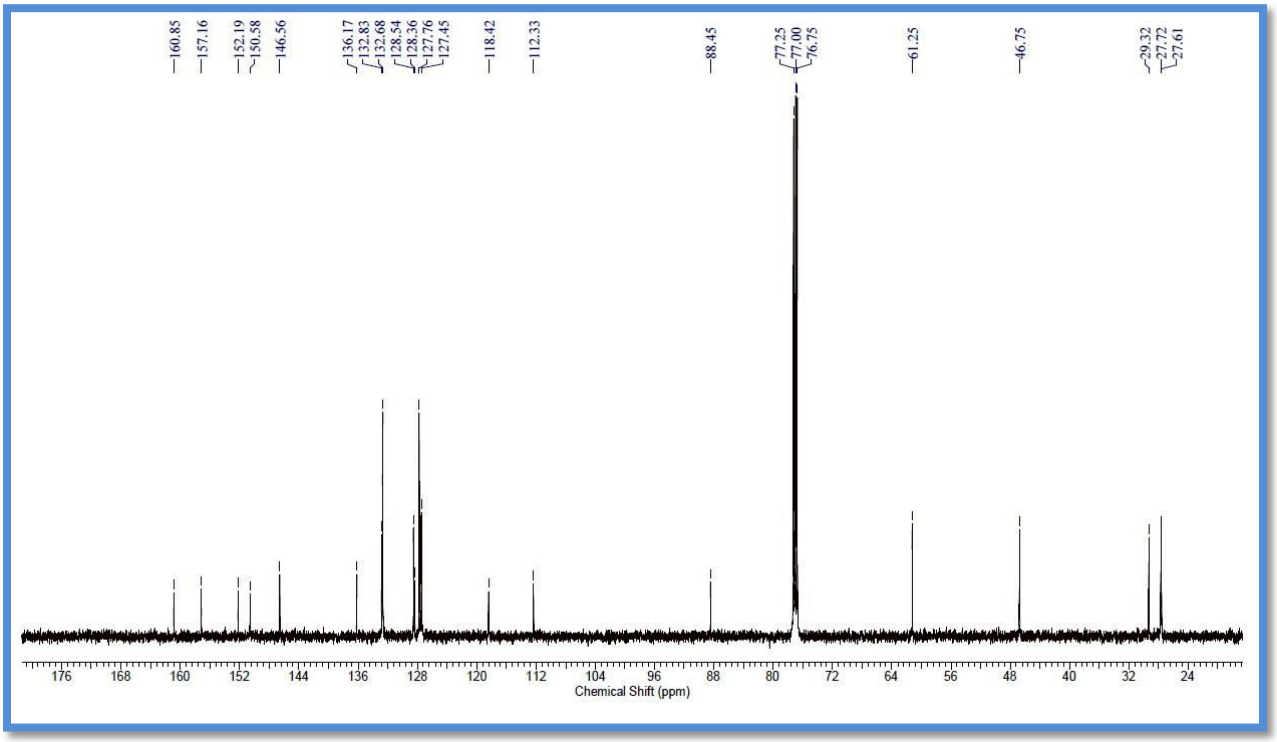
^{13}C-NMR of compound 4g



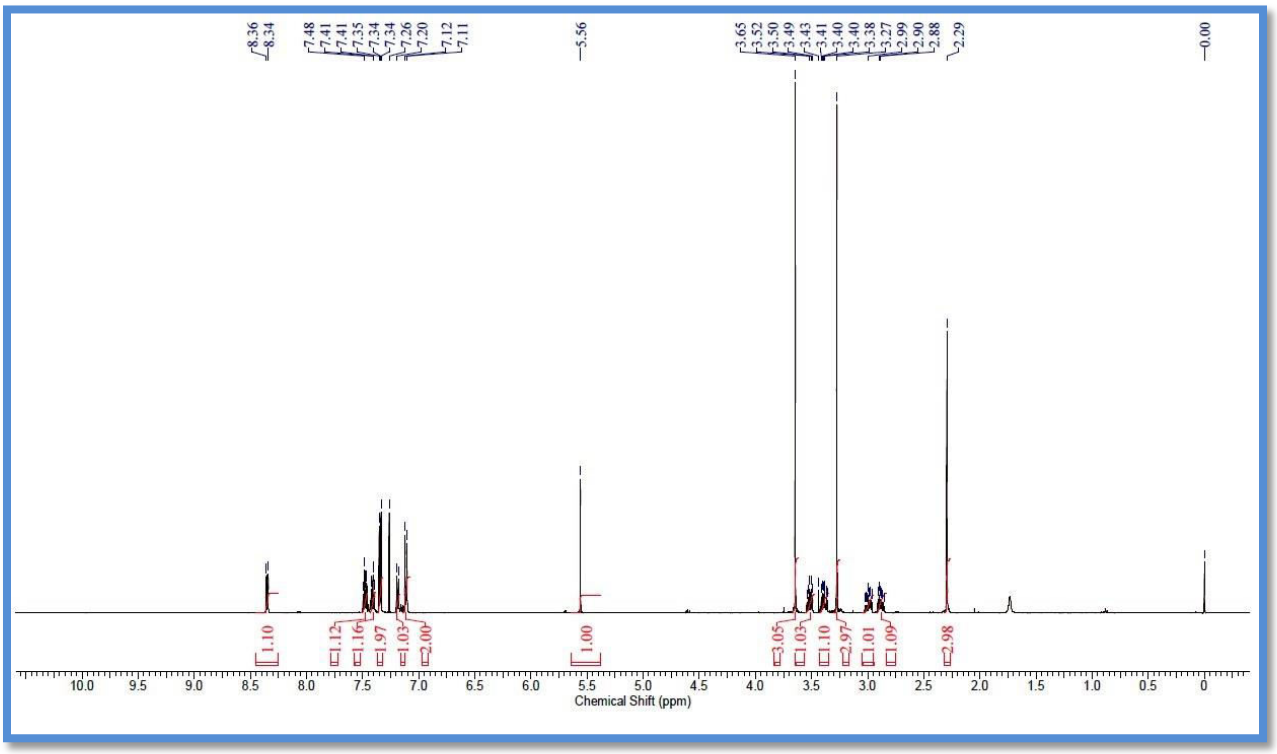
^{1}H-NMR of compound 4h



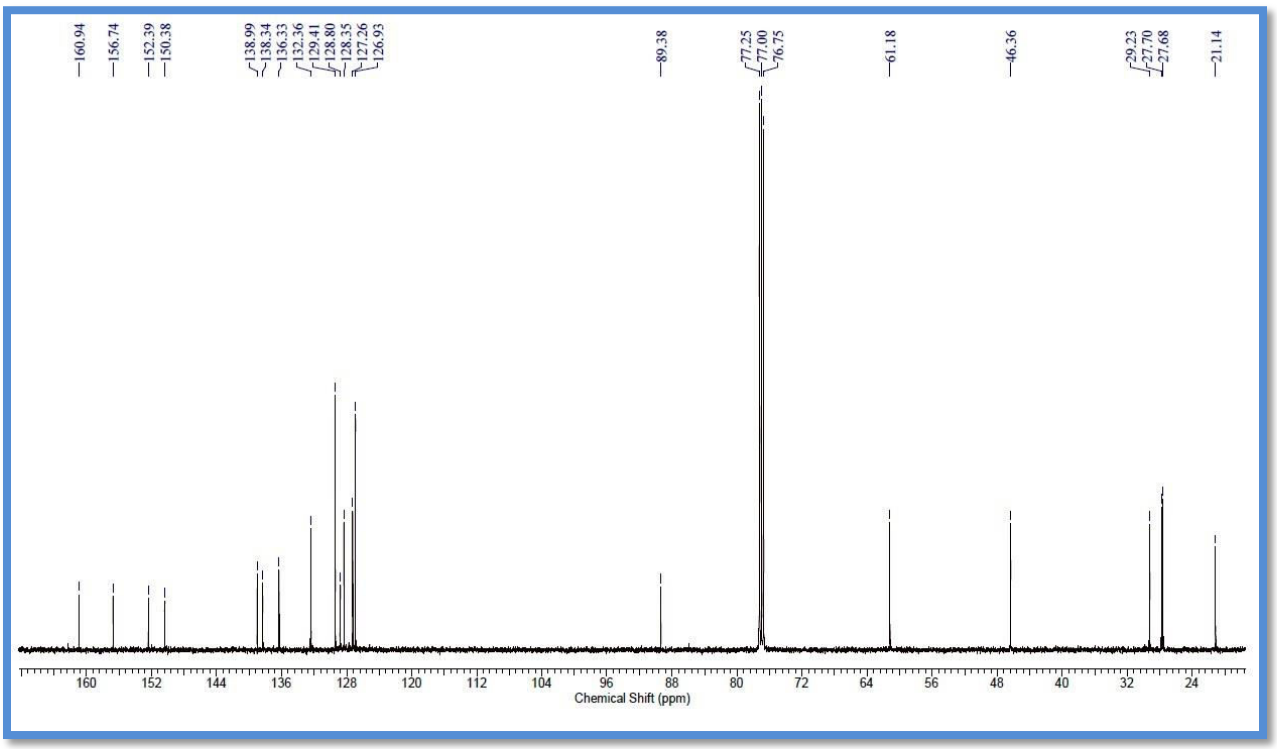
^{13}C-NMR of compound 4h



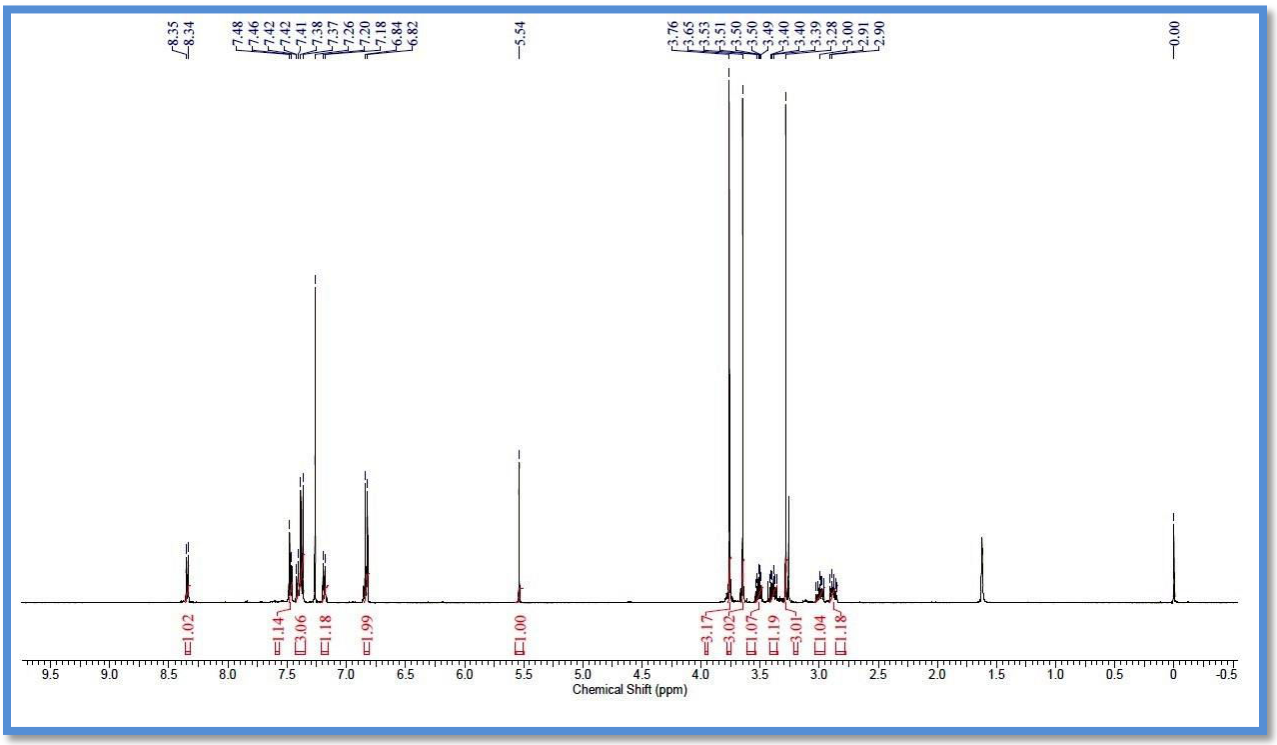
^{1}H-NMR of compound 4i



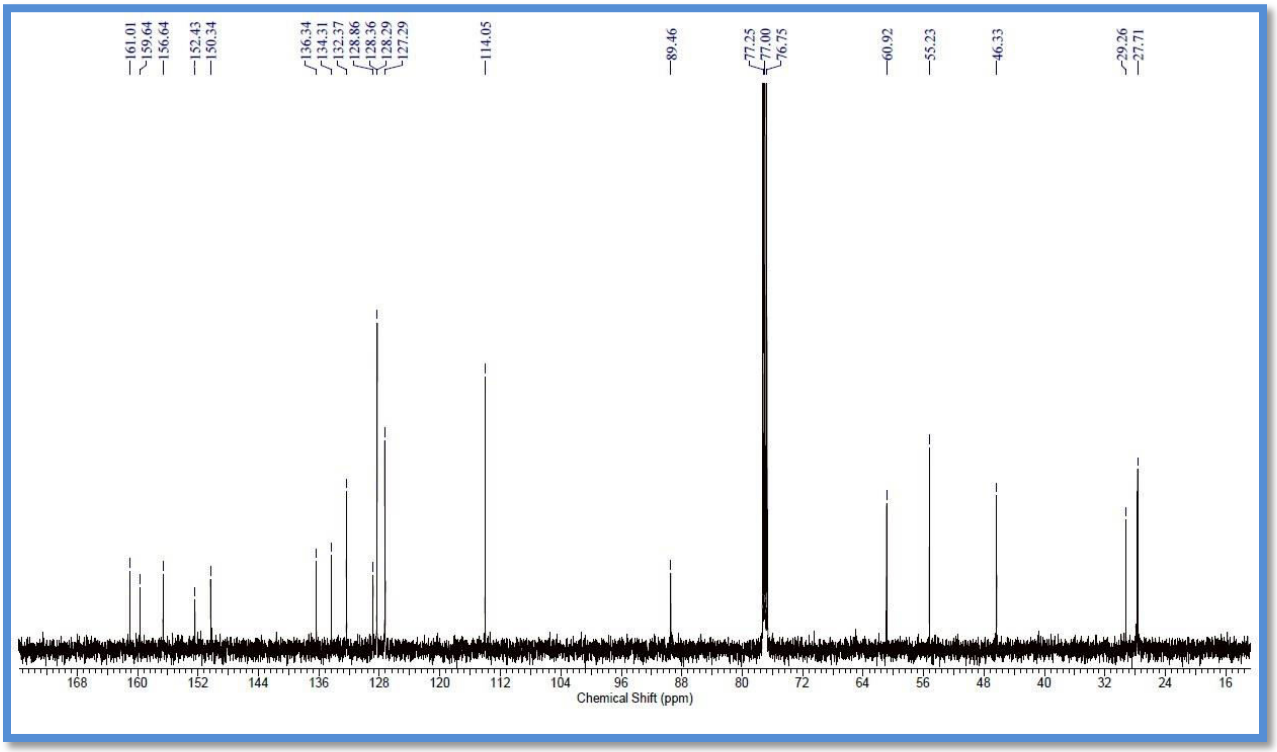
^{13}C-NMR of compound 4i



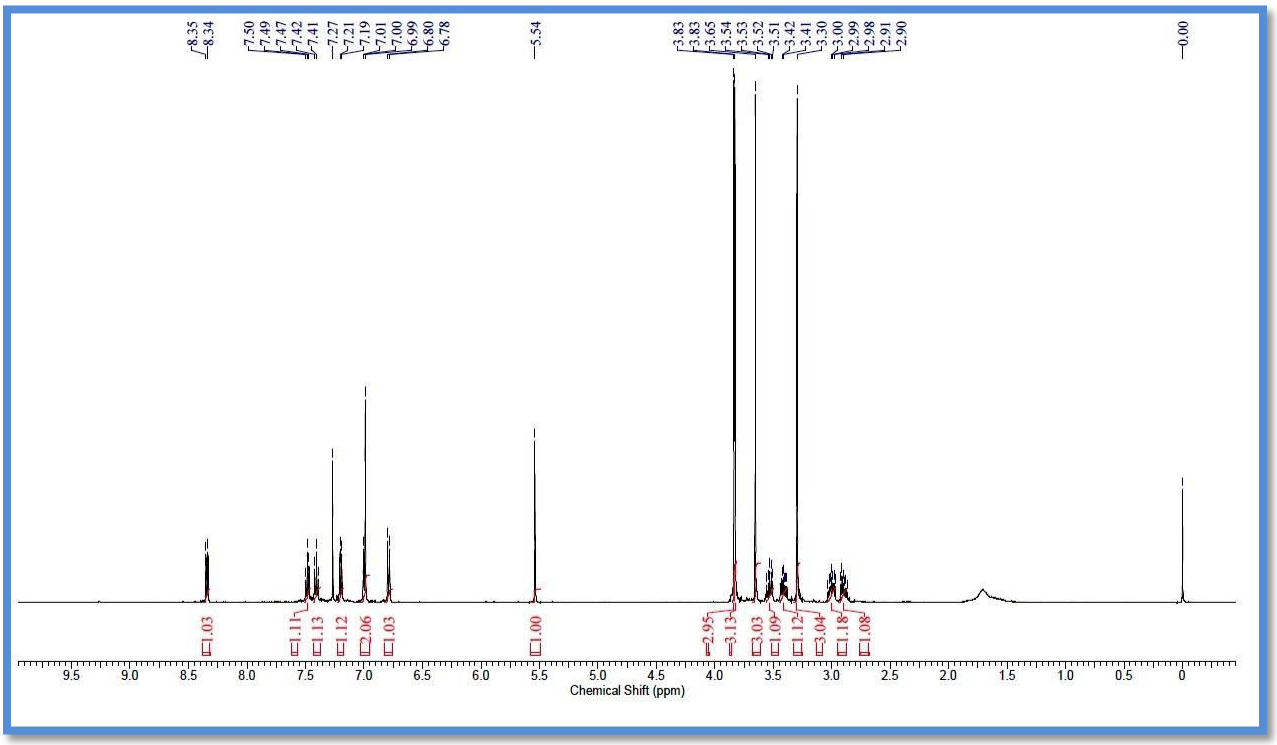
^{1}H-NMR of compound 4j



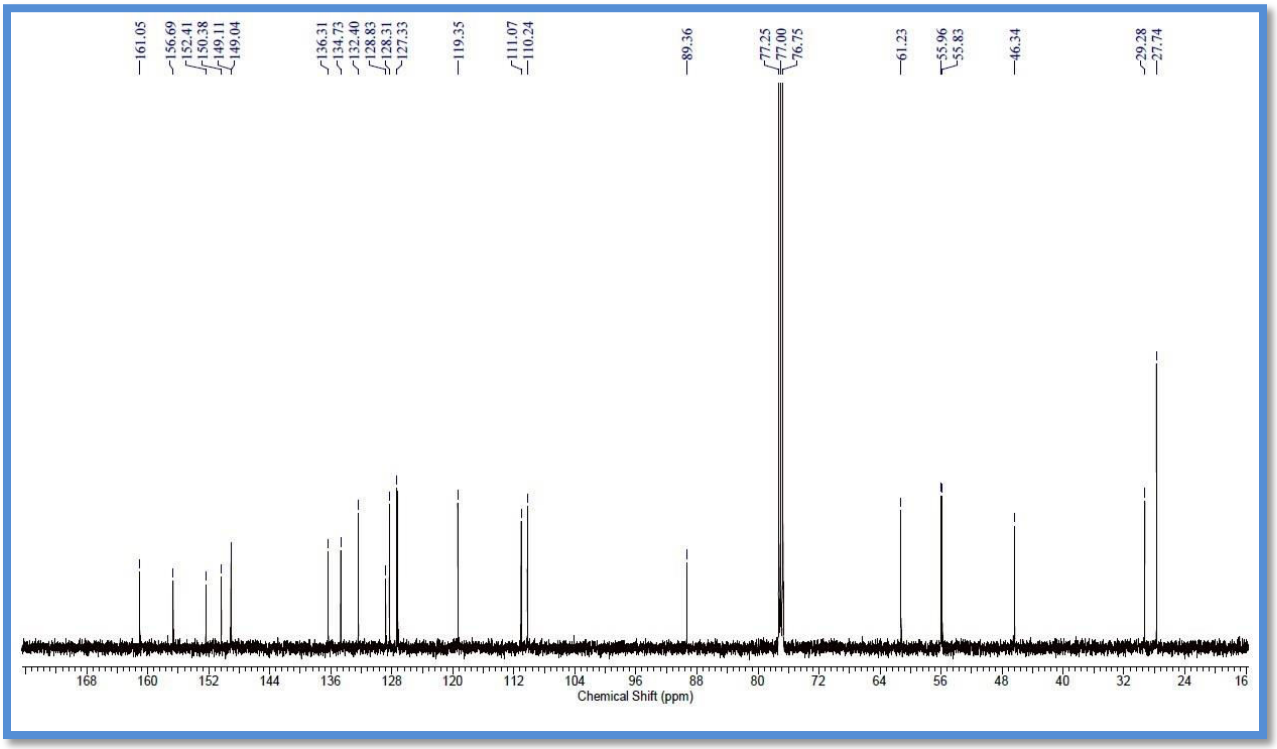
^{13}C-NMR of compound 4j



^{1}H-NMR of compound 4k



^{13}C-NMR of compound 4k



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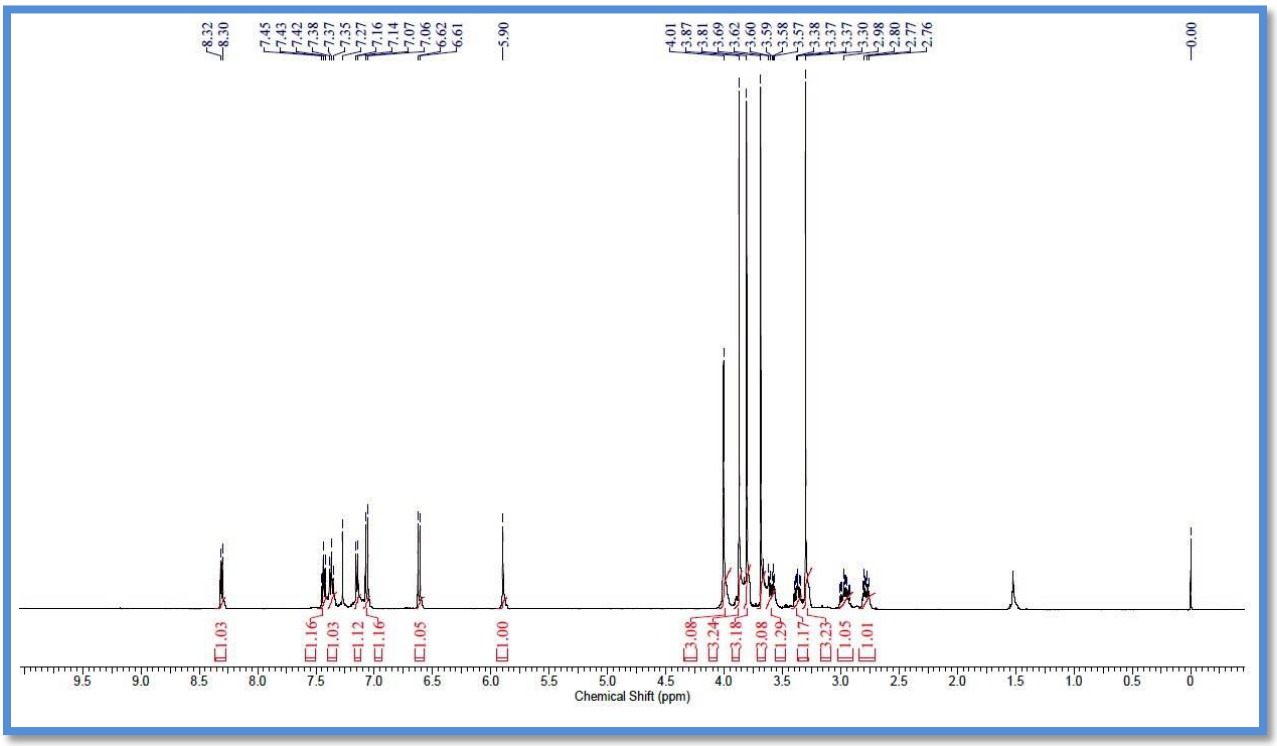
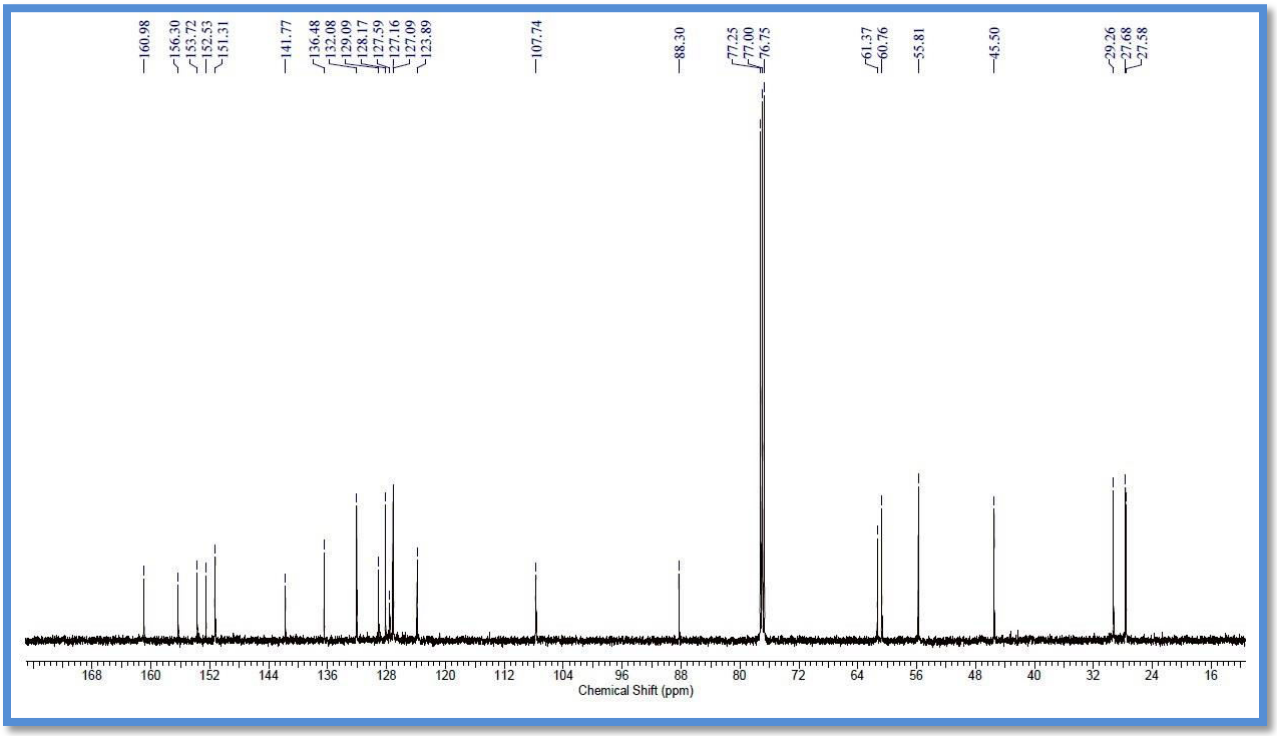
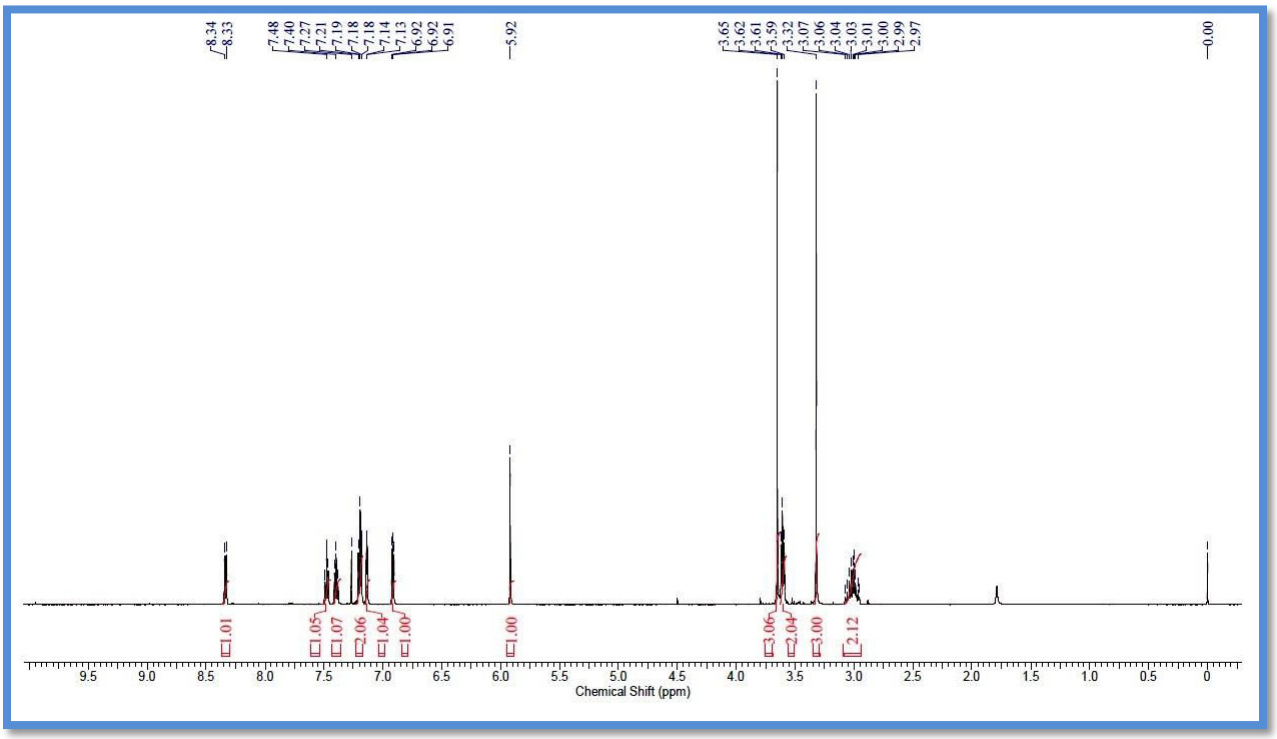


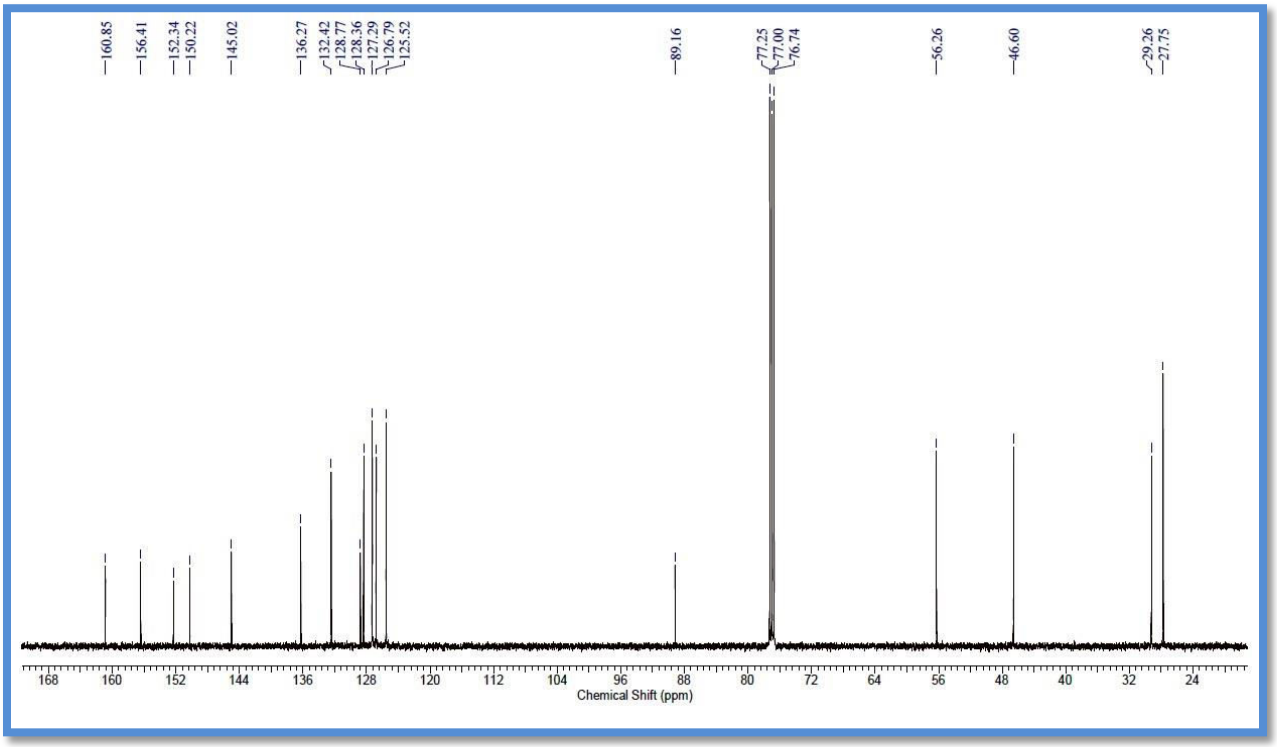
^{13}C-NMR of compound 4l



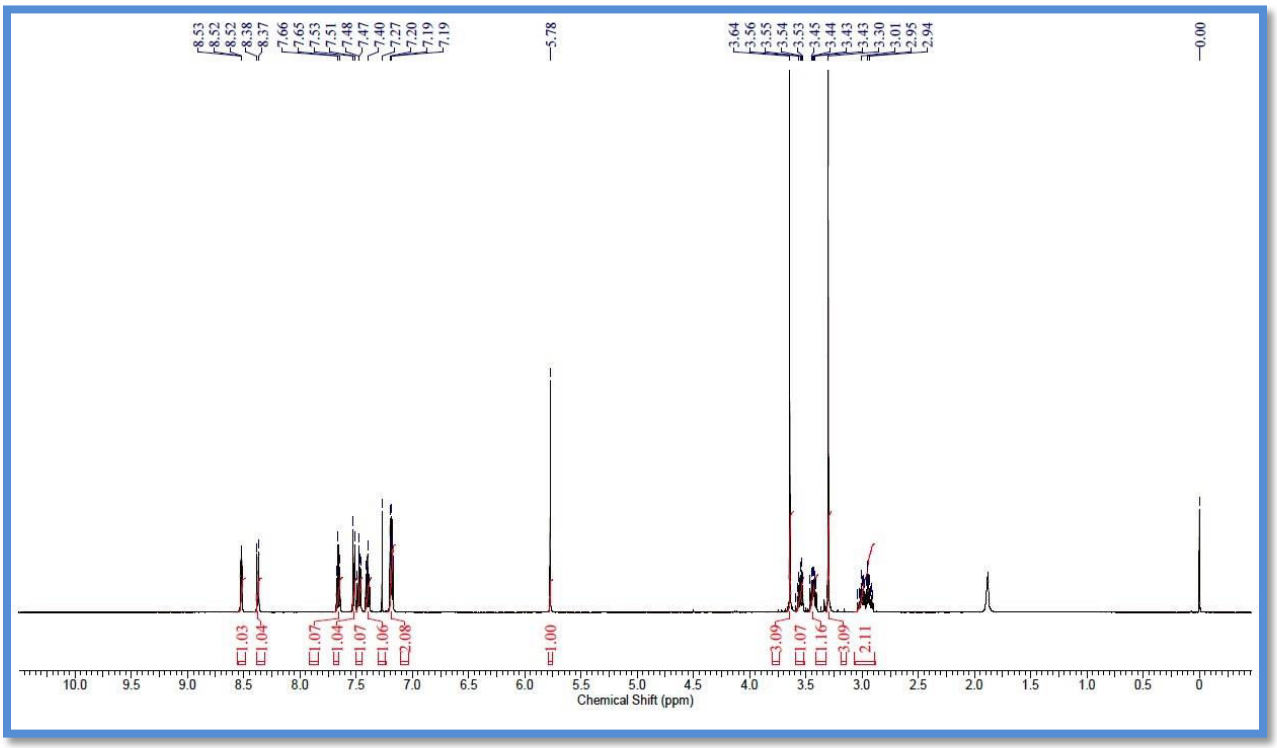
^{1}H-NMR of compound 4m



^{13}C-NMR of compound 4m



^{1}H-NMR of compound 4n



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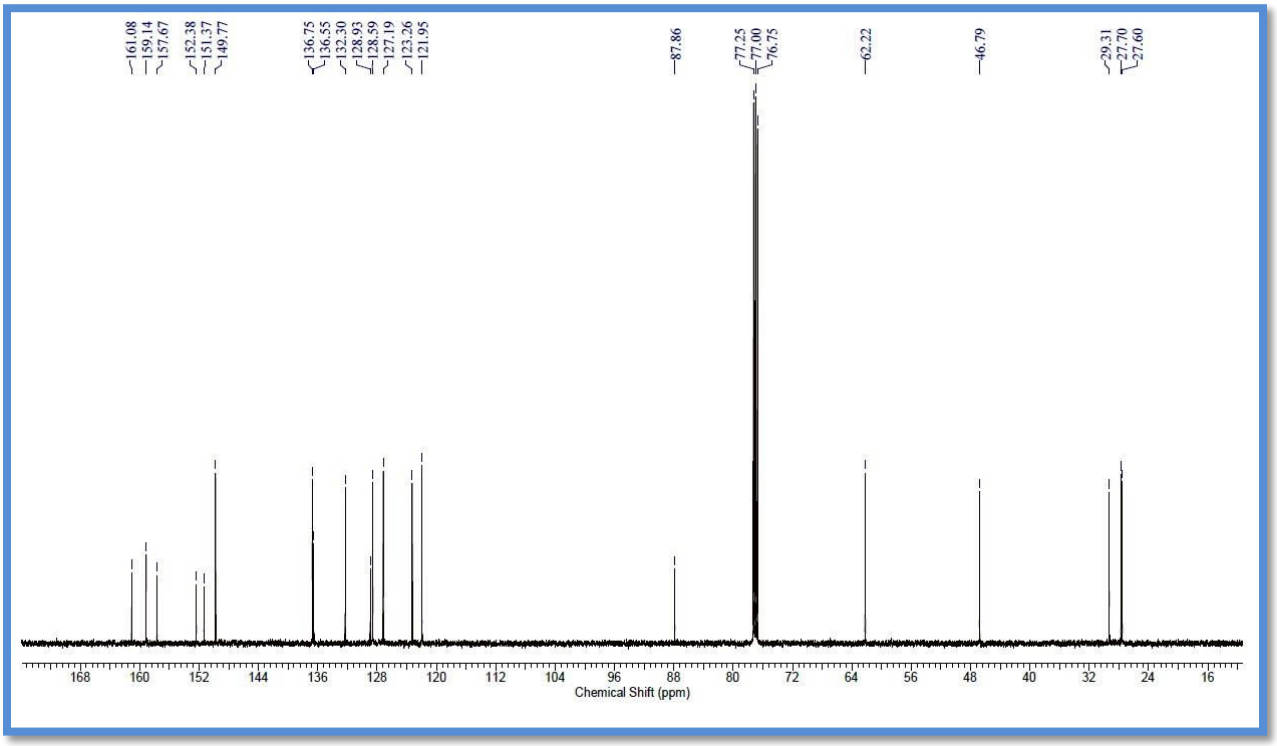
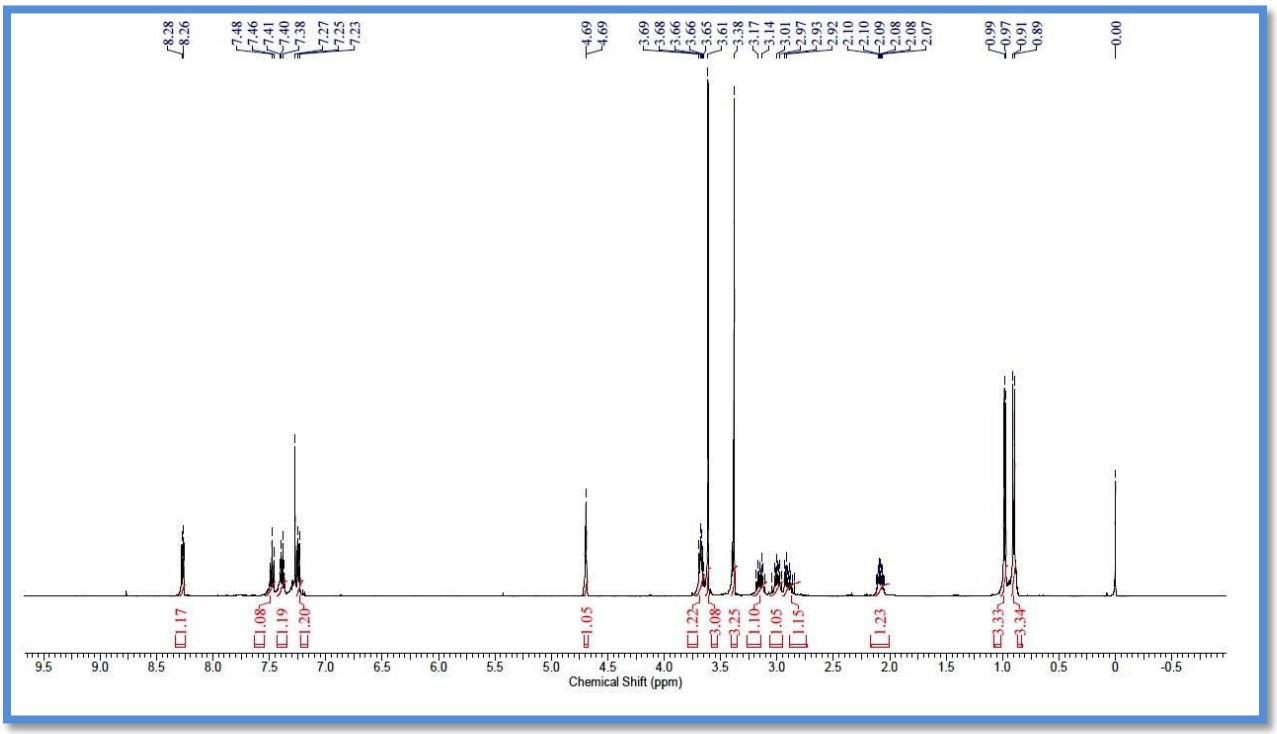
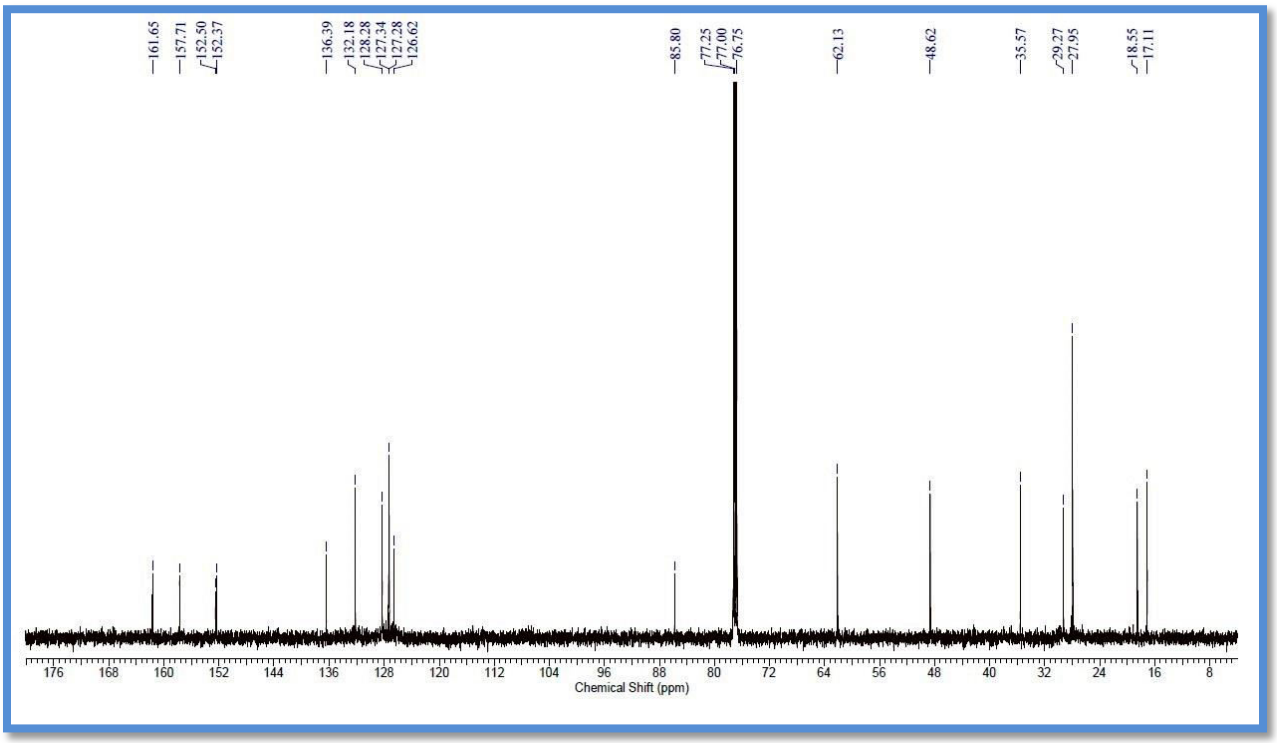


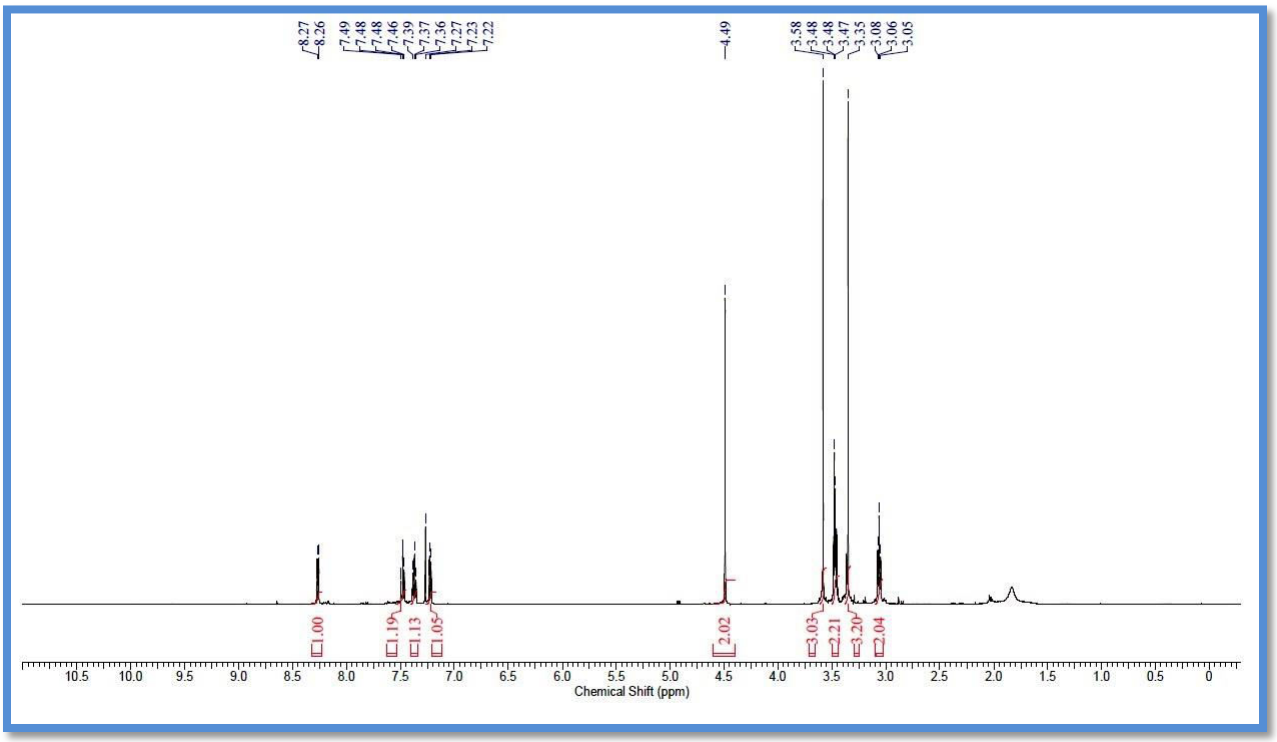
^{1}H-NMR of compound 4o



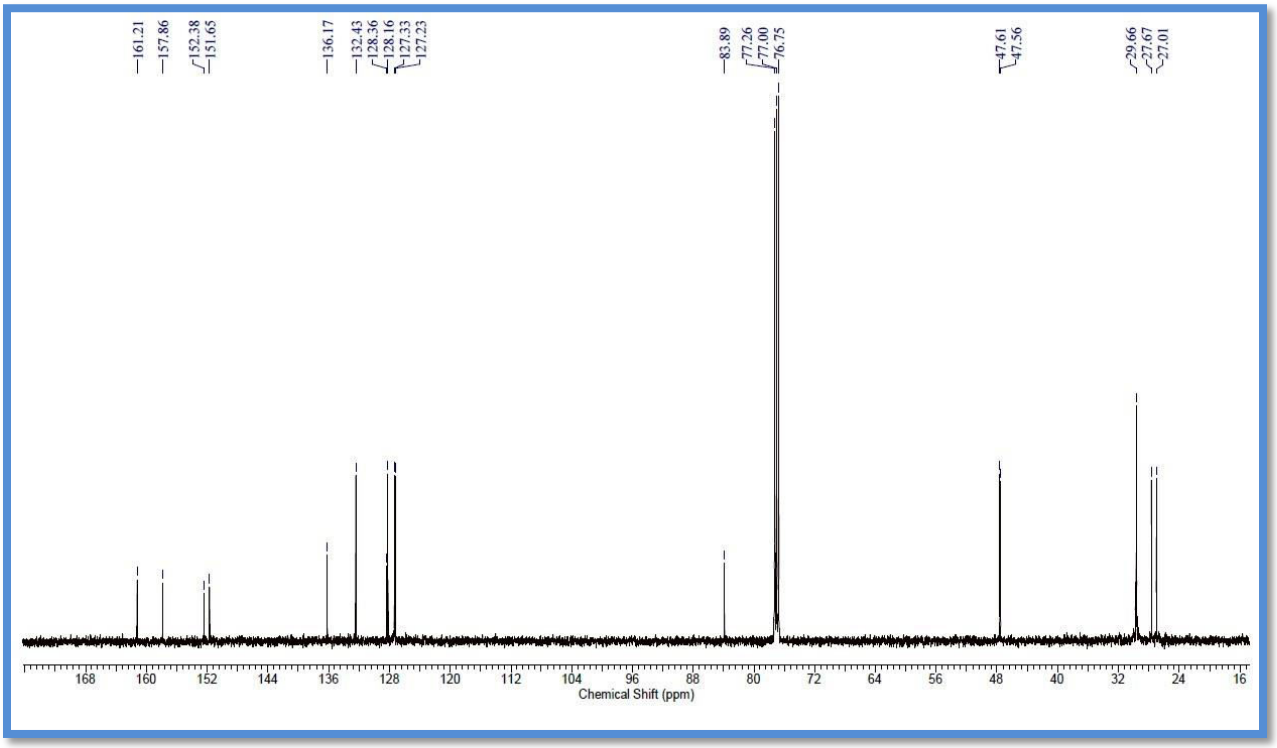
^{13}C-NMR of compound 4o



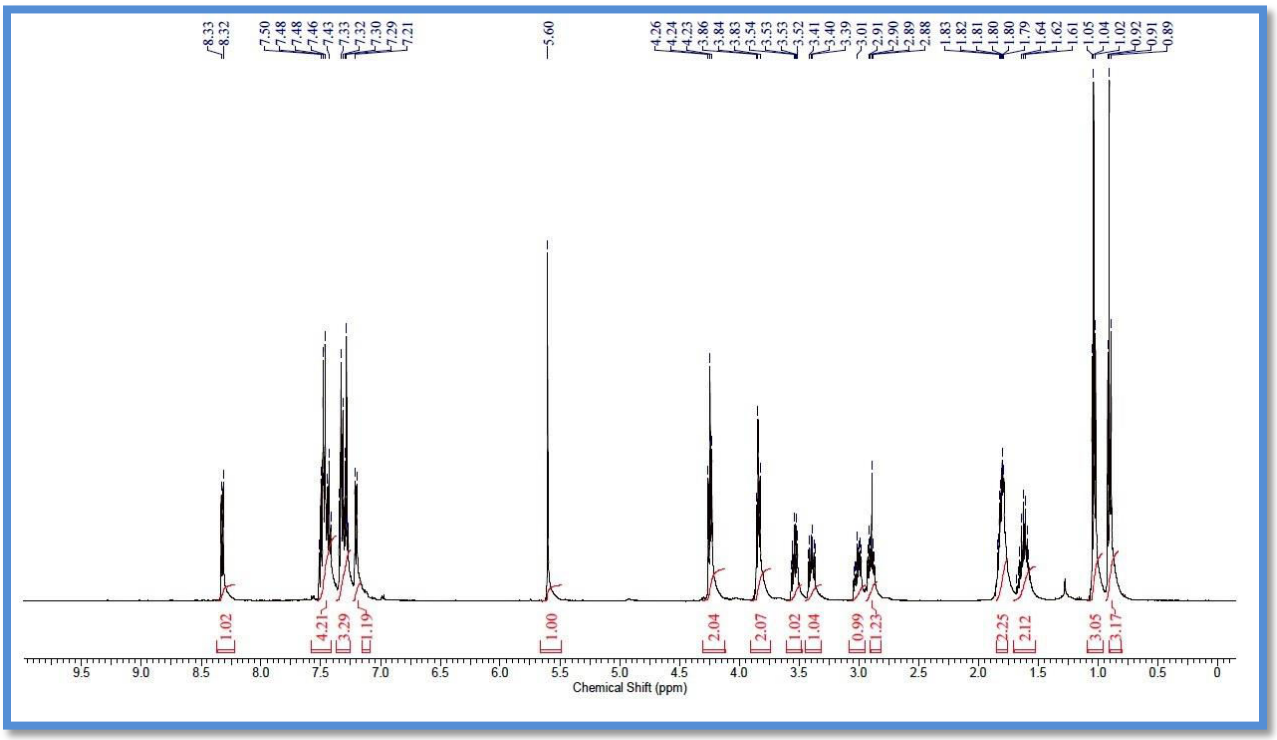
^{1}H-NMR of compound 4p



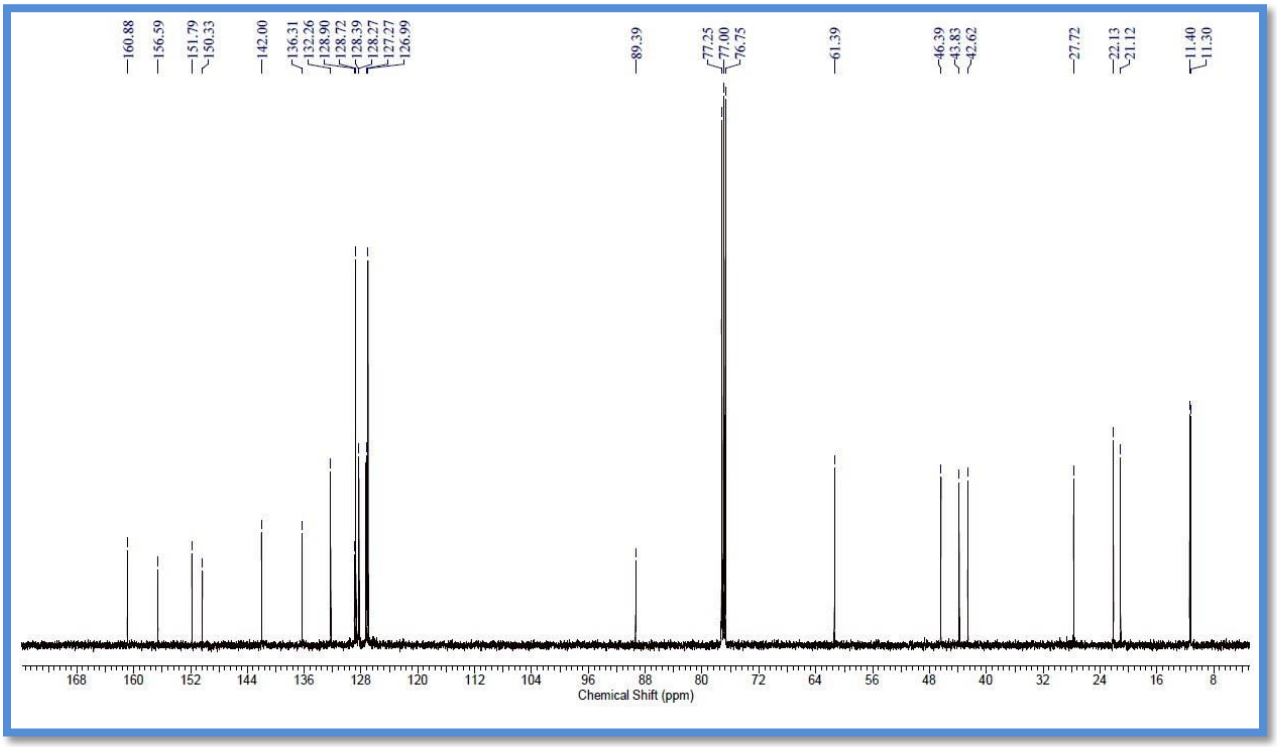
^{13}C-NMR of compound 4p



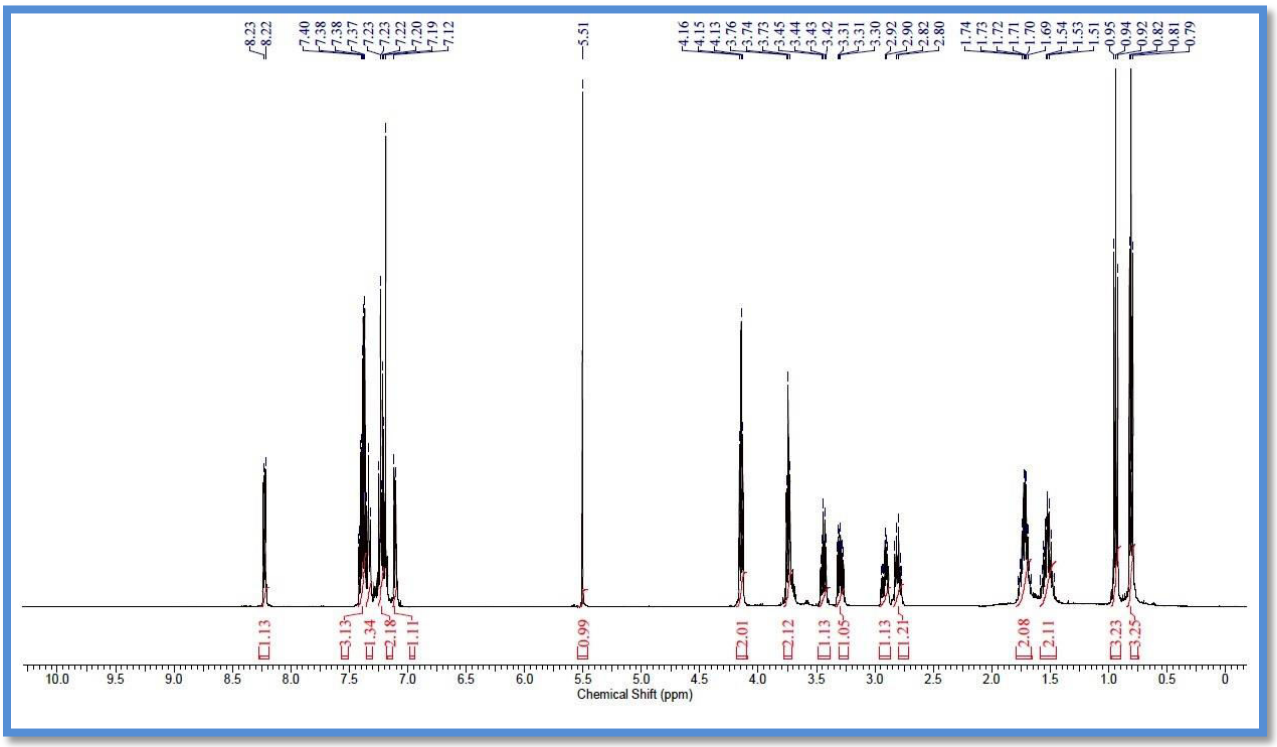
^{1}H-NMR of compound 4q



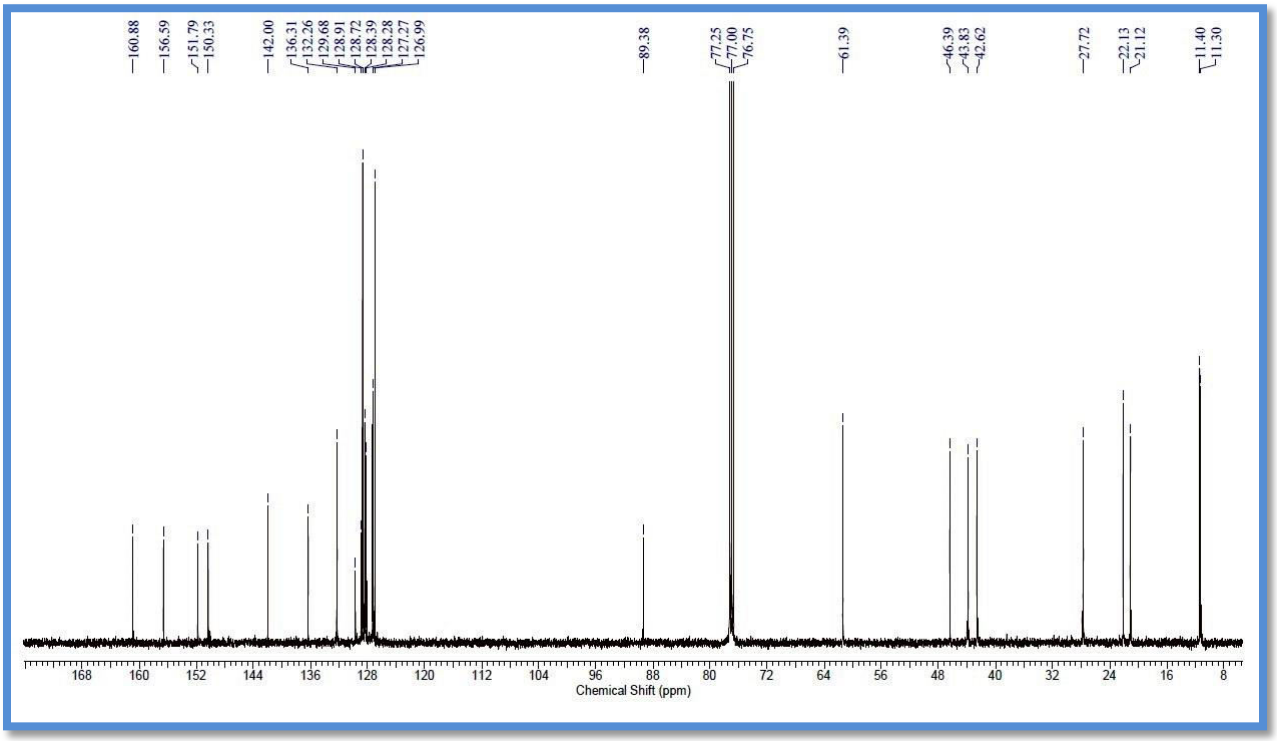
^{13}C-NMR of compound 4q



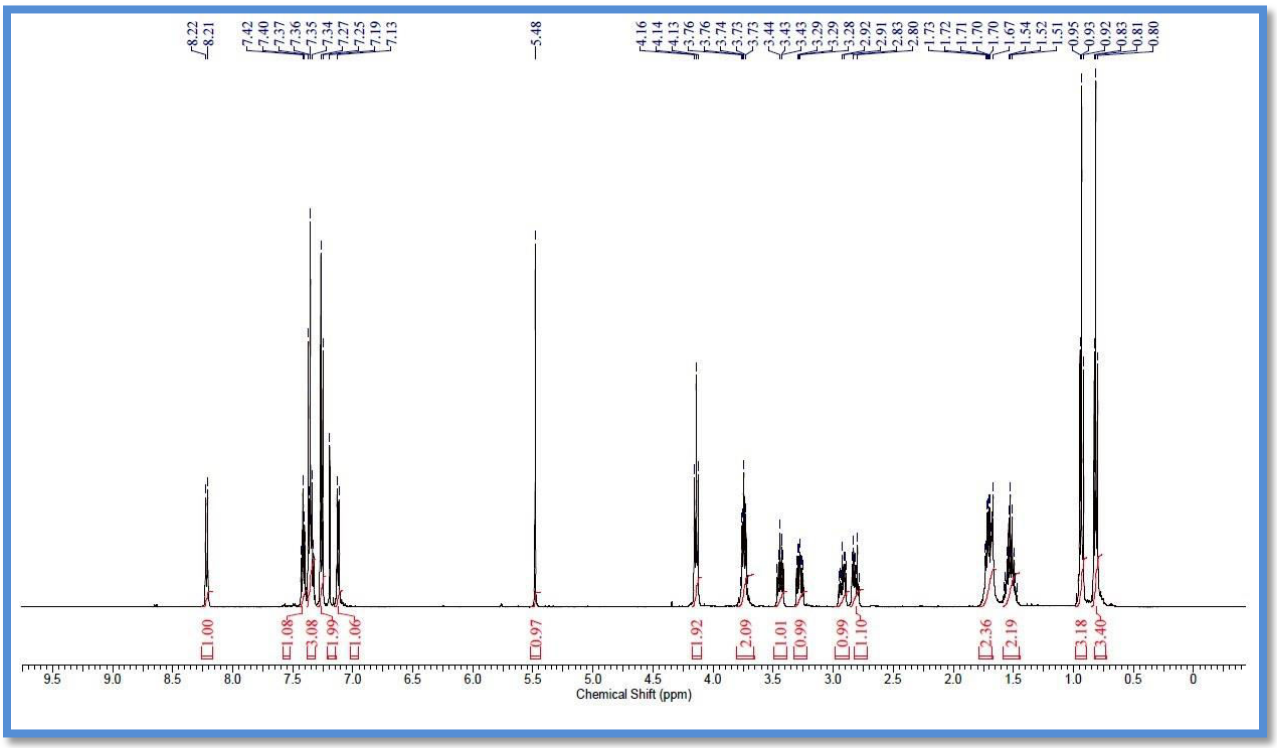
^{1}H-NMR of compound 4r



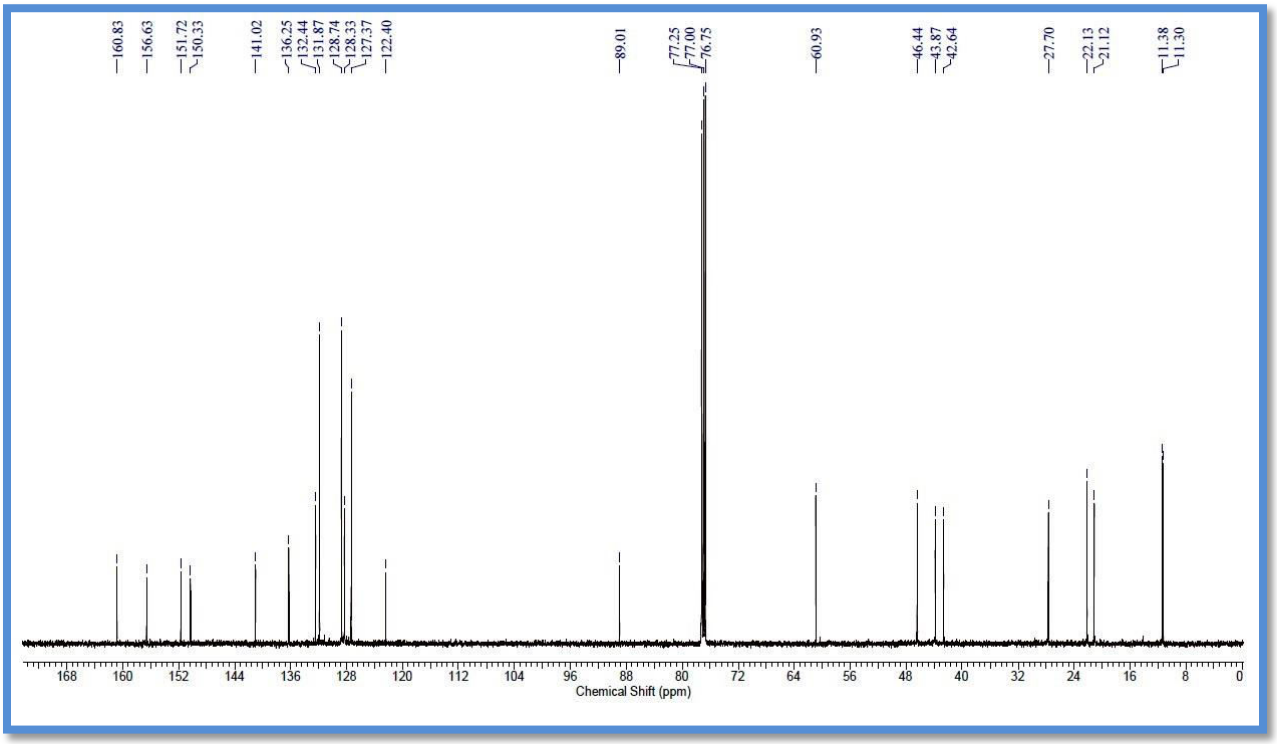
^{13}C-NMR of compound 4r



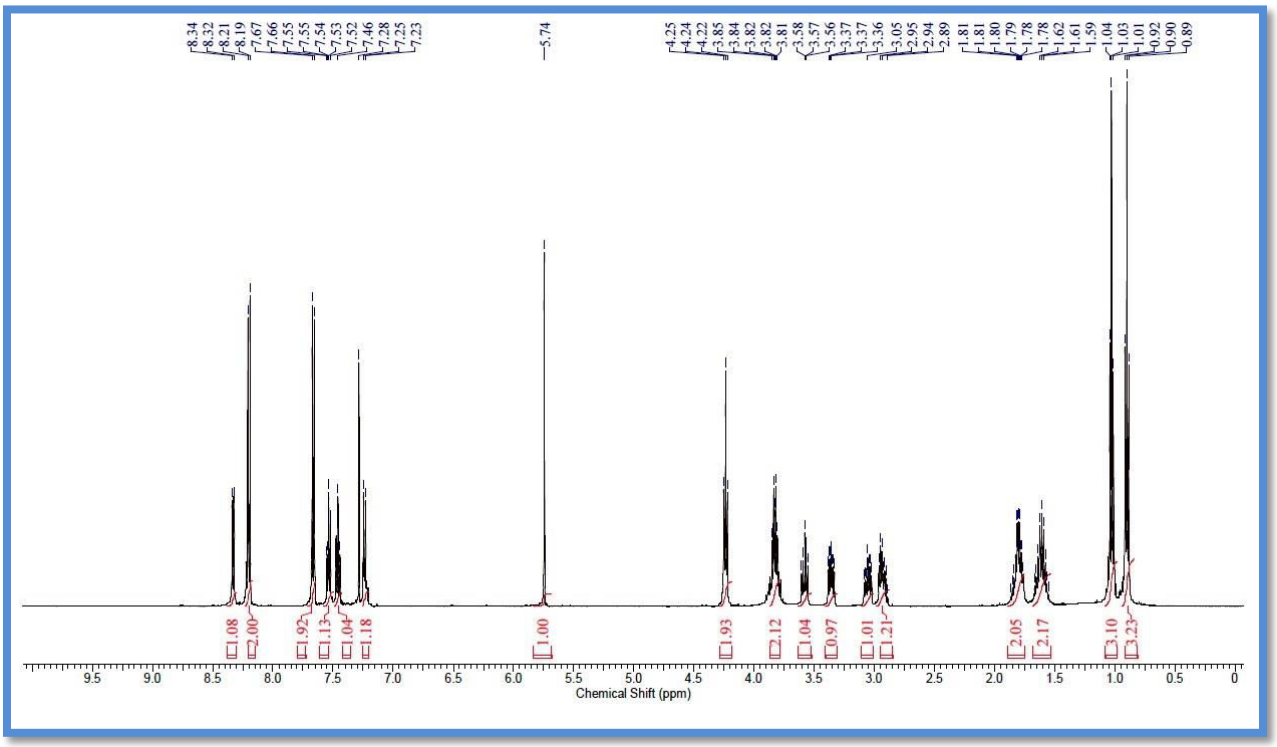
^{1}H-NMR of compound 4s



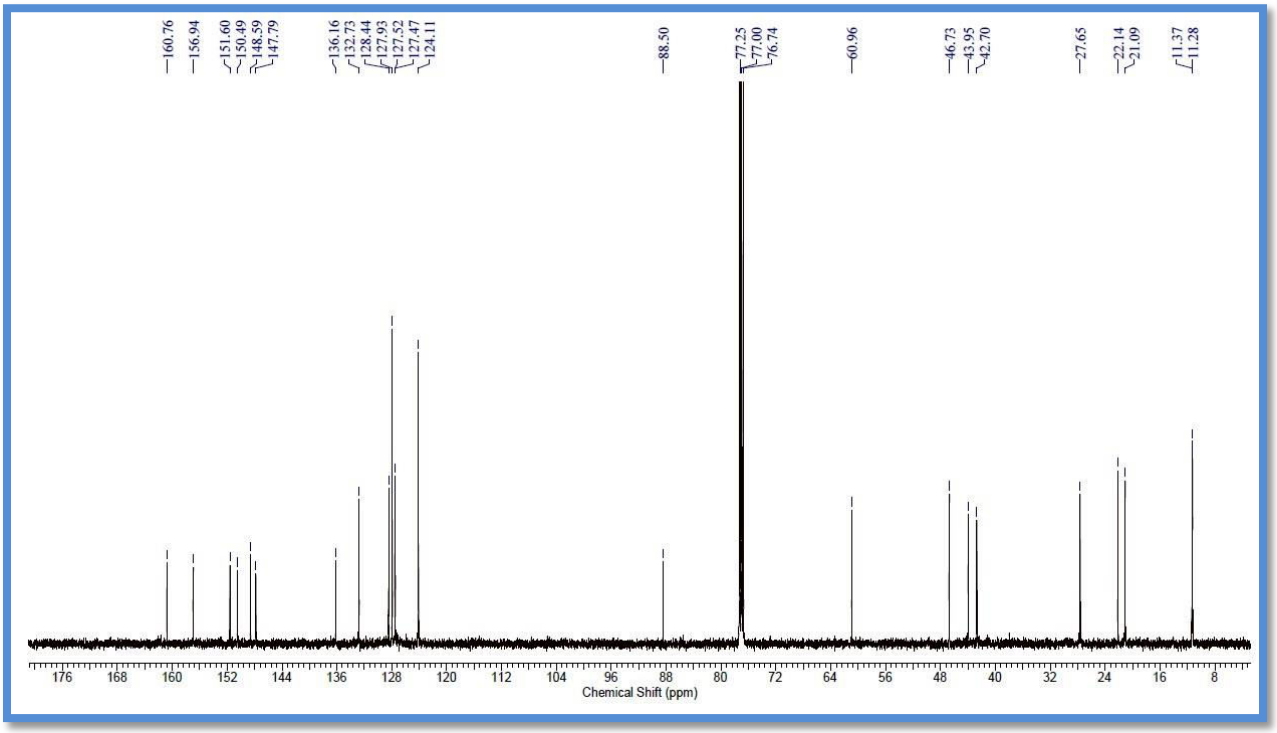
^{13}C-NMR of compound 4s



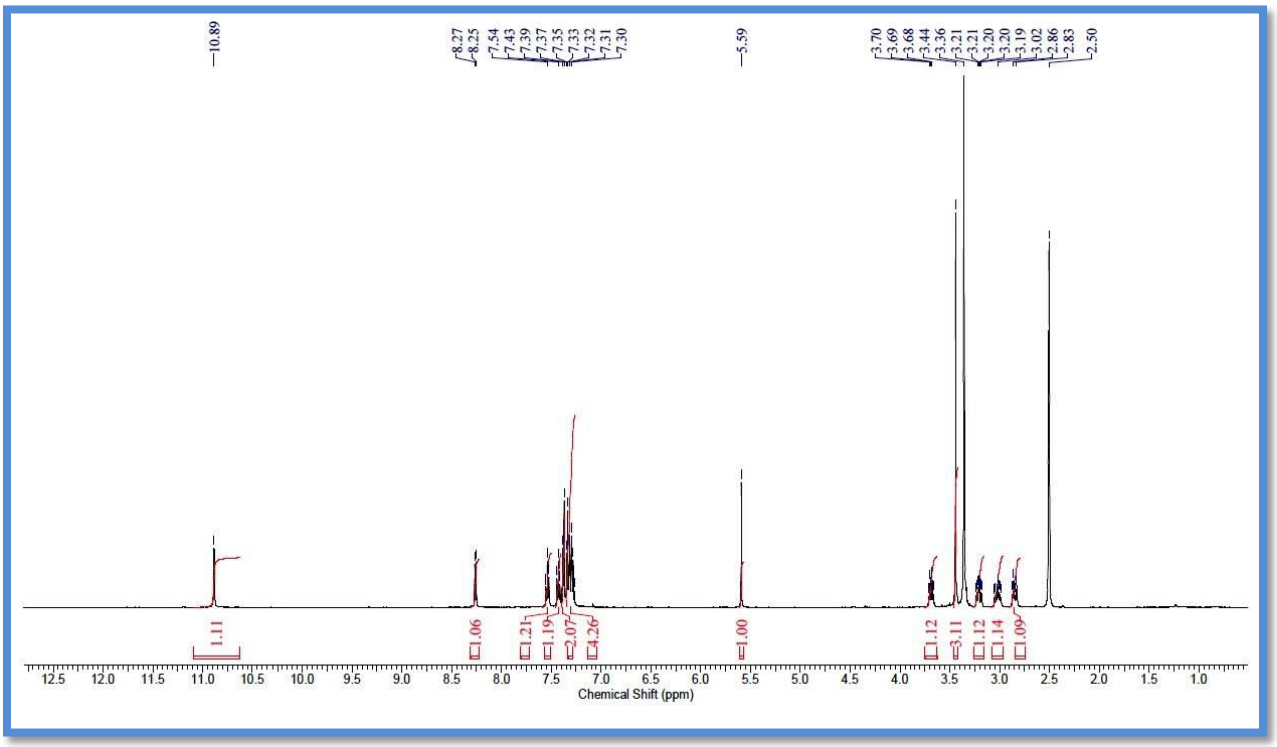
^{1}H-NMR of compound 4t



^{13}C-NMR of compound 4t



^{1}H-NMR of compound 4u



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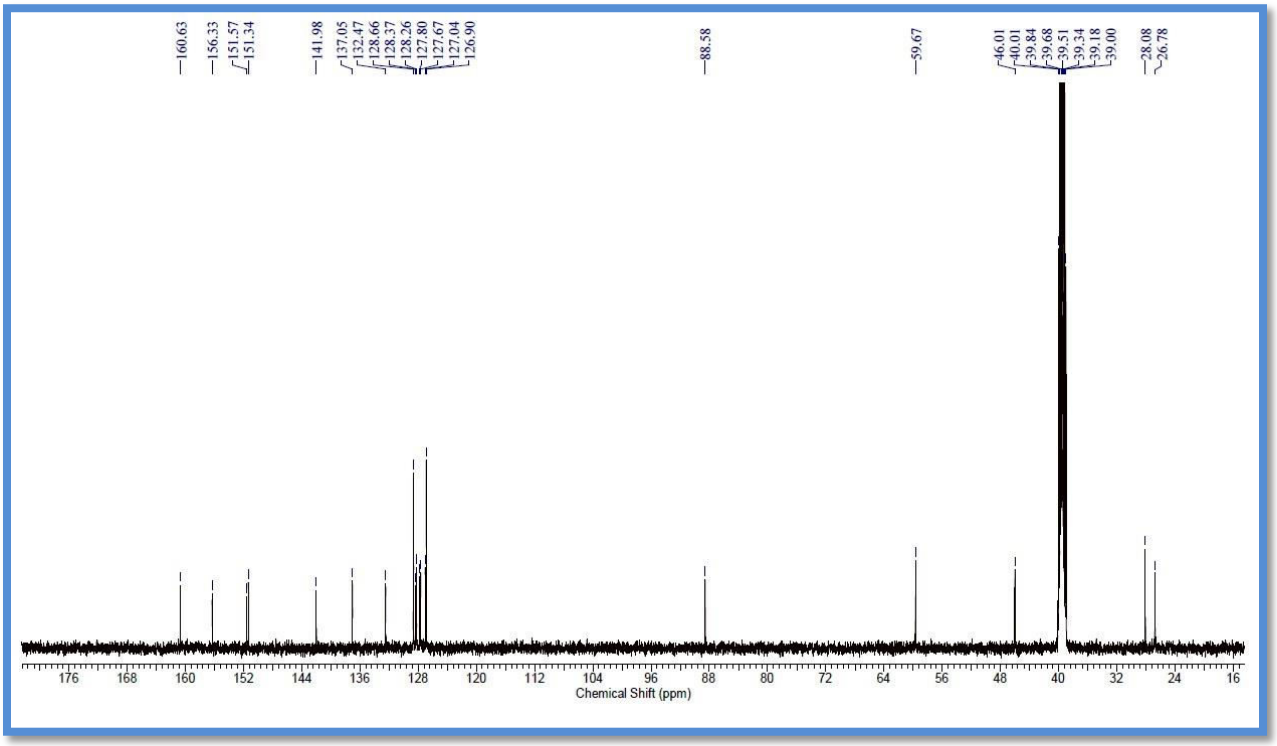
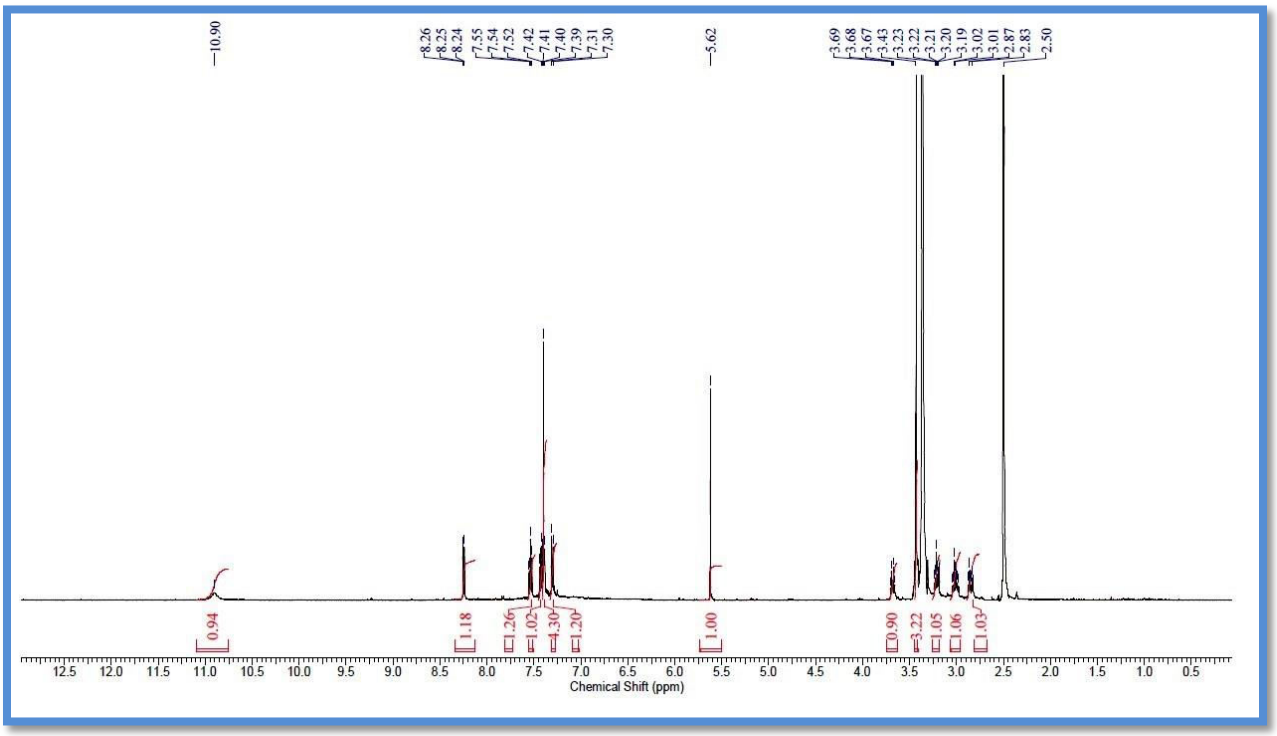
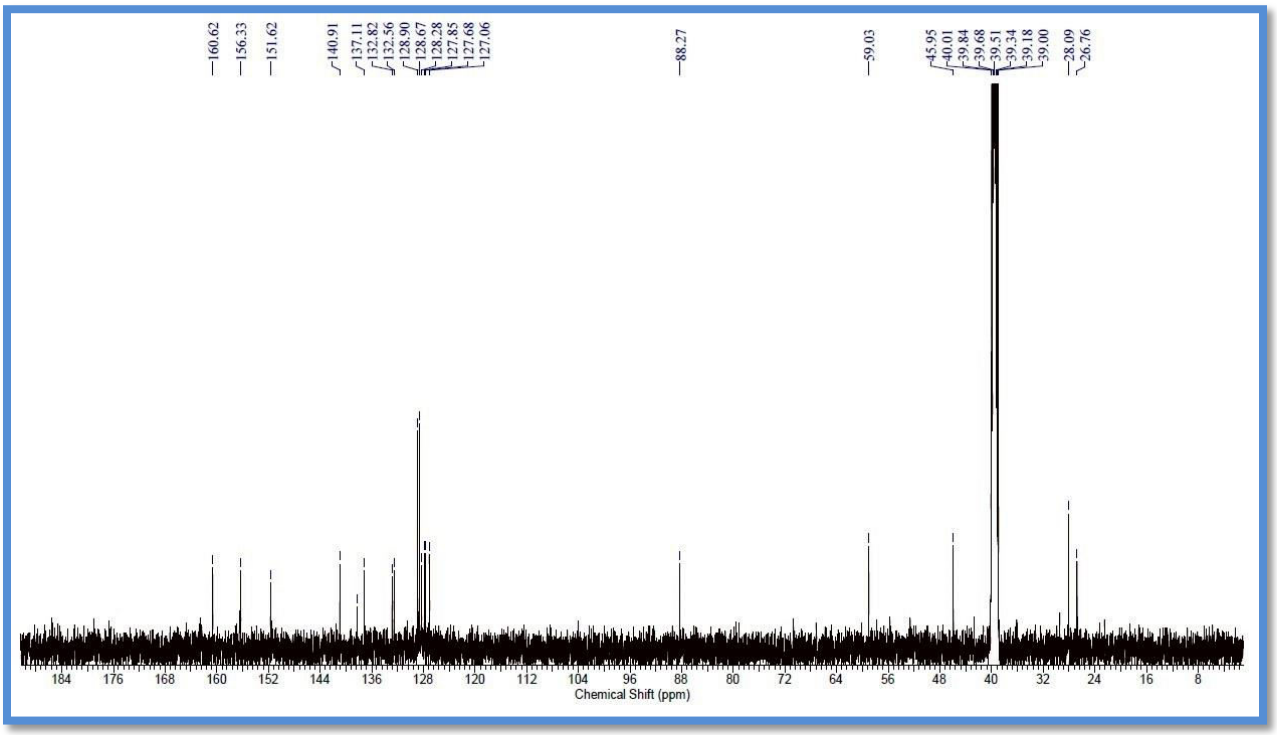


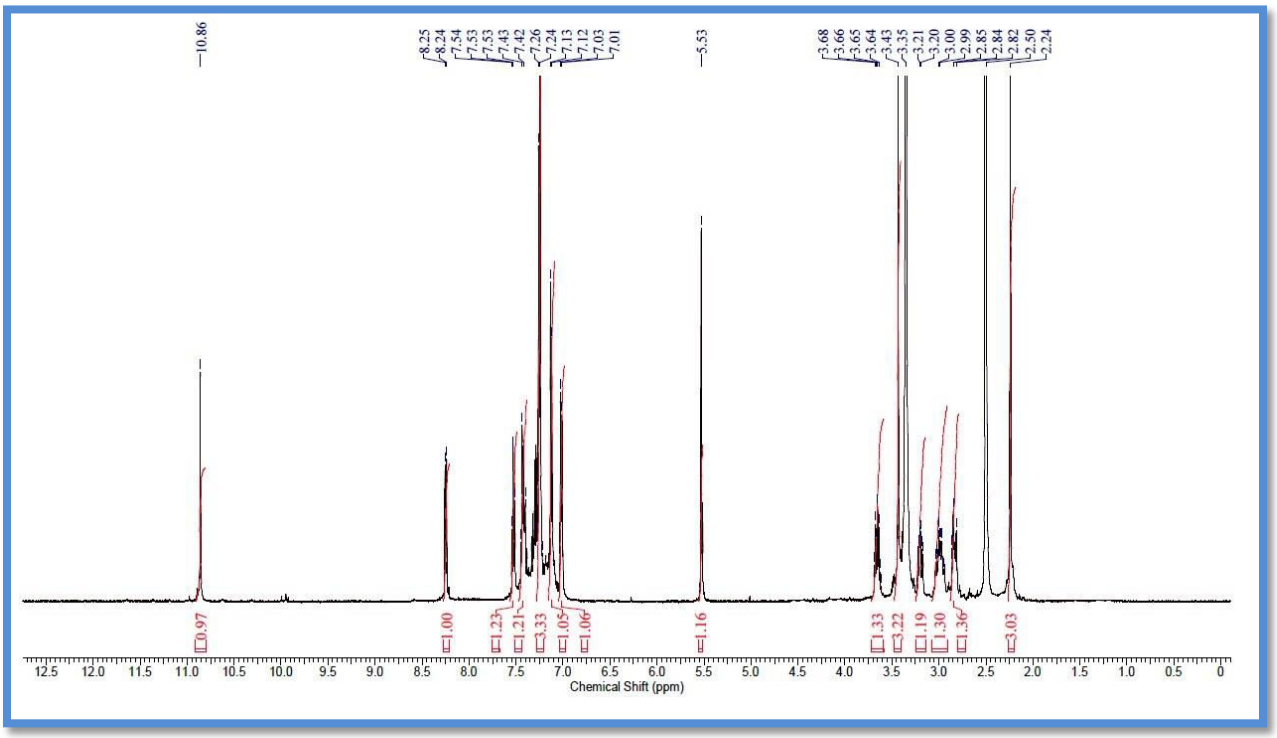
^{1}H-NMR of compound 4v



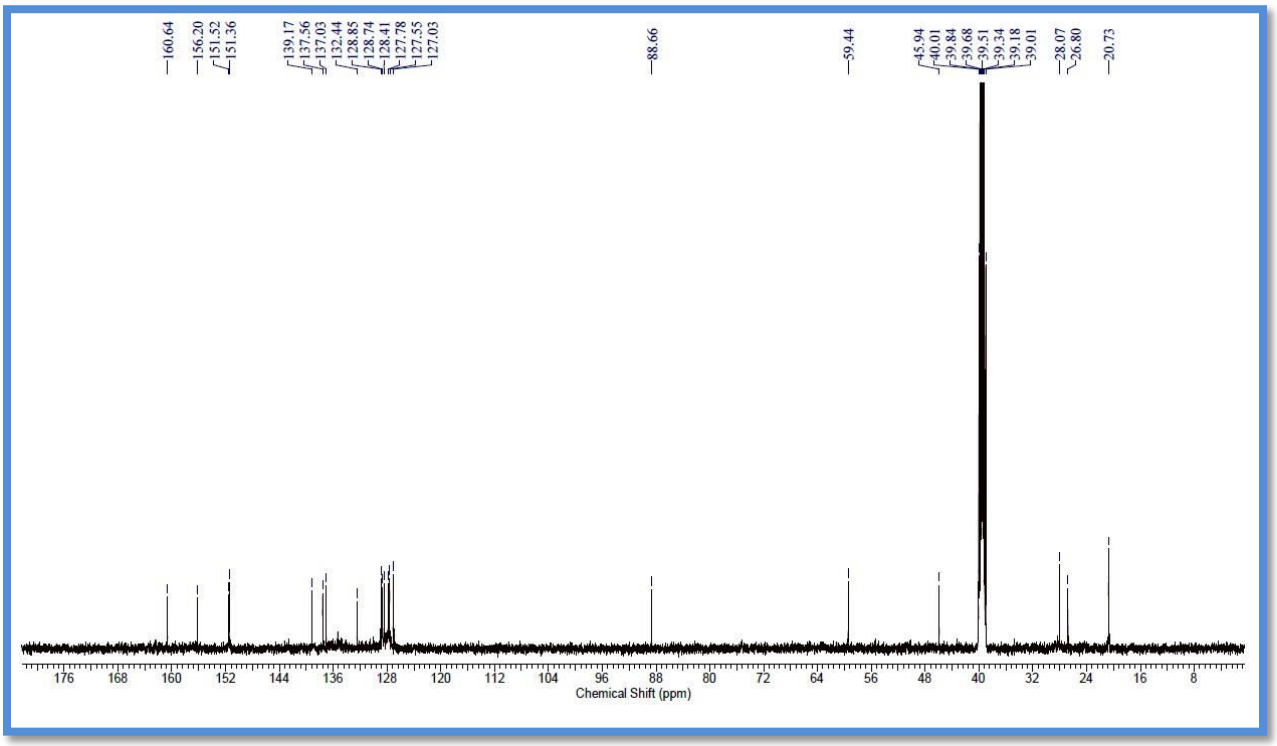
^{13}C-NMR of compound 4v



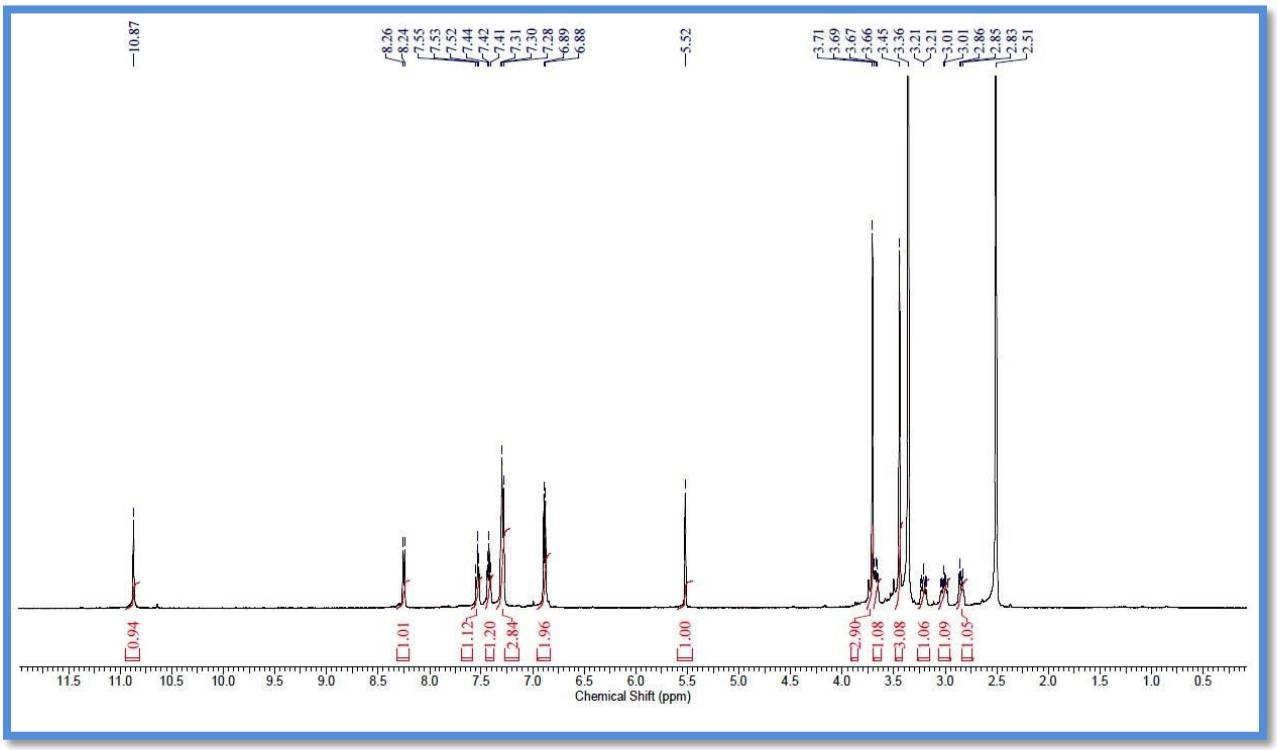
^{1}H-NMR of compound 4w



^{13}C-NMR of compound 4w



^{1}H-NMR of compound 4x



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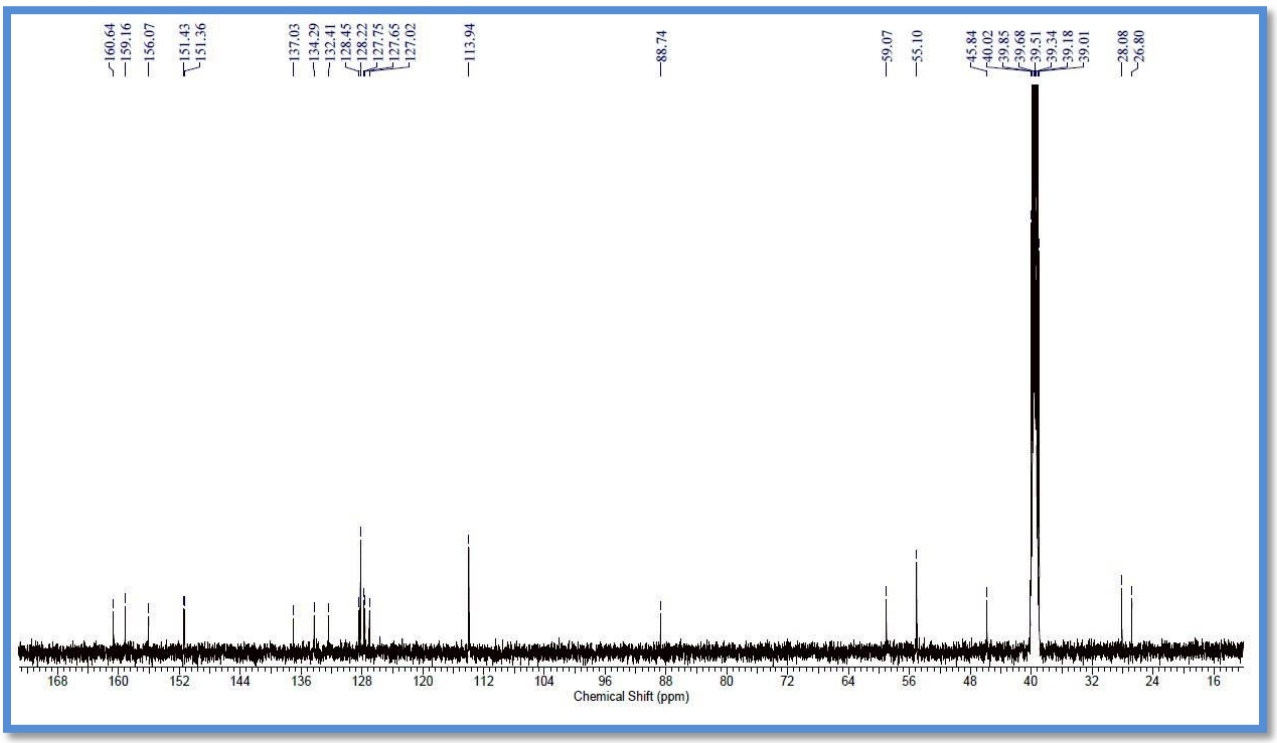

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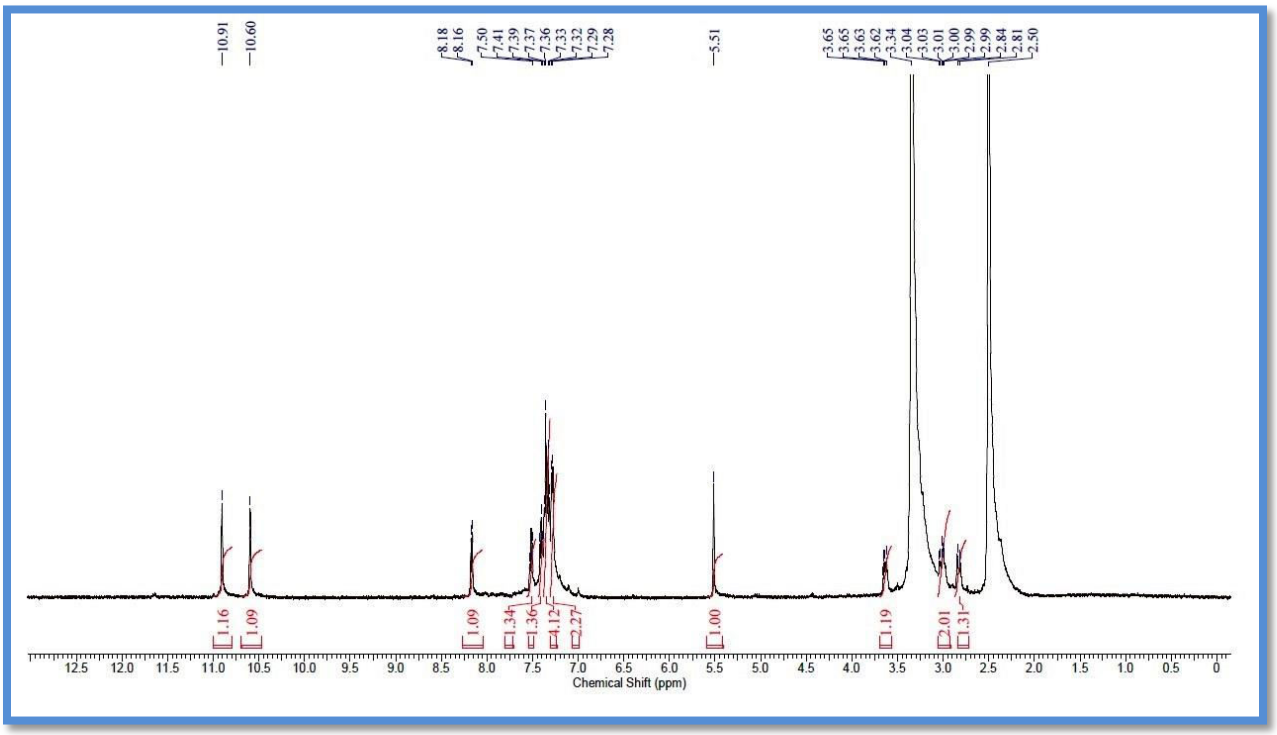

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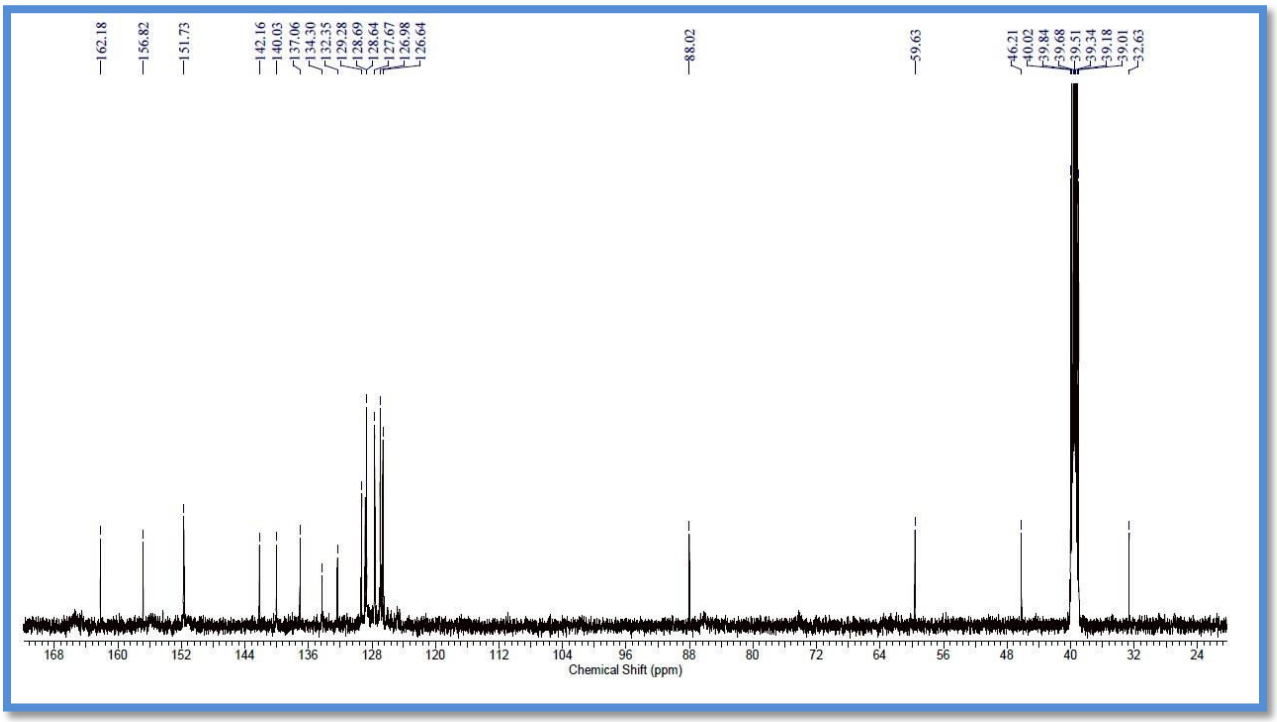
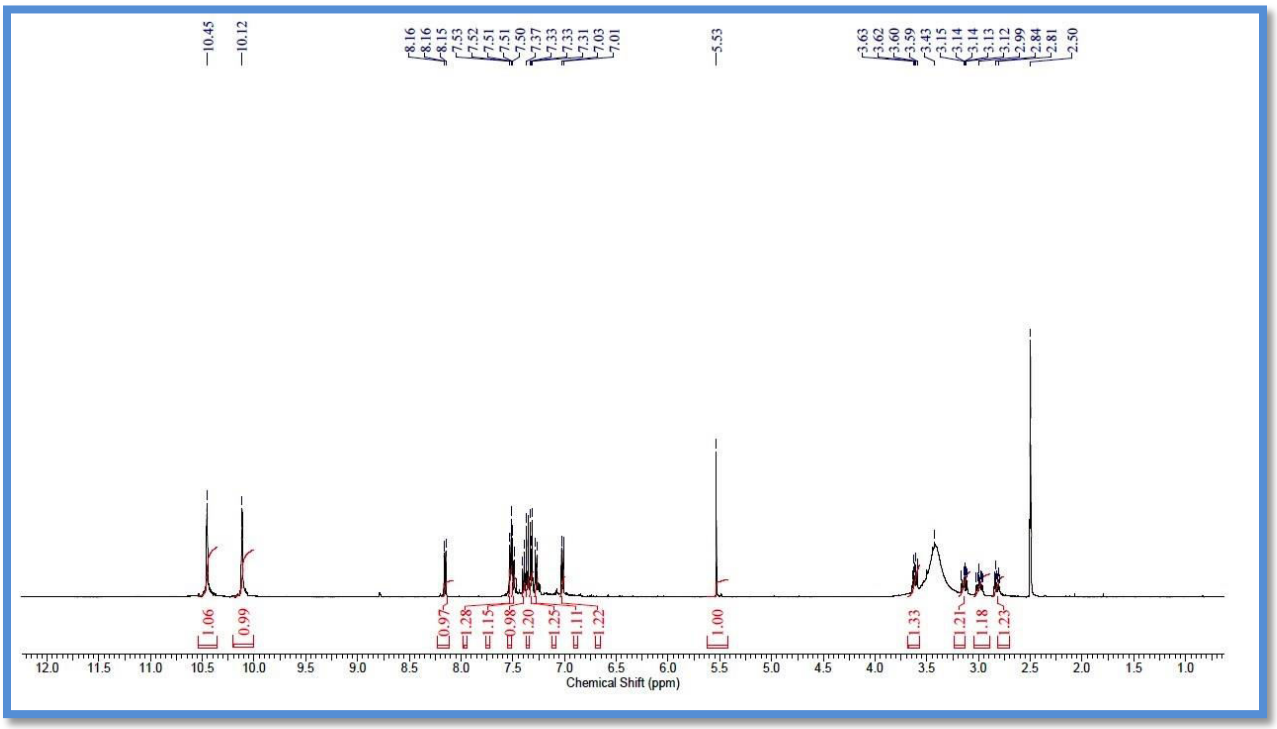
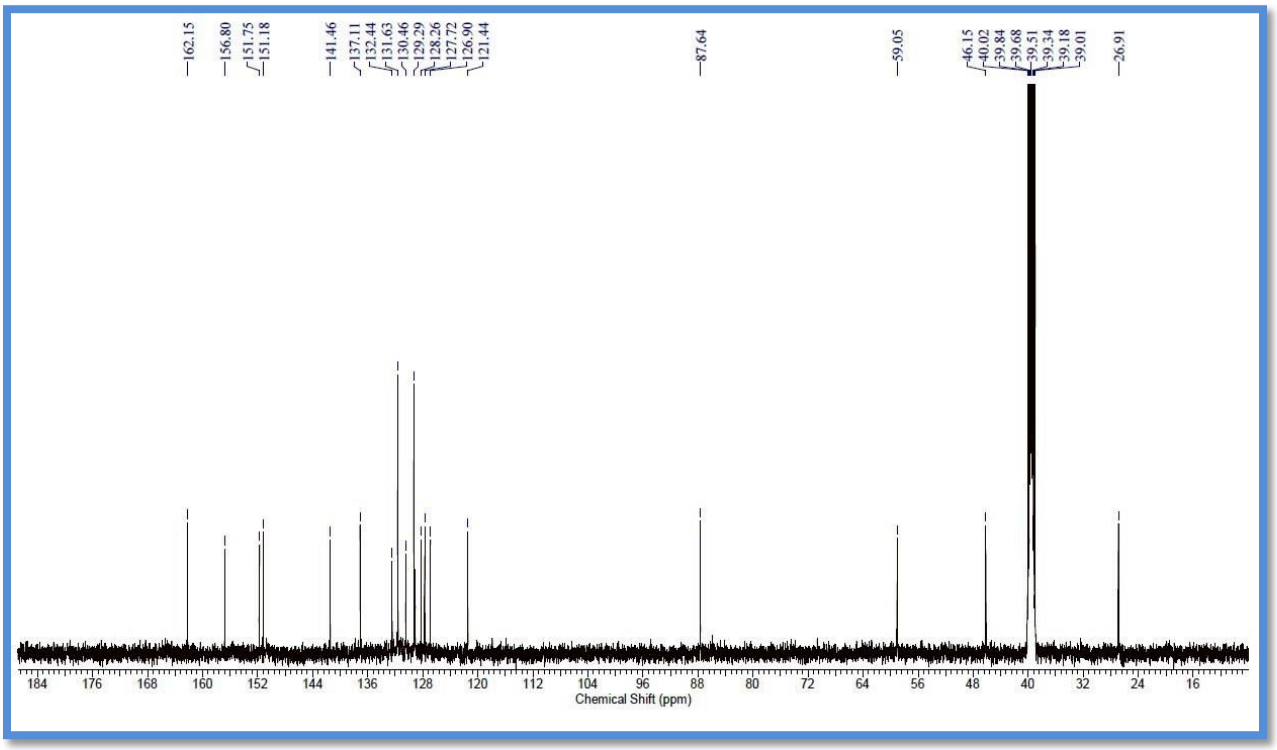


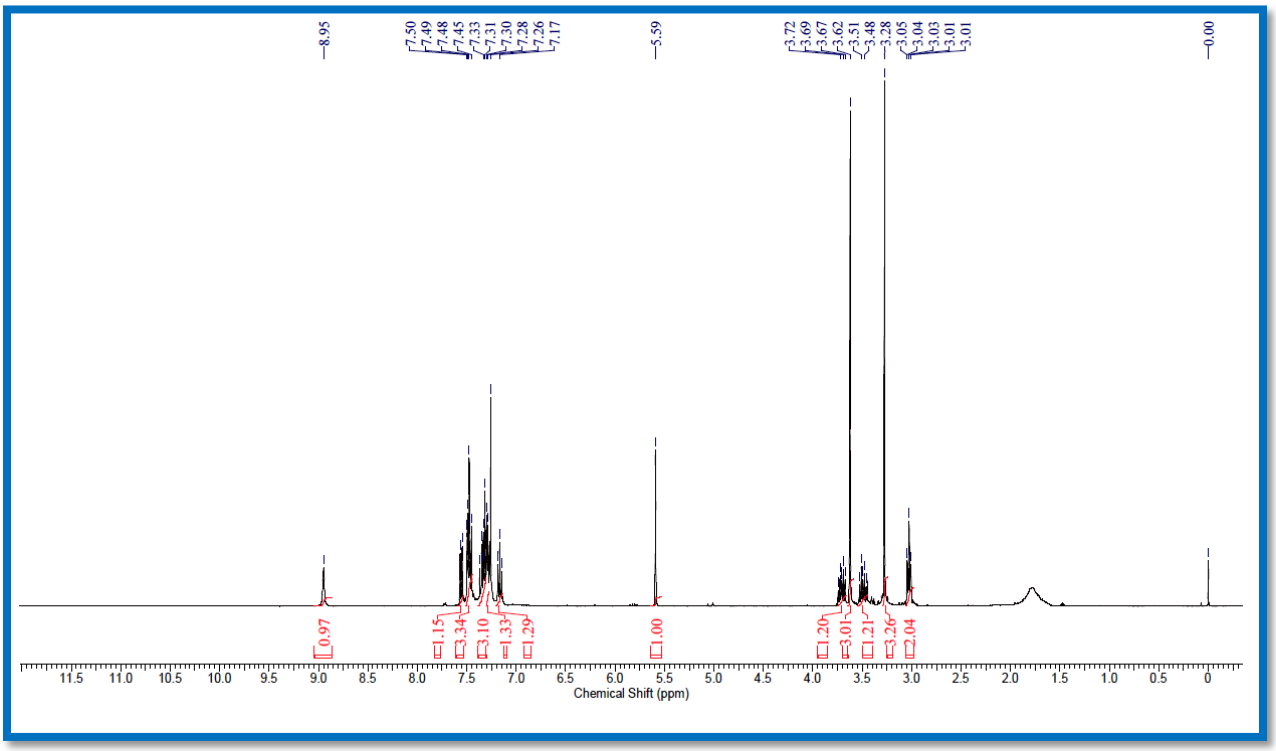
^{1}H-NMR of compound 4z



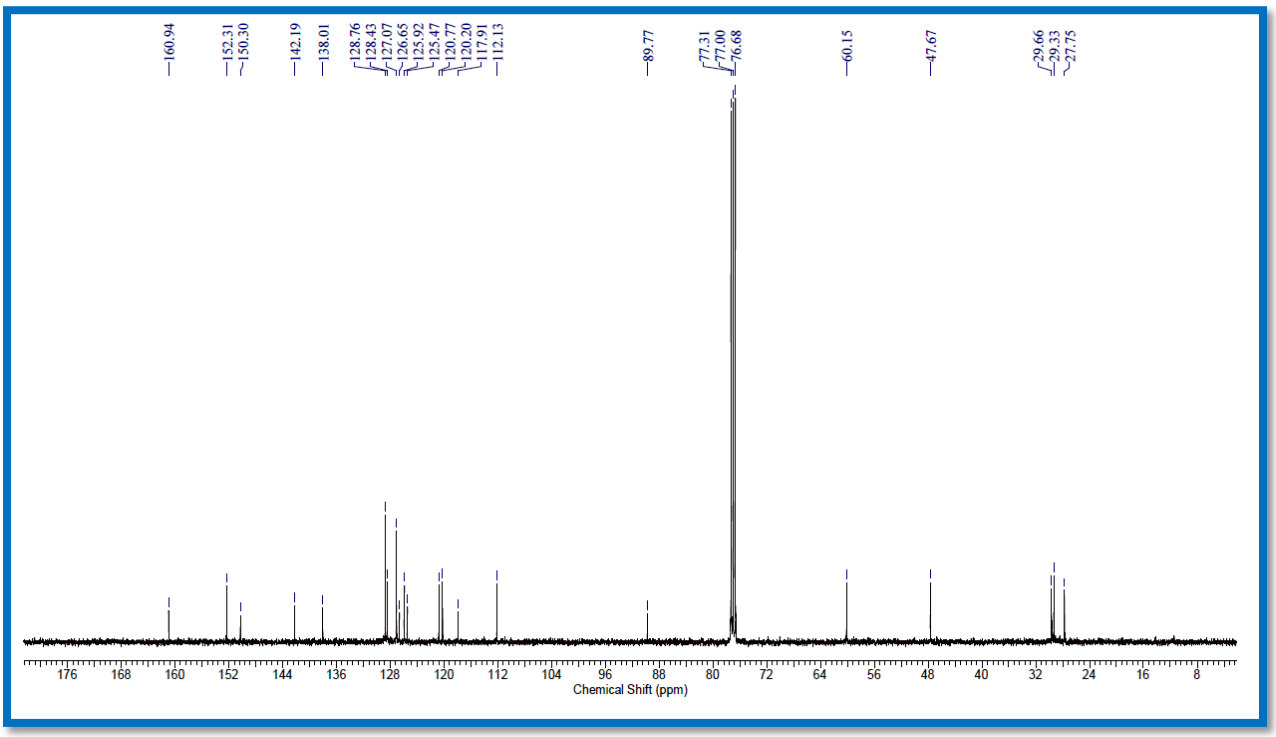
^{13}C-NMR of compound 4z



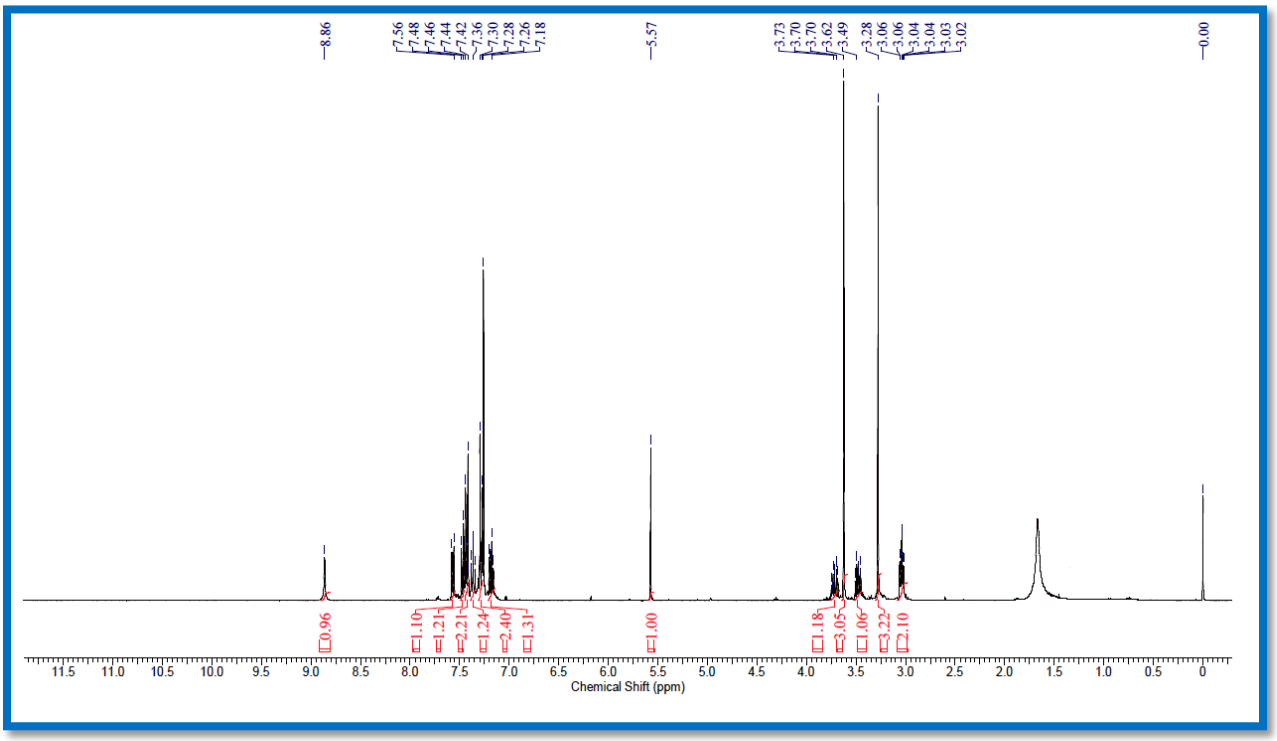
^{1}H-NMR of compound 4z1



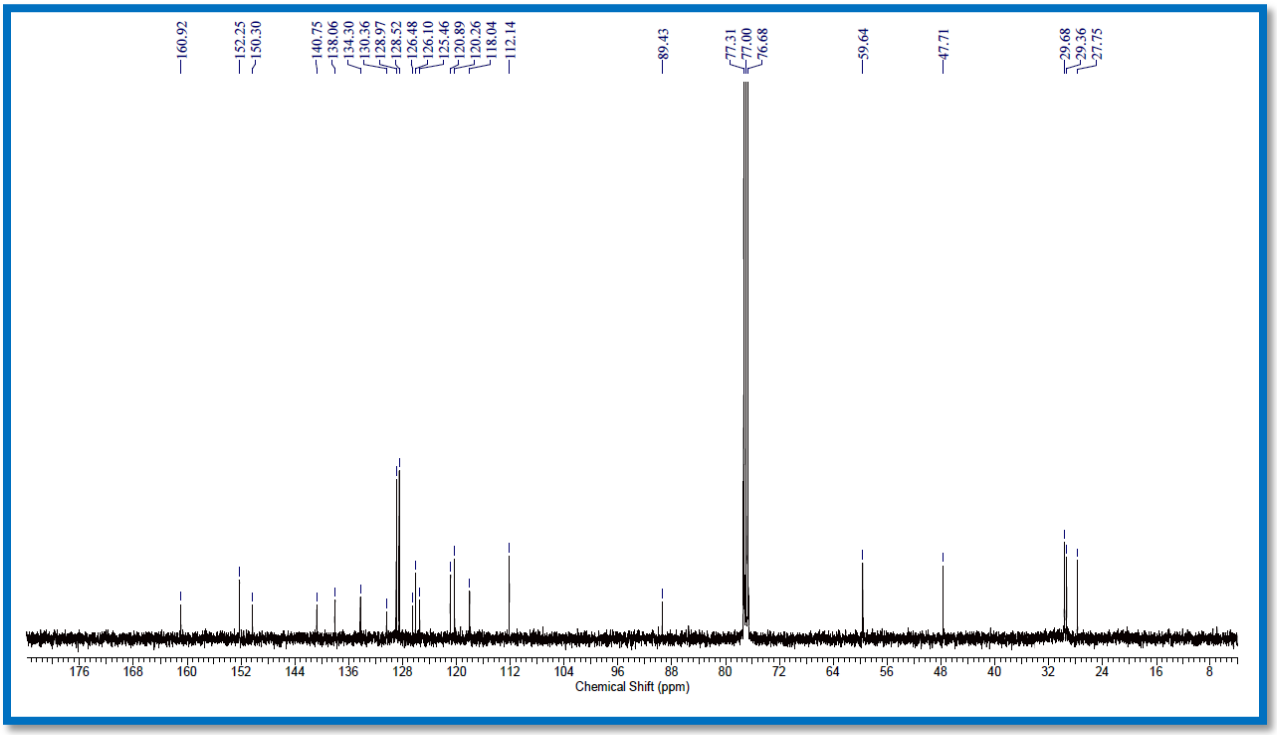
^{13}C-NMR of compound 4z1



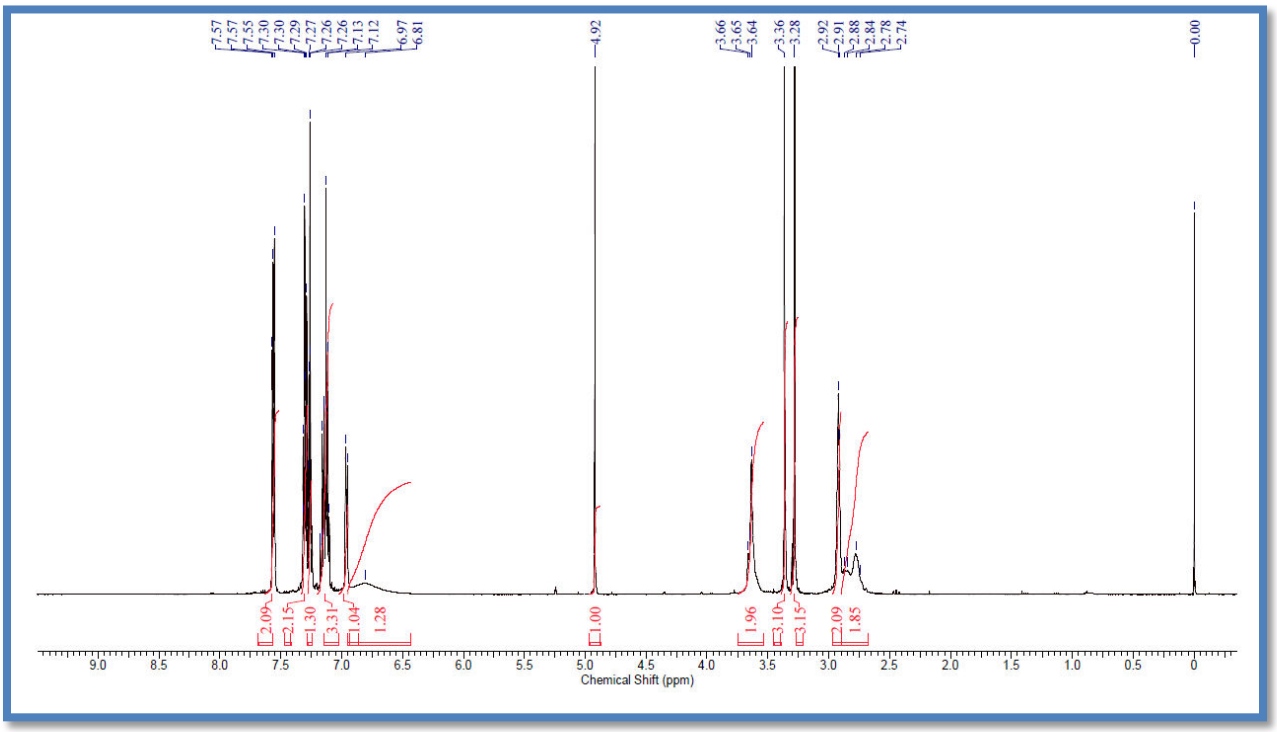
^{1}H-NMR of compound 4z2



^{13}C-NMR of compound 4z2



^{1}H-NMR of compound 4'a



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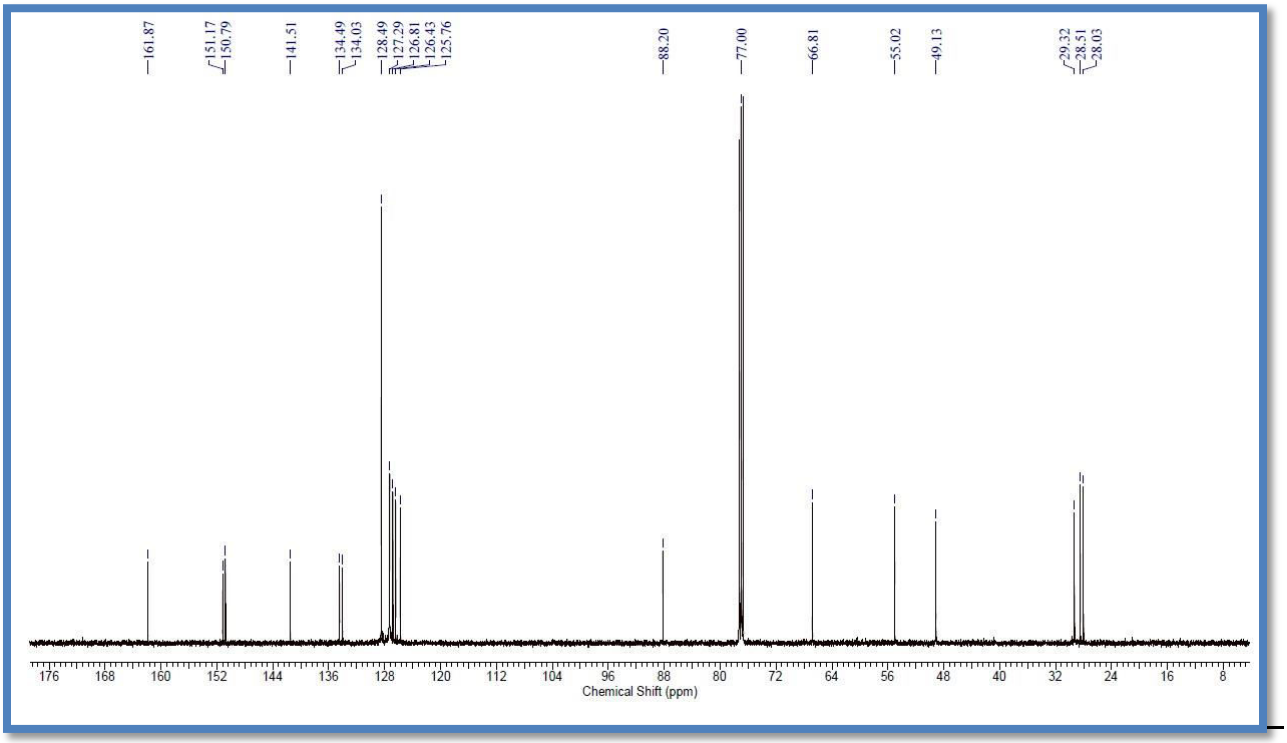
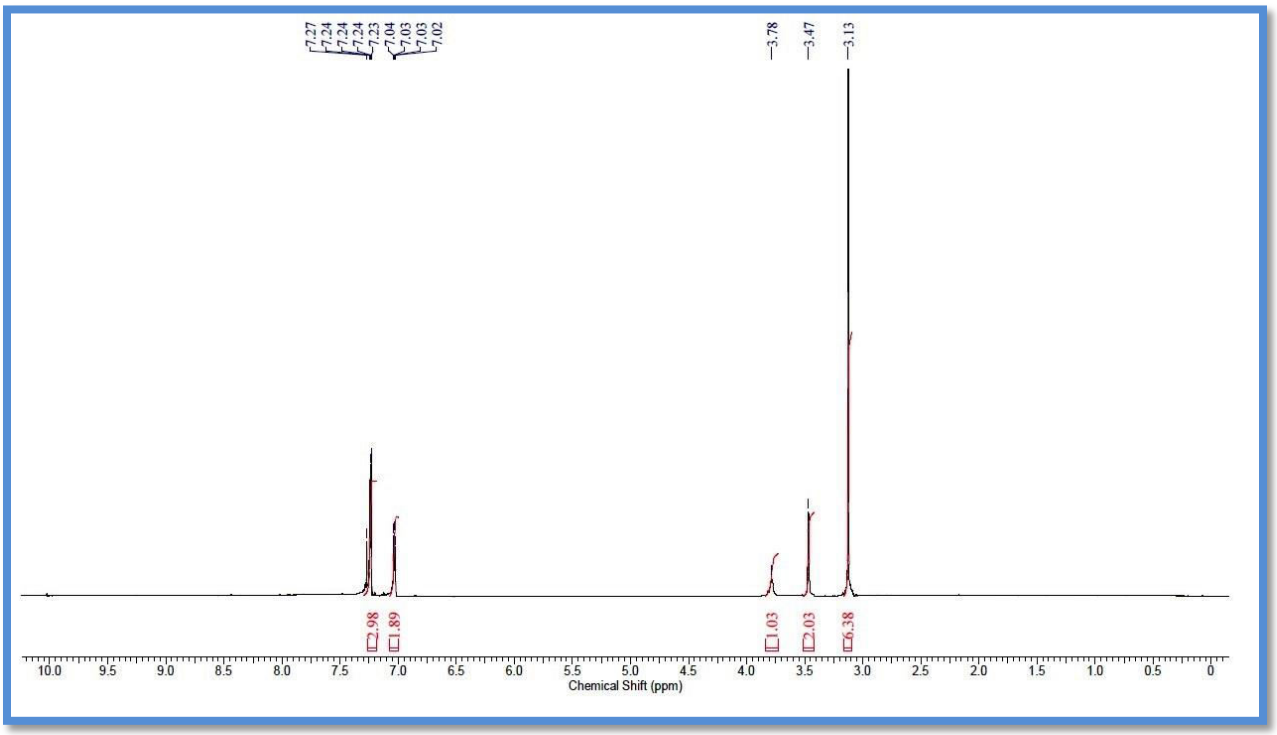
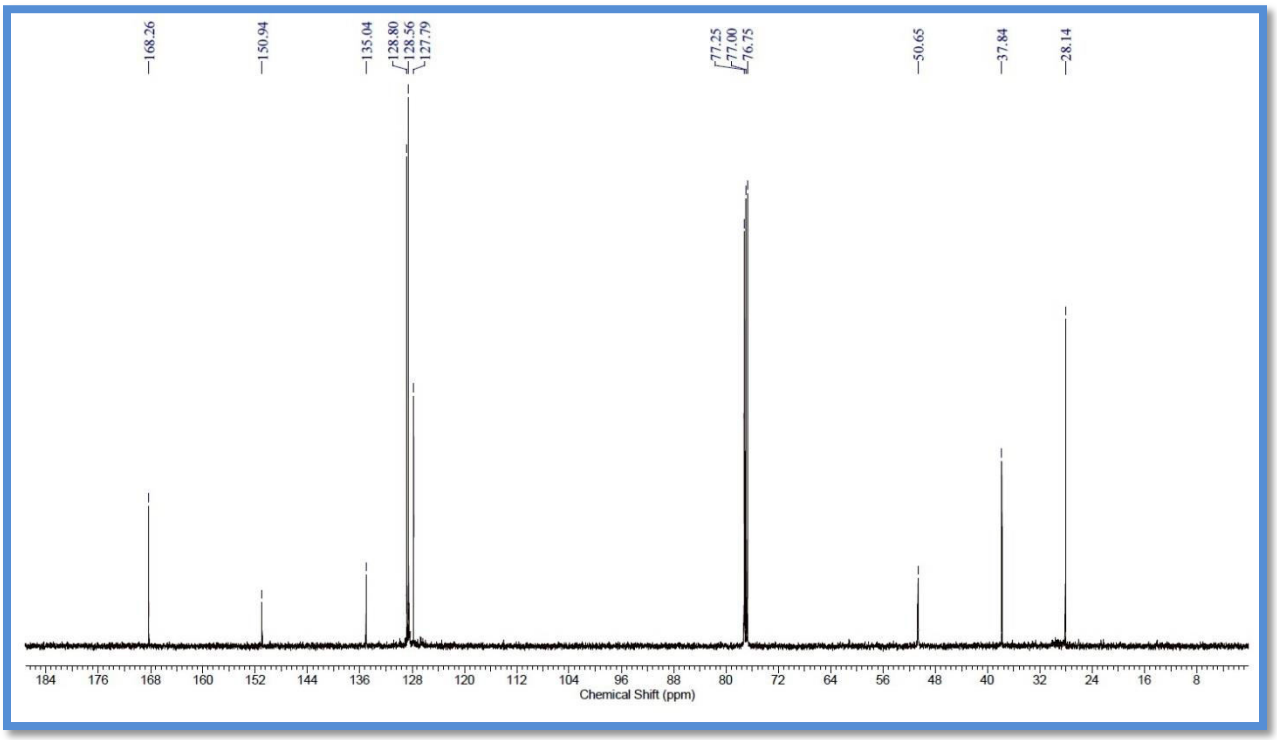


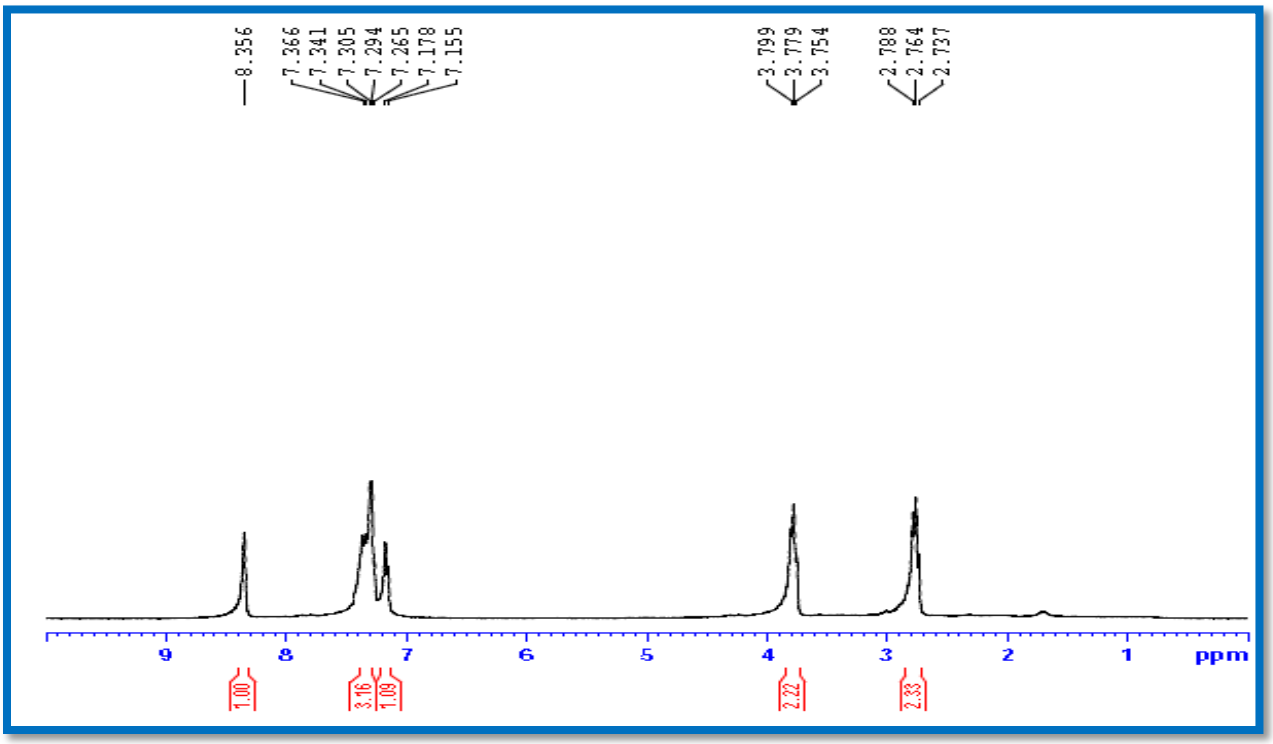
^{1}H-NMR of compound 7



^{13}C-NMR of compound 7



^{1}H-NMR of compound 8



^{13}C-NMR of compound 8