Erbium Triflate Promoted Multicomponent Synthesis of Highly Substituted Imidazoles

Kandasamy Rajaguru,[†] Rajendran Suresh,[‡] Arumugam Mariappan,[†] Shanmugam Muthusubramanian,[†] and Nattamai Bhuvanesh[#]

[†] *Department of Organic Chemistry, School of Chemistry, Madurai Kamaraj University, Madurai - 625 021, India*
[‡] *Syngene International Limited, Biocon, Bangalore – 560 100, India*
[#] *X-ray Diffraction Laboratory, Department of Chemistry, Texas A & M University, College Station, TX 77842, USA*

e-mail: muthumanian2001@yahoo.com

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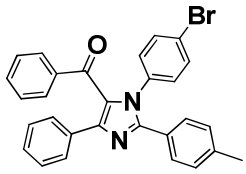
Experimental Section

General Remarks: Nuclear Magnetic Resonance (^{1}H and ^{13}C NMR) spectra were recorded on a 300 MHz spectrometer in $CDCl_3$ using TMS as an internal standard. Chemical shifts are reported in parts per million (δ), coupling constants (*J* values) are reported in Hertz (Hz) and spin multiplicities are indicated by the following symbols: s (singlet), d (doublet), t (triplet), m (multiplet). ^{13}C NMR spectra were routinely run with broadband decoupling.

General procedure for the synthesis of aryl(1,2,4-triaryl-1H-imidazol-5-yl)methanone (4): A mixture of substituted benzaldehyde **2** (0.2 mmol), substituted aniline **3** (0.2 mmol), and $Er(OTf)_3$ (0.004 mmol), in acetonitrile (10 mL) was stirred at room temperature for 10 minutes. Then, the substituted α-azido chalcone **1** (0.2 mmol) was added to the reaction mixture and refluxed for 3 h. The completion of the reaction was monitored by TLC. The reaction mixture was then quenched with saturated solutions of sodium bicarbonate and extracted with ethyl acetate. The organic layer was washed with water and brine, dried over anhydrous sodium sulfate and concentrated in vacuum. Crude product was purified by column chromatography using ethyl acetate − hexane as the solvent to get **4**.

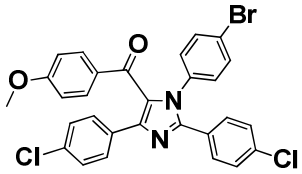
Characterization data for compounds (4a-4r)

(1-(4-Bromophenyl)-4-phenyl-2-p-tolyl-1H-imidazol-5-yl)(phenyl)methanone (4a)



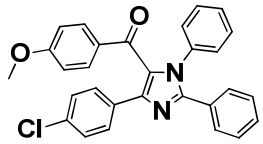
Isolated as white solid; mp 190-192 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$): δ 7.68 (d, *J* = 7.9 Hz, 2H), 7.49 –7.42 (m, 4H), 7.34 (d, *J* = 8.2 Hz, 2H), 7.24 – 7.08 (m, 10H), 2.32 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 187.3, 150.1, 147.1, 139.4, 137.5, 136.2, 133.2, 132.9, 132.4, 129.8, 129.4, 129.1, 129.0, 128.7, 128.0, 127.9, 127.8, 126.2, 122.7, 21.3 (One of the ring quaternary carbons is not picked up). Anal. Calcd for: $C_{29}H_{21}BrN_2O$: C, 70.59; H, 4.29; N, 5.68%. Found: C, 70.52; H, 4.25; N, 5.71%.

(1-(4-Bromophenyl)-2,4-bis(4-chlorophenyl)-1H-imidazol-5-yl)(4-methoxyphenyl) methanone (4b)



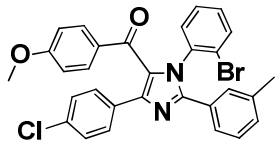
Isolated as white solid; mp 203-205 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.70 (d, *J* = 8.7 Hz, 2H), 7.49 (d, *J* = 8.5 Hz, 2H), 7.45 (d, *J* = 8.5 Hz, 2H), 7.37 (d, *J* = 8.5 Hz, 2H), 7.28 (d, *J* = 8.5 Hz, 2H), 7.17 (d, *J* = 8.4 Hz, 2H), 7.10 (d, *J* = 8.4 Hz, 2H), 6.72 (d, *J* = 8.7 Hz, 2H), 3.79 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.1, 164.1, 148.1, 143.6, 135.8, 135.6, 133.8, 132.6, 132.3, 131.7, 130.2, 129.8, 129.3, 128.7, 128.4, 127.7, 123.2, 113.8, 55.5. (Two of the ring quaternary carbons are not picked up). Anal. Calcd for: $C_{29}H_{19}BrCl_2N_2O_2$: C, 60.23; H, 3.31; N, 4.84%. Found: C, 60.20; H, 3.37; N, 4.81%.

(4-(4-Chlorophenyl)-1,2-diphenyl-1H-imidazol-5-yl)(4-methoxyphenyl)methanone (**4c**)



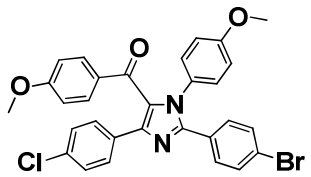
Isolated as brown solid; mp 215-217 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$): δ 7.72 (d, *J* = 8.5 Hz, 2H), 7.50 (d, *J* = 8.2 Hz, 2H), 7.44 (d, *J* = 6.9 Hz, 2H), 7.34 – 7.15 (m, 10H), 6.72 (d, *J* = 8.5 Hz, 2H), 3.78 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.6, 163.9, 149.2, 143.3, 137.0, 133.5, 132.3, 132.1, 130.6, 129.8, 129.6, 129.3, 129.2, 129.1, 128.8, 128.3, 128.2, 127.8, 113.8, 55.4 (One of the ring quaternary carbons is not picked up). Anal. Calcd for: $C_{29}H_{21}ClN_2O_2$: C, 74.91; H, 4.55; N, 6.03%. Found C, 74.88; H, 4.58; N, 6.09%.

(1-(2-Bromophenyl)-4-(4-chlorophenyl)-2-(m-tolyl)-1H-imidazol-5-yl)(4-methoxyphenyl) methanone (**4d**)



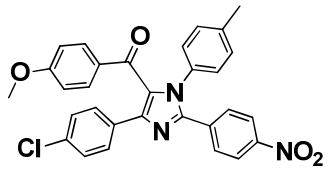
Isolated as brown solid; mp 236-237 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.70 (d, *J* = 8.0 Hz, 2H), 7.58 (d, *J* = 7.9 Hz, 1H), 7.51 (d, *J* = 7.6 Hz, 1H), 7.43 – 7.35 (m, 5H), 7.29 (d, *J* = 7.6 Hz, 1H), 7.14 – 7.06 (m, 4H), 6.67 (d, *J* = 8.1 Hz, 2H), 3.77 (s, 3H), 2.31 (s, 3H). ^{13}C NMR (75 MHz, $CDCl_3$): δ 185.5, 163.6, 150.0, 144.9, 139.5, 136.7, 133.6, 133.3, 132.5, 132.1, 131.5, 131.1, 130.6, 130.4, 129.9, 129.0, 128.8, 128.6, 128.1, 126.4, 122.7, 113.8, 113.4, 55.4, 21.3 (One of the ring quaternary carbons is not picked up). Anal. Calcd for: $C_{30}H_{22}BrClN_2O_2$: C, 64.59; H, 3.97; N, 5.02%. Found C, 64.37; H, 3.89; N, 4.98%.

(2-(4-Bromophenyl)-4-(4-chlorophenyl)-1-(4-methoxyphenyl)-1H-imidazol-5-yl)(4-methoxyphenyl)methanone (**4e**)



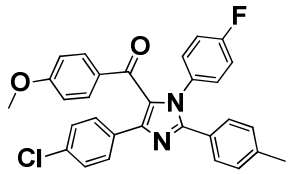
Isolated as pale brown solid; mp 206-208 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.71 (d, *J* = 8.8 Hz, 2H), 7.47 (d, *J* = 8.4 Hz, 2H), 7.41 (d, *J* = 8.6 Hz, 2H), 7.34 (d, *J* = 8.5 Hz, 2H), 7.17 (d, *J* = 8.3 Hz, 4H), 6.85 (d, *J* = 8.8 Hz, 2H), 6.72 (d, *J* = 8.8 Hz, 2H), 3.80 (s, 3H), 3.79 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.5, 163.9, 159.70, 148.2, 143.0, 133.5, 132.2, 131.8, 131.5, 130.3, 130.2, 129.6, 129.2, 128.8, 128.5, 128.3, 123.5, 114.5, 113.7, 55.4, 55.4 (One of the ring quaternary carbons is not picked up). Anal. Calcd for: $C_{30}H_{22}BrClN_2O_3$: C, 62.79; H, 3.86; N, 4.88%. Found C, 62.72; H, 3.89; N, 4.85%.

(4-(4-Chlorophenyl)-2-(4-nitrophenyl)-1-p-tolyl-1H-imidazol-5-yl)(4-methoxyphenyl) methanone (**4f**)



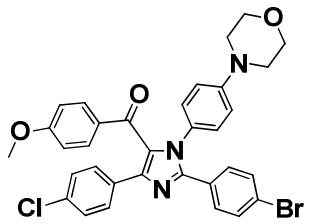
Isolated as yellow solid; mp 223-225 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.12 (d, *J* = 8.4 Hz, 2H), 7.72 (d, *J* = 8.4 Hz, 2H), 7.65 (d, *J* = 8.5 Hz, 2H), 7.50 (d, *J* = 8.1 Hz, 2H), 7.19 (d, *J* = 9.0 Hz, 4H), 7.12 (d, *J* = 7.9 Hz, 2H), 6.74 (d, *J* = 8.4 Hz, 2H), 3.79 (s, 3H), 2.37 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.3, 164.1, 147.6, 146.4, 143.1, 139.7, 135.6, 133.7, 133.6, 132.3, 131.5, 130.3, 130.0, 129.5, 129.4, 128.4, 127.3, 123.4, 113.8, 55.5, 21.2 (One of the ring quaternary carbons is not picked up). Anal. Calcd for: $C_{30}H_{22}ClN_3O_4$: C, 68.77; H, 4.23; N, 8.02%. Found C, 68.79; H, 4.26; N, 8.09%.

(4-(4-Chlorophenyl)-1-(4-fluorophenyl)-2-p-tolyl-1H-imidazol-5-yl)(4-methoxyphenyl) methanone (**4g**)



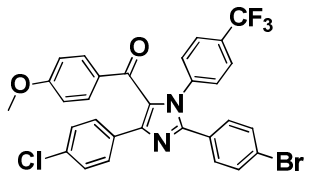
Isolated as off-white solid; mp 200-202 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.70 (d, *J* = 8.8 Hz, 2H), 7.46 (d, *J* = 8.4 Hz, 2H), 7.32 (d, *J* = 8.1 Hz, 2H), 7.21 (d, *J* = 8.8, 4.8 Hz, 2H), 7.16 (d, *J* = 8.4 Hz, 2H), 7.09 (d, *J* = 8.0 Hz, 2H), 7.03 (d, *J* = 8.4 Hz, 2H), 6.71 (d, *J* = 8.8 Hz, 2H), 3.79 (s, 3H), 2.33 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.3, 162.0, 151.6, 143.7, 139.4, 133.5, 133.1, 132.3, 131.9, 130.3, 129.9, 129.8, 129.7, 129.5, 129.1, 128.9, 128.3, 126.3, 116.2, 113.7, 55.5, 21.3. Anal. Calcd for: $C_{30}H_{22}ClFN_2O_2$: C, 72.51; H, 4.46; N, 5.64%. Found C, 72.55; H, 4.44; N, 5.68%.

(2-(4-Bromophenyl)-4-(4-chlorophenyl)-1-(4-morpholinophenyl)-1H-imidazol-5-yl)(4-methoxyphenyl)methanone (**4h**)



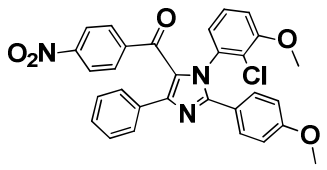
Isolated as off-white solid; mp 194-196 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$): δ 7.71 (d, *J* = 8.9 Hz, 2H), 7.46 (d, *J* = 8.6 Hz, 2H), 7.41 (d, *J* = 8.8 Hz, 2H), 7.36 (d, *J* = 8.9 Hz, 2H), 7.16 (d, *J* = 8.7 Hz, 2H), 7.10 (d, *J* = 8.9 Hz, 2H), 6.81 (d, *J* = 9.0 Hz, 2H), 6.72 (d, *J* = 8.9 Hz, 2H), 3.85–3.82 (m, 4H), 3.79 (s, 3H), 3.18–3.16 (m, 4H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.6, 164.0, 151.2, 148.2, 142.9, 133.5, 132.3, 132.0, 131.5, 130.6, 130.4, 129.8, 129.6, 128.8, 128.5, 128.3, 128.0, 123.4, 115.2, 113.8, 66.7, 55.5, 48.44. Anal. Calcd for: $C_{33}H_{27}BrClN_3O_3$: C, 63.02; H, 4.33; N, 6.68%. Found C, 63.06; H, 4.37; N, 6.61%.

(2-(4-Bromophenyl)-4-(4-chlorophenyl)-1-(4-(trifluoromethyl)phenyl)-1H-imidazol-5-yl)(4-methoxyphenyl)methanone (4i)



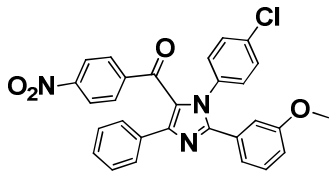
Isolated as off-white solid; mp 190-191 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.70 (d, *J* = 8.8 Hz, 2H), 7.64 (d, *J* = 8.4 Hz, 2H), 7.45 (d, *J* = 1.6 Hz, 2H), 7.43 (d, *J* = 1.7 Hz, 2H), 7.35 (d, *J* = 8.3 Hz, 2H), 7.27 (d, *J* = 8.5 Hz, 2H), 7.17 (d, *J* = 8.5 Hz, 2H), 6.73 (d, *J* = 8.9 Hz, 2H), 3.80 (s,3H).^{13}C NMR (75 MHz, $CDCl_3$) δ 185.9, 164.2, 148.4, 144.1, 139.9, 133.9, 132.3, 131.7, 131.6, 130.5, 130.1, 129.9, 129.4, 128.4, 128.3, 128.9, 126.6, 124.1, 113.9, 55.5 (Two of the ring quaternary carbons are not picked up). Anal. Calcd for: $C_{30}H_{19}BrClF_3N_2O_2$: C, 58.89; H, 3.13; N, 4.58%. Found: C, 58.85; H, 3.11; N, 4.55%.

(1-(2-chloro-3-methoxyphenyl)-2-(4-methoxyphenyl)-4-phenyl-1H-imidazol-5-yl)(4-nitrophenyl)methanone (4j)



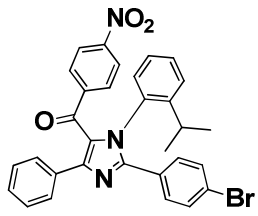
Isolated as yellow solid; mp 119-121 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.98 (d, *J* = 8.9 Hz, 2H), 7.67 (d, *J* = 7.1 Hz, 2H), 7.61 (d, *J* = 8.9 Hz, 2H), 7.44 (d, *J* = 8.9 Hz, 3H), 7.37 (d, *J* = 7.5 Hz, 1H), 7.23 – 7.18 (m, 3H), 7.08 (t, *J* = 8.0 Hz, 1H), 6.83 (d, *J* = 8.9 Hz, 2H), 3.79 (s, 3H), 3.65 (s, 3H).^{13}C NMR (75 MHz, $CDCl_3$): δ 186.8, 160.8, 152.0, 150.61, 147.0, 144.1, 140.0, 137.3, 133.4, 132.0, 131.9, 130.2, 129.9, 129.8, 129.7, 128.8, 128.4, 128.1, 124.4, 123.1, 121.5, 114.1, 60.8, 55.3. Anal. Calcd for: $C_{30}H_{22}ClN_3O_5$: C, 66.73; H, 4.11; N, 7.78%. Found C, 66.78; H, 4.16; N, 7.70%.

(1-(4-Chlorophenyl)-2-(3-methoxyphenyl)-4-phenyl-1H-imidazol-5-yl)(4-nitrophenyl) methanone (4k)



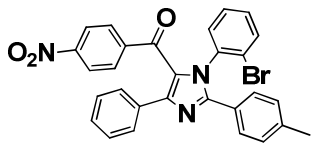
Isolated as pale yellow solid; mp 180-181 ^{o}C ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.03 (d, *J* = 8.8 Hz, 2H), 7.71 – 7.67 (m, 4H), 7.42 (d, *J* = 7.5 Hz, 1H), 7.34 (d, *J* = 8.6 Hz, 2H), 7.27 – 7.18 (m, 5H), 7.04 (s, 1H), 6.98 (d, *J* = 7.6 Hz, 1H), 6.92 (d, *J* = 8.3 Hz, 1H), 3.71 (s, 3H). ^{13}C NMR (75 MHz, $CDCl_3$): δ 187.5, 159.5, 149.9, 147.1, 143.0, 139.6, 137.3, 135.3, 135.1, 133.9, 130.3, 129.9, 129.7, 129.6, 129.1, 128.6, 123.3, 121.4, 116.0, 114.3, 55.2 (Two of the ring quaternary carbons are not picked up). Anal. Calcd for: $C_{29}H_{20}ClN_3O_4$: C, 68.30; H, 3.95; N, 8.24%. Found C, 68.36; H, 3.92; N, 8.28%.

(2-(4-Bromophenyl)-1-(2-isopropylphenyl)-4-phenyl-1H-imidazol-5-yl)(4-nitrophenyl) methanone (4l)



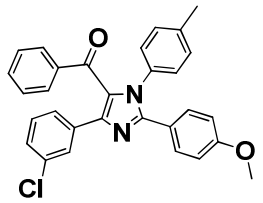
Isolated as brown solid; mp 166-167 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.01 (d, *J* = 8.3 Hz, 2H), 7.69 (t, *J* = 9.4 Hz, 4H), 7.48 – 7.30 (m, 8H), 7.27 – 7.20 (m, 3H), 2.54 – 2.45 (m, 1H), 1.01 (d, *J* = 6.7 Hz, 3H), 0.72 (d, *J* = 6.7 Hz, 3H). ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.7, 149.1, 146.8, 145.8, 143.0, 139.6, 137.0, 133.7, 133.7, 131.5, 130.6, 130.4, 130.0, 129.7, 129.2, 128.6, 128.5, 128.0, 127.3, 126.5, 124.1, 123.2, 27.9, 23.8, 22.8. Anal. Calcd for: $C_{31}H_{24}BrN_3O_3$: C, 65.73; H, 4.27; N, 7.42%. Found C, 65.77; H, 4.29; N, 7.39%.

(1-(2-Bromophenyl)-4-phenyl-2-p-tolyl-1H-imidazol-5-yl)(4-nitrophenyl)methanone (4m)



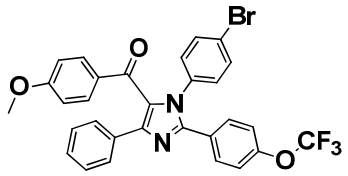
Isolated as off-white solid; mp 221-223 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$): ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.98 (d, *J* = 8.5 Hz, 2H), 7.71 (d, *J* = 7.8 Hz, 2H), 7.63 – 7.59 (m, 3H), 7.53 (d, *J* = 7.8 Hz, 1H), 7.45 – 7.29 (m, 5H), 7.18 (t, *J* = 7.7 Hz, 2H), 7.10 (d, *J* = 7.9 Hz, 2H), 2.32 (s, 3H). ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.5, 150.7, 146.9, 144.4, 140.0, 139.8, 136.9, 136.3, 133.3, 133.3, 130.9, 130.8, 129.9, 129.8, 129.1, 128.6, 128.2, 125.9, 123.0, 122.6, 21.3 (Two of the ring quaternary carbons are not picked up). Anal. Calcd for: $C_{29}H_{20}BrN_3O_3$: C, 64.69; H, 3.74; N, 7.80%. Found C, 64.64; H, 3.78; N, 7.77%.

(4-(3-Chlorophenyl)-2-(4-methoxyphenyl)-1-p-tolyl-1H-imidazol-5-yl)(phenyl)methanone (4n)



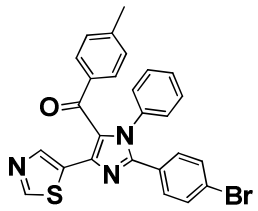
Isolated as off-white solid; mp 233-235 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.67 (d, *J* = 6.7 Hz, 2H), 7.56 (s, 1H), 7.42 – 7.34 (m, 3H), 7.25 (d, *J* = 2.8 Hz, 2H), 7.21 (d, *J* = 6.4 Hz, 2H), 7.11 – 7.00 (m, 5H), 6.79 (d, *J* = 8.8 Hz, 2H), 3.79 (s, 3H), 2.30 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 187.4, 160.3, 149.8, 145.1, 139.3, 137.8, 137.0, 135.3, 133.9, 133.0, 130.5, 129.7, 129.2, 129.1, 129.0, 128.9, 128.3, 128.1, 127.7, 127.1, 124.9, 113.7, 55.2, 21.3. Anal. Calcd for: $C_{30}H_{23}ClN_2O_2$: C, 75.23; H, 4.84; N, 5.85;%. Found C, 75.27; H, 4.81; N, 5.87%.

(1-(4-Bromophenyl)-4-phenyl-2-(4-(trifluoromethoxy)phenyl)-1H-imidazol-5-yl)(4-methoxyphenyl)methanone (**4o**)



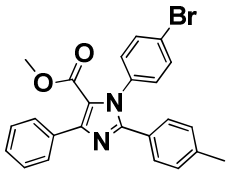
Isolated as off-white solid; mp 214-215 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$): δ 7.74 (d, *J* = 8.9 Hz, 2H), 7.54 – 7.51 (m, 5H), 7.21 – 7.13 (m, 8H), 6.71 (d, *J* = 8.9 Hz, 2H), 3.77 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$): δ 186.1, 163.8, 149.7, 147.8, 145.2, 135.8, 133.0, 132.6, 132.3, 130.5, 130.1, 129.2, 129.1, 128.6, 128.1, 127.9, 127.8, 123.1, 120.6, 120.0, 113.6, 55.3. Anal. Calcd for: $C_{30}H_{20}BrF_3N_2O_3$: C, 60.72; H, 3.40; N, 4.72%. Found C, 60.70; H, 3.37; N, 4.71%.

(2-(4-Bromophenyl)-1-phenyl-4-(thiazol-5-yl)-1H-imidazol-5-yl)(p-tolyl)methanone (**4p**)



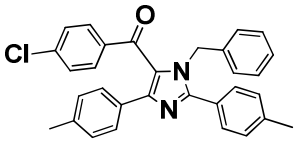
Isolated as brown solid; mp 248-250 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.66 (s, 1H), 7.73 (s, 1H), 7.68 (d, *J* = 8.1 Hz, 2H), 7.42 – 7.27 (m, 7H), 7.21 (d, *J* = 1.6 Hz, 2H), 7.12 (d, *J* = 7.9 Hz, 2H), 2.34 (s, 3H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 186.7, 152.9, 148.4, 145.0, 142.2, 136.3, 136.0, 134.7, 131.5, 130.4, 129.9, 129.6, 129.5, 129.4, 129.2, 127.9, 127.6, 123.9, 21.7. (One of the ring quaternary carbons is not picked up). Anal. Calcd for: $C_{26}H_{18}BrN_3OS$: C, 62.40; H, 3.63; N, 8.40%. Found C, 62.39; H, 3.61; N, 8.44%.

Methyl 1-(4-bromophenyl)-4-phenyl-2-p-tolyl-1H-imidazole-5-carboxylate (4q)



Isolated as off-white solid; mp 142-144 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.79 (d, *J* = 8.1 Hz, 2H), 7.56 (d, *J* = 8.6 Hz, 2H), 7.47 – 7.38 (m, 3H), 7.28 (d, *J* = 8.2 Hz, 2H), 7.18 (d, *J* = 8.6 Hz, 2H), 7.07 (d, *J* = 7.9 Hz, 2H), 3.59 (s, 3H), 2.31 (s, 3H). ^{13}C NMR (75 MHz, $CDCl_3$): δ 160.8, 150.4, 148.6, 139.5, 136.9, 134.1, 132.2, 129.8, 129.4, 129.1, 129.0, 128.3, 127.8, 126.4, 122.9, 121.2, 51.3, 21.2. Anal. Calcd for: $C_{24}H_{19}BrN_2O_2$: C, 64.44; H, 4.28; N, 6.26%. Found C, 64.47; H, 4.26; N, 6.29%.

(1-Benzyl-2,4-di-p-tolyl-1H-imidazol-5-yl)(4-chlorophenyl)methanone (4r)



Isolated as white solid; mp 185-187 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.57 (d, *J* = 8.1 Hz, 2H), 7.44 (d, *J* = 8.6 Hz, 2H), 7.27 (d, *J* = 7.6 Hz, 2H), 7.24 – 7.10 (m, 5H), 7.04 (d, *J* = 8.6 Hz, 2H), 6.95 (d, *J* = 6.6 Hz, 2H), 6.89 (d, *J* = 7.9 Hz, 2H), 5.59 (s, 2H), 2.41 (s, 3H), 2.23 (s, 3H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 187.1, 152.4, 148.9, 139.9, 138.6, 137.7, 137.4, 136.2, 131.1, 130.6, 129.4, 129.4,* 129.2, 128.6, 128.5, 128.1, 127.5, 126.8, 126.3, 49.2, 21.4, 21.1. Anal. Calcd for: $C_{31}H_{25}ClN_2O$ C, 78.06; H, 5.28; N, 5.87%. Found C, 78.09; H, 5.25; N, 5.84%. (*Two carbons merged here)

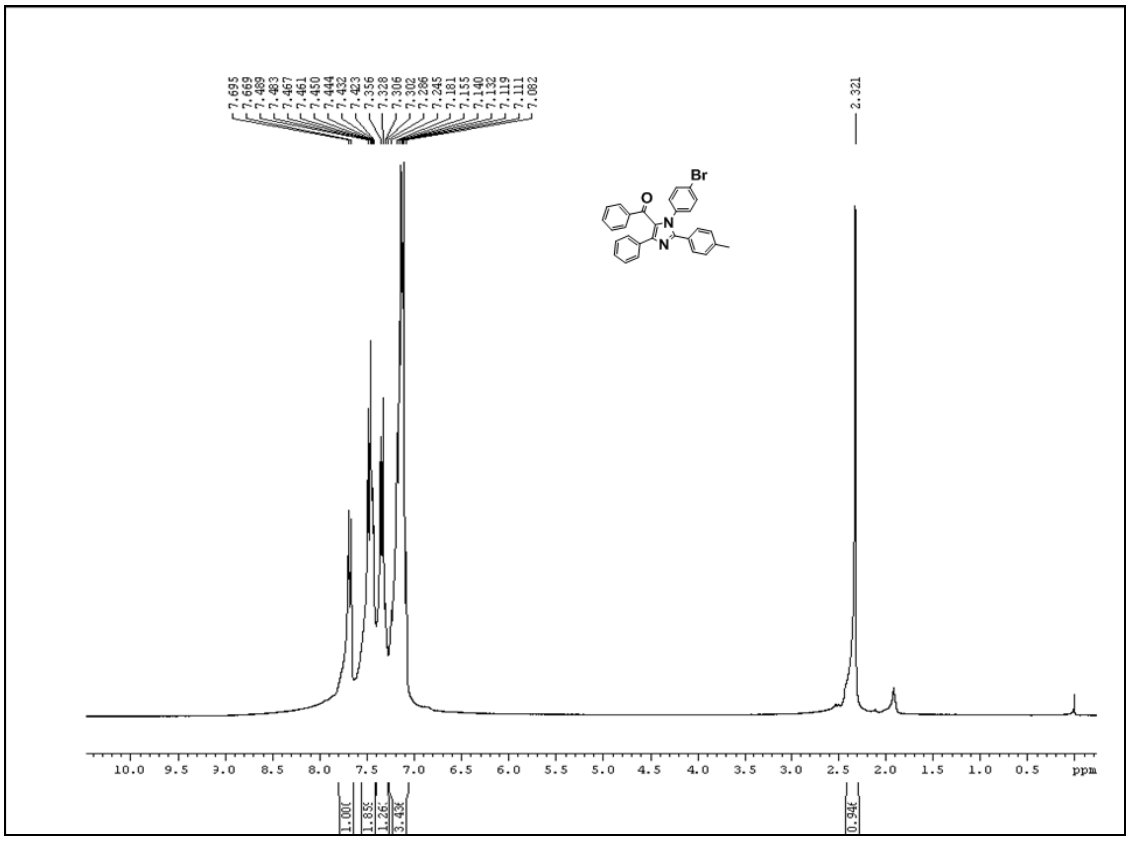


Fig 1. ^{1}H-NMR spectrum of **4a**

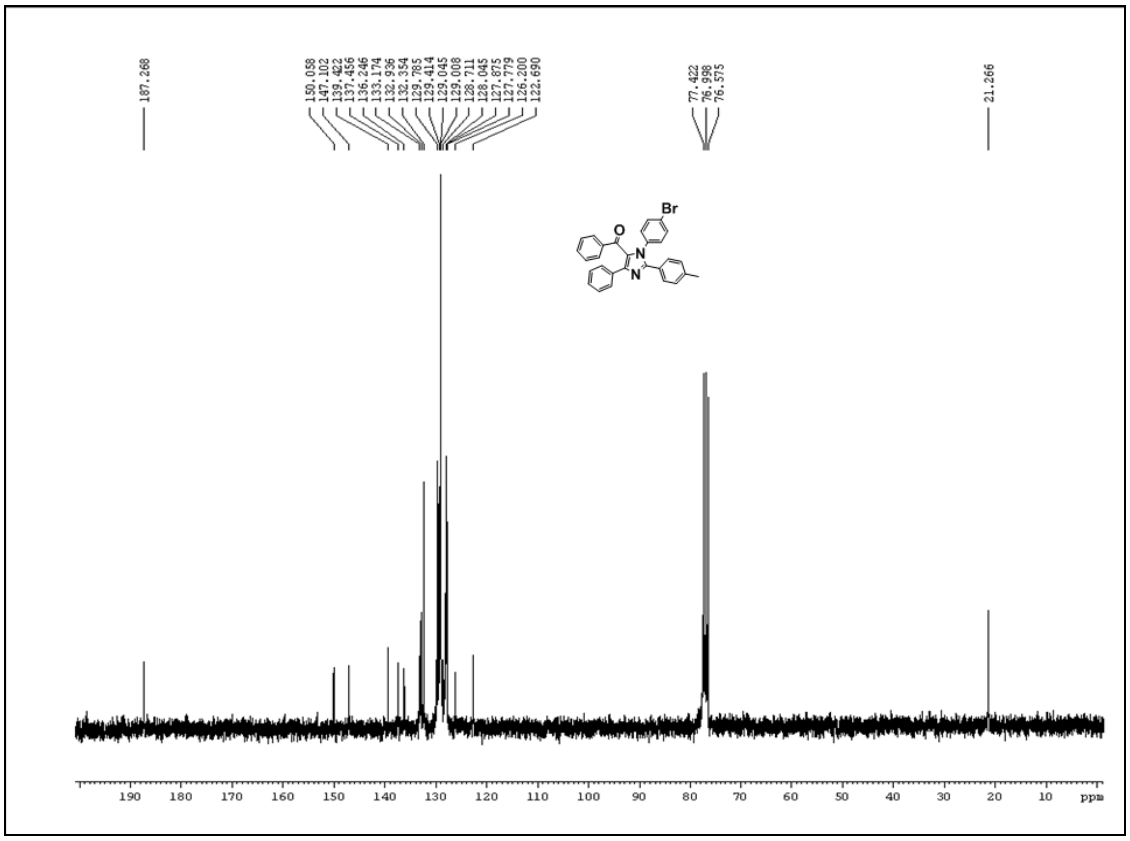


Fig 2. ^{13}C-NMR spectrum of **4a**

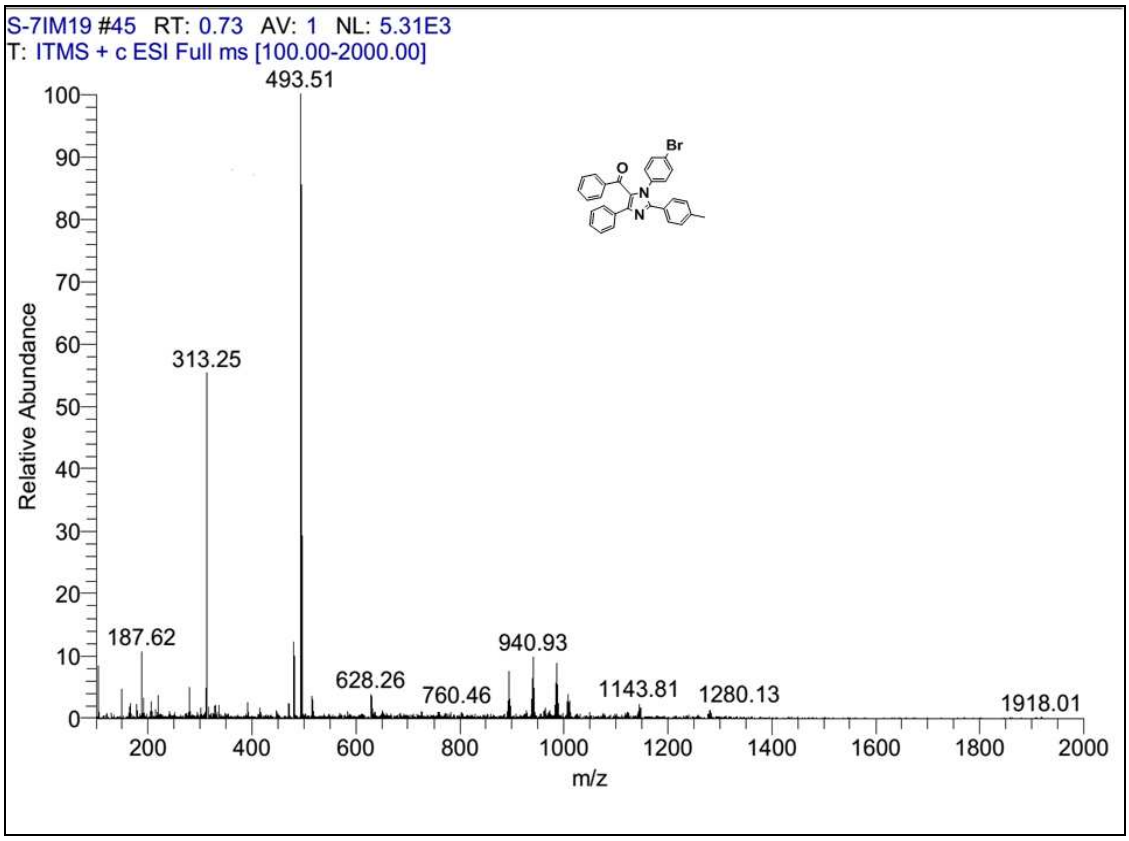


Fig 3. Mass spectrum of **4a**

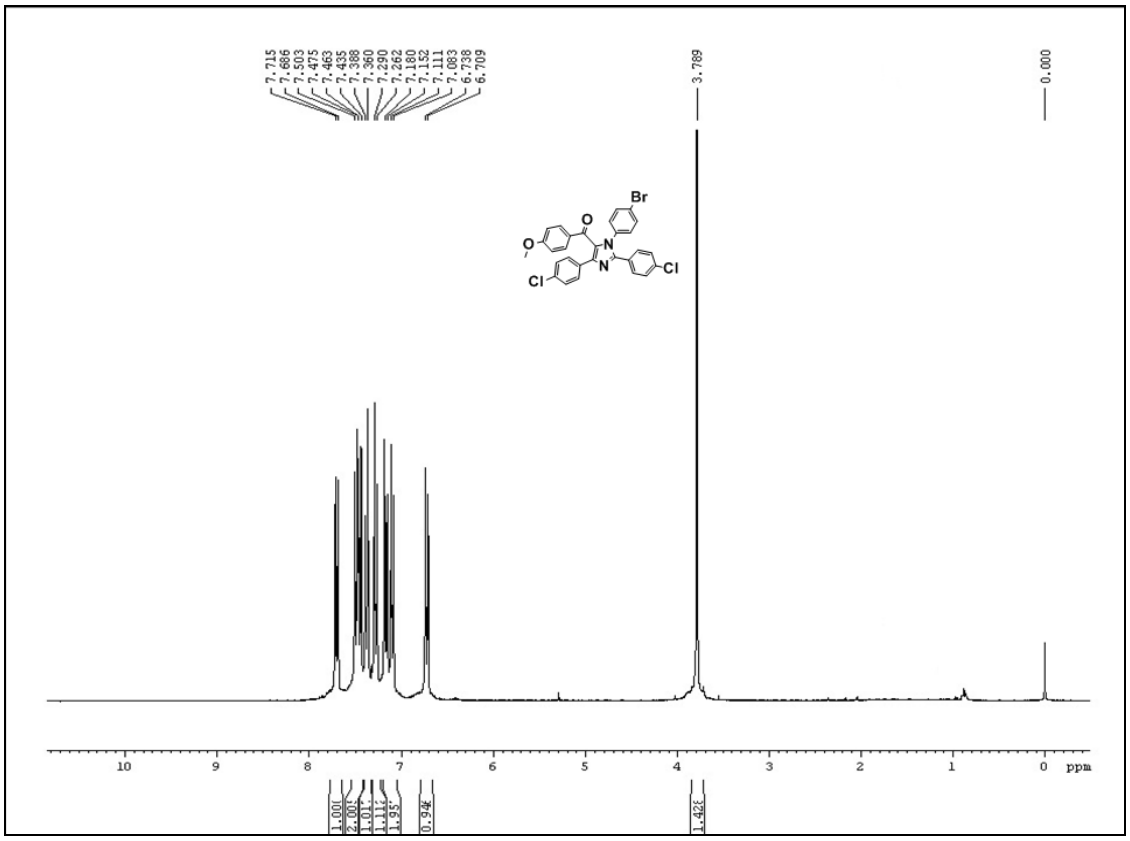


Fig 4. ^{1}H-NMR spectrum of **4b**

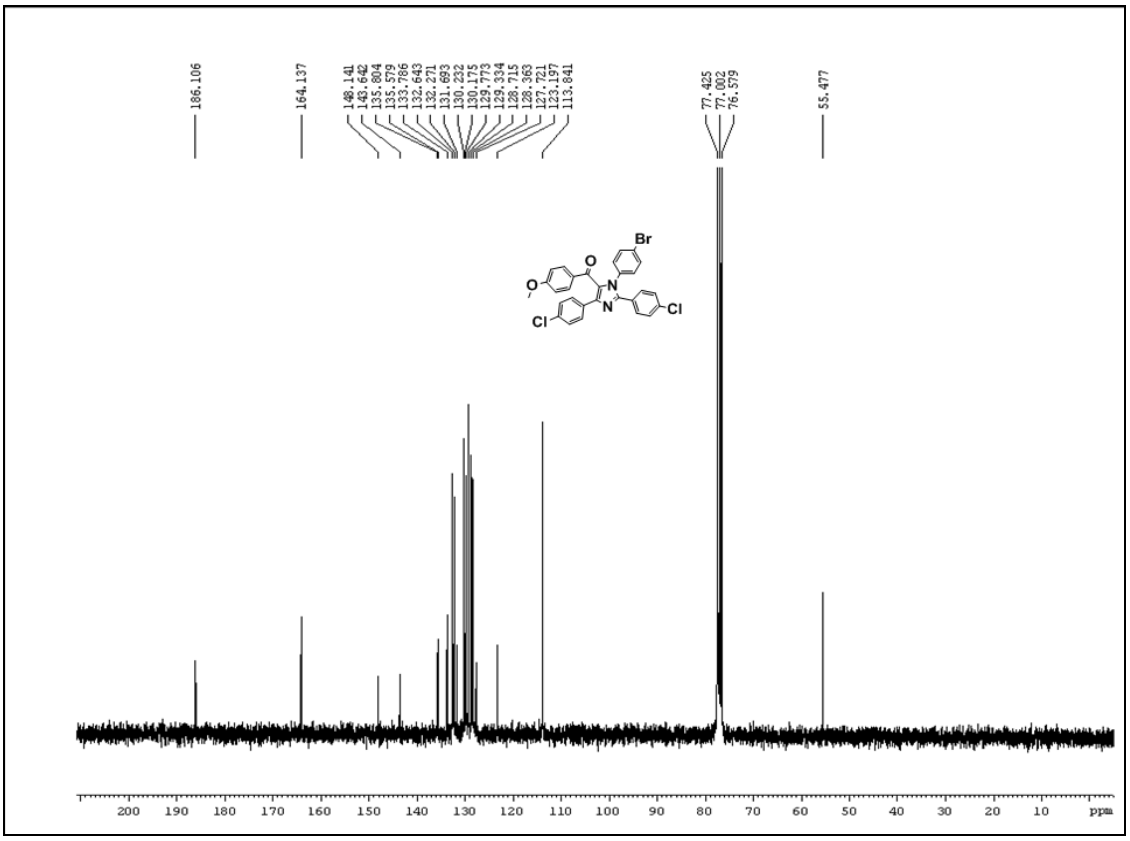


Fig 5. ^{13}C-NMR spectrum **4b**

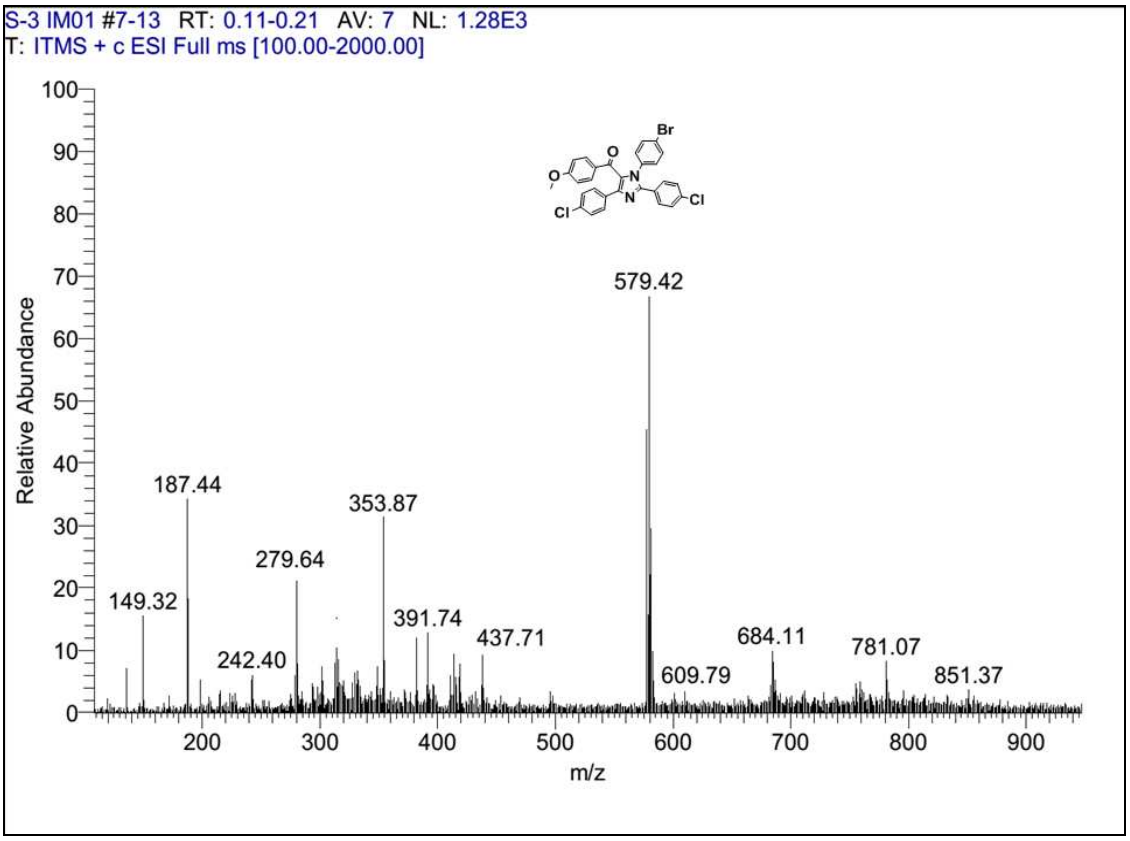


Fig 6. Mass spectrum **4b**

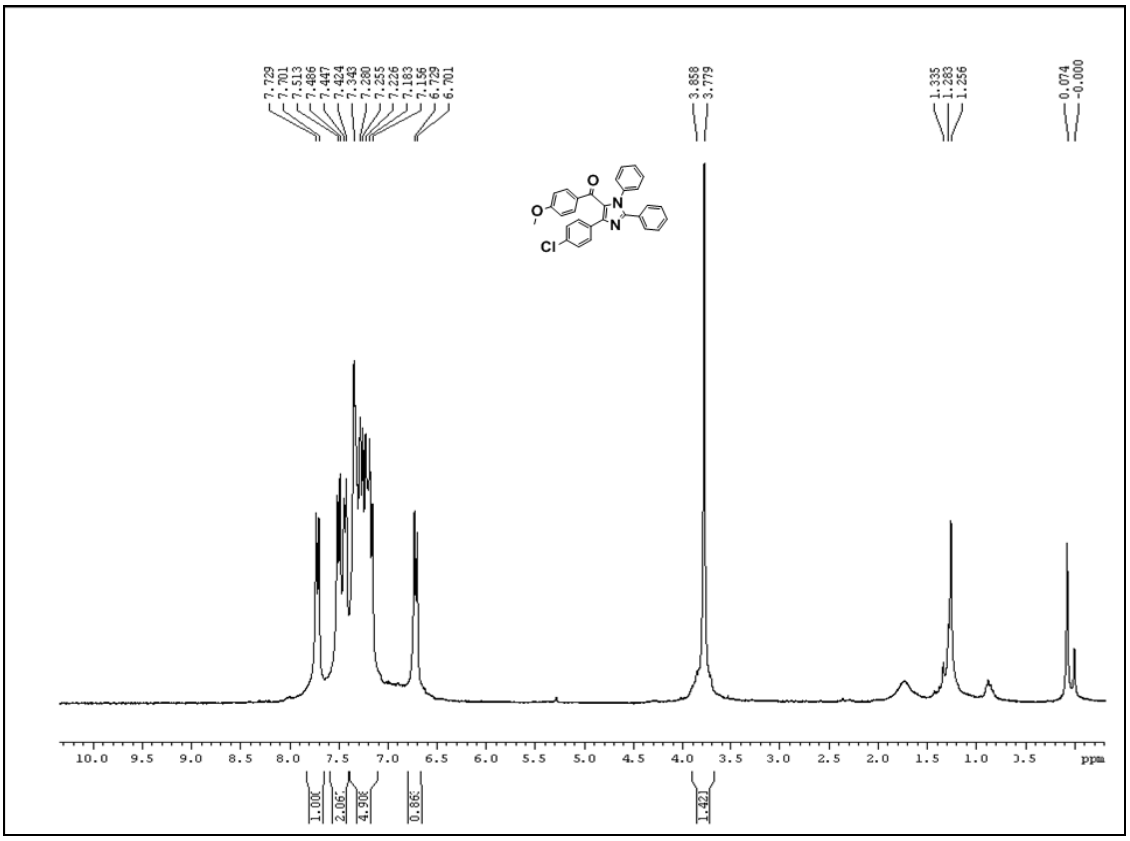


Fig 7. ^{1}H-NMR spectrum of **4c**

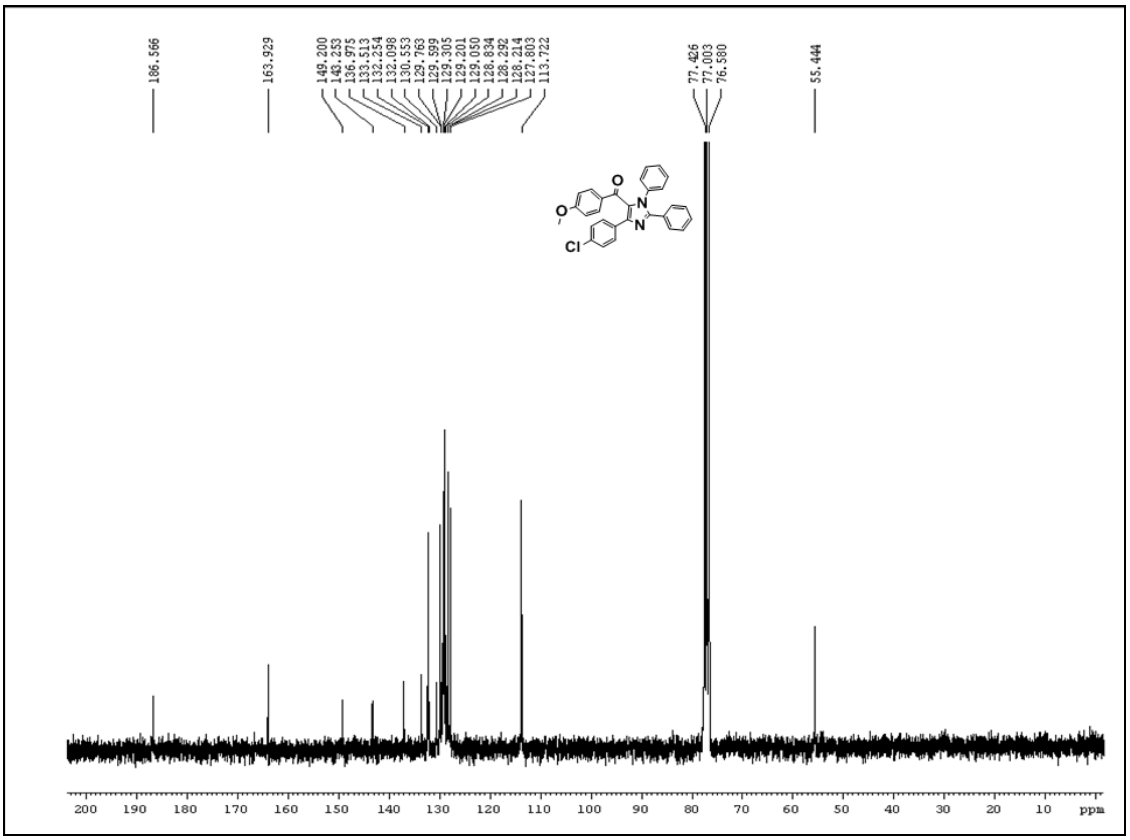


Fig 8. ^{13}C-NMR spectrum of **4c**

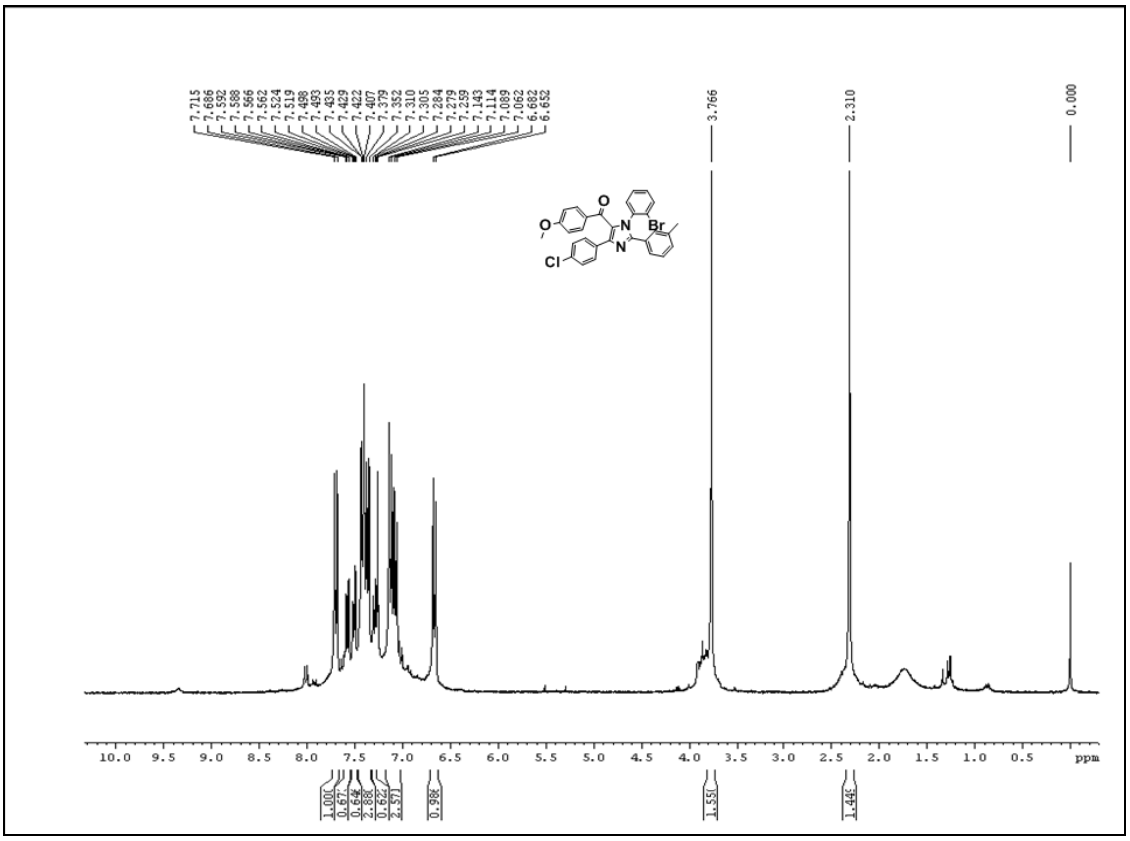


Fig 9. ^{1}H-NMR spectrum of **4d**

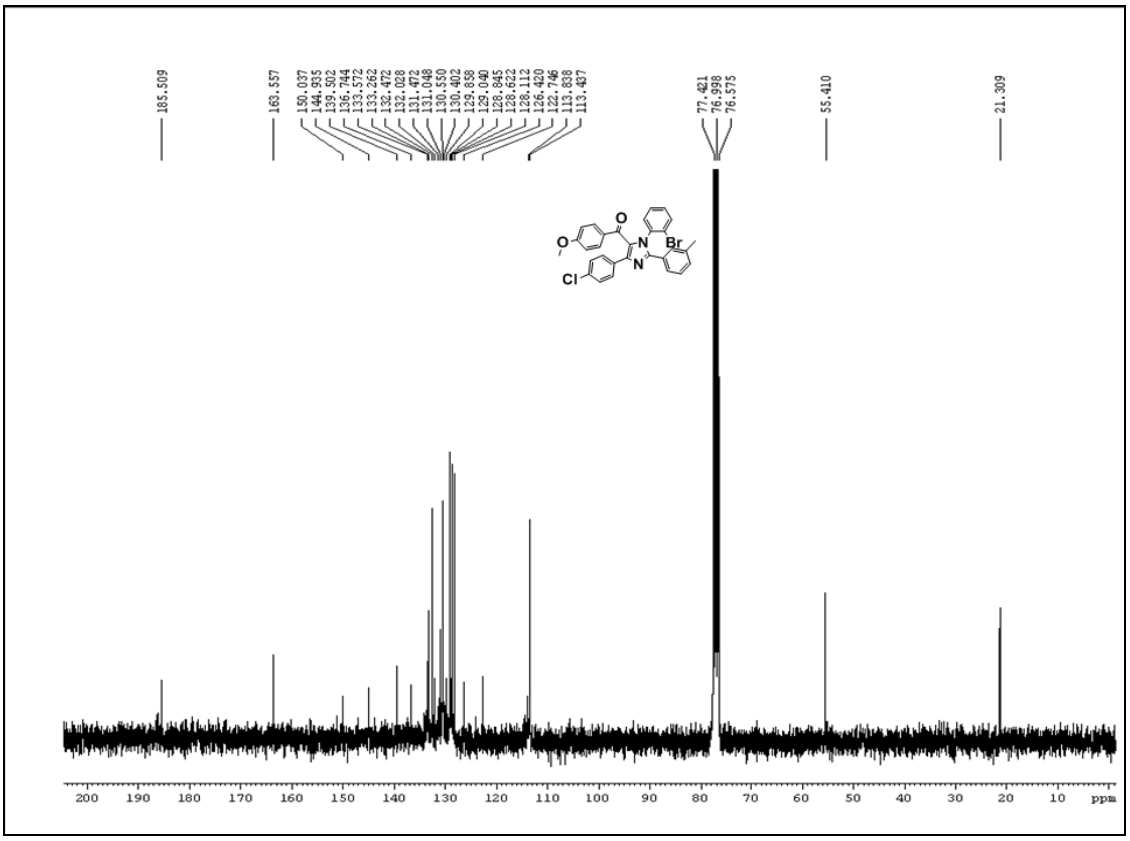


Fig 10. ^{13}C-NMR spectrum of **4d**

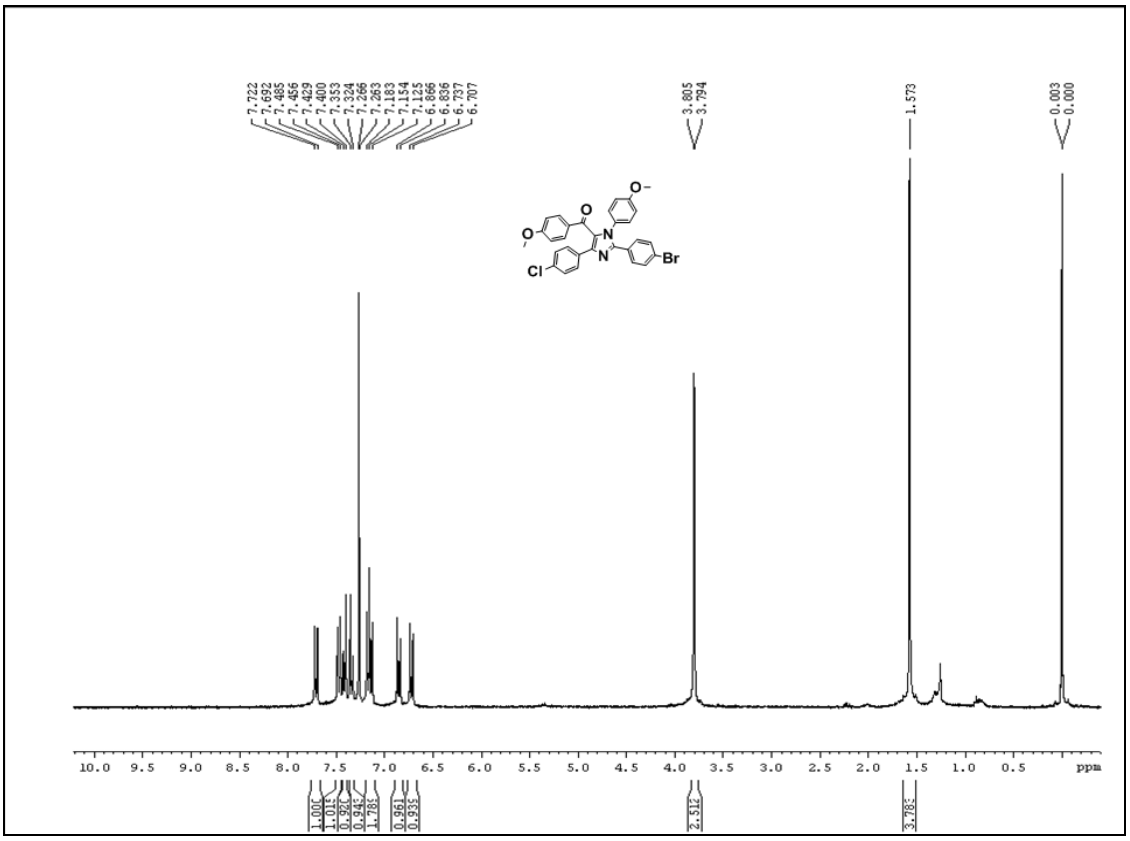


Fig 11. ^{1}H-NMR spectrum of **4e**

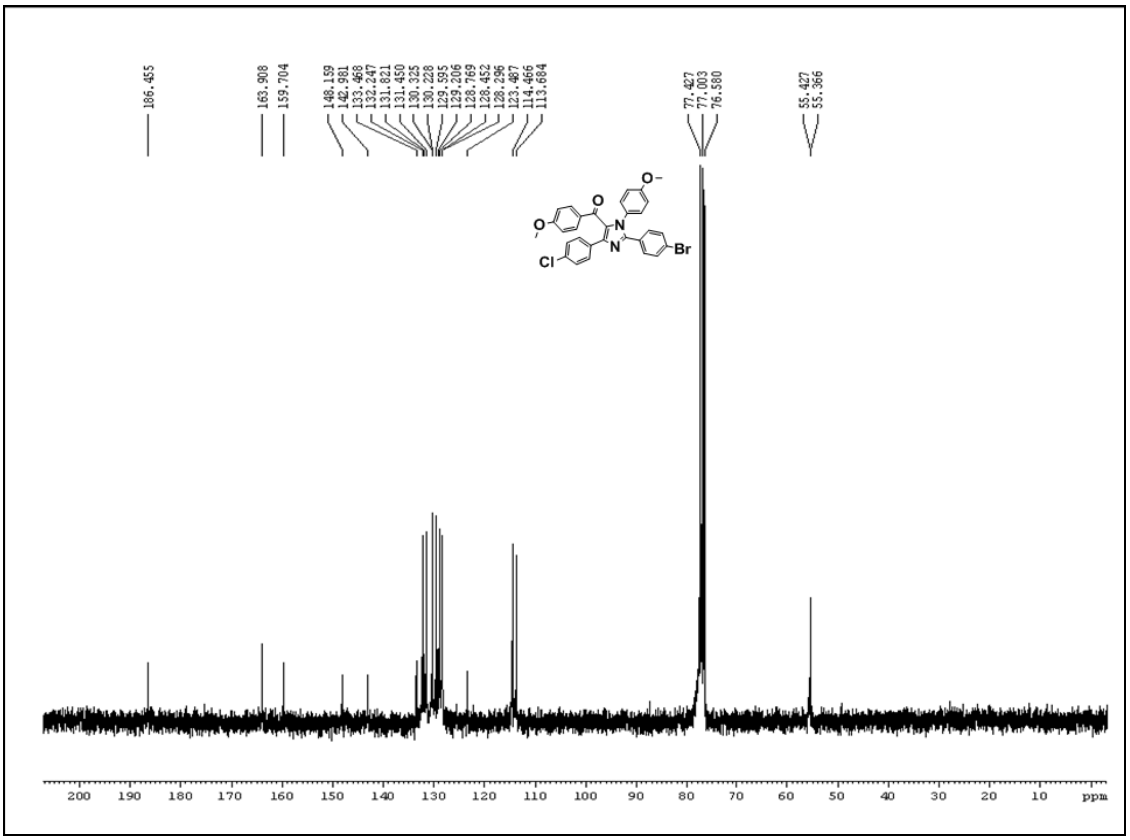


Fig 12. ^{13}C-NMR spectrum of **4e**

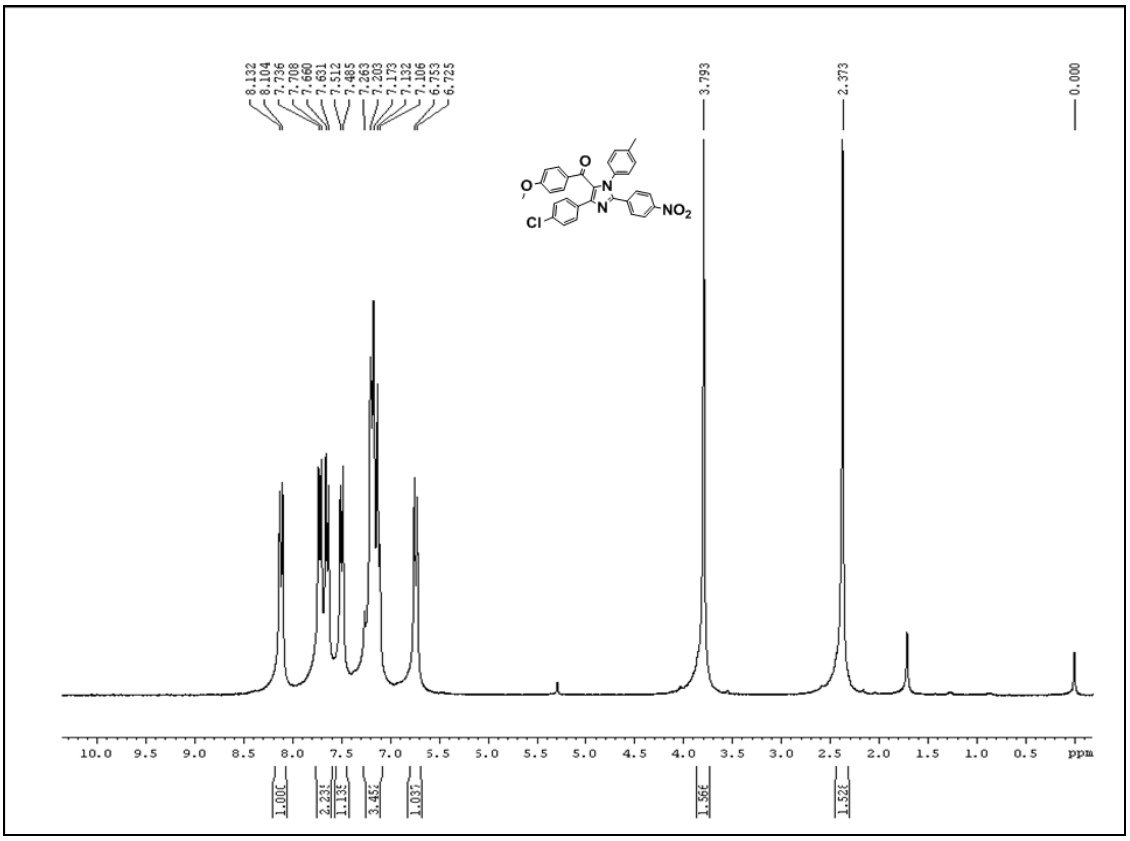


Fig 13. ^{1}H-NMR spectrum of **4f**

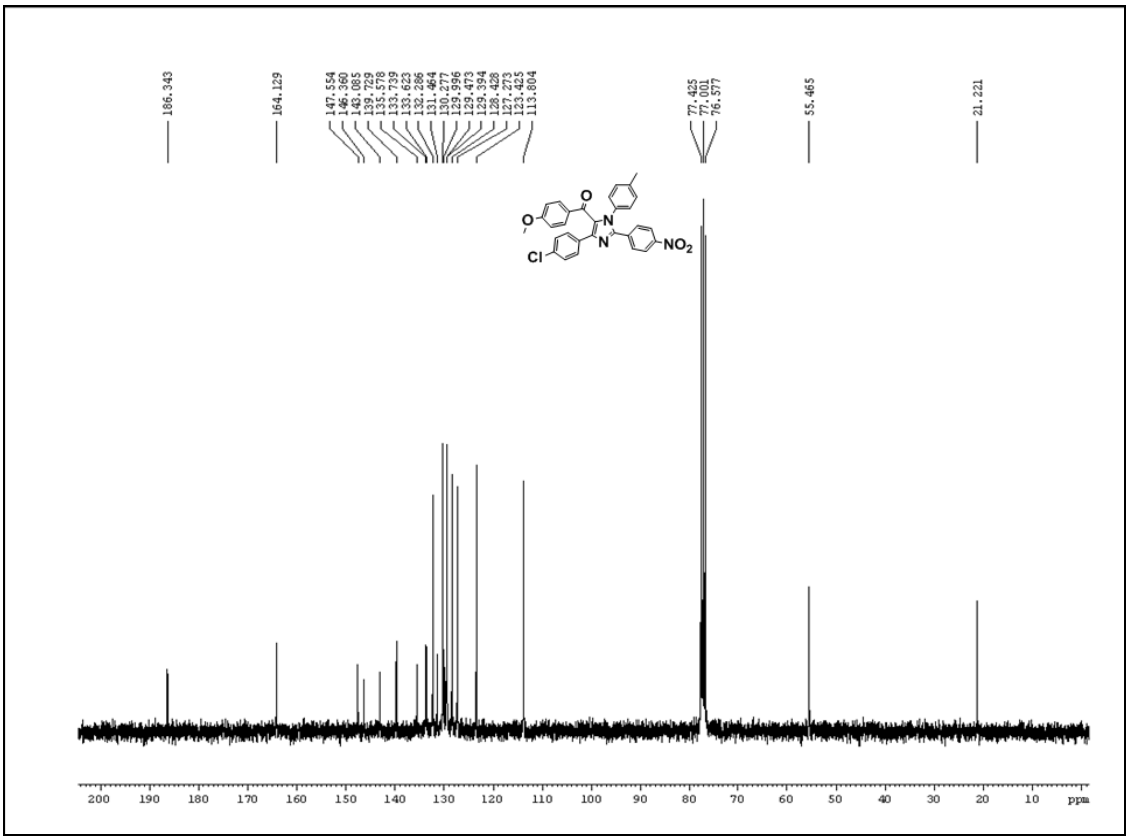


Fig 14. ^{13}C-NMR spectrum of **4f**

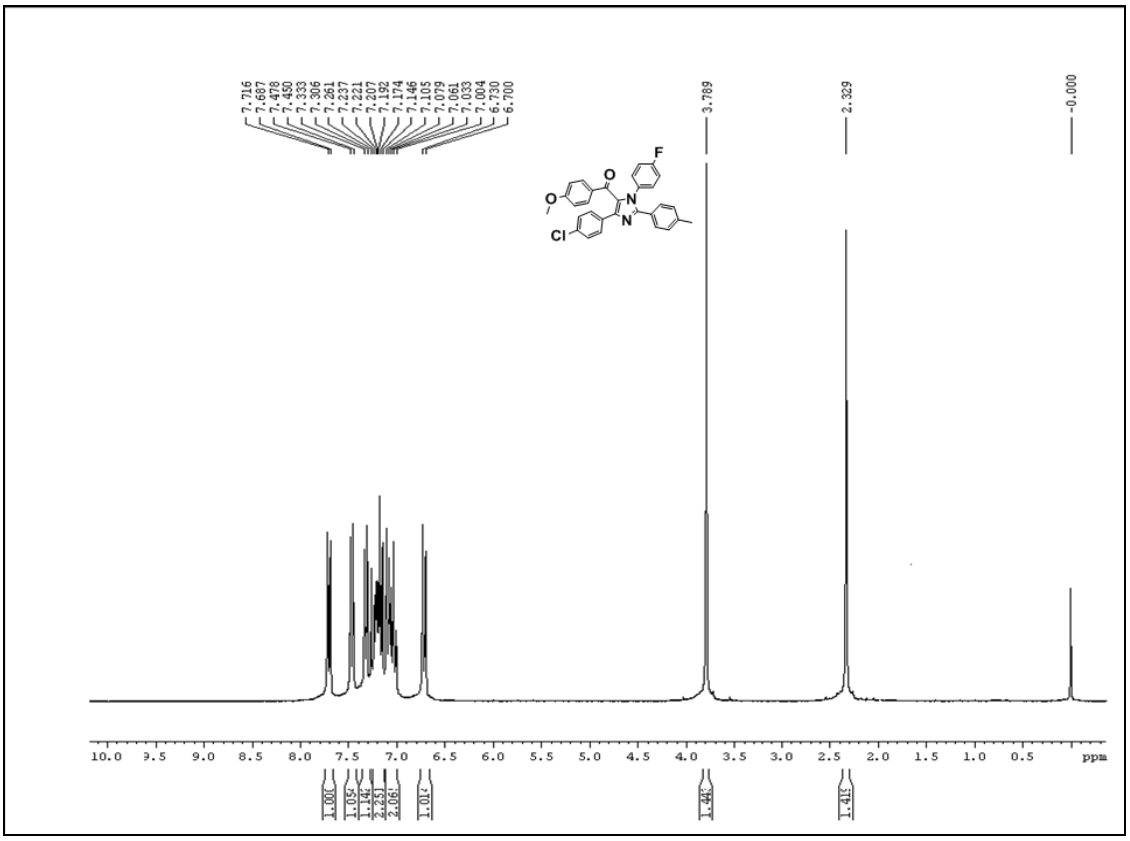


Fig 15. ^{1}H-NMR spectrum of **4g**

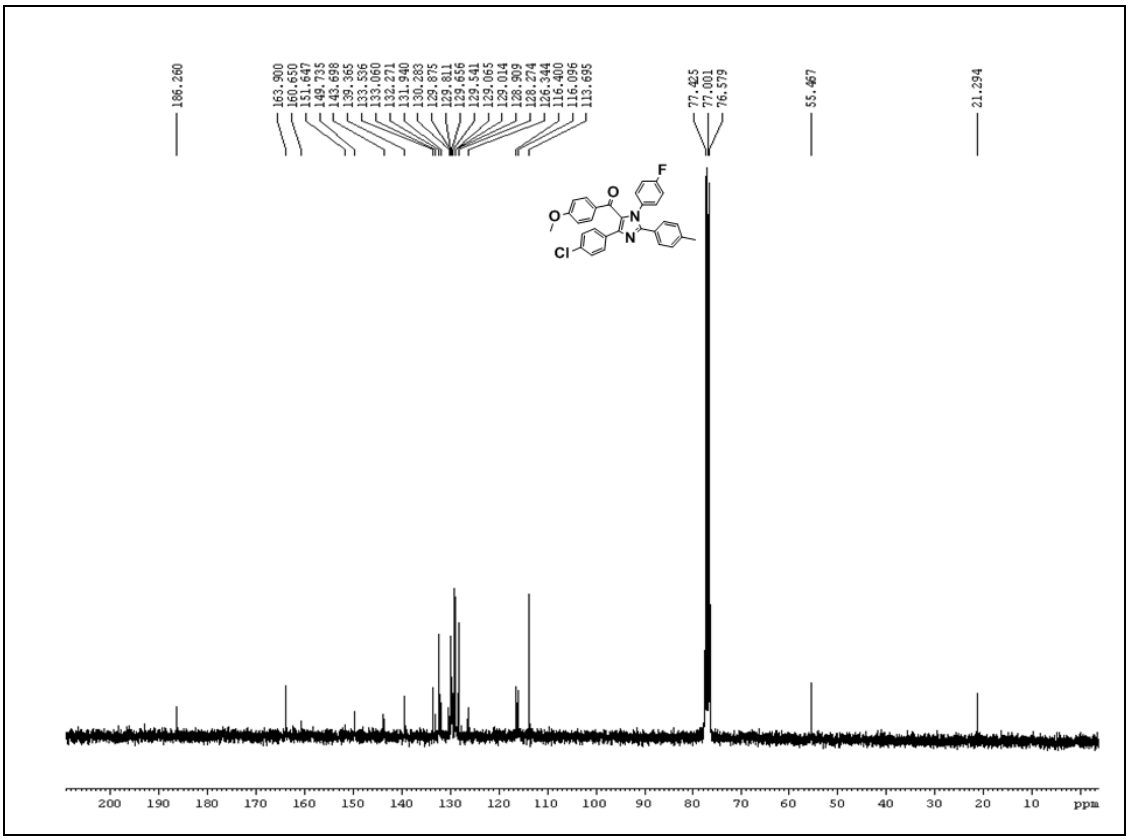


Fig 16. ^{13}C-NMR spectrum of **4g**

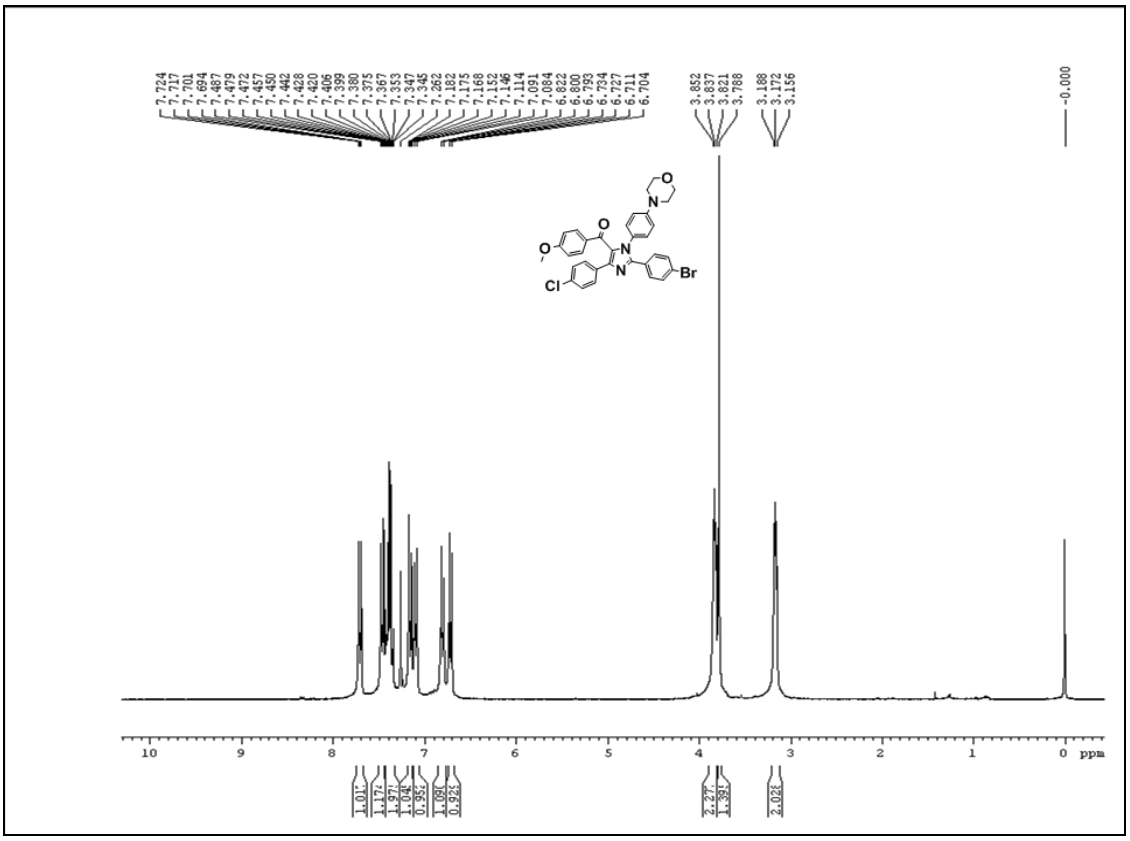


Fig 17. ^{1}H-NMR spectrum of **4h**

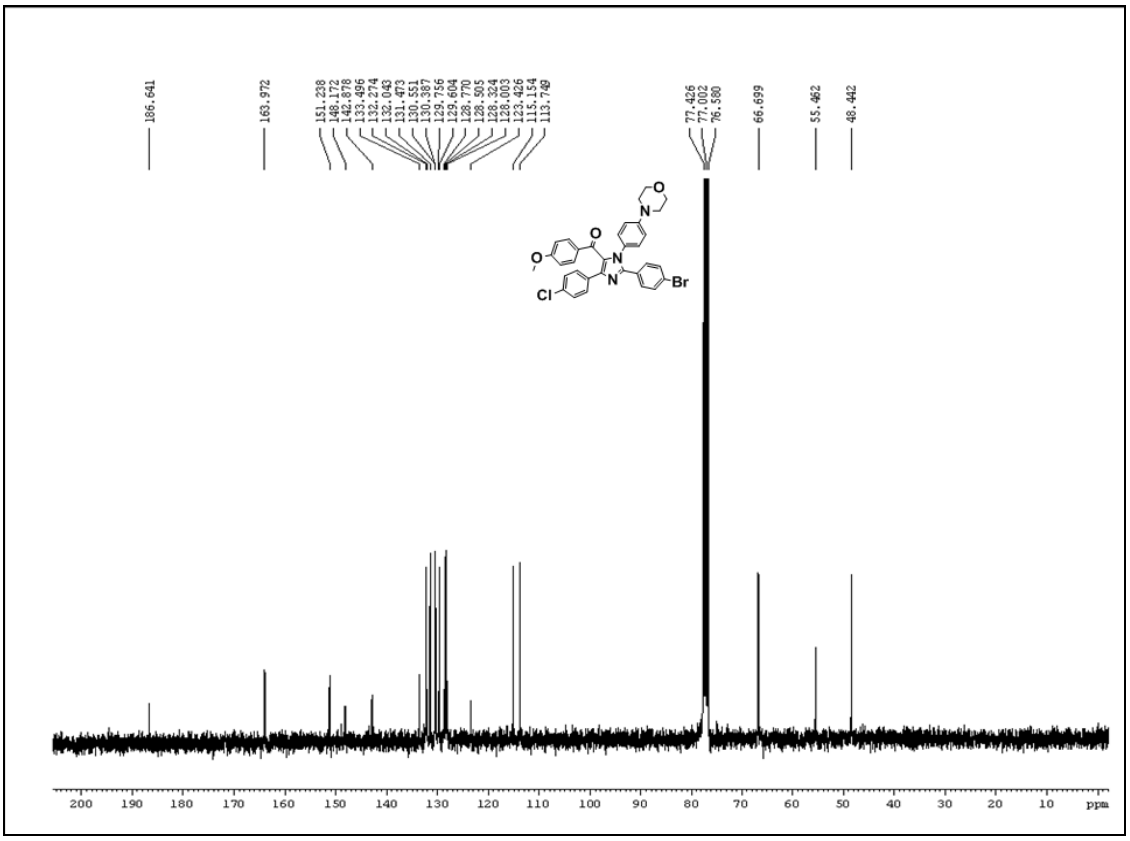


Fig 18. ^{13}C-NMR spectrum of **4h**

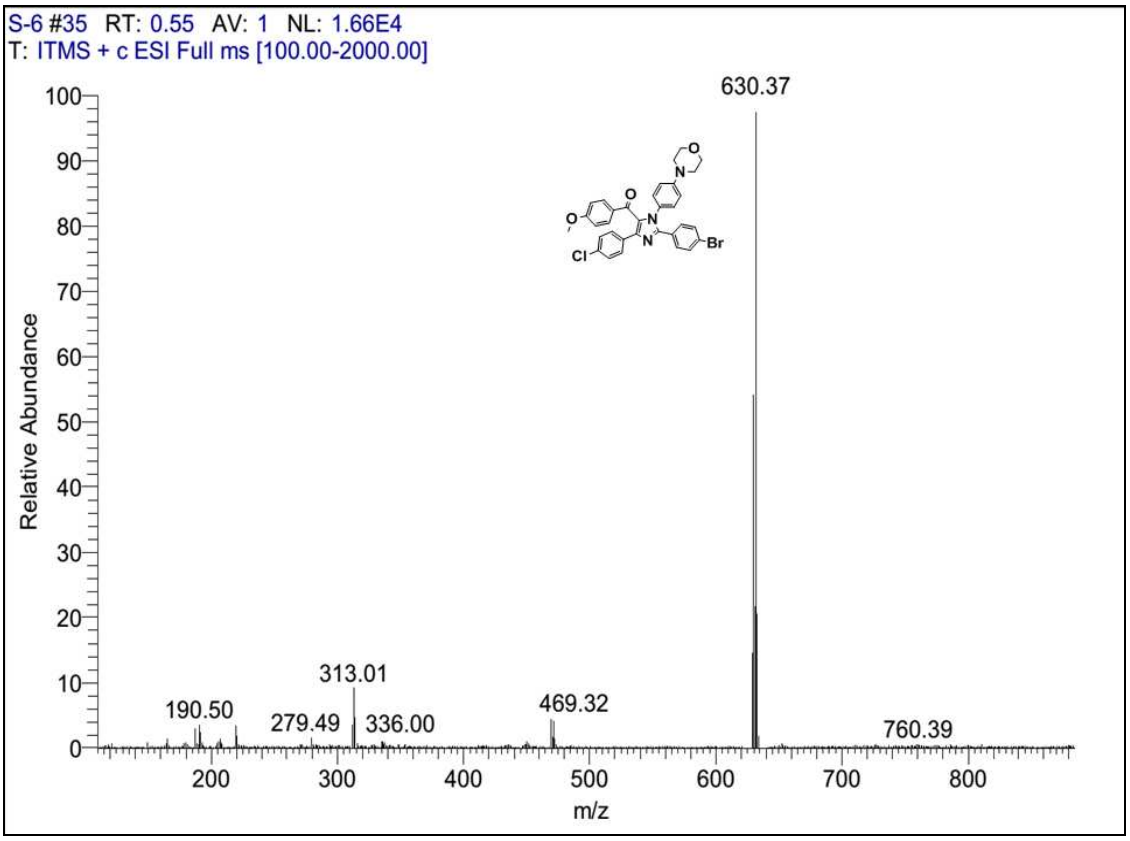


Fig 19. Mass spectrum of **4h**

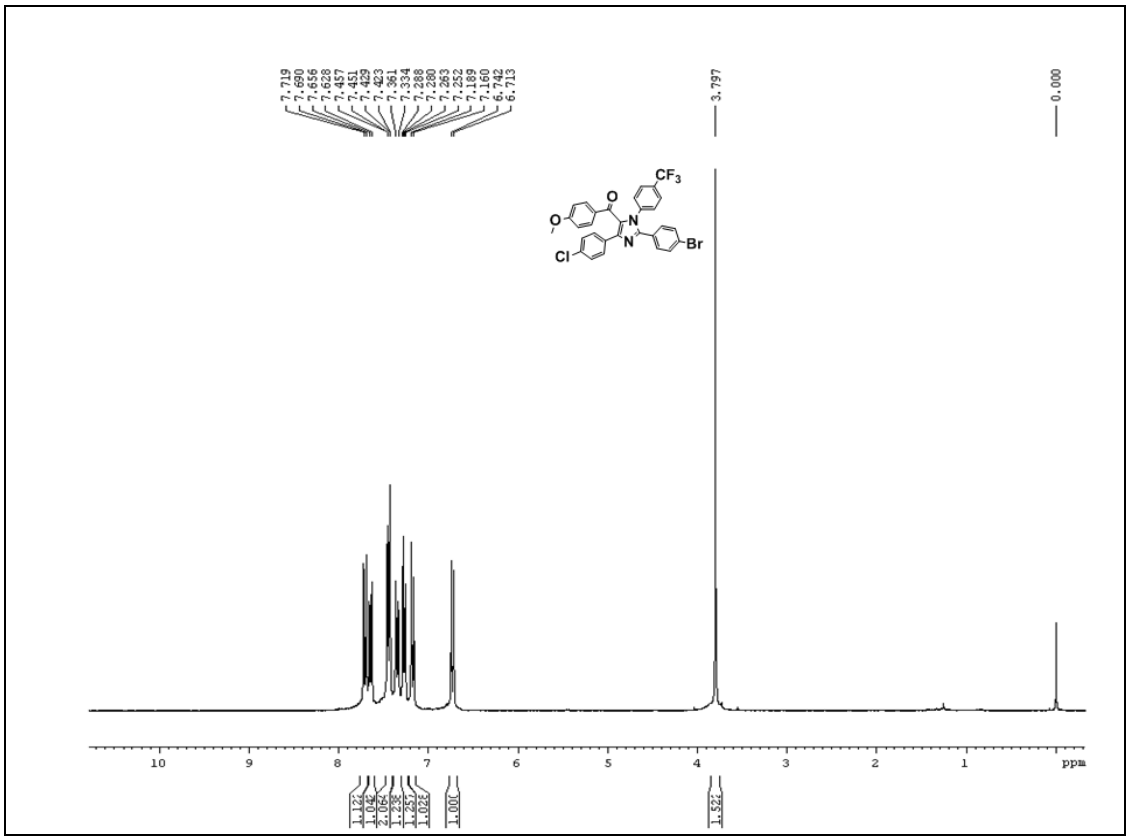


Fig 20. ^{1}H-NMR spectrum of **4i**

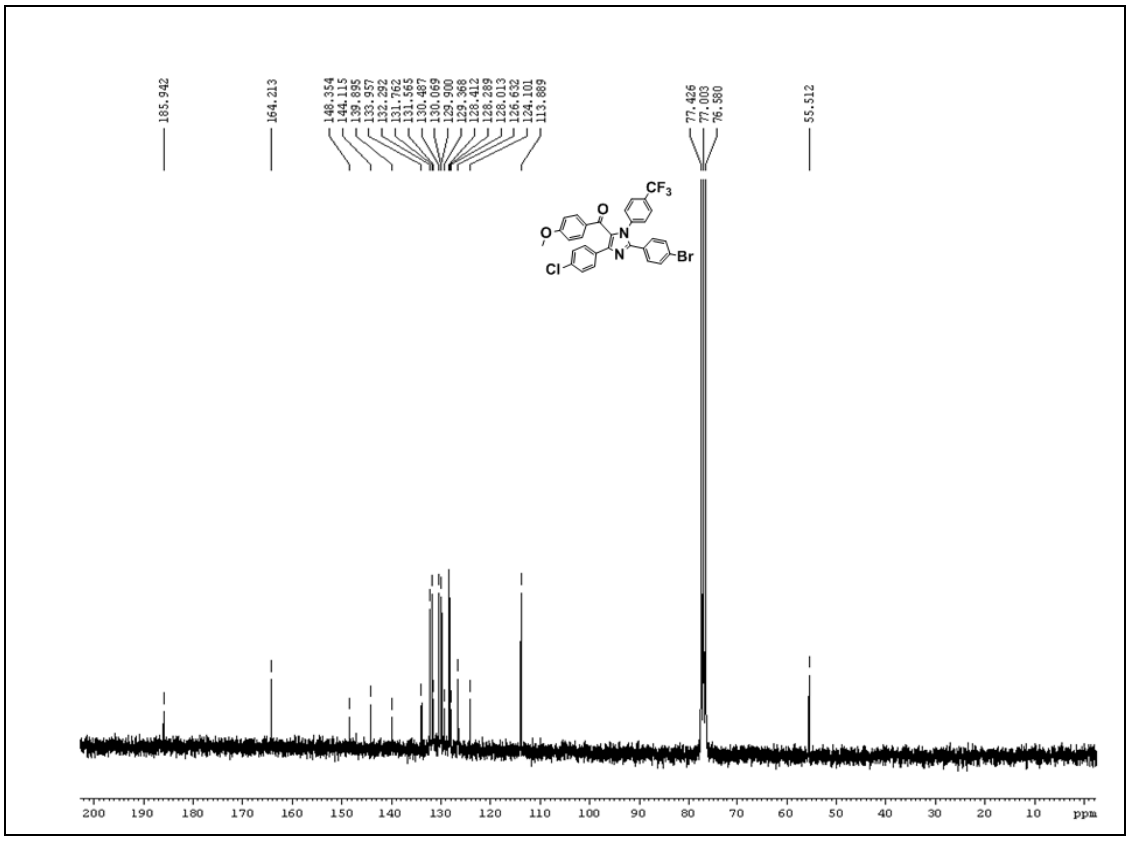


Fig 21. ^{13}C-NMR spectrum of **4i**

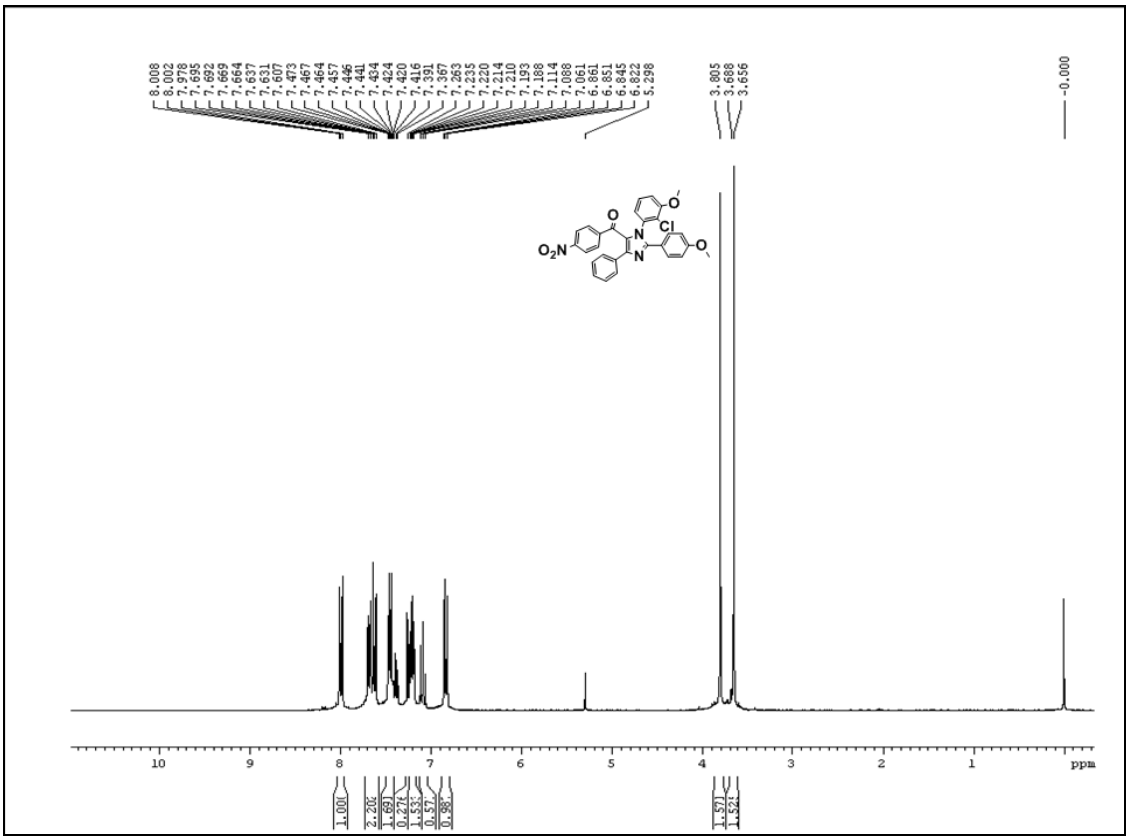


Fig 22. ^{1}H-NMR spectrum of **4j**

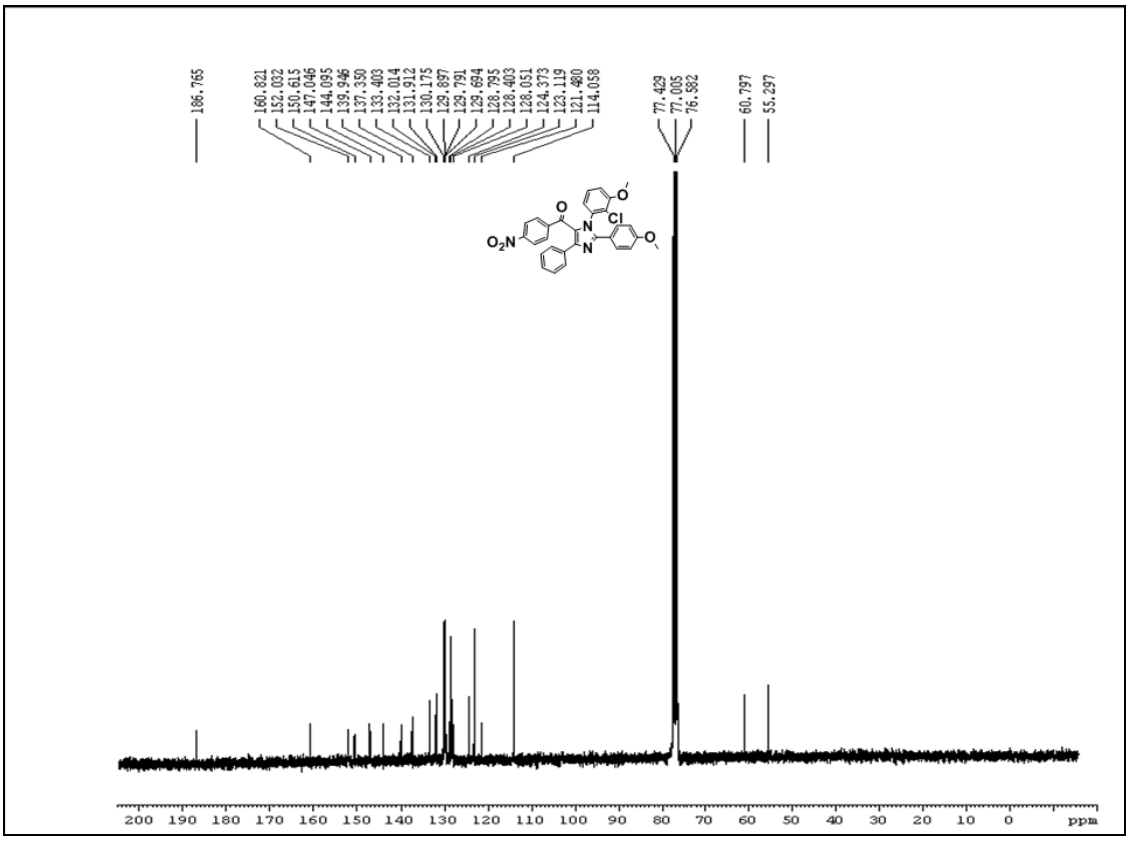


Fig 23. ^{13}C-NMR spectrum of **4j**

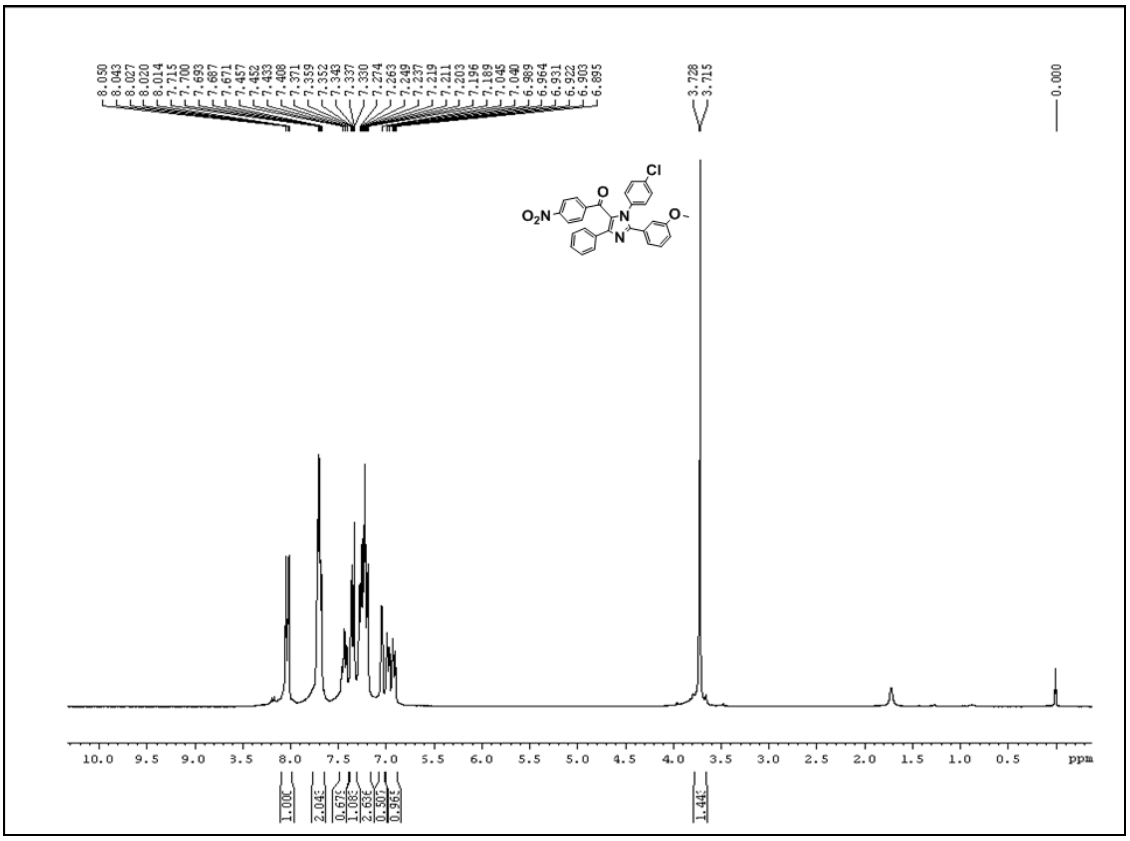


Fig 24. ^{1}H-NMR spectrum of **4k**

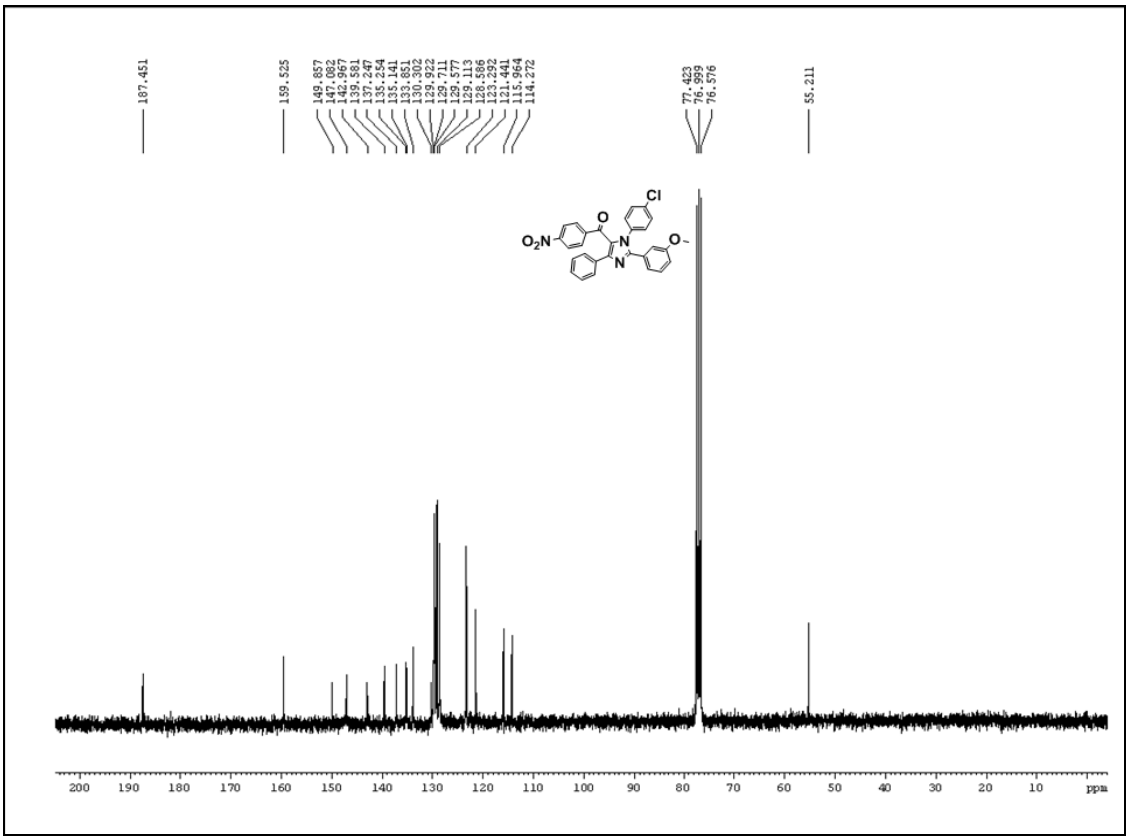


Fig 25. ^{13}C-NMR spectrum of **4k**

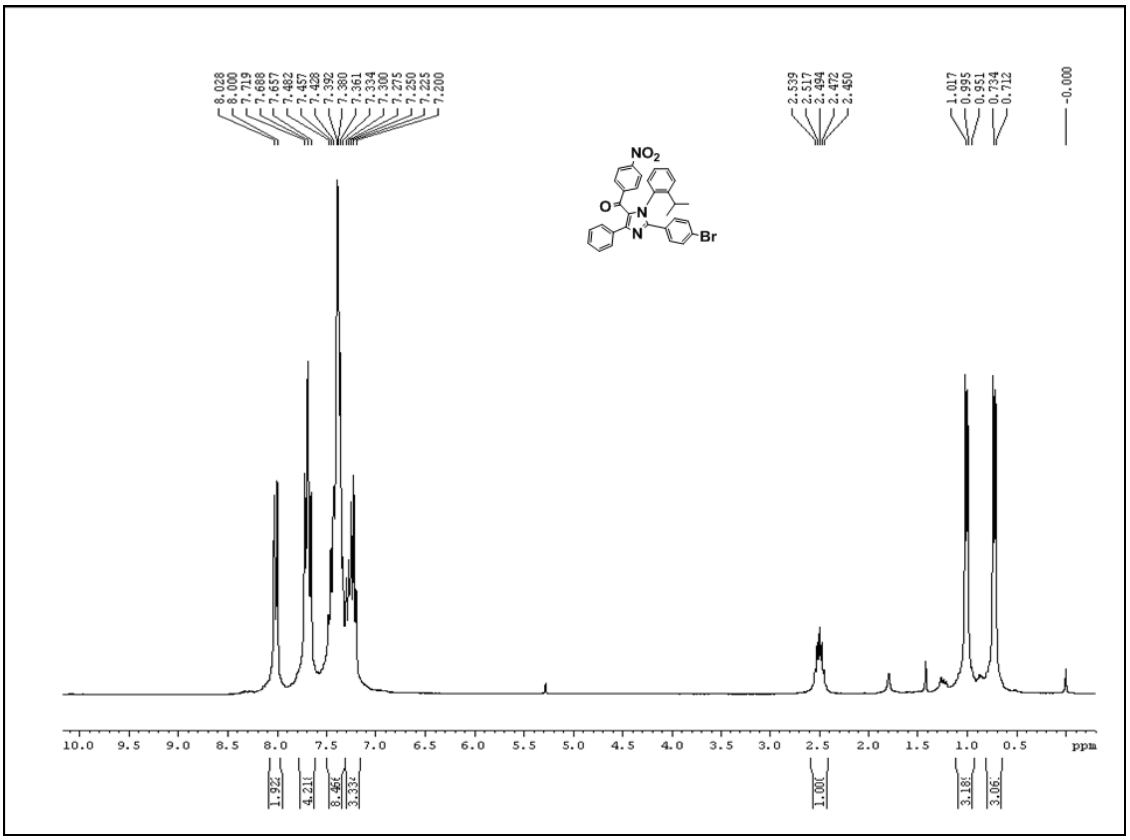


Fig 26. ^{1}H-NMR spectrum of **4l**

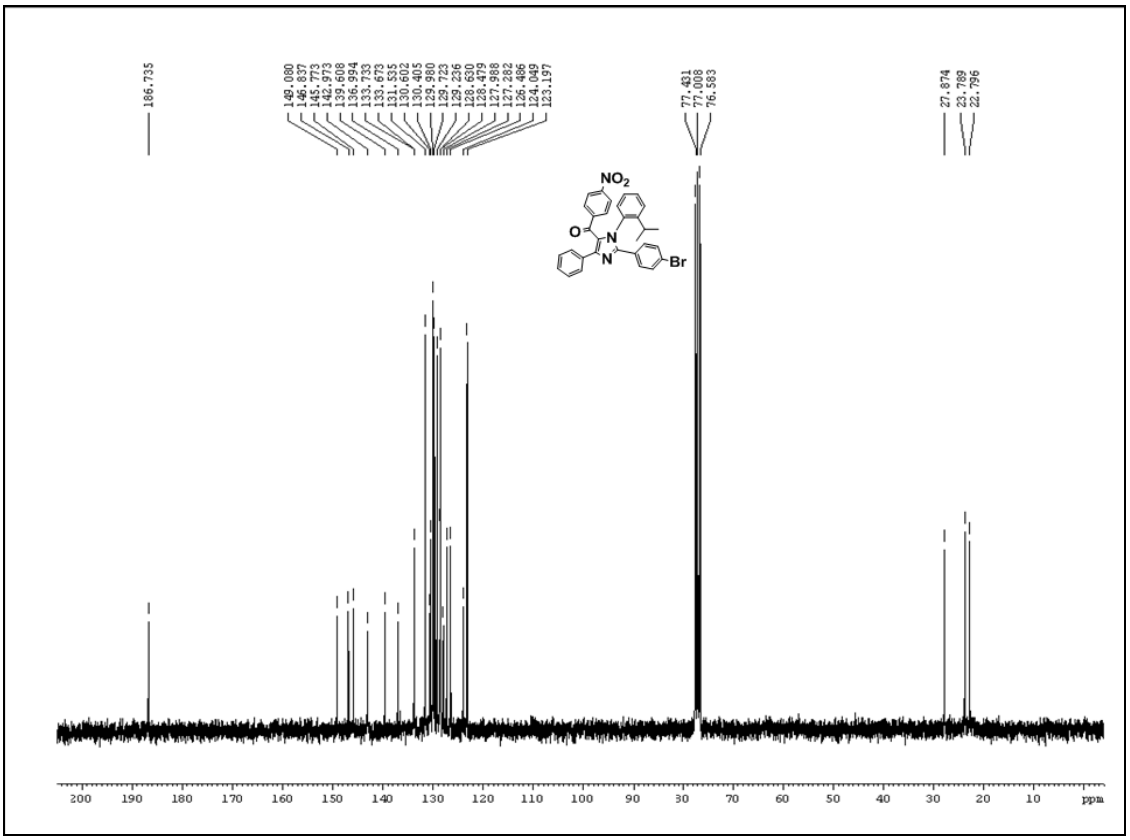


Fig 27. ^{13}C-NMR spectrum of **4l**

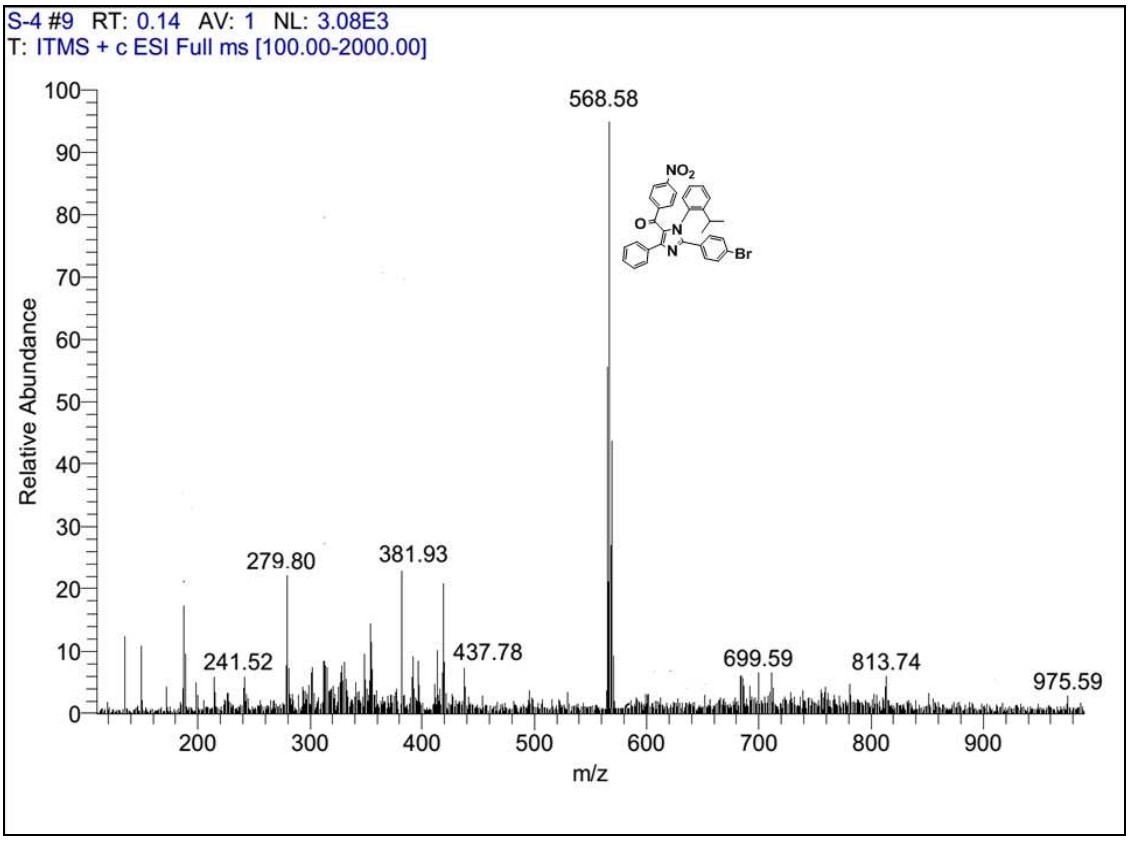


Fig 28. Mass spectrum of **4l**

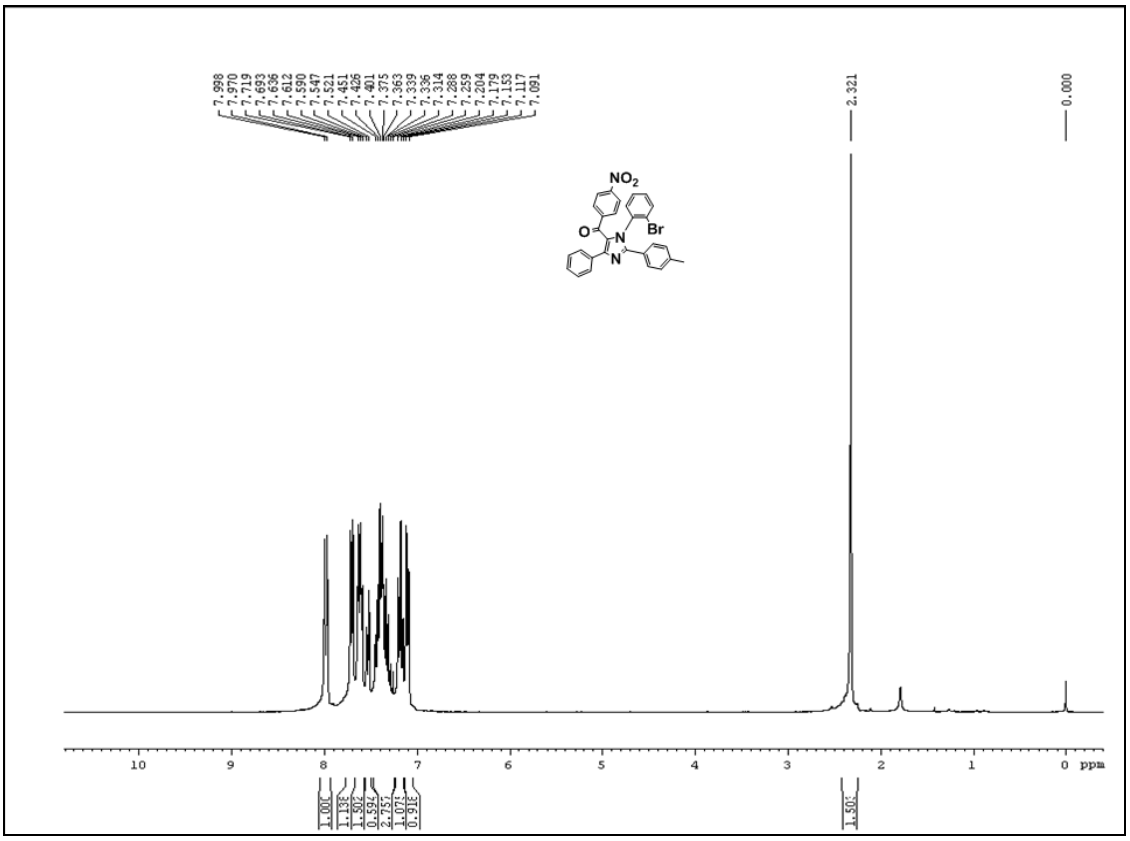


Fig 29. ^{1}H-NMR spectrum of **4m**

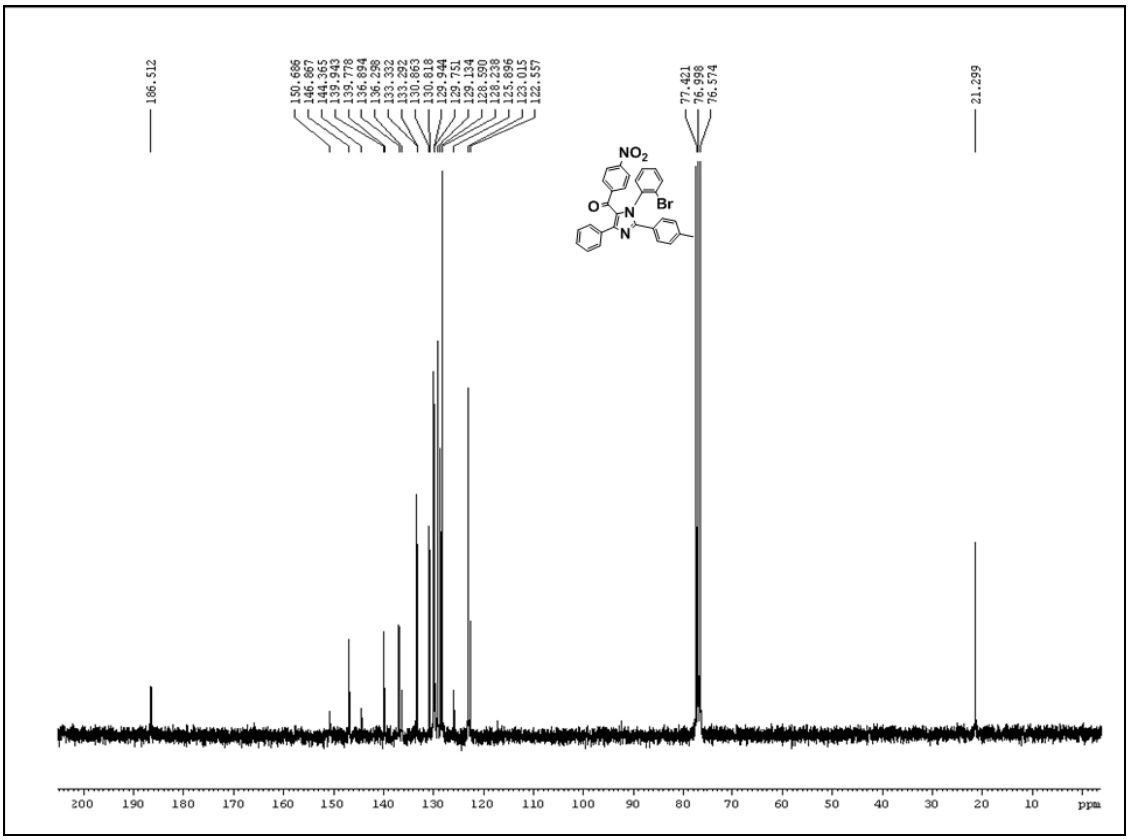


Fig 30. ^{13}C-NMR spectrum of **4m**

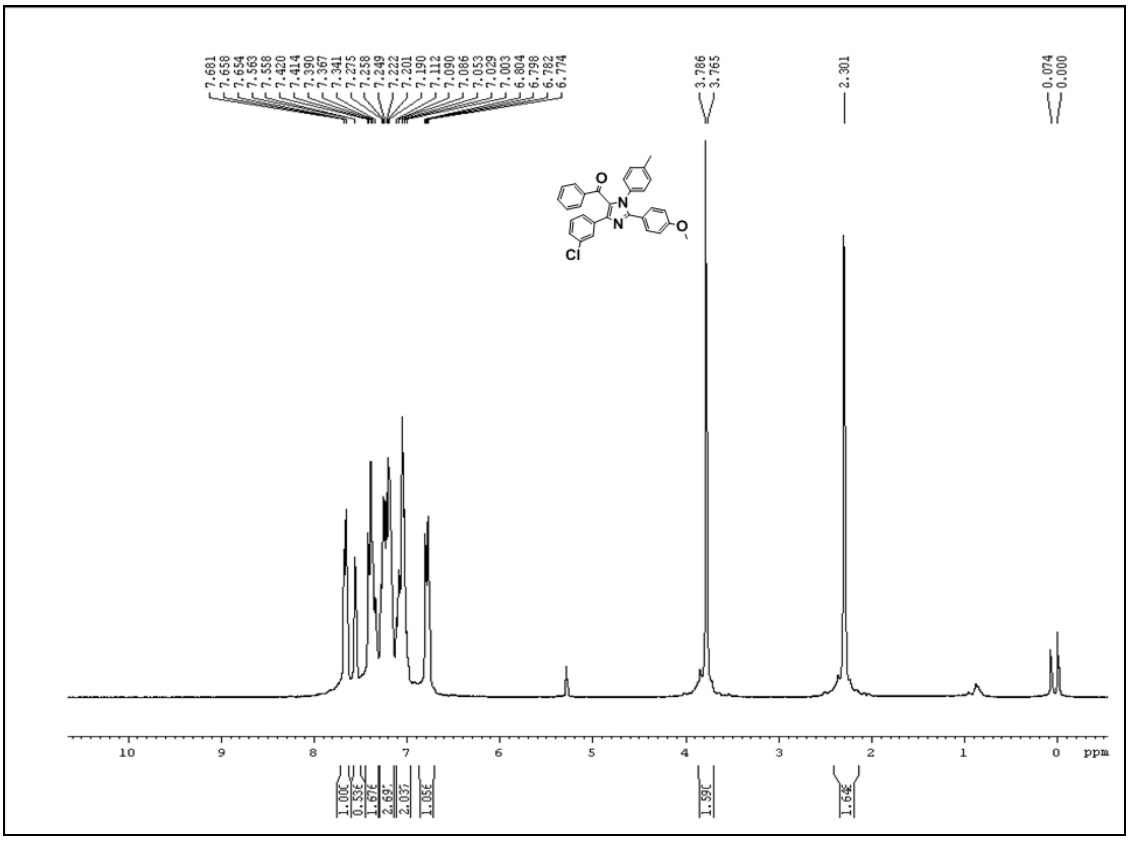


Fig 31. ^{1}H-NMR spectrum of **4n**

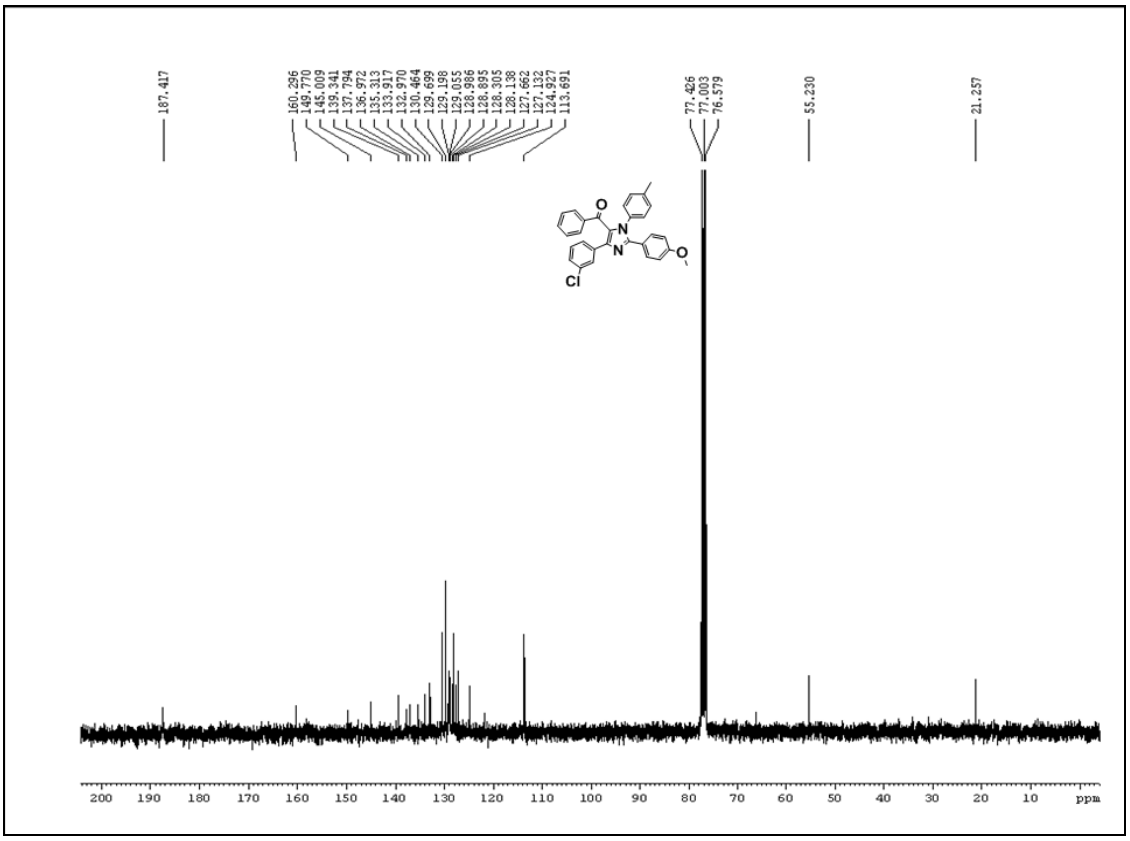


Fig 32. ^{13}C-NMR spectrum of **4n**

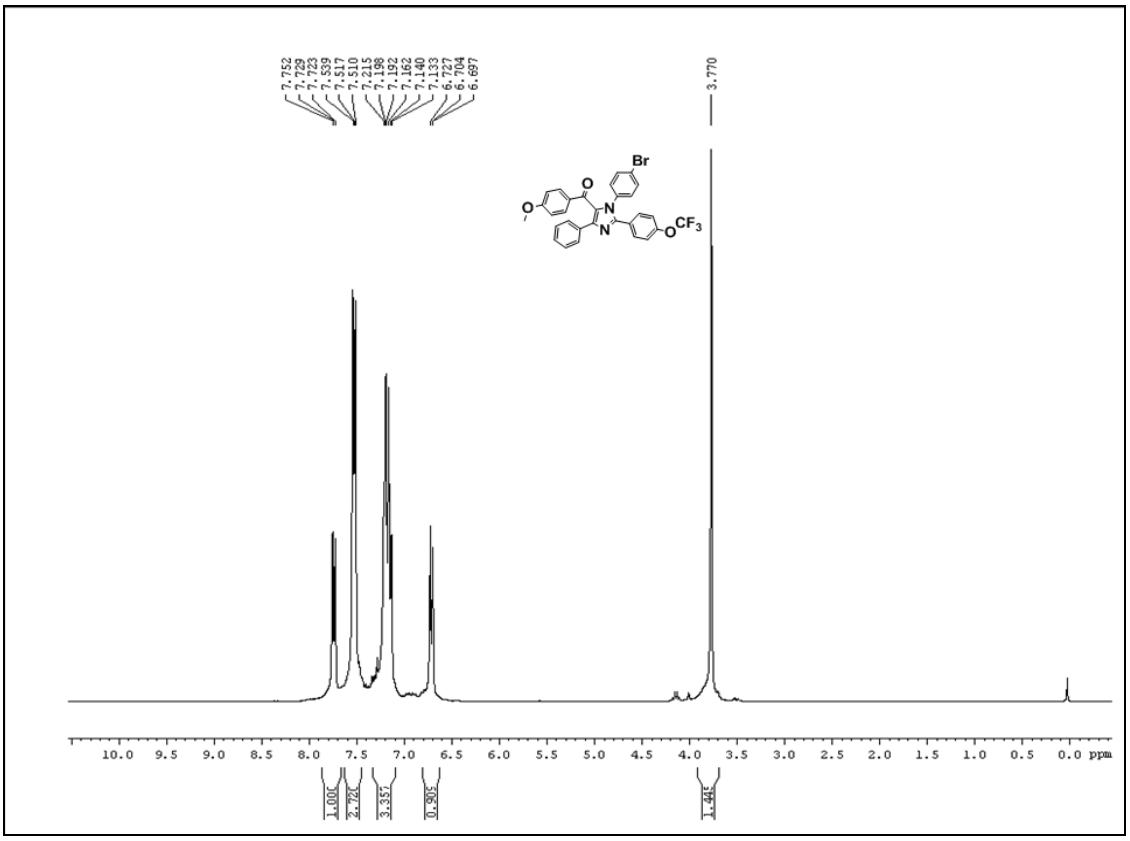


Fig 33. ^{1}H-NMR spectrum of **4o**

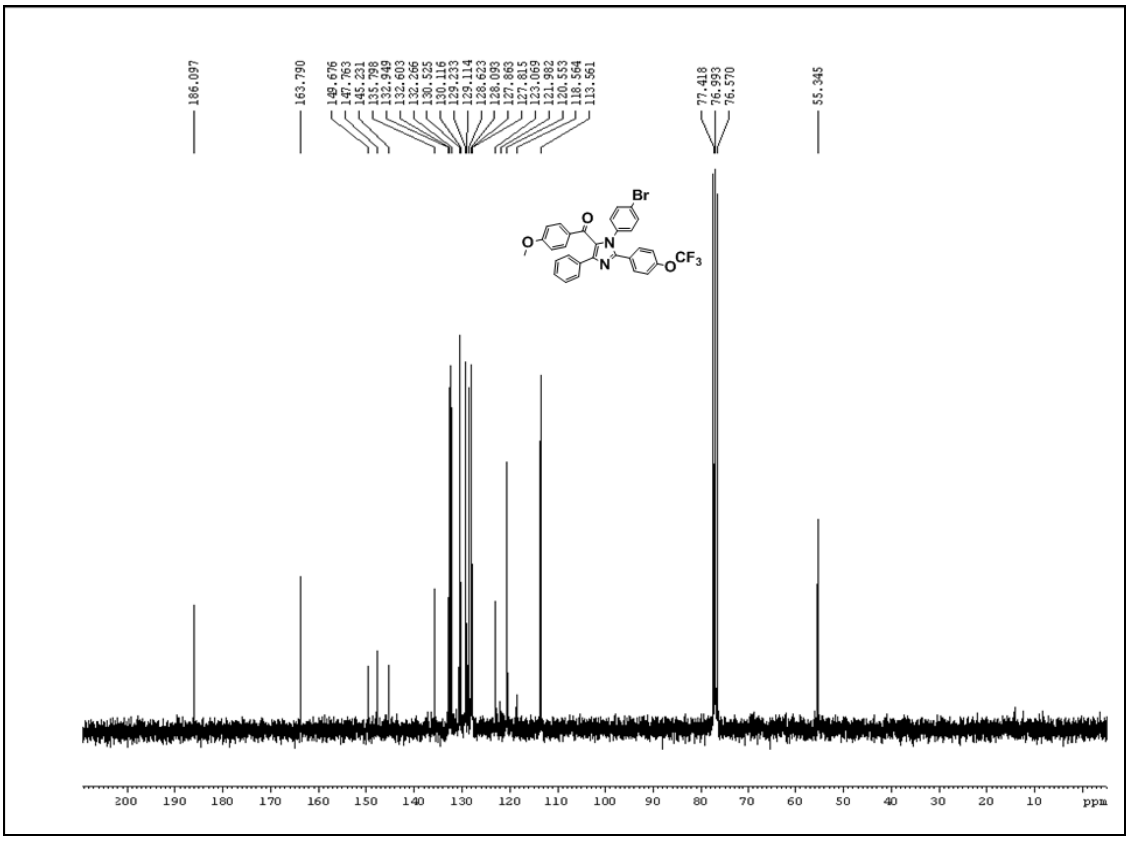


Fig 34. ^{13}C-NMR spectrum of **4o**

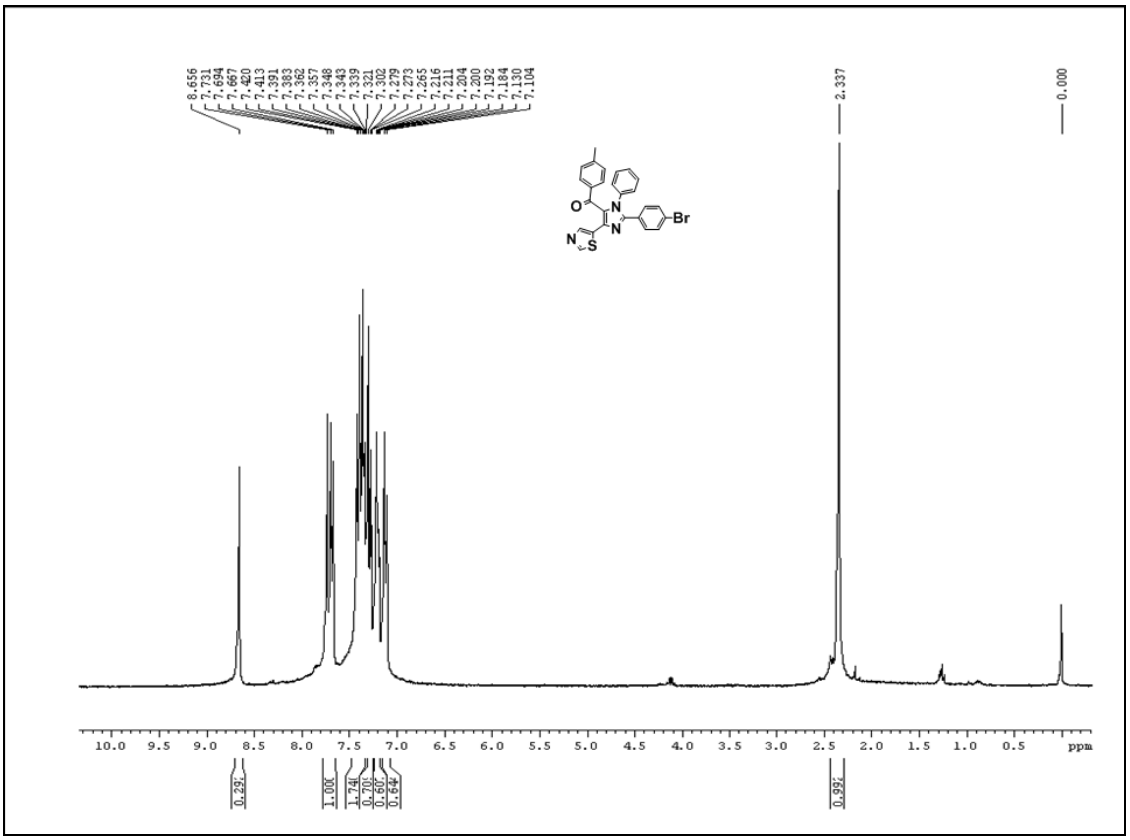


Fig 35. ^{1}H-NMR spectrum of **4p**

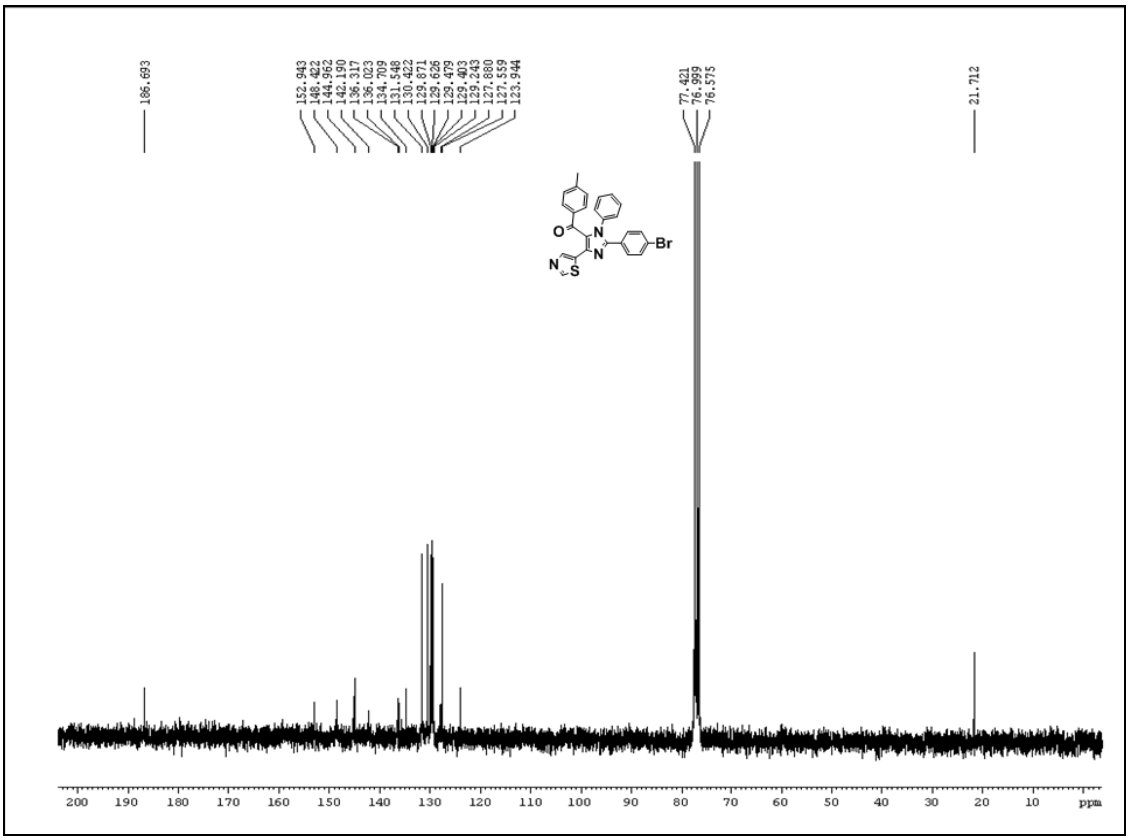


Fig 36. ^{13}C-NMR spectrum of **4p**

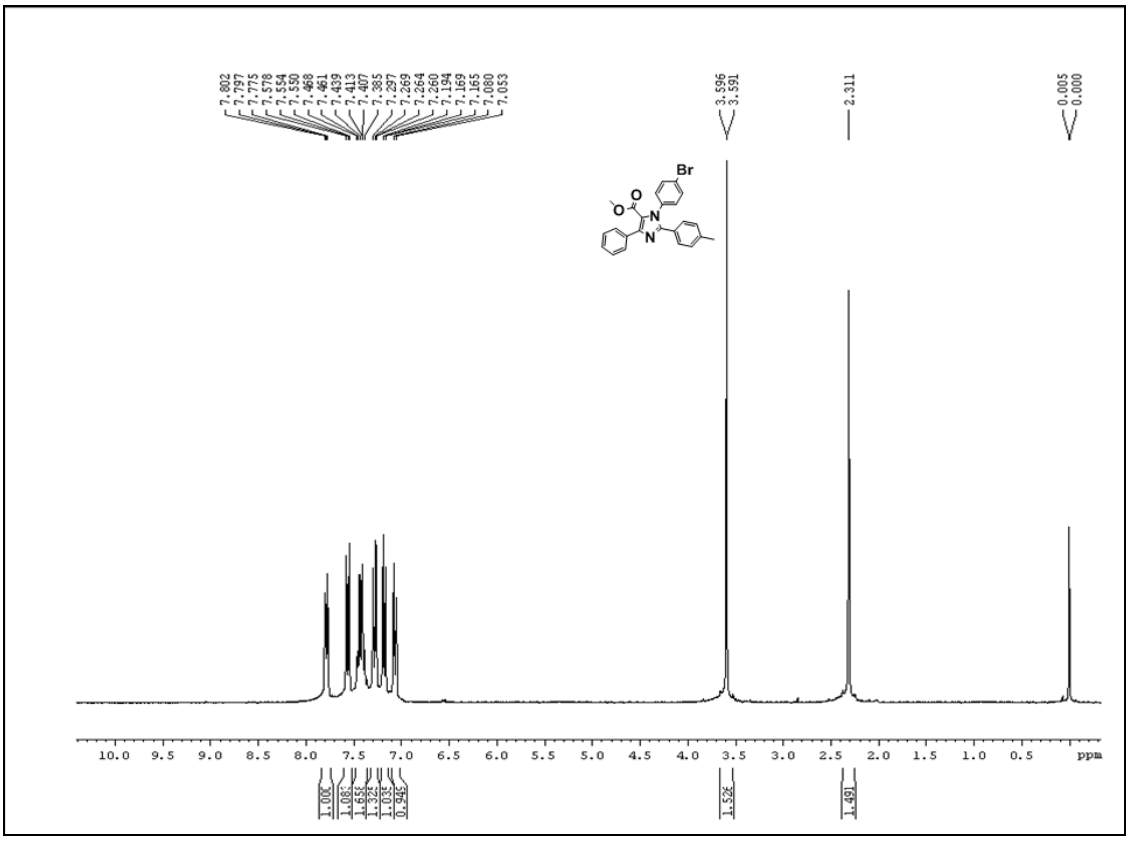


Fig 37. ^{1}H-NMR spectrum of **4q**

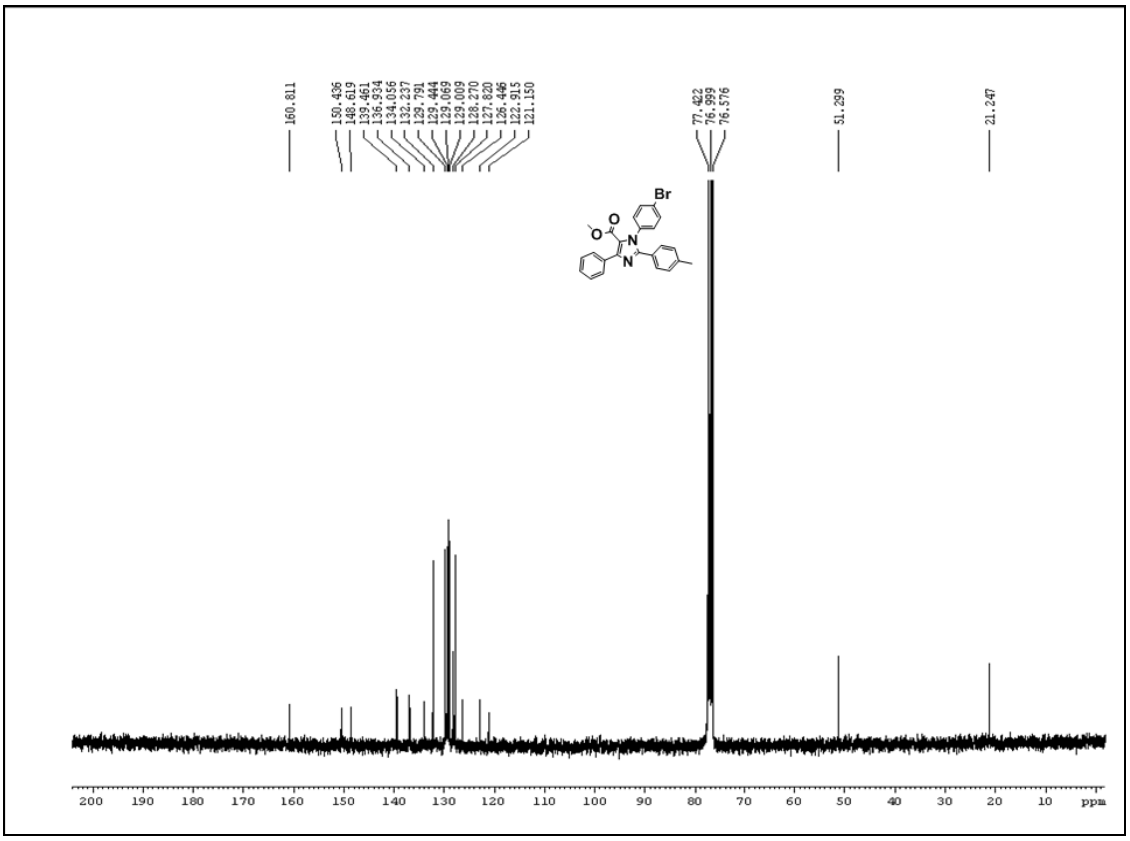


Fig 38. ^{13}C-NMR spectrum of **4q**

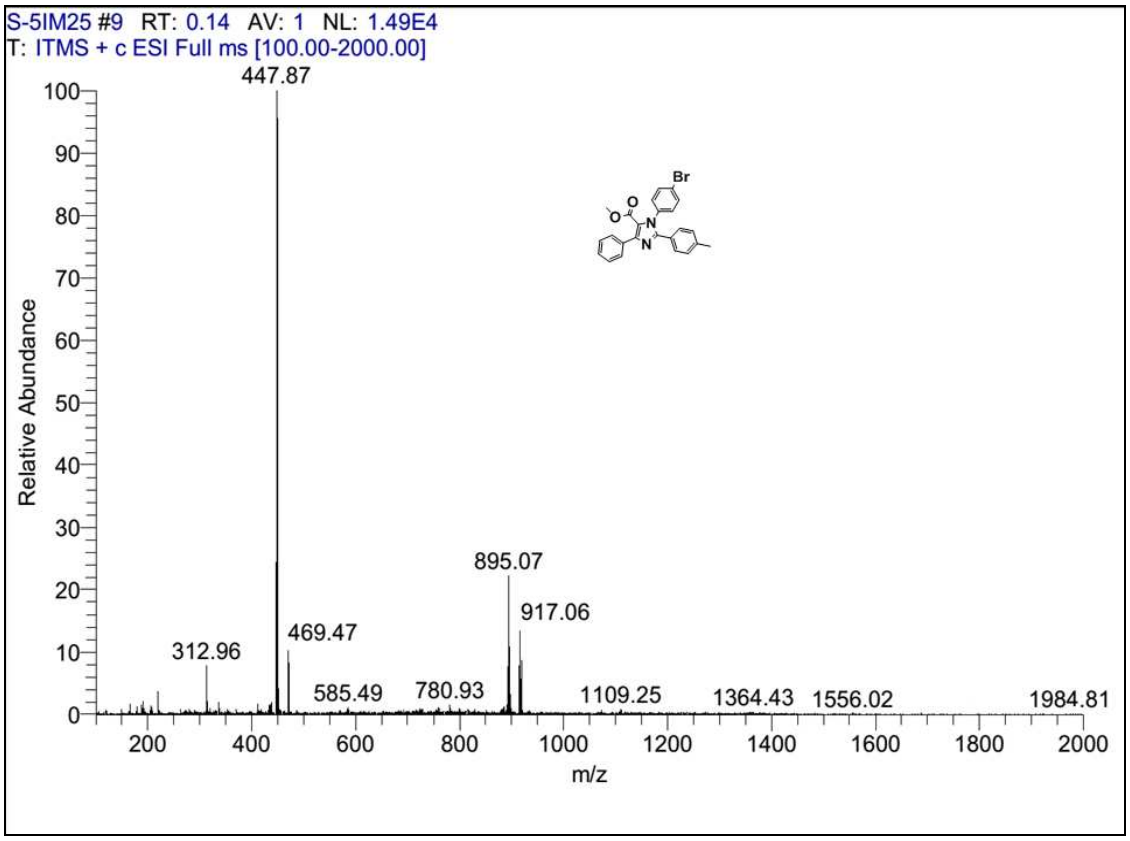


Fig 39. Mass spectrum of **4q**

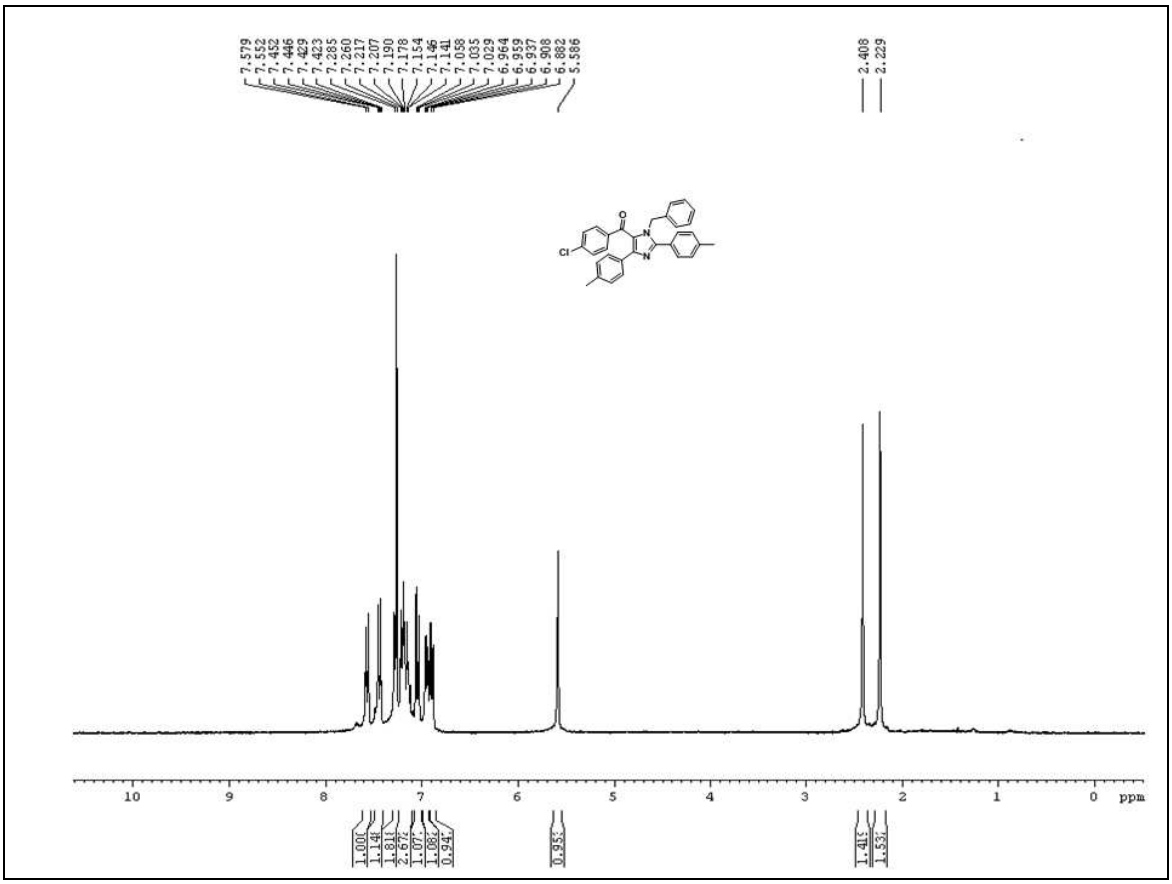


Fig 40. ^{1}H-NMR spectrum of **4r**

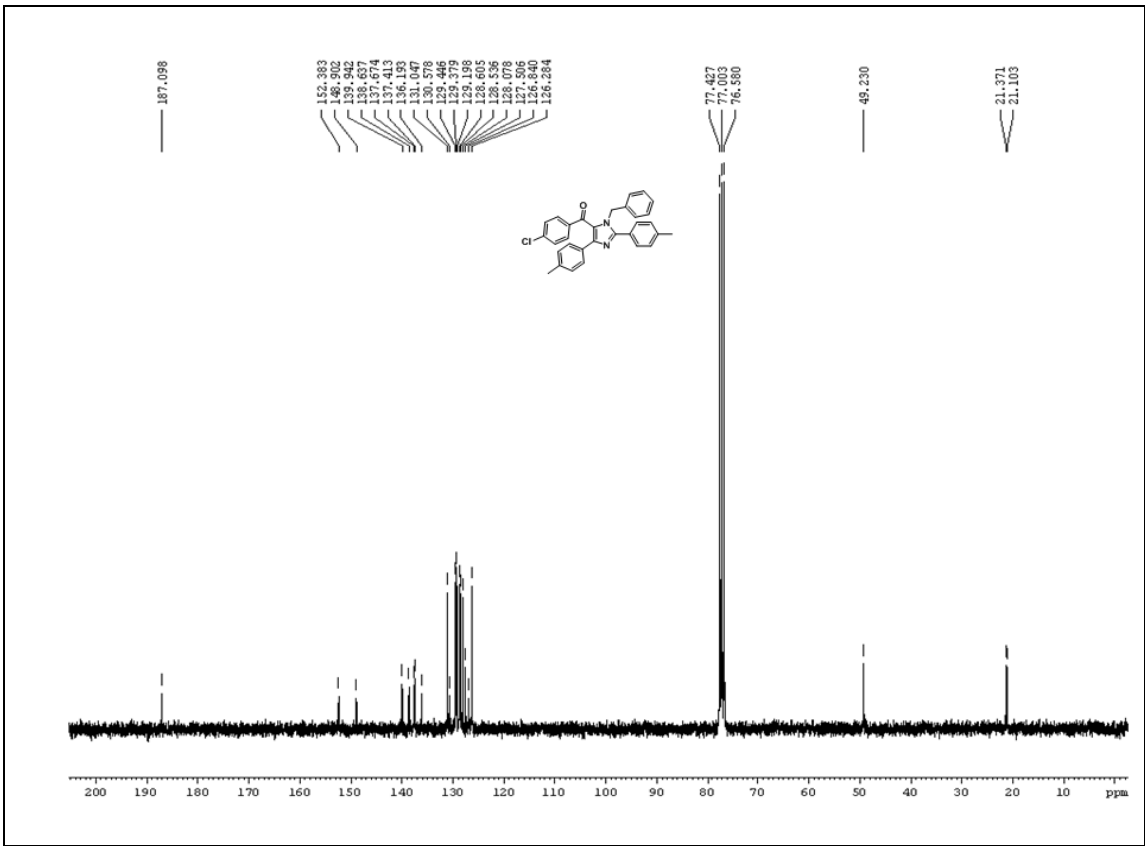


Fig 41. ^{13}C-NMR spectrum of **4r**