Pyridine appended 2-hydrazinylthiazole Derivatives: Design, Synthesis, *in vitro* and *in silico* Antimycobacterial Studies

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1. Spectral data of pyridine appended 2-hydrazinylthiazole derivatives (1b-30b)

2-(2-(propan-2-ylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(1b)**

Yellowish orange solid from EtOH (84 %). m.p. 177-179 ºC; ^{1}H NMR (400 MHz, DMSO-d_6) δ 10.90 (s, 1H), 8.73 (d, *J* = 4.8 Hz, 1H), 8.48 (t, *J* = 7.9 Hz, 1H), 8.36 (d, *J* = 8.0 Hz, 1H), 8.10 (s, 1H), 7.85 – 7.79 (m, 1H), 1.98 (s, 3H), 1.97 (s, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO-d_6): 170.93, 152.45, 146.36, 145.02, 142.79, 124.76, 122.90, 113.61, 24.83, 17.98; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3379.6, 3015.1, 1620.8, 1578.3, 1449.5; Anal. Calcd. for $C_{11}H_{12}N_4S$: C, 56.87; H, 5.21; N, 24.12; S, 13.80; Found: C, 56.83; H, 5.19; N, 24.35; S, 13.63; HR-MS (ESI+): Calcd. m/z for $C_{11}H_{12}N_4S$: 232.0783; Found m/z $[M+Na]^+$: 255.0670.

(E)-2-(2-benzylidenehydrazinyl)-4-(pyridin-2-yl)thiazole **(2b)**

Yellow solid from EtOH (89 %). m.p. 168-170 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 11.90 (s, 1H), 8.69 (d, J = 5.4 Hz, 1H), 8.36 – 8.25 (m, 2H), 8.14 (s, 1H), 8.00 (s, 1H), 7.68 (dd, J = 5.3, 3.1 Hz, 2H), 7.51 – 7.36 (m, 5H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.75, 165.39, 152.60, 149.02, 147.08, 145.22, 138.64, 137.85, 129.13, 124.76, 122.85, 122.56, 115.77, 108.12, 103.25; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3438.5, 1620.9, 1576.9, 1431.4; Anal. Calcd. for $C_{15}H_{12}N_4S$: C, 64.26; H, 4.31; N, 19.99; S, 11.44; Found: C, 64.29; H, 4.32; N, 19.98; S, 11.41; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{12}N_4S$: 280.0783; Found m/z $[M+H]^+$: 281.0872.

(E)-2-(2-(1-phenylethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(3b)**

Yellow solid from EtOH (83 %). m.p. 243-245 ºC; ^{1}H NMR (400 MHz, DMSO-d_6) δ 11.50 (s, 1H), 8.74 (d, *J* = 5.5 Hz, 1H), 8.45 (t, *J* = 7.8 Hz, 1H), 8.35 (s, 1H), 8.11 (s, 1H), 7.80 (d, *J* = 7.8 Hz, 3H), 7.42 (dd, *J* = 7.0, 4.4 Hz, 3H), 2.36 (s, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO-d_6): 170.91, 147.94, 143.88, 143.59, 137.35, 129.12, 128.35, 126.13, 124.55, 122.91, 113.89, 14.34; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3405.7, 3052.1, 1619.3, 1573.8, 1496.3; Anal. Calcd. for $C_{16}H_{14}N_4S$: C, 65.28; H, 4.79; N, 19.03; S, 10.89; Found: C, 65.33; H, 4.78; N, 19.05; S, 10.84; HR-MS (ESI+): Calcd. m/z for $C_{16}H_{14}N_4S$: 294.0939; Found m/z $[M+H]^+$: 295.1015.

(E)-2-(2-(naphthalen-1-ylmethylene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(4b)**

Yellow solid from EtOH (81 %). m.p. 188-190 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.83 (s, 1H), 8.80 – 8.70 (m, 4H), 8.36 (s, 2H), 8.11 (s, 1H), 8.04 – 7.99 (m, 3H), 7.61 (ddd, J = 5.3, 4.5, 2.6 Hz, 4H): ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 161.67, 142.57, 129.84, 128.94, 126.35, 125.66, 118.04; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3407.2, 1624.4, 1595.2, 1448.0; Anal.

Calcd. for $C_{19}H_{14}N_4S$: C, 69.07; H, 4.27; N, 16.96; S, 9.70; Found: C, 69.29; H, 4.29; N, 16.92; S, 9.50; HR-MS (ESI+): Calcd. m/z for $C_{19}H_{14}N_4S$: 330.0939; Found m/z $[M+H]^+$: 331.0998.

(E)-2-(2-(2-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(5b)**

Light yellow solid from EtOH (89 %). m.p. 233-235 ^oC; ^1H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.72 (d, J = 6.4 Hz, 1H), 8.48 (t, J = 8.6 Hz, 1H), 8.39 (d, J = 8.0 Hz, 1H), 8.36 (s, 1H), 8.18 (s, 1H), 7.87 – 7.79 (m, 2H), 7.45 (dd, J = 15.3, 7.4 Hz, 1H), 7.28 (dd, J = 15.0, 7.8 Hz, 2H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.28, 161.60, 159.12, 146.19, 145.01, 143.62, 143.08, 135.84, 131.69, 126.04, 125.13, 124.95, 123.28, 121.74, 121.65, 116.30, 116.10, 114.81; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3437.4, 1620.5, 1582.5, 1441.0, 1063.0; Anal. Calcd. for $C_{15}H_{11}FN_4S$: C, 60.39; H, 3.72; N, 18.78; S, 10.75; Found: C, 60.44; H, 3.75; N, 18.75; S, 10.71; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}FN_4S$: 298.0688; Found m/z $[M+H]^+$: 299.0770.

(E)-2-(2-(3-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(6b)**

Light yellow solid from EtOH (90 %). m.p. 227-229 ^oC; ^1H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.70 (dd, J = 4.4, 3.6 Hz, 1H), 8.31 (d, J = 25.3 Hz, 1H), 8.18 – 8.07 (m, 2H), 7.87 (d, J = 133.8 Hz, 1H), 7.48 (ddd, J = 10.2, 8.0, 4.1 Hz, 3H), 7.23 (dd, J = 5.5, 4.3 Hz, 1H): ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.25, 167.30, 163.76, 161.34, 147.07, 144.77, 144.09, 141.72, 141.41, 140.04, 136.76, 131.06, 124.60, 124.10, 123.51, 122.94, 122.78, 116.34, 113.69, 112.59; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3394.9, 1621.4, 158.1, 1421.4, 1063.1; Anal. Calcd. for $C_{15}H_{11}FN_4S$: C, 60.39; H, 3.72; N, 18.78; S, 10.75; Found: C, 60.41; H, 3.77; N, 18.75; S, 10.71; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}FN_4S$: 298.0688; Found m/z $[M+Na]^+$: 321.0560.

(E)-2-(2-(4-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(7b)**

Yellow crystalline from EtOH (91 %). m.p. 223-225 ^oC; ^1H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.71 (d, J = 4.9 Hz, 1H), 8.46 – 8.37 (m, 1H), 8.33 (d, J = 8.0 Hz, 1H), 8.16 (s, 1H), 8.08 (s, 1H), 7.75 (ddd, J = 8.8, 7.7, 4.6 Hz, 3H), 7.29 (t, J = 8.8 Hz, 2H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.44, 164.14, 161.67, 146.15, 144.97, 143.52, 143.00, 142.05, 130.71, 128.72, 128.63, 124.86, 123.25, 116.14, 115.92, 114.53; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3430.6, 1621.8, 1569.0, 1437.4, 1065.9; Anal. Calcd. for $C_{15}H_{11}FN_4S$: C, 60.39; H, 3.72; N, 18.78; S, 10.75; Found: C, 60.43; H, 3.75; N, 18.76; S, 10.70; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}FN_4S$: 298.0688; Found m/z $[M+H]^+$: 299.0766.

(E)-2-(2-(1-(4-fluorophenyl)ethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(8b)**

Yellow solid from EtOH (92 %). m.p. 239-241 ^oC; ^1H NMR (400 MHz, δ, ppm, DMSO-d_6): 11.41 (s, 1H), 8.73 (d, *J* = 4.2 Hz, 1H), 8.42 (t, *J* = 7.9 Hz, 1H), 8.32 (d, *J* = 8.0 Hz, 1H), 8.09 (s, 1H), 7.88 – 7.79 (m, 3H), 7.79 – 7.74 (m, 1H), 7.25 (d, *J* = 8.9 Hz, 2H), 2.35 (s, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 170.88, 147.45, 134.14, 128.08, 127.99, 124.55, 122.84,

115.52, 115.30, 14.45; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3384.5, 3046.5, 1618.5, 1576.9, 1448.6, 1125.4; Anal. Calcd. for $C_{16}H_{13}FN_4S$: C, 61.52; H, 4.20; N, 17.94; S, 10.26; Found: C, 61.58; H, 4.21; N, 17.92; S, 10.21; HR-MS (ESI+): Calcd. m/z for $C_{16}H_{13}FN_4S$: 312.0845; Found m/z $[M+H]^+$: 313.0918.

(E)-4-(pyridin-2-yl)-2-(2-(3-(trifluoromethyl)benzylidene)hydrazinyl)thiazole **(9b)**

Light yellow solid from EtOH (94 %). m.p. 178-180 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.42 (s, 1H), 8.58 (d, J = 4.7 Hz, 1H), 8.13 (s, 1H), 7.97 (d, J = 10.1 Hz, 2H), 7.88 (t, J = 7.8 Hz, 2H), 7.73 – 7.64 (m, 2H), 7.57 (s, 1H), 7.30 (t, J = 6.7 Hz, 1H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 168.32, 152.20, 149.45, 139.63, 137.27, 135.60, 130.04, 129.92, 129.85, 129.54, 125.45, 122.70, 122.50, 119.99, 107.87; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3427.0, 1593.1, 1561.0, 1478.6, 1125.5; Anal. Calcd. for $C_{16}H_{11}F_3N_4S$: C, 55.17; H, 3.18; N, 16.08; S, 9.20; Found: C, 55.19; H, 3.21; N, 16.05; S, 9.18; HR-MS (ESI+): Calcd. m/z for $C_{16}H_{11}F_3N_4S$: 348.0657; Found m/z $[M+Na]^+$: 371.0542.

(E)-4-(pyridin-2-yl)-2-(2-(4-(trifluoromethyl)benzylidene)hydrazinyl)thiazole **(10b)**

Yellow solid from EtOH (90 %). m.p. 157-159 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.54 (s, 1H), 8.70 (d, J = 5.3 Hz, 1H), 8.42 – 8.19 (m, 3H), 8.03 (d, J = 8.0 Hz, 1H), 7.89 (d, J = 8.2 Hz, 2H), 7.80 (d, J = 8.4 Hz, 2H), 7.70 (d, J = 5.9 Hz, 1H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.09, 146.96, 143.95, 141.04, 138.06, 129.38, 127.01, 125.84, 124.60, 122.81, 113.90; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3391.9, 1619.5, 1583.3, 1442.8, 1121.7; Anal. Calcd. for $C_{16}H_{11}F_3N_4S$: C, 55.17; H, 3.18; N, 16.08; S, 9.20; Found: C, 55.18; H, 3.22; N, 16.05; S, 9.18; HR-MS (ESI+): Calcd. m/z for $C_8H_9N_3S$: 348.0657; Found m/z $[M+H]^+$: 349.0704.

(E)-2-(2-(2-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(11b)**

Light yellow solid from EtOH (89 %). m.p. 231-233 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.56 (s, 1H), 8.72 (d, J = 4.8 Hz, 1H), 8.53 (s, 1H), 8.47 – 8.39 (m, 1H), 8.35 (d, J = 8.0 Hz, 1H), 8.14 (s, 1H), 7.98 – 7.90 (m, 1H), 7.79 (dd, J = 9.5, 3.4 Hz, 1H), 7.55 – 7.50 (m, 1H), 7.47 – 7.40 (m, 2H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 168.98, 147.15, 143.36, 138.14, 132.71, 131.06, 129.99, 127.57, 126.13, 124.28, 123.21, 113.63; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3364.3, 1622.8, 1582.2, 1446.1, 750.4; Anal. Calcd. for $C_{15}H_{11}ClN_4S$: C, 57.23; H, 3.52; N, 17.80; S, 10.18; Found: C, 57.25; H, 3.55; N, 17.78; S, 10.15; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}ClN_4S$: 314.0393; Found m/z $[M+H]^+$: 315.0471.

(E)-2-(2-(3-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(12b)**

Yellow solid from EtOH (87 %). m.p. 194-196 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.39 (s, 1H), 8.69 (d, J = 5.0 Hz, 1H), 8.34 (t, J = 7.6 Hz, 1H), 8.27 (d, J = 7.8 Hz, 1H), 8.12

(s, 1H), 8.03 (s, 1H), 7.70 (d, J = 11.9 Hz, 2H), 7.64 (d, J = 6.6 Hz, 1H), 7.51 – 7.42 (m, 2H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 168.97, 166.82, 144.43, 140.86, 136.29, 133.80, 130.82, 129.13, 125.66, 125.05, 124.27, 122.91, 122.61, 112.98; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3395.9, 1621.6, 1573.9, 1446.4, 784.9; HR-MS (ESI+): Anal. Calcd. for $C_{15}H_{11}ClN_4S$: C, 57.23; H, 3.52; N, 17.80; S, 10.18; Found: C, 57.24; H, 3.56; N, 17.77; S, 10.16; Calcd. m/z for $C_{15}H_{11}ClN_4S$: 314.0393; Found m/z $[M+H]^+$: 315.0462.

(E)-2-(2-(4-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(13b)**

Yellow solid from EtOH (88 %). m.p. 220-222 ^oC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.70 (d, J = 5.0 Hz, 1H), 8.42 – 8.27 (m, 2H), 8.14 (s, 1H), 8.07 – 8.03 (m, 1H), 7.73 (d, J = 5.7 Hz, 1H), 7.70 (d, J = 8.6 Hz, 2H), 7.51 (d, J = 8.5 Hz, 2H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.30, 145.98, 145.13, 143.38, 142.89, 141.75, 134.07, 132.98, 129.01, 128.11, 124.89, 123.30, 114.79 ; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3305.5, 1621.8, 1592.2, 1448.7, 743.1; Anal. Calcd. for $C_{15}H_{11}ClN_4S$: C, 57.23; H, 3.52; N, 17.80; S, 10.18; Found: C, 57.26; H, 3.56; N, 17.77; S, 10.14; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}ClN_4S$: 314.0393; Found m/z $[M+H]^+$: 315.0488.

(E)-2-(2-(2,4-dichlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(14b)**

Light yellow solid from EtOH (84 %). m.p. 236-238 ^oC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.65 (s, 1H), 8.71 (d, J = 6.3 Hz, 1H), 8.45 (s, 1H), 8.39 (d, J = 6.0 Hz, 1H), 8.31 (d, J = 8.0 Hz, 1H), 8.11 (s, 1H), 7.90 (d, J = 8.6 Hz, 1H), 7.75 (t, J = 6.5 Hz, 1H), 7.67 (s, 1H), 7.51 (d, J = 8.6 Hz, 1H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 168.76, 166.82, 146.59, 144.27, 143.63, 137.35, 134.38, 133.00, 130.32, 129.31, 127.93, 127.33, 124.54, 122.77, 114.08; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3357.4, 1622.3, 1593.5, 1475.3, 784.6; Anal. Calcd. for $C_{15}H_{10}Cl_2N_4S$: C, 51.59; H, 2.89; N, 16.04; S, 9.18; Found: C, 51.61; H, 2.93; N, 16.01; S, 9.15; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{10}Cl_2N_4S$: 348.0003; Found m/z $[M+H]^+$: 349.0097.

(E)-2-(2-(3-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**15b)**

Yellow solid from EtOH (85 %). m.p. 183-185 ^oC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.57 (s, 1H), 8.68 (s, 1H), 8.33 – 7.96 (m, 4H), 7.84 (s, 1H), 7.66 (d, J = 5.6 Hz, 2H), 7.57 (d, J = 7.8 Hz, 1H), 7.41 (d, J = 7.5 Hz, 1H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 168.98, 167.02, 141.09, 136.70, 136.51, 131.12, 128.67, 125.60, 122.32; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3382.9, 1578.7, 1448.5, 614.8; Anal. Calcd. for $C_{15}H_{11}BrN_4S$: C, 50.15; H, 3.09; N, 15.60; S, 8.92; Found: C, 50.18; H, 3.11; N, 15.58; S, 8.89; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}BrN_4S$: 357.9888; Found m/z $[M+H]^+$: 358.9962.

(E)-2-(2-(4-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(16b)**

Light yellow solid from EtOH (89 %). m.p. 205-207 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.27 (s, 1H), 8.67 (d, J = 2.8 Hz, 1H), 8.24 (dd, J = 15.2, 8.3 Hz, 2H), 8.10 (d, J = 3.0 Hz, 1H), 8.03 – 7.91 (m, 1H), 7.70 – 7.58 (m, 5H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 167.43, 144.55, 142.90, 141.54, 140.76, 139.89, 132.04, 130.38, 126.82, 122.74, 121.37, 121.09, 111.29; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3430.6, 1624.7, 568.6; Anal. Calcd. for $C_{15}H_{11}BrN_4S$: C, 50.15; H, 3.09; N, 15.60; S, 8.92; Found: C, 50.17; H, 3.12; N, 15.59; S, 8.88; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}BrN_4S$: 357.9888; Found m/z $[M+H]^+$: 358.9976.

(E)-2-(2-(4-methylbenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(17b)**

Greenish yellow solid from EtOH (86 %). m.p. 176-178 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.72 (d, J = 5.5 Hz, 1H), 8.52 – 8.44 (m, 1H), 8.38 (d, J = 8.0 Hz, 1H), 8.16 (t, J = 2.7 Hz, 2H), 7.82 (dd, J = 9.6, 3.5 Hz, 1H), 7.56 (d, J = 8.1 Hz, 2H), 7.25 (d, J = 8.0 Hz, 2H), 2.32 (s, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.54, 145.98, 145.24, 143.37, 143.26, 142.79, 139.61, 131.36, 129.59, 126.58, 124.93, 123.36, 114.67, 21.13; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3408.5, 2916.2, 1584.8, 1441.1; Anal. Calcd. for $C_{16}H_{14}N_4S$: C, 65.28; H, 4.79; N, 19.03; S, 10.89; Found: C, 65.34; H, 4.81; N, 18.99; S, 10.86; HR-MS (ESI+): Calcd. m/z for $C_{16}H_{14}N_4S$: 294.0939; Found m/z $[M+H]^+$: 295.1026.

(E)-2-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol **(18b)**

Yellow solid from EtOH (83 %). m.p. 187-189 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 10.12 (s, 1H), 8.70 (dd, J = 5.5, 0.8 Hz, 1H), 8.45 (s, 1H), 8.42 – 8.36 (m, 1H), 8.32 (d, J = 8.0 Hz, 1H), 8.06 (s, 1H), 7.76 (d, J = 7.0 Hz, 1H), 7.67 (dd, J = 7.8, 1.6 Hz, 1H), 7.27 – 7.21 (m, 1H), 6.90 (dd, J = 15.9, 7.9 Hz, 2H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.27, 156.17, 146.25, 144.84, 143.61, 143.12, 140.80, 131.07, 126.01, 124.83, 123.19, 120.07, 119.60, 116.27, 114.13; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3435.7, 3109.5, 1622.2, 1591.8, 1447.7; Anal. Calcd. for $C_{15}H_{12}N_4OS$: C, 60.80; H, 4.08; N, 18.91; S, 10.82; Found: C, 60.83; H, 4.11; N, 18.89; S, 10.78; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{12}N_4OS$: 296.0732; Found m/z $[M+H]^+$: 297.0790.

(E)-3-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol **(19b)**

Yellow solid from EtOH (84 %). m.p. 189-191 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.16 (s, 1H), 10.91 (s, 1H), 8.70 (dd, J = 5.9, 1.0 Hz, 1H), 8.33 (t, J = 17.4 Hz, 2H), 8.11 – 8.00 (m, 2H), 7.74 (s, 1H), 7.28 (t, J = 4.8 Hz, 1H), 7.20 (dd, J = 8.4, 1.9 Hz, 1H), 7.05 – 6.98 (m, 1H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.28, 157.61, 146.50, 143.14, 135.20, 129.89, 124.59, 122.89, 118.06, 117.02, 113.87, 112.20; IR (KBr, $\upsilon_{max,}$ cm^{-}1): 3423.5, 1621.8, 1587.1, 1433.6; Anal. Calcd. for $C_{15}H_{12}N_4OS$: C, 60.80; H, 4.08; N, 18.91; S, 10.82; Found: C, 60.82; H, 4.13; N, 18.87; S, 10.81; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{12}N_4OS$: 296.0732; Found m/z $[M+H]^+$: 297.0798.

(E)-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol (**20b)**

Dark green solid from EtOH (82 %). m.p. 194-196 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 9.89 (s, 1H), 8.66 (d, J = 5.0 Hz, 1H), 8.27 – 8.15 (m, 2H), 8.03 (s, 1H), 7.87 (s, 1H), 7.61 (t, J = 5.9 Hz, 1H), 7.51 (d, J = 8.6 Hz, 2H), 6.84 (d, J = 8.6 Hz, 2H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.27, 159.11, 148.44, 145.29, 143.15, 142.16, 128.24, 125.21, 124.03, 122.07, 115.85, 111.57; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3290.6, 3206.2, 1600.6, 1441.2, 1209.0; Anal. Calcd. for $C_{15}H_{12}N_4OS$: C, 60.80; H, 4.08; N, 18.91; S, 10.82; Found: C, 60.82; H, 4.11; N, 18.88; S, 10.80; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{12}N_4OS$: 296.0732; Found m/z $[M+H]^+$: 297.0821.

(E)-2-(2-(4-methoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(21b)**

Light brown solid from EtOH (85 %). m.p. 204-206 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.77 - 8.66 (m, 1H), 8.43 (d, J = 7.7 Hz, 1H), 8.36 (d, J = 8.2 Hz, 1H), 8.11 (d, J = 9.9 Hz, 2H), 7.79 (dd, J = 7.1, 6.0 Hz, 1H), 7.62 (d, J = 8.8 Hz, 2H), 7.01 (d, J = 8.8 Hz, 2H), 3.80 (s, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.51, 160.61, 146.38, 144.68, 143.66, 143.19, 128.14, 126.65, 124.73, 123.10, 114.46, 113.98, 55.37; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3433.5, 3001.5, 1587.4, 1446.7, 1251.0; Anal. Calcd. for $C_{16}H_{14}N_4OS$: C, 61.92; H, 4.55; N, 18.05; S, 10.33; Found: C, 61.95; H, 4.58; N, 18.03; S, 10.29; HR-MS (ESI+): Calcd. m/z for $C_{16}H_{14}N_4OS$: 310.0888; Found m/z $[M+H]^+$: 311.0982.

(E)-2-(2-(3,4-dimethoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(22b)**

Yellow solid from EtOH (87 %). m.p. 219-221 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.70 (dd, J = 5.9, 1.0 Hz, 1H), 8.41 – 8.33 (m, 1H), 8.30 (d, J = 8.0 Hz, 1H), 8.08 (s, 1H), 8.03 (d, J = 2.7 Hz, 1H), 7.79 – 7.69 (m, 1H), 7.29 (d, J = 1.9 Hz, 1H), 7.19 (d, J = 1.9 Hz, 1H), 7.02 (d, J = 8.4 Hz, 1H), 3.82 (s, 3H), 3.80 (s, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.60, 150.56, 149.14, 143.43, 126.88, 124.18, 123.17, 120.91, 111.80, 108.43, 55.73, 55.53; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3424.2, 1622.0, 1515.0, 1067.4; Anal. Calcd. for $C_{17}H_{16}N_4O_2S$: C, 59.98; H, 4.74; N, 16.46; S, 9.42; Found: C, 59.99; H, 4.78; N, 16.44; S, 9.39; HR-MS (ESI+): Calcd. m/z for $C_{17}H_{16}N_4O_2S$: 340.0994; Found m/z $[M+H]^+$: 341.1082.

(E)-4-(pyridin-2-yl)-2-(2-(3,4,5-trimethoxybenzylidene)hydrazinyl)thiazole **(23b)**

Yellow solid from EtOH (84 %). m.p. 223-225 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.71 (d, J = 4.9 Hz, 1H), 8.35 (t, J = 19.3 Hz, 2H), 8.09 (s, 2H), 7.78 (s, 1H), 7.00 (s, 2H), 3.83 (s, 6H), 3.70 (s, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.46, 153.27, 146.45, 144.71, 143.83, 143.29, 142.91, 138.95, 129.68, 124.78, 123.11, 114.20, 103.77, 60.24, 55.96; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3444.8, 1588.6, 1456.9, 1124.2; Anal. Calcd. for $C_{18}H_{18}N_4O_3S$: C, 58.36; H, 4.90;

N, 15.13; S, 8.65; Found: C, 58.40; H, 4.91; N, 15.11; S, 8.62; HR-MS (ESI+): Calcd. m/z for $C_{18}H_{18}N_4O_3S$: 370.1100; Found m/z $[M+H]^+$: 371.1194.

(E)-2-methoxy-5-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol **(24b)**

Light yellow solid from EtOH (89 %). m.p. 242-244 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.72 (d, J = 5.2 Hz, 1H), 8.51 – 8.30 (m, 2H), 8.10 (d, J = 36.2 Hz, 2H), 7.81 (s, 1H), 7.20 (s, 1H), 6.99 (dd, J = 21.6, 8.3 Hz, 2H), 3.80 (s, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO-d_6): 169.49, 149.59, 146.89, 146.24, 143.61, 143.03, 126.89, 124.76, 123.16, 119.78, 114.09, 111.98, 111.89, 55.66; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3433.6, 3251.9, 3080.1, 1614.3, 1586.2, 1437.4; Anal. Calcd. for $C_{16}H_{14}N_4O_2S$: C, 58.88; H, 4.32; N, 17.17; S, 9.82; Found: C, 58.92; H, 4.34; N, 17.14; S, 9.79; HR-MS (ESI+): Calcd. m/z for $C_{16}H_{14}N_4O_2S$: 326.0837; Found m/z $[M+H]^+$: 327.0916.

(E)-2-ethoxy-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol **(25b)**

Yellow solid from EtOH (84 %). m.p. 233-235ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.70 (d, J = 4.8 Hz, 1H), 8.38 (d, J = 7.6 Hz, 1H), 8.31 (d, J = 8.1 Hz, 1H), 8.03 (s, 2H), 7.79 – 7.70 (m, 1H), 7.24 (d, J = 1.9 Hz, 1H), 7.08 (dd, J = 8.3, 1.9 Hz, 1H), 6.85 (d, J = 8.1 Hz, 1H), 4.07 (d, J = 7.0 Hz, 2H), 1.37 (t, J = 7.0 Hz, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.48, 153.43, 148.88, 147.07, 143.93, 143.70, 138.05, 125.38, 120.83, 115.69, 110.51, 63.80, 14.67; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3546.1, 3431.9, 1585.2, 1439.2, 1279.6; Anal. Calcd. for $C_{17}H_{16}N_4O_2S$: C, 59.98; H, 4.74; N, 16.46; S, 9.42; Found: C, 60.01; H, 4.78; N, 16.42; S, 9.39; HR-MS (ESI+): Calcd. m/z for $C_{17}H_{16}N_4O_2S$: 340.0994; Found m/z $[M+H]^+$: 341.1087.

(E)-2-(2-(3-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(26b)**

Light orange solid from EtOH (86 %). m.p. 219-221 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 12.68 (s, 1H), 8.74 – 8.68 (m, 1H), 8.44 (dd, J = 8.0, 6.1 Hz, 1H), 8.39 (dd, J = 10.7, 9.5 Hz, 1H), 8.30 (d, J = 8.0 Hz, 1H), 8.27 – 8.17 (m, 2H), 8.12 – 8.06 (m, 2H), 7.78 – 7.55 (m, 2H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.01, 167.02, 148.25, 140.26, 135.96, 132.55, 130.49, 124.51, 123.69, 122.70, 120.36, 102.55; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3304.10, 1560.47, 1512.16, 1336.23; Anal. Calcd. for $C_{15}H_{11}N_5O_2S$: C, 55.38; H, 3.41; N, 21.53; S, 9.85; Found: C, 55.44; H, 3.43; N, 21.48; S, 9.82; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}N_5O_2S$: 325.0633; Found m/z $[M+H]^+$: 326.0713.

(E)-2-(2-(4-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole **(27b)**

Orange solid from EtOH (83 %). m.p. 227-229 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 8.62 (d, J = 4.9 Hz, 1H), 8.27 (d, J = 8.8 Hz, 2H), 8.16 (s, 1H), 8.01 (t, J = 5.8 Hz, 2H), 7.89 (d, J = 8.8 Hz, 2H), 7.76 (s, 1H), 7.45 (dd, J = 7.7, 3.4 Hz, 1H); ^{13}C NMR (101 MHz, δ, ppm,

DMSO- d_6): 168.51, 147.30, 146.48, 140.60, 139.32, 127.16, 124.20, 123.80, 121.57, 121.37; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3433.3, 1561.4, 1512.7, 1336.6; Anal. Calcd. for $C_{15}H_{11}N_5O_2S$: C, 55.38; H, 3.41; N, 21.53; S, 9.85; Found: C, 55.45; H, 3.42; N, 21.49; S, 9.81; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{11}N_5O_2S$: 325.0633; Found m/z $[M+H]^+$: 326.0716.

(E)-N,N-dimethyl-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)aniline **(28b)**

Light brown solid from EtOH (82 %). m.p. 174-176 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO- d_6): 8.66 (dd, J = 5.3, 0.7 Hz, 1H), 8.22 (dd, J = 17.7, 7.6 Hz, 2H), 8.02 (s, 1H), 7.88 (s, 1H), 7.63 (t, J = 6.3 Hz, 1H), 7.50 (d, J = 8.9 Hz, 2H), 6.78 (d, J = 8.9 Hz, 2H), 2.97 (s, 6H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 169.28, 150.97, 148.28, 145.80, 143.71, 127.85, 124.03, 122.11, 112.35, 105.38, 40.08; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3428.9, 2923.3, 1600.3, 1574.5, 1437.6; Anal. Calcd. for $C_{17}H_{17}N_5S$: C, 63.13; H, 5.30; N, 21.65; S, 9.91; Found: C, 63.15; H, 5.32; N, 21.64; S, 9.89; HR-MS (ESI+): Calcd. m/z for $C_{17}H_{17}N_5S$: 323.1205; Found m/z $[M+H]^+$: 324.1292.

(E)-4-(pyridin-2-yl)-2-(2-(pyridin-2-ylmethylene)hydrazinyl)thiazole **(29b)**

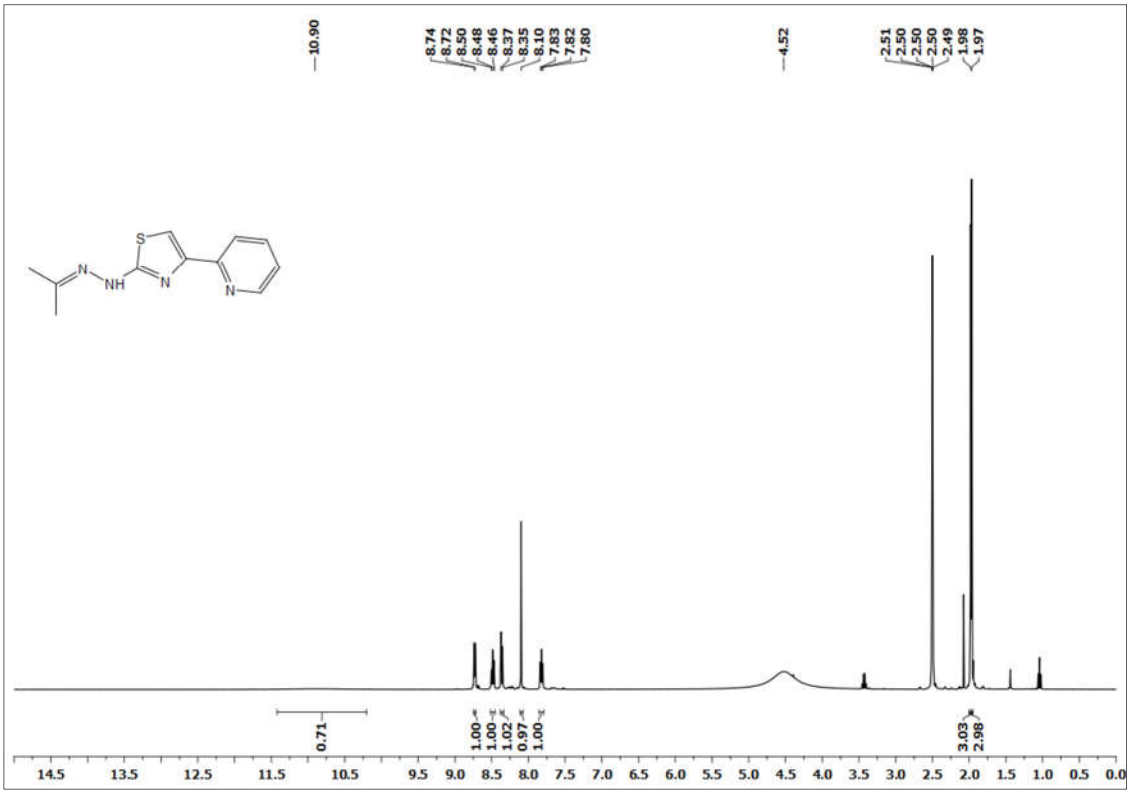
Light yellow solid from EtOH (87 %). m.p. 195-197 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO- d_6): 12.48 (s, 1H), 8.57 (ddd, J = 4.8, 2.9, 1.4 Hz, 2H), 8.07 (s, 1H), 7.90 – 7.83 (m, 4H), 7.59 (s, 1H), 7.35 (ddd, J = 6.8, 4.9, 2.1 Hz, 1H), 7.30 (ddd, J = 6.8, 4.8, 2.0 Hz, 1H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 168.12, 153.22, 152.21, 150.93, 149.51, 149.48, 141.75, 137.30, 136.84, 123.74, 122.74, 120.05, 119.26, 108.04; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3434.3, 1625.3, 1569.8, 1475.0; Anal. Calcd. for $C_{14}H_{11}N_5S$: C, 59.77; H, 3.94; N, 24.89; S, 11.40; Found: C, 59.84; H, 3.96; N, 24.84; S, 11.36; HR-MS (ESI+): Calcd. m/z for $C_{14}H_{11}N_5S$: 281.0735; Found m/z $[M+H]^+$: 282.0786.

(E)-4-(pyridin-2-yl)-2-(2-(1-(pyridin-3-yl)ethylidene)hydrazinyl)thiazole **(30b)**

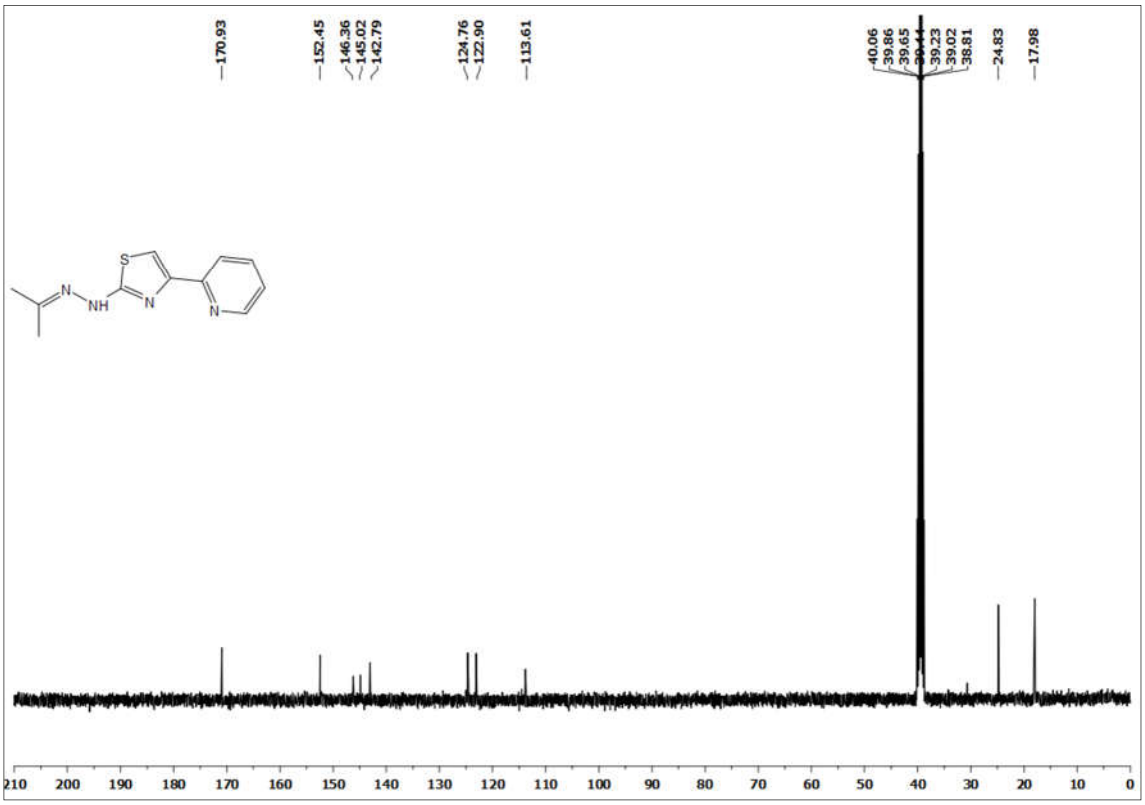
Greenish yellow solid from EtOH (84 %). m.p. 185-187 ºC; ^{1}H NMR (400 MHz, δ, ppm, DMSO-d_6): 9.13 (d, J = 2.0 Hz, 1H), 8.86 (d, J = 5.5 Hz, 1H), 8.78 – 8.73 (m, 1H), 8.70 (dd, J = 4.9, 1.2 Hz, 1H), 8.29 – 8.17 (m, 1H), 8.09 – 7.95 (m, 3H), 7.67 – 7.48 (m, 1H), 2.43 (d, J = 9.3 Hz, 3H); ^{13}C NMR (101 MHz, δ, ppm, DMSO- d_6): 168.16, 156.96, 149.57, 146.07, 141.15, 124.56, 124.27, 122.04, 121.56, 116.04; IR (KBr, $\upsilon_{max,}$ cm^{-1}): 3373.0, 3027.7, 1617.5, 1575.2, 1451.0; Anal. Calcd. for $C_{15}H_{13}N_5S$: C, 61.00; H, 4.44; N, 23.71; S, 10.85; Found: C, 61.03; H, 4.45; N, 23.69; S, 10.83; HR-MS (ESI+): Calcd. m/z for $C_{15}H_{13}N_5S$: 295.0892; Found m/z $[M+H]^+$: 296.0970.

2. ^{1}H, ^{13}C NMR, Mass and IR Spectra of the compounds 1b to 30b

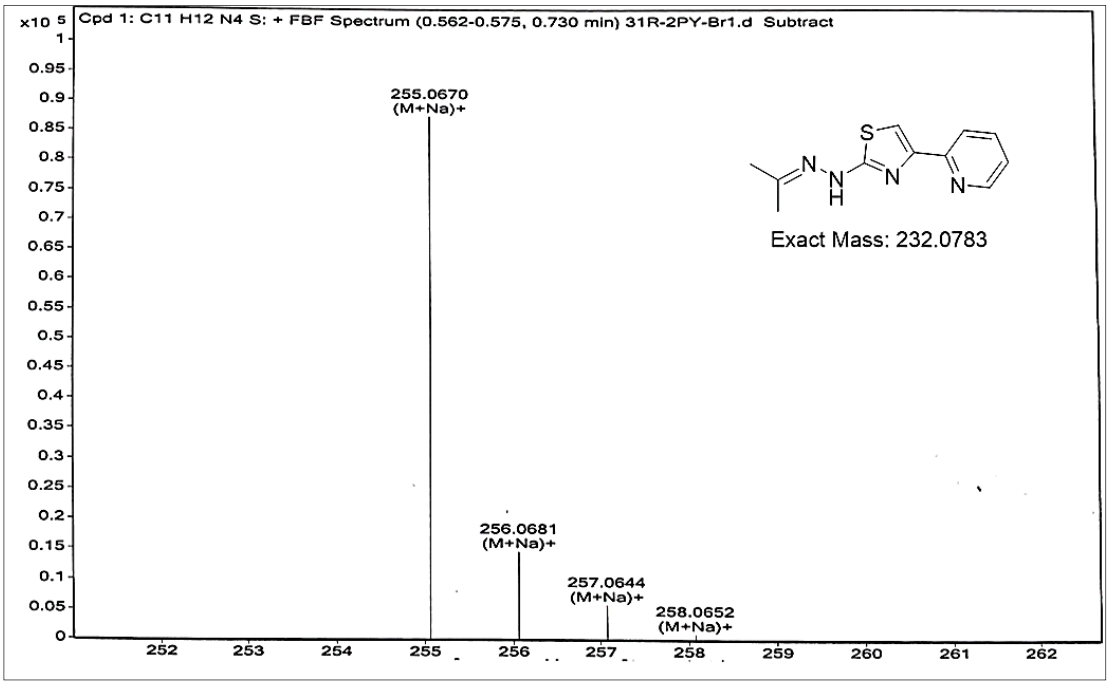
^{1}H NMR Spectrum of 2-(2-(propan-2-ylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(1b)**



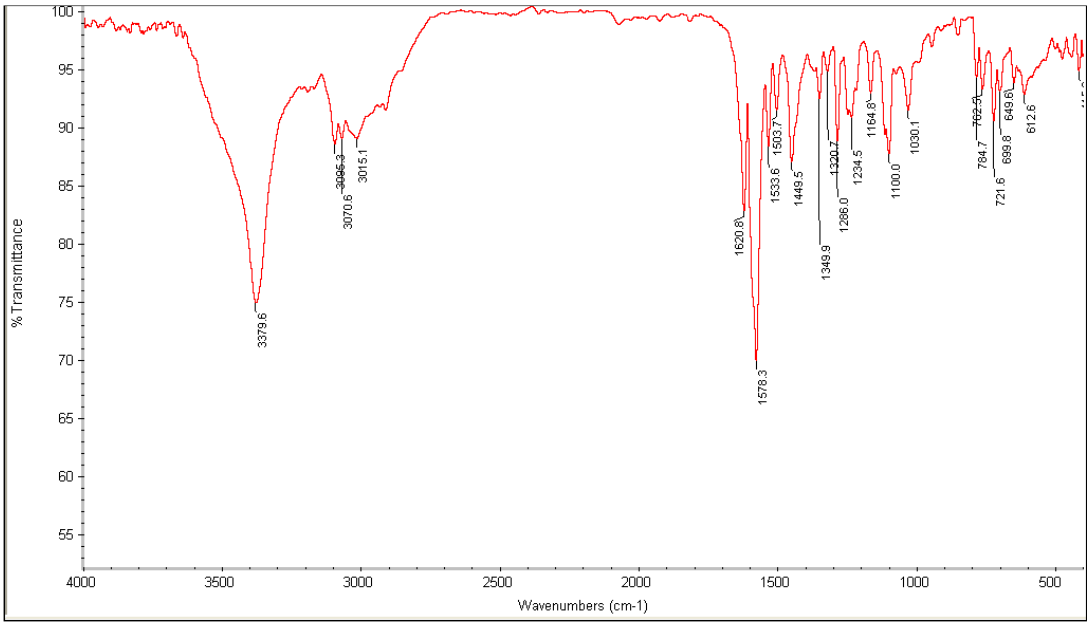
^{13}C NMR Spectrum of 2-(2-(propan-2-ylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(1b)**



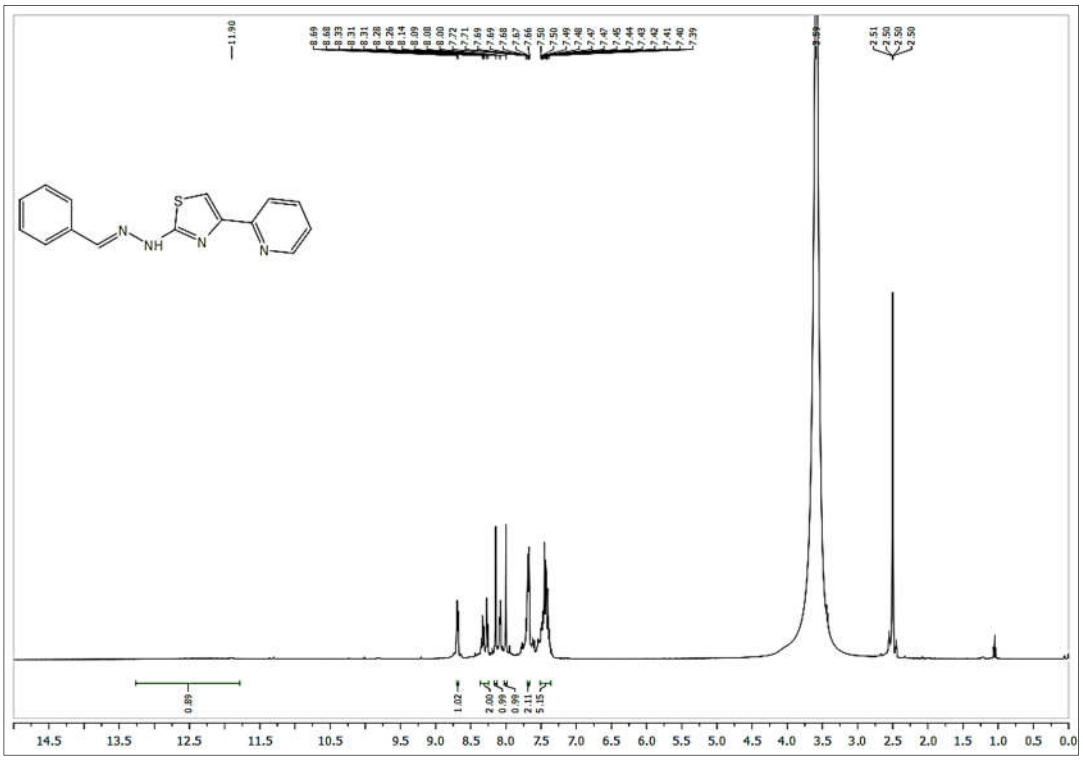
Mass Spectrum of 2-(2-(propan-2-ylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**1b**)



IR Spectrum of 2-(2-(propan-2-ylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(1b)**



^{1}H NMR Spectrum of (E)-2-(2-benzylidenehydrazinyl)-4-(pyridin-2-yl)thiazole, (**2b**)



Mass Spectrum of (E)-2-(2-benzylidenehydrazinyl)-4-(pyridin-2-yl)thiazole, (**2b**)

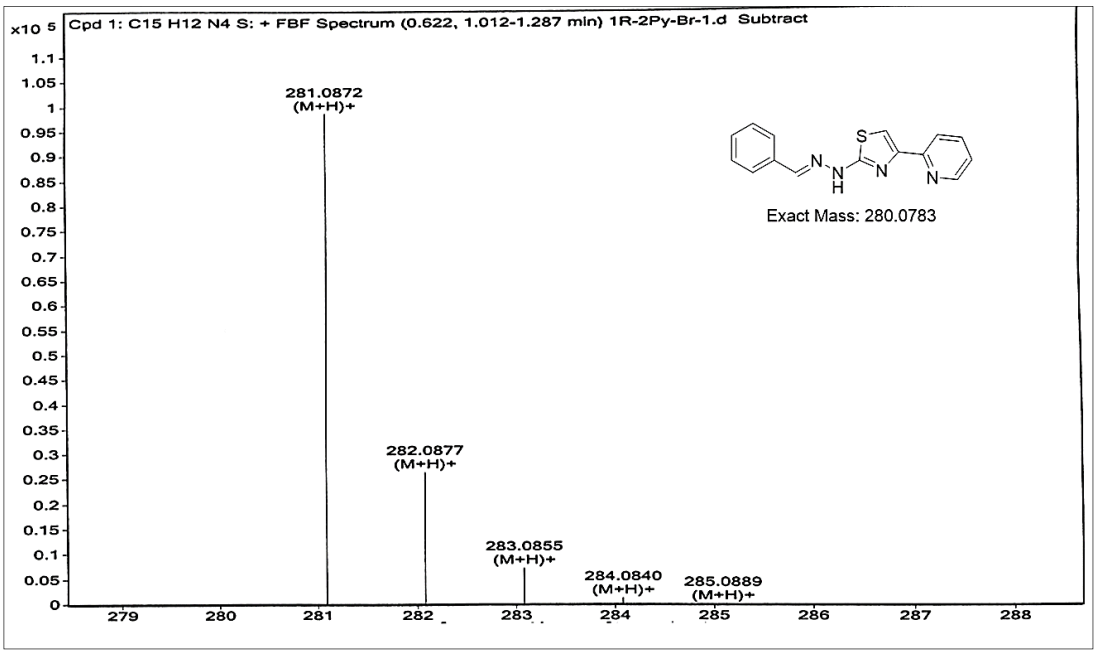
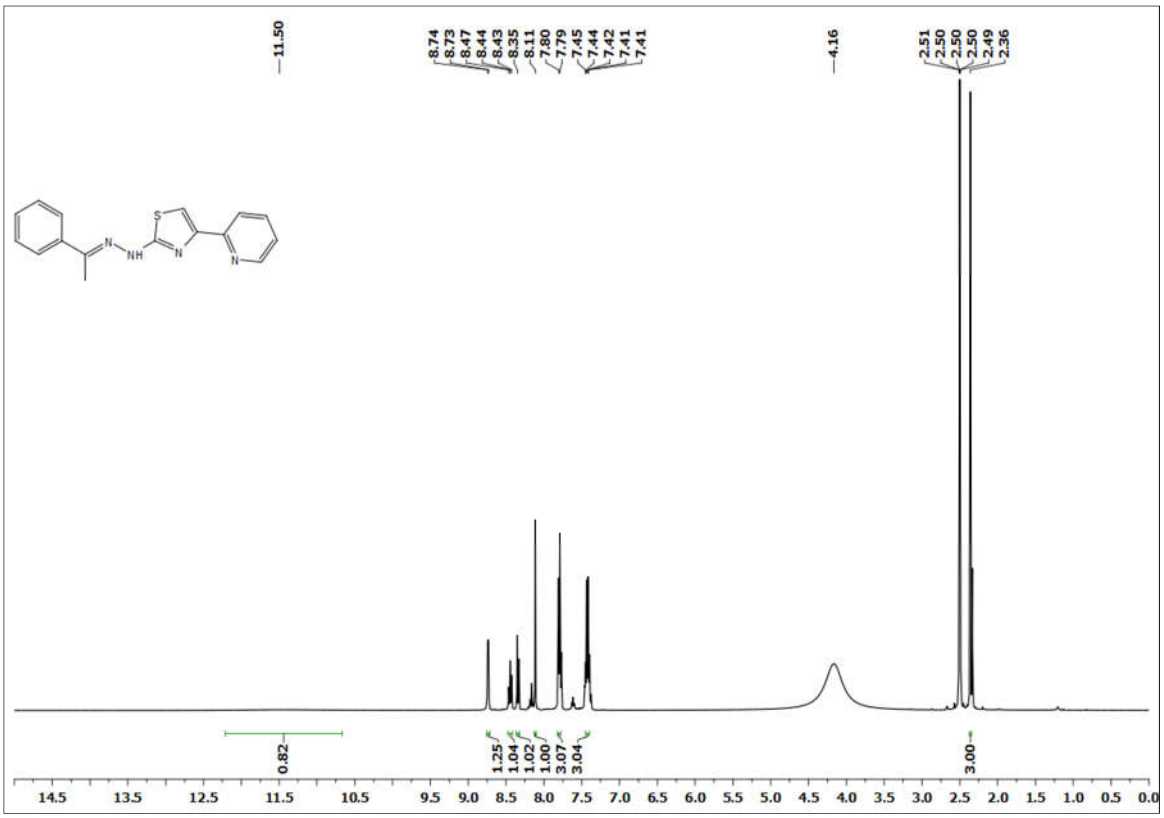
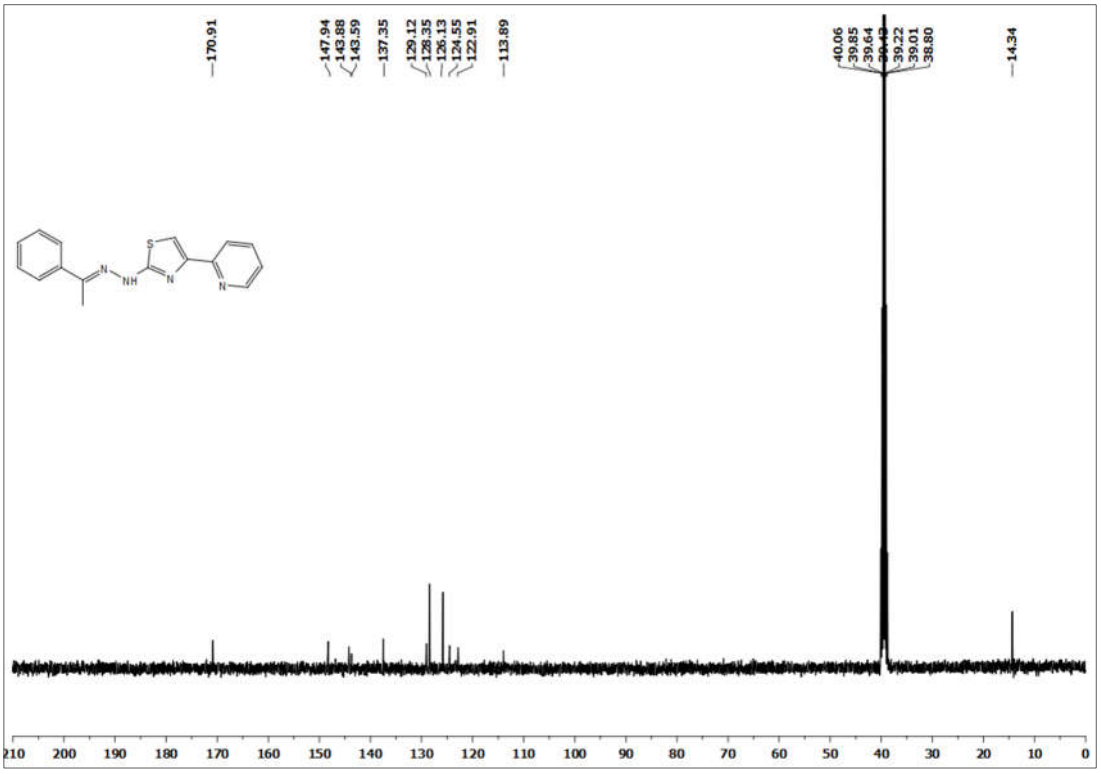


Figure 4.8. IR Spectrum of (E)-2-(2-benzylidenehydrazinyl)-4-(pyridin-2-yl)thiazole, (**2b**)

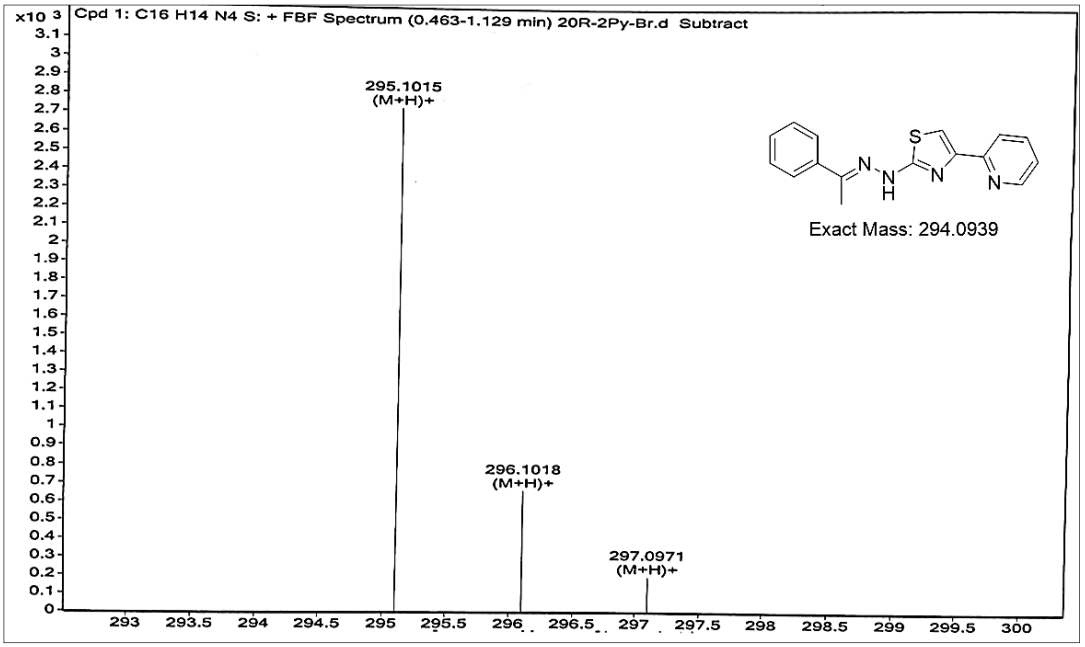
^{1}H NMR Spectrum of (E)-2-(2-(1-phenylethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(3b)**



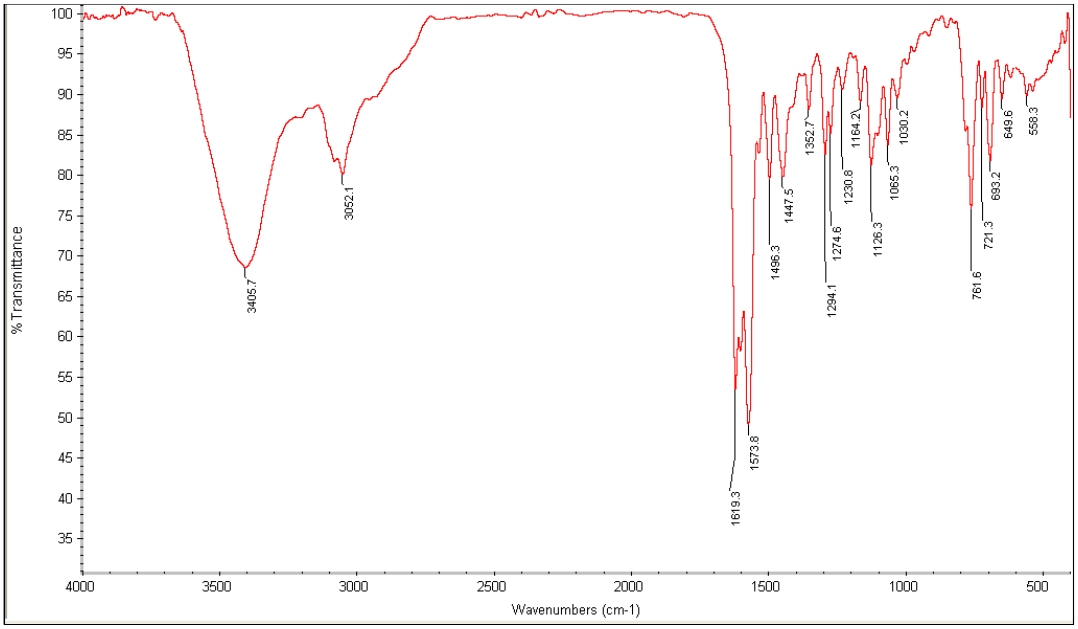
^{13}C NMR Spectrum of (E)-2-(2-(1-phenylethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(3b)**



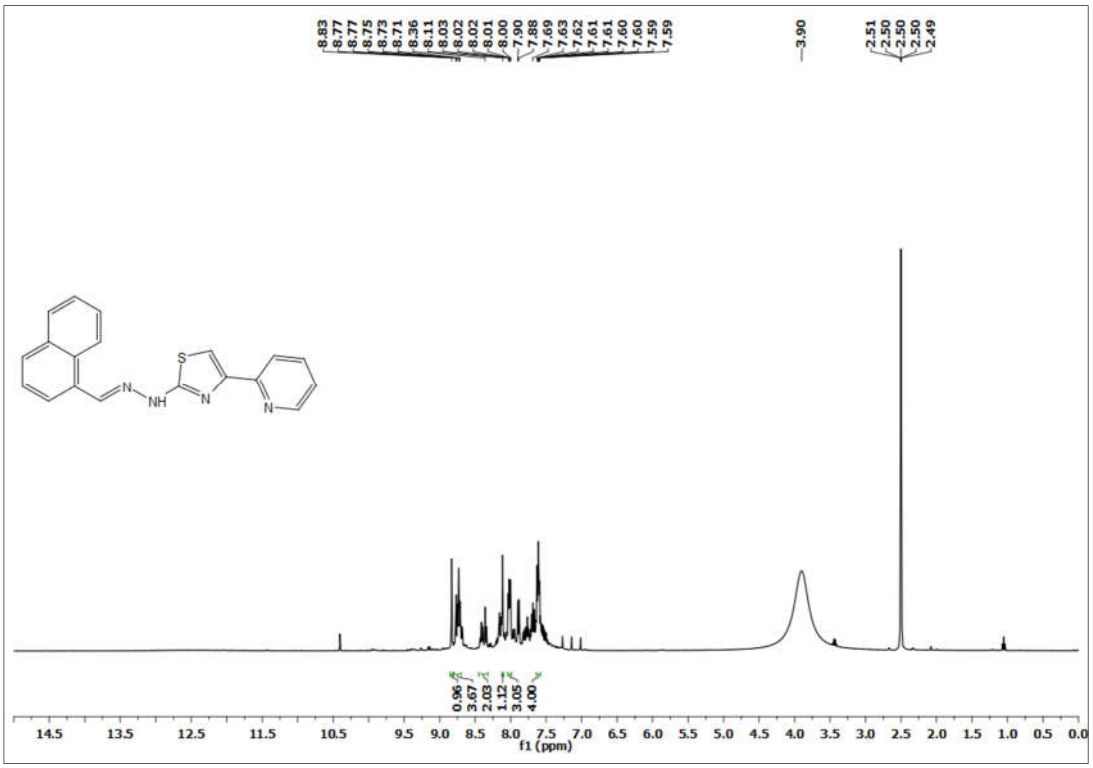
Mass Spectrum of (E)-2-(2-(1-phenylethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**3b)**



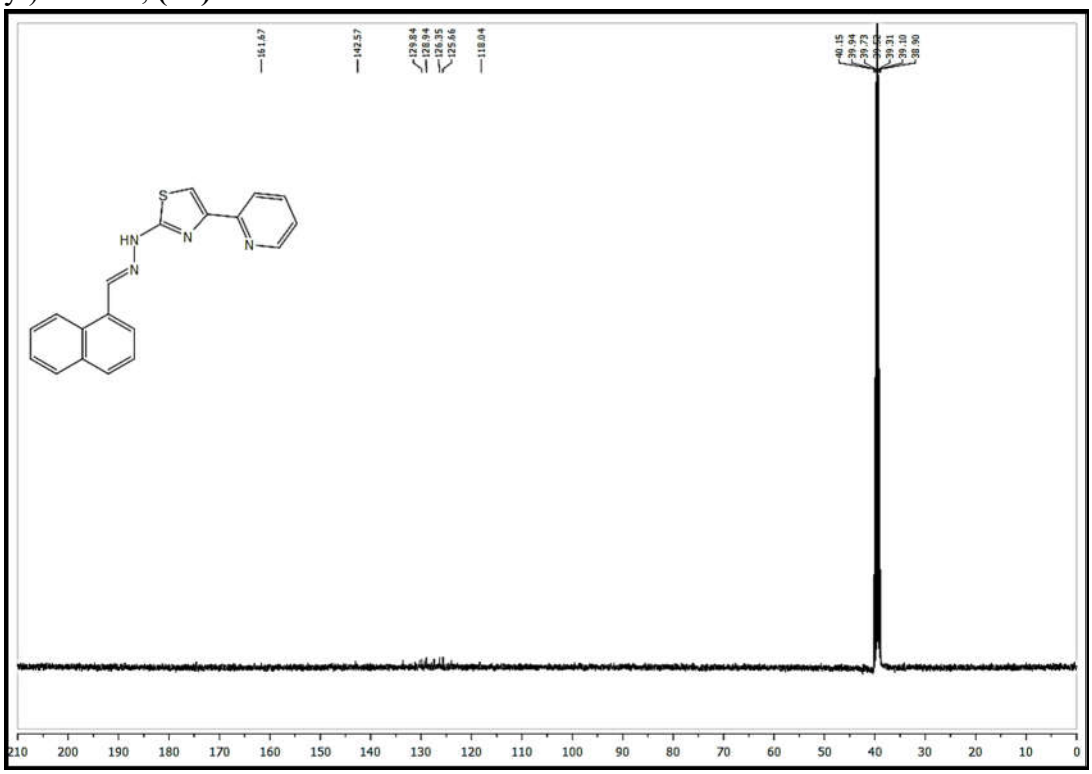
IR Spectrum of (E)-2-(2-(1-phenylethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(3b)**



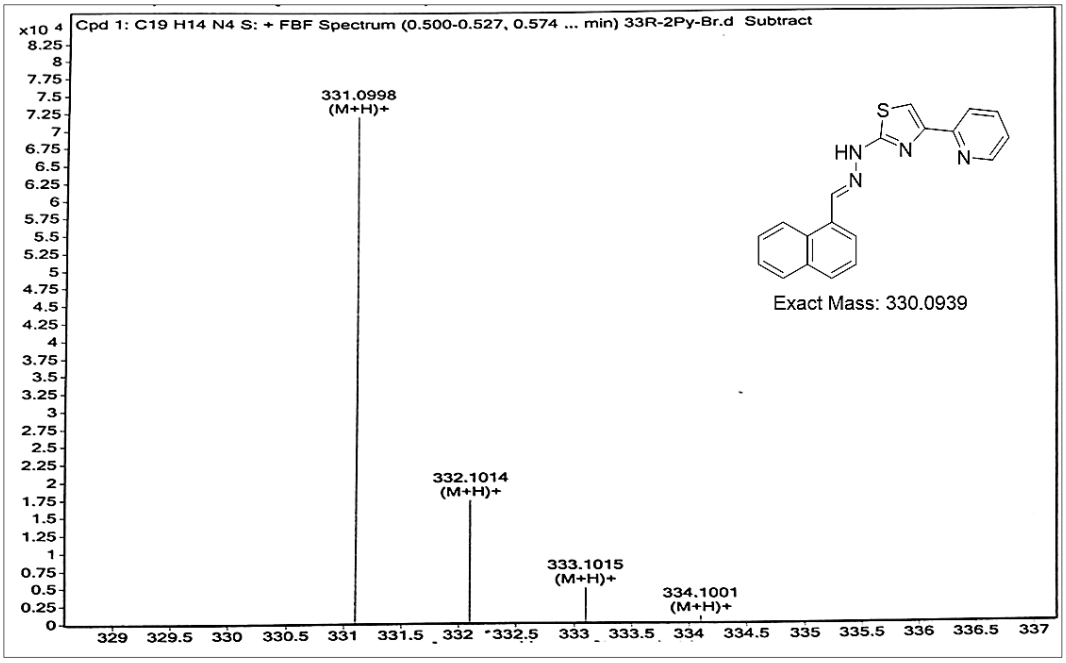
^{1}H NMR Spectrum of (E)-2-(2-(naphthalen-1-ylmethylene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(4b)**



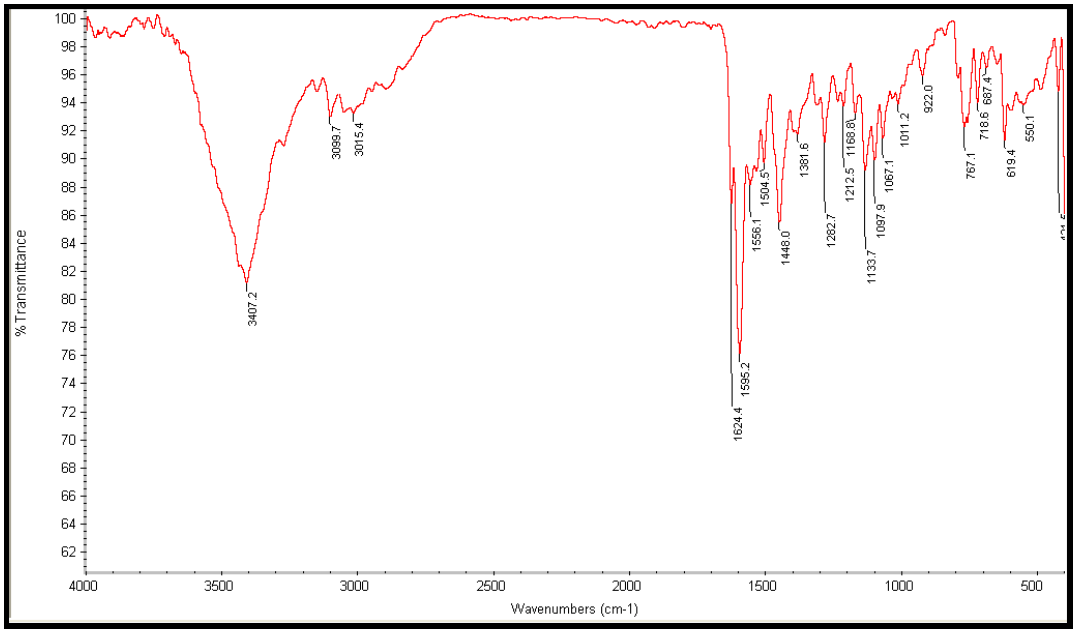
^{13}C NMR Spectrum of (E)-2-(2-(naphthalen-1-ylmethylene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(4b)**



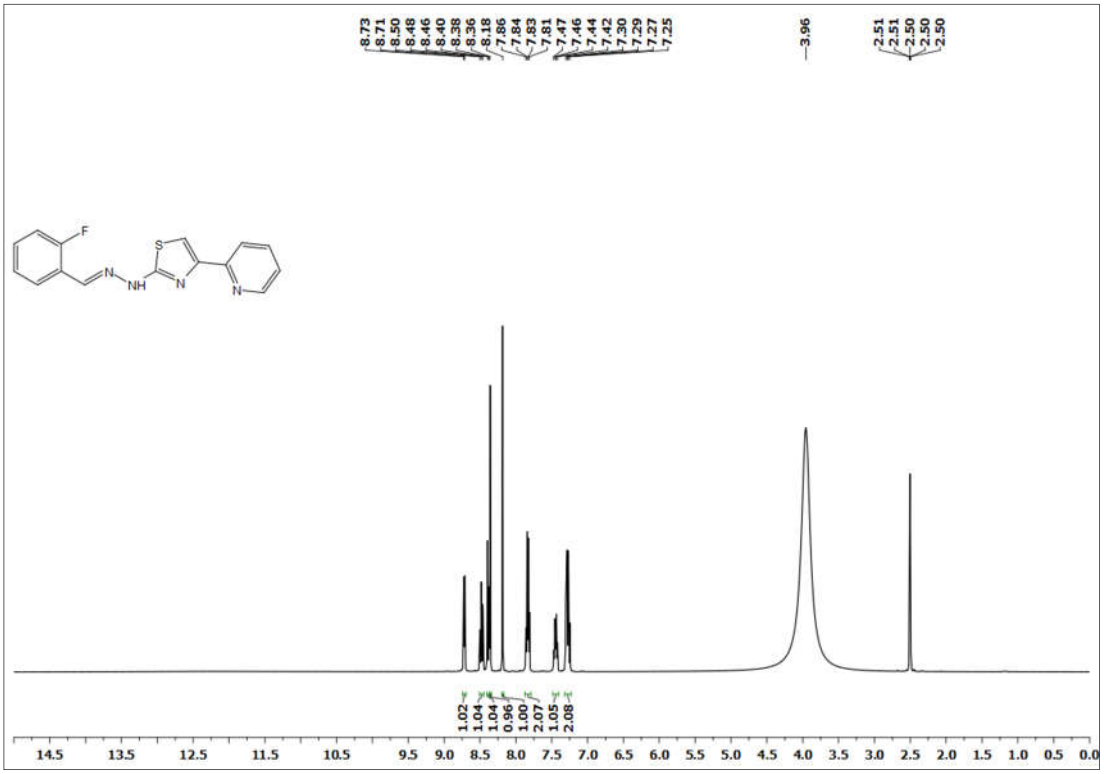
Mass Spectrum of (E)-2-(2-(naphthalen-1-ylmethylene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**4b**)



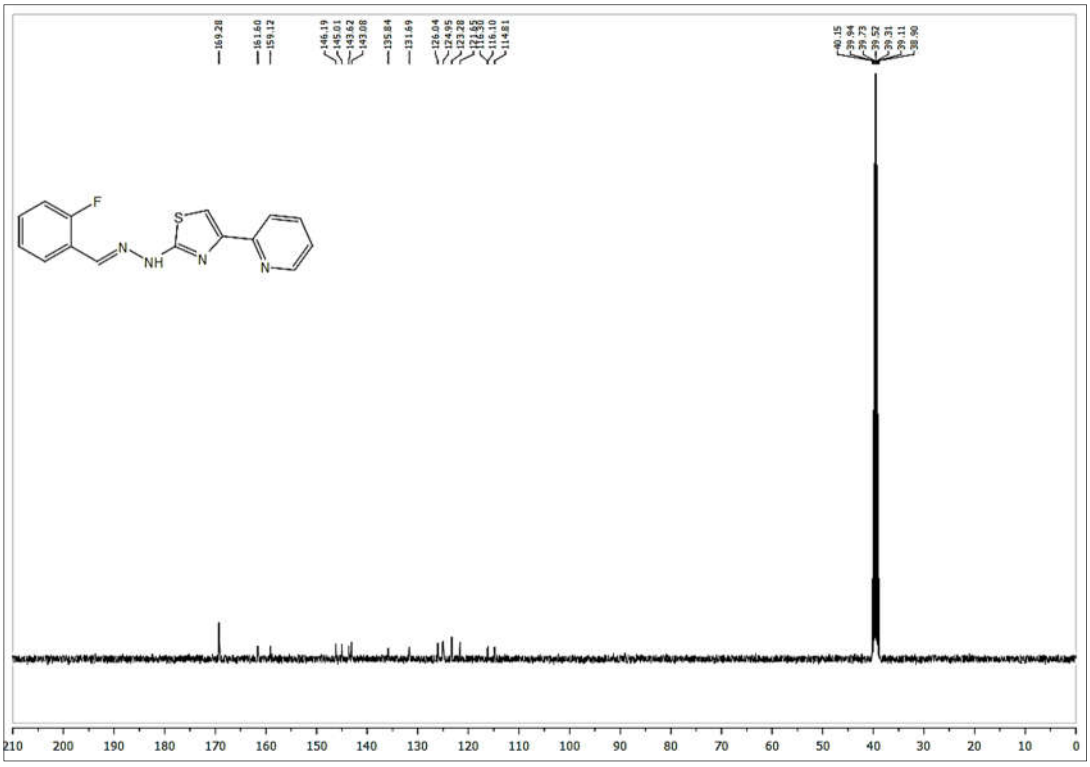
IR Spectrum of (E)-2-(2-(naphthalen-1-ylmethylene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(4b)**



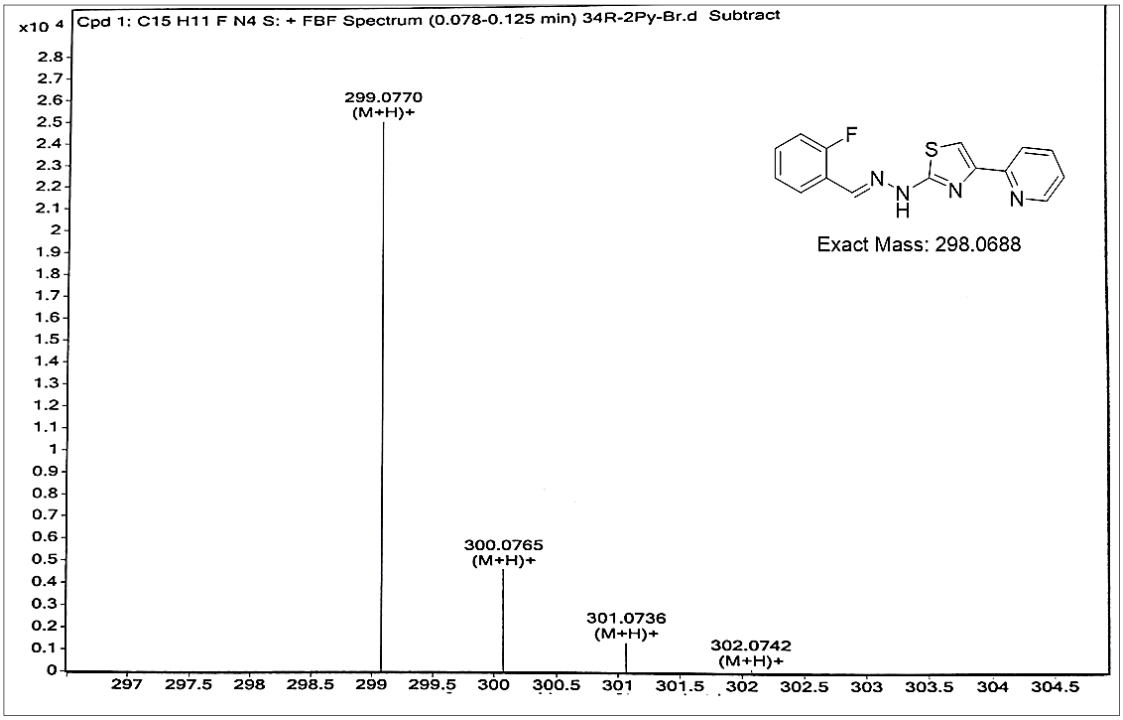
^{1}H NMR Spectrum of (E)-2-(2-(2-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(5b)**



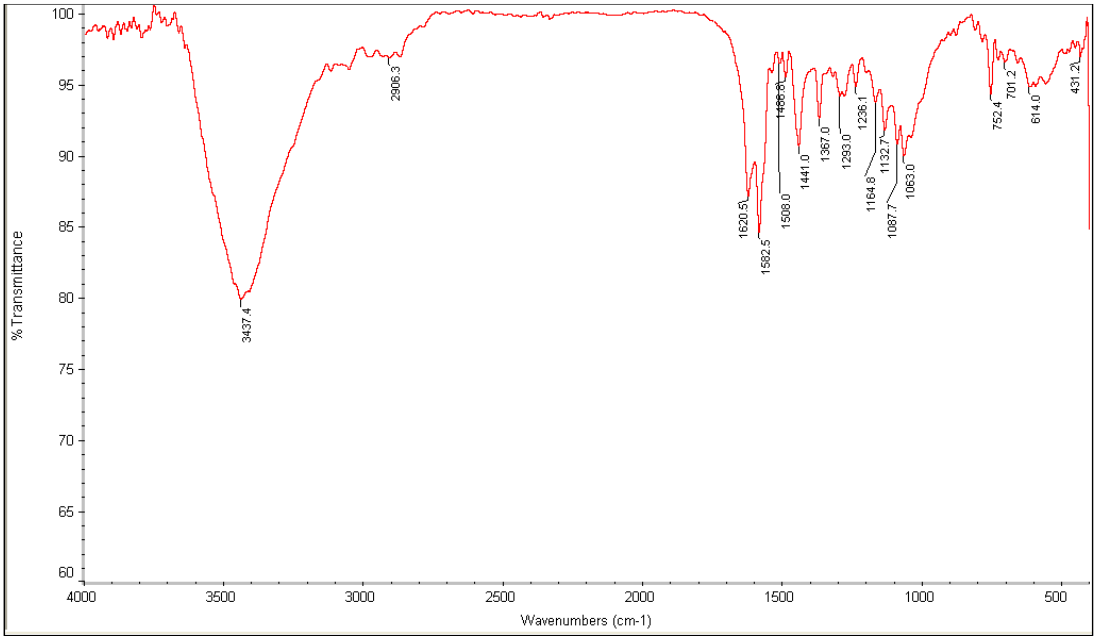
^{13}C NMR Spectrum of (E)-2-(2-(2-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(5b)**



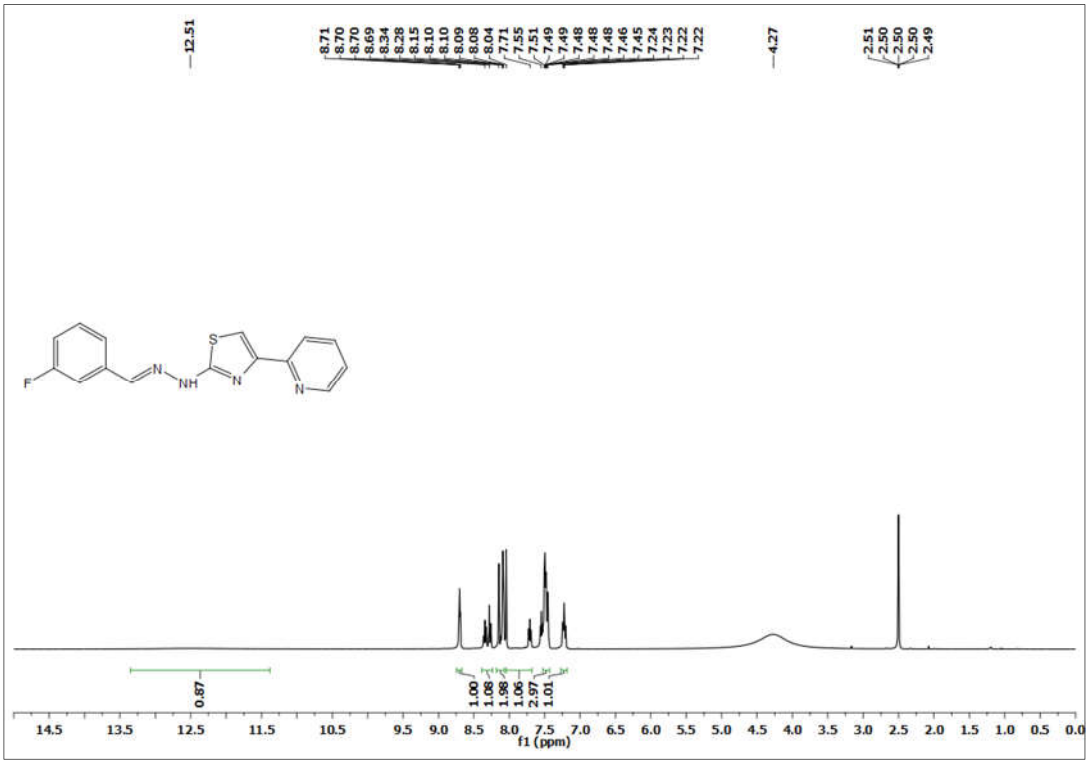
Mass Spectrum of (E)-2-(2-(2-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**5b**)



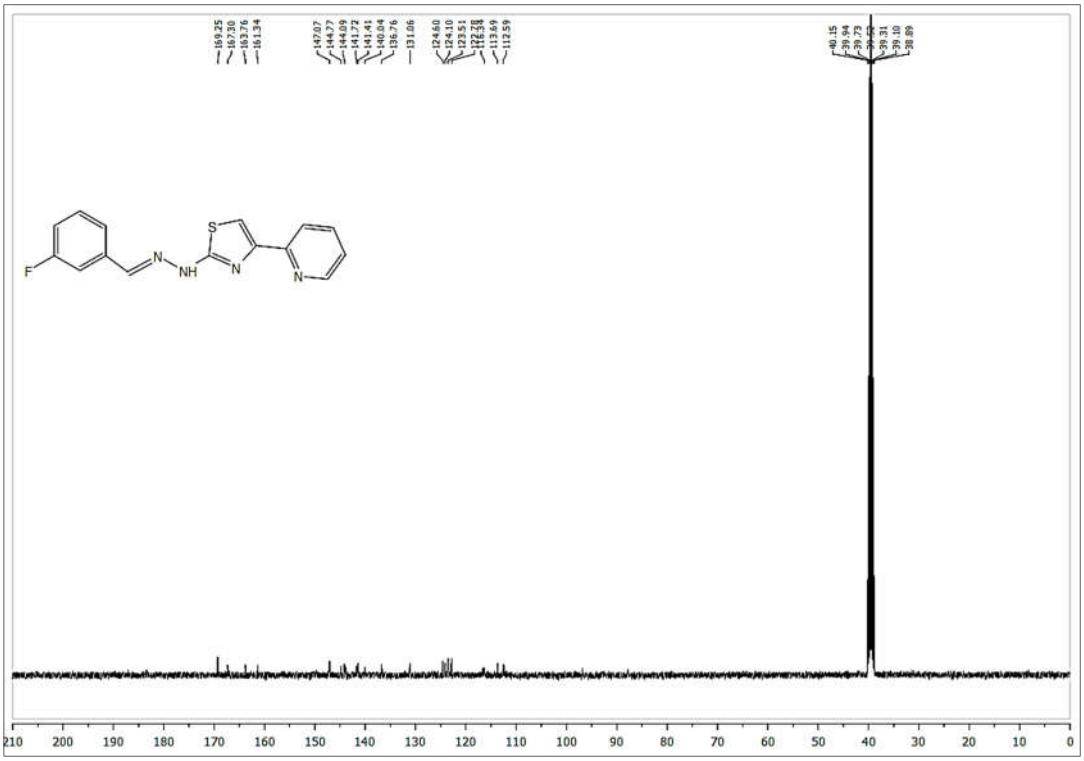
IR Spectrum of (E)-2-(2-(2-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(5b)**



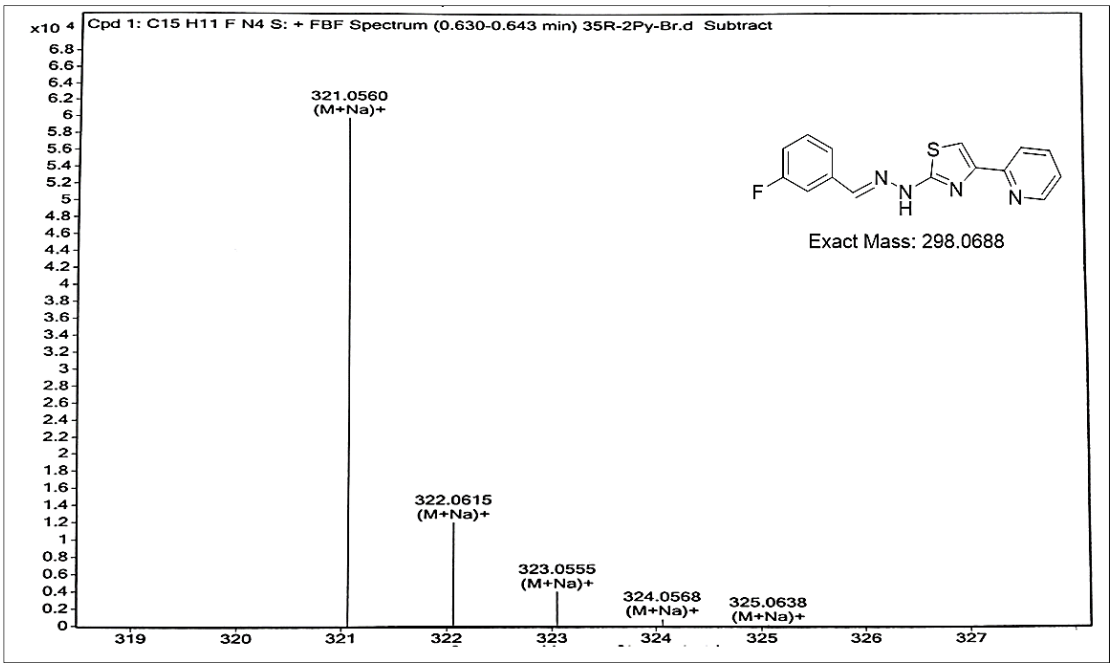
^{1}H NMR Spectrum of (E)-2-(2-(3-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(6b)**



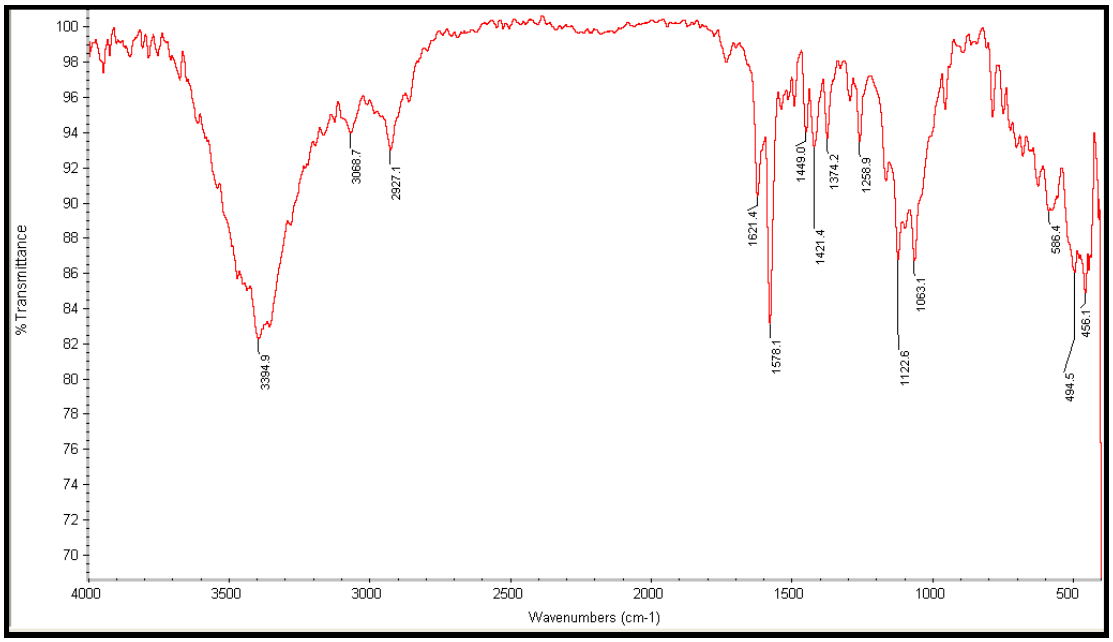
^{13}C NMR Spectrum of (E)-2-(2-(3-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(6b)**



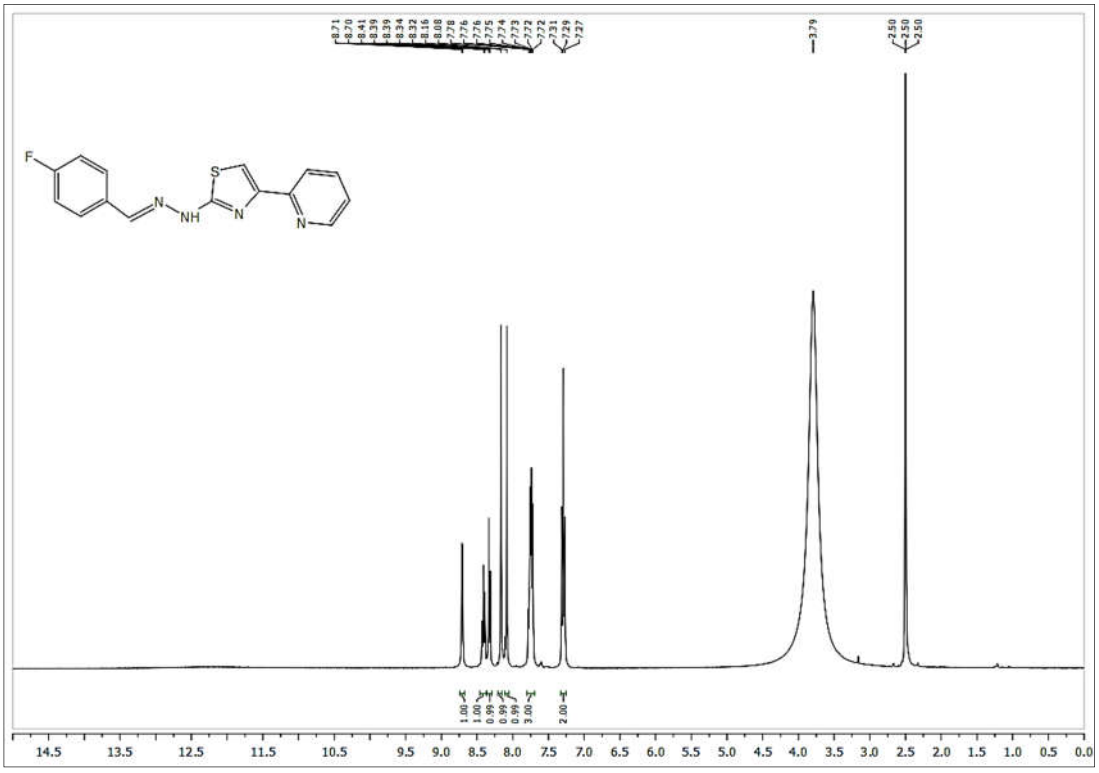
Mass Spectrum of (E)-2-(2-(3-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**6b**)



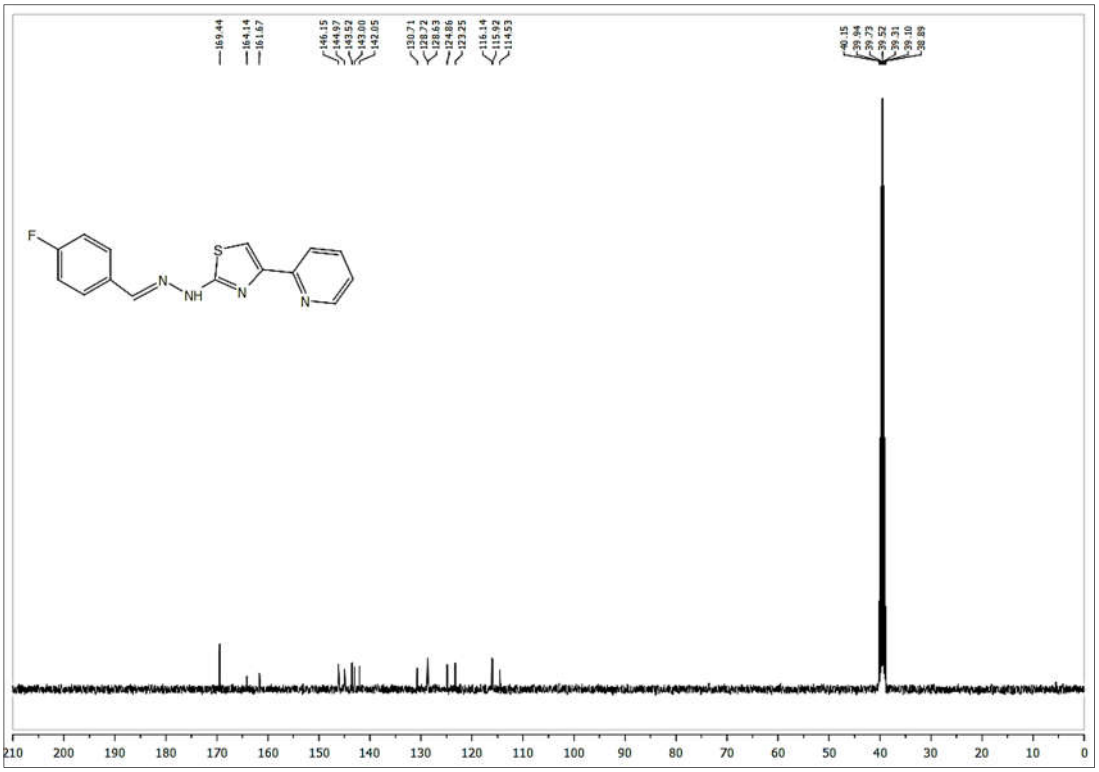
IR Spectrum of (E)-2-(2-(3-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(6b)**



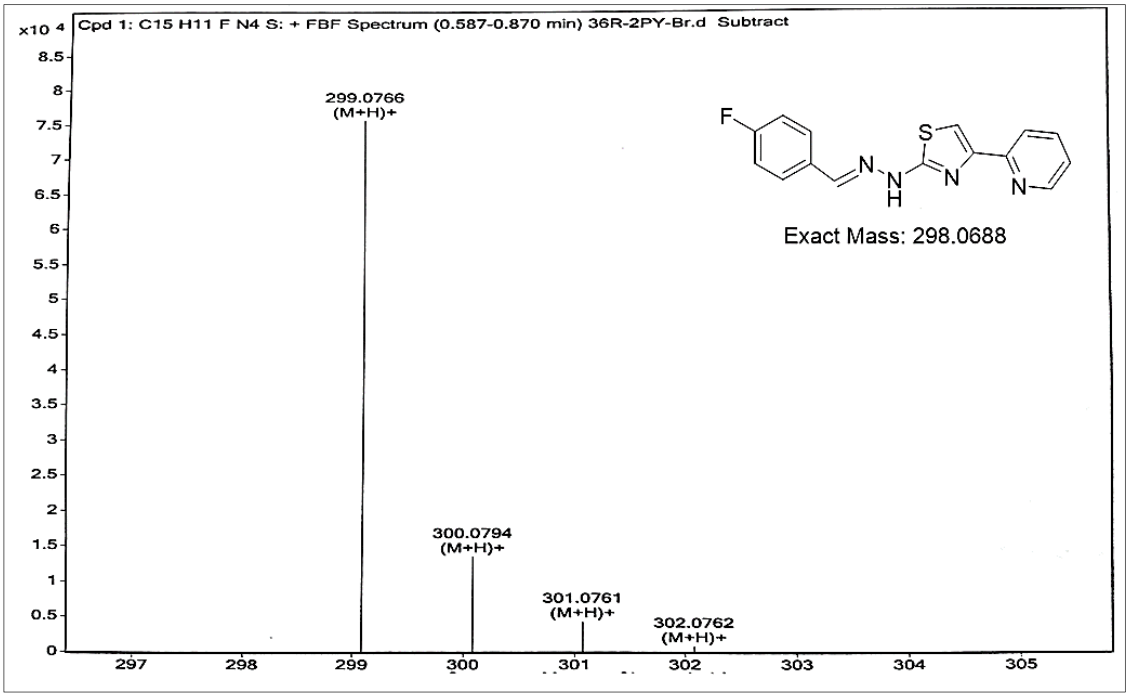
^{1}H NMR Spectrum of (E)-2-(2-(4-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(7b)**



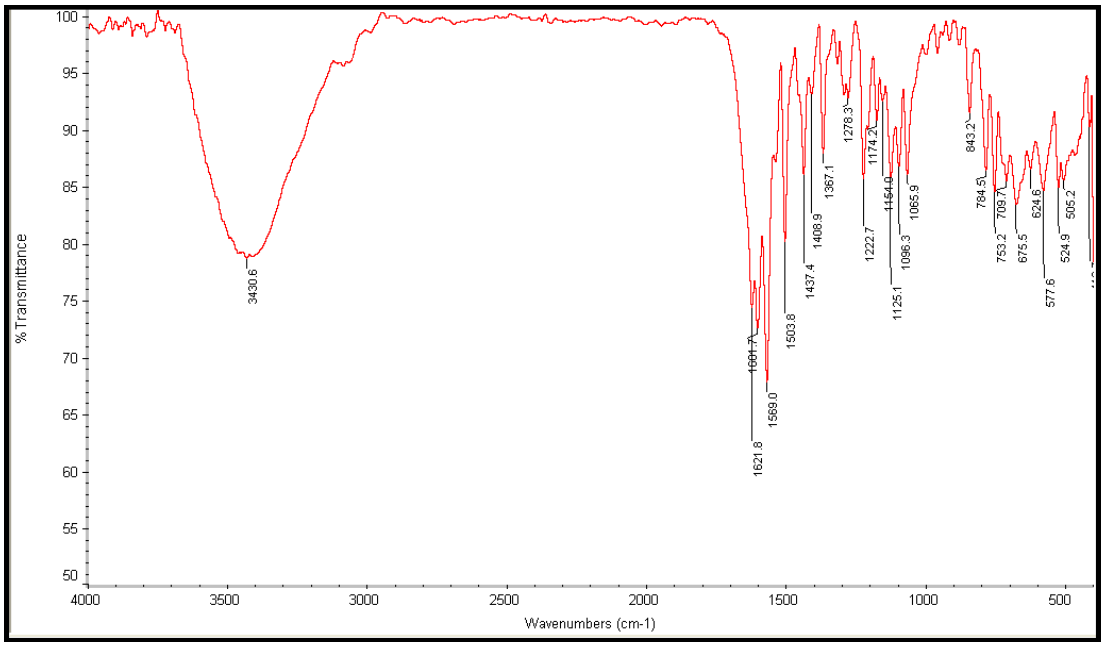
^{13}C NMR Spectrum of (E)-2-(2-(4-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(7b)**



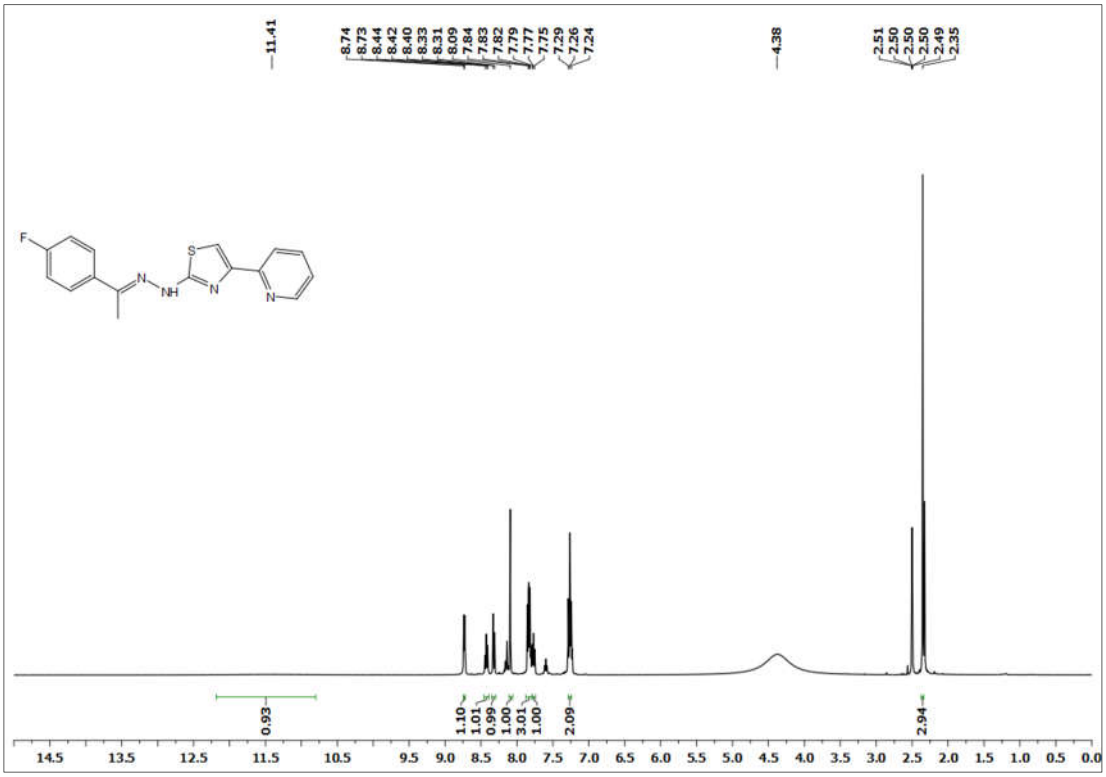
Mass Spectrum of (E)-2-(2-(4-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**7b**)



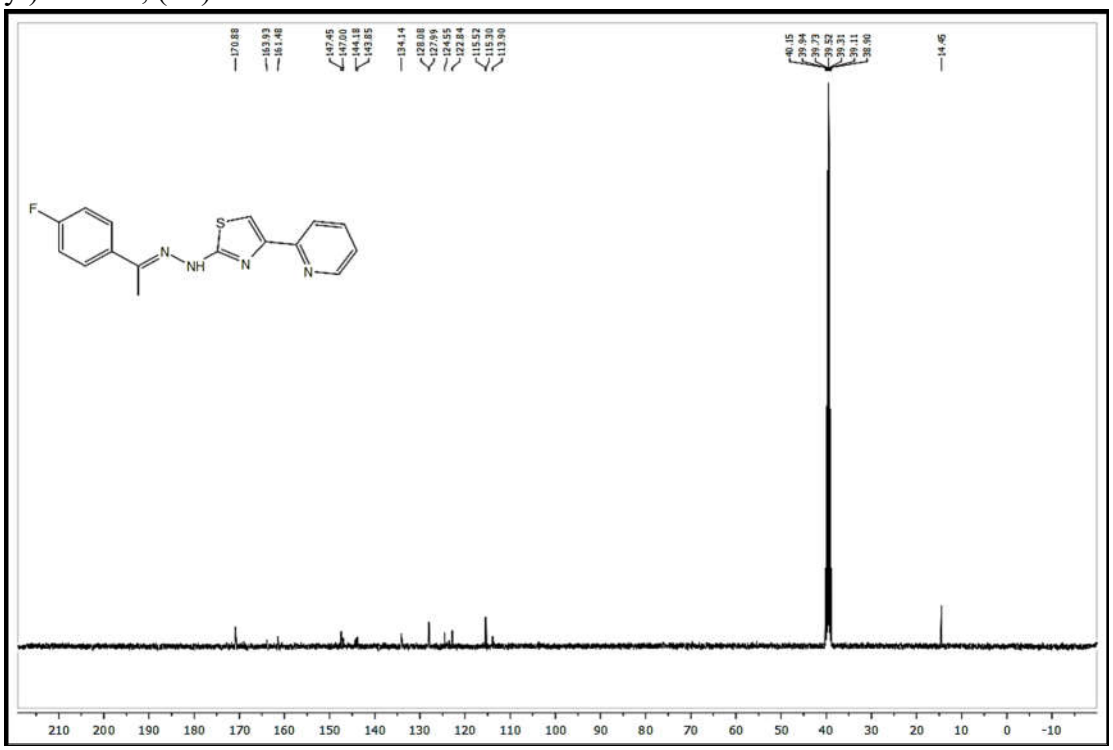
IR Spectrum of (E)-2-(2-(4-fluorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(7b)**



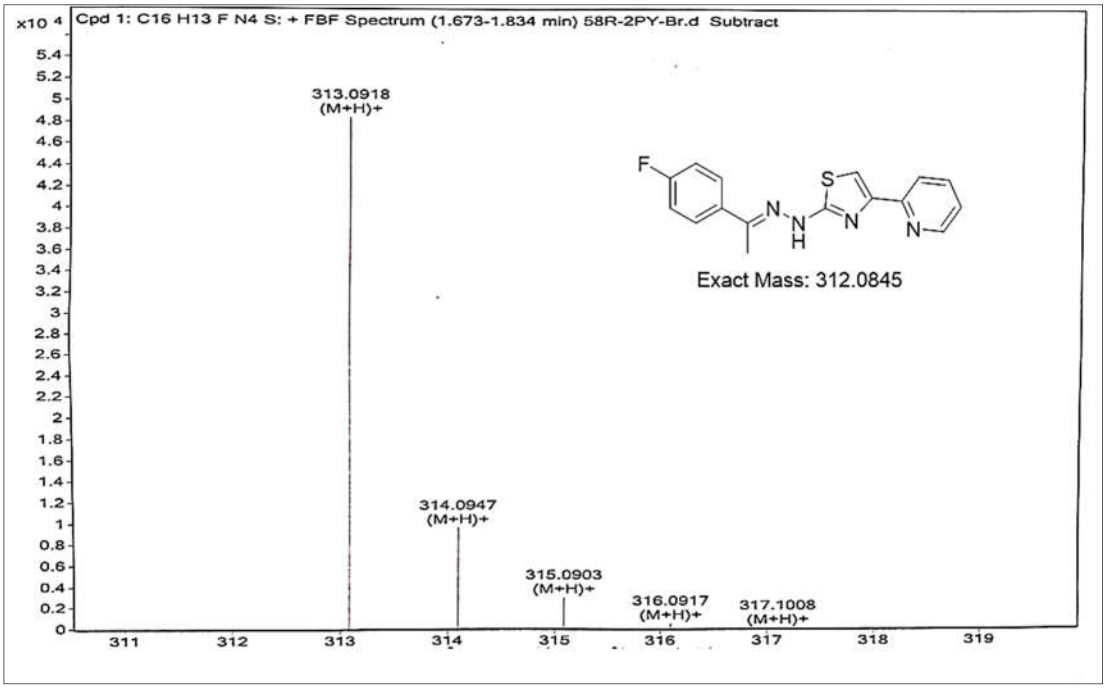
^{1}H NMR Spectrum of (E)-2-(2-(1-(4-fluorophenyl)ethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, (**8b**)



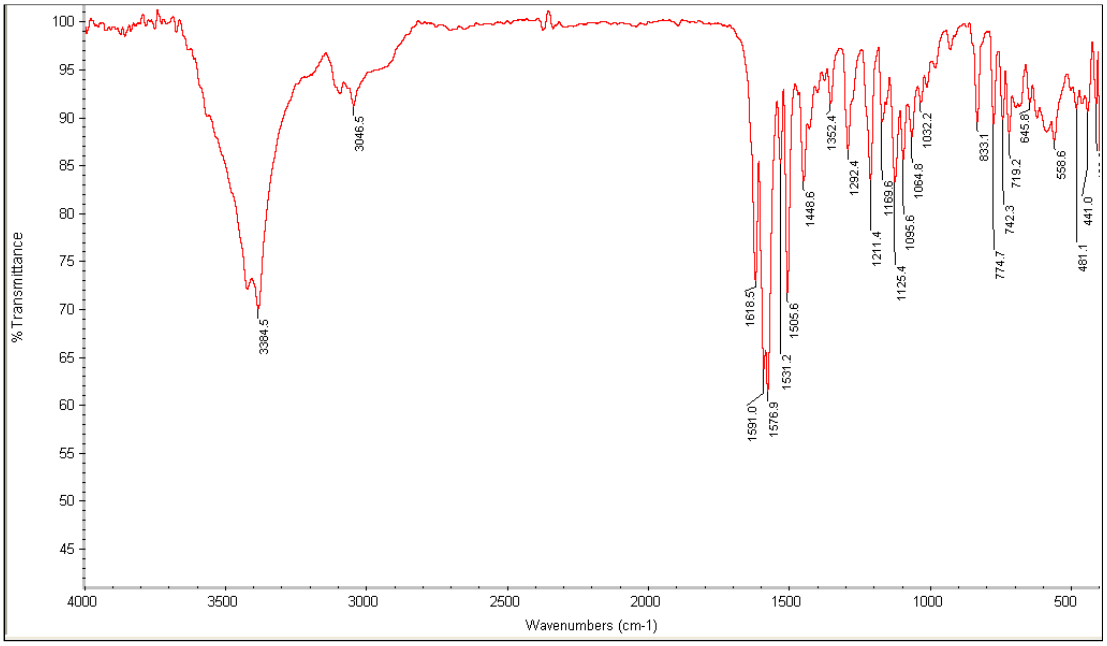
^{13}C NMR Spectrum of (E)-2-(2-(1-(4-fluorophenyl)ethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, (**8b**)



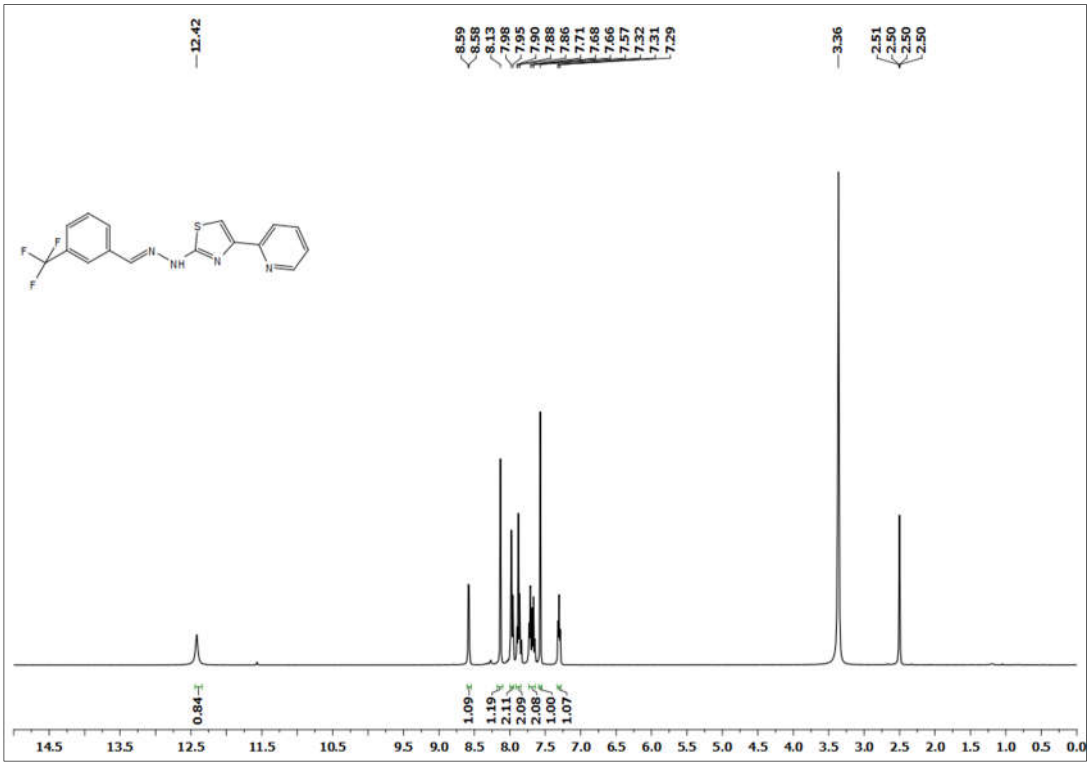
Mass Spectrum of (E)-2-(2-(1-(4-fluorophenyl)ethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**8b**)



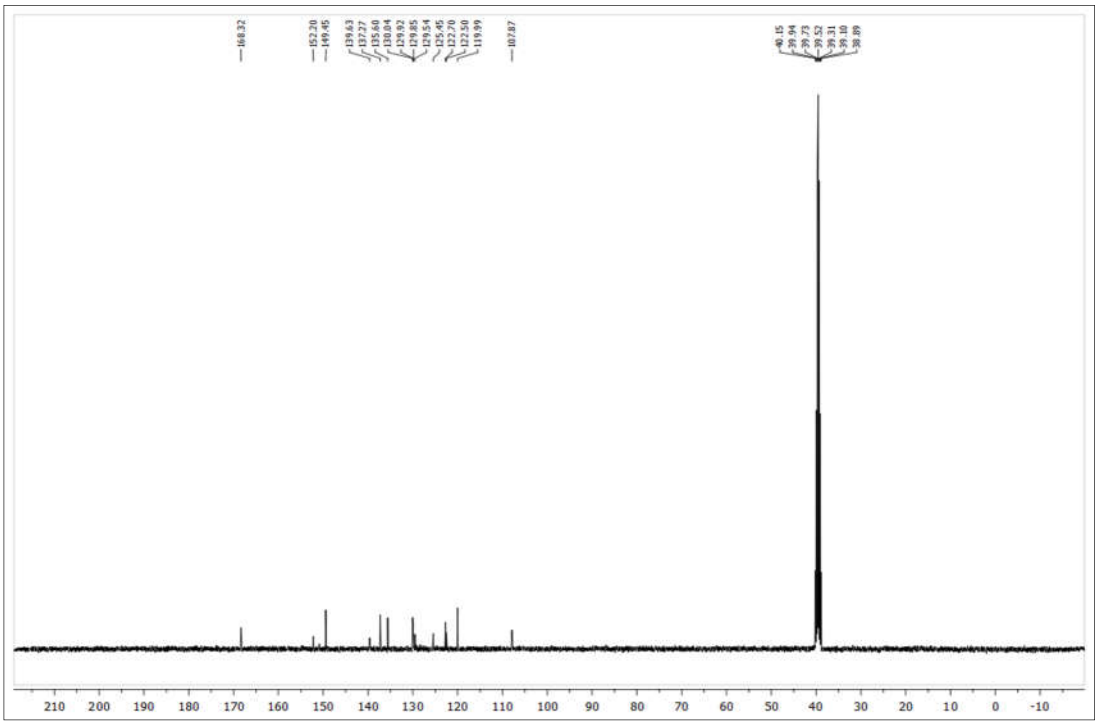
IR Spectrum of (E)-2-(2-(1-(4-fluorophenyl)ethylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, (**8b**)



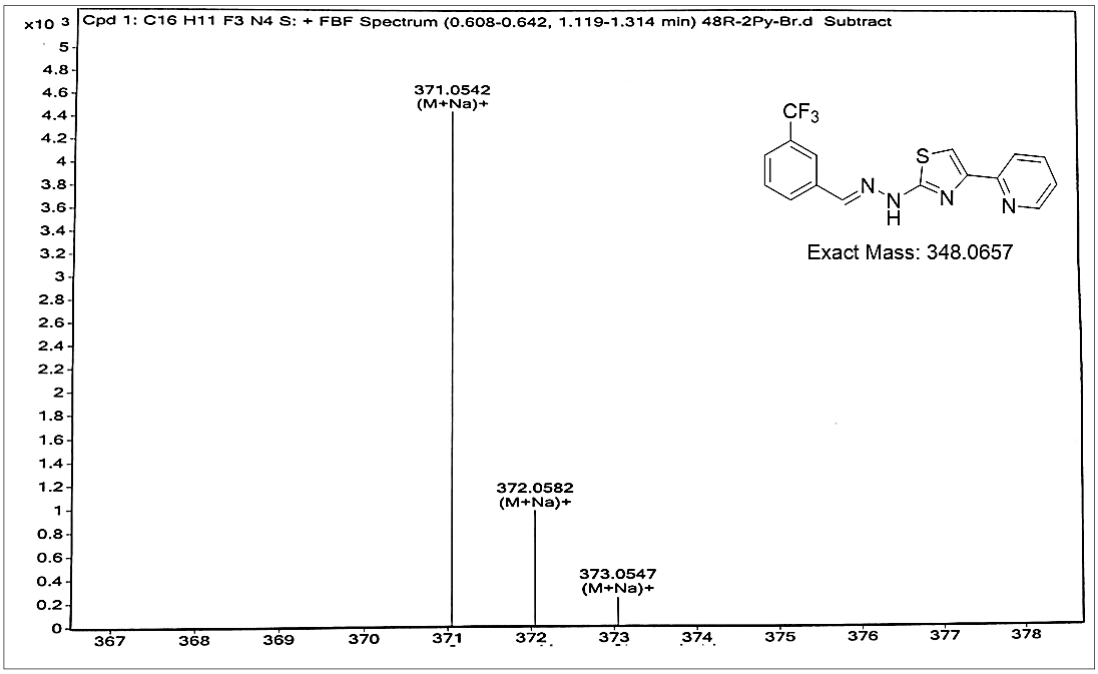
^{1}H NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(3-(trifluoromethyl)benzylidene)hydrazinyl) thiazole, (**9b**)



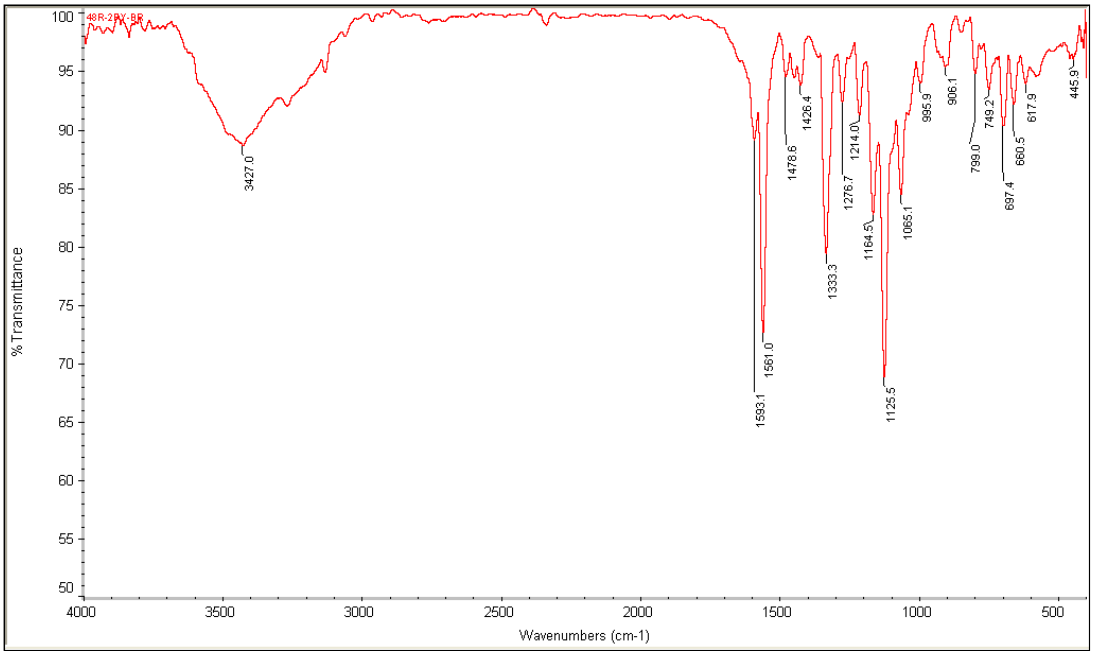
^{13}C NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(3-(trifluoromethyl) enzylidene) hydrazinyl)thiazole, (**9b**)



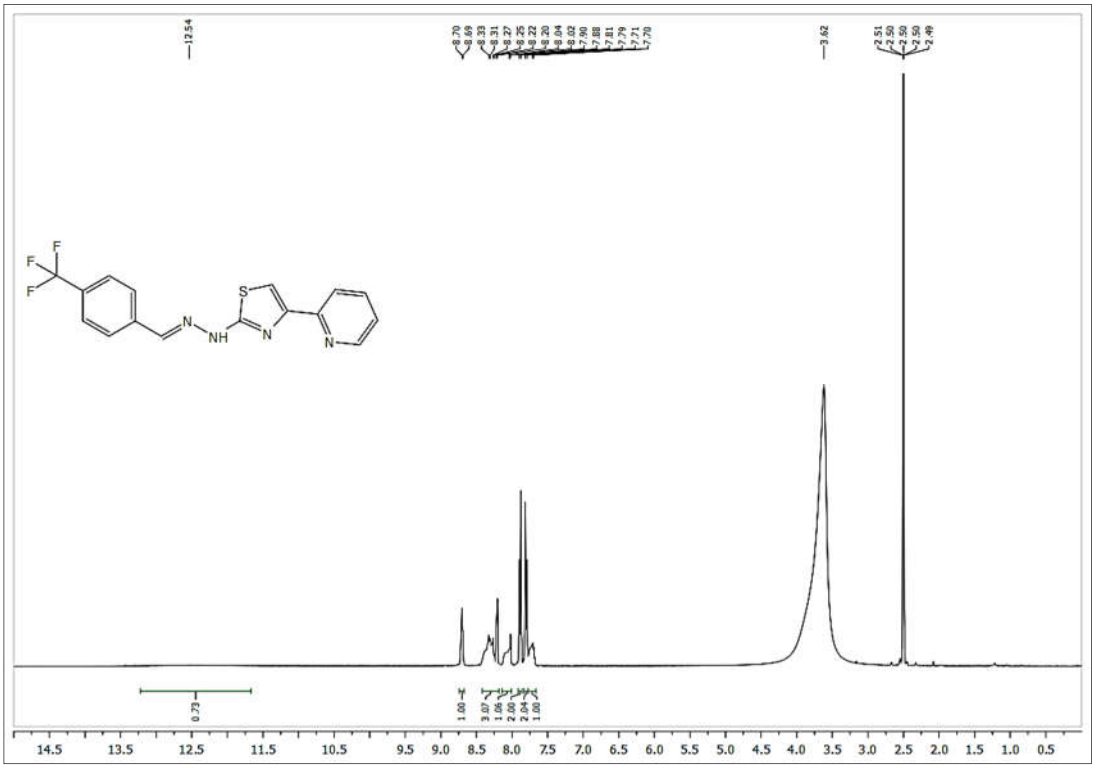
Mass Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(3-(trifluoromethyl)benzylidene)hydrazinyl) thiazole (**9b**)



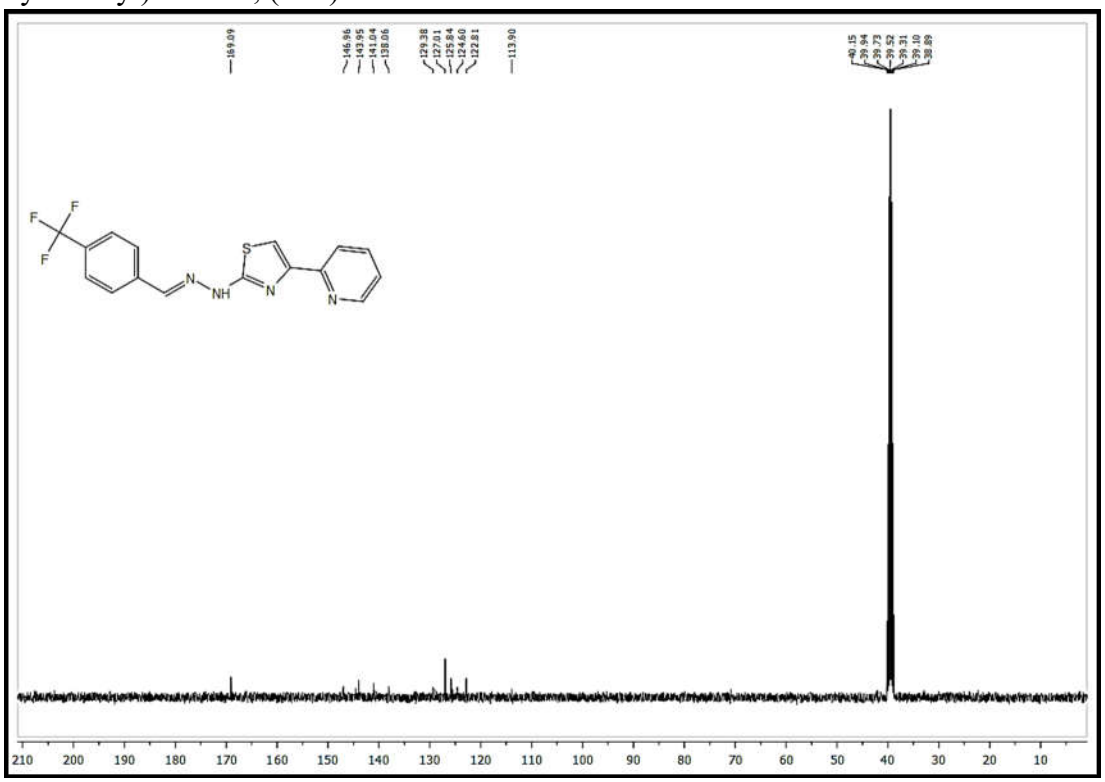
IR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(3-(trifluoromethyl)benzylidene)hydrazinyl) thiazole, (**9b**)



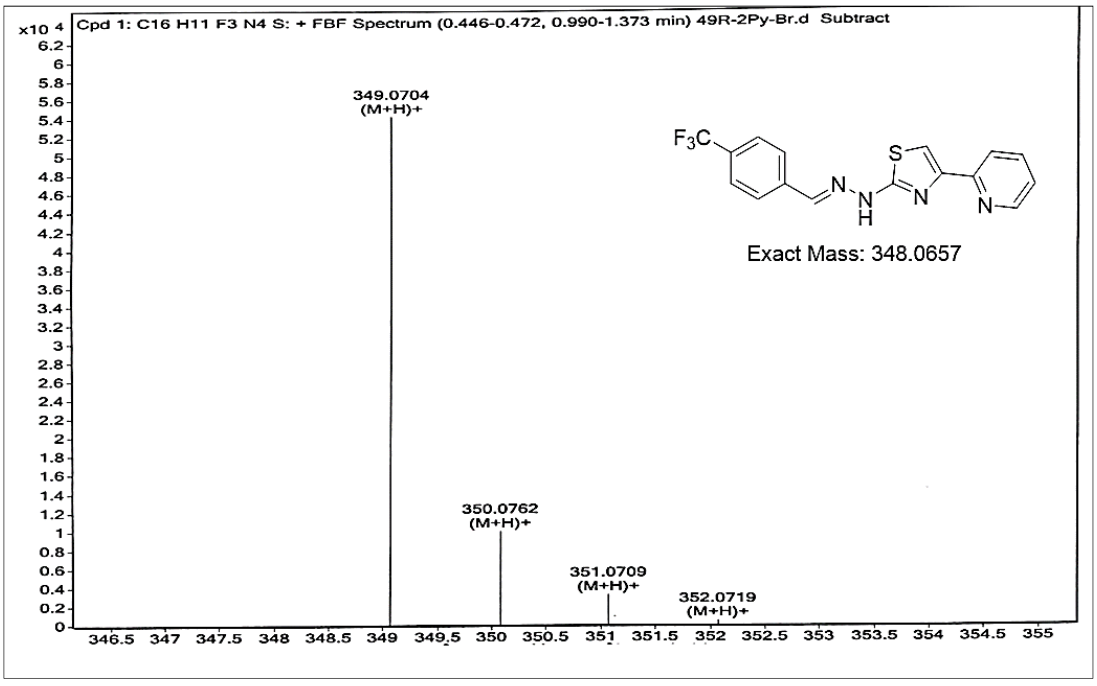
^{1}H NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(4-(trifluoromethyl) benzylidene) hydrazinyl)thiazole, (**10b**)



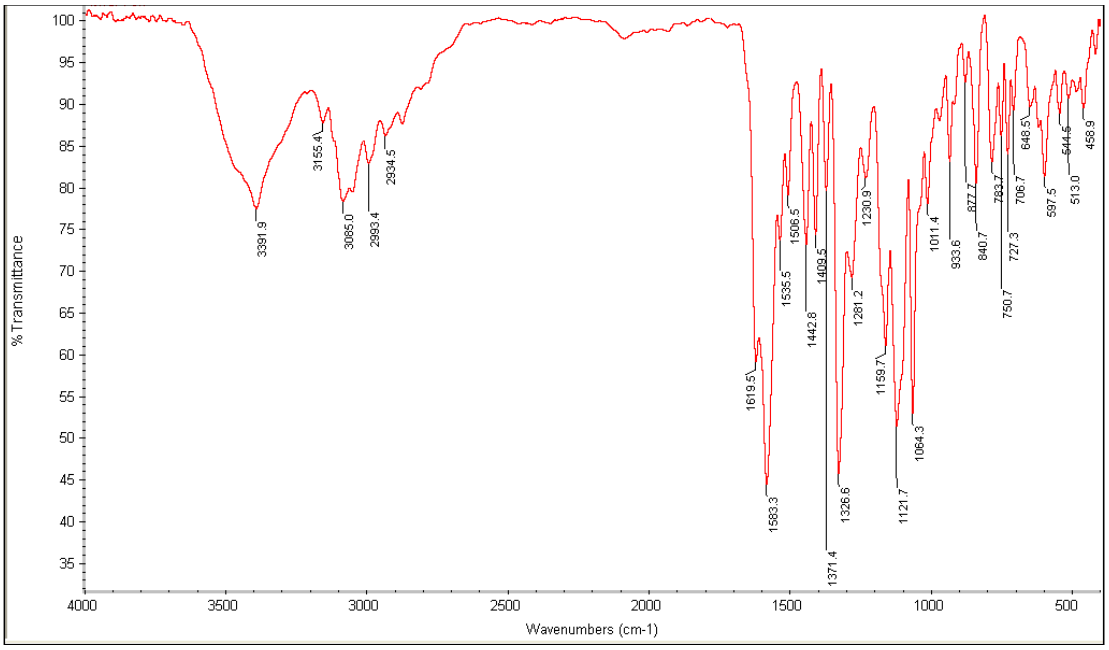
^{13}C NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(4-(trifluoromethyl) benzylidene) hydrazinyl)thiazole, (**10b**)



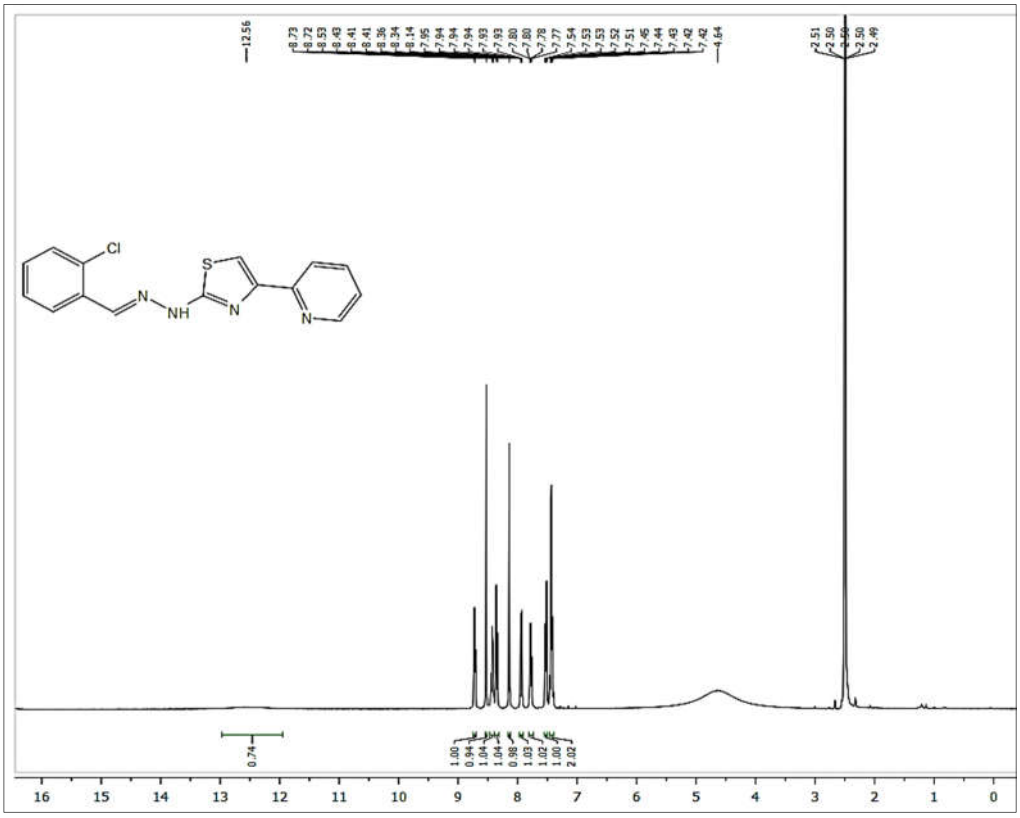
Mass Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(4-(trifluoromethyl)benzylidene)hydrazinyl) thiazole (**10b**)



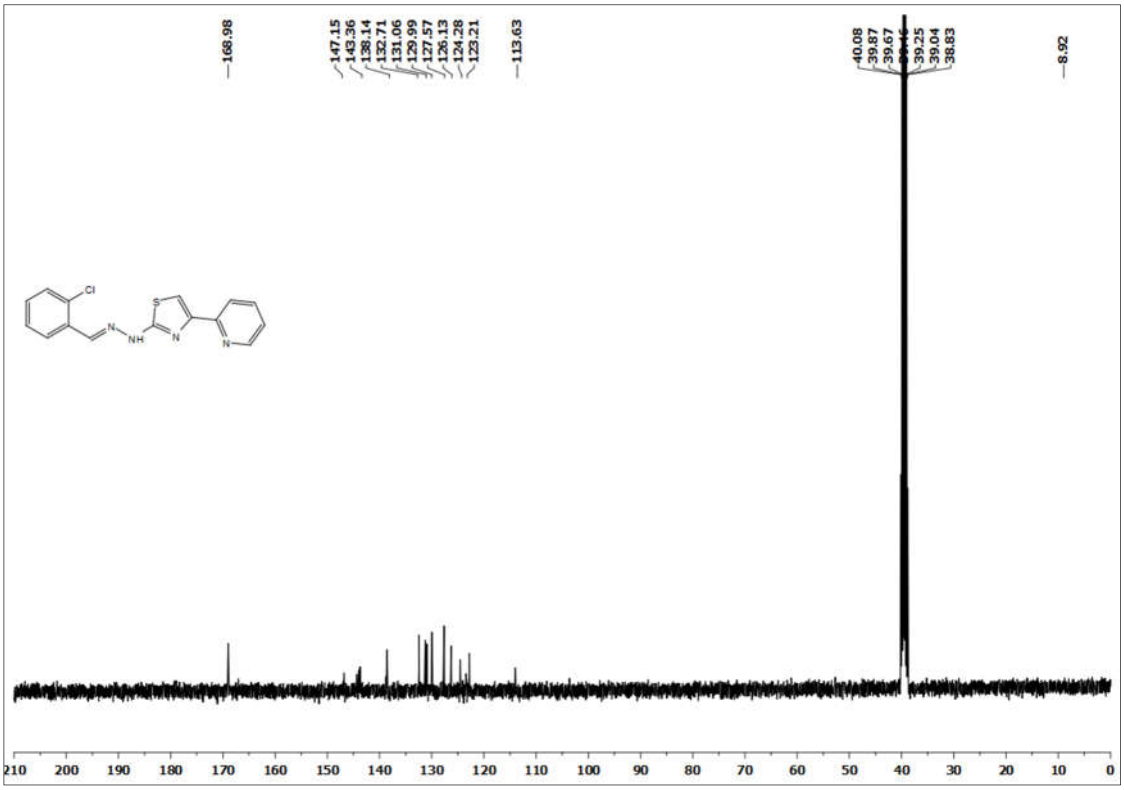
IR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(4-(trifluoromethyl)benzylidene)hydrazinyl) thiazole, (**10b**)



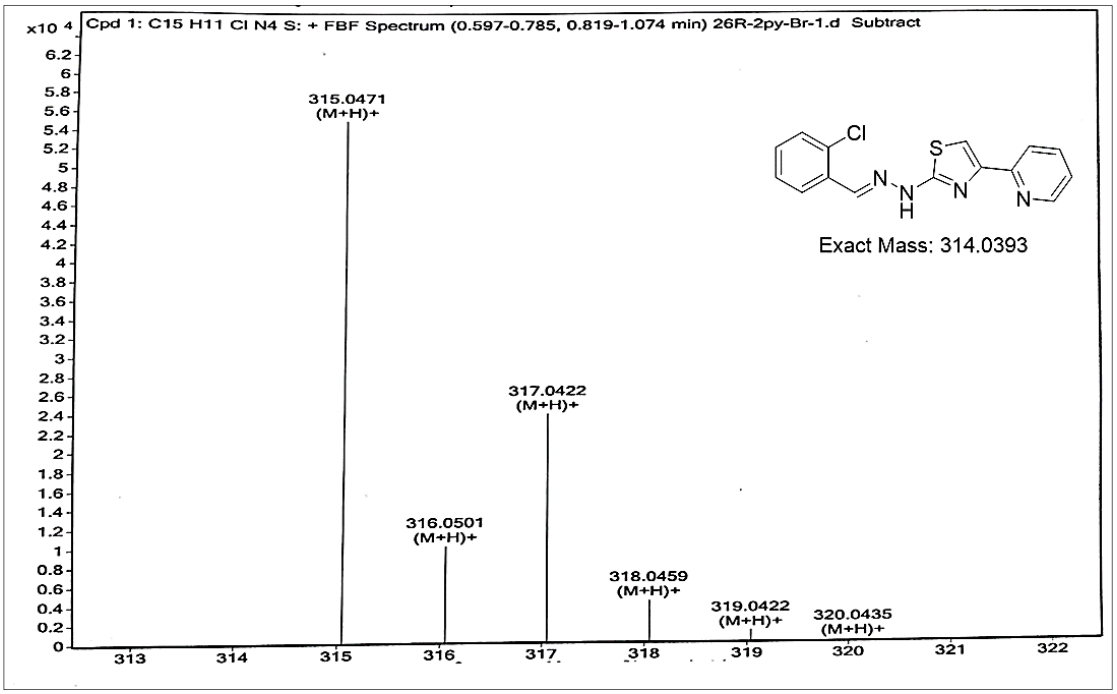
^{1}H NMR Spectrum of (E)-2-(2-(2-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(11b)**



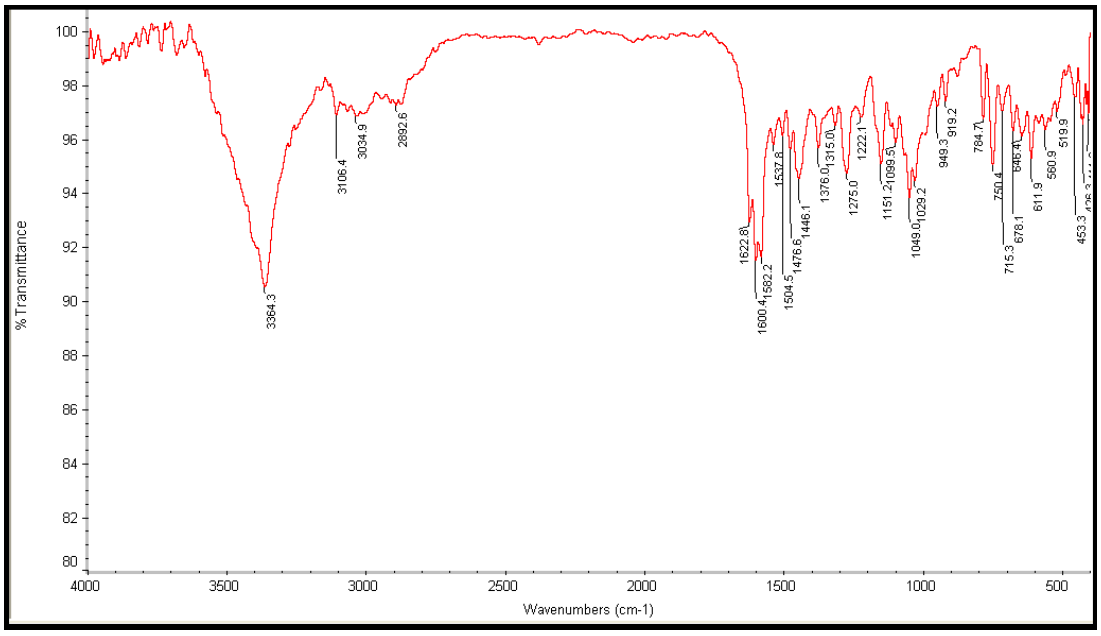
^{13}C NMR Spectrum of (E)-2-(2-(2-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(11b)**



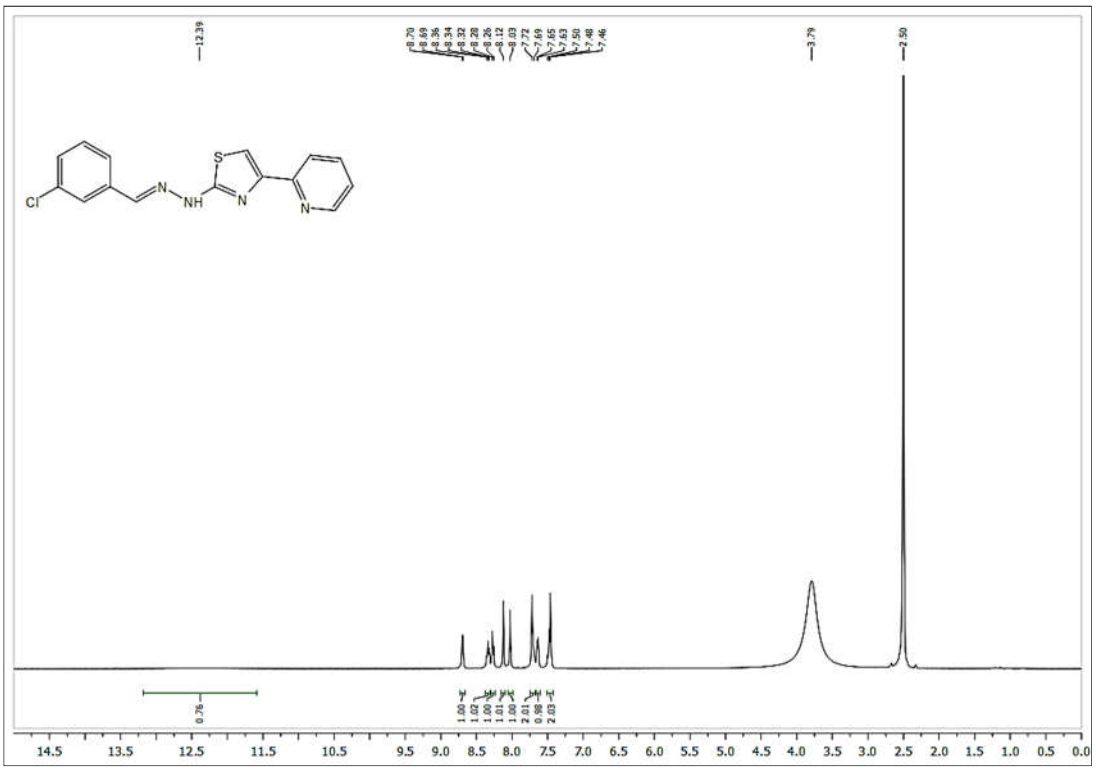
Mass Spectrum of (E)-2-(2-(2-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(11b)**



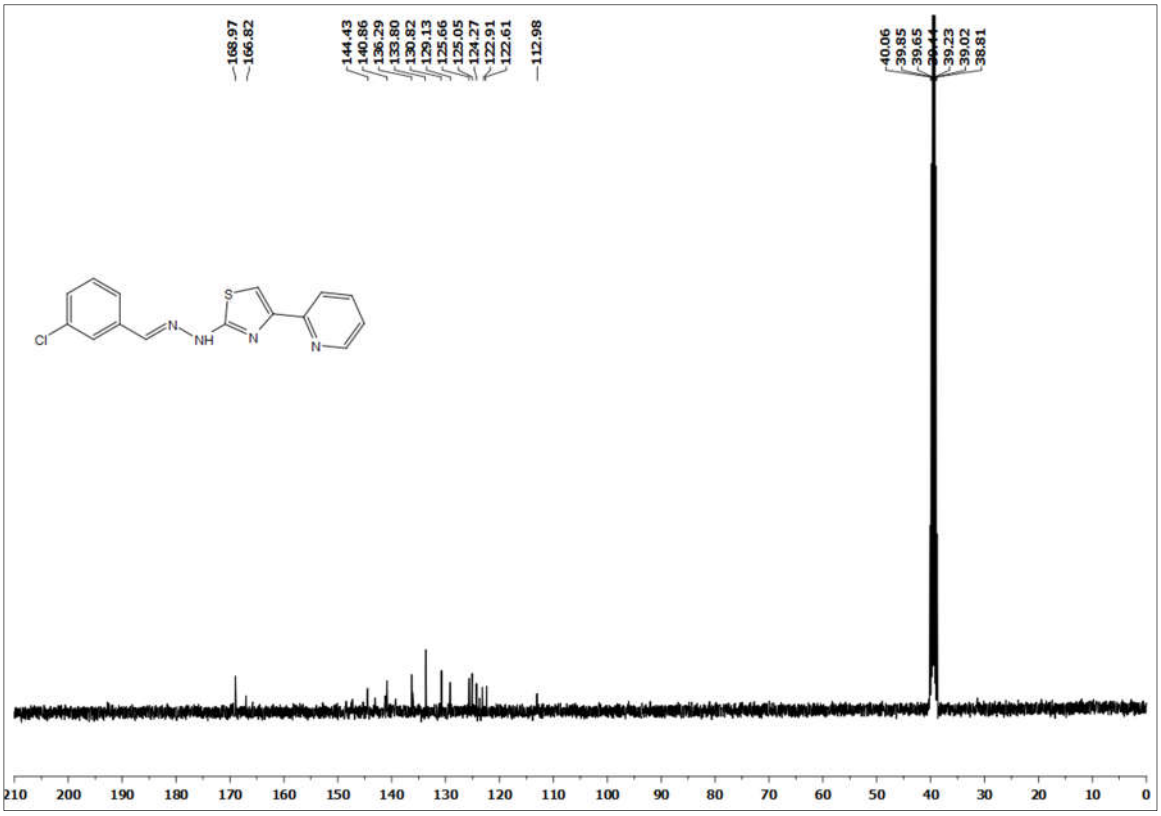
IR Spectrum of (E)-2-(2-(2-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(11b)**



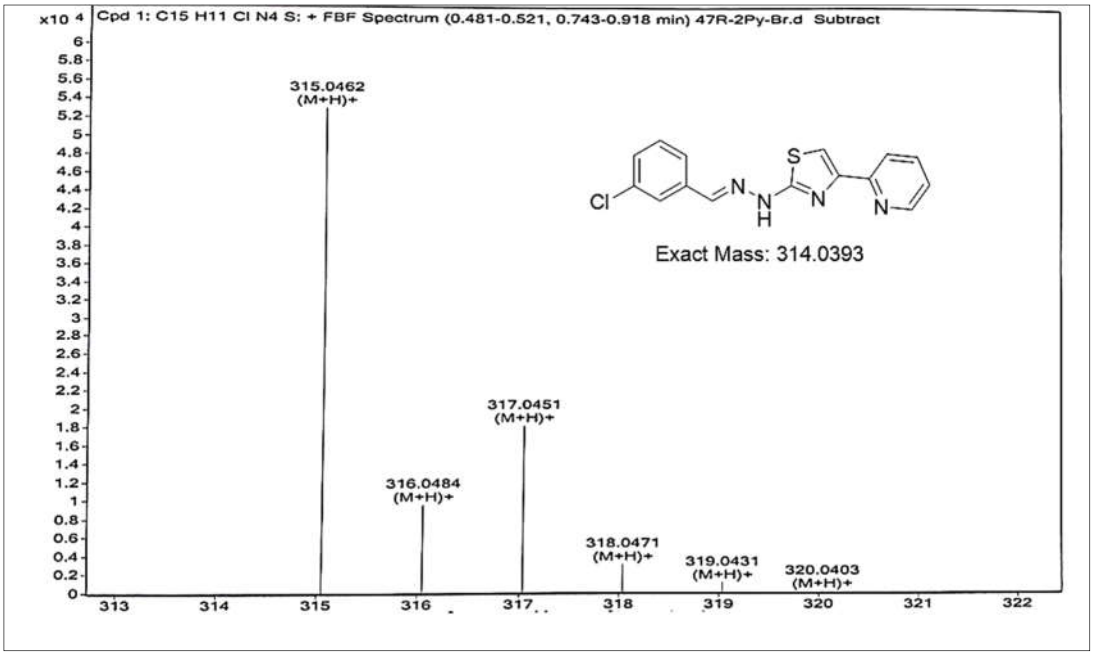
^{1}H NMR Spectrum of (E)-2-(2-(3-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl) thiazole, **(12b)**



^{13}C NMR Spectrum of (E)-2-(2-(3-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl) thiazole, **(12b)**

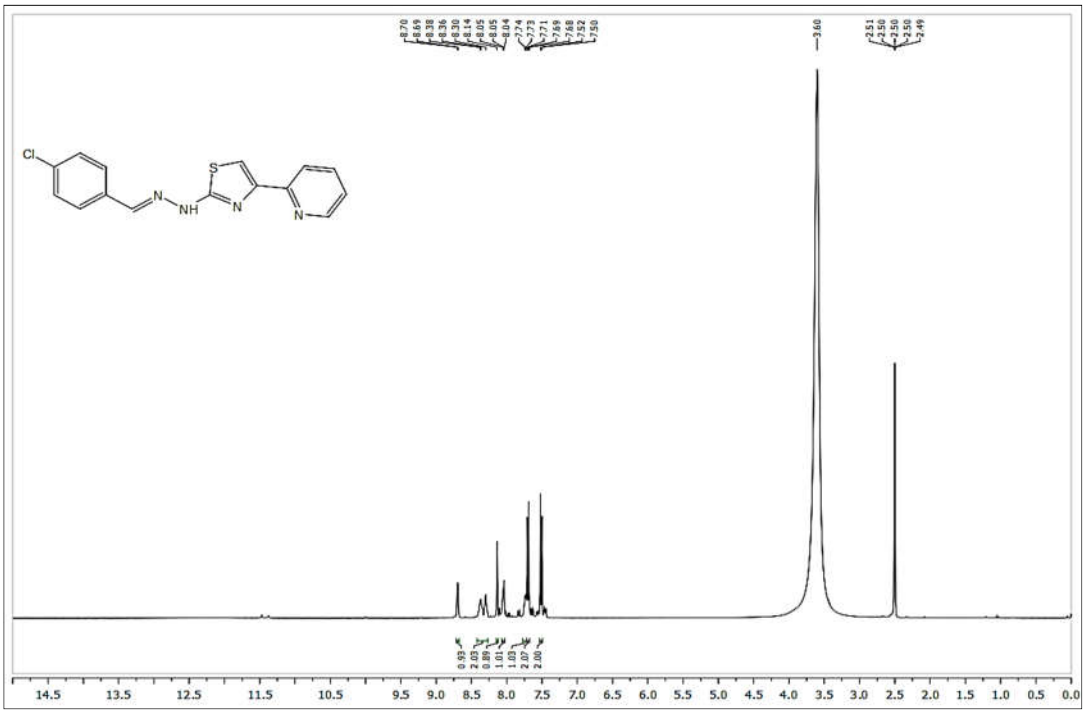


Mass Spectrum of (E)-2-(2-(3-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(12b)**

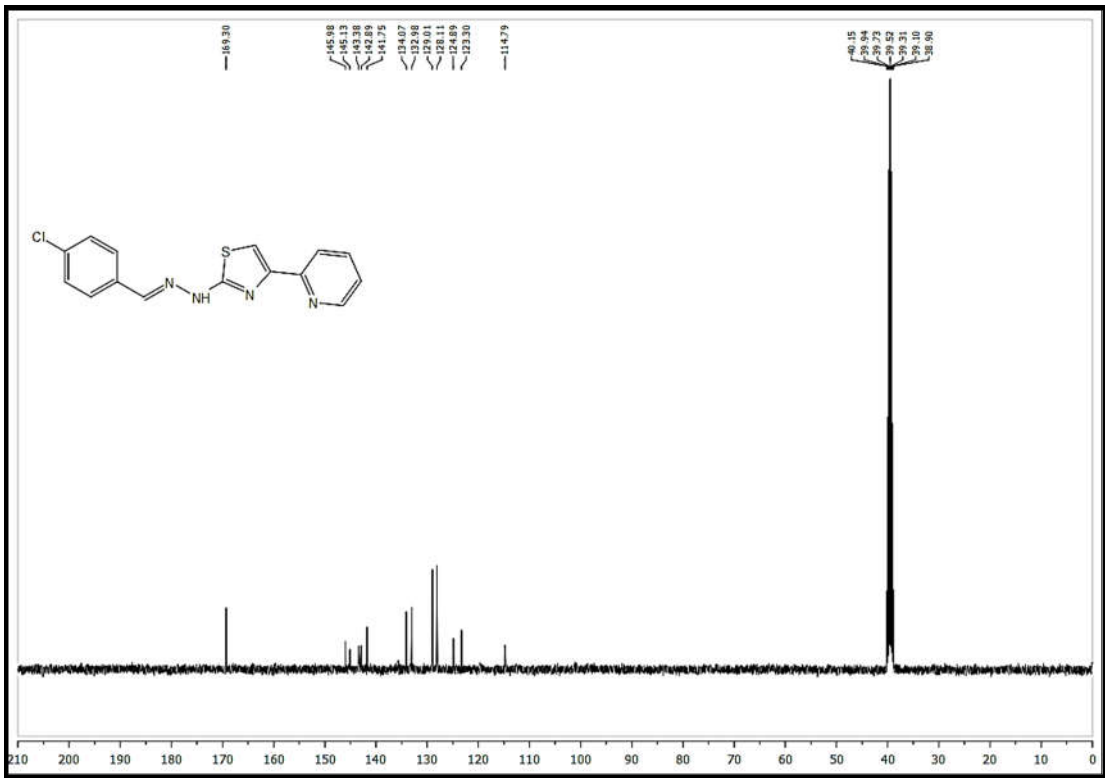


IR Spectrum of (E)-2-(2-(3-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, (**12b**)

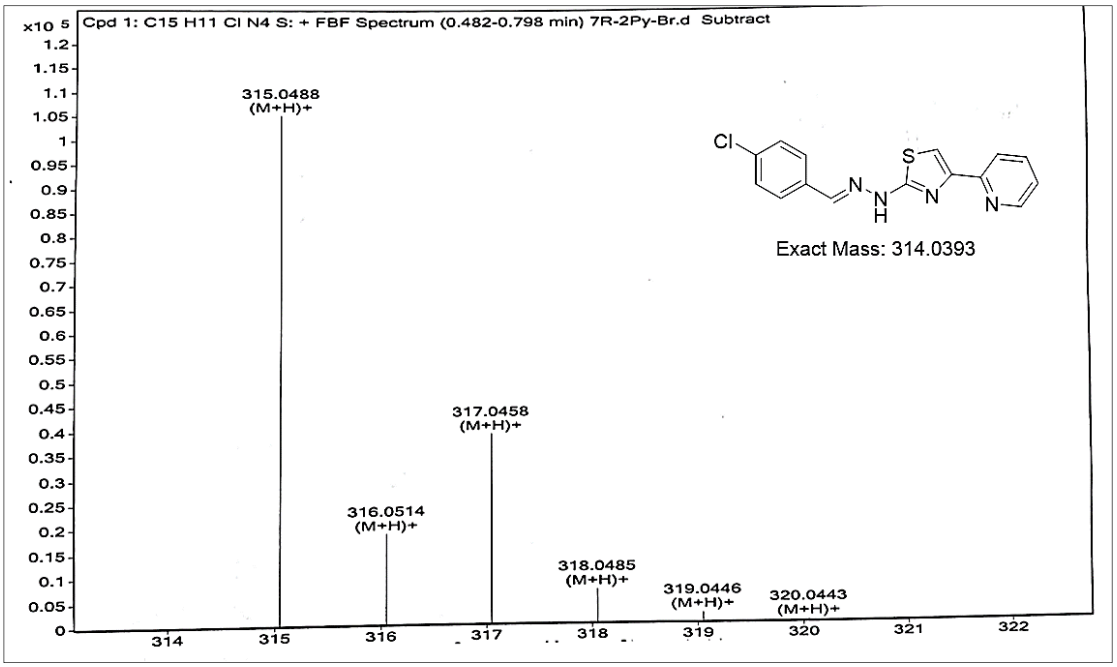
^{1}H NMR Spectrum of (E)-2-(2-(4-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(13b)**



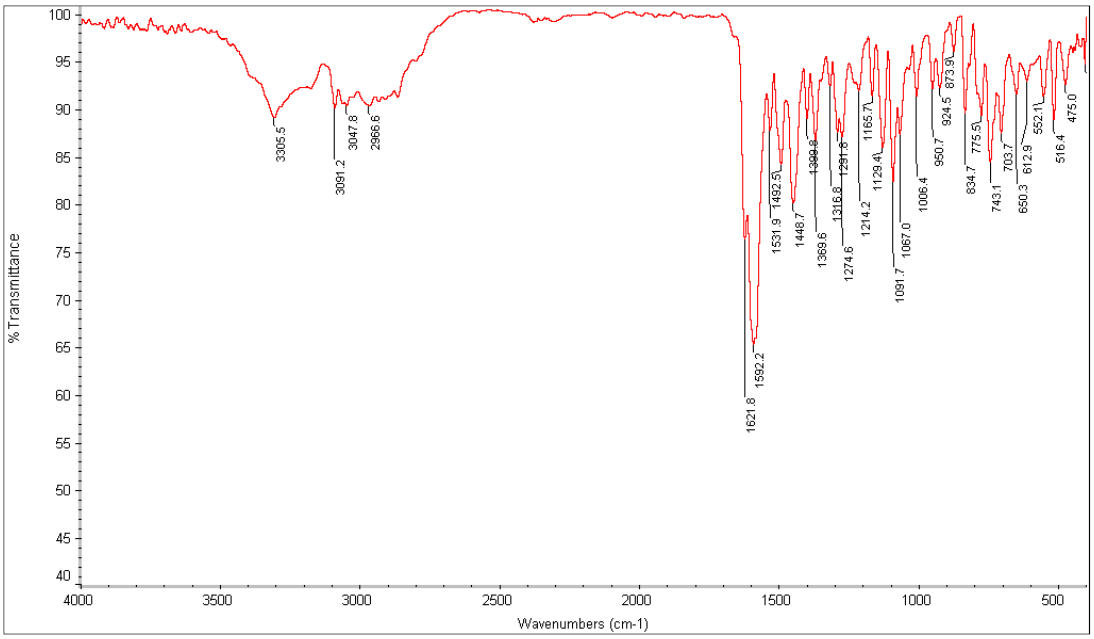
^{13}C NMR Spectrum of (E)-2-(2-(4-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(13b)**



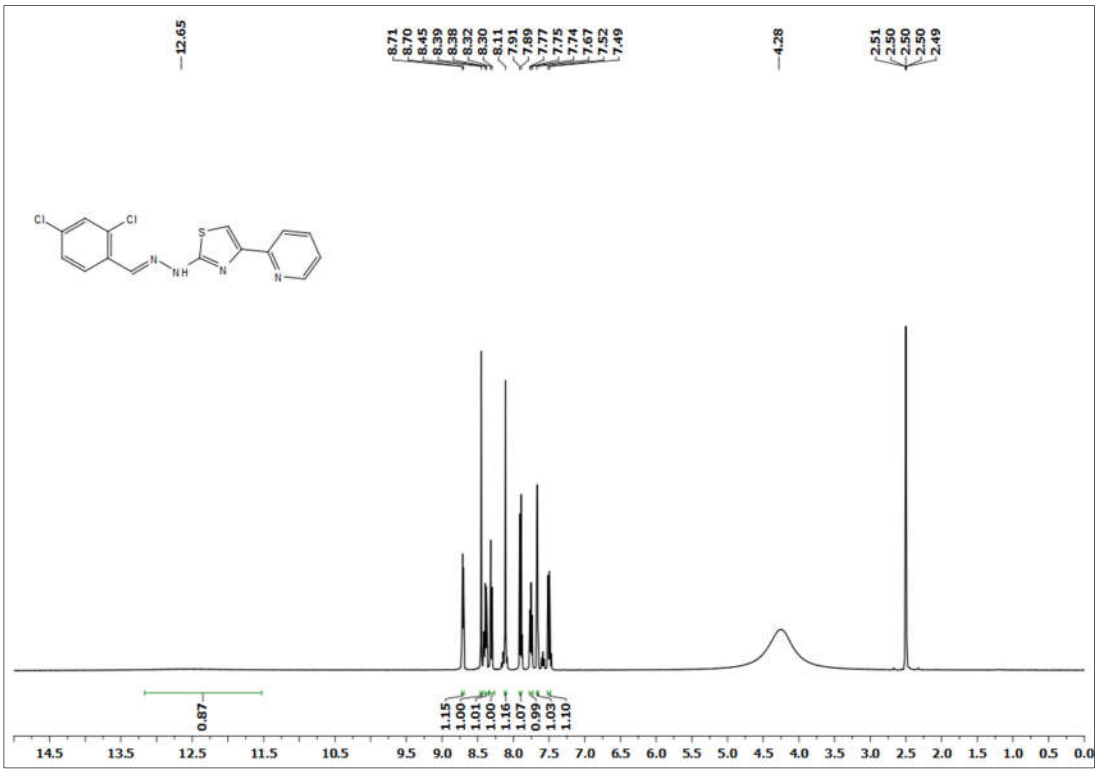
Mass Spectrum of (E)-2-(2-(4-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**13b**)



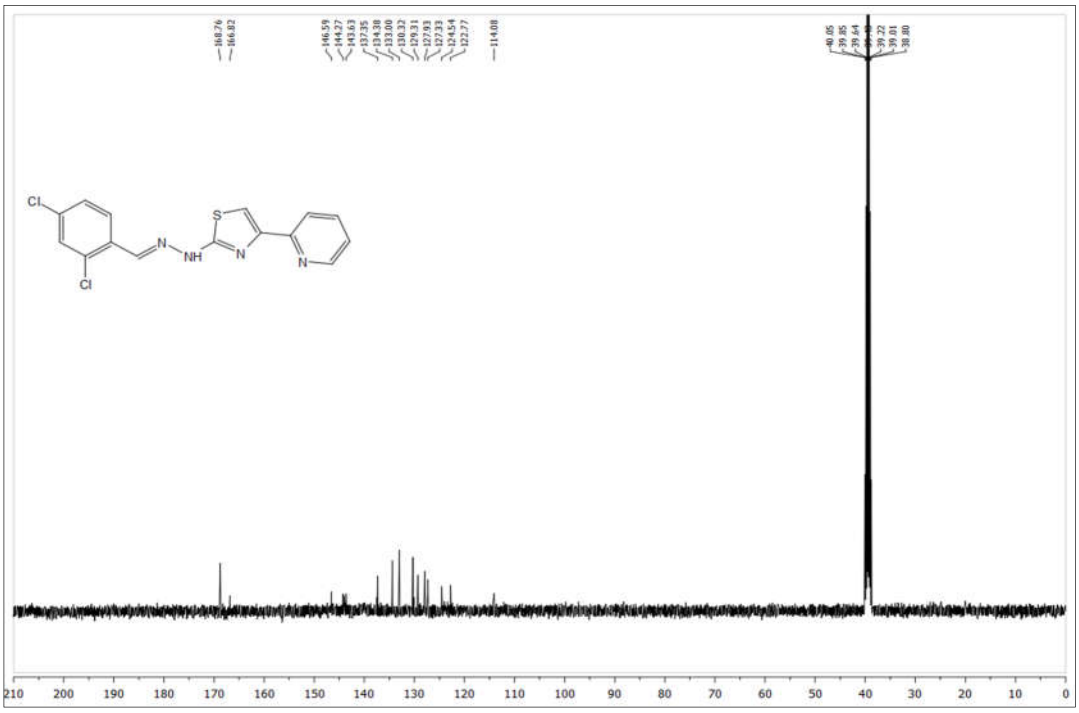
IR Spectrum of (E)-2-(2-(4-chlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(13b)**



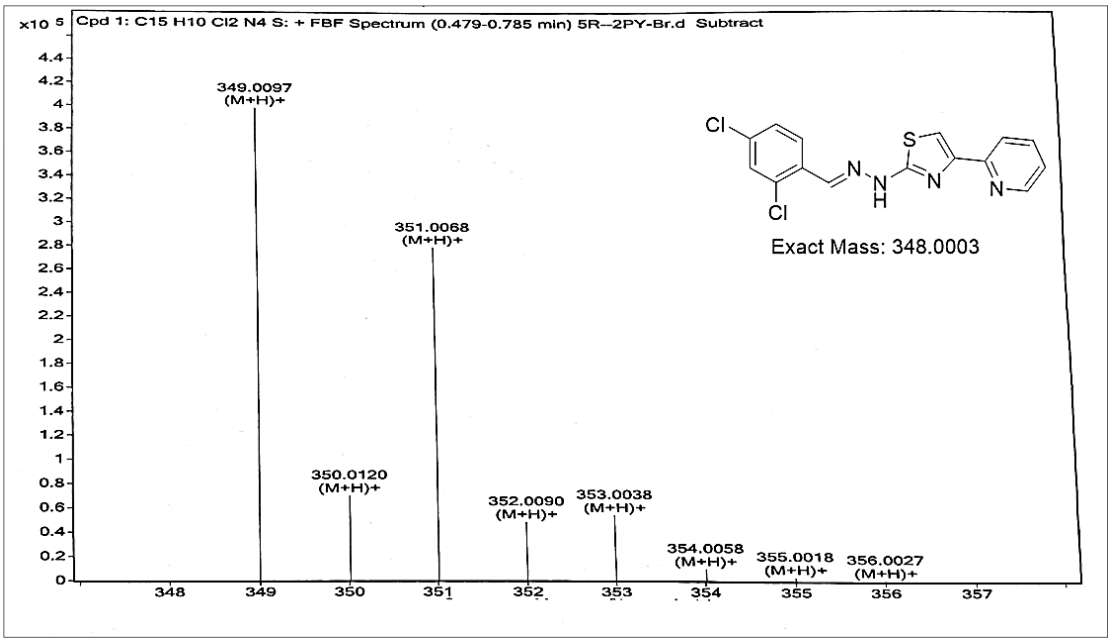
^{1}H NMR Spectrum of (E)-2-(2-(2,4-dichlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl) thiazole, **(14b)**



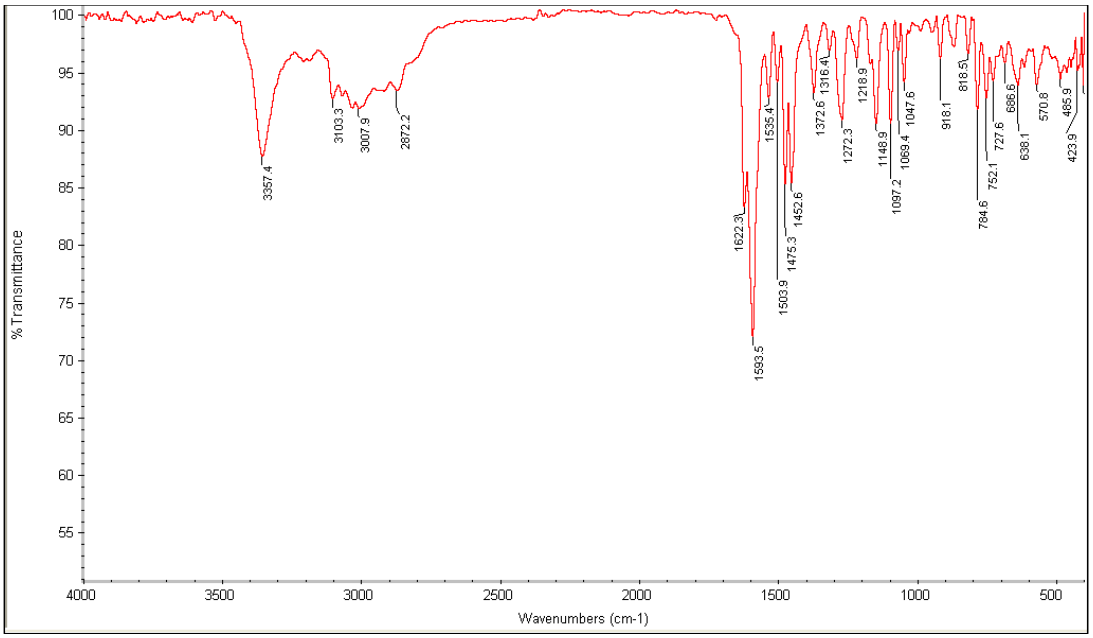
^{13}C NMR Spectrum of (E)-2-(2-(2,4-dichlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl) thiazole, **(14b)**



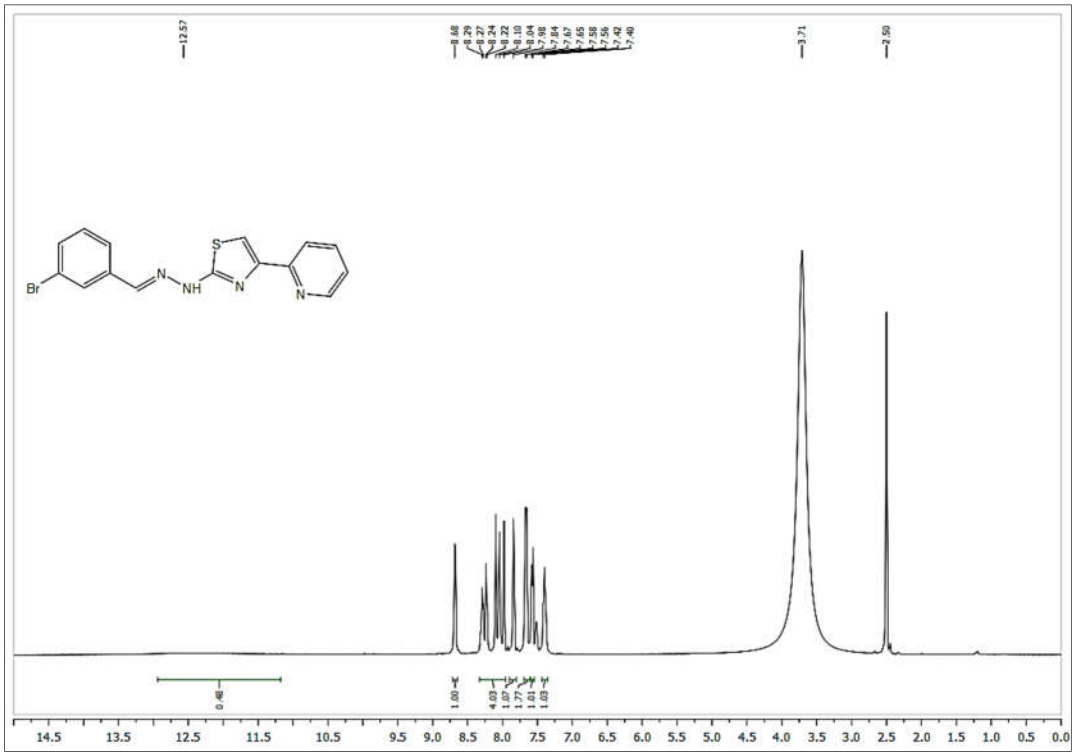
Mass Spectrum of (E)-2-(2-(2,4-dichlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**14b**)



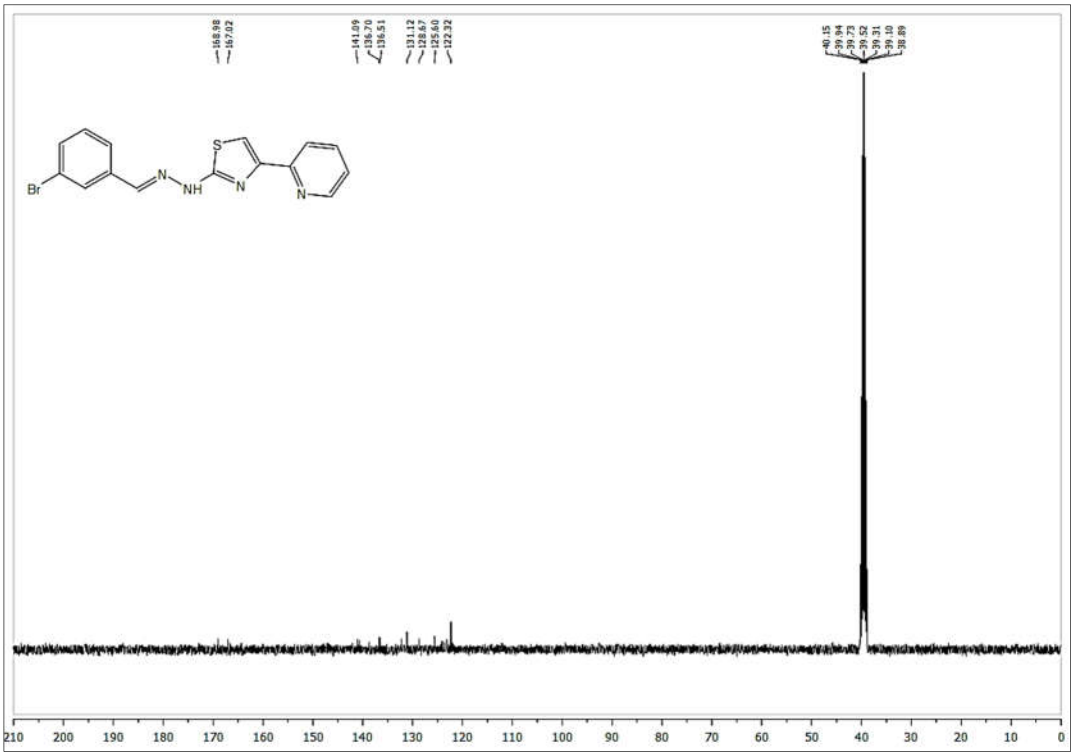
IR Spectrum of (E)-2-(2-(2,4-dichlorobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(14b)**



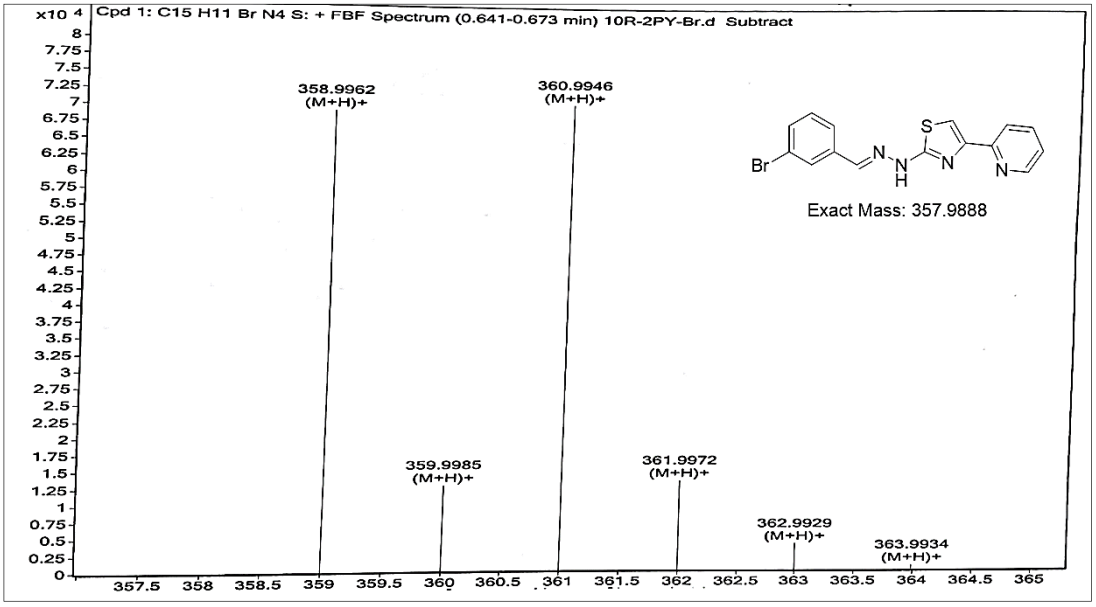
^{1}H NMR Spectrum of (E)-2-(2-(3-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(15b)**



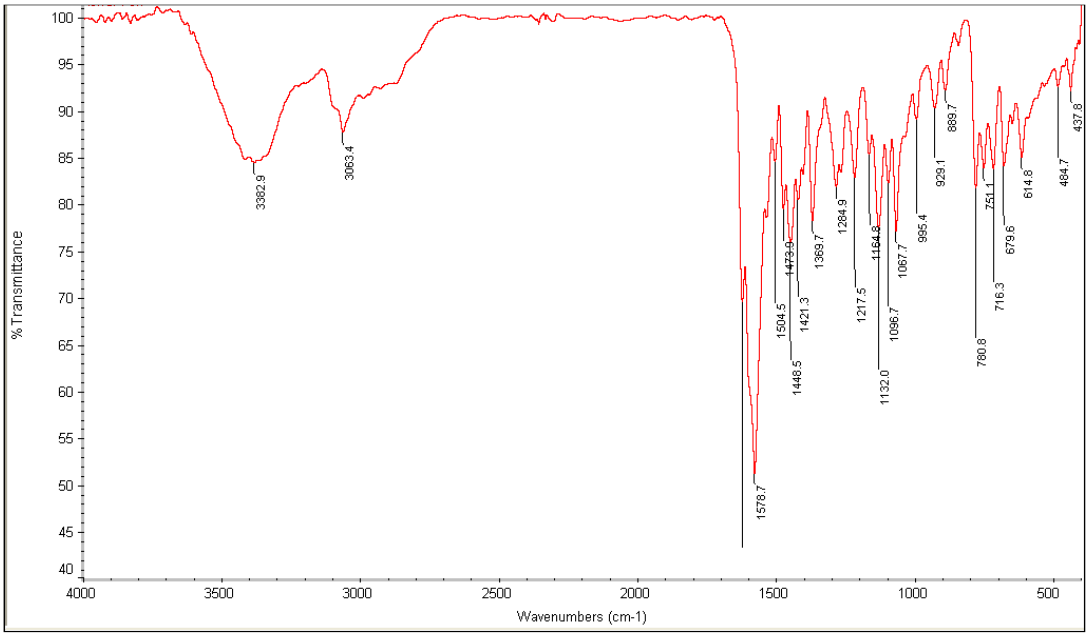
^{13}C NMR Spectrum of (E)-2-(2-(3-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(15b)**



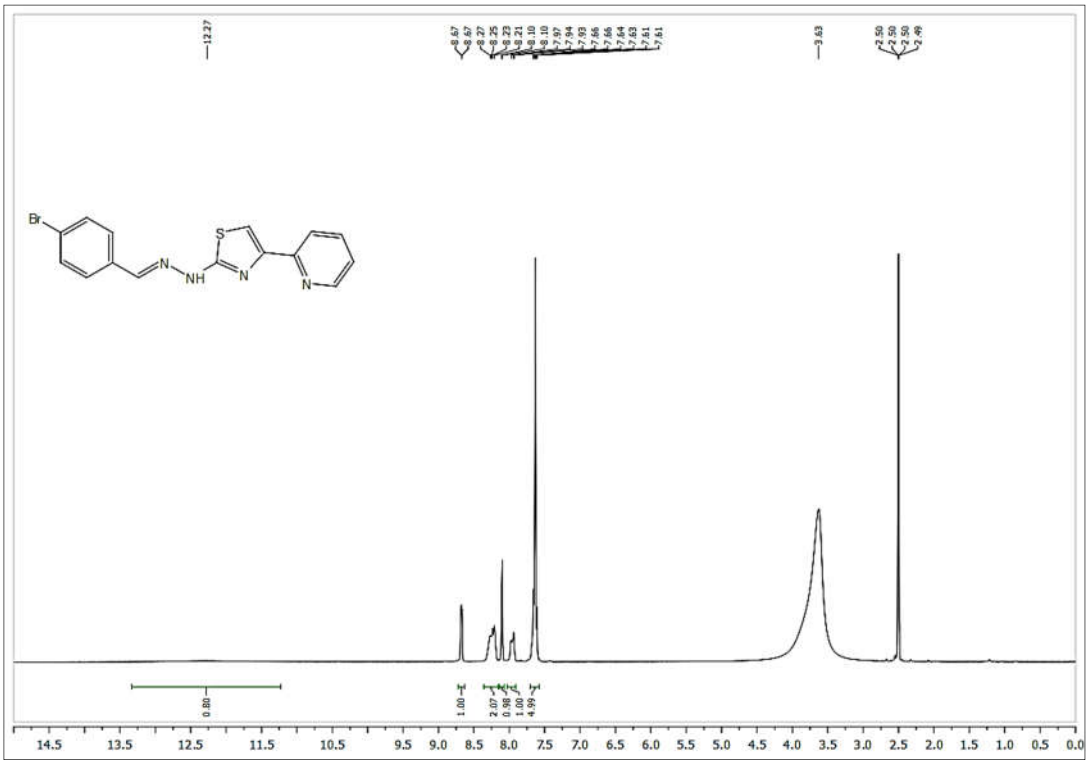
Mass Spectrum of (E)-2-(2-(3-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**15b)**



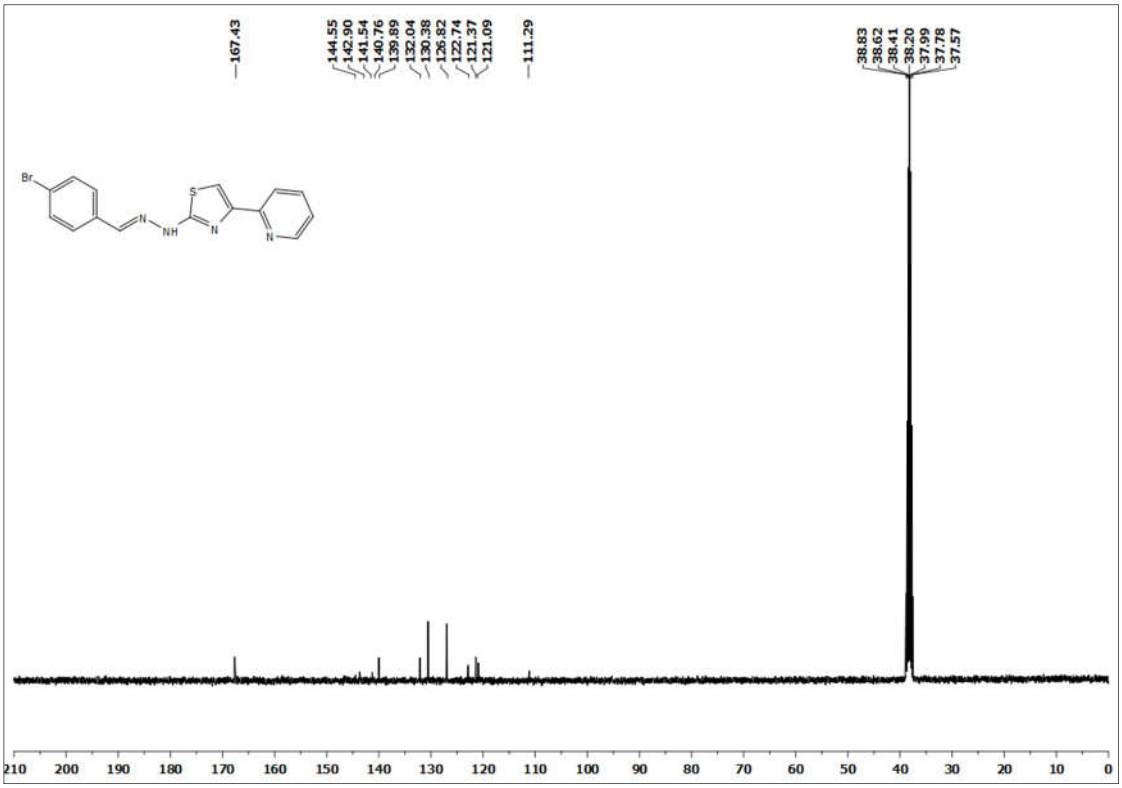
IR Spectrum of (E)-2-(2-(3-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(15b)**



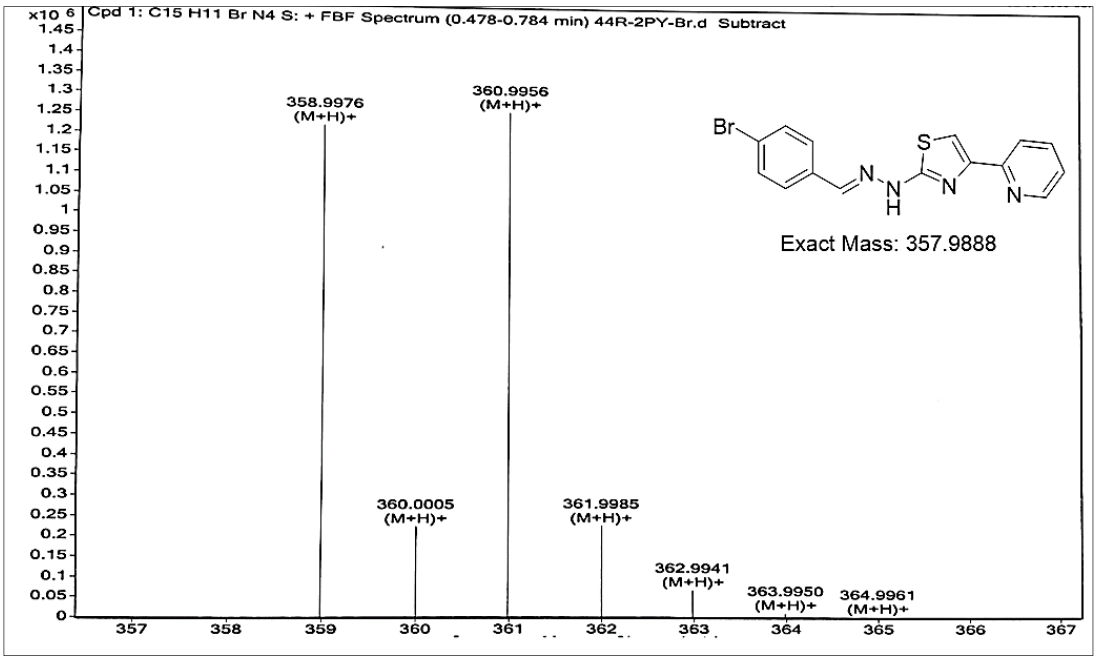
^{1}H NMR Spectrum of (E)-2-(2-(4-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(16b)**



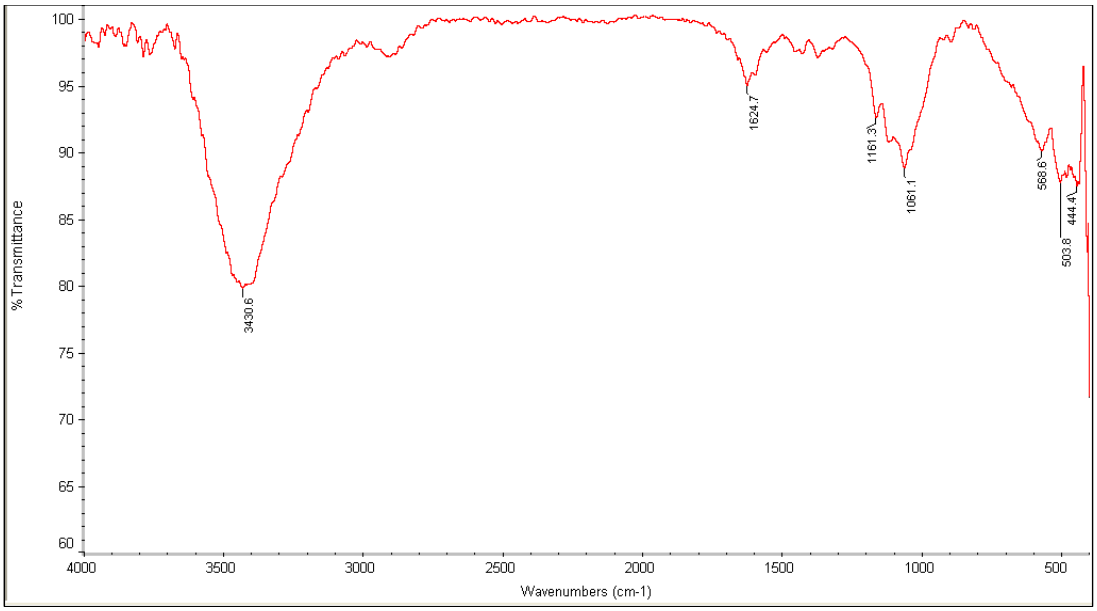
^{13}C NMR Spectrum of (E)-2-(2-(4-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(16b)**



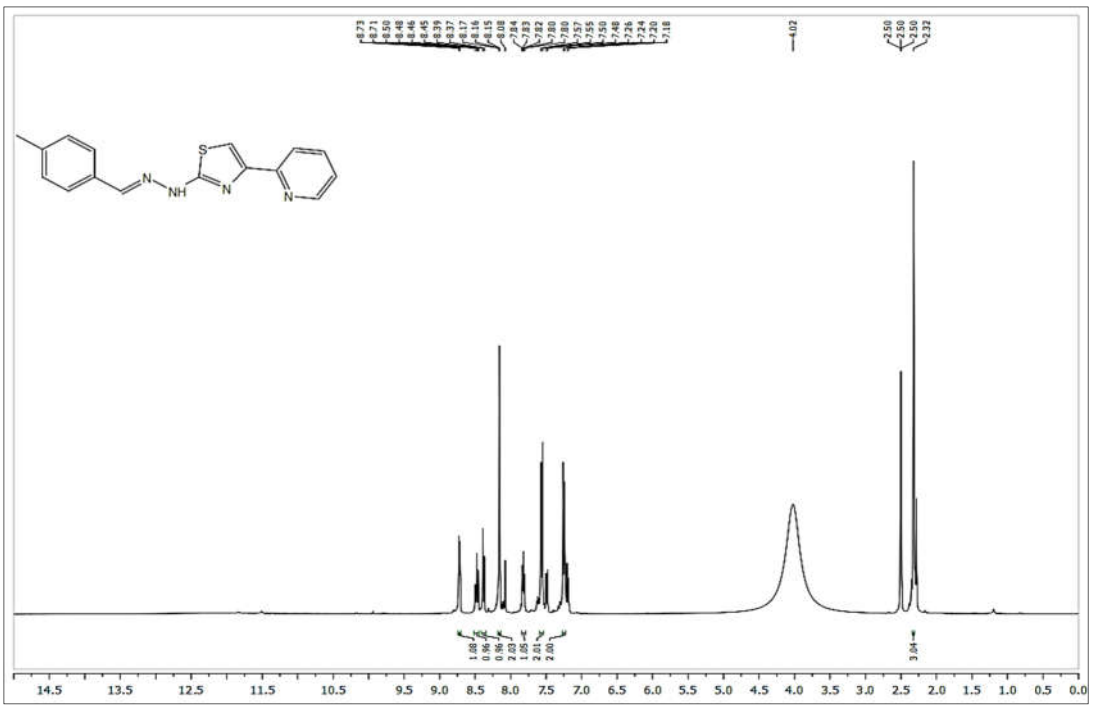
Mass Spectrum of (E)-2-(2-(4-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(16b)**



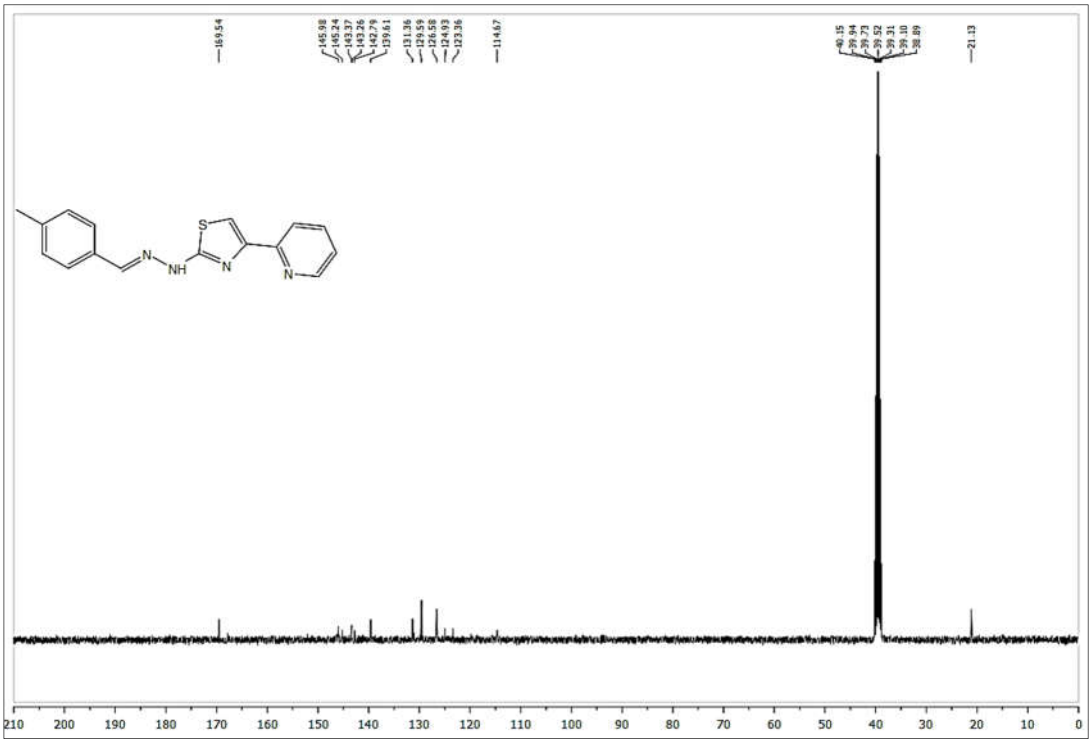
IR Spectrum of (E)-2-(2-(4-bromobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(16b)**



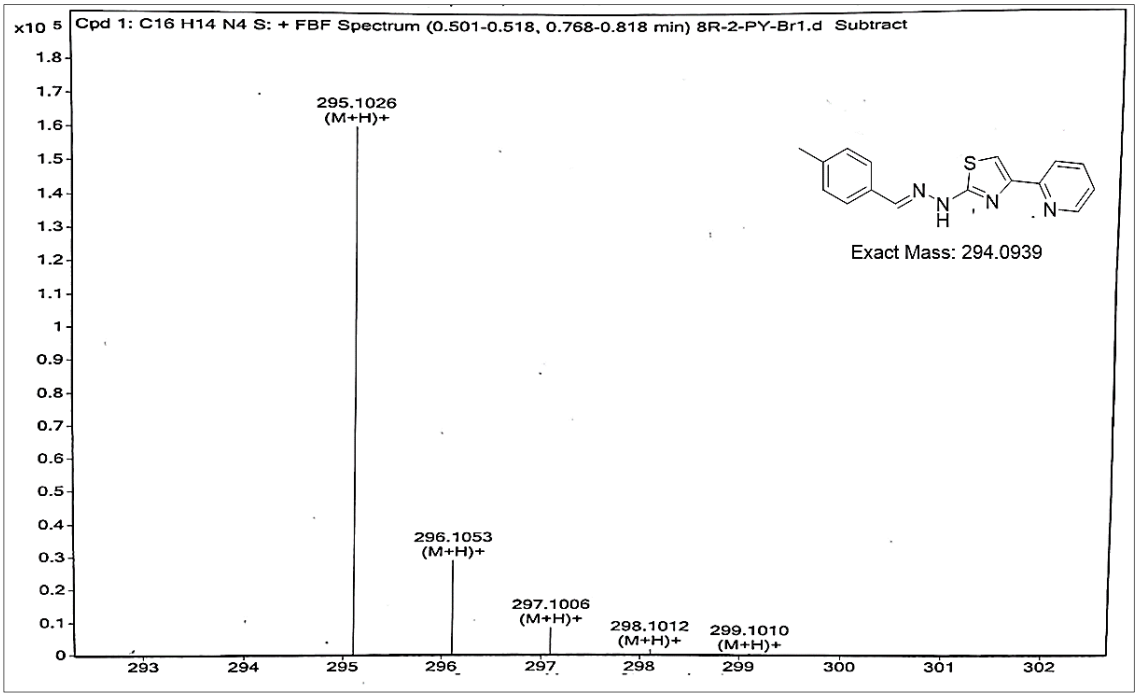
^{1}H NMR Spectrum of (E)-2-(2-(4-methylbenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(17b)**



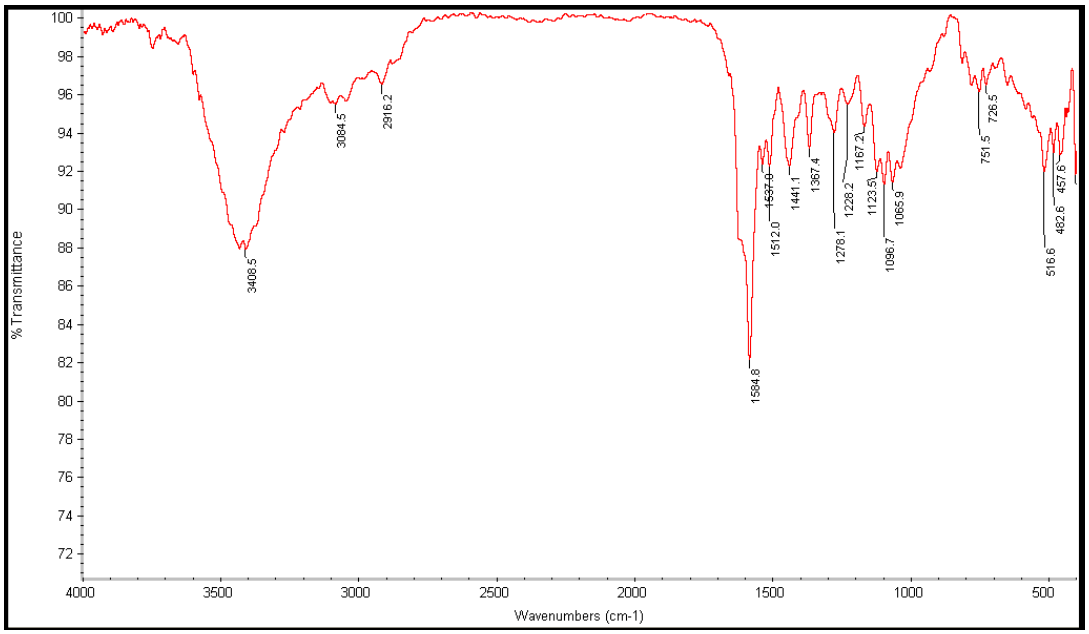
^{13}C NMR Spectrum of (E)-2-(2-(4-methylbenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(17b)**



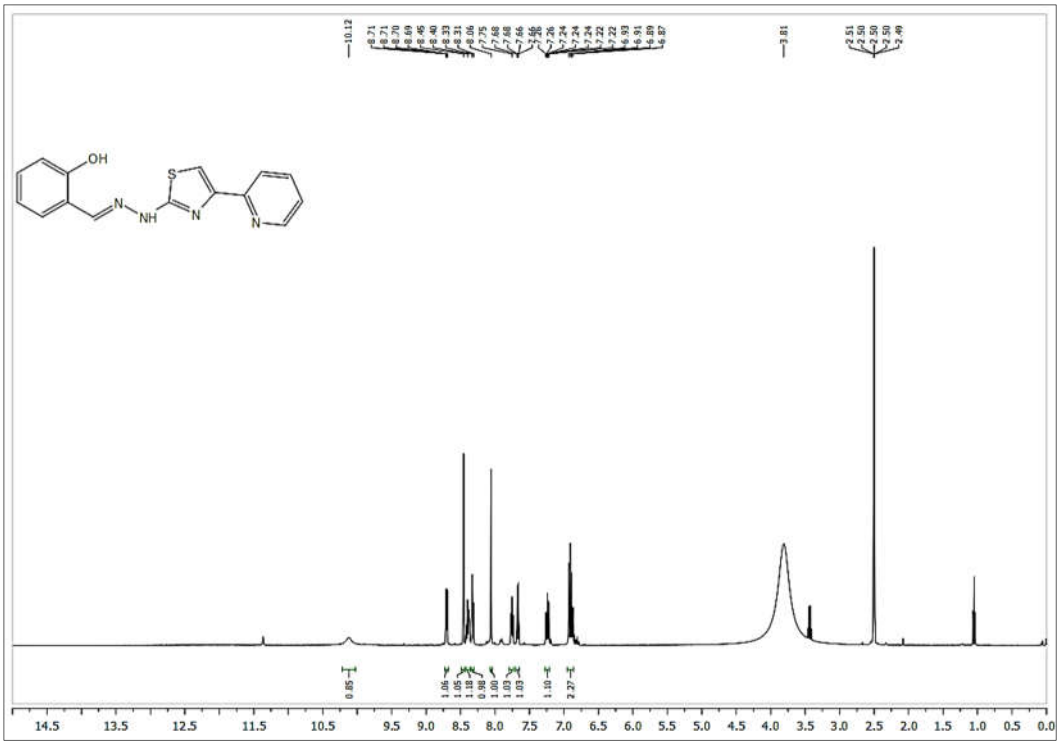
Mass Spectrum of (E)-2-(2-(4-methylbenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**17b**)



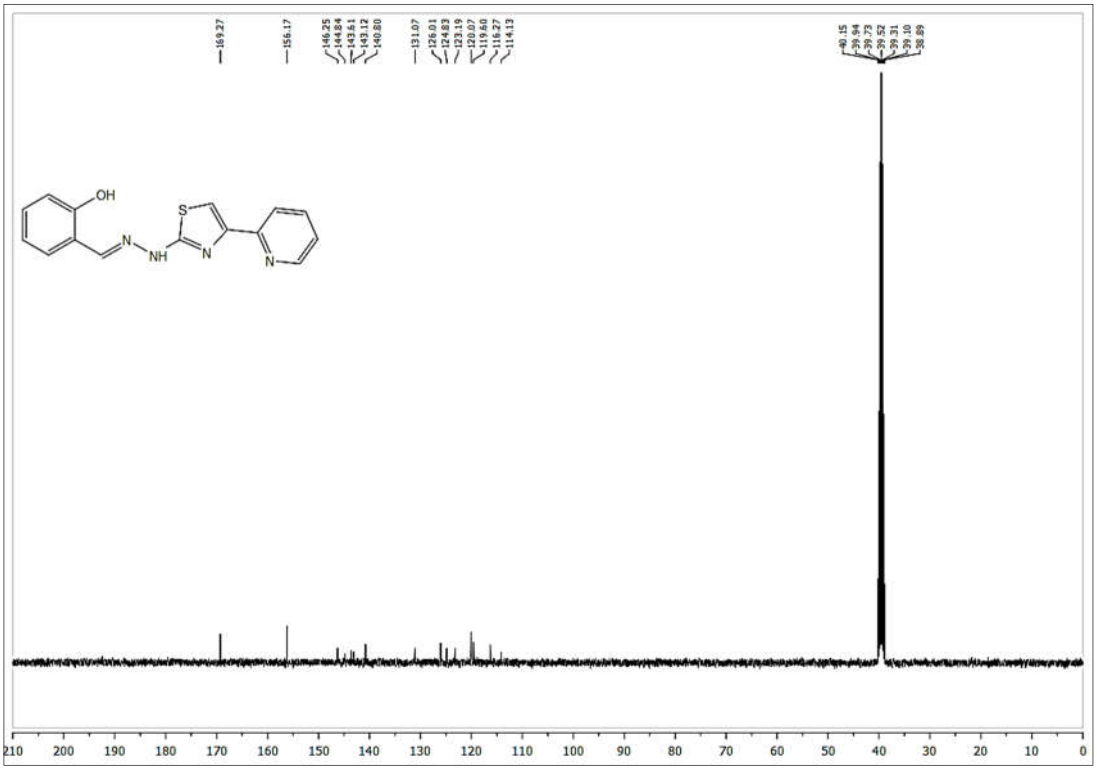
IR Spectrum of (E)-2-(2-(4-methylbenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(17b)**



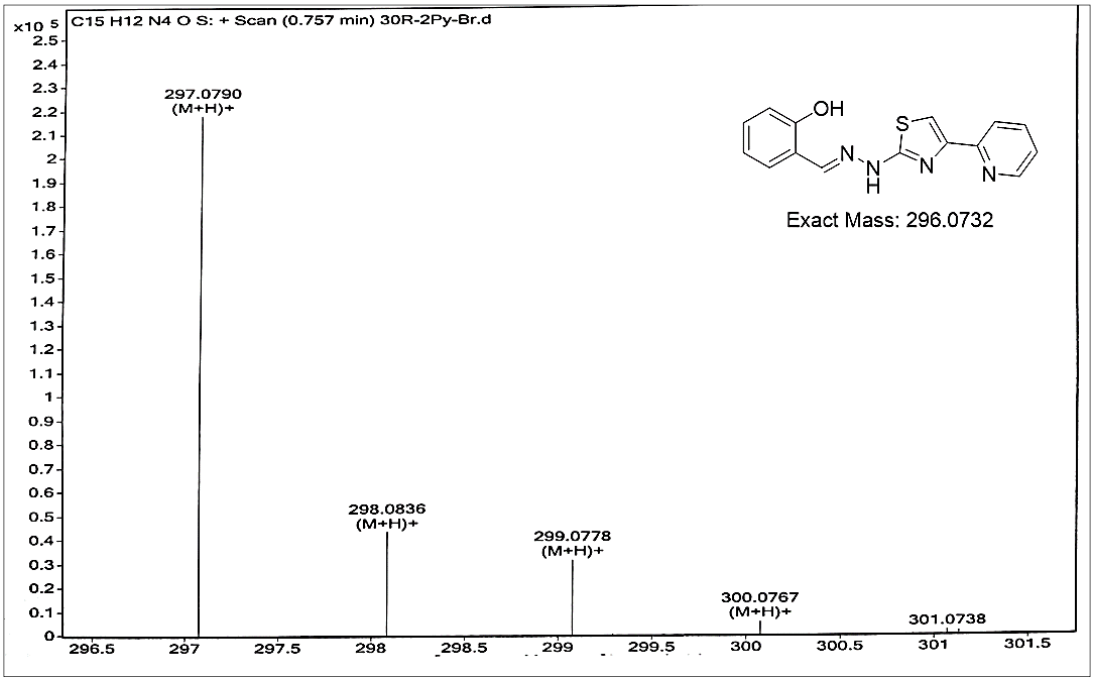
^{1}H NMR Spectrum of (E)-2-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, **(18b)**



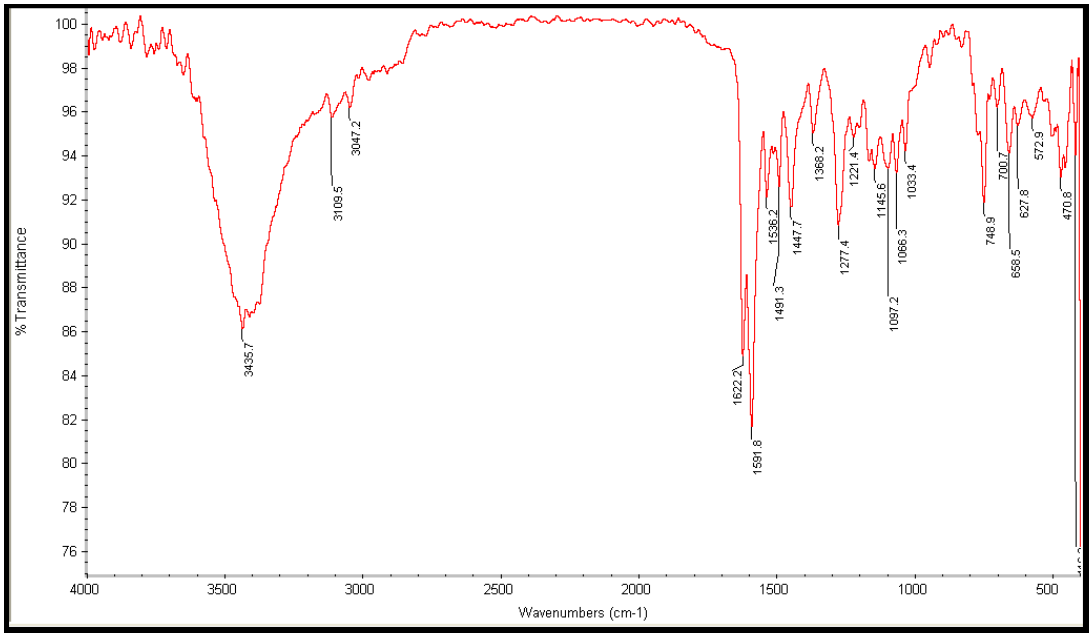
^{13}C NMR Spectrum of (E)-2-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, **(18b)**



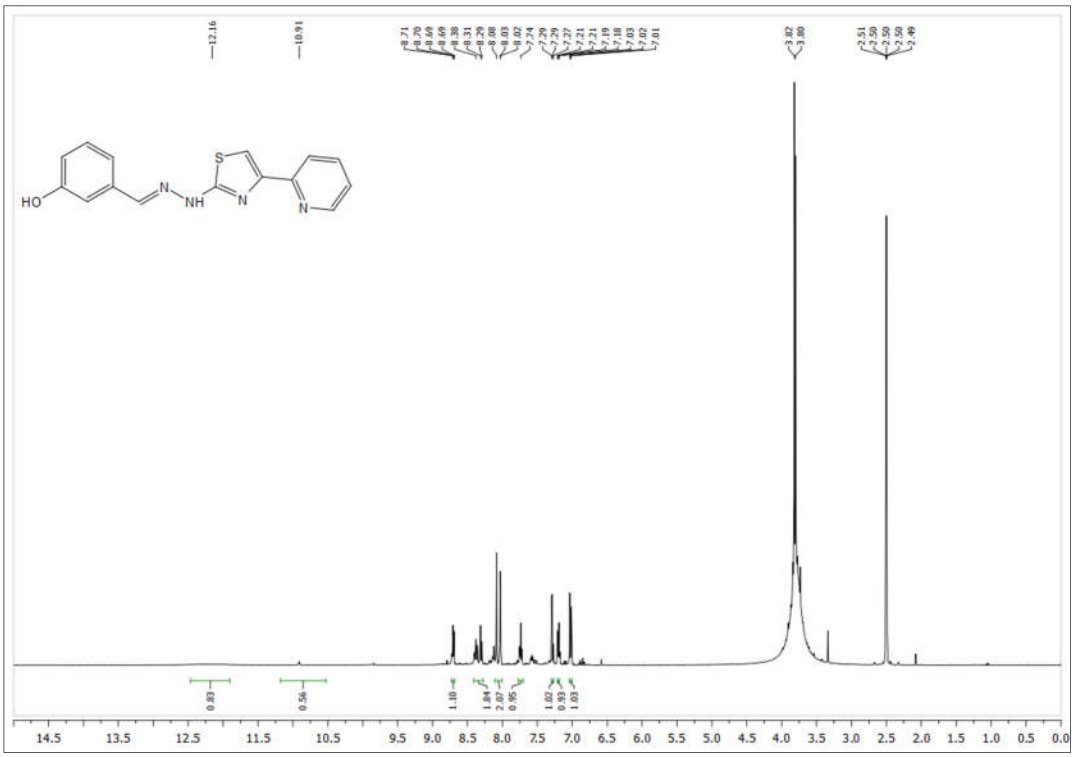
Mass Spectrum of (E)-2-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol (**18b**)



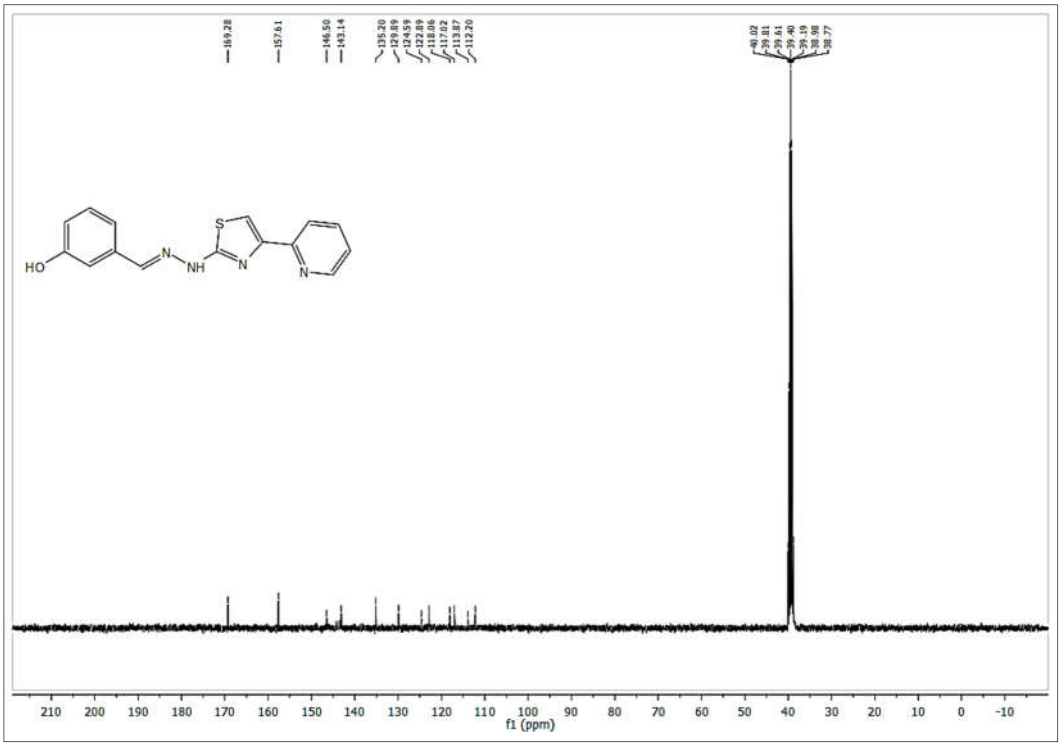
IR Spectrum of (E)-2-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, **(18b)**



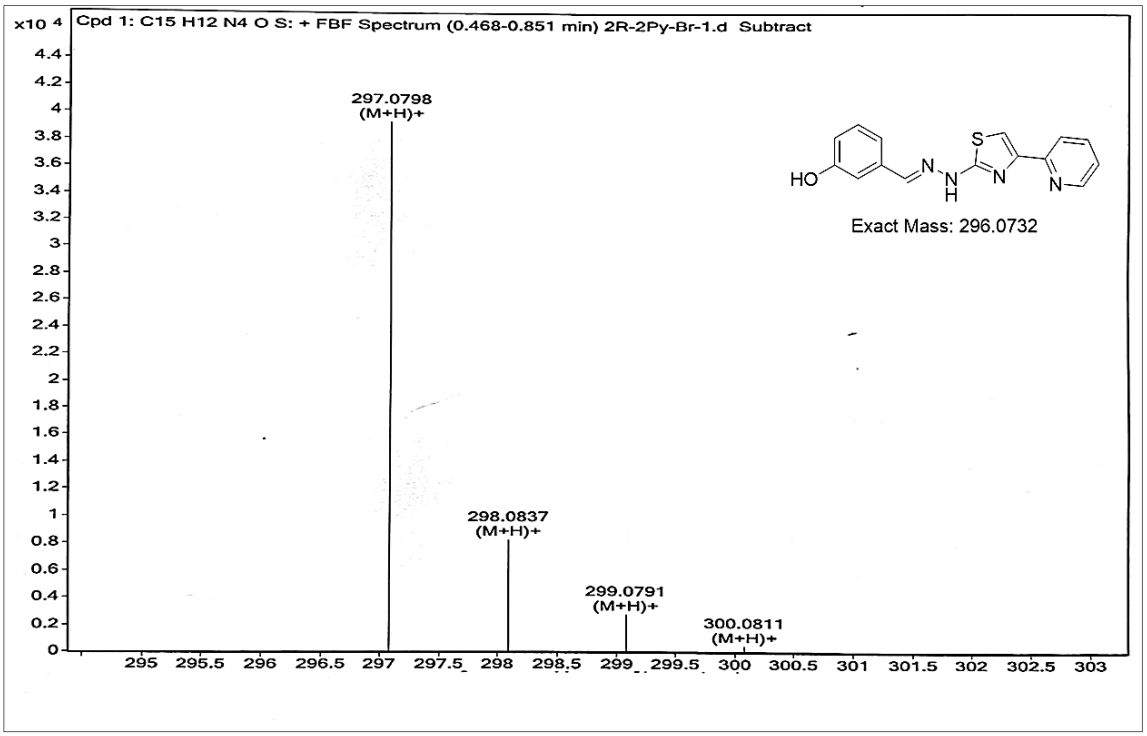
^{1}H NMR Spectrum of (E)-3-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl) phenol, (**19b**)



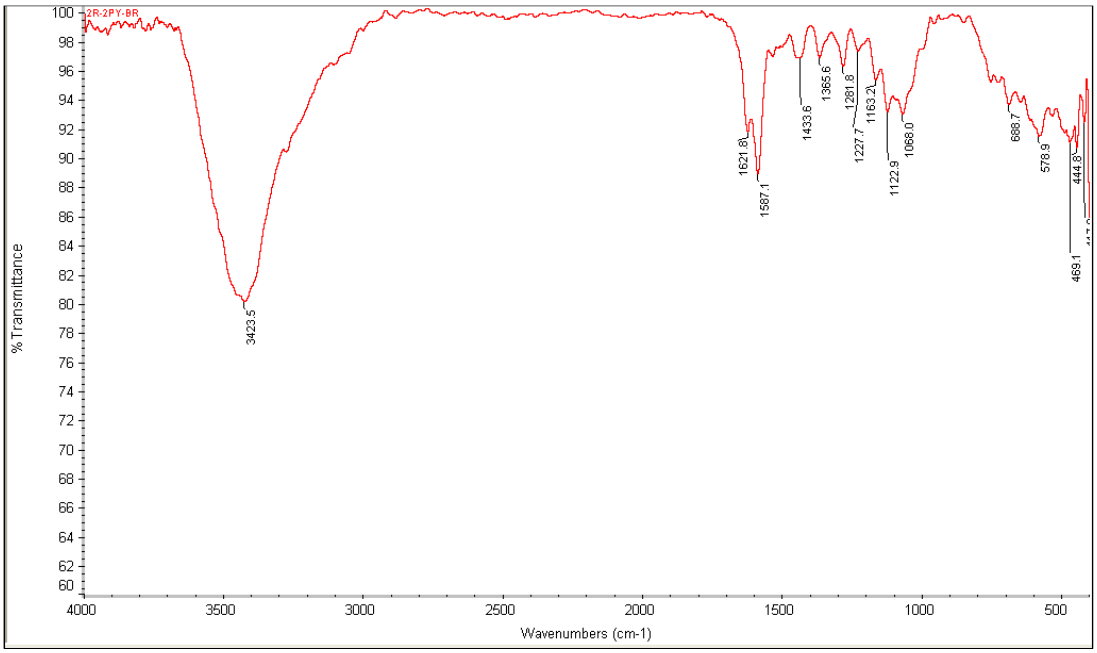
^{13}C NMR Spectrum of (E)-3-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, **(19b)**



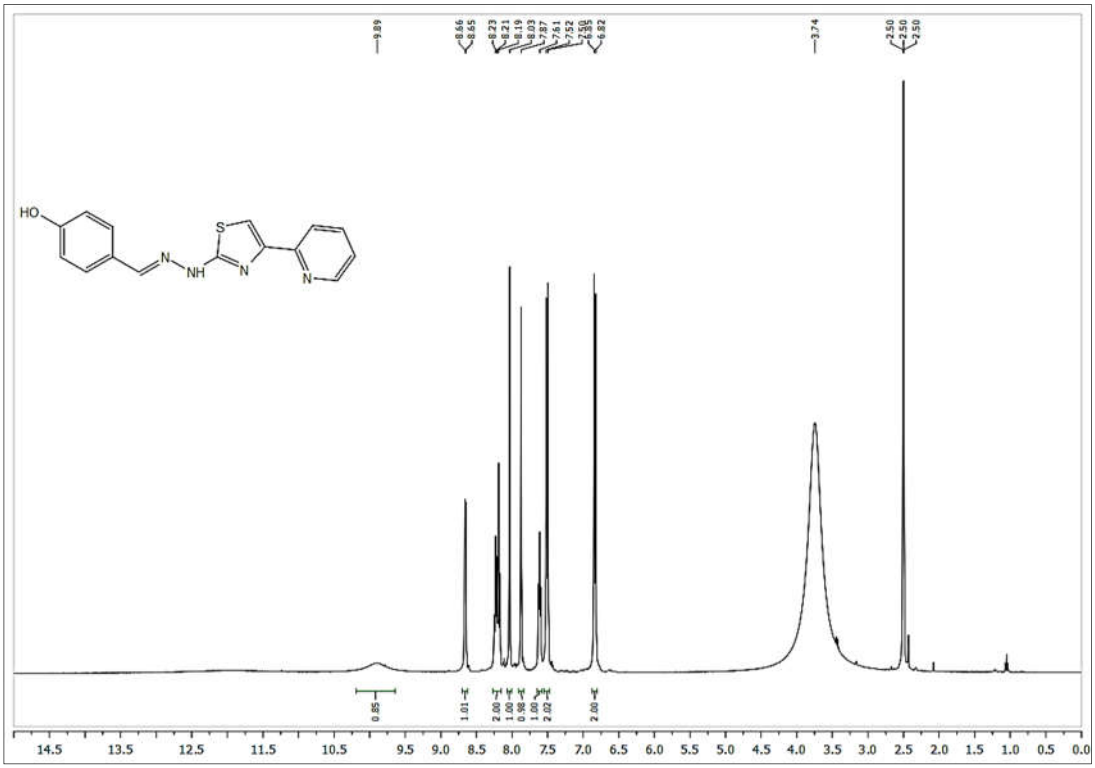
Mass Spectrum of (E)-3-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol (**19b**)



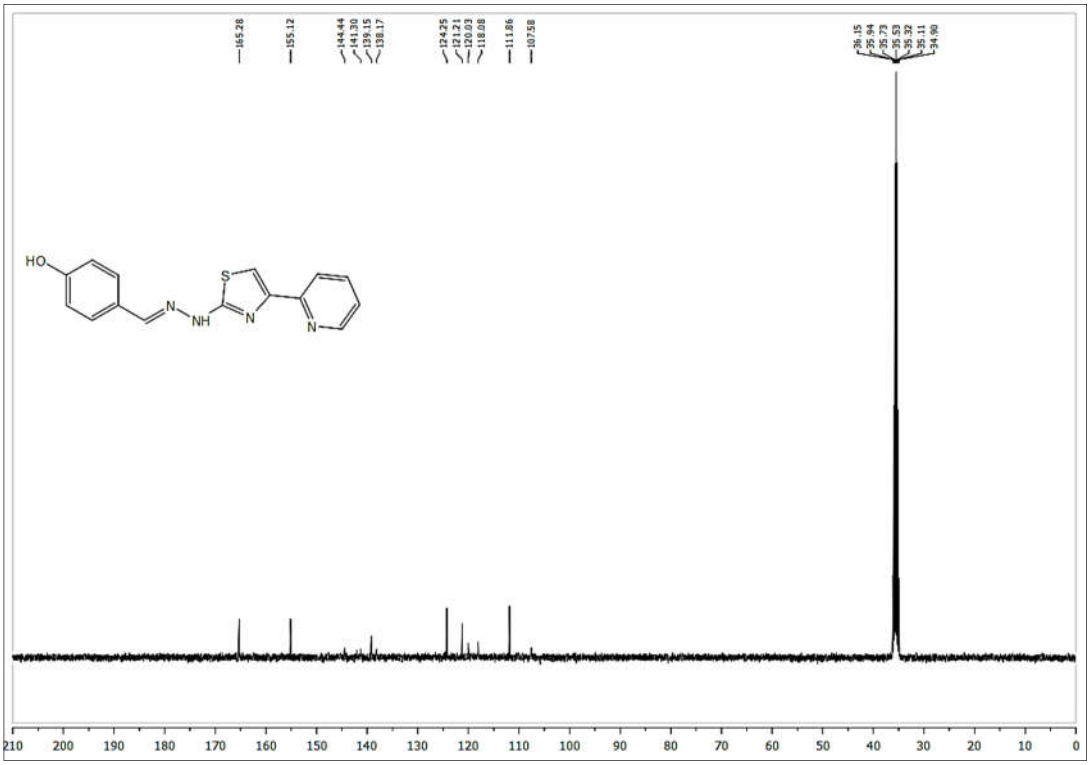
IR Spectrum of (E)-3-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, **(19b)**



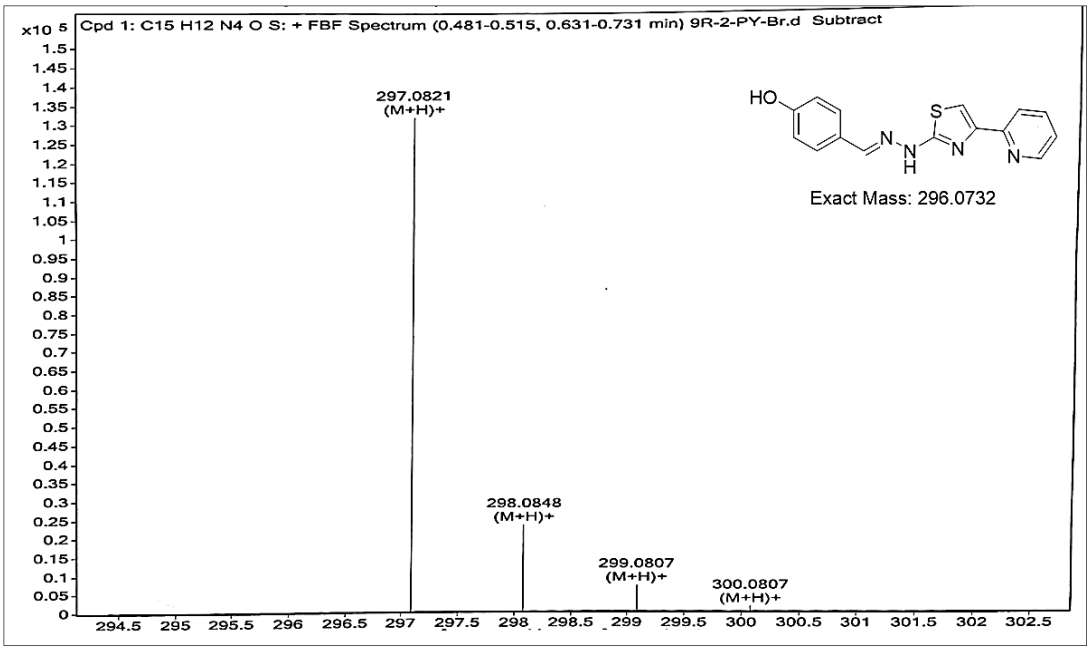
^{1}H NMR Spectrum of (E)-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl) phenol, **(20b)**



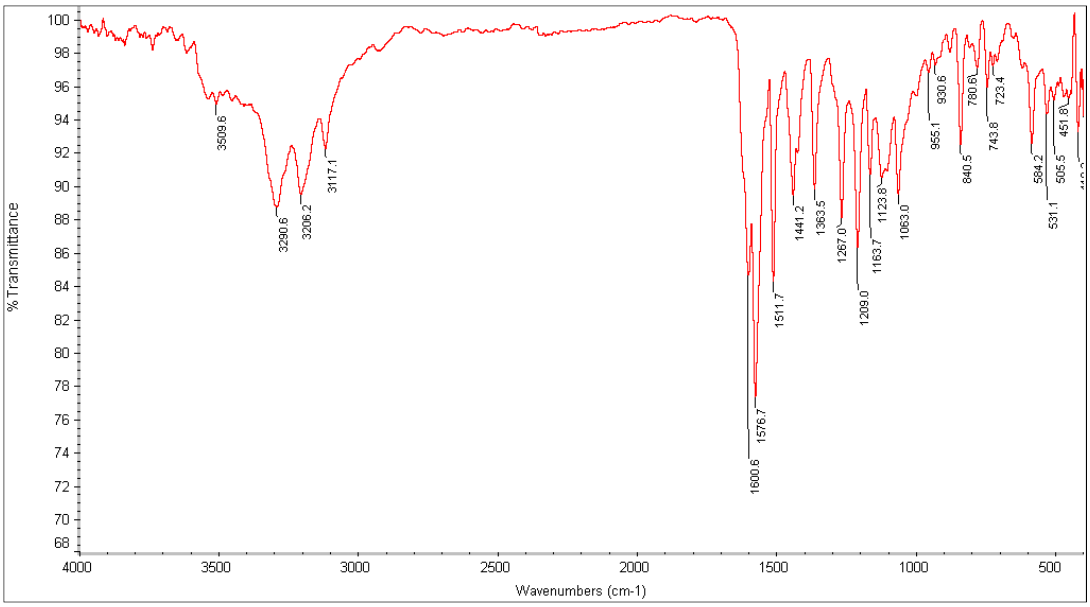
^{13}C NMR Spectrum of (E)-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, **(20b)**



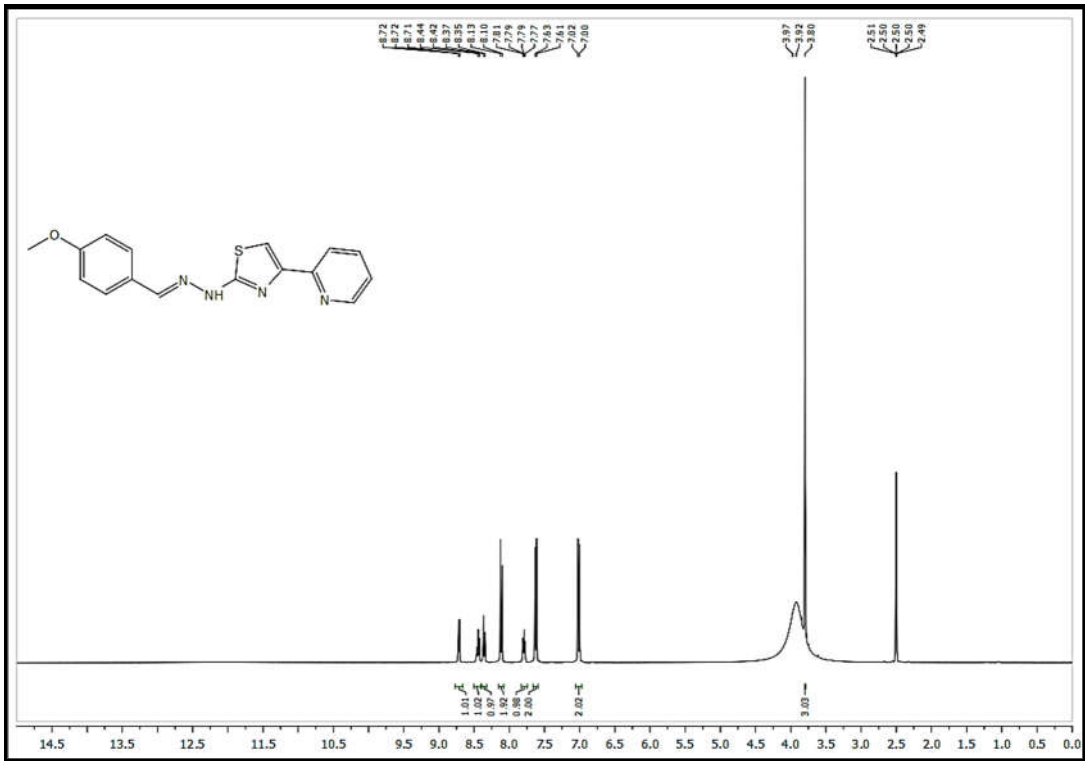
Mass Spectrum of (E)-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl) phenol, **(20b)**



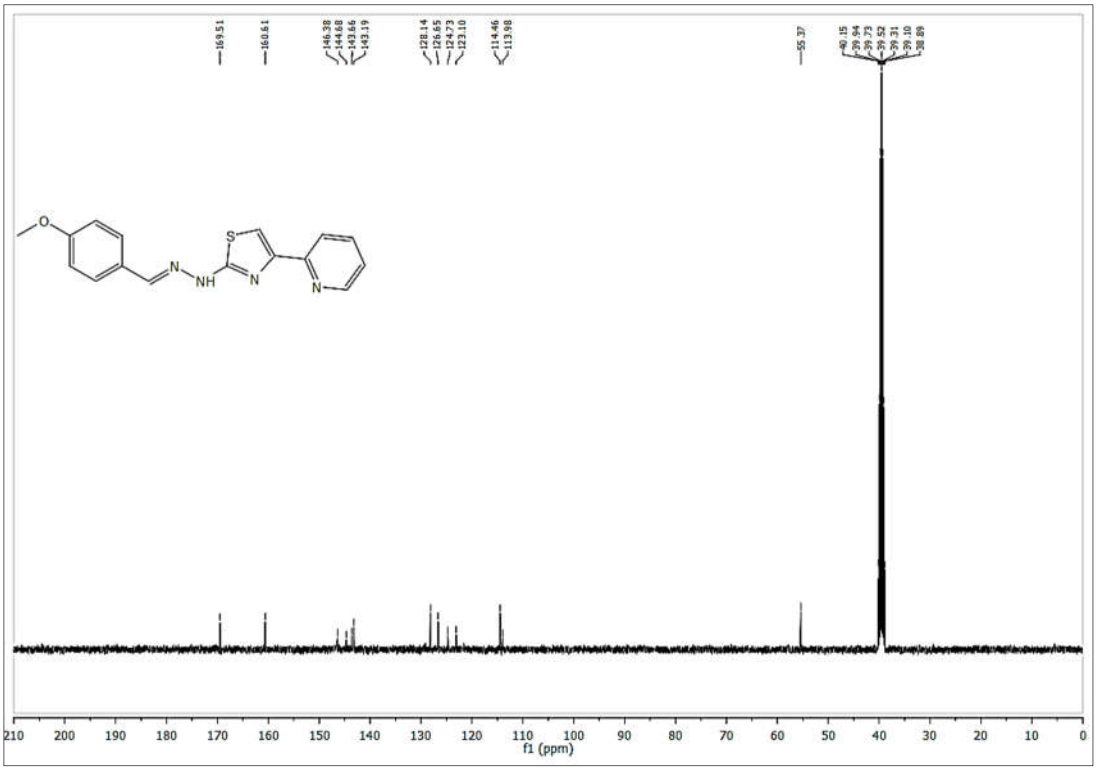
IR Spectrum of (E)-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl) phenol, **(20b)**



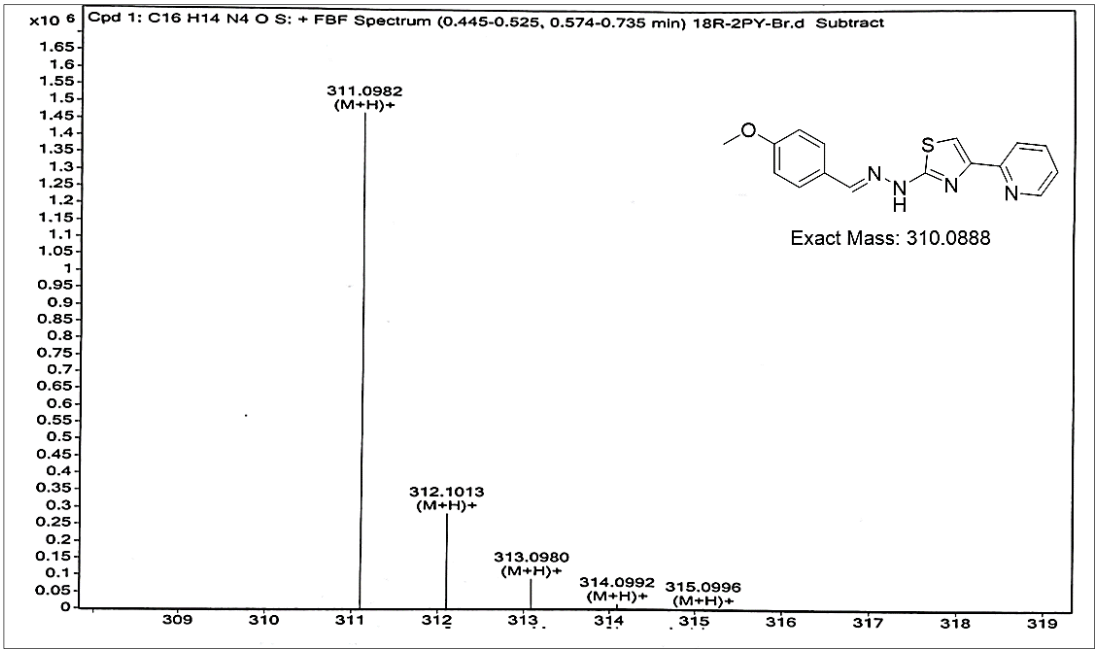
^{1}H NMR Spectrum of (E)-2-(2-(4-methoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl) thiazole, (**21b)**



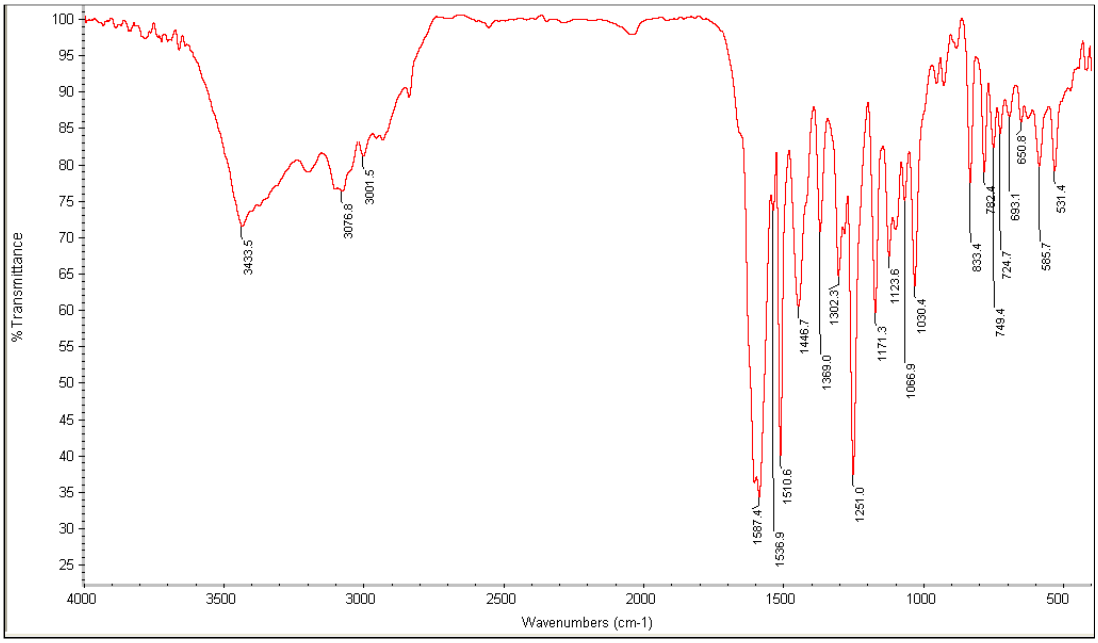
^{13}C NMR Spectrum of (E)-2-(2-(4-methoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, (**21b)**



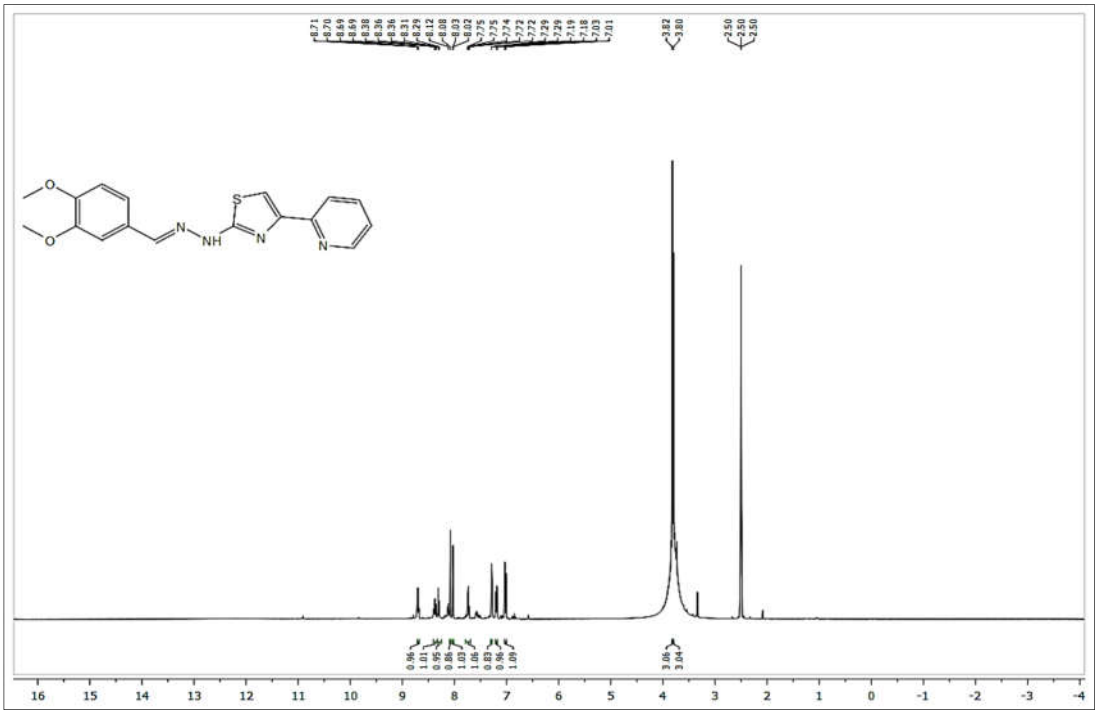
Mass Spectrum of (E)-2-(2-(4-methoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**21b)**



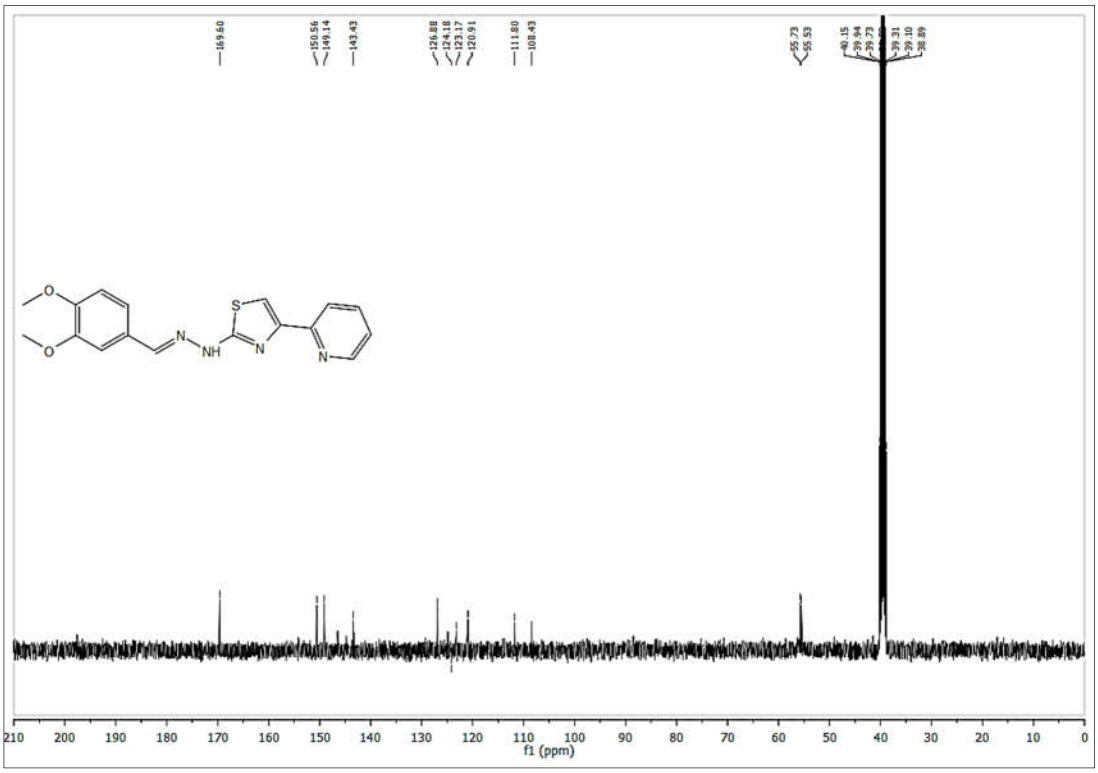
IR Spectrum of (E)-2-(2-(4-methoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, (**21b)**



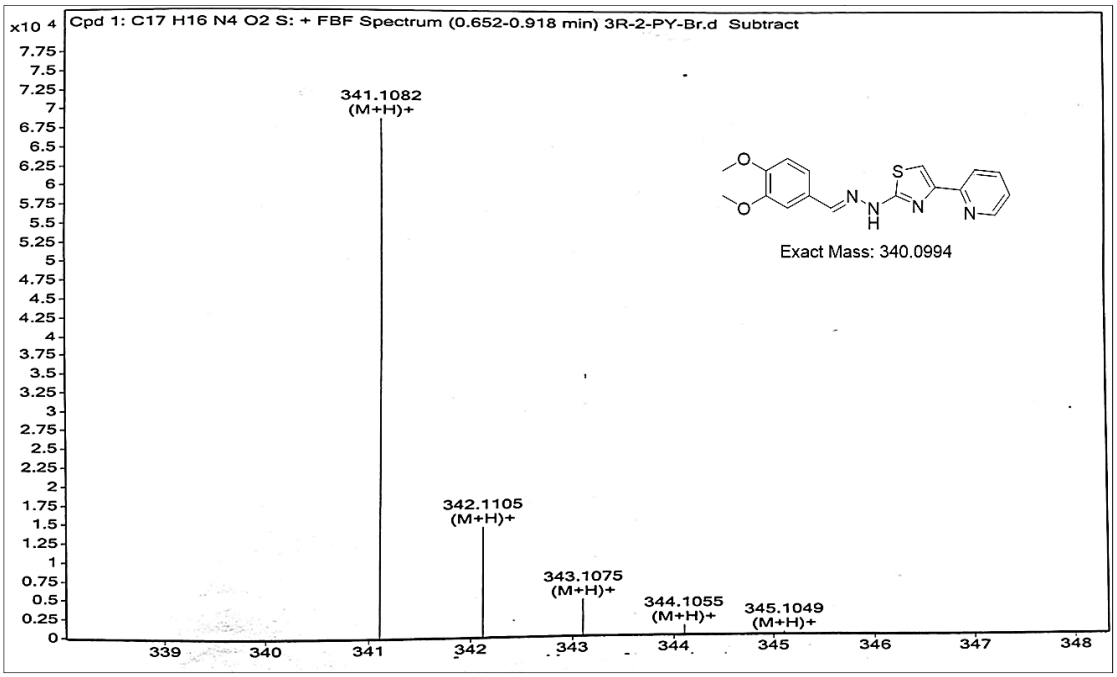
^{1}H NMR Spectrum of (E)-2-(2-(3,4-dimethoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(22b)**



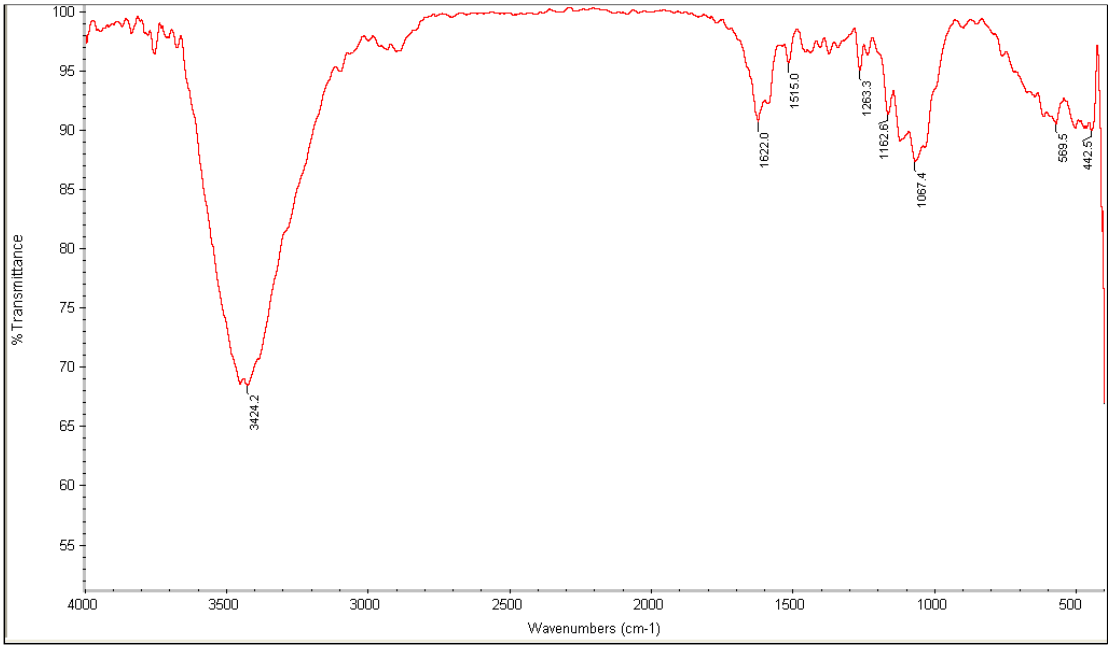
^{13}C NMR Spectrum of (E)-2-(2-(3,4-dimethoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(22b)**



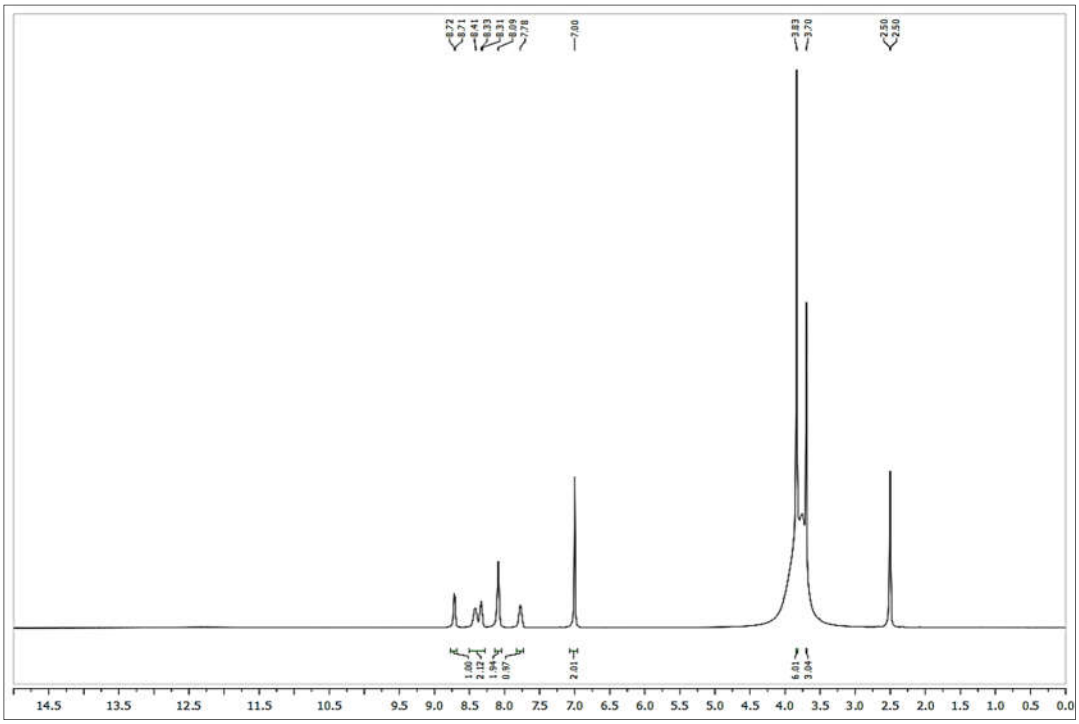
Mass Spectrum of (E)-2-(2-(3,4-dimethoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**22b**)



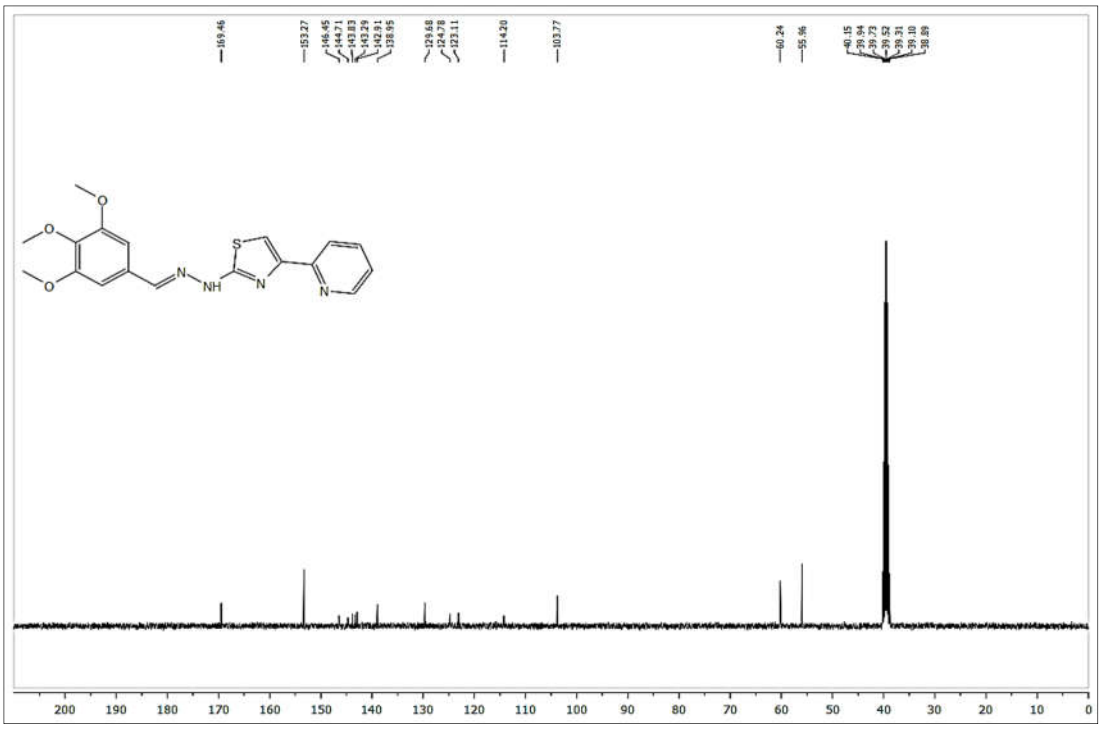
IR Spectrum of (E)-2-(2-(3,4-dimethoxybenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(22b)**



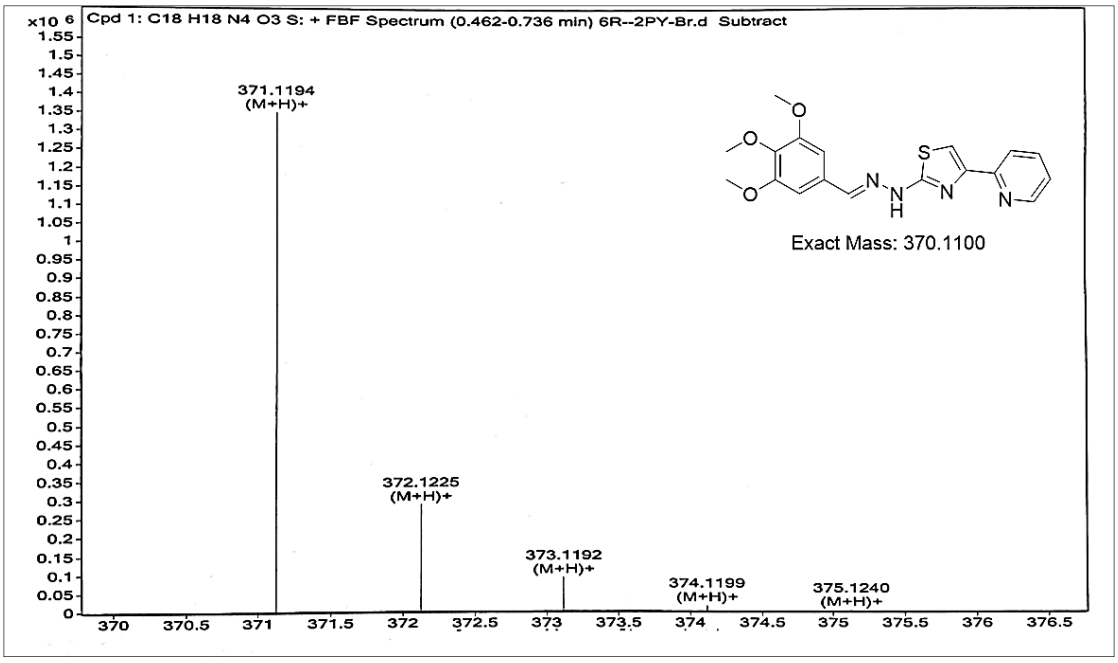
^{1}H NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(3,4,5-trimethoxybenzylidene)hydrazinyl) thiazole, **(23b)**



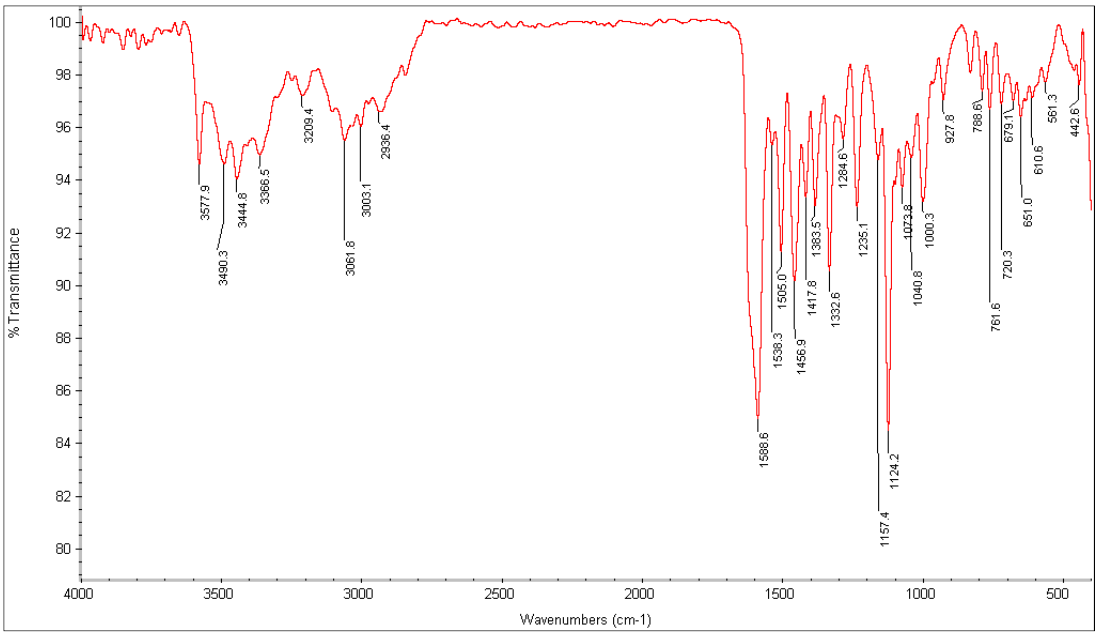
^{13}C NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(3,4,5-trimethoxybenzylidene) hydrazinyl)thiazole, **(23b)**



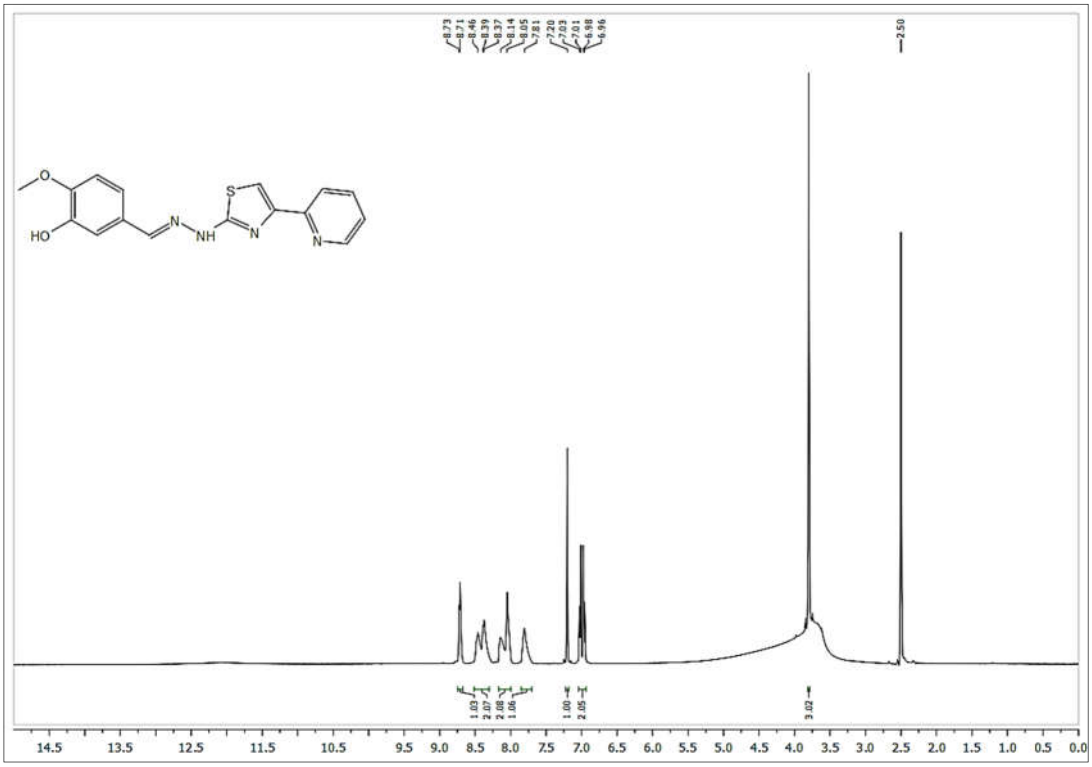
Mass Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(3,4,5-trimethoxybenzylidene)hydrazinyl) thiazole (**23b**)



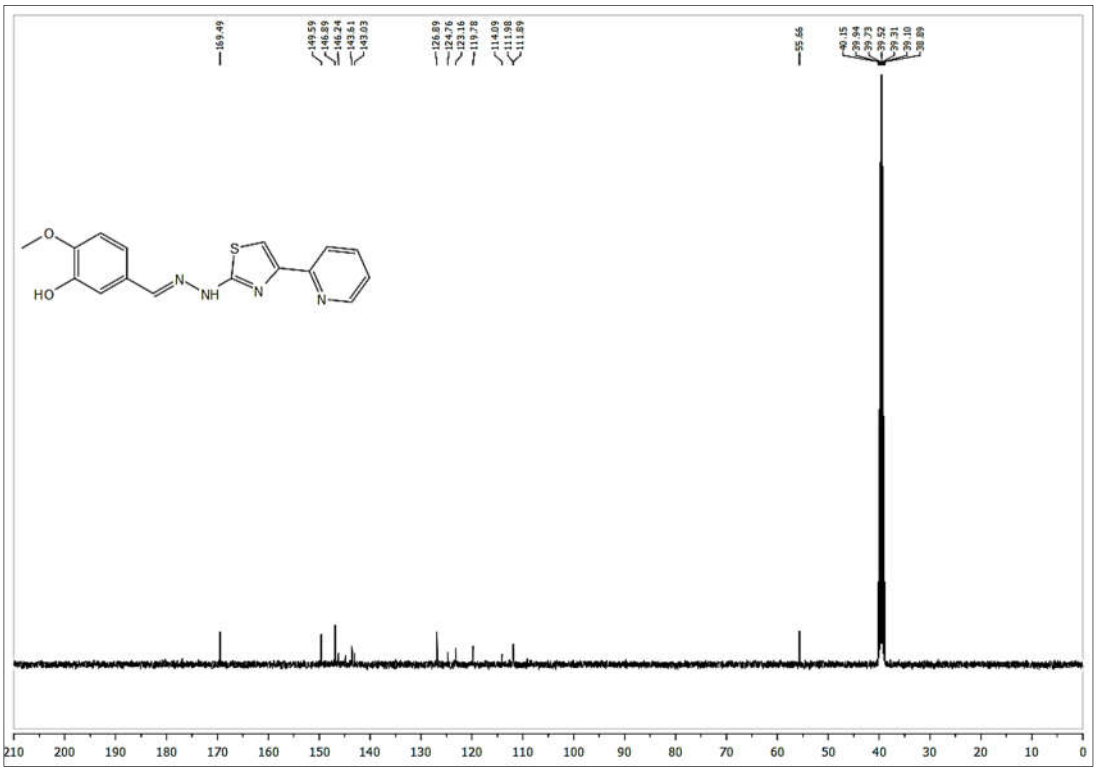
IR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(3,4,5-trimethoxybenzylidene) hydrazinyl)thiazole, **(23b)**



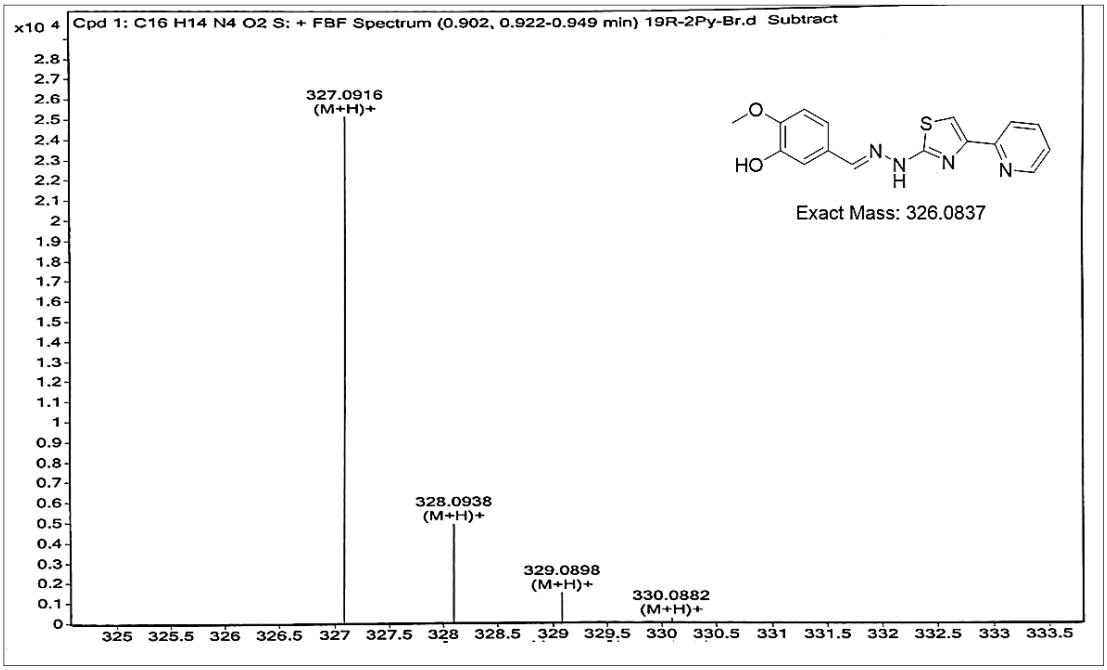
^{1}H NMR Spectrum of (E)-2-methoxy-5-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl) phenol, (**24b)**



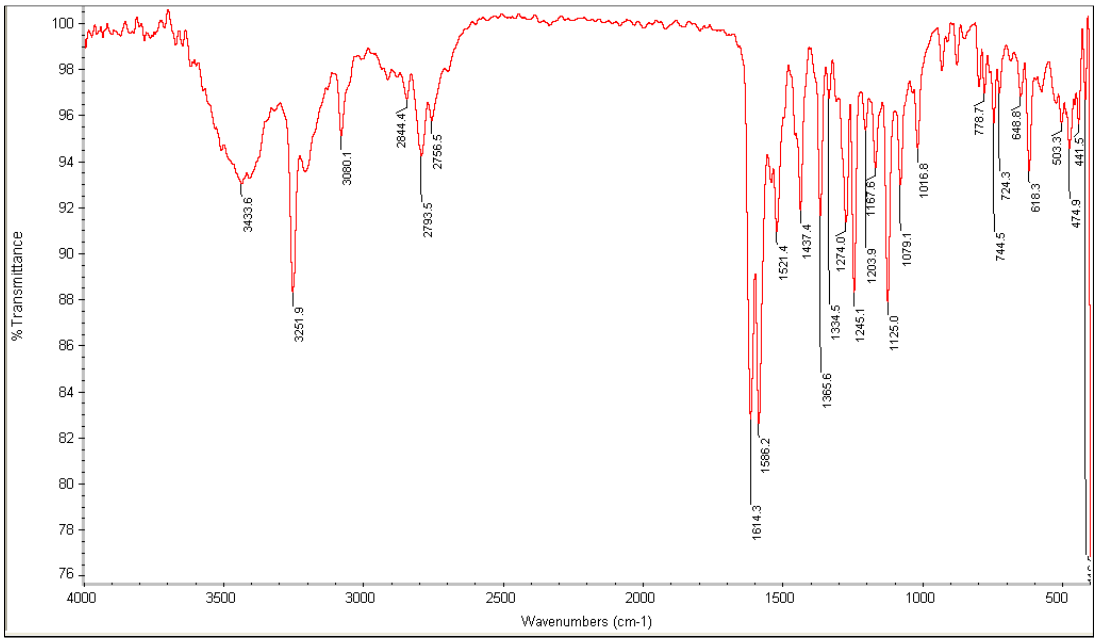
^{13}C NMR Spectrum of (E)-2-methoxy-5-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, (**24b)**



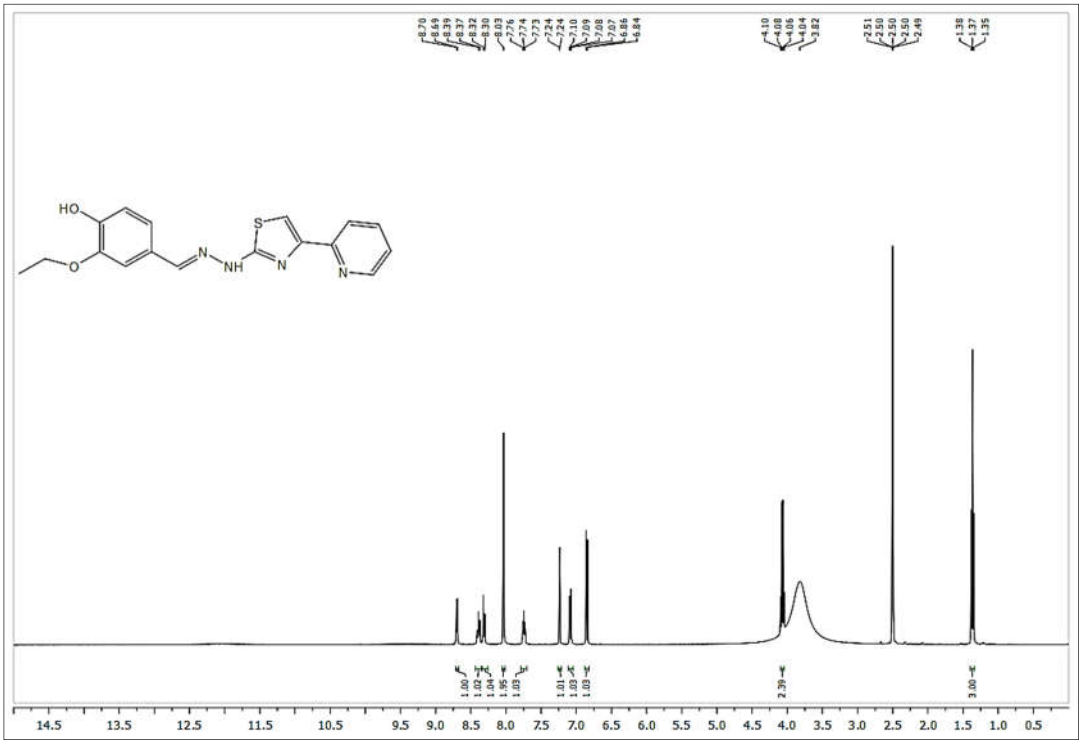
Mass Spectrum of (E)-2-methoxy-5-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl) phenol (**24b)**



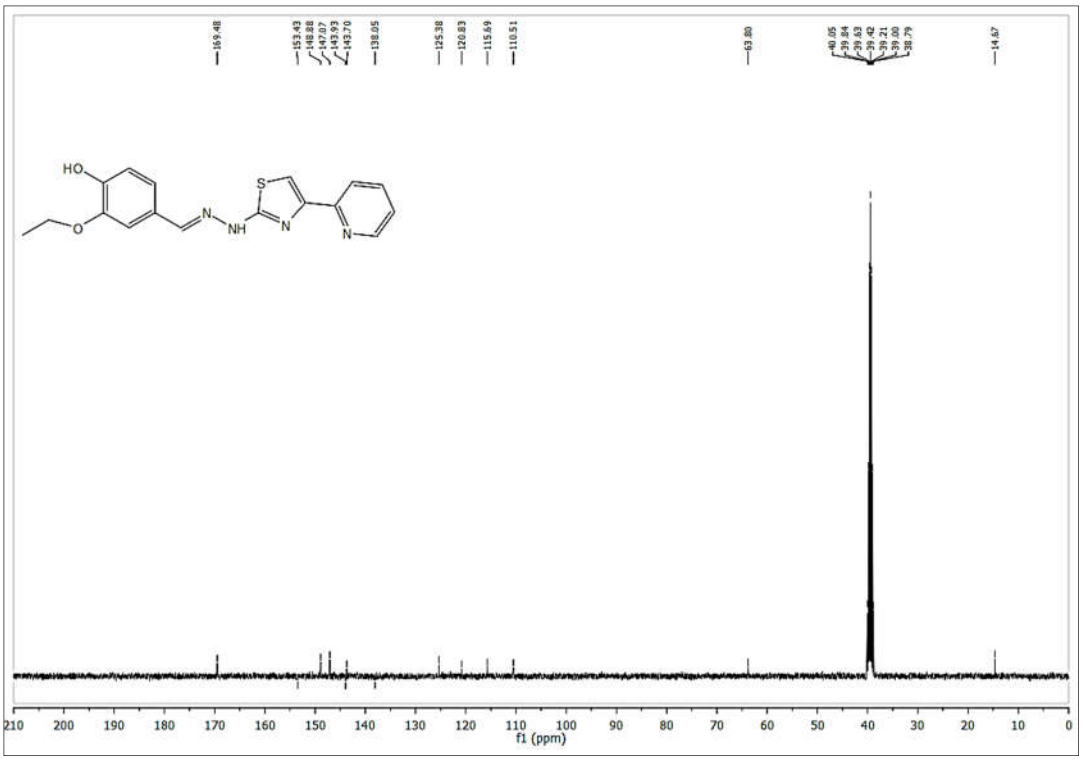
IR Spectrum of (E)-2-methoxy-5-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, (**24b)**



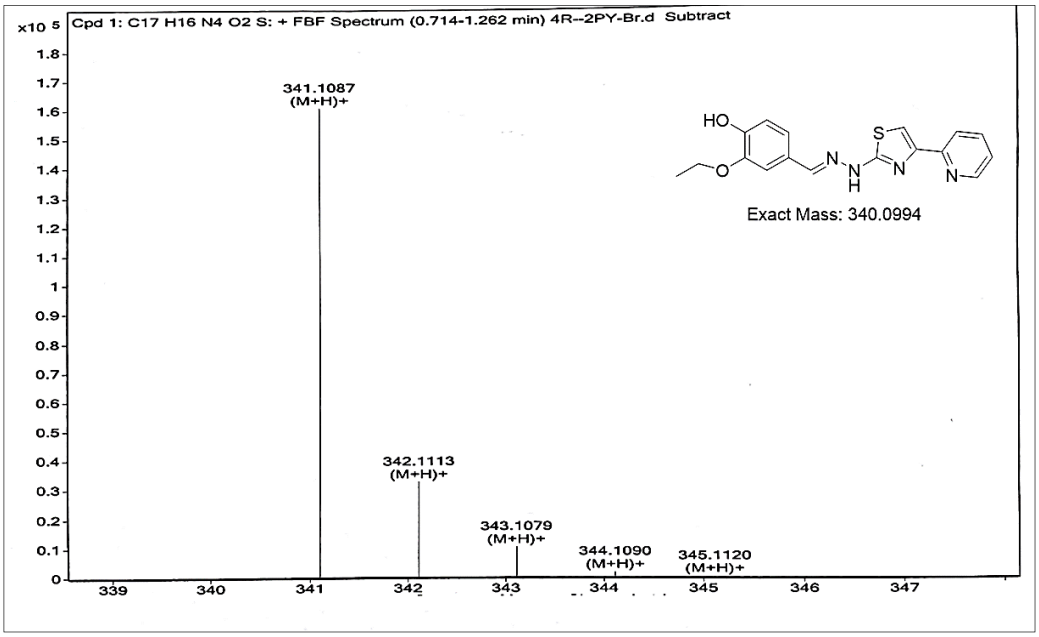
^{1}H NMR Spectrum of (E)-2-ethoxy-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl) phenol, **(25b)**



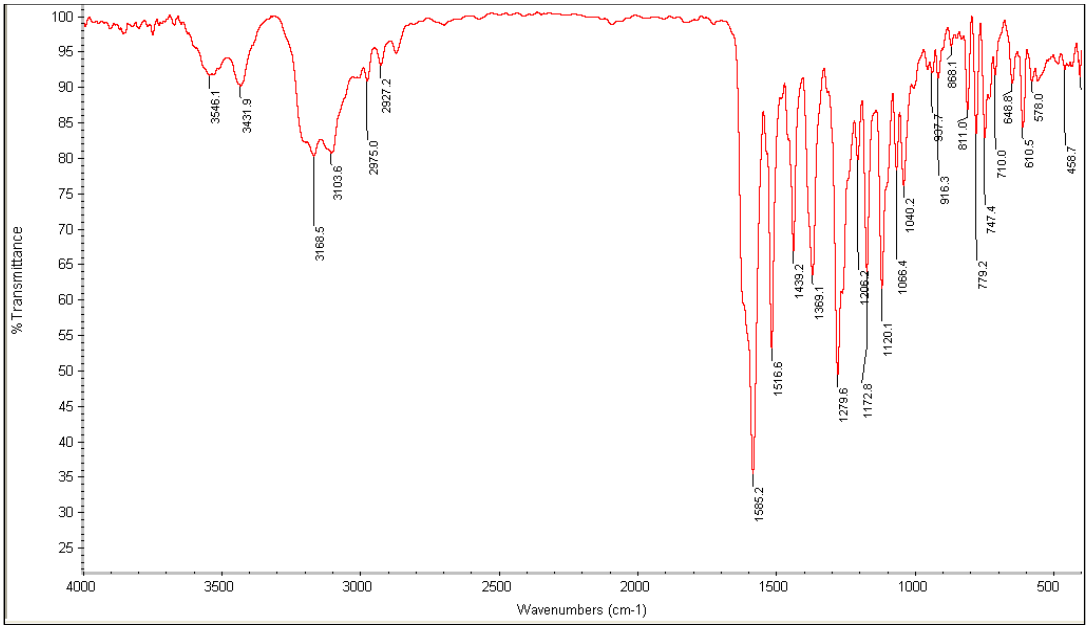
^{13}C NMR Spectrum of (E)-2-ethoxy-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)phenol, **(25b)**



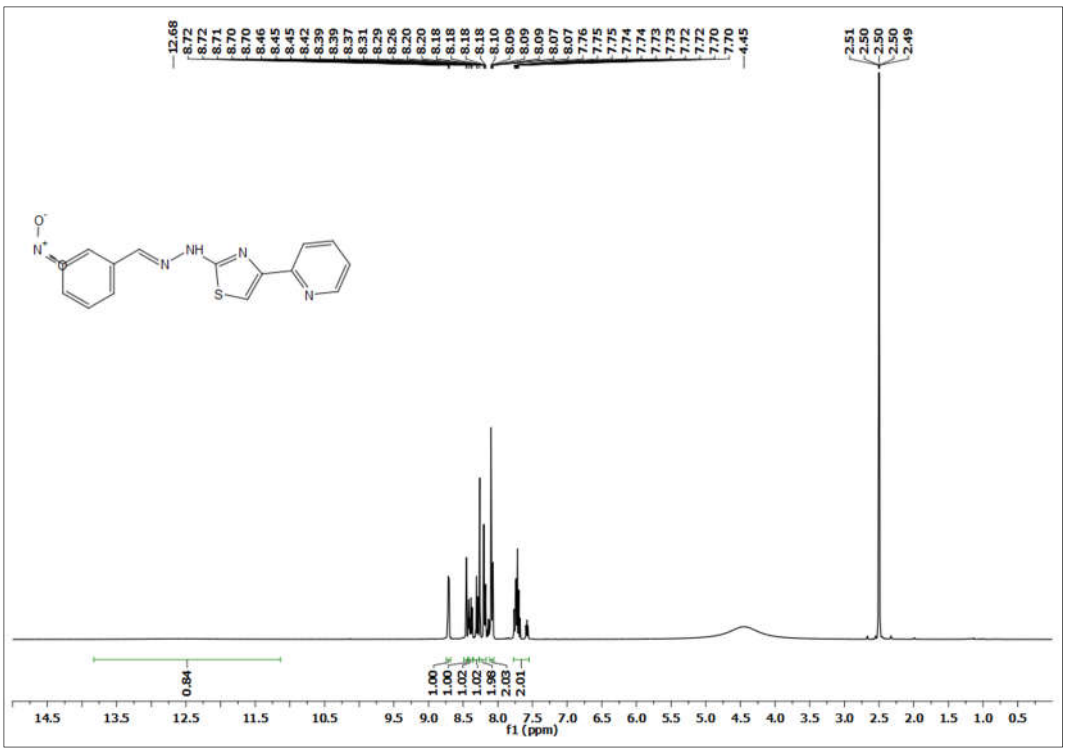
Mass Spectrum of (E)-2-ethoxy-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol (**25b**)



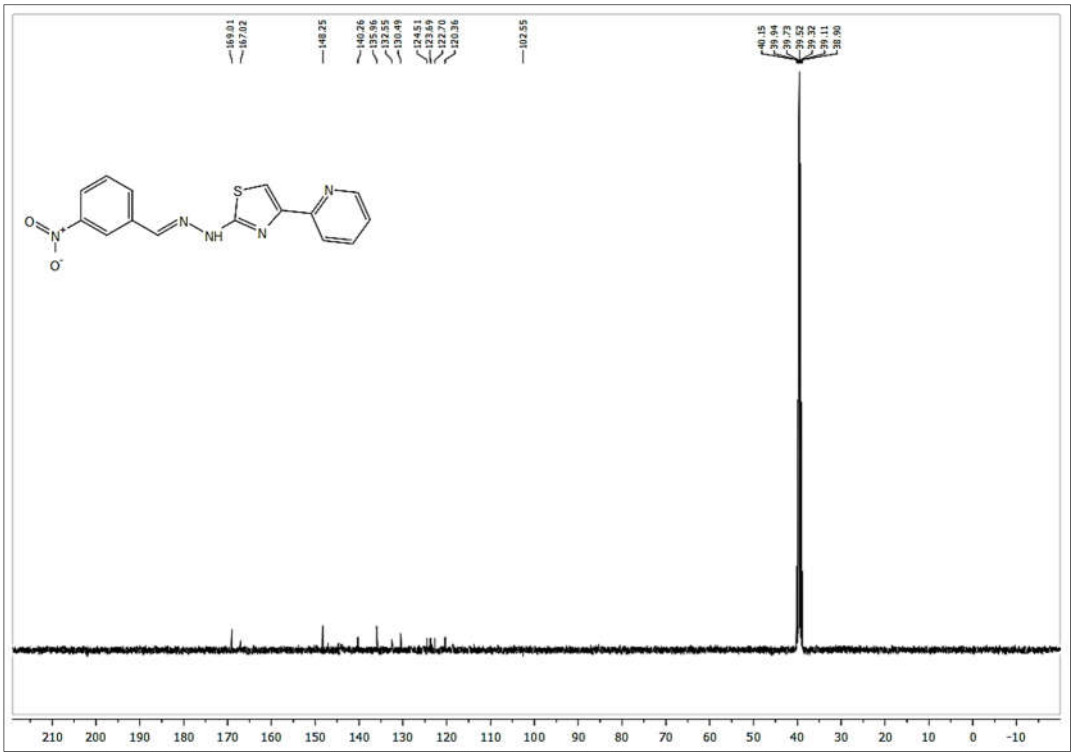
IR Spectrum of (E)-2-ethoxy-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)phenol, **(25b)**



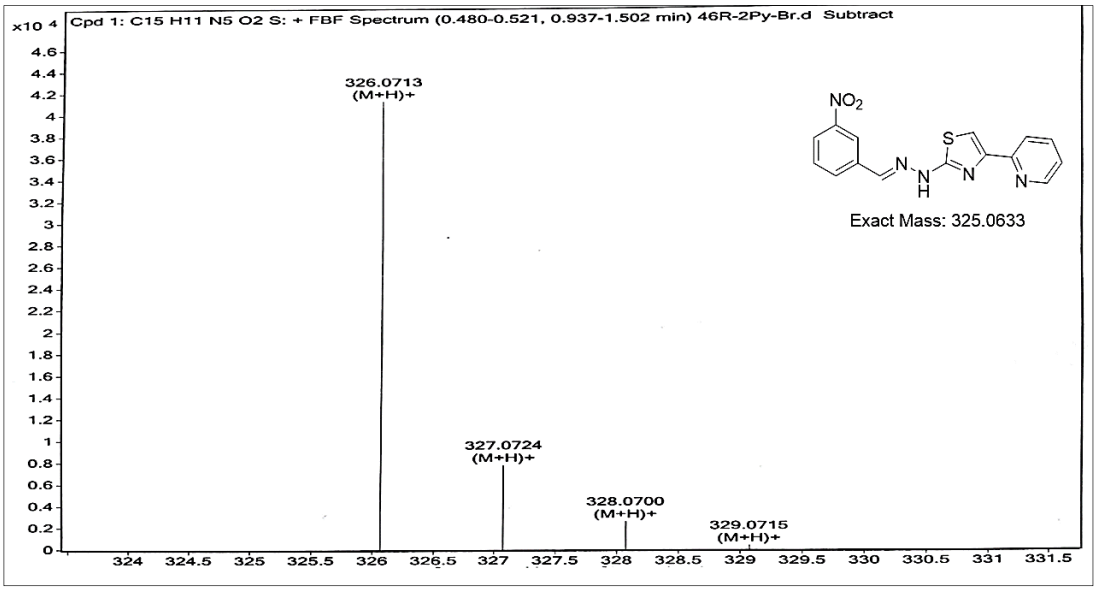
^{1}H NMR Spectrum of (E)-2-(2-(3-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(26b)**



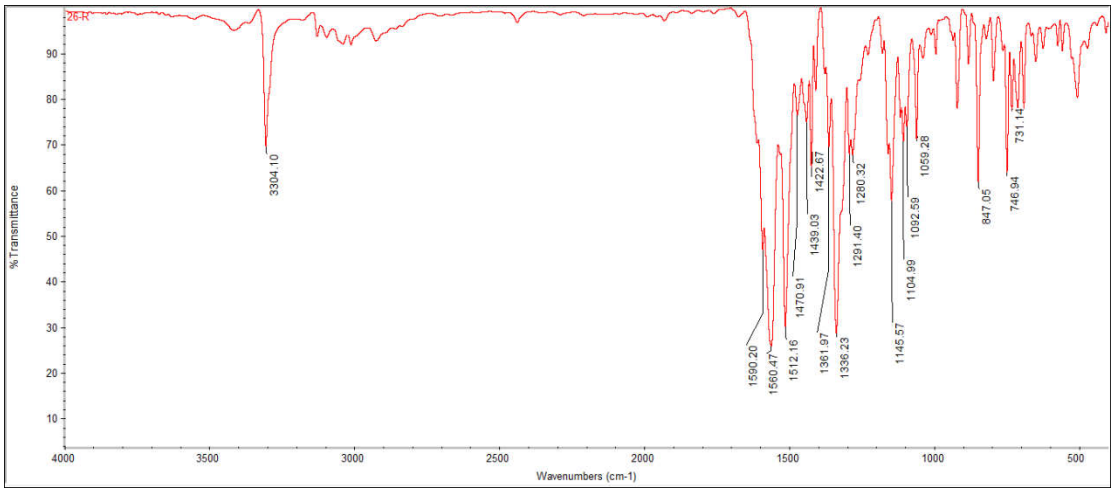
^{13}C NMR Spectrum of (E)-2-(2-(3-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(26b)**



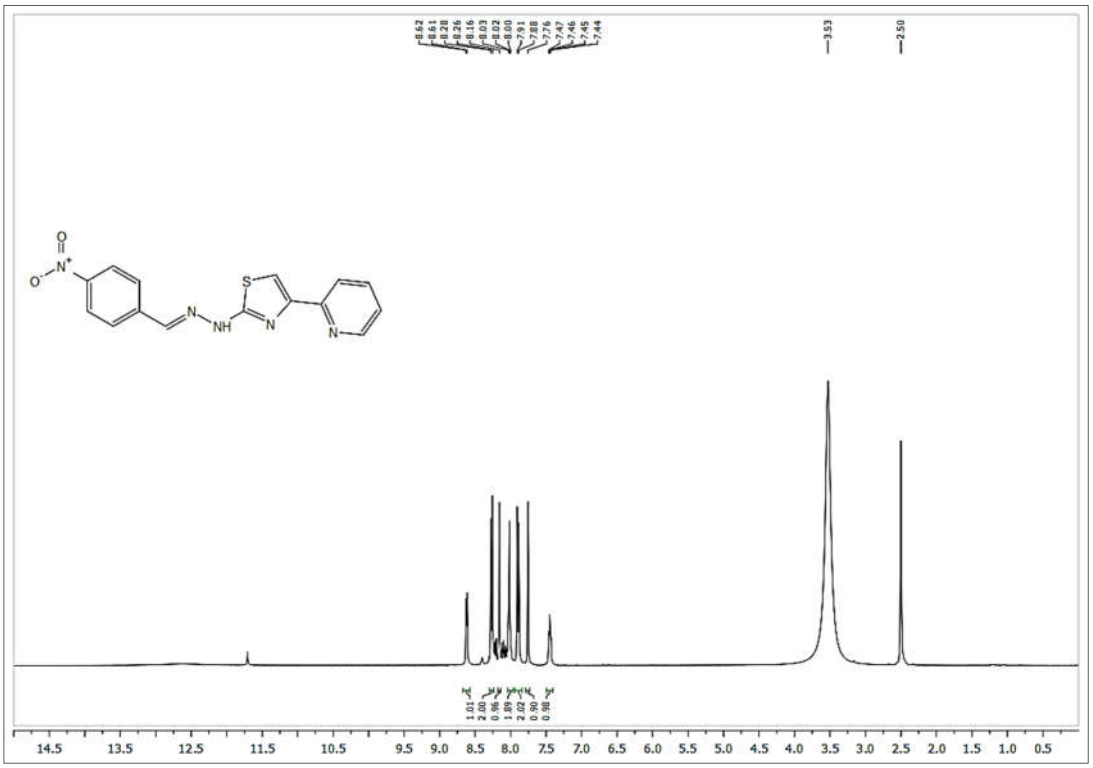
Mass Spectrum of (E)-2-(2-(3-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(26b)**



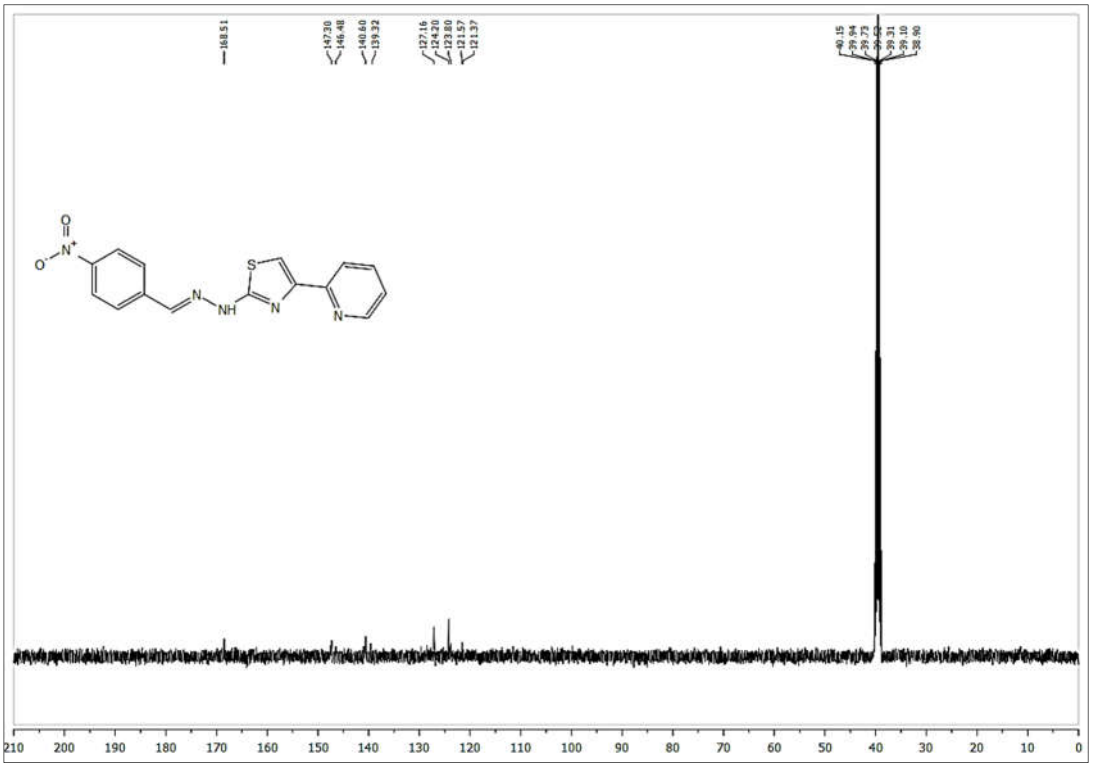
IR Spectrum of (E)-2-(2-(3-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(26b)**



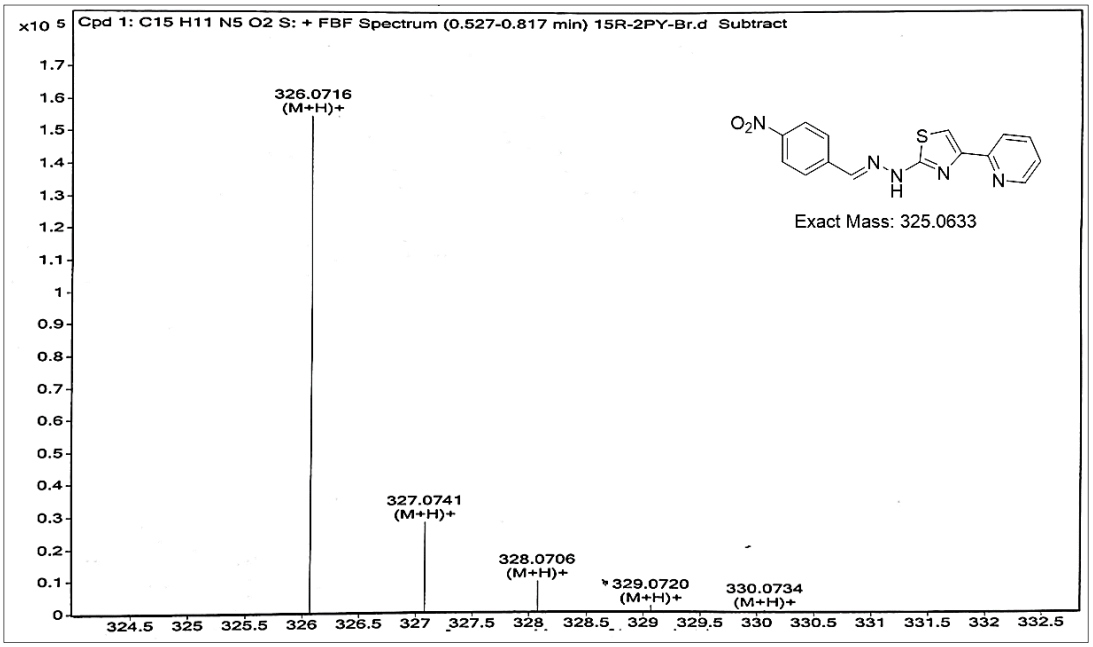
^{1}H NMR Spectrum of (E)-2-(2-(4-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(27b)**



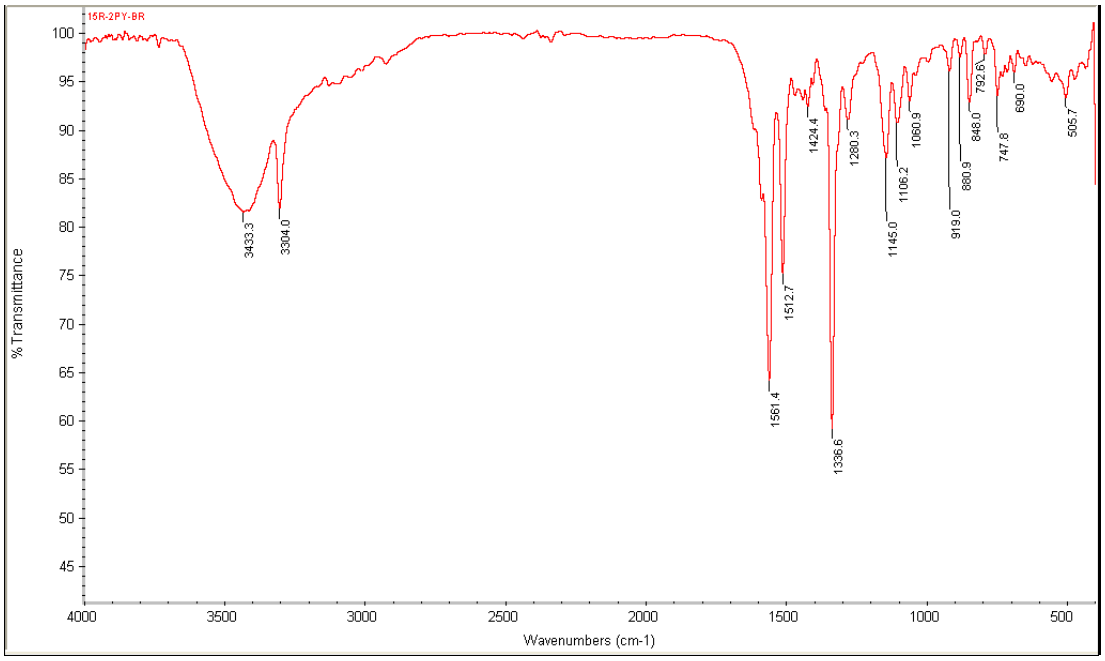
^{13}C NMR Spectrum of (E)-2-(2-(4-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(27b)**



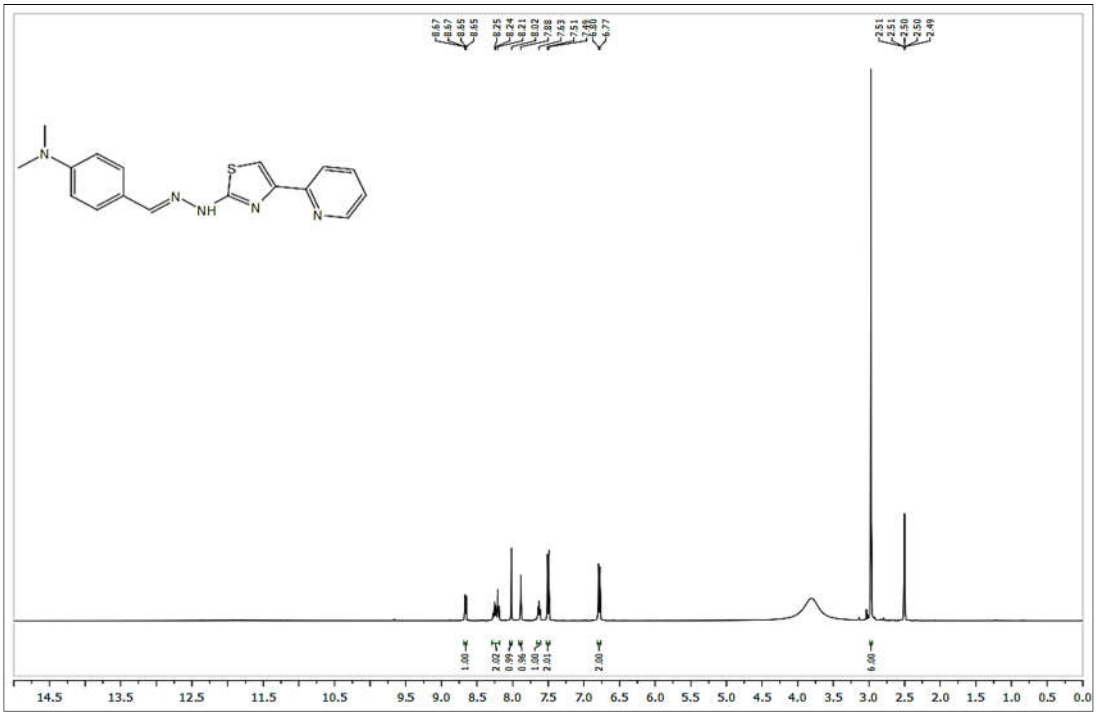
Mass Spectrum of (E)-2-(2-(4-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole (**27b)**



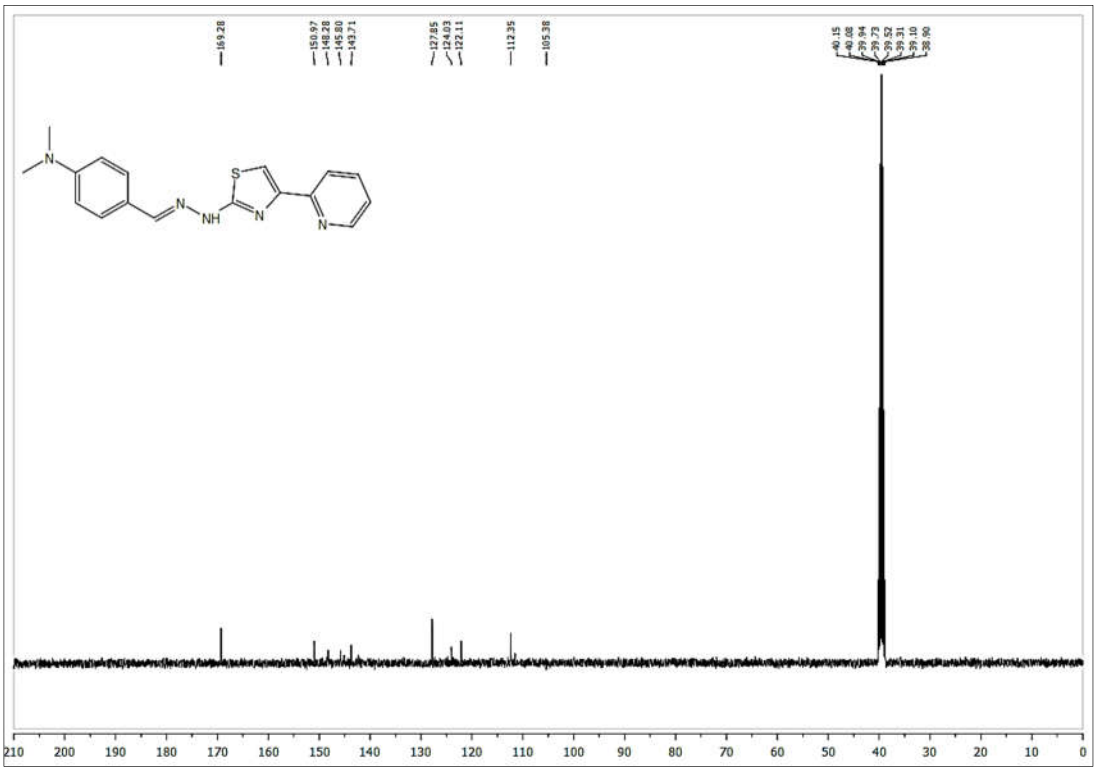
IR Spectrum of (E)-2-(2-(4-nitrobenzylidene)hydrazinyl)-4-(pyridin-2-yl)thiazole, **(27b)**



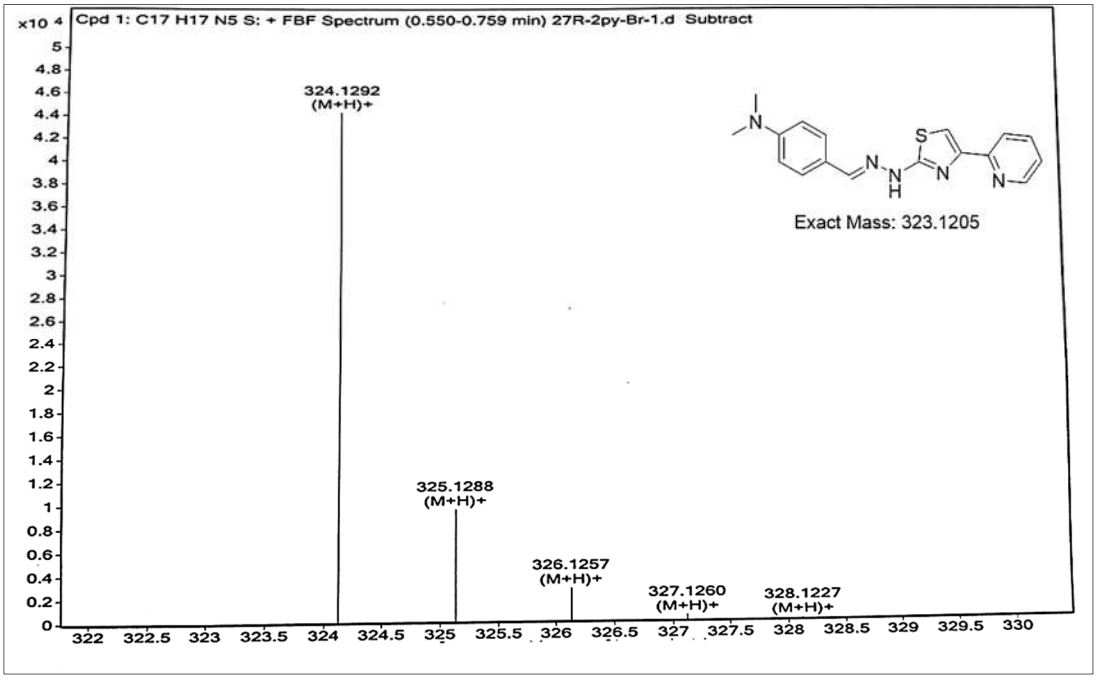
^{1}H NMR Spectrum of (E)-N,N-dimethyl-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl)aniline, (**28b**)



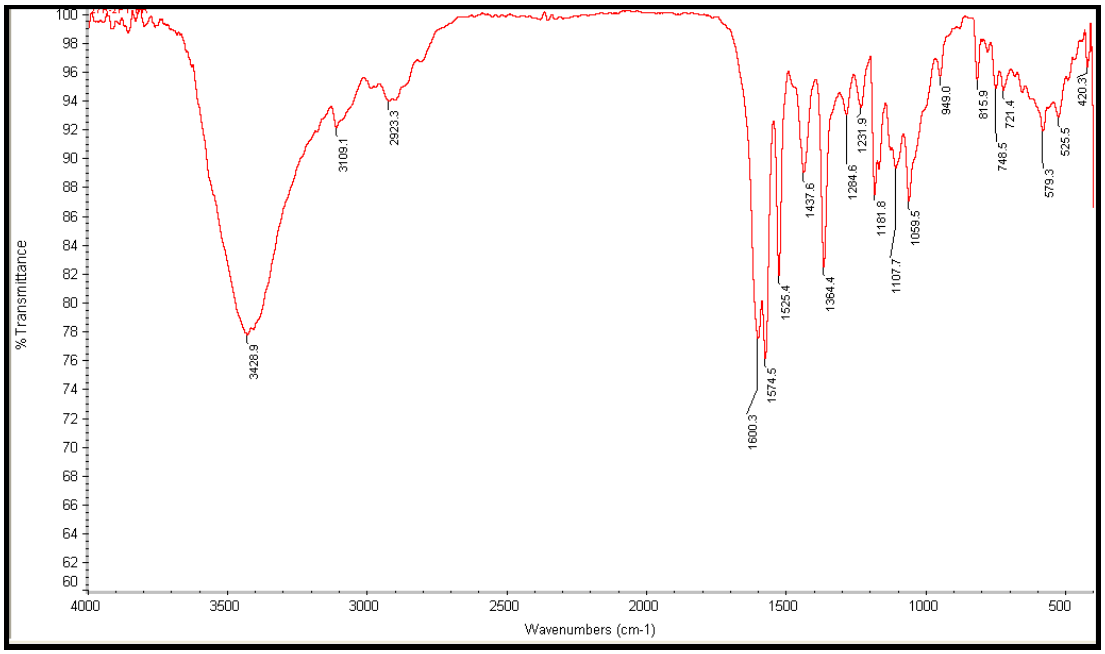
^{13}C NMR Spectrum of (E)-N,N-dimethyl-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono) methyl)aniline, (**28b**)



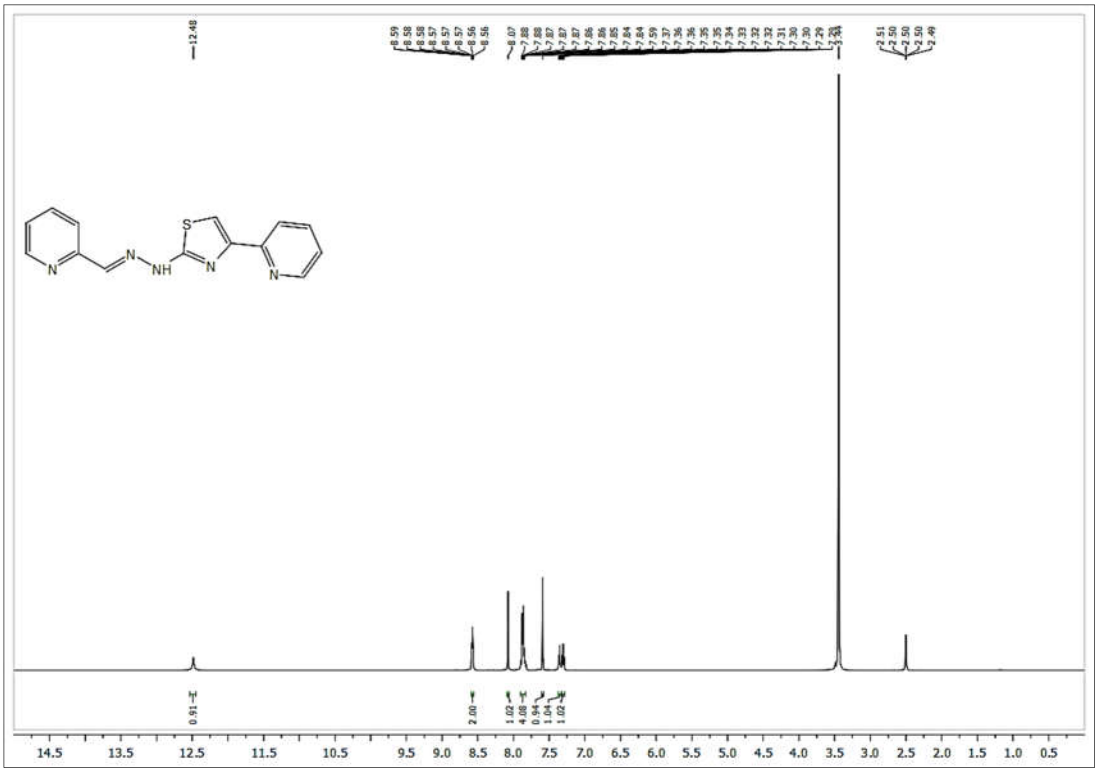
Mass Spectrum of (E)-N,N-dimethyl-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl) aniline (**28b**)



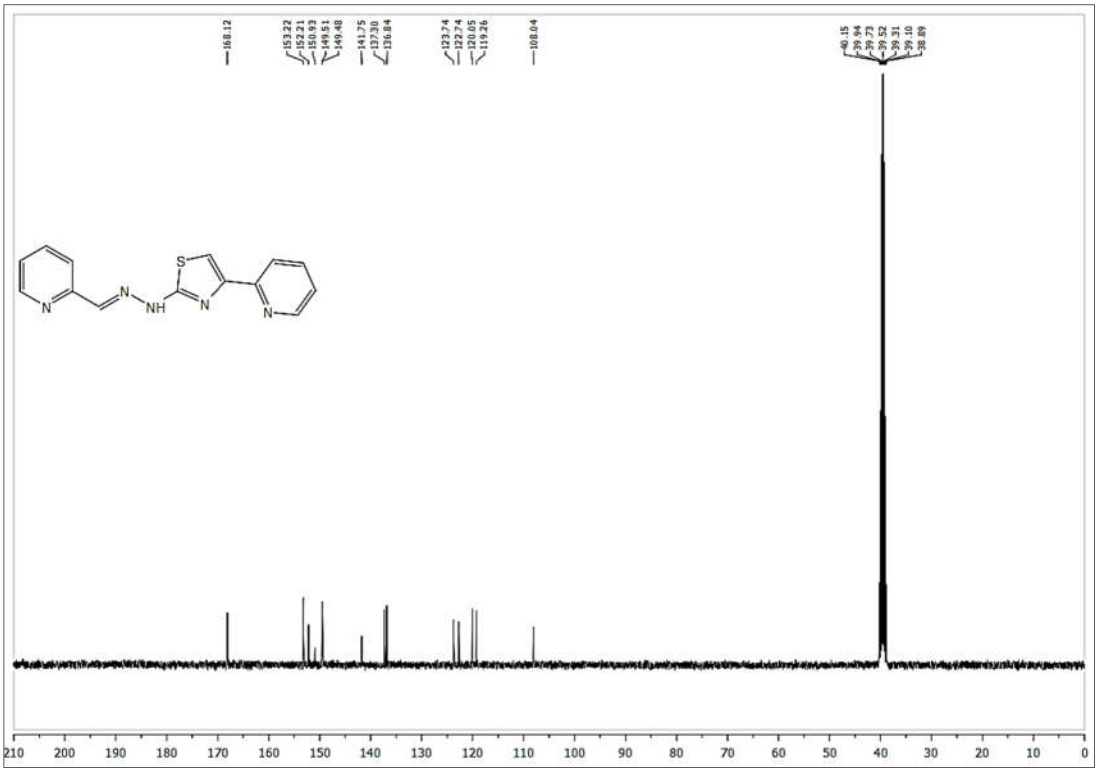
IR Spectrum of (E)-N,N-dimethyl-4-((2-(4-(pyridin-2-yl)thiazol-2-yl)hydrazono)methyl) aniline, (**28b**)



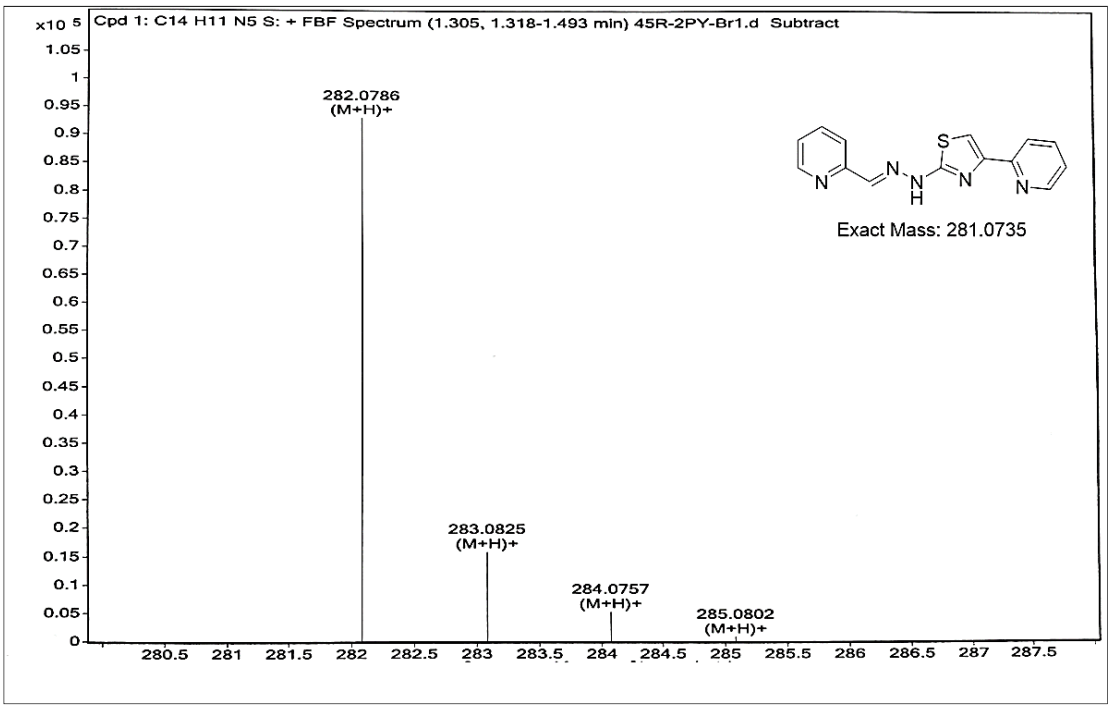
^{1}H NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(pyridin-2-ylmethylene) hydrazinyl)thiazole, **(29b)**



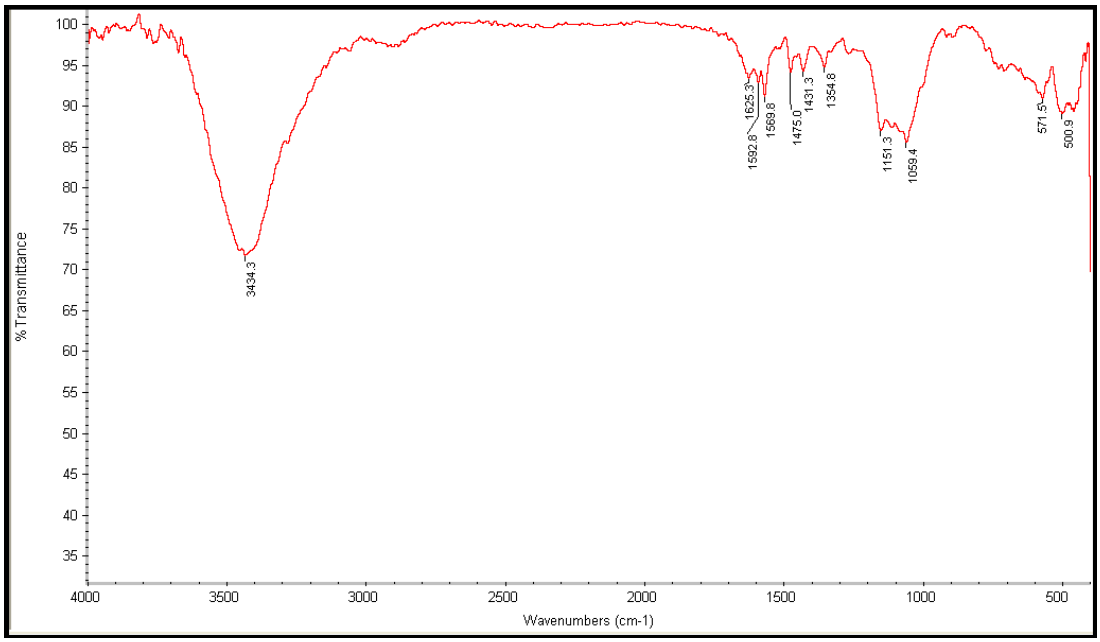
^{13}C NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(pyridin-2-ylmethylene) hydrazinyl)thiazole, **(29b)**



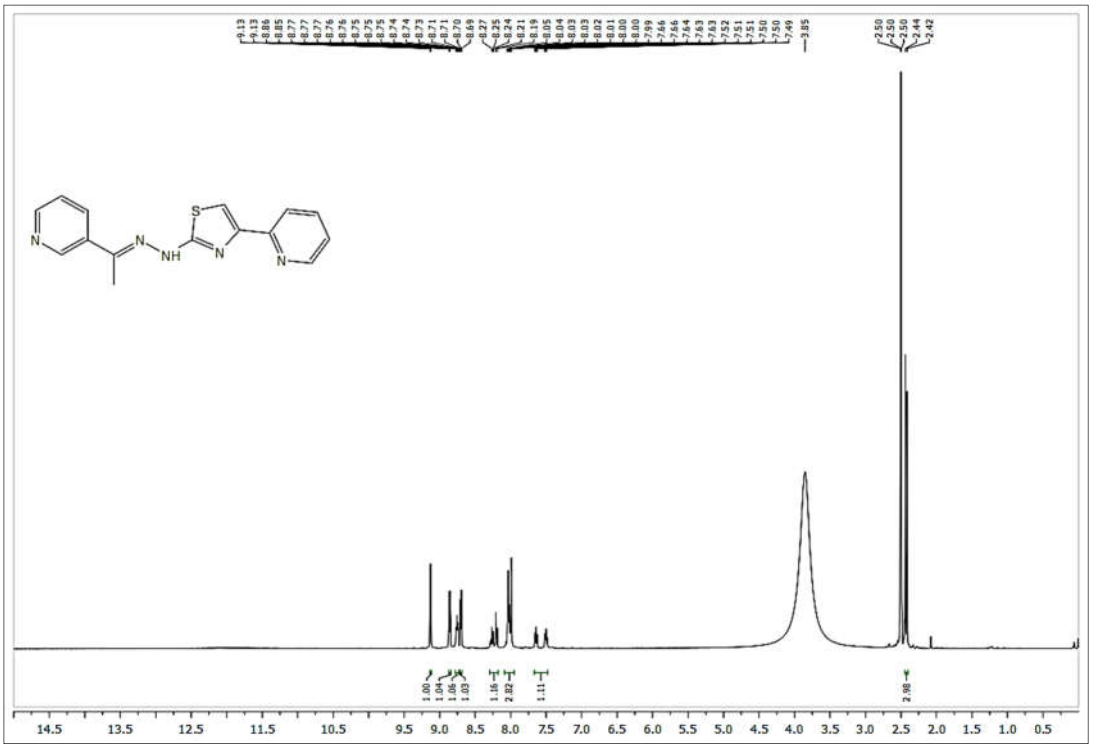
Mass Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(pyridin-2-ylmethylene)hydrazinyl)thiazole (**29b**)



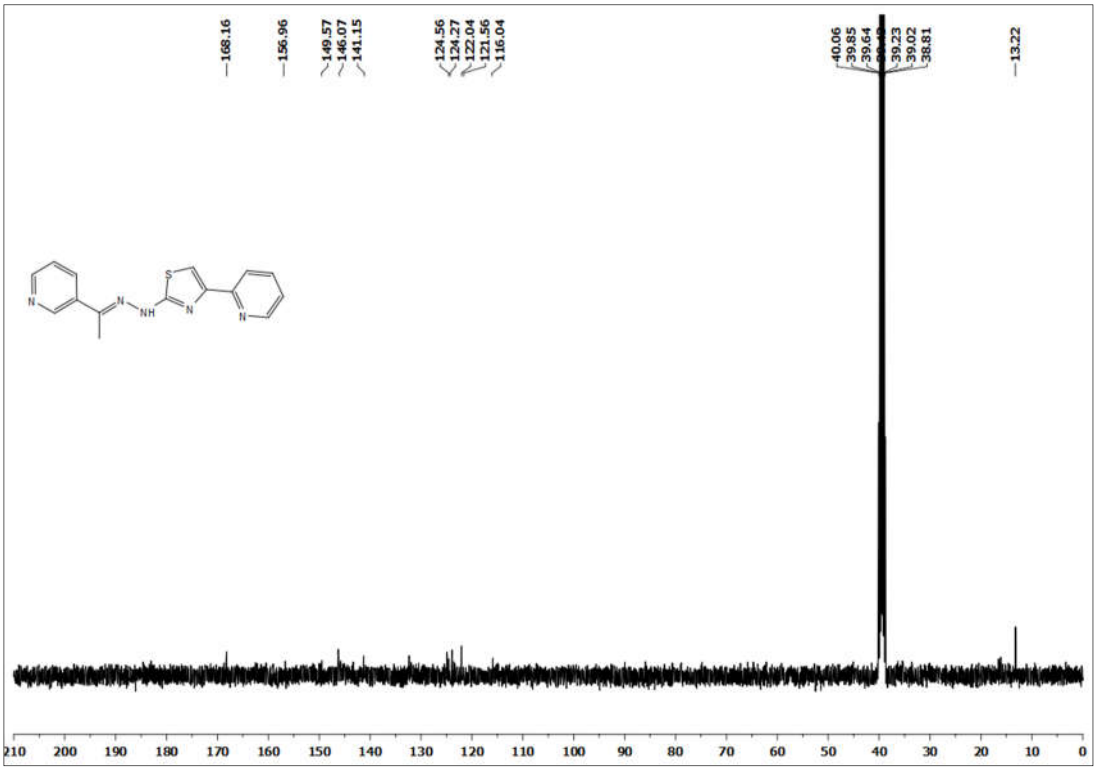
IR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(pyridin-2-ylmethylene)hydrazinyl) thiazole, **(29b)**



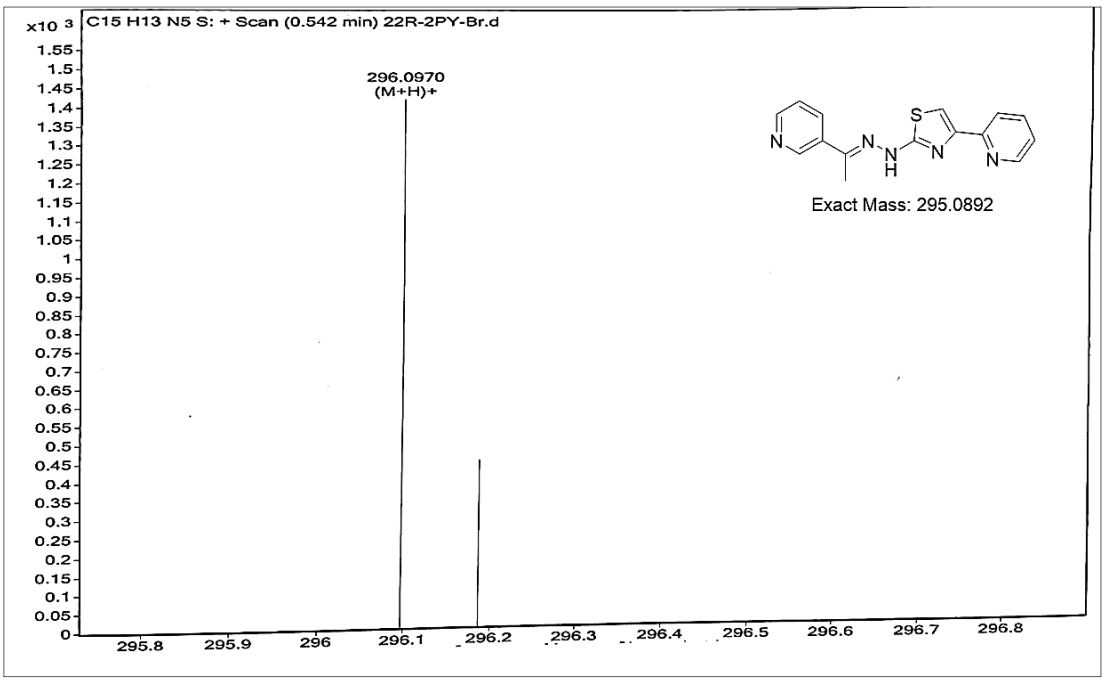
^{1}H NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(1-(pyridin-3-yl)ethylidene) hydrazinyl)thiazole, **(30b)**



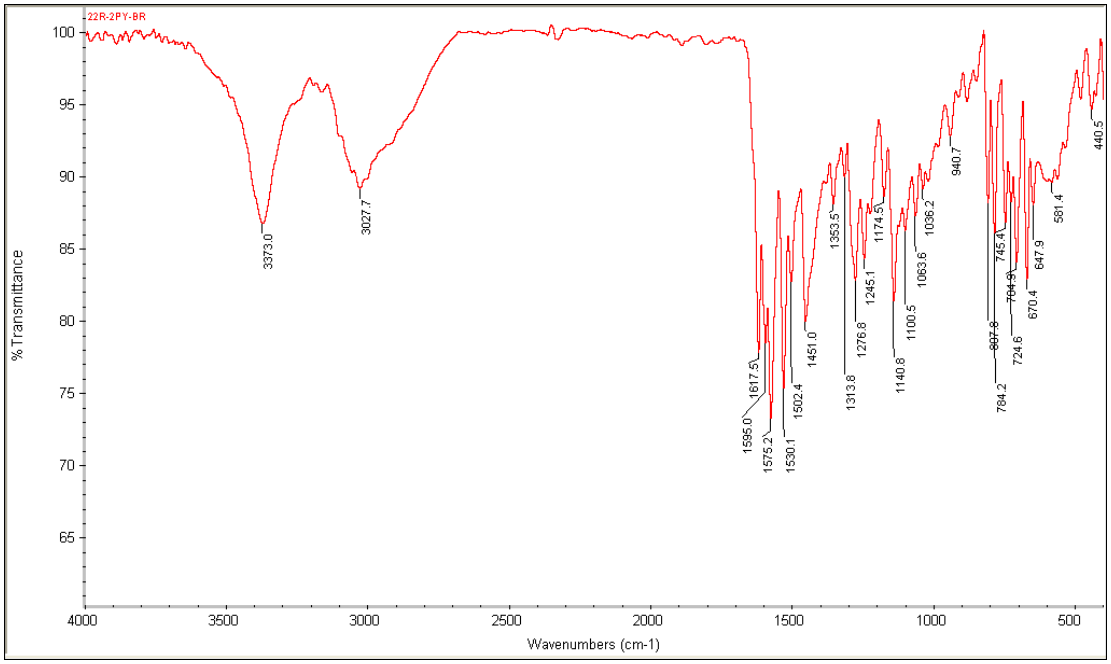
^{13}C NMR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(1-(pyridin-3-yl)ethylidene) hydrazinyl)thiazole, **(30b)**



Mass Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(1-(pyridin-3-yl)ethylidene) hydrazinyl)thiazole, **(30b)**



IR Spectrum of (E)-4-(pyridin-2-yl)-2-(2-(1-(pyridin-3-yl)ethylidene) hydrazinyl) thiazole, **(30b)**



STable 1. Crystal Parameters of the compound 29b.	
Empirical formula	$C_{14}H_{11}N_5S$
Formula weight	281.0735
Temperature/K	293(2)
Crystal system	monoclinic
Space group	$P2_1/c$
a/Å	8.9740(7)
b/Å	28.5391(18)
c/Å	11.4550(9)
α/°	90.00
β/°	112.364(9)
γ/°	90.00
Volume/Å^3	2713.1(3)
Z	13
ρ_{calc}mg/mm^3	1.418
μ/mm^{-1}	0.323
F(000)	1235.0
2Θ range for data collection	5.72 to 58.1°

Index ranges	$-11 \leq h \leq 10, -38 \leq k \leq 35, -14 \leq l \leq 14$
Reflections collected	15232
Independent reflections	6268[R(int) = 0.0347]
Goodness-of-fit on F^2	1.030

STable 2. *In vitro* evaluation of pyridine **appended 2-hydrazinylthiazole derivatives against *Mtb*, H37Rv strain and *in silico* hydrophobic interacting residues of *Mtb,* KasA protein.**

Code	Concentrations in µM (*Mtb* % of inhibition)			Hydrophobic interacting residue
1b	430.46 (61.13)	-	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Pro 316, Ile 317, Gly 318, Phe 402, Phe 404
2b	**356.68 (98.08)**	**89.17 (96.35)**	**7.13 (54.53)**	Arg 214, Met 213, Ala 215, His 311, Gly 318, Phe 402, Thr 313, Pro 280, Val 278, Phe 404, Ile 317, Ala 279
3b	**339.70 (97.88)**	**84.92 (97.91)**	**6.79 (59.54)**	Met 213, Arg 214,Ala 215, Ile 317, Ala 279, Pro 280, Thr 313, Thr 315, Gly 318, Asp 319, Glu 322, Phe 402, Gly 403, Phe 404
4b	302.65 (79.85)	75.66 (63.33)	-	Met 213, Ala 215, Val 278, Ala 279, Pro 280, Thr 313, Ile 317, Gly 318, His 311, Phe 402, Phe 404
5b	**335.18 (97.7)**	**83.79 (95.45)**	**6.70 (55.52)**	Met 213, Arg 214, Ala 215, Val 278, Ala 279,Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
6b	335.18 (96.48)	83.79 (97.03)	6.70 (27.68)	Met 213, Arg 214, Ala 215, Val 278,Ala 279, Pro 280, His 311, Thr 313,Ile 317,Gly 318, Phe 402, Phe 404
7b	335.18 (96.55)	83.79 (94.76)	6.70 (38.56)	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, His 313, Ile 317, Gly318, Phe 402, Phe 404
8b	**320.13 (96.34)**	**80.03 (97.93)**	**6.40 (61.95)**	Met 213, Arg 215, Val 278, Ala 279, Pro 280, His 311, His 313, Ile 317, Gly318, Phe 402, Phe 404

9b	287.07 (96.67)	71.77 (55.42)	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
10b	287.07 (98.14)	71.77 (95.84)	5.74 (39.74)	Met 231, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311
11b	317.66 (97.5)	79.42 (53.87)	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
12b	317.66 (84.84)	79.42 (46.31)	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
13b	317.66 (97.78)	79.42 (69.94)	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
14b	286.33 (94.69)	71.58 (22.84)	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
15b	278.36 (93.9)	69.59 (2.03)	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
16b	278.36 (95.03)	69.59 (32.22)	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404

17b	339.70 (61.67)	-	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
18b	337.43 (64.96)	-	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
19b	337.43 (68.71)	-	-	Met 213, Arg 214, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
20b	337.43 (70.53)	84.36 (79.61)	-	Met 213, Ala 215, Phe 275, His 276, Val 278, Ala 279, Pro 280, Ile 317, Phe 404
21b	322.19 (70.59)	80.55 (74.09)	-	Met 213, Arg 214, Val 278, Ala 279, Pro 280, Ile 317, Gly 318
22b	293.76 (55.34)	-	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
23b	269.96 (58.88)	-	-	Met 213,Arg 214,Ala 215,Val 278,Ala 279,Pro 280,His 311,Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
24b	306.39 (64.63)	-	-	Met 213, Arg 214, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
25b	293.76 (70.53)	73.44 (70.87)	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Ile 317, Phe 404
26b	307.36 (53.08)	-	-	Met 212, Arg 214, Ala 215, Ile 317, Phe 404

27b	307.36 (44.54)	-	-	Met 212, Met 213, Arg 214, Phe 404, Ala 215, Ile 317
28b	309.19 (51.79)	-	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
29b	355.44 (60.56)	-	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
30b	338.56 (68.85)	-	-	Met 213, Arg 214, Ala 215, Val 278, Ala 279, Pro 280, His 311, Thr 313, Ile 317, Gly 318, Phe 402, Phe 404
Rifam-picin	-	-	**2.4** (**99.34**)	**-**