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Silver catalyzed decarboxylative acylation of pyridine *N*-oxides using α-oxocarboxylic acids

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General procedure for the synthesis of (substituted benzoyl)pyridine 1-oxide (3). The mixture of α-keto carboxylic acid **2** (0.92 mmol), substituted pyridine N-oxide **1** (0.46 mmol), silvercarbonate (10 mol %) and $K_2S_2O_8$ (1.38 mmol) in DCM: H_2O (3:1, 2 mL) were stirred at 50 °C for 12 h. The reaction mixture was filtered through celite pad, washed with dichloromethane. The organic layer was washed with water and brine, dried over sodium sulfate, and concentrated in vacuum. Crude product was purified by flash column chromatography using 70-80 % ethylacetate in hexane mixture as the solvent to get derivative **3**.

1. **2-(4-Chlorobenzoyl)-4-methylpyridine 1-oxide (3a).** Isolated as pale yellow solid; mp 174-175 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.43 (s, 3H), 7.23-7.25 (m, 2H), 7.45 (d, *J* = 8.4 Hz, 2H), 7.78 (d, *J* = 8.4 Hz, 2H), 8.13 (d, *J* = 6.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 20.4, 126.3, 128.0, 129.2, 130.6, 133.5, 137.8, 139.3, 140.6, 146.0, 188.3; UPLC (M+1) 248; Anal. Calcd for: $C_{13}H_{10}ClNO_2$: C, 63.04; H, 4.07; N, 5.66 %. Found C, 63.10; H, 4.03; N, 5.70 %.
2. **4-Chloro-2-(4-chlorobenzoyl)pyridine 1-oxide (3b).** Isolated as offwhite solid; mp 191-192 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.39 - 7.41 (m, 2H), 7.46 (d, *J* = 8.4 Hz, 2H), 7.76 (d, *J* = 8.4 Hz, 2H), 8.15 (d, *J* = 6.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 125.9, 127.5, 129.3, 129.5, 130.6, 132.0, 133.0, 140.7, 141.0, 186.7; UPLC (M+1) 269; Anal. Calcd for: $C_{12}H_7Cl_2NO_2$: C, 53.76; H, 2.63; N, 5.22 %. Found C, 53.70; H, 2.67; N, 5.17 %.
3. **2-(4-Chlorobenzoyl)-4-methoxypyridine 1-oxide (3c).** Isolated as offwhite solid; mp 124-125 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 3.91 (s, 3H), 6.93-6.97 (m, 2H), 7.45 (d, *J* = 8.0 Hz, 2H), 7.80 (d, *J* = 8.0 Hz, 2H), 8.14 (d, *J* = 4.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 56.4, 110.4, 113.7, 129.2, 130.7, 133.3, 140.7,* 140.9, 156.0, 187.7.; UPLC (M+1) 264; Anal. Calcd for: $C_{13}H_{10}ClNO_3$: C, 59.22; H, 3.82; N, 5.31 %. Found C, 59.26; H, 3.85; N, 5.27 %.) * Two carbons merged here.
4. **4-Methyl-2-(4-(trifluoromethyl)benzoyl)pyridine 1-oxide (3d).** Isolated as offwhite solid; mp 156-157 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.29-7.31 (m, 2H), 7.74 (d, *J* = 8.0 Hz, 2H), 7.94 (d, *J* = 8.0 Hz, 2H), 8.15 (d, *J* = 5.6 Hz, 1H); The solubility in NMR solvents is so poor and an intense ^{13}C NMR spectrum could not be recorded; UPLC (M+1) 282; Anal. Calcd for: $C_{14}H_{10}F_3NO_2$: C, 59.79; H, 3.58; N, 4.98 %. Found C, 59.71; H, 3.64; N, 5.03 %.
5. **4-Chloro-2-(4-(trifluoromethyl)benzoyl)pyridine 1-oxide (3e).** Isolated as pale yellow solid; mp 134-135 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.43-7.47 (m, 2H), 7.45 (d, *J* = 8.0 Hz, 2H), 7.93 (d, *J* = 8.0 Hz, 2H), 8.16 (d, *J* = 6.4 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 29.7, 123.3, 125.9, 126.0, 127.8, 129.3, 132.2, 135.2, 137.4, 140.8, 187.2; UPLC (M+1) 302; Anal. Calcd for: $C_{13}H_7ClF_3NO_2$: C, 51.76; H, 2.34; N, 4.64 %. Found C, 51.82; H, 2.38; N, 4.60 %.
6. **4-Chloro-2-(3-methoxybenzoyl)pyridine 1-oxide (3f).** Isolated as pale brown solid; mp 145-147 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 3.86 (s, 3H), 7.15-7.45 (m, 6H), 8.16 (t, *J* = 5.2 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 55.4, 112.7, 121.2, 122.2, 125.5, 126.4, 127.0, 129.9, 131.8, 135.8, 140.7, 160.0, 187.6; UPLC (M+1) 264; Anal. Calcd for: $C_{13}H_{10}ClNO_3$: C, 59.22; H, 3.82; N, 5.31 %. Found C, 59.17; H, 3.88; N, 5.38 %.
7. **4-Chloro-2-(2-methylbenzoyl)pyridine 1-oxide (3g).** Isolated as offwhite solid; mp 169-171 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.66 (s, 3H), 7.22-7.48 (m, 6H), 8.13 (d, *J* = 7.2 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 21.4, 125.7, 126.0, 127.1, 130.6, 131.8,

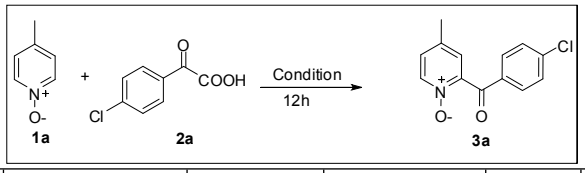
132.4, 133.0, 134.2, 140.5, 140.8, 148.6, 189.5; UPLC (M+1) 248; Anal. Calcd for: $C_{13}H_{10}ClNO_2$: C, 63.04; H, 4.07; N, 5.66 %. Found C, 63.07; H, 4.08; N, 5.71 %.

8. **4-Cyano-2-(2-methylbenzoyl)pyridine 1-oxide (3h).** Isolated as offwhite solid; mp 194-196 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.65 (s, 3H), 7.25 (t, *J* = 8.0 Hz, 1H), 7.32-7.37 (m, 2H), 7.48 (t, *J* = 7.2 Hz, 1H), 7.60 (dd, *J* = 7.2, 2.8 Hz, 1H), 7.71 (d, *J* = 2.8 Hz, 1H), 8.20 (d, *J* = 7.2 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 21.3, 107.8, 115.4, 126.0, 128.7, 129.2, 130.4, 132.5, 133.4, 133.8, 140.6, 141.1, 148.9, 188.8; UPLC (M+1) 239; Anal. Calcd for: $C_{14}H_{10}N_2O_2$: C, 70.58; H, 4.23; N, 11.76 %. Found C, 70.51; H, 4.28; N, 11.71 %.
9. **4-Chloro-2-propionylpyridine 1-oxide (3i).** Isolated as pale yellow solid; mp 84-86 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 1.21 (t, *J* = 7.2 Hz, 3H), 3.23 (q, *J* = 7.2 Hz, 2H), 7.32-7.34 (m, 1H), 7.66 (s, 1H), 8.12 (d, *J* = 6.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 7.9, 36.4, 126.8, 127.9, 131.8, 141.5, 147.0, 197.2; UPLC (M+1) 186; Anal. Calcd for: $C_8H_8ClNO_2$: C, 51.77; H, 4.34; N, 7.55 %. Found C, 51.73; H, 4.30; N, 7.60 %.
10. **2-Benzoyl-4-methylpyridine 1-oxide (3j).** Isolated as offwhite solid; mp 144-146 °C**;** ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.43 (s, 3H), 7.22-7.23 (m, 2H), 7.48 (t, *J* = 7.8 Hz, 2H), 7.62 (t, *J* = 7.2 Hz, 1H), 7.85 (d, *J* = 7.8 Hz, 2H), 8.15 (d, *J* = 7.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 20.4, 126.0, 127.7, 128.8, 129.3, 134.2, 135.0, 137.8, 139.3, 146.4, 189.4; UPLC (M+1) 214; Anal. Calcd for: $C_{13}H_{11}NO_2$: C, 73.23; H, 5.20; N, 6.57 %. Found C, 73.29; H, 5.24; N, 6.62 %.
11. **2-Benzoyl-4-chloropyridine 1-oxide (3k).** Isolated as offwhite solid; mp 120-122 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.37-7.39 (m, 2H), 7.49 (t, *J* = 7.6 Hz, 1H), 7.63 (t, *J* = 7.6 Hz, 2H), 7.84 (d, *J* = 7.6 Hz, 2H), 8.16 (d, *J* = 6.4 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 125.7, 127.1, 128.9,* 129.2, 131.8, 134.4, 134.5, 140.7, 187.8; UPLC (M+1) 234; Anal. Calcd for: $C_{12}H_8ClNO_2$: C, 61.69; H, 3.45; N, 5.99 %. Found C, 61.65; H, 3.48; N, 6.05 %. * Two carbons merged here.
12. **2-Benzoyl-4-methoxypyridine 1-oxide (3l).** Isolated as offwhite solid; mp 94-95 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.99 (s, 3H), 6.91 – 6.96 (m, 2H), 7.48 (t, *J* = 7.6 Hz, 2H), 7.61 (t, *J* = 7.2 Hz, 1H), 7.86 (d, *J* = 7.6 Hz, 2H), 8.14 (d, *J* = 7.2 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 56.3, 110.1, 113.4, 128.8, 129.3, 134.2, 134.8, 138.2, 140.7, 157.7, 188.7; UPLC (M+1) 230; Anal. Calcd for: $C_{13}H_{11}NO_3$: C, 68.11; H, 4.84; N, 6.11 %. Found C, 68.15; H, 4.79; N, 6.17 %.
13. **2-Benzoyl-4-cyanopyridine 1-oxide (3m).** Isolated as pale yellow solid; mp 170-171 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.51 (t, *J* = 7.2 Hz, 2H), 7.63-7.68 (m, 3H), 7.80 (d, *J* = 7.2 Hz, 2H), 8.27 (d, *J* = 6.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 107.9, 115.3, 128.6, 129.1, 129.2, 129.3, 134.3, 134.8, 141.0, 148.1, 187.3; UPLC (M+1) 225; Anal. Calcd for: $C_{13}H_8N_2O_2$: C, 69.64; H, 3.60; N, 12.49 %. Found C, 69.73; H, 3.67; N, 12.41 %.
14. **2-Benzoylquinoline 1-oxide (3n).** Isolated as offwhite solid; mp 155-157 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.42-7.50 (m, 3H), 7.62 (t, *J* = 7.2 Hz, 1H), 7.74 (t, *J* = 7.2 Hz, 1H), 7.81-7.84 (m, 4H), 7.96 (d, *J* = 7.6 Hz, 1H), 8.73 (d, *J* = 8.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 119.8, 120.4, 125.8, 128.3, 128.9, 129.2, 129.6, 130.7, 131.0, 134.1, 135.2, 141.6, 142.8, 190.3; UPLC (M+1) 250; Anal. Calcd for: $C_{16}H_{11}NO_2$: C, 77.10; H, 4.45; N, 5.62 %. Found C, 77.06; H, 4.41; N, 5.68 %.
15. **2-(4-Chlorobenzoyl)quinoline 1-oxide (3o).** Isolated as offwhite solid; mp 137-138 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.44-7.47 (m, 3H), 7.74-7.90 (m, 5H), 7.97 (d, *J* = 7.6 Hz,

1H), 8.72 (d, *J* = 8.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 119.8, 120.5, 126.0, 128.3, 129.3, 129.8, 130.5, 130.8, 131.2, 133.7, 140.5, 141.6, 142.3, 189.1; UPLC (M+1) 284; Anal. Calcd for: $C_{16}H_{10}ClNO_2$: C, 67.74; H, 3.55; N, 4.94 %. Found C, 67.69; H, 3.59; N, 4.90 %.

16. **2-Benzoyl-6-methylpyridine 1-oxide (3p).** Isolated as offwhite solid; mp 111-113 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.56 (s, 3H), 7.28-7.35 (m, 2H), 7.42 (dd, *J* = 7.0, 2.4 Hz, 1H), 7.48 (t, *J* = 7.2 Hz, 2H), 7.60 (t, *J* = 7.8 Hz, 1H), 7.82 (d, *J* = 7.6 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 17.4, 122.9, 125.3, 127.3, 128.8, 129.1, 133.9, 135.4, 147.0, 149.9, 189.7; UPLC (M+1) 214; Anal. Calcd for: $C_{13}H_{11}NO_2$: C, 73.23; H, 5.20; N, 6.57 %. Found C, 73.26; H, 5.23; N, 6.60 %.
17. **4-Benzoyl-2-methylpyridine 1-oxide (3p').** Isolated as offwhite solid; mp 120-121 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.56 (s, 3H), 7.54 (t, *J* = 7.6 Hz, 2H), 7.59 (d, *J* = 4.8 Hz, 1H), 7.66 (t, *J* = 7.6 Hz, 1H), 7.73 (s, 1H), 8.34 (d, *J* = 6.4 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 17.8, 124.3, 127.0, 128.7, 129.6, 132.6, 133.2, 136.3, 139.3, 149.3, 192.6; UPLC (M+1) 214; Anal. Calcd for: $C_{13}H_{11}NO_2$: C, 73.23; H, 5.20; N, 6.57 %. Found C, 73.19; H, 5.18; N, 6.51 %.
18. **2-Benzoyl-5-carboxypyridine 1-oxide (3r).** Isolated as pale yellow solid; mp 148-149 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.54 (t, *J* = 7.2 Hz, 2H), 7.68 (t, *J* = 7.2 Hz, 1H), 7.94 (d, *J* = 7.2 Hz, 2H), 8.06 (d, *J* = 8.0 Hz, 1H), 8.50 (dd, *J* = 8.0, 1.6 Hz, 1H), 9.15 (d, *J* = 1.6 Hz, 1H), 13.8 (br s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 124.5, 128.8, 129.0, 131.0, 133.8, 136.0, 138.9, 149.6, 157.8, 166.1, 193.4; UPLC (M+1) 244; Anal. Calcd for: $C_{13}H_9NO_4$: C, 64.20; H, 3.73; N, 5.76 %. Found C, 64.27; H, 3.67; N, 5.72 %.
19. **2,4-Dibenzoyl-5-carboxypyridine 1-oxide (3r').** Isolated as pale brown solid; mp 189-190 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.53-7.60 (m, 4H), 7.68-7.80 (m, 4H), 8.01 (s, 1H), 8.02 (d, *J* = 7.6 Hz, 2H), 9.25 (s, 1H), 13.9 (br s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 122.0, 126.8, 128.8, 129.3, 129.6, 131.2, 134.1, 134.4, 135.7, 135.9, 150.2, 150.4, 157.9, 165.6, 192.9, 194.5; UPLC (M+1) 348; Anal. Calcd for: $C_{20}H_{13}NO_5$: C, 69.16; H, 3.77; N, 4.03 %. Found C, 69.20; H, 3.81; N, 3.98 %.
20. **4-Benzoyl-2,6-dimethylpyridine 1-oxide (3s).** Isolated as offwhite solid; mp 215-216 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ 2.41 (s, 6H), 7.55-7.60 (m, 2H), 7.67-7.70 (m, 3H), 7.75-7.78 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 18.4, 124.4, 128.6, 129.6, 131.7, 133.0, 136.5, 149.2, 193.2; UPLC (M+1) 228; Anal. Calcd for: $C_{14}H_{13}NO_2$: C, 73.99; H, 5.77; N, 6.16 %. Found C, 74.03; H, 5.72; N, 6.22 %.
21. **2-Benzoylpyridine 1-oxide (3t).** Isolated as pale brown viscous liquid; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.42-7.49 (m, 5H), 7.61 (t, *J* = 7.6 Hz, 1H), 7.83 (d, *J* = 7.2 Hz, 2H), 8.27 (bd, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 125.6, 125.9, 126.9, 128.8, 128.9, 129.3, 134.2, 135.0, 140.1, 189.3; UPLC (M+1) 200; Anal. Calcd for: $C_{12}H_9NO_2$: C, 72.35; H, 4.55; N, 7.03 %. Found C, 72.31; H, 4.62; N, 7.00 %.
22. **6,6'-Dibenzoyl-3,3'-bipyridine 1,1'-dioxide (3t').** Isolated as offwhite solid; mp 161-162 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.58 (t, *J* = 7.6 Hz, 4H), 7.73 (t, *J* = 7.6 Hz, 2H), 7.80 (d, *J* = 7.6 Hz, 4H), 7.88 (d, *J* = 8.0 Hz, 2H), 8.02 (d, *J* = 8.0 Hz, 2H), 8.92 (s, 2H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 125.2, 126.0, 129.4, 129.5, 134.8, 135.3, 138.5,* 146.3, 189.8; UPLC (M+1) 397; Anal. Calcd for: $C_{24}H_{16}N_2O_4$: C, 72.72; H, 4.07; N, 7.07 %. Found C, 72.63; H, 4.02; N, 7.02 %. * Two carbons merged here.

Table Screening impact on reagents and solvents



Entry	Catalyst (10 mol %)	Oxidant (2eq)	Solvent	Temp °C	Yield of **3a** (%)[a]
1	$Cu(OAc)_2$	TBHP	DMF	100	28
2	$Cu(OAc)_2$	Ag_2O	DMF	100	45
3	$CuSO_4.5H_2O$	TBHP	DMF	100	28
4	$CuSO_4.5H_2O$	$K_2S_2O_8$	DMF	100	32
5	$FeSO_4.7H_2O$	$K_2S_2O_8$	DCM/H_2O	50	40
6	$Pd(OAc)_2$	$K_2S_2O_8$	DMSO	80	38
7	$Mn(OAc)_3$	-	HOAc	80	25
8	-	CAN	CH_3CN	80	-
9	AgOAc	$K_2S_2O_8$	DCM/H_2O	50	22
10	$AgNO_3$	TBHP	DCM/H_2O	50	41
11	$AgNO_3$	$NH_4S_2O_8$	DCM/H_2O	50	53
12	$AgNO_3$	$K_2S_2O_8$	DCM/H_2O	50	59
13[b]	Ag_2CO_3	$K_2S_2O_8$	DCM/H_2O	50	65
14[c]	**Ag_2CO_3**	**$K_2S_2O_8$**	**DCM/H_2O**	**50**	**81**
15	Ag_2CO_3	O_2	DCM/H_2O	50	16
16	Ag_2CO_3	Oxone	DCM/H_2O	50	11

[a]Isolated yield; [b]Reaction condition: heteroarene *N*-oxide (0.46 mmol), acid (0.69 mmol), catalyst (10 mol%), oxidant (0.92 mmol) in DCM:water mixture (2 mL, 3:1), 50 °C, 12 h; [c]Reaction condition: 0.92 mmol of acid and 1.38 mmol of oxidant used.

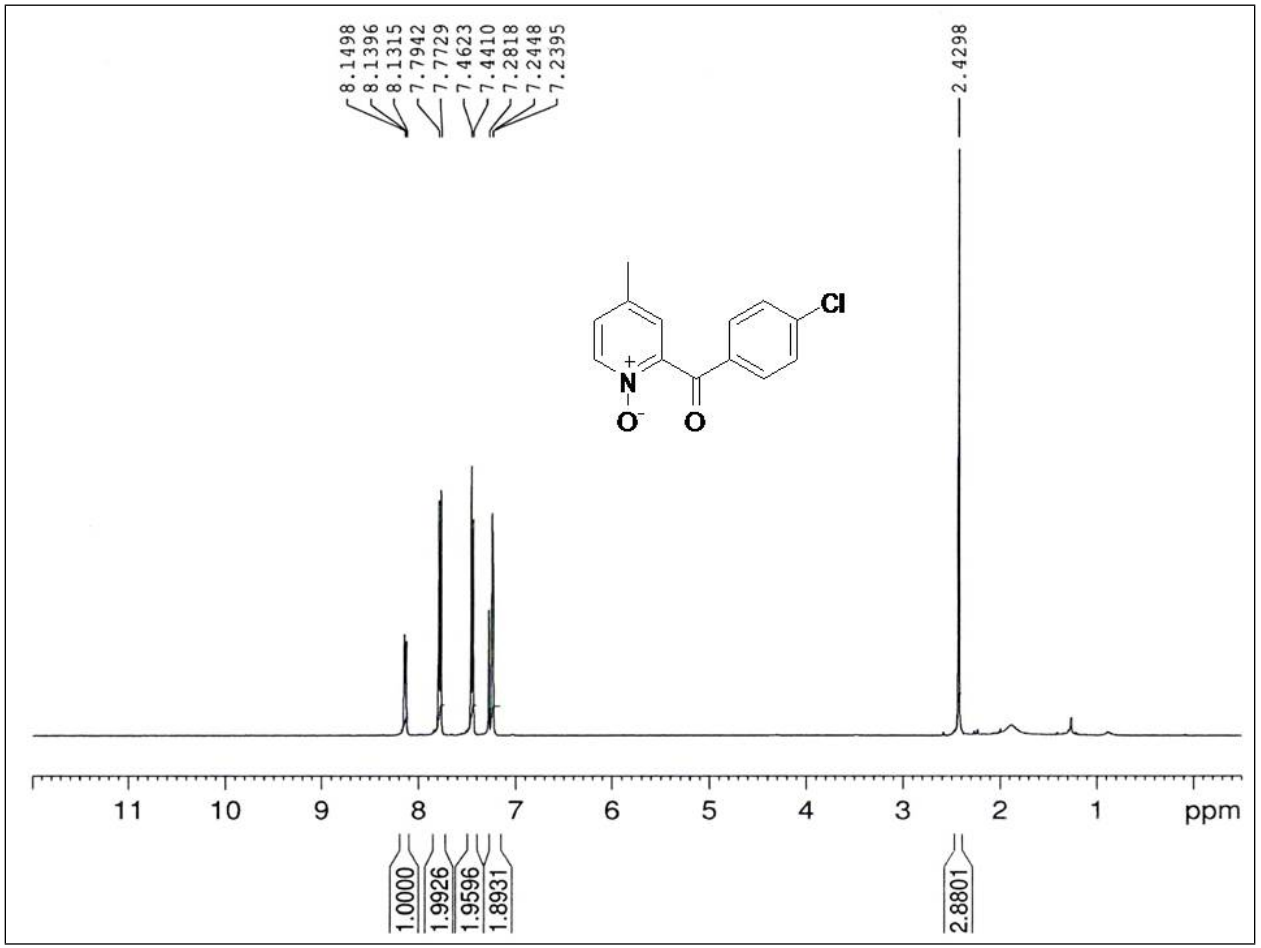


Fig 1. ^{1}H-NMR spectrum of **3a**

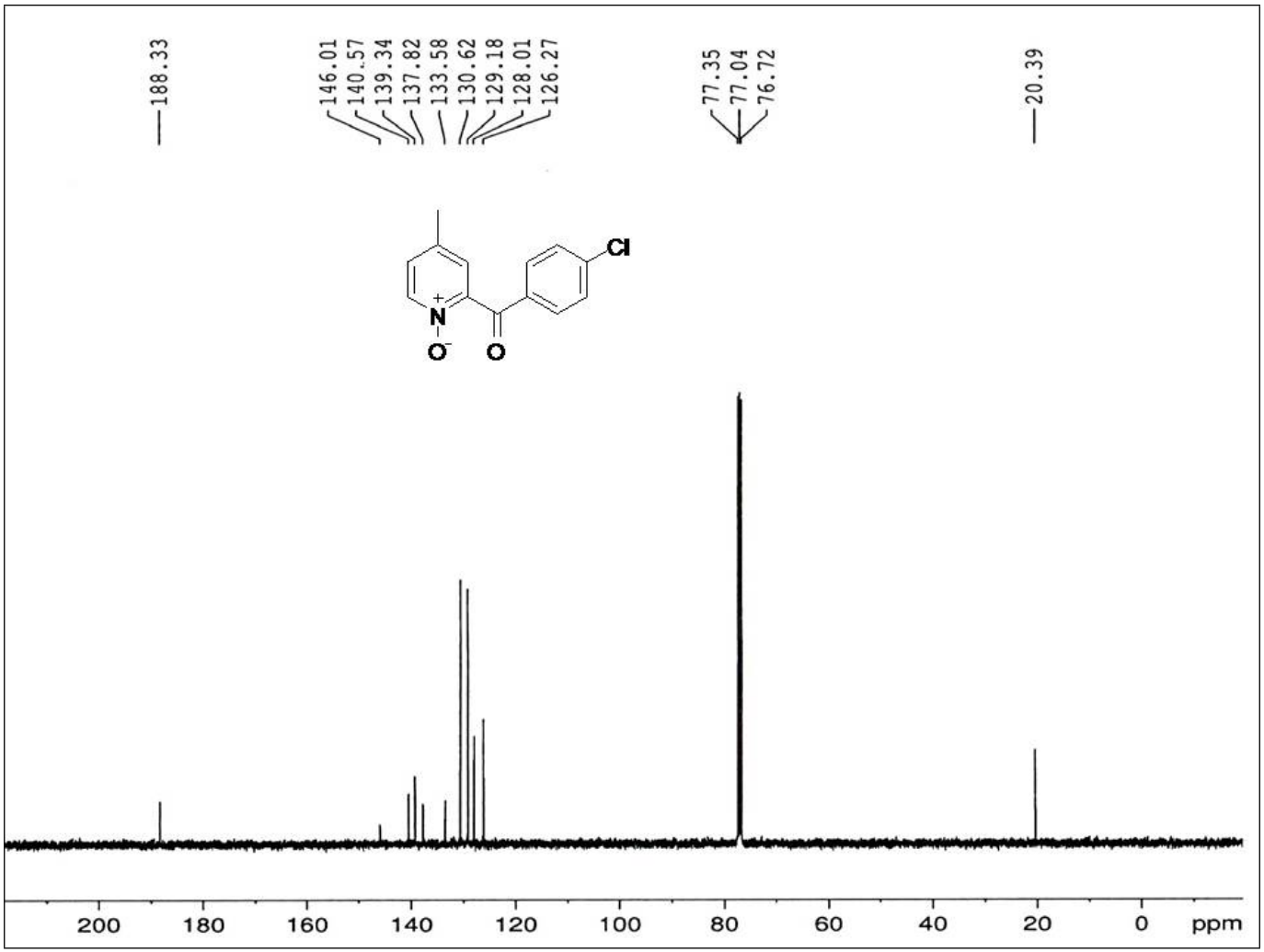


Fig 2. ^{13}C-NMR spectrum of **3a**

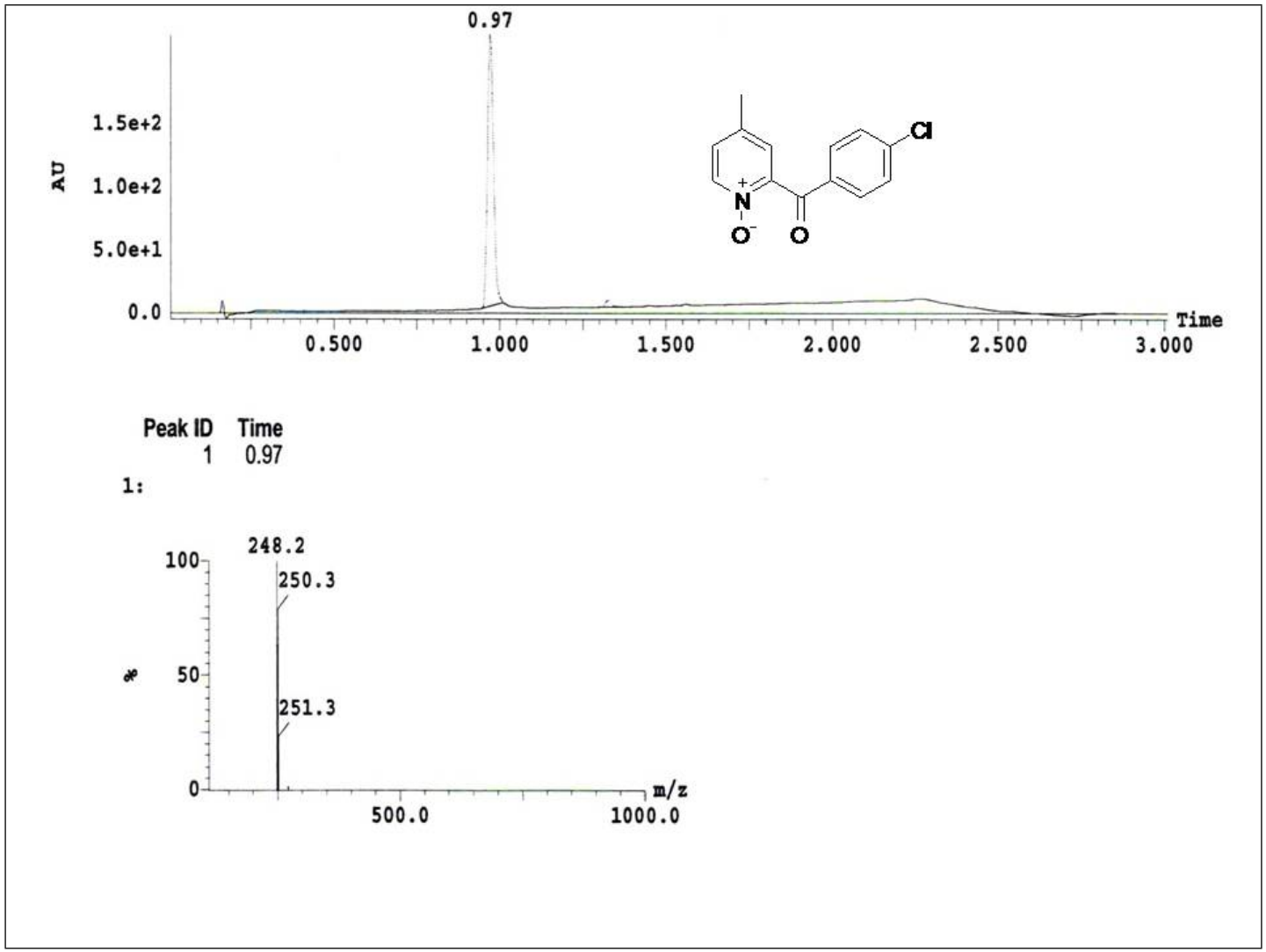


Fig 3. Mass spectrum of **3a**

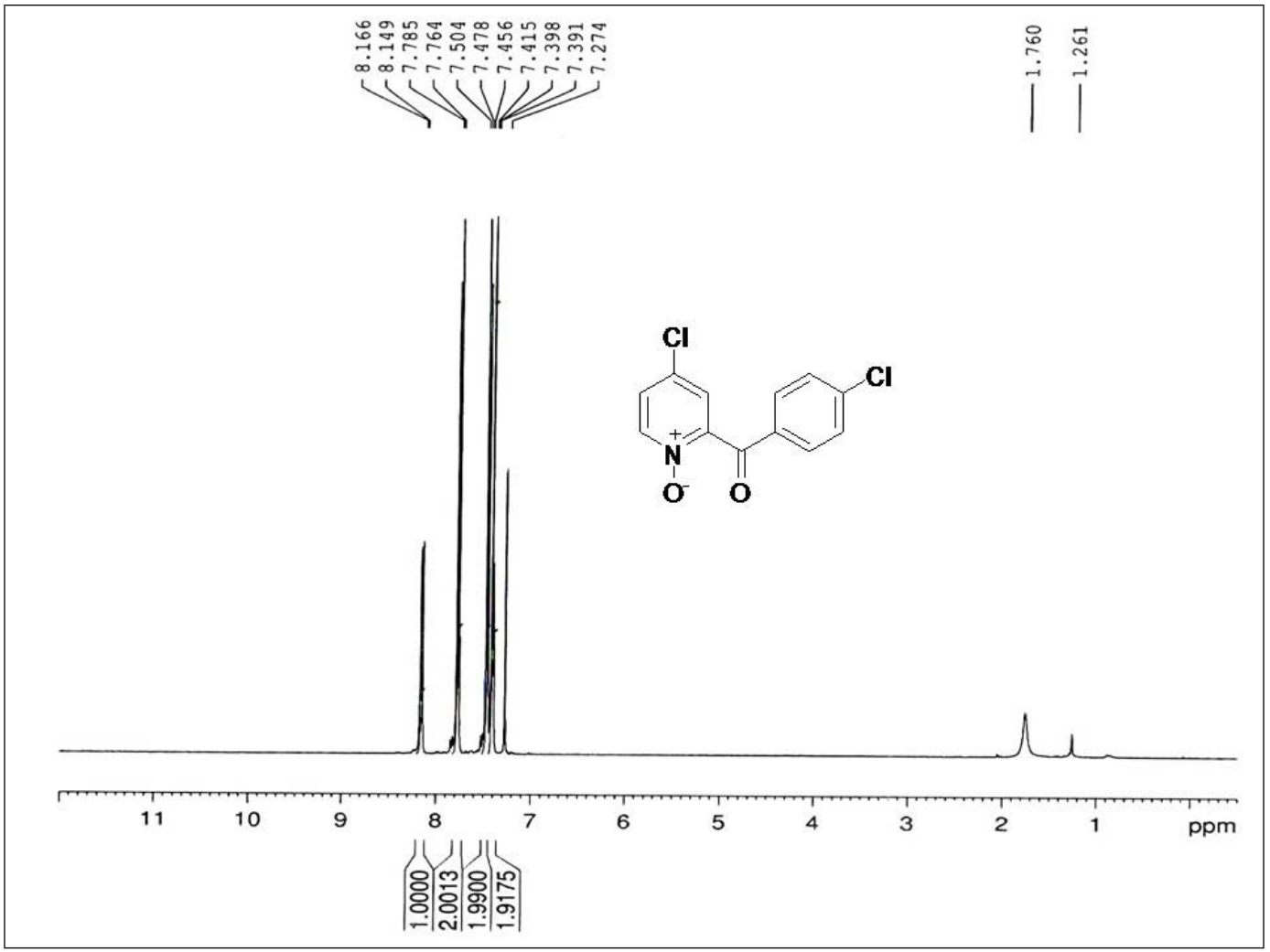


Fig 4. ^{1}H-NMR spectrum of **3b**

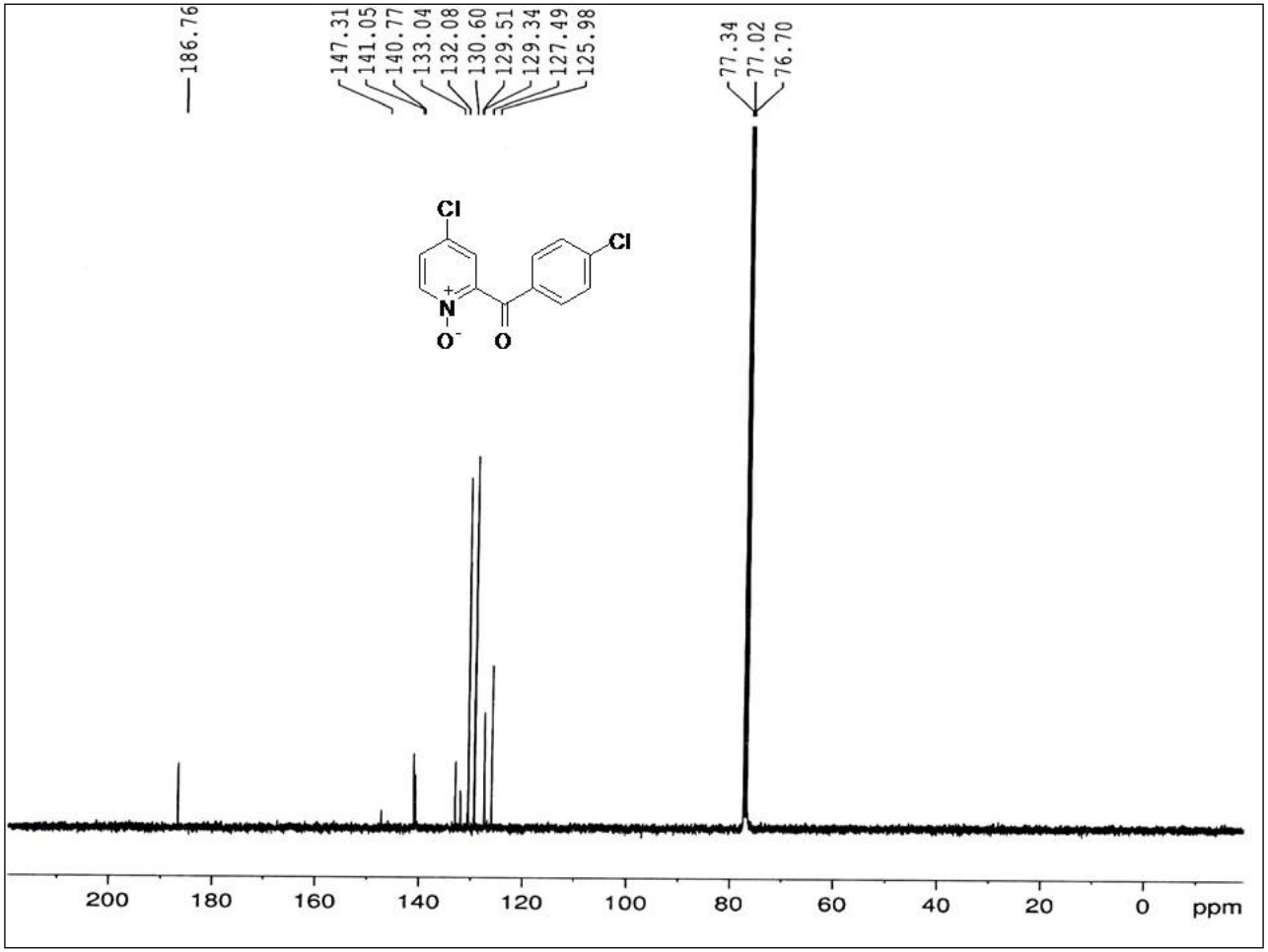


Fig 5. ^{13}C-NMR spectrum of **3b**

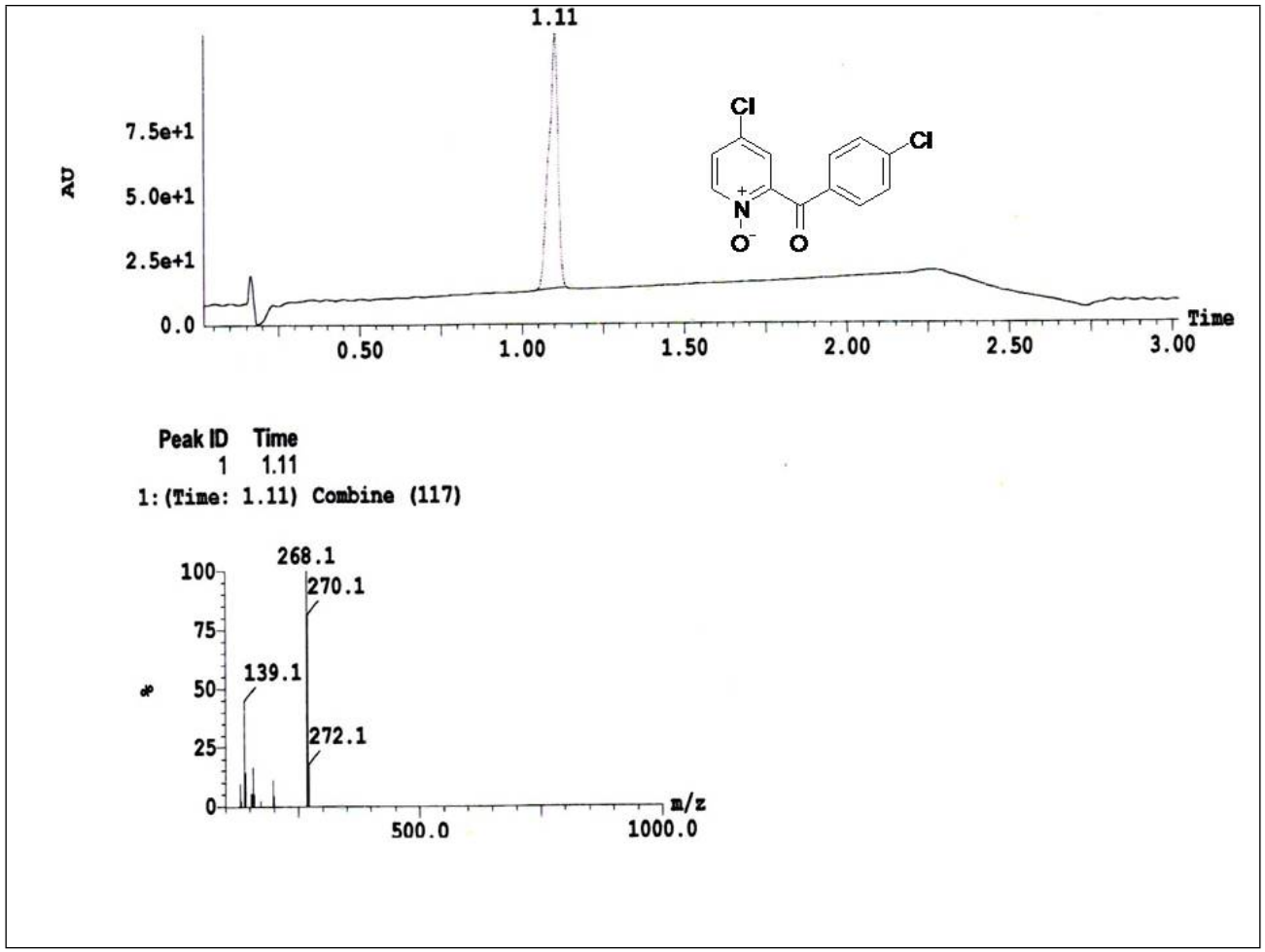


Fig 6. Mass spectrum of **3b**

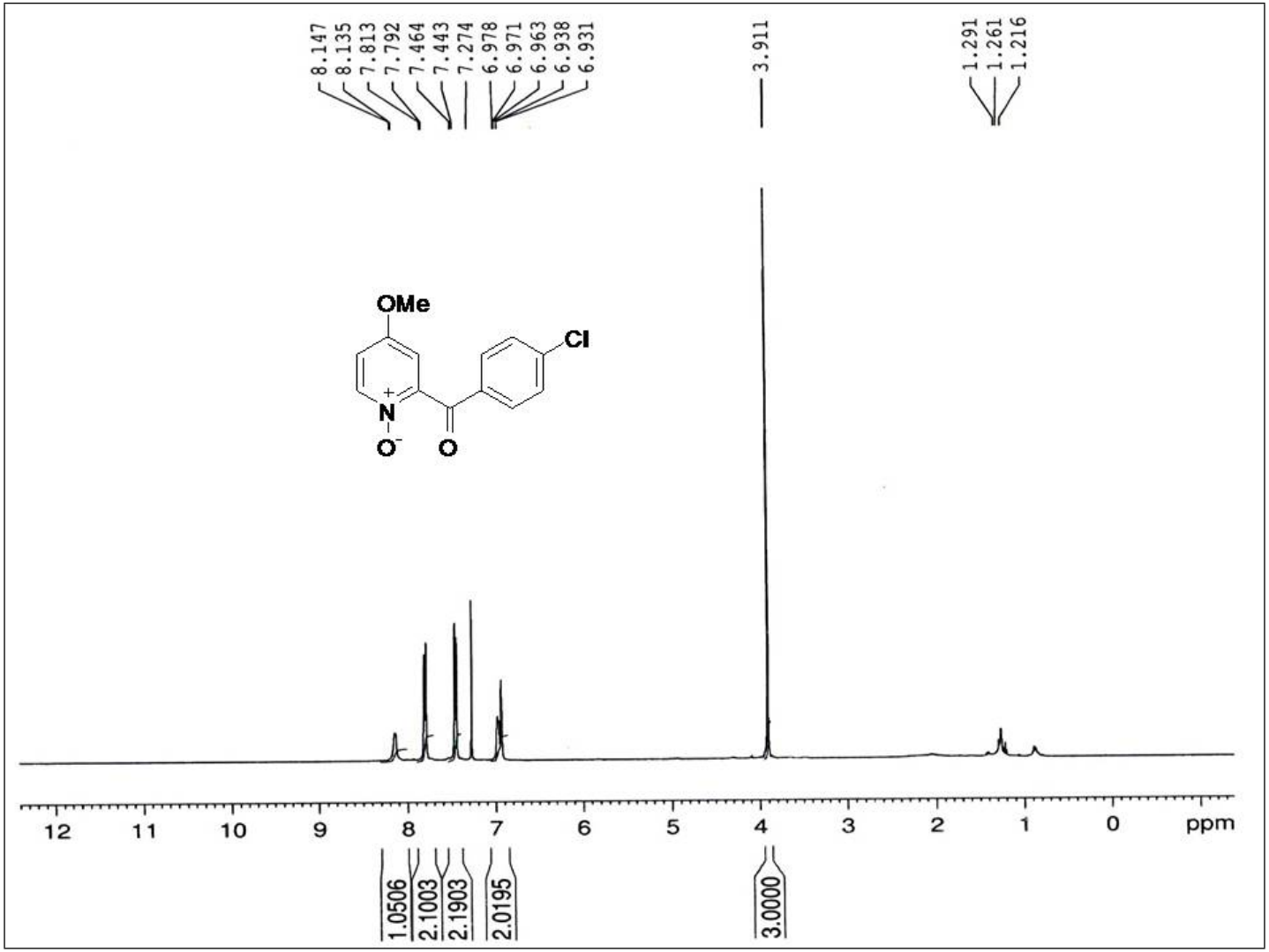


Fig 7. [1]H-NMR spectrum of **3c**

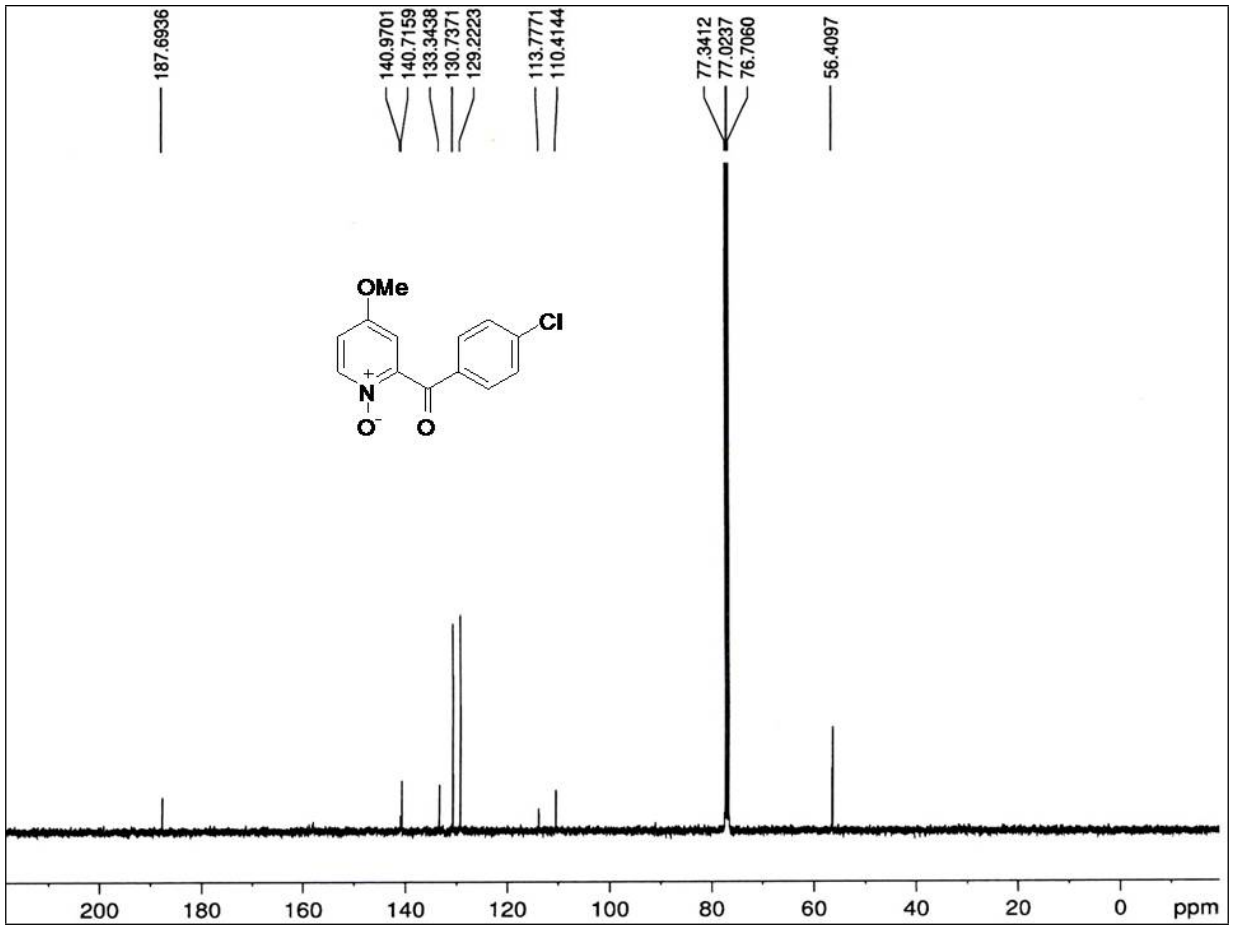


Fig 8. ^{13}C-NMR spectrum of **3c**

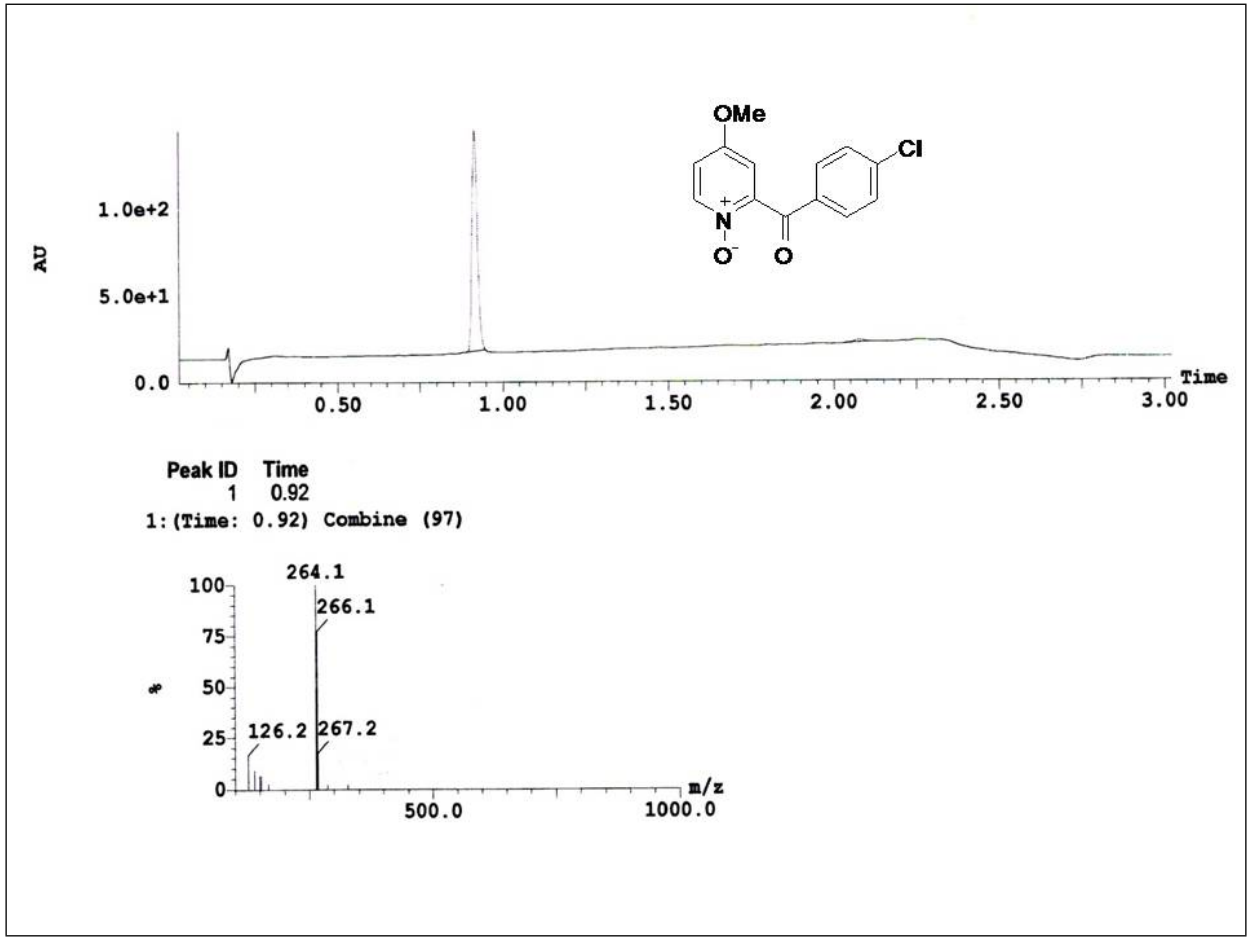


Fig 9. Mass spectrum of **3c**

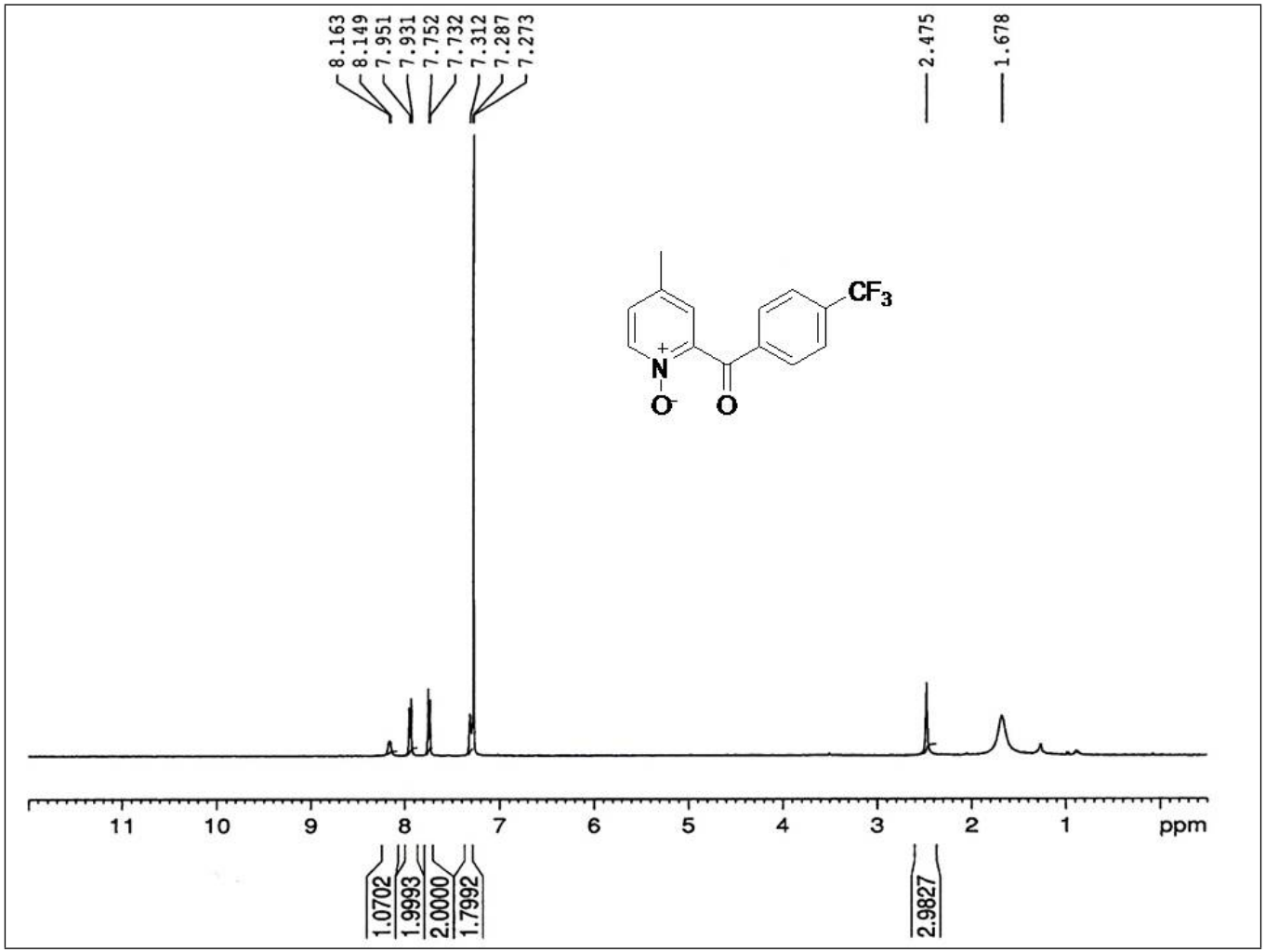


Fig 10. ^{1}H-NMR spectrum of **3d**

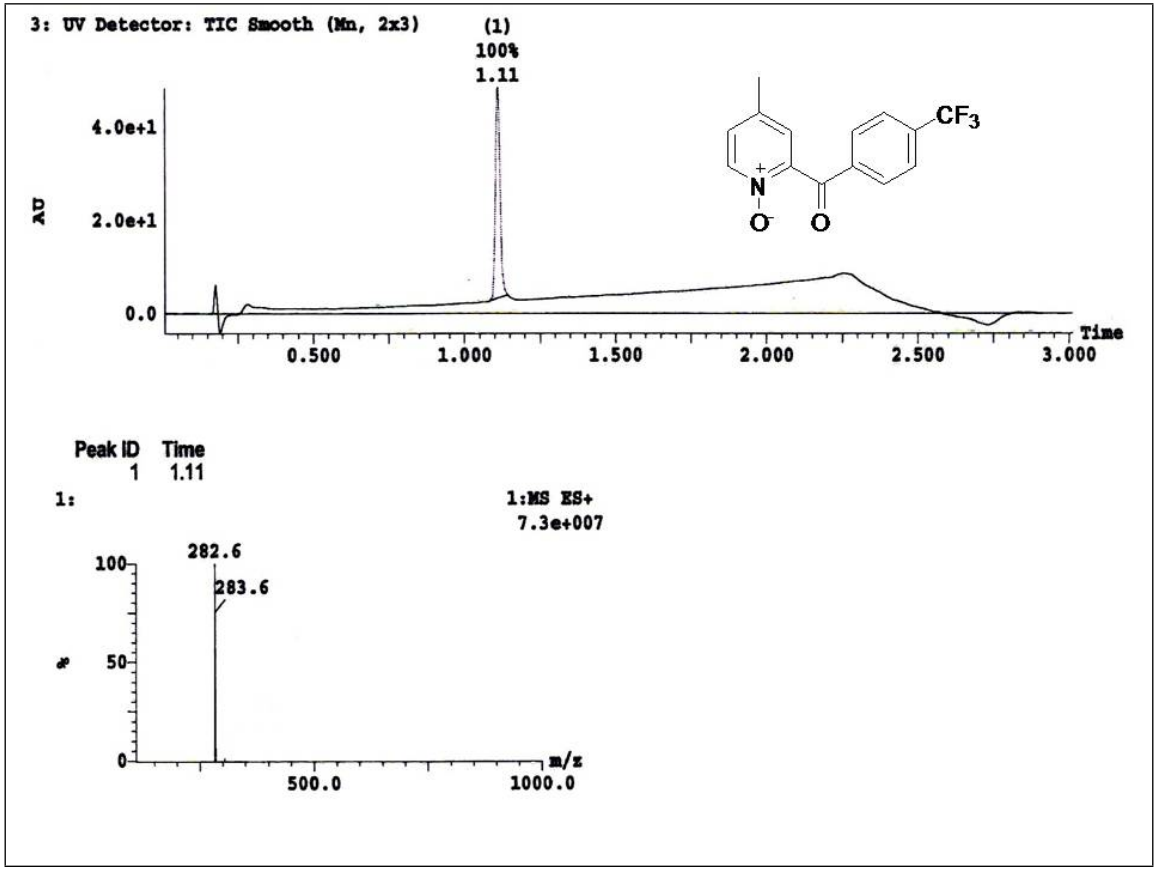


Fig 11. Mass spectrum of **3d**

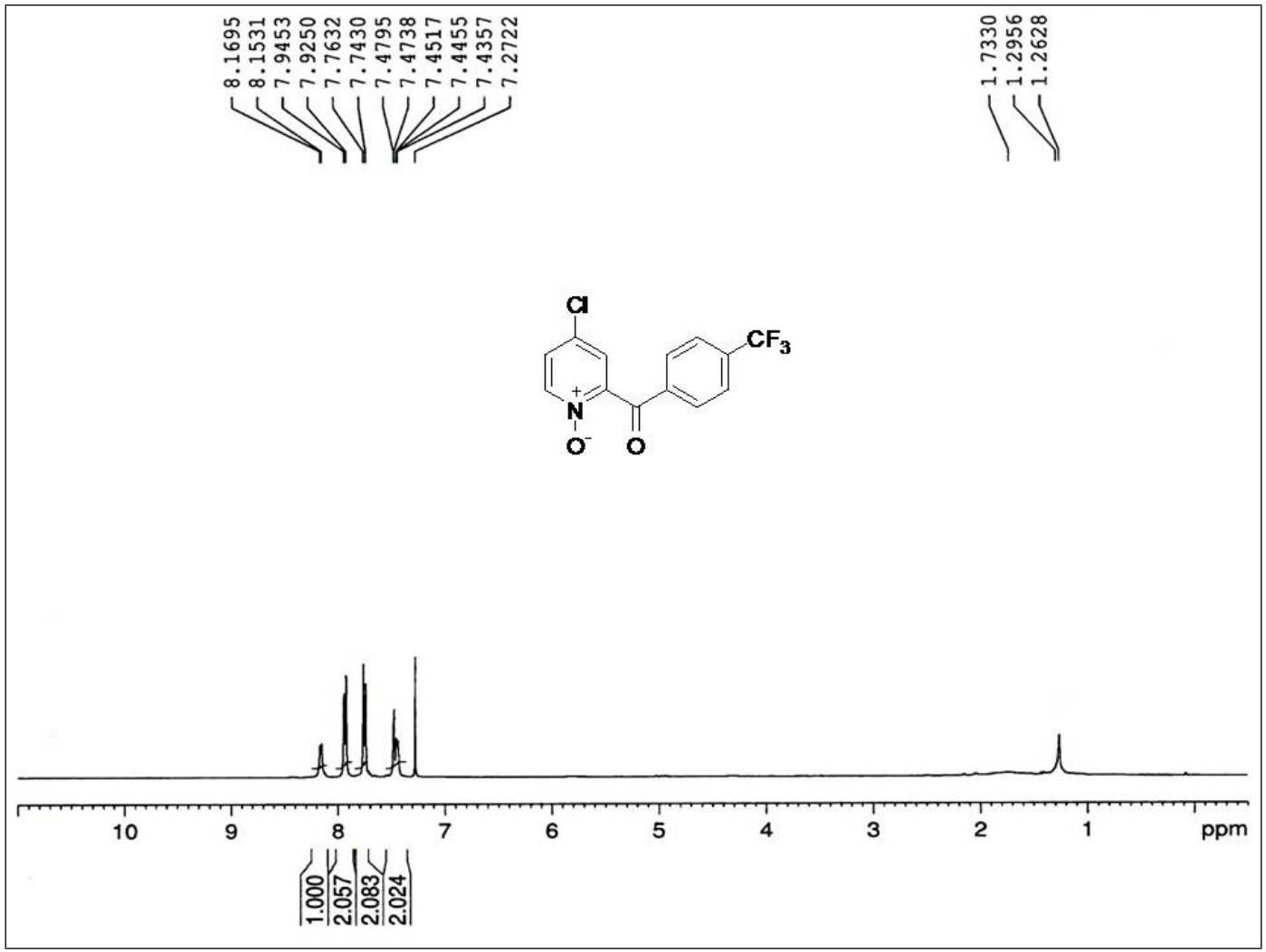


Fig 12. ^{1}H-NMR spectrum of **3e**

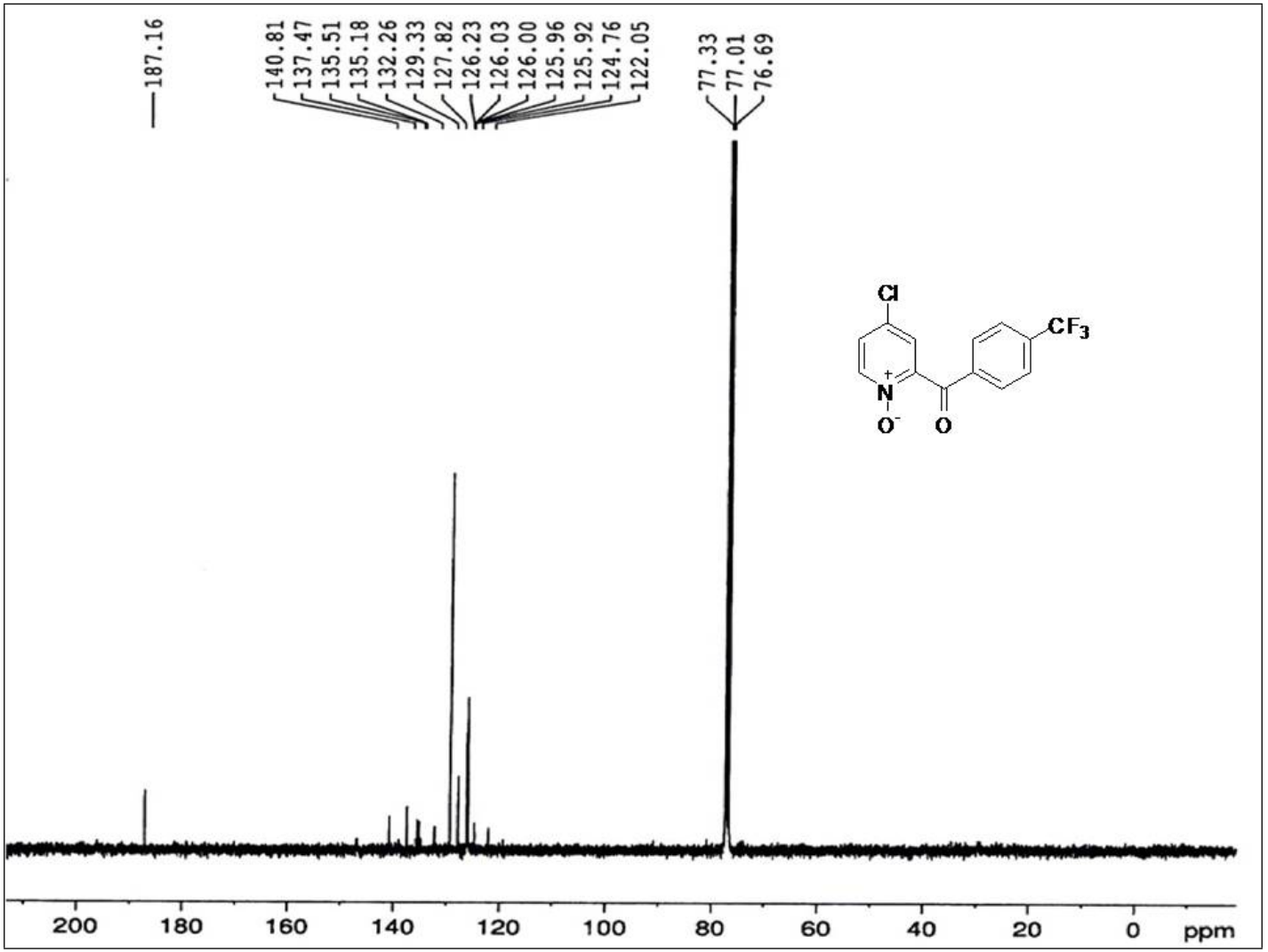


Fig 13. ^{13}C-NMR spectrum of **3e**

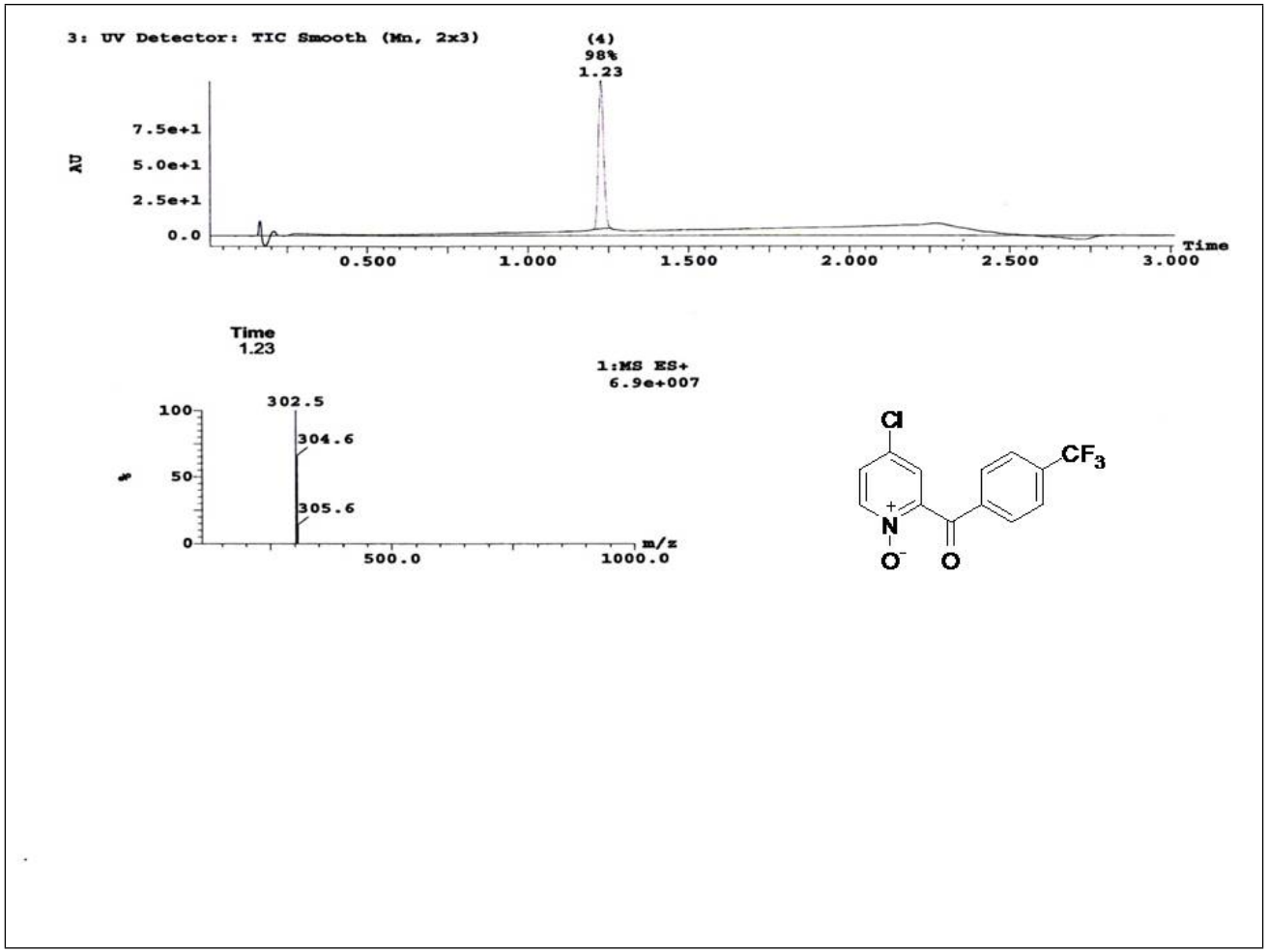


Fig 14. Mass spectrum of **3e**

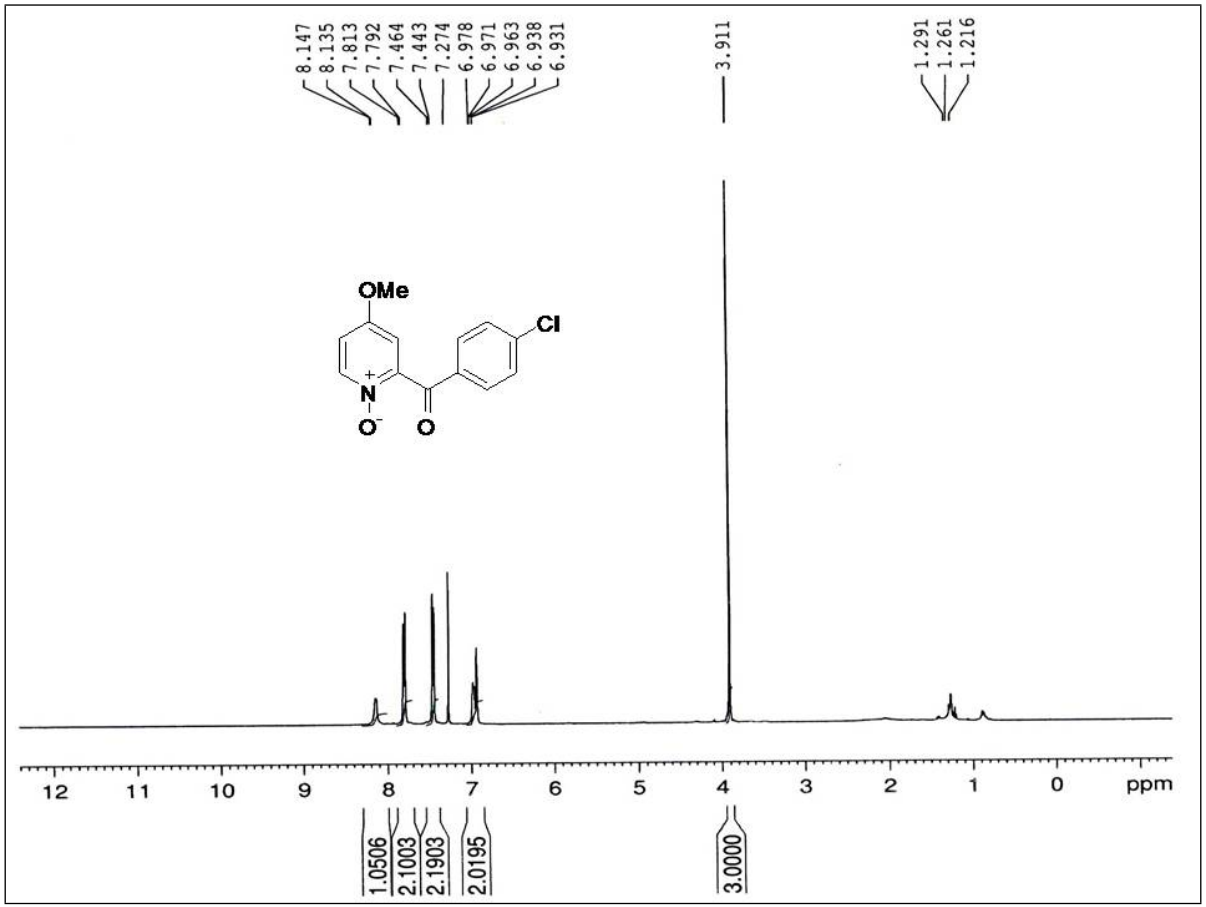


Fig 15. ^{1}H-NMR spectrum of **3f**

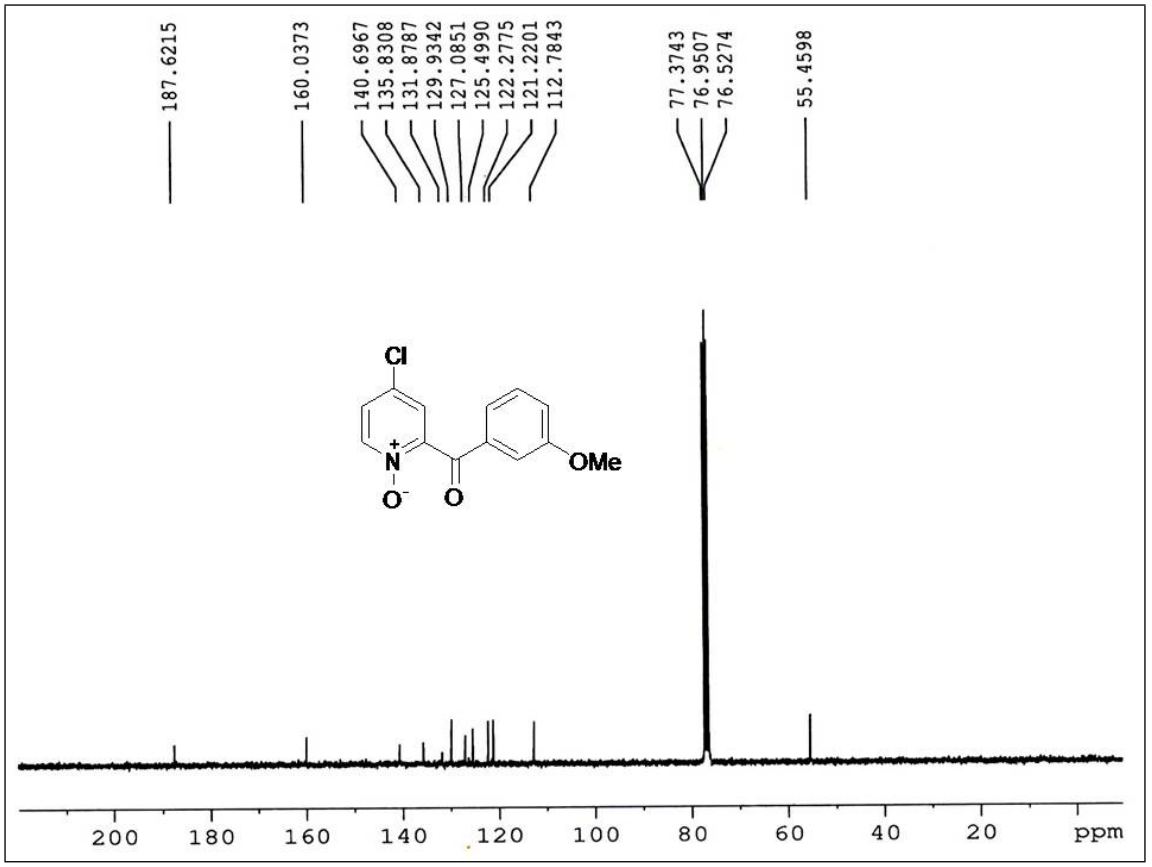


Fig 16. ^{13}C-NMR spectrum of **3f**

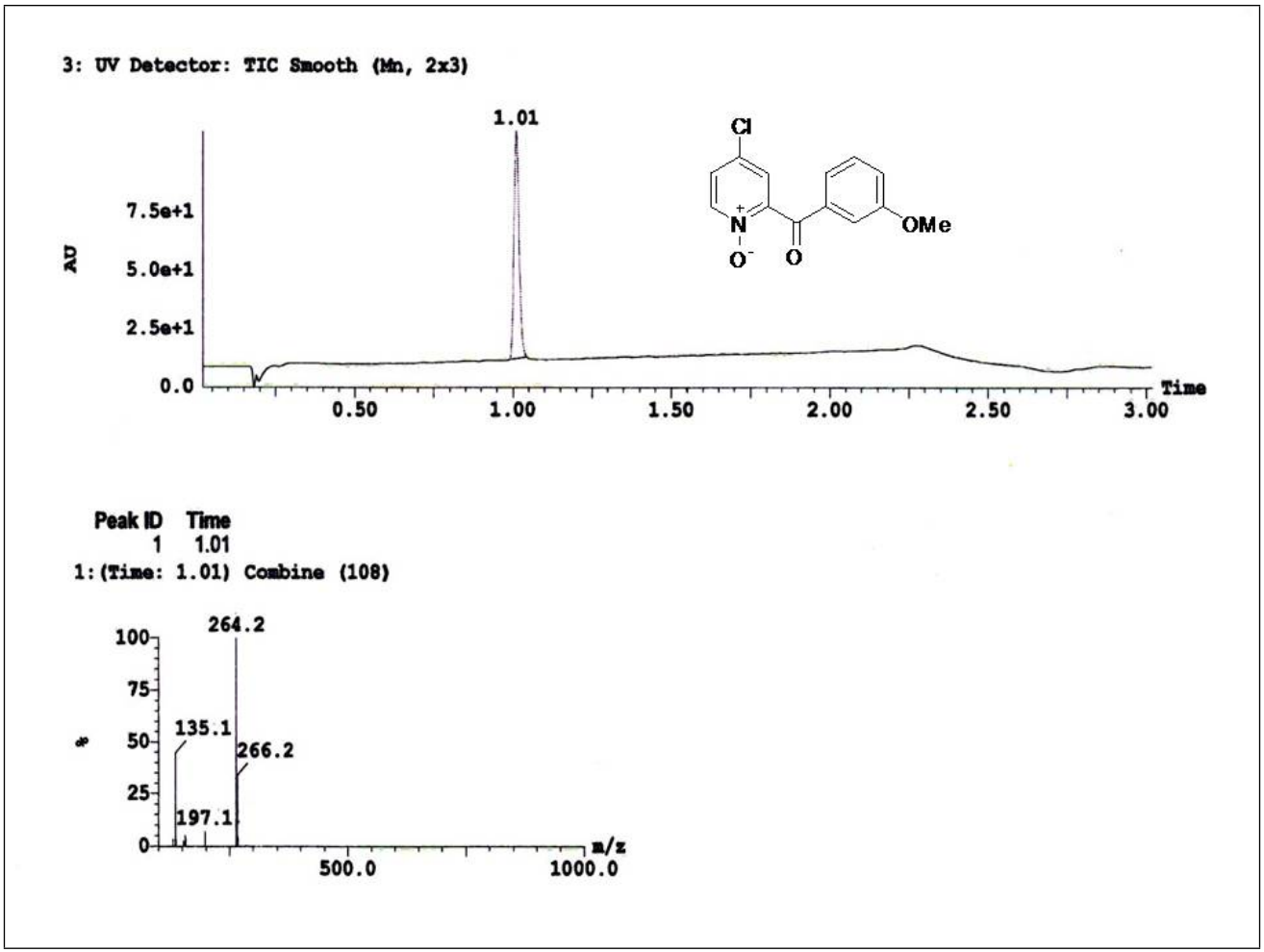


Fig 17. Mass spectrum of **3f**

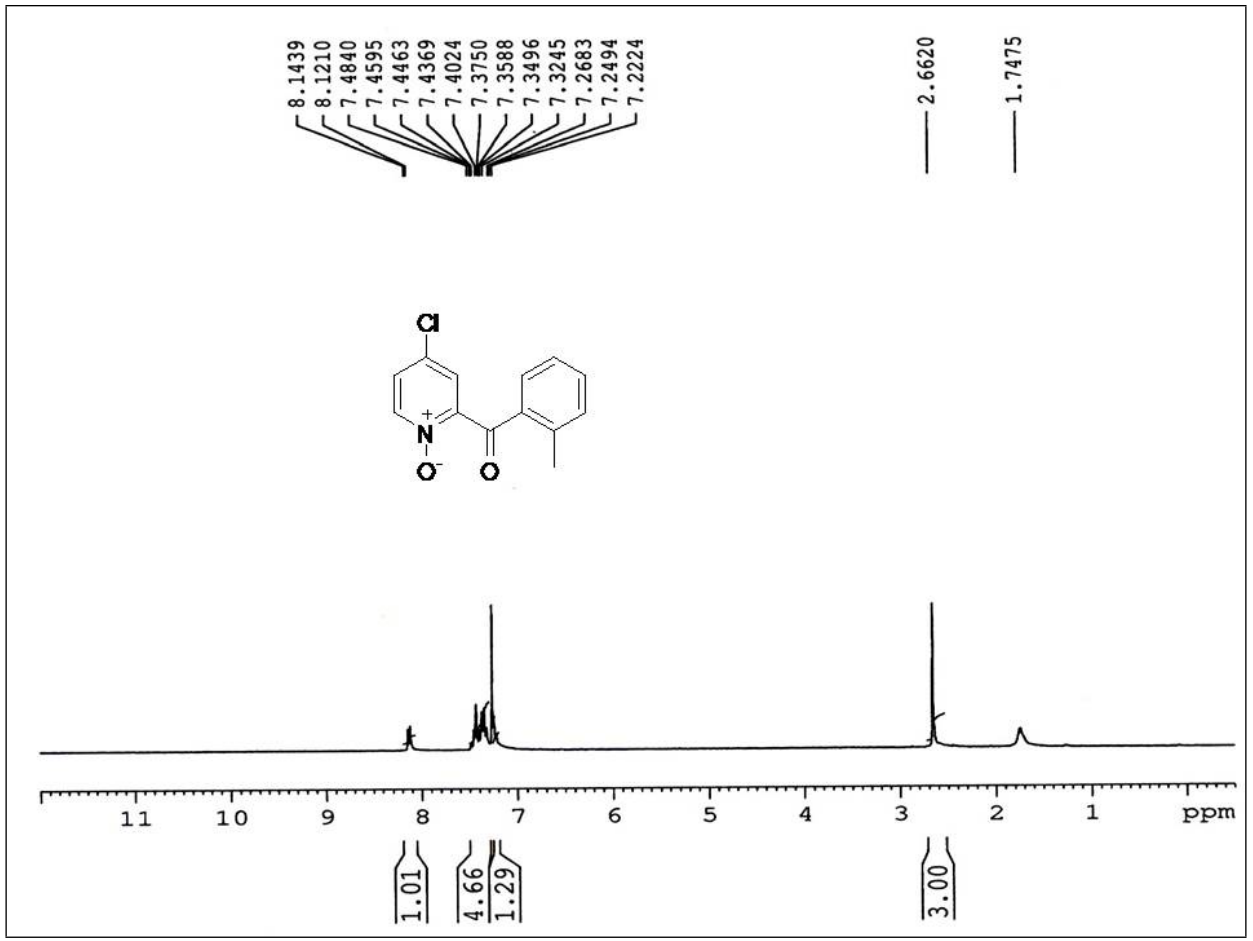


Fig 18. ^{1}H-NMR spectrum of **3g**

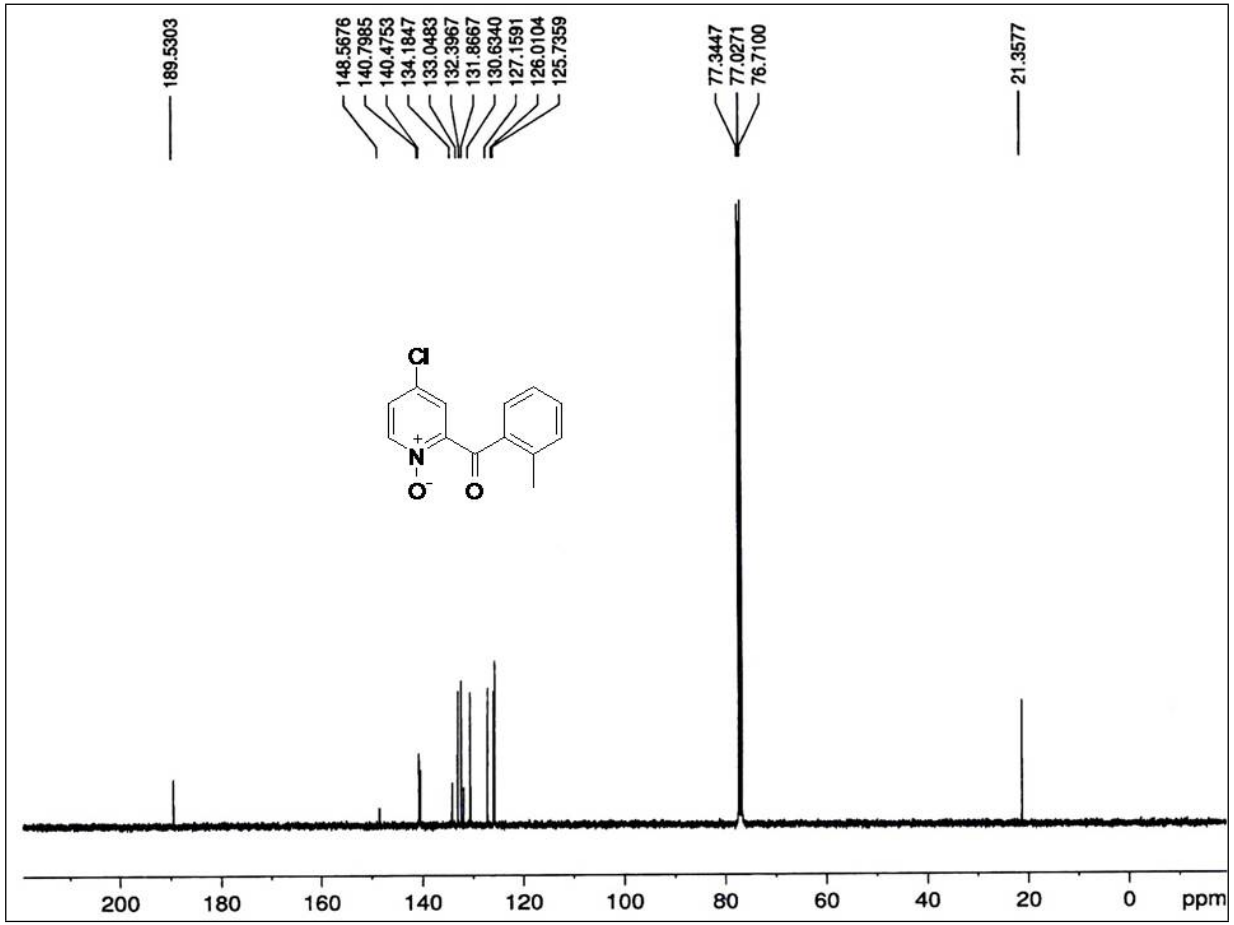


Fig 19. ^{13}C-NMR spectrum of **3g**

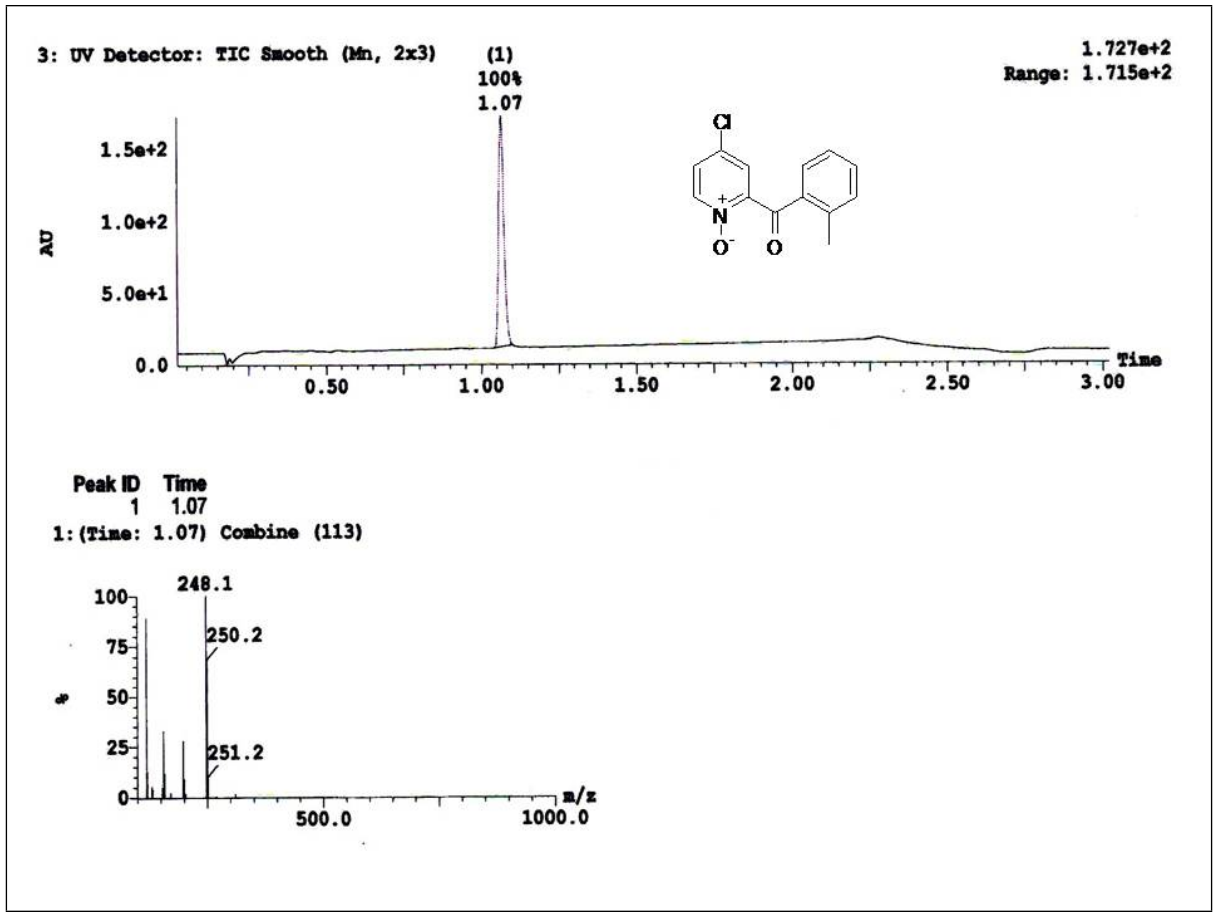


Fig 20. Mass spectrum of **3g**

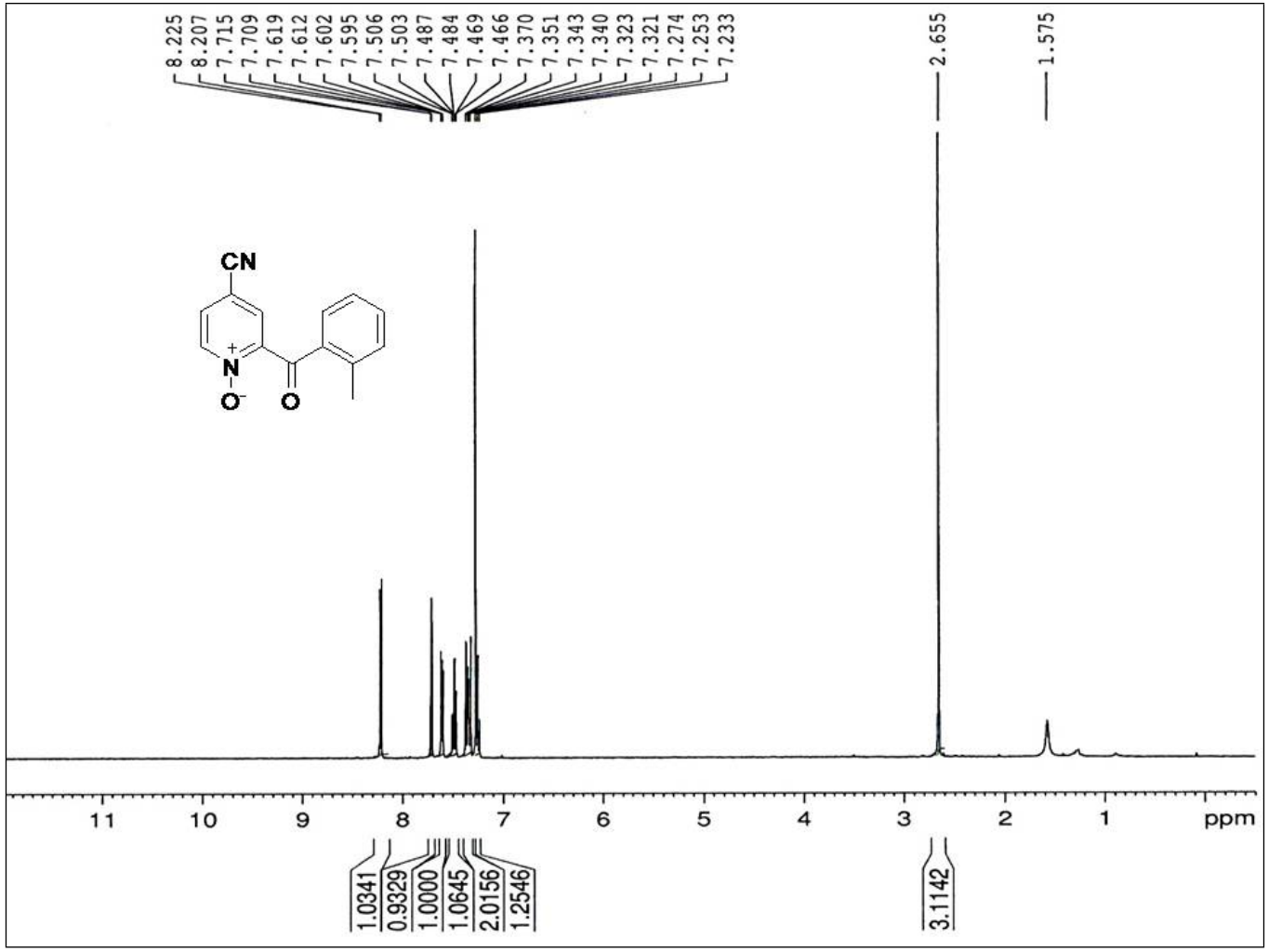


Fig 21. ^{1}H-NMR spectrum of **3h**

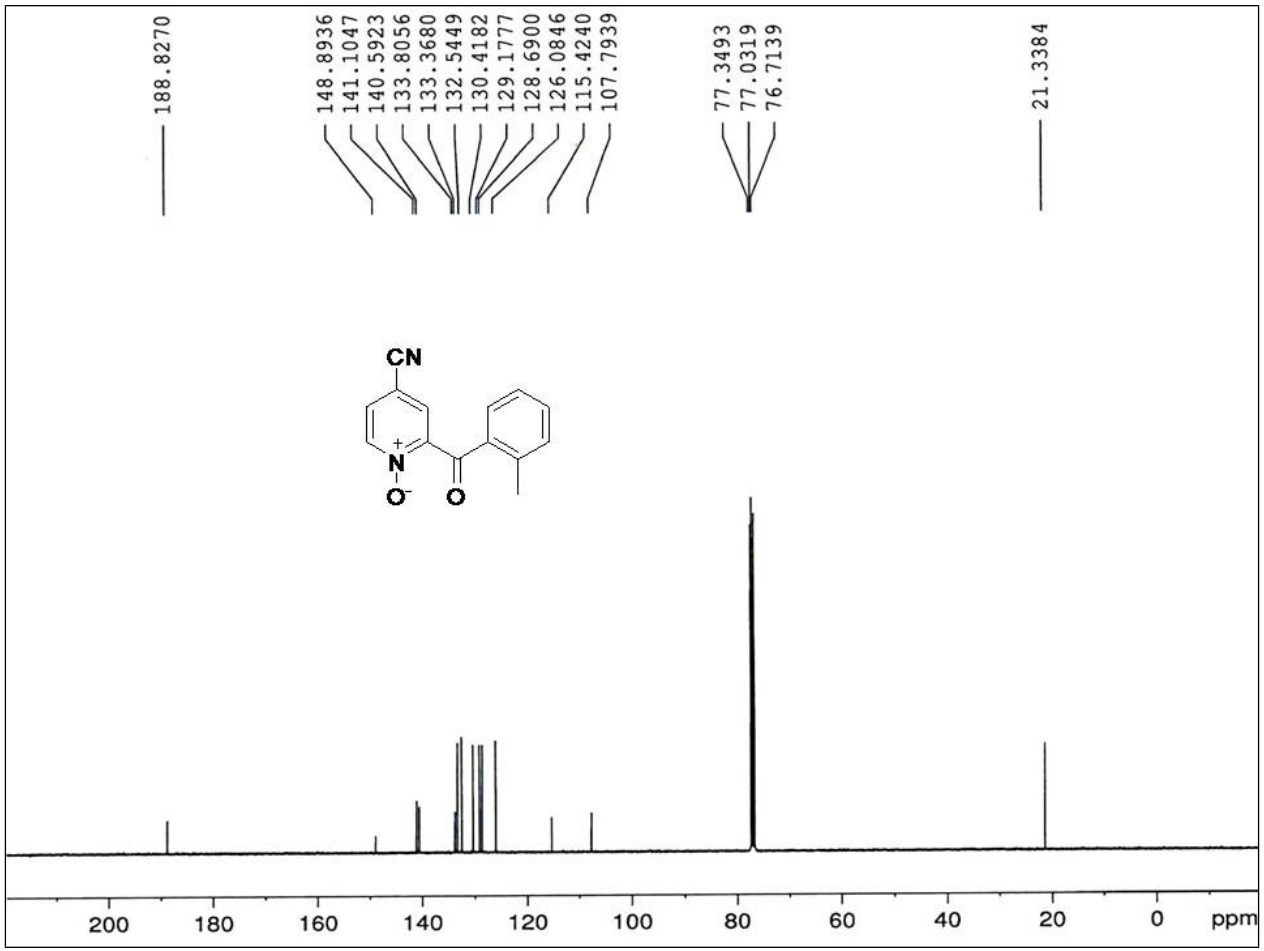


Fig 22. ^{13}C-NMR spectrum of **3h**

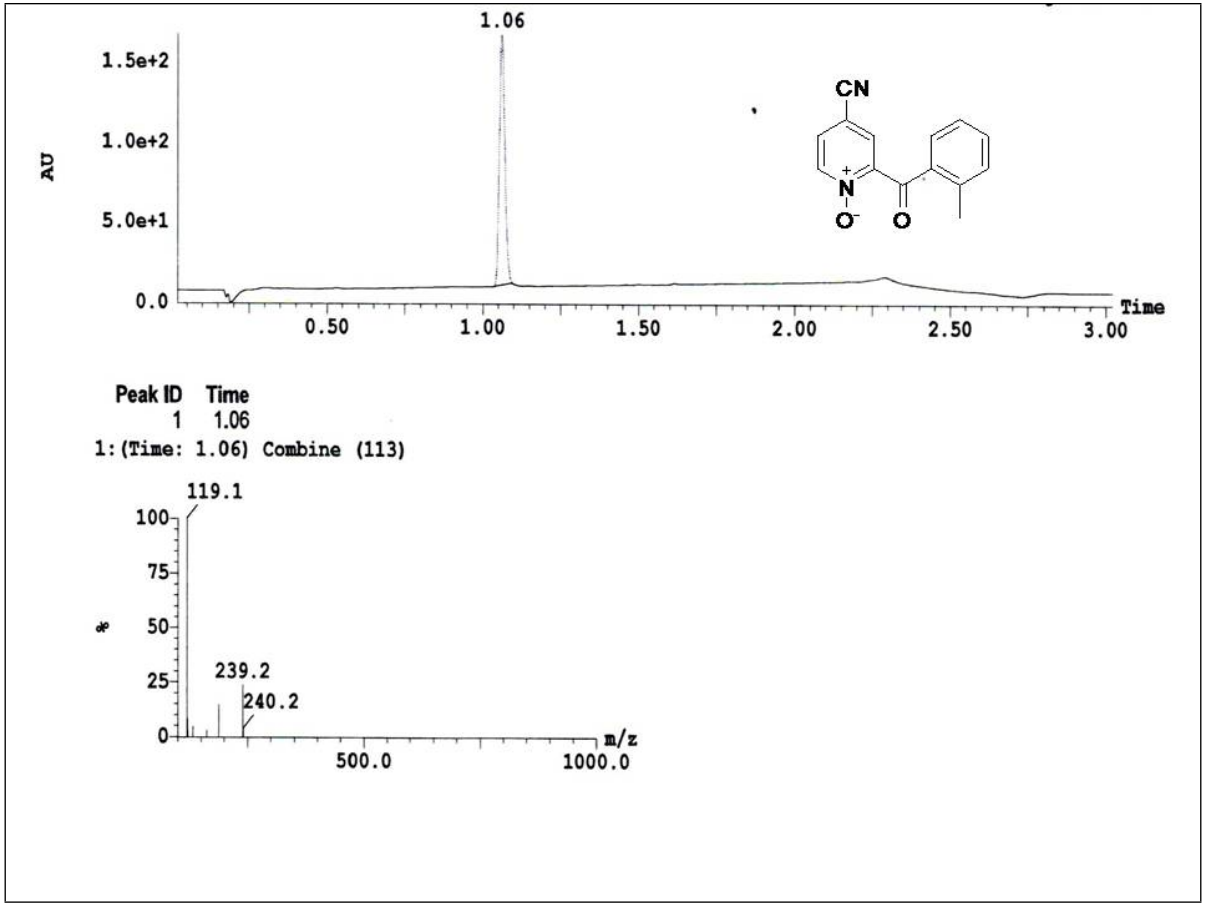


Fig 23. Mass spectrum of **3h**

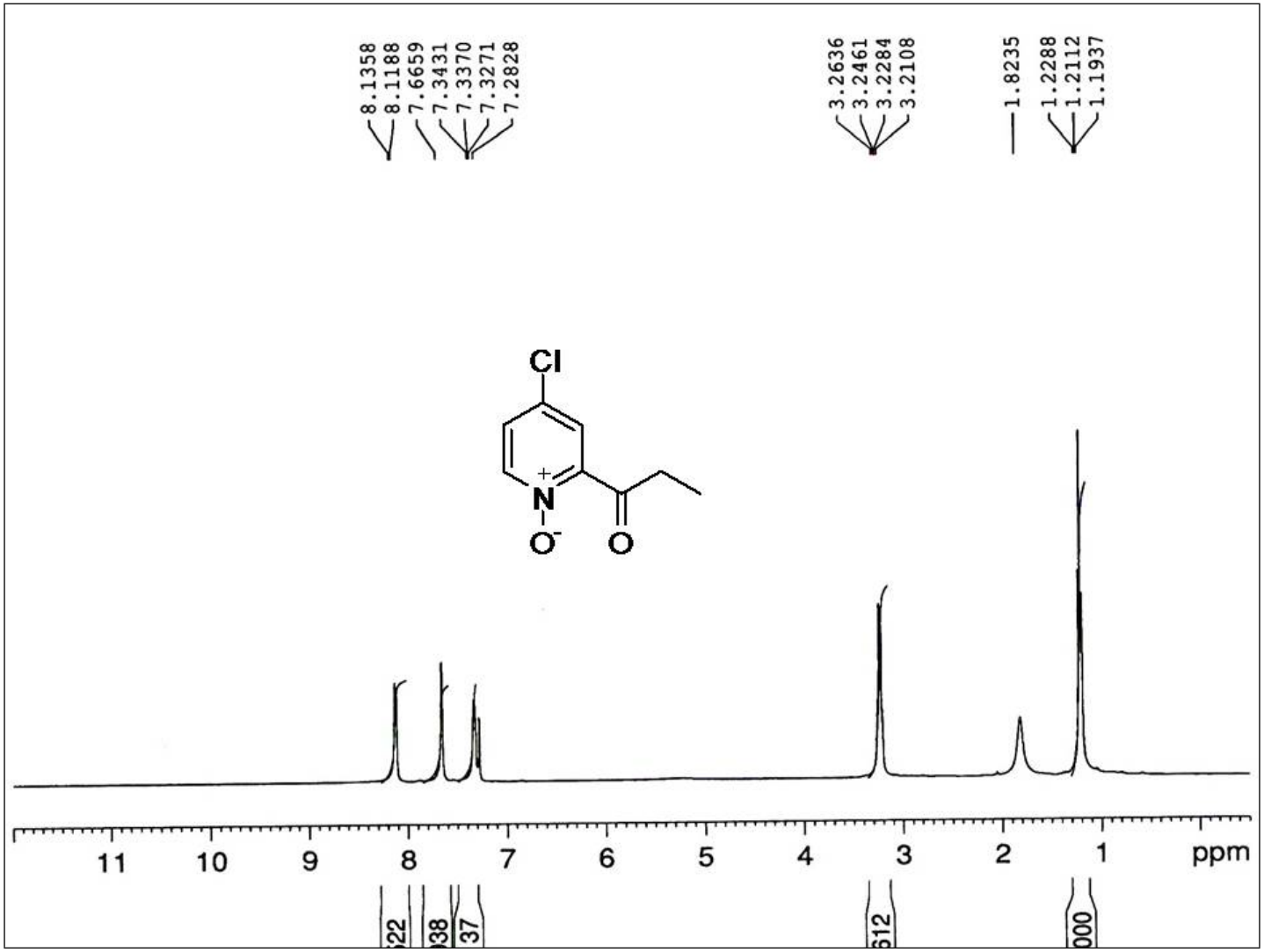


Fig 24. [1]H-NMR spectrum of **3i**

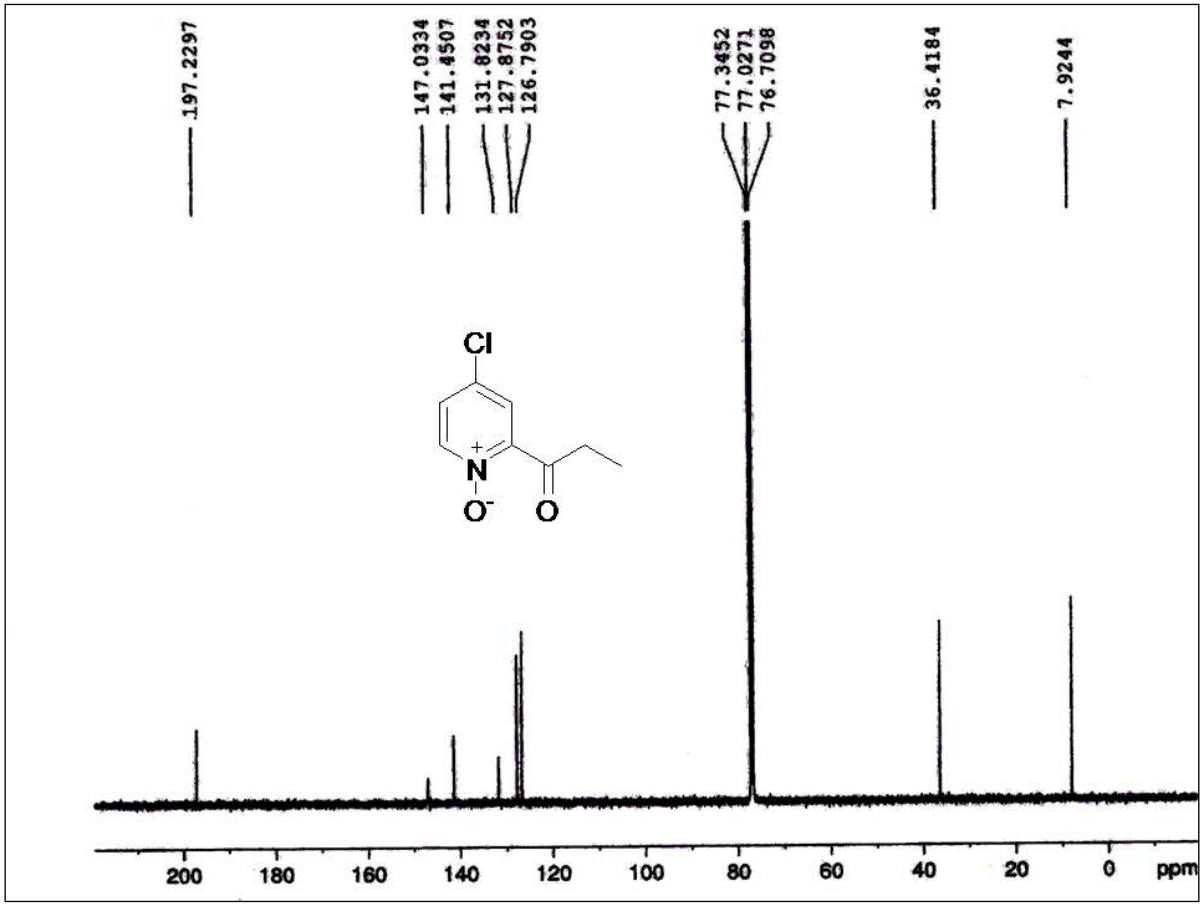


Fig 25. ^{13}C-NMR spectrum of **3i**

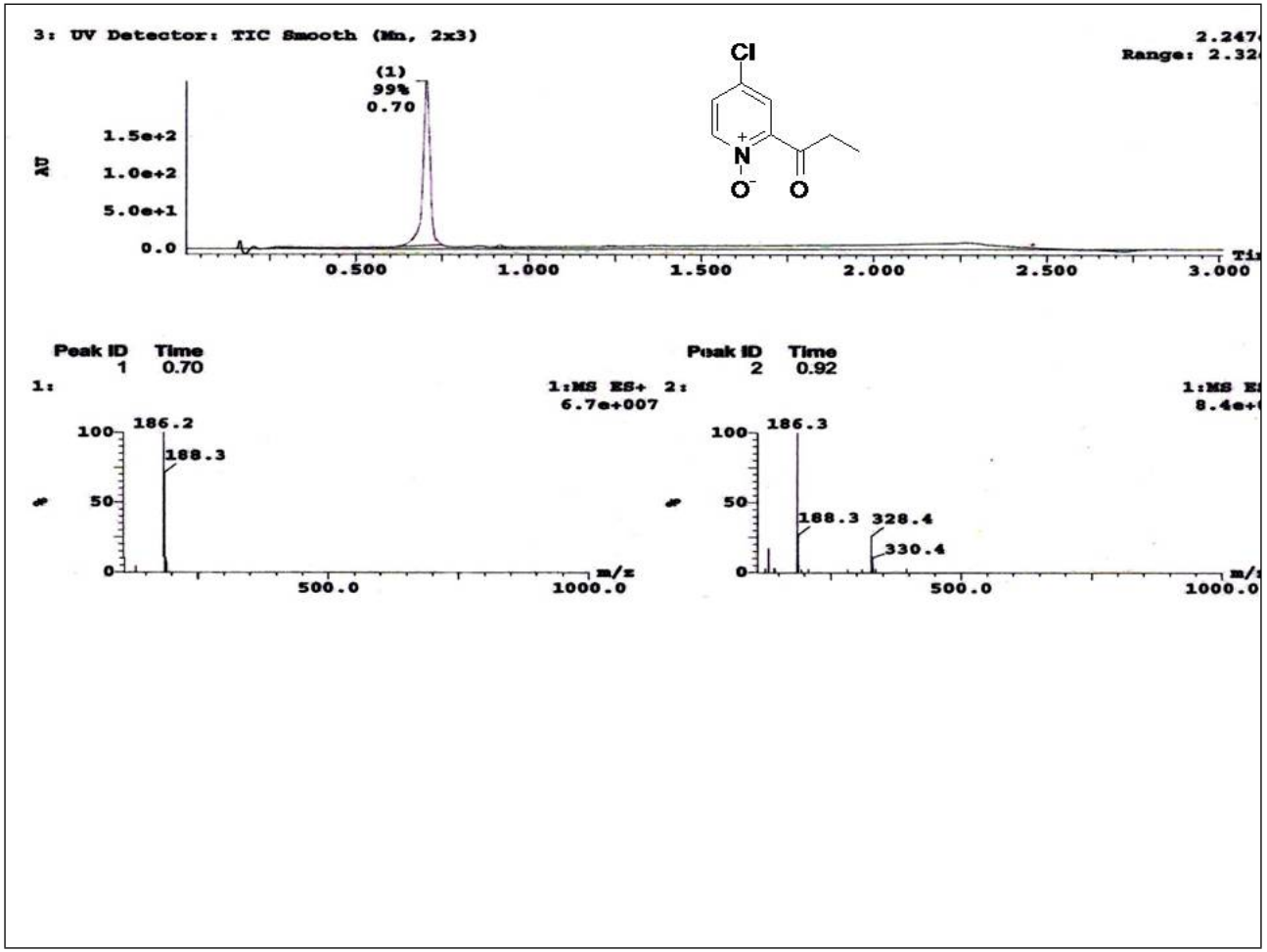


Fig 26. Mass spectrum of **3i**

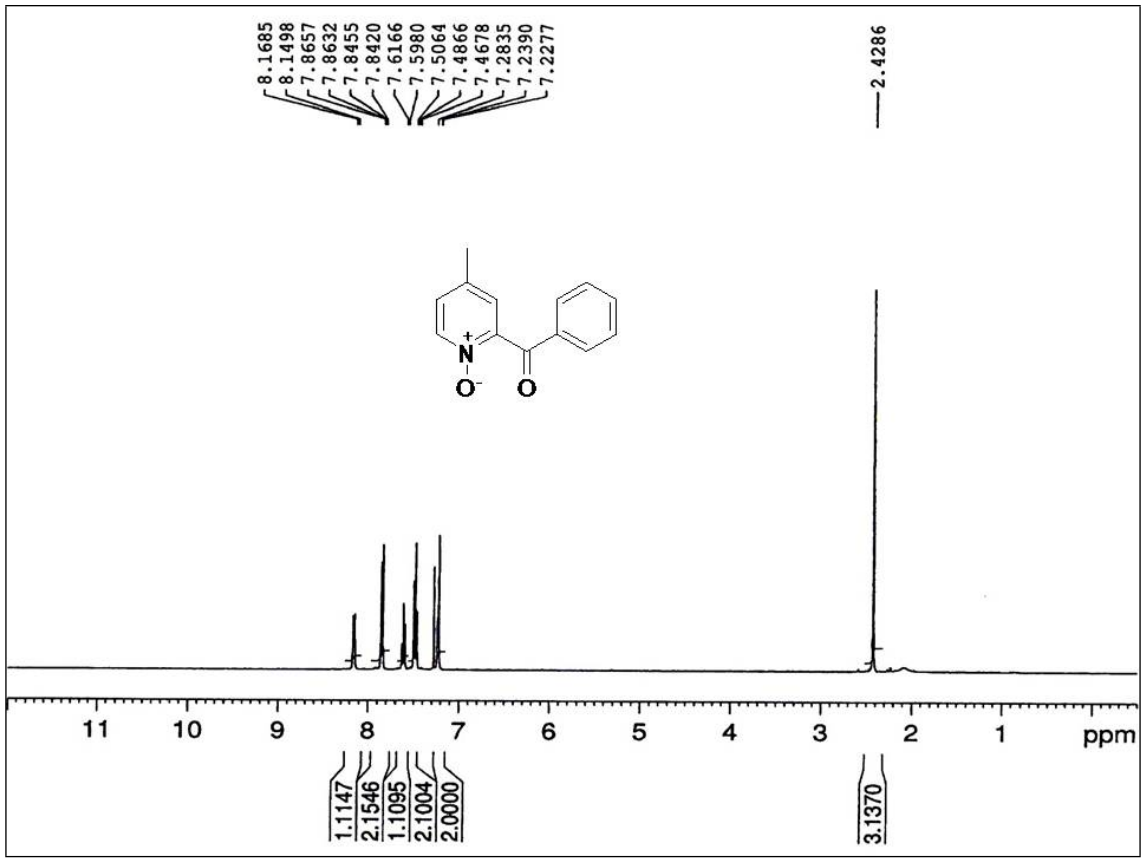


Fig 27. ^{1}H-NMR spectrum of **3j**

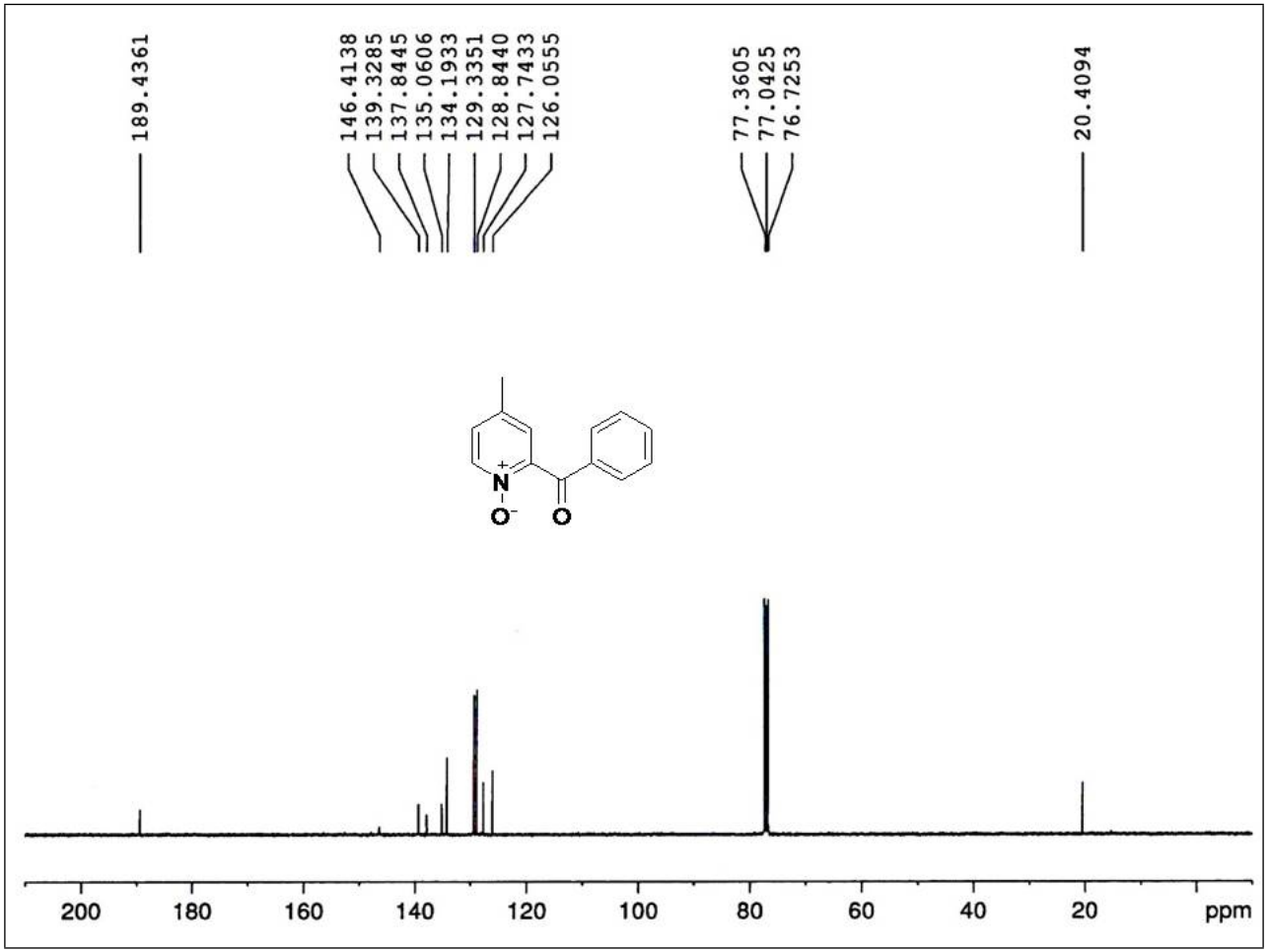


Fig 28. ^{13}C-NMR spectrum of **3j**

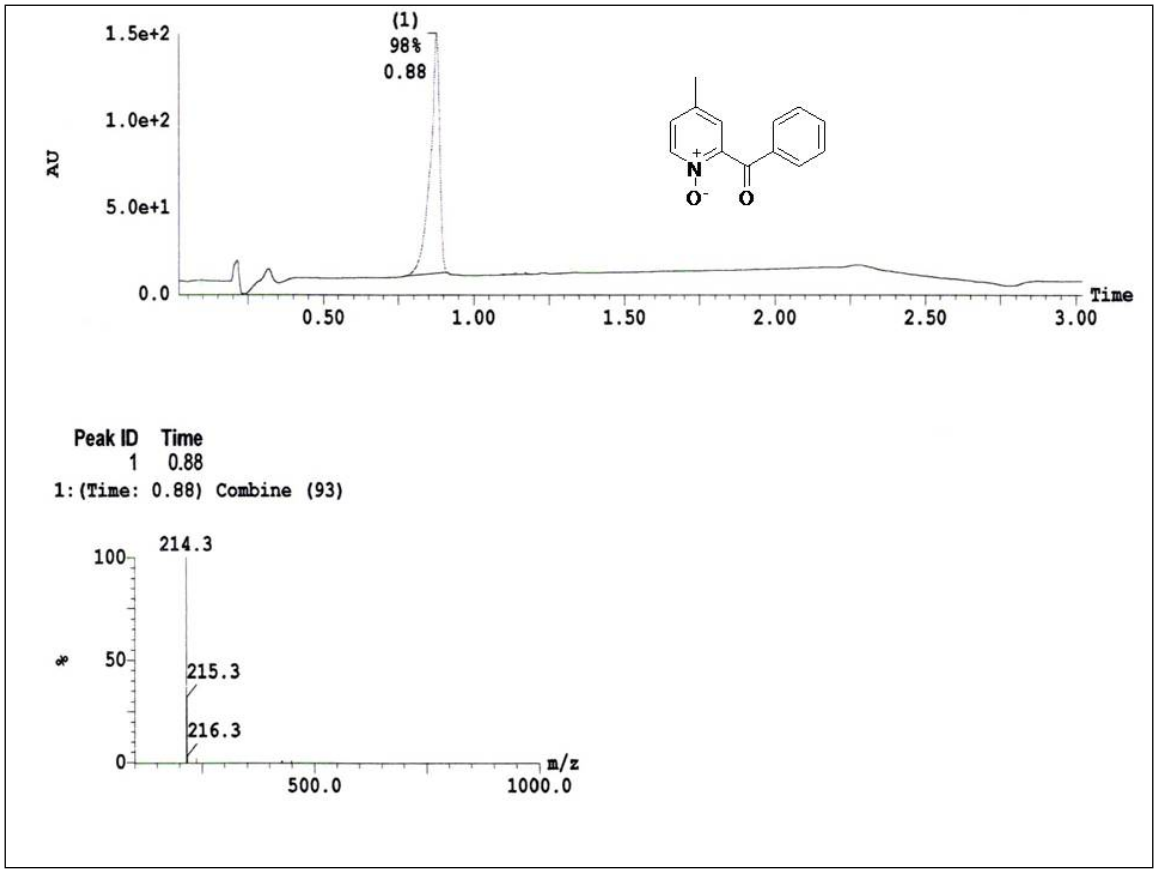


Fig 29. Mass spectrum of **3j**

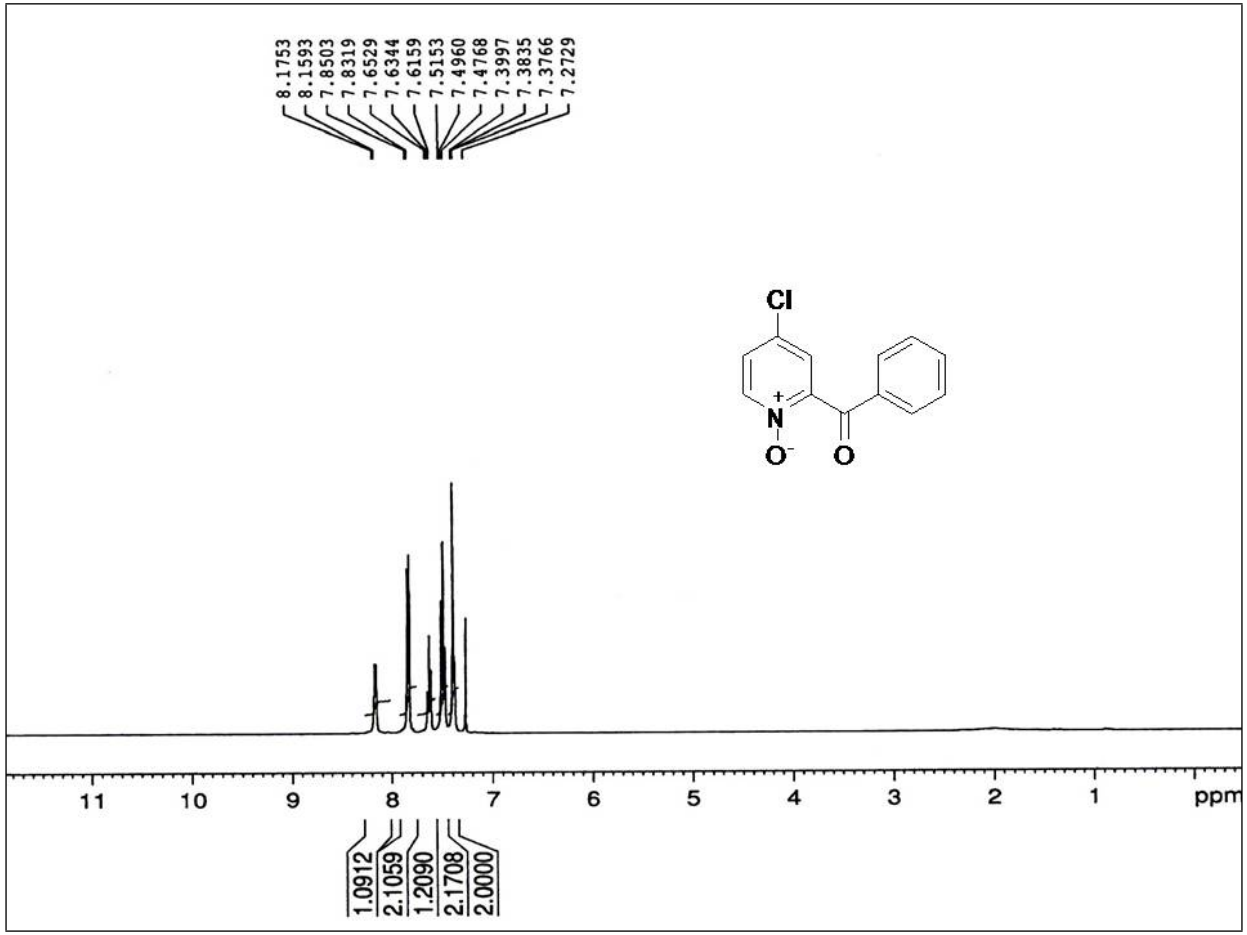


Fig 30. ^{1}H-NMR spectrum of **3k**

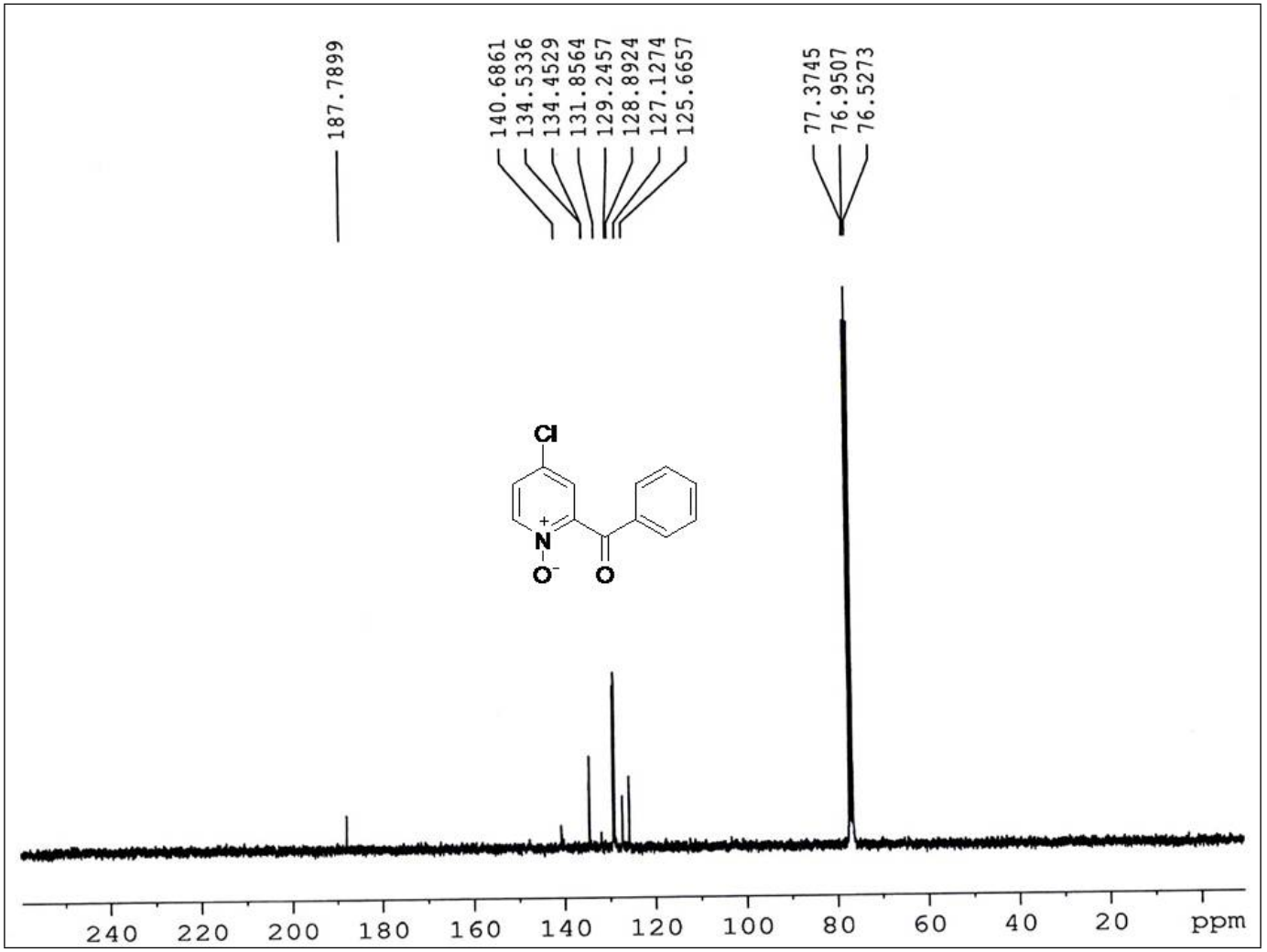


Fig 31. ^{13}C-NMR spectrum of **3k**

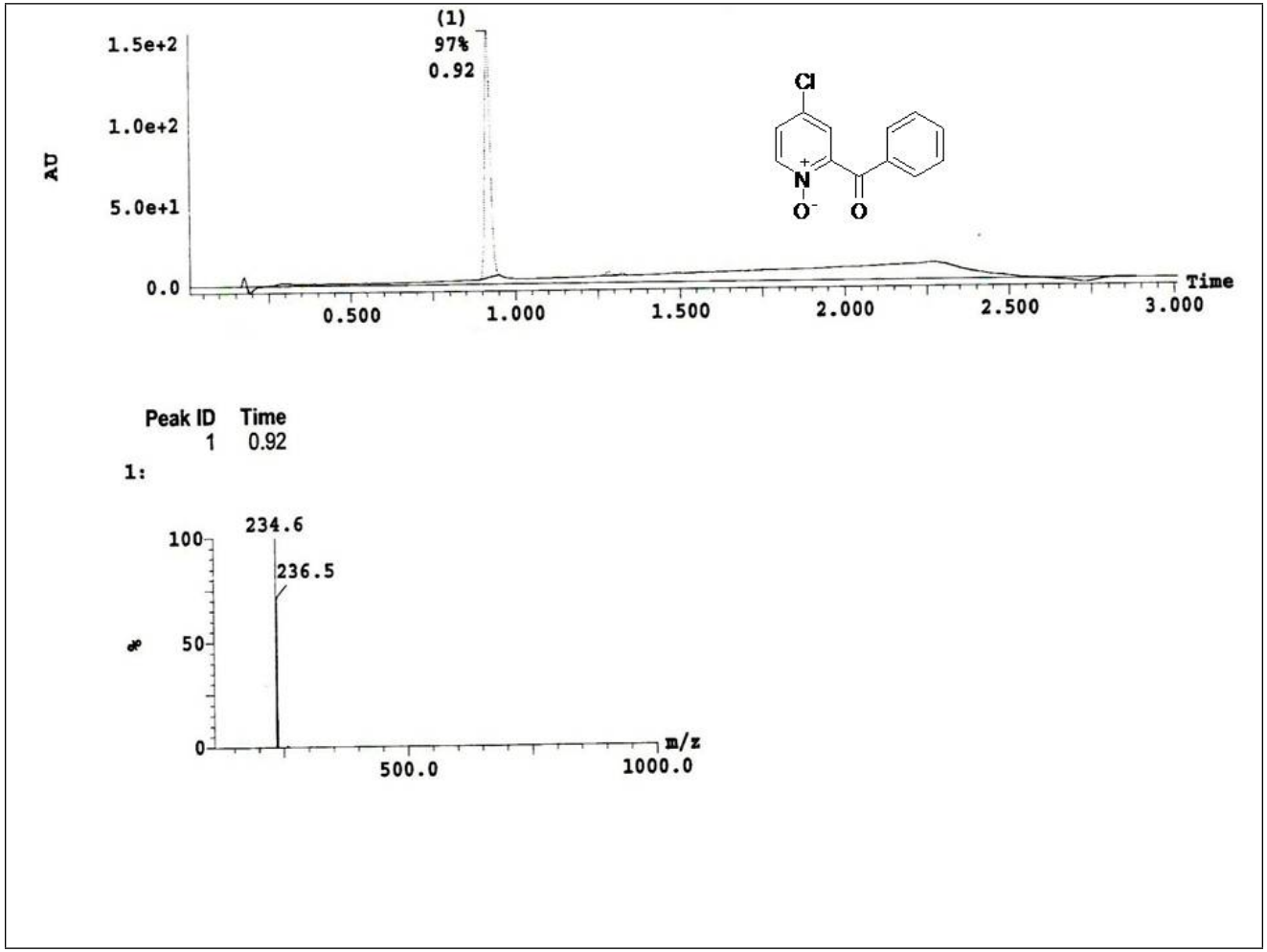


Fig 32. Mass spectrum of **3k**

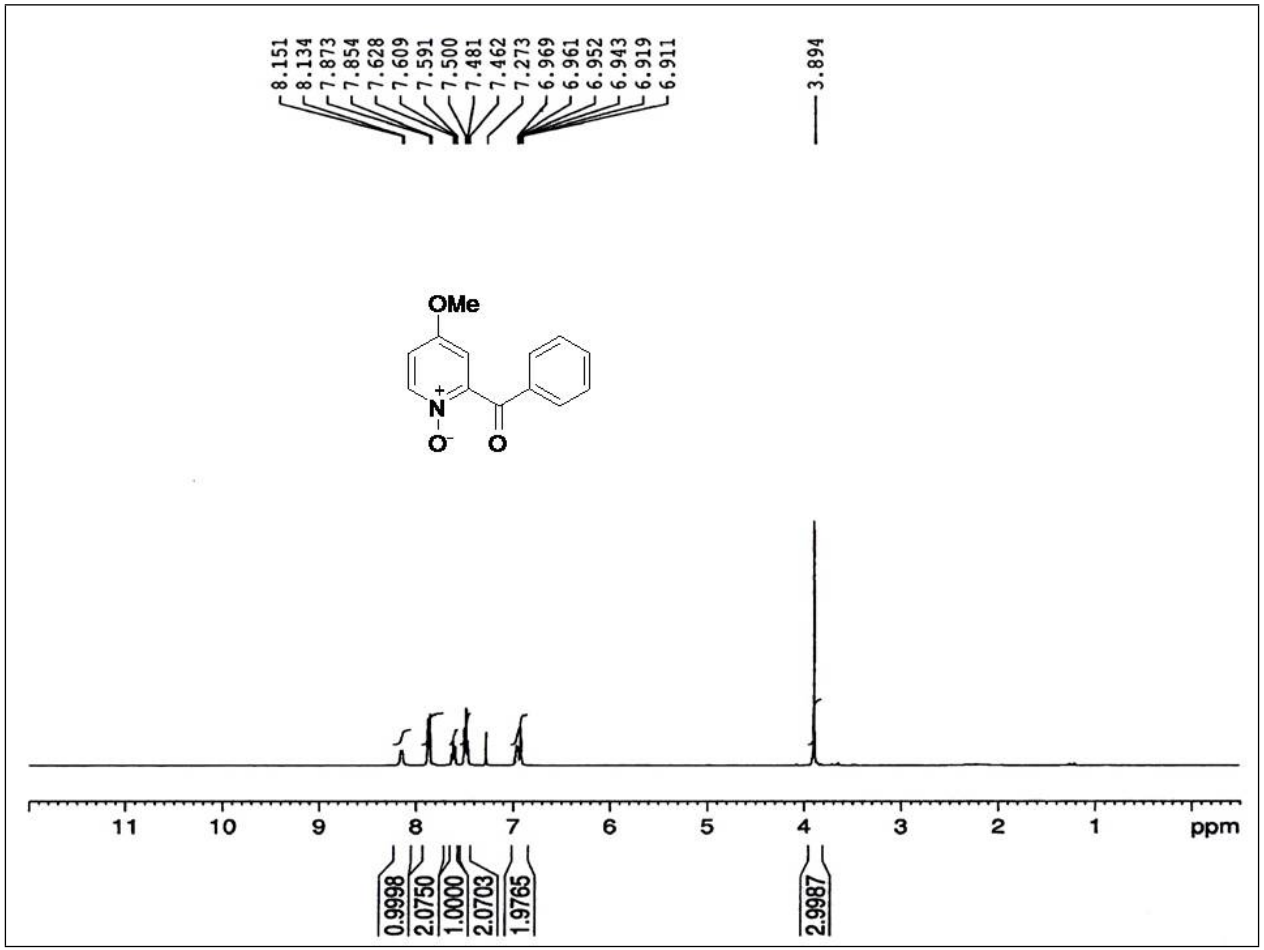


Fig 33. ^{1}H-NMR spectrum of **3l**

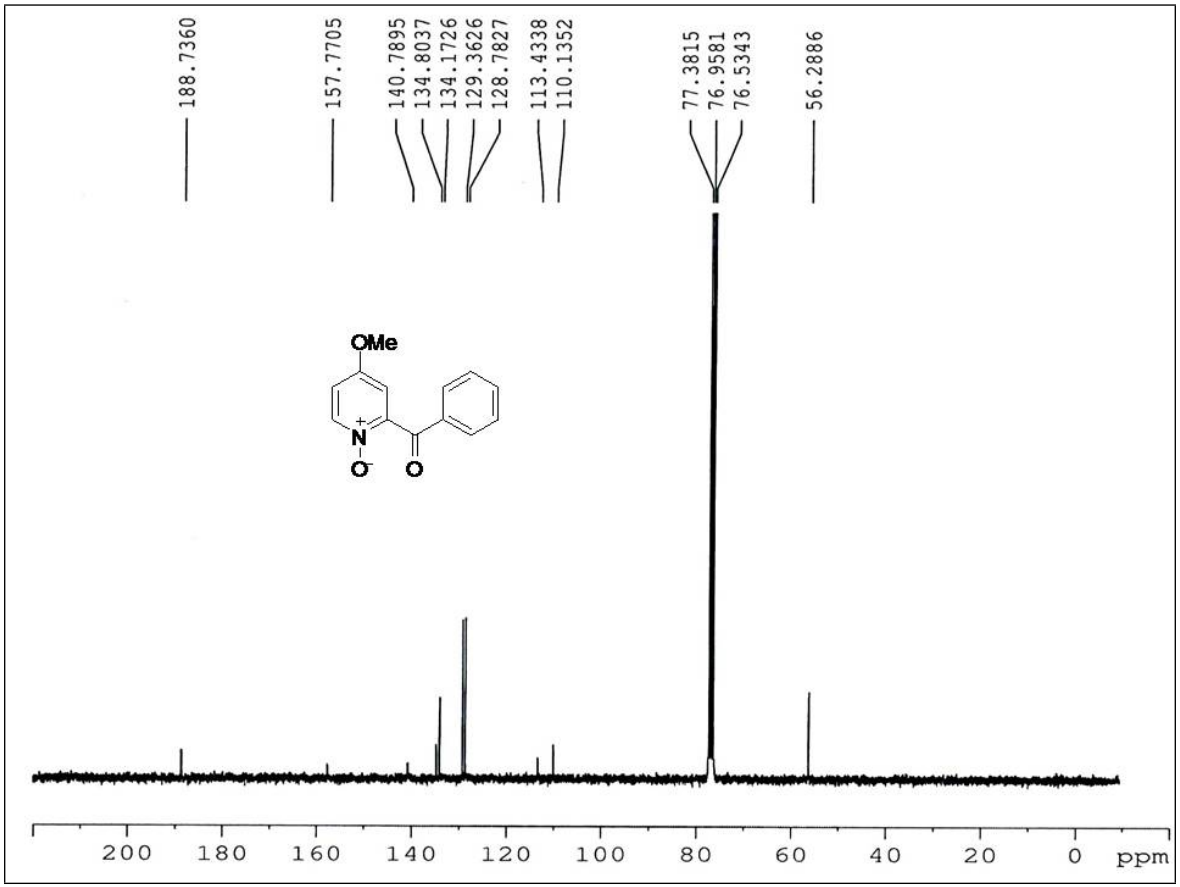


Fig 34. ^{13}C-NMR spectrum of **3l**

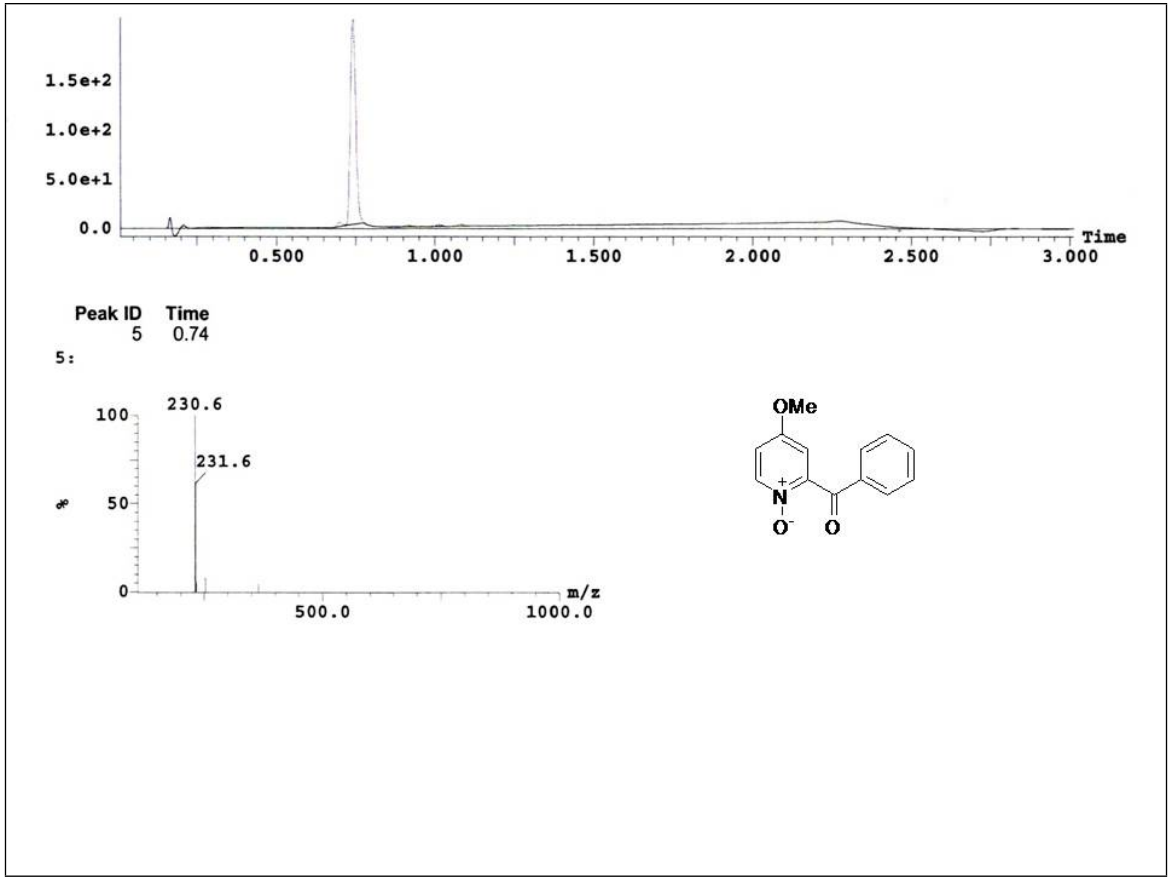


Fig 35. Mass spectrum of **3l**

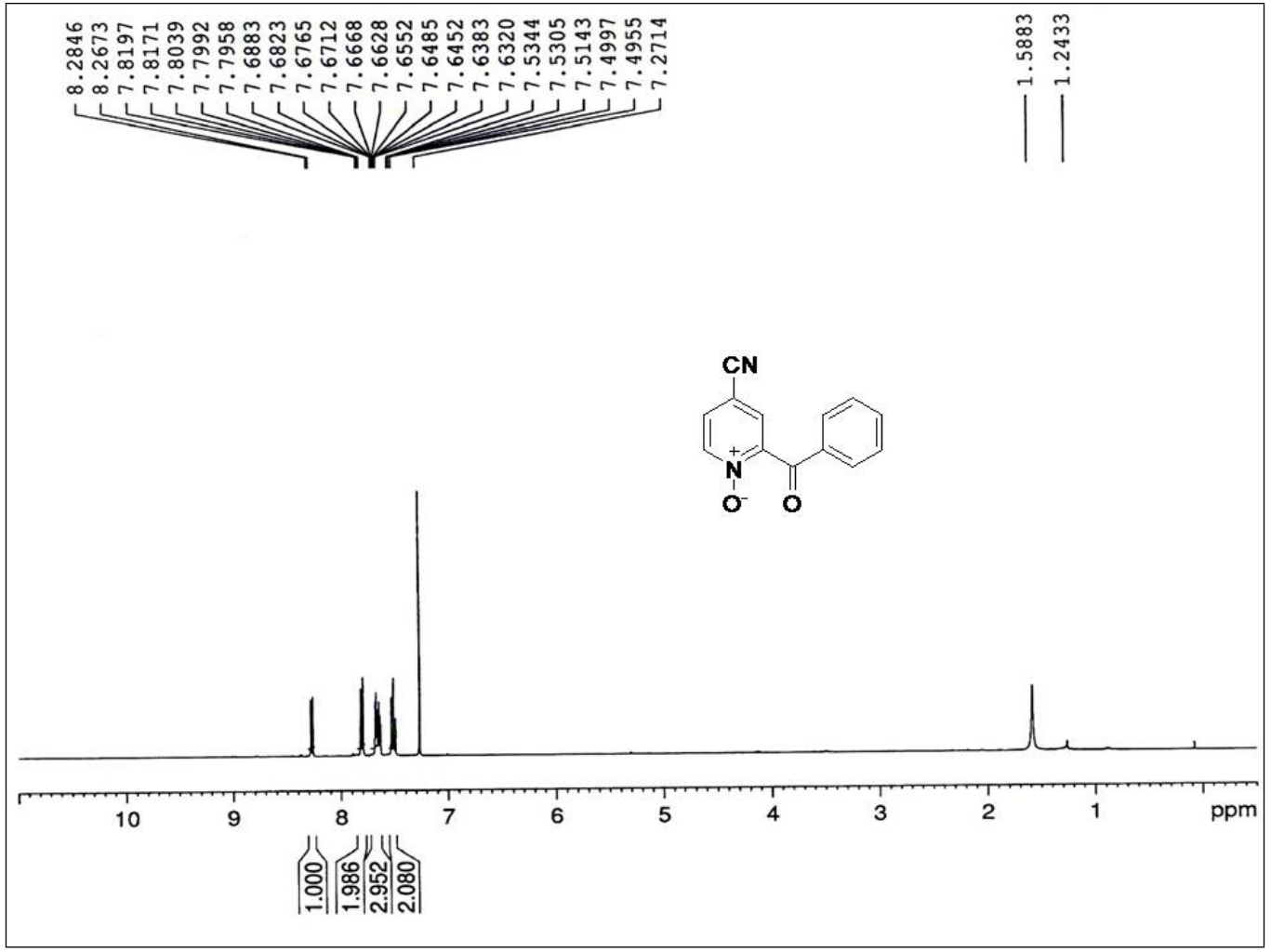


Fig 36. ^{1}H-NMR spectrum of **3m**

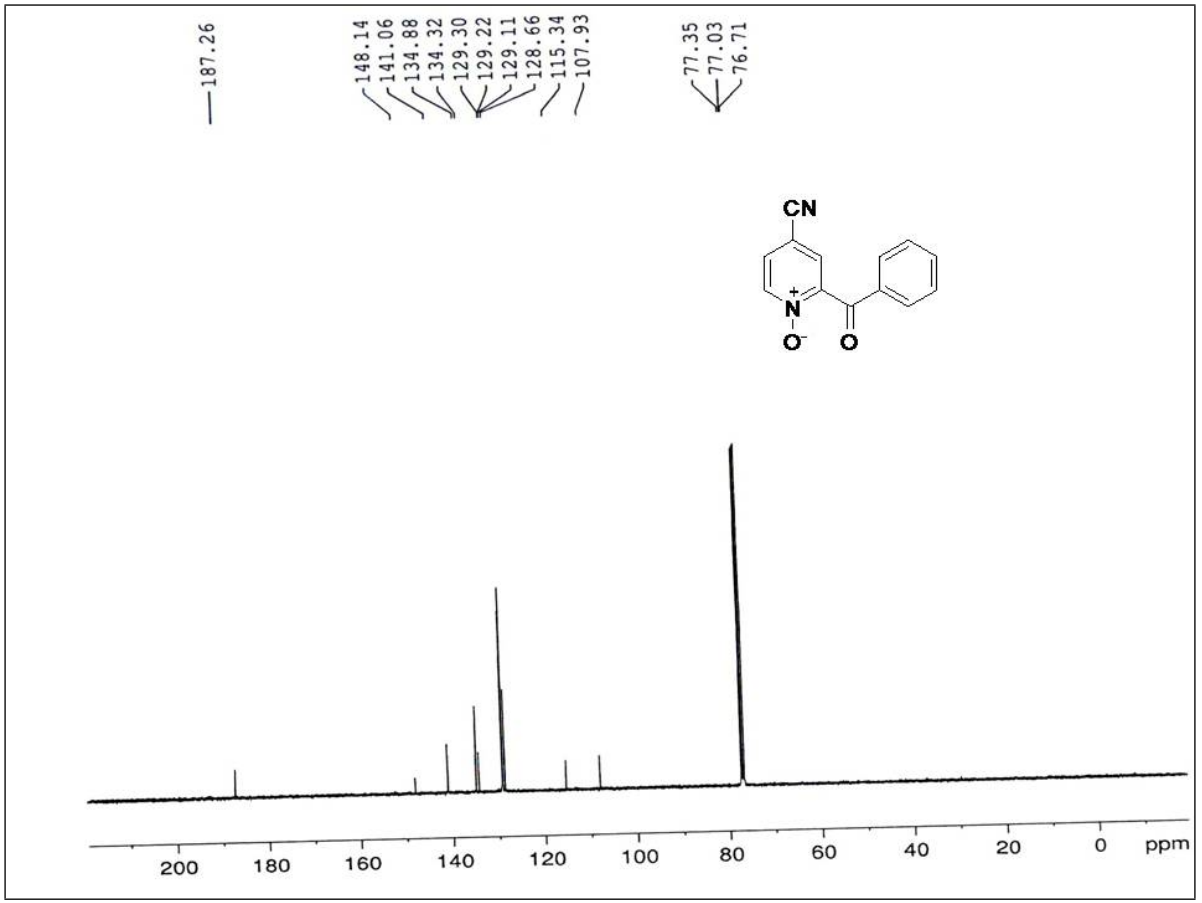


Fig 37. ^{13}C-NMR spectrum of **3m**

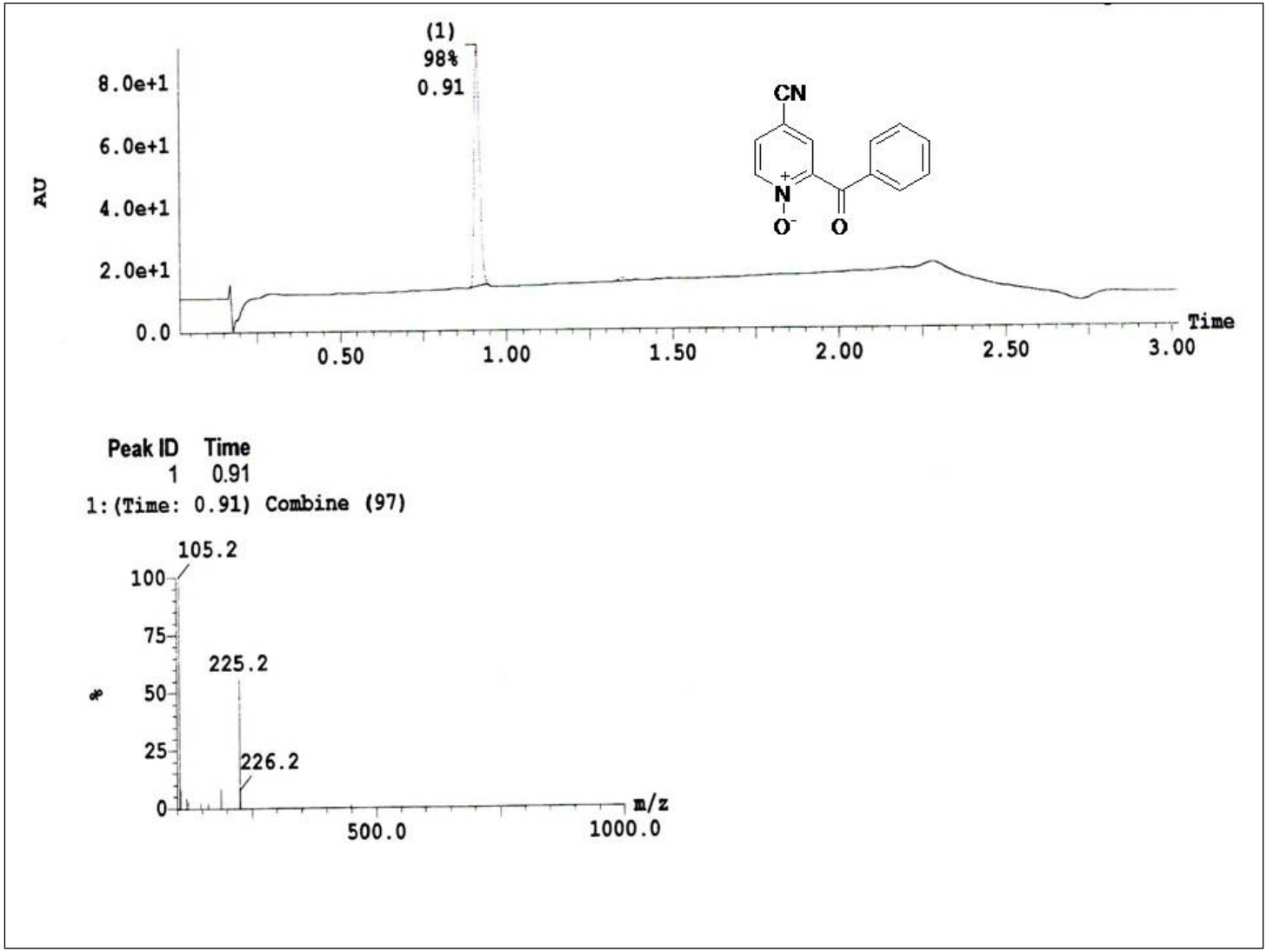


Fig 38. Mass spectrum of **3m**

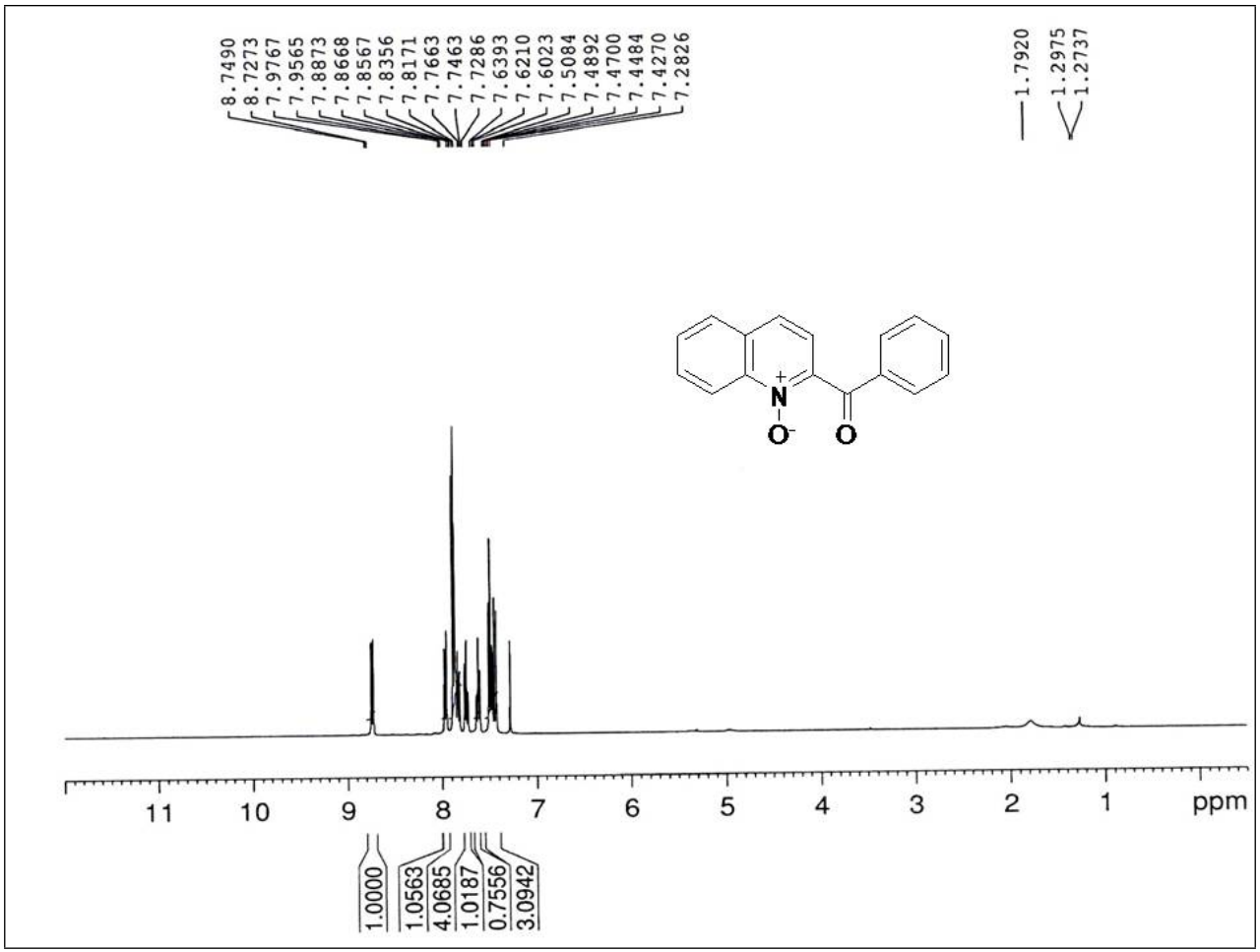


Fig 39. ^{1}H-NMR spectrum of **3n**

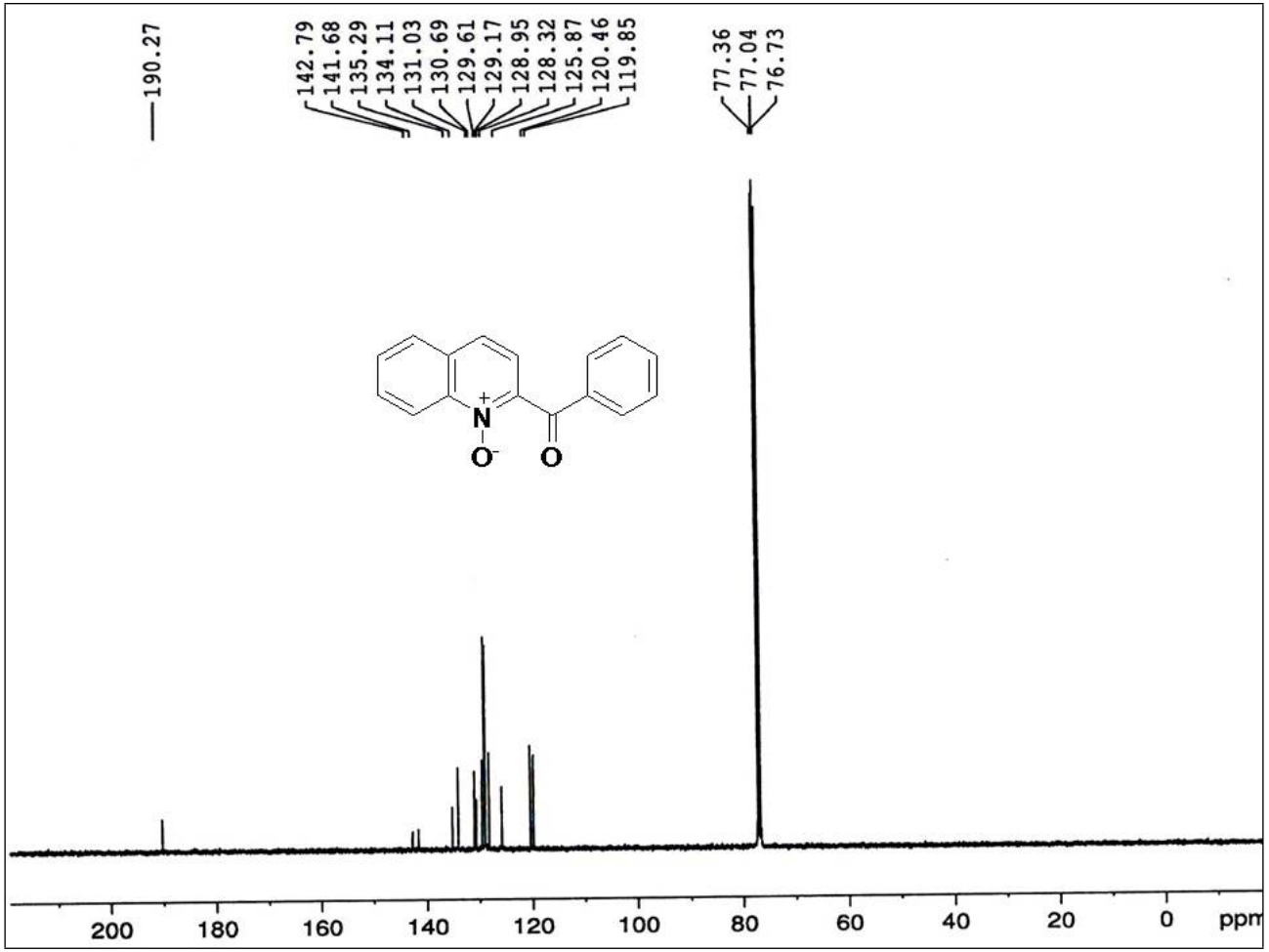


Fig 40. ^{13}C-NMR spectrum of **3n**

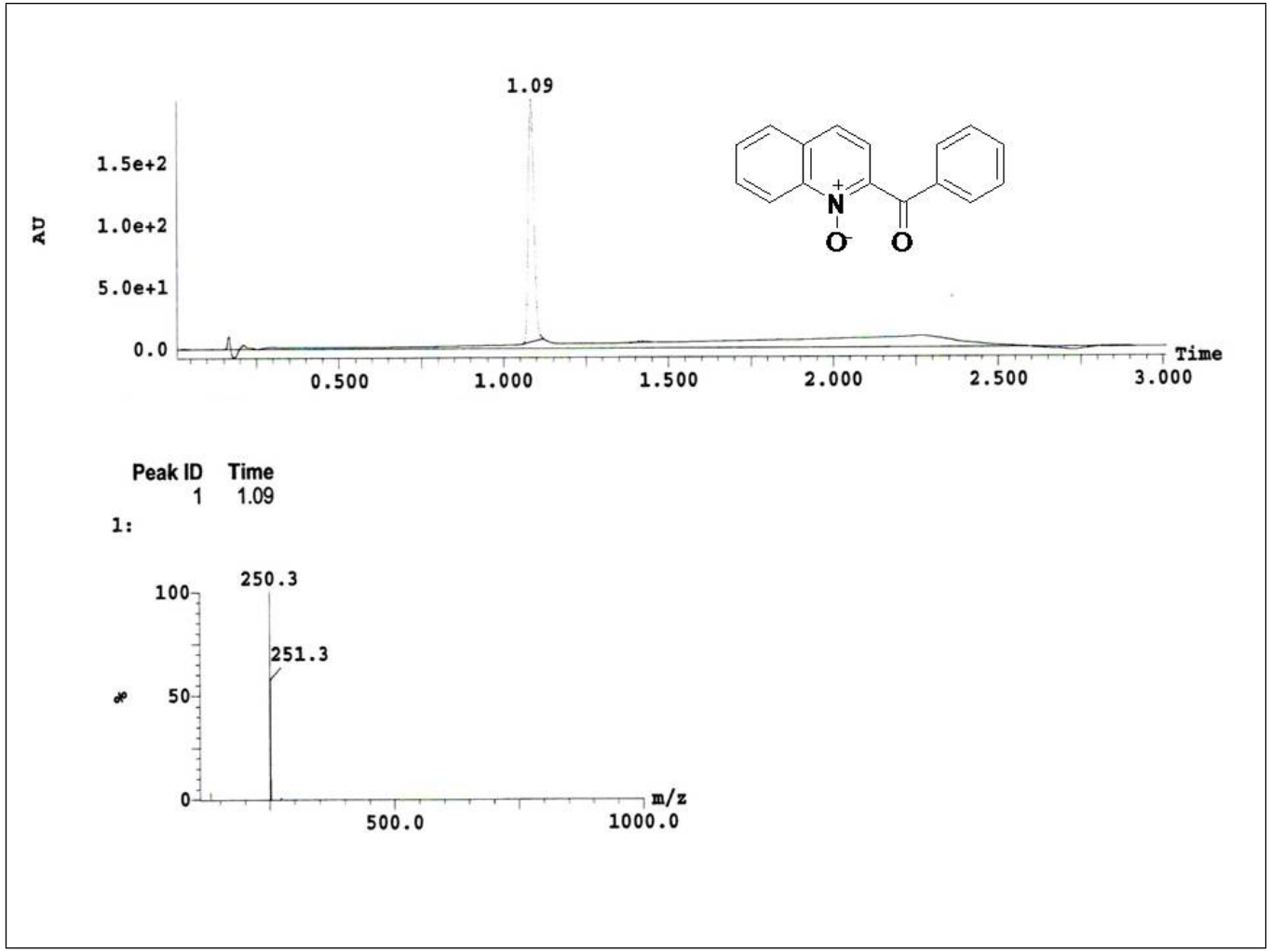


Fig 41. Mass spectrum of **3n**

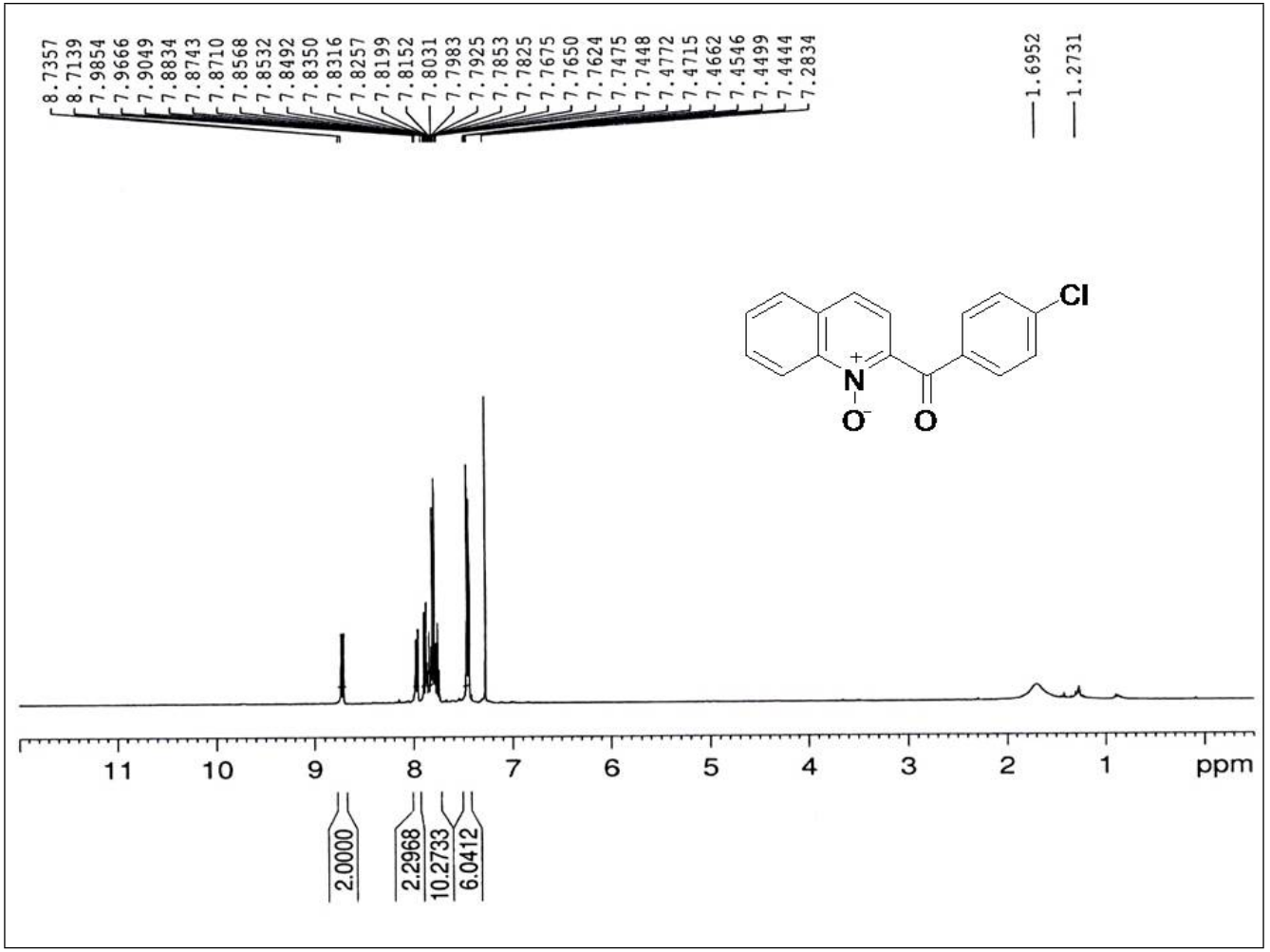


Fig 42. ^{1}H-NMR spectrum of **3o**

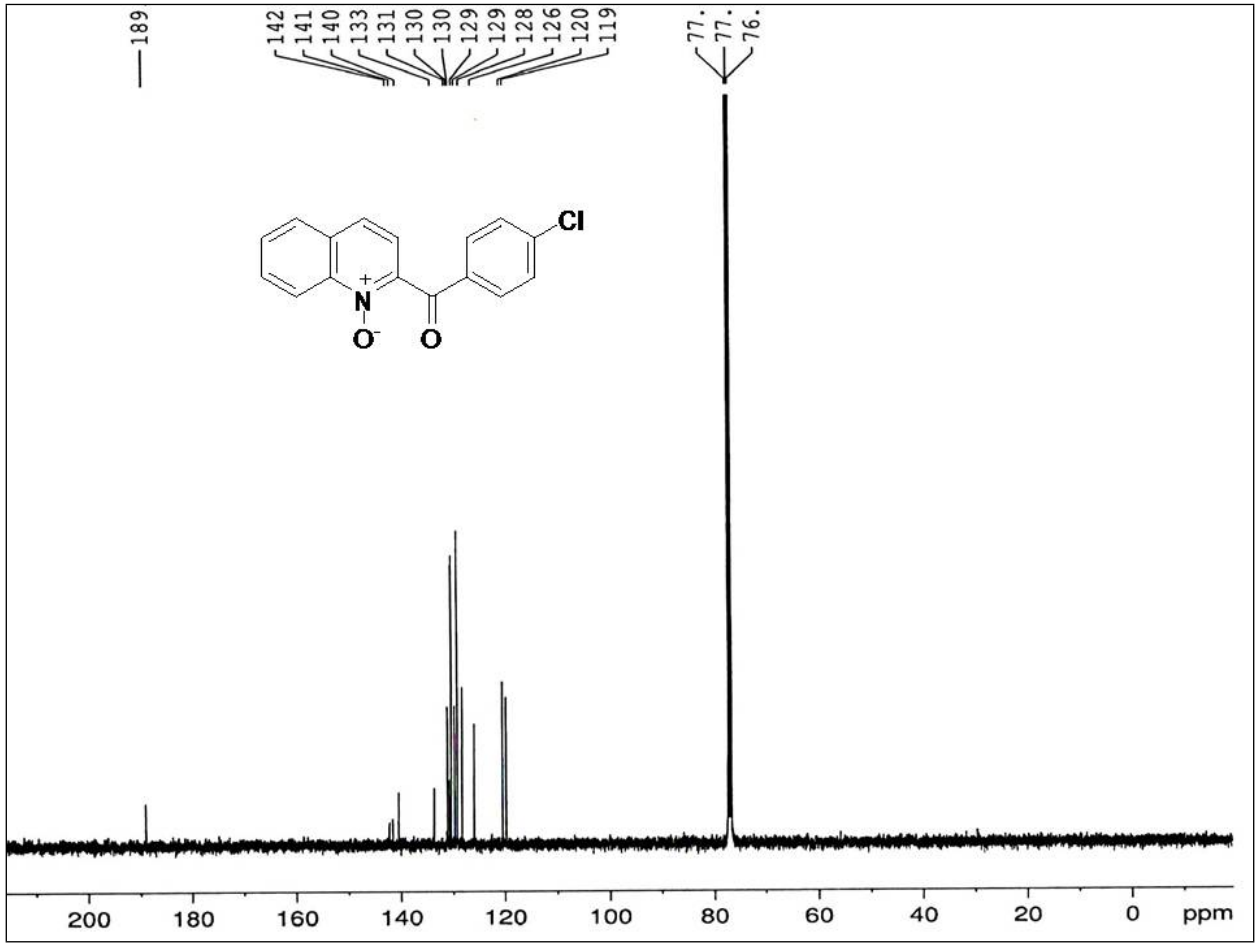


Fig 43. ^{13}C-NMR spectrum of **3o**

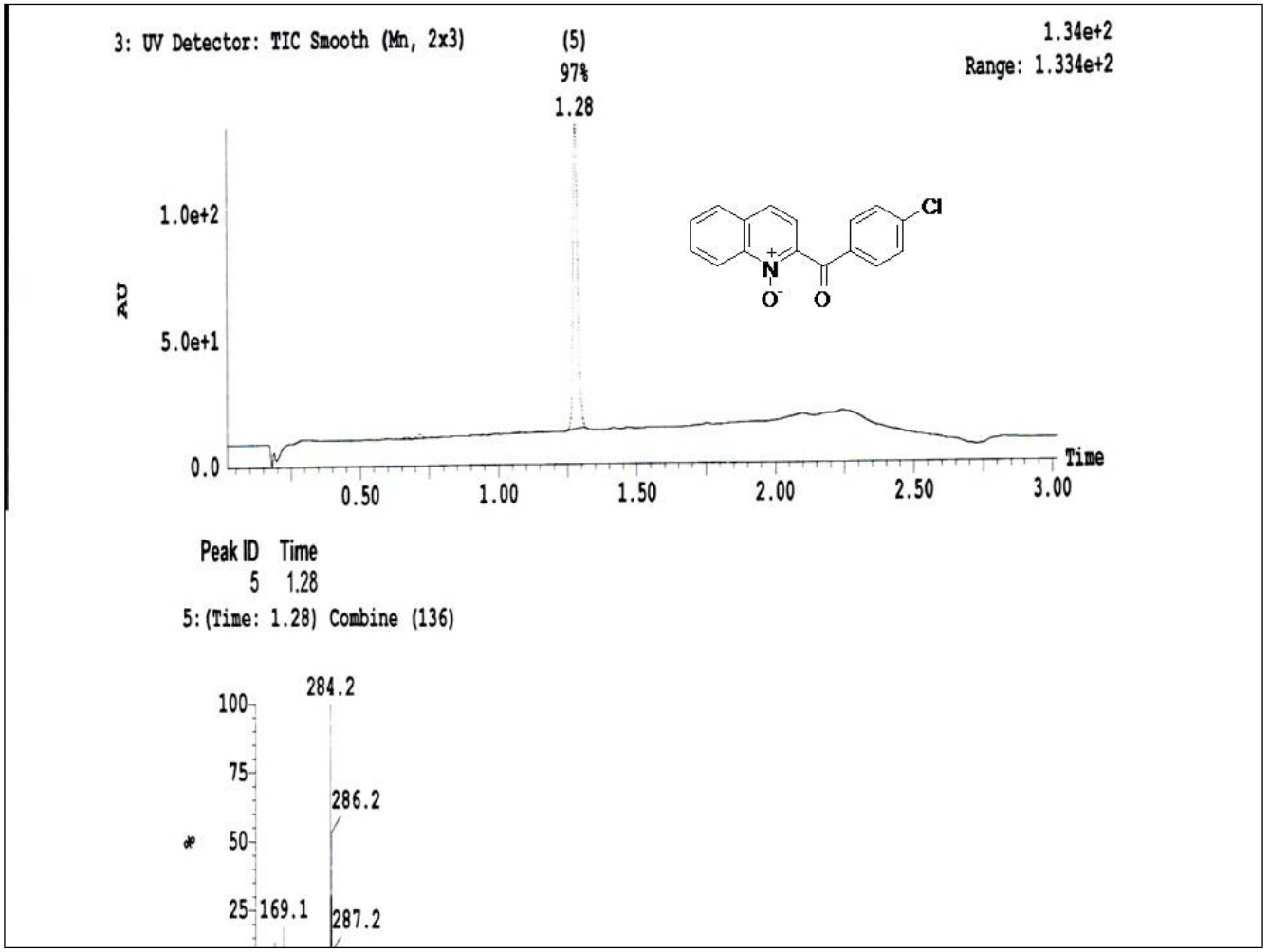


Fig 44. Mass spectrum of **3o**

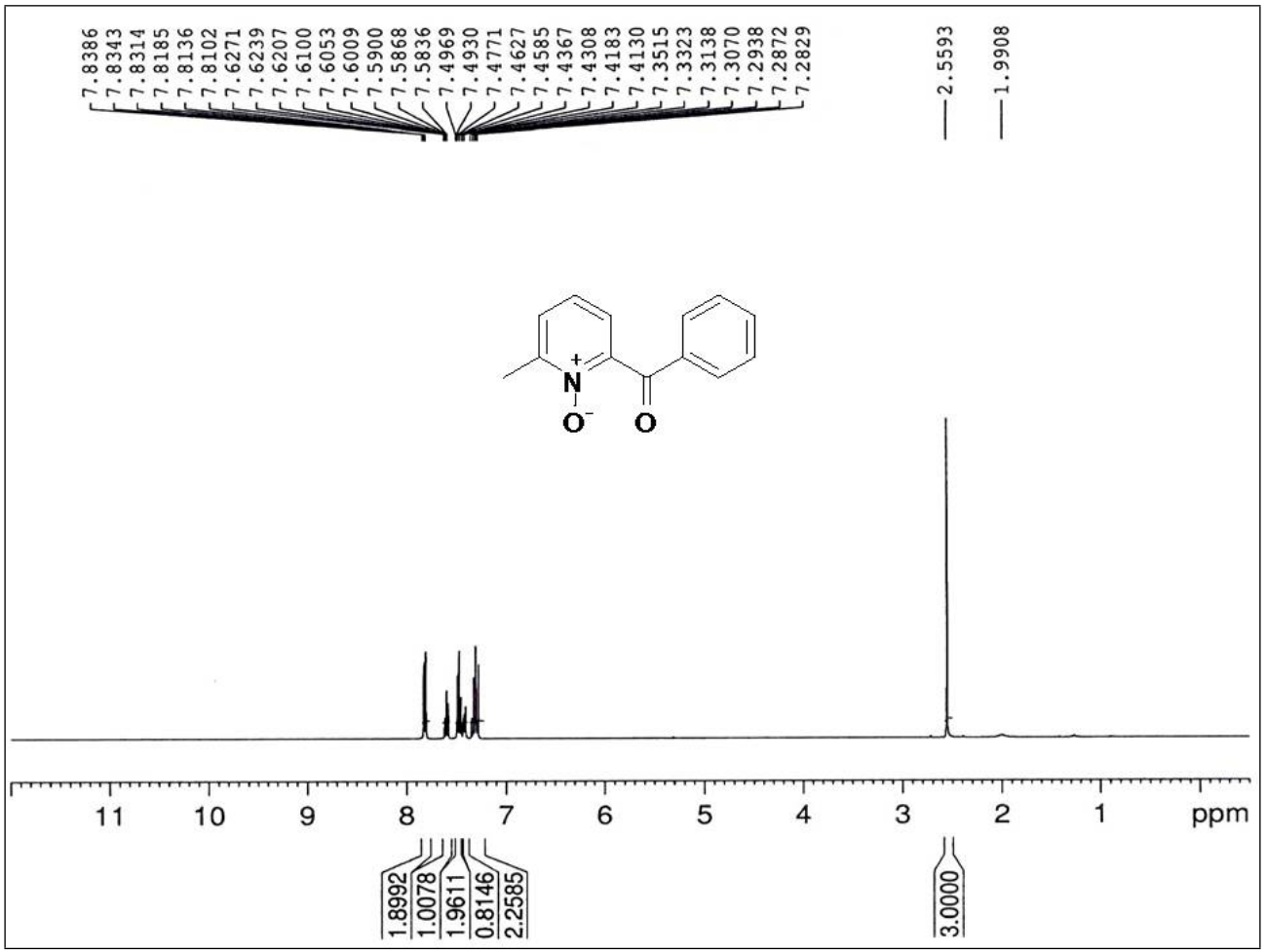


Fig 45. ^{1}H-NMR spectrum of **3p**

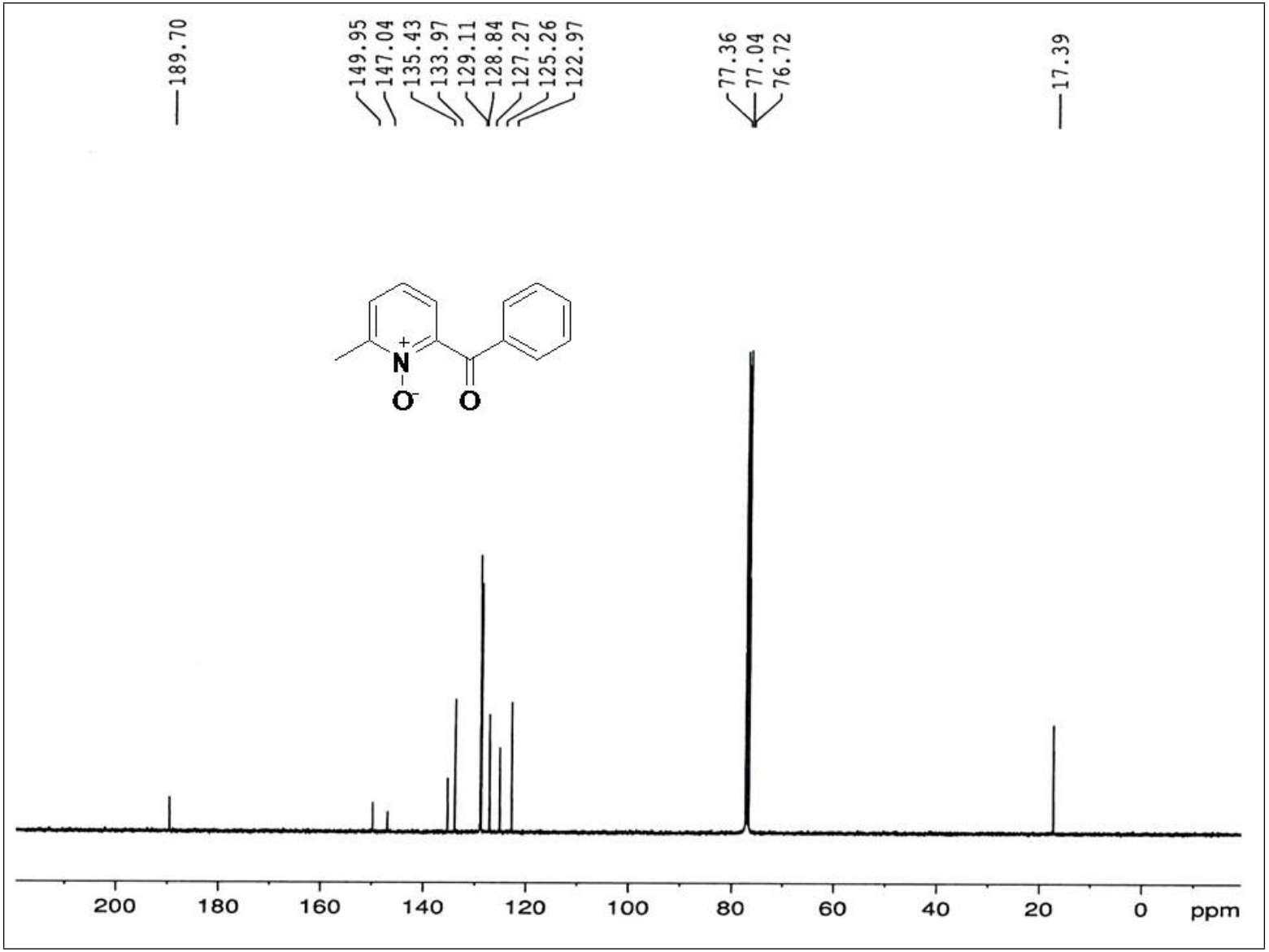


Fig 46. ^{13}C-NMR spectrum of **3p**

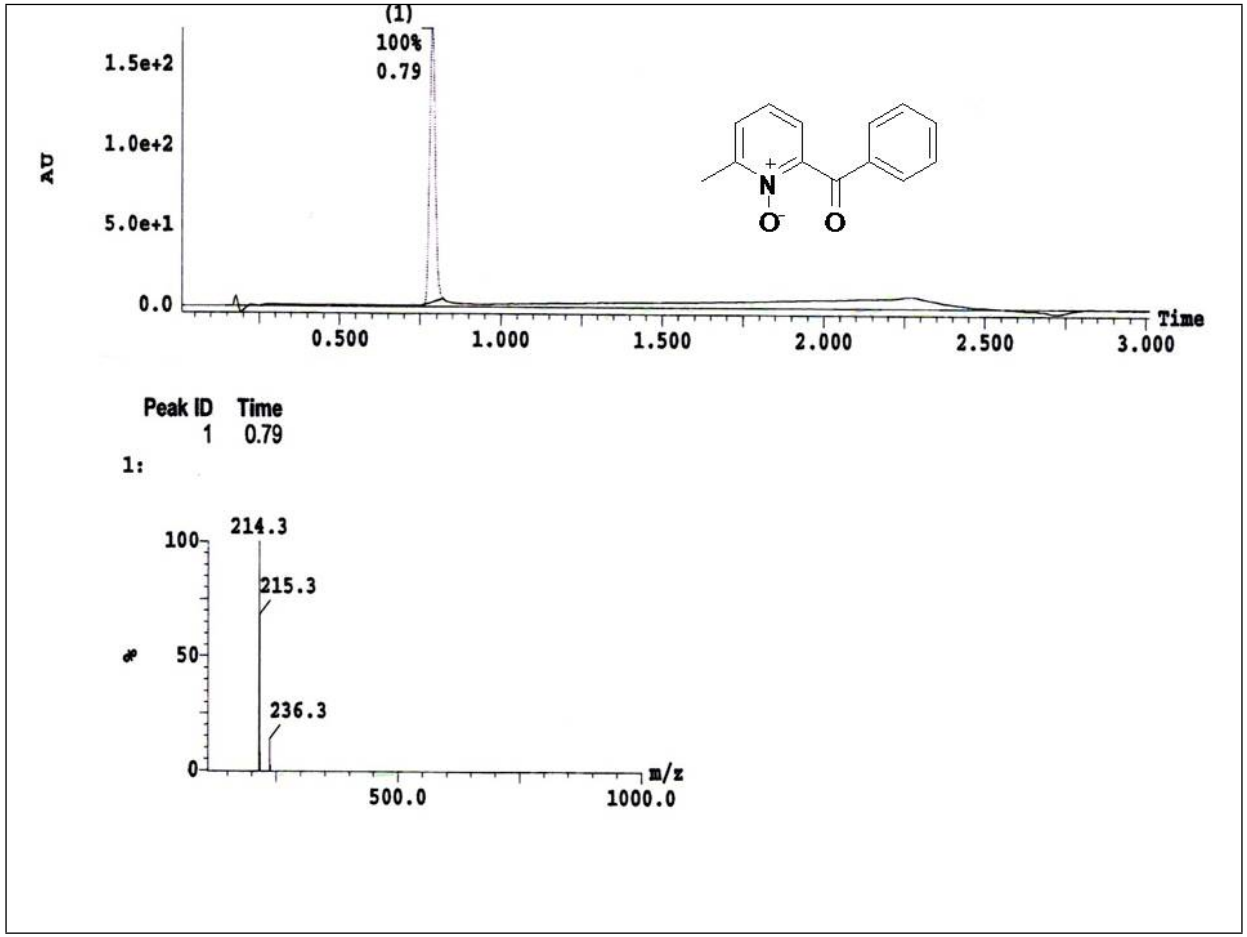


Fig 47. Mass spectrum of **3p**

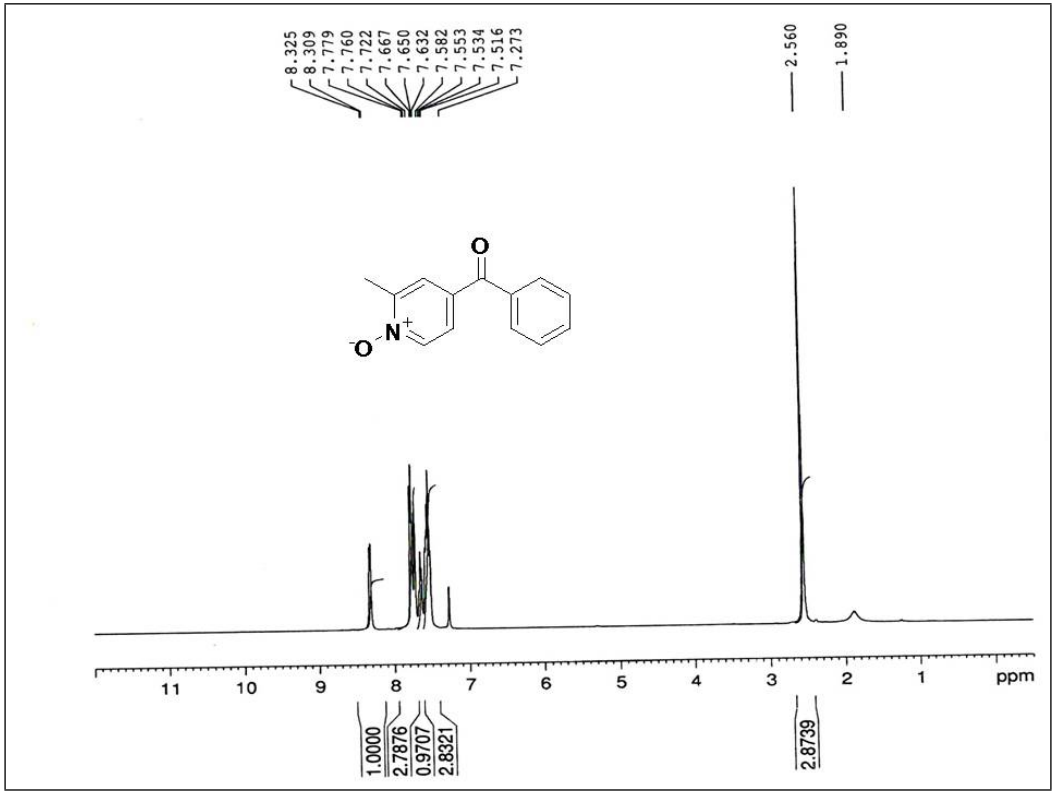


Fig 48. ^{1}H-NMR spectrum of **3p'**

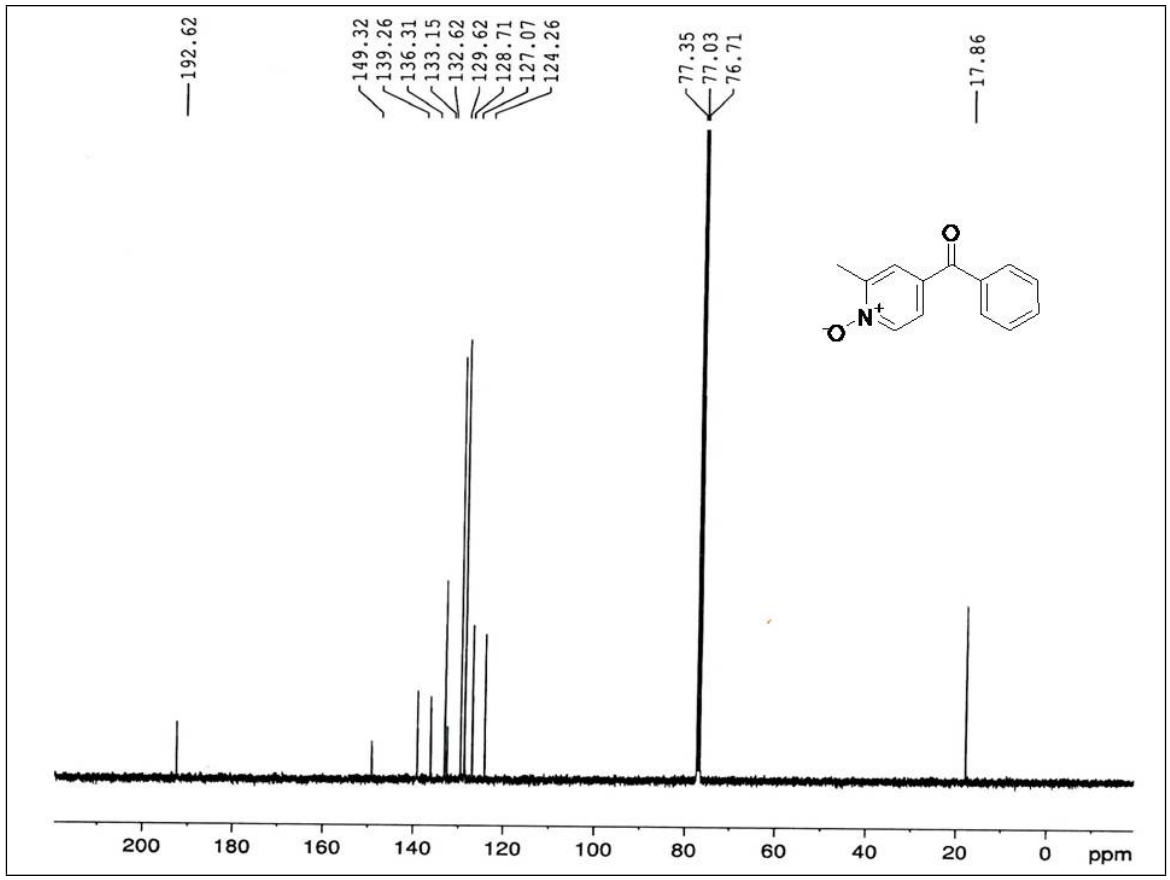


Fig 49. ^{13}C-NMR spectrum of **3p'**

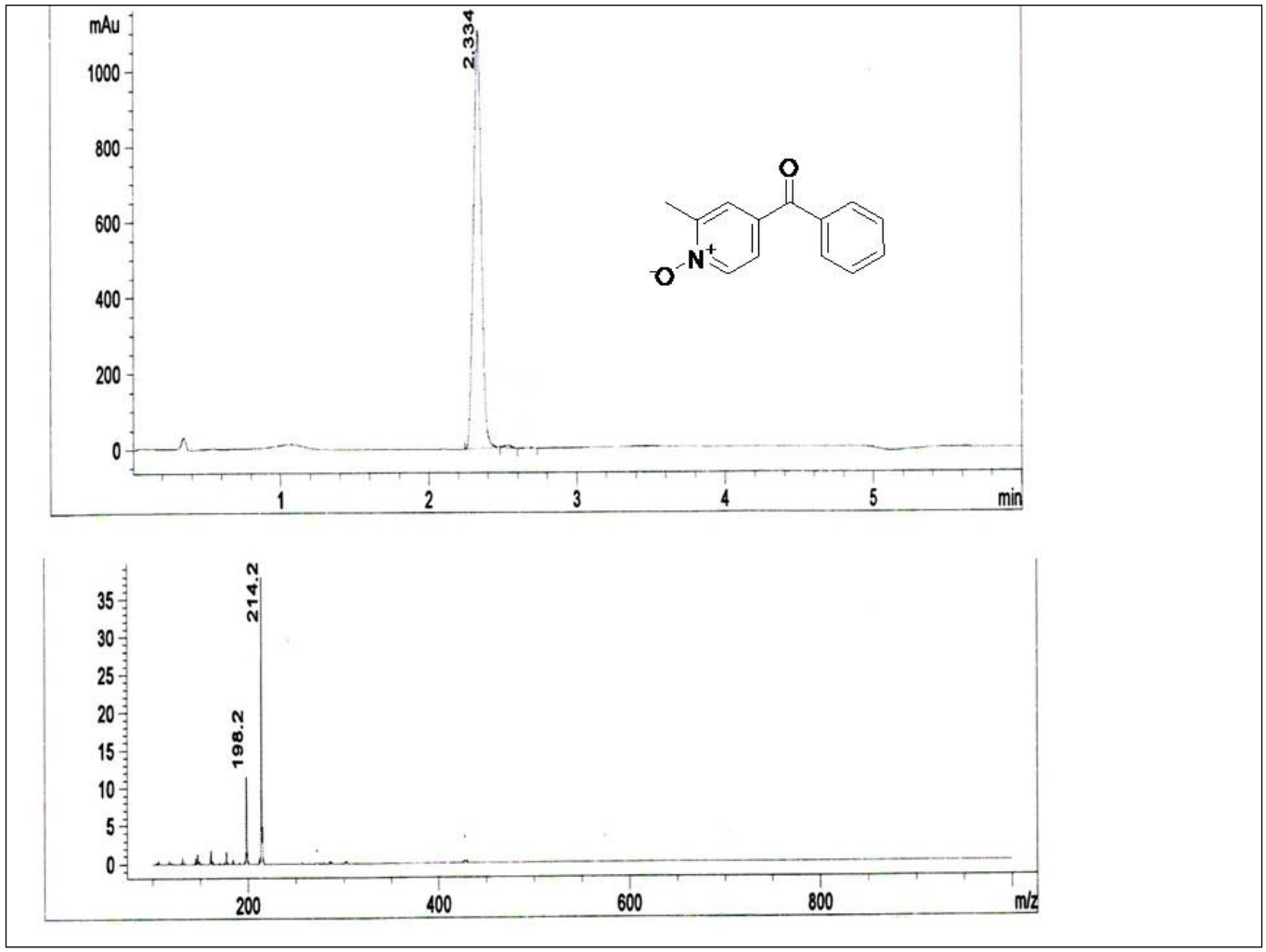


Fig 50. Mass spectrum of **3p'**

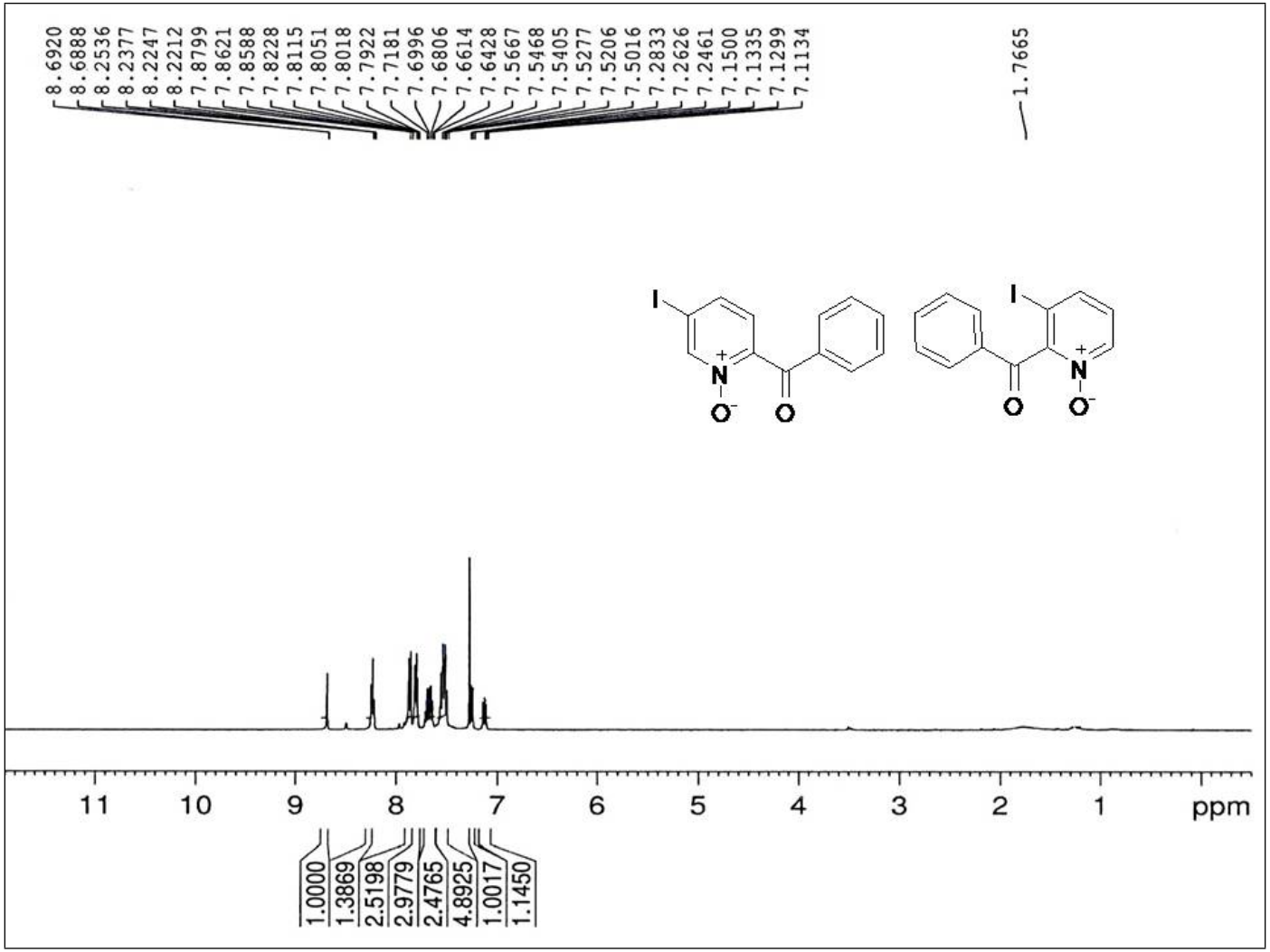


Fig 51. [1]H-NMR spectrum of **3q** & **3q'**

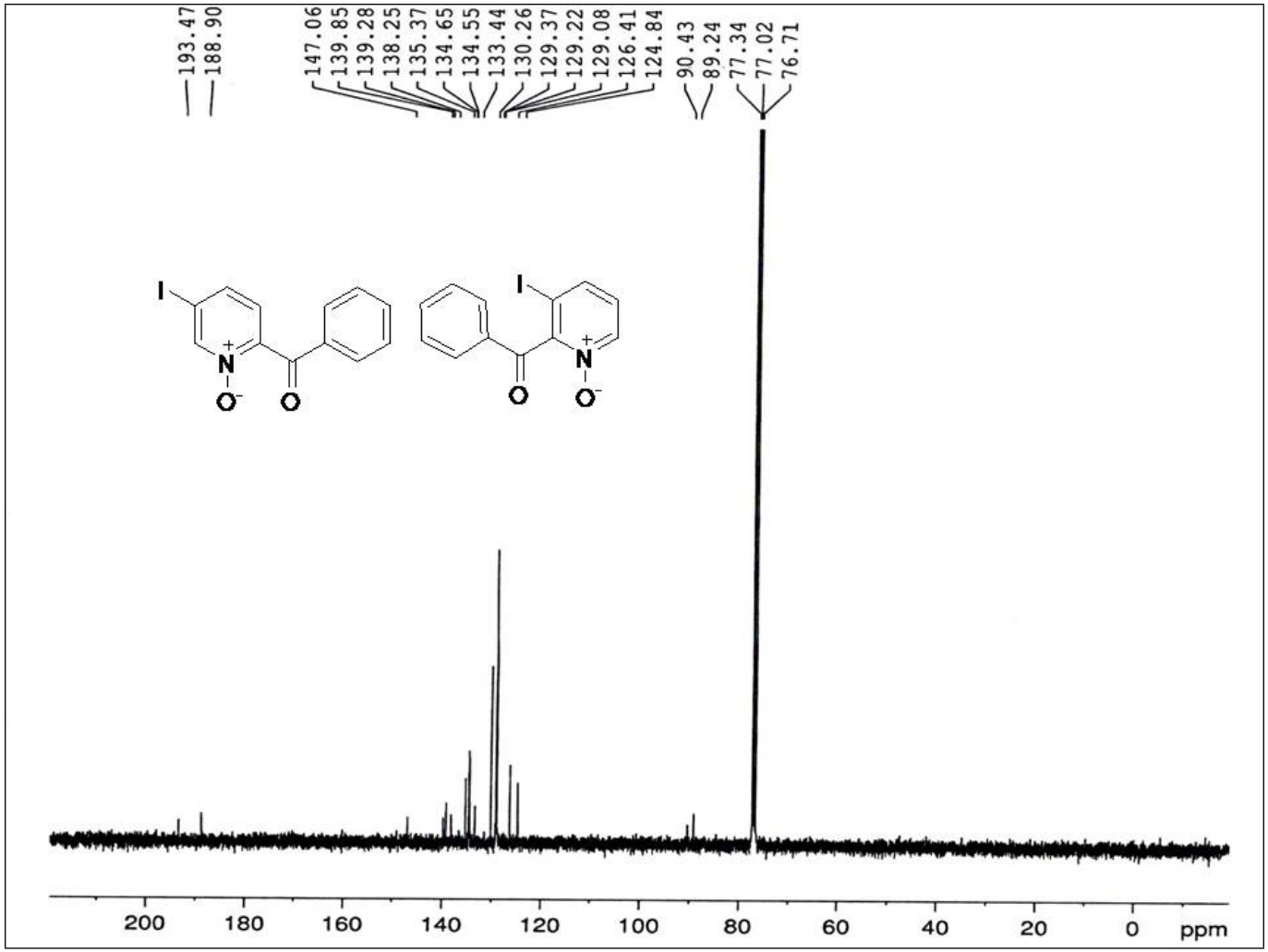


Fig 52. ^{13}C-NMR spectrum of **3q** & **3q'**

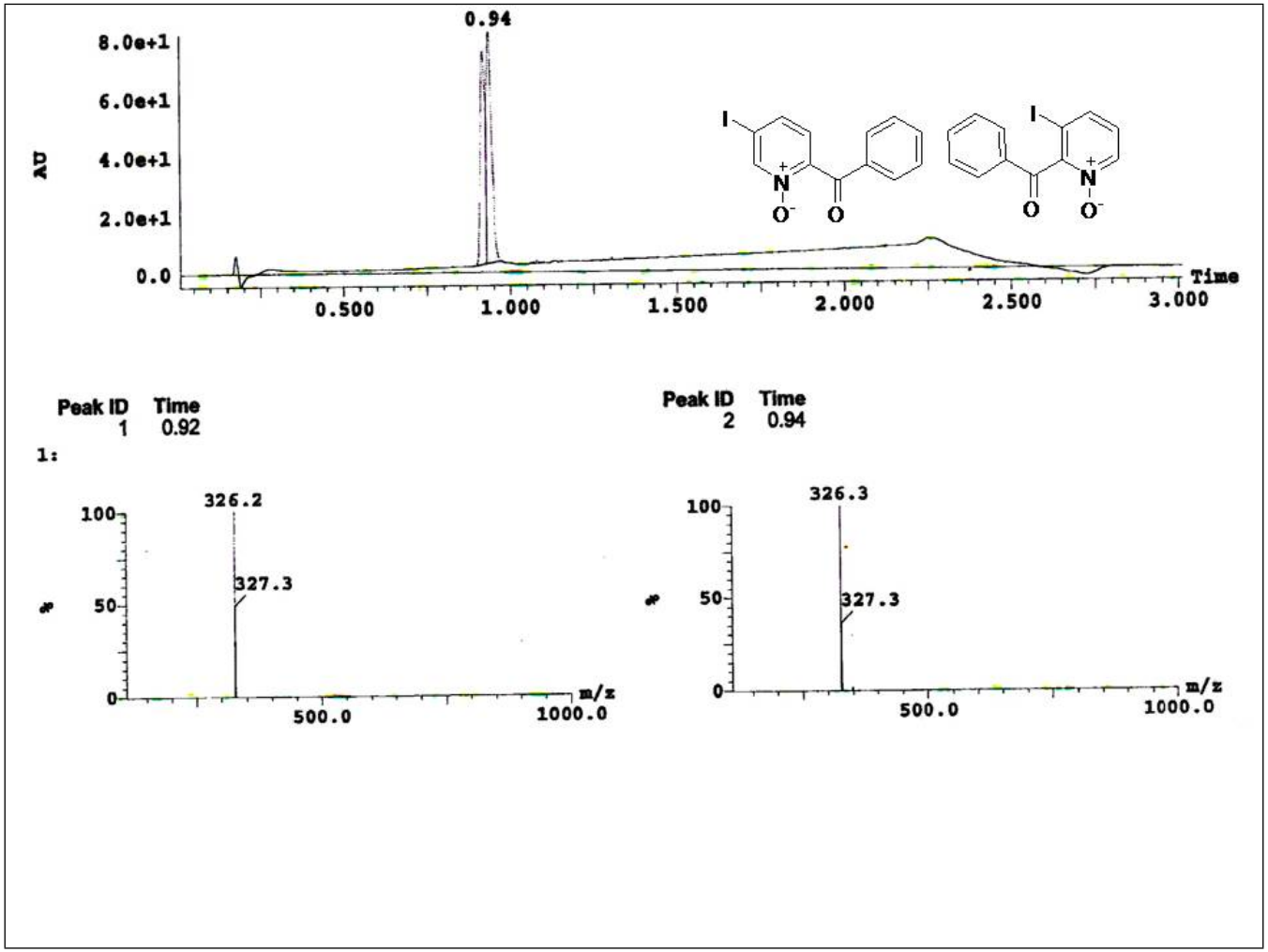


Fig 53. Mass spectrum of **3q** & **3q'**

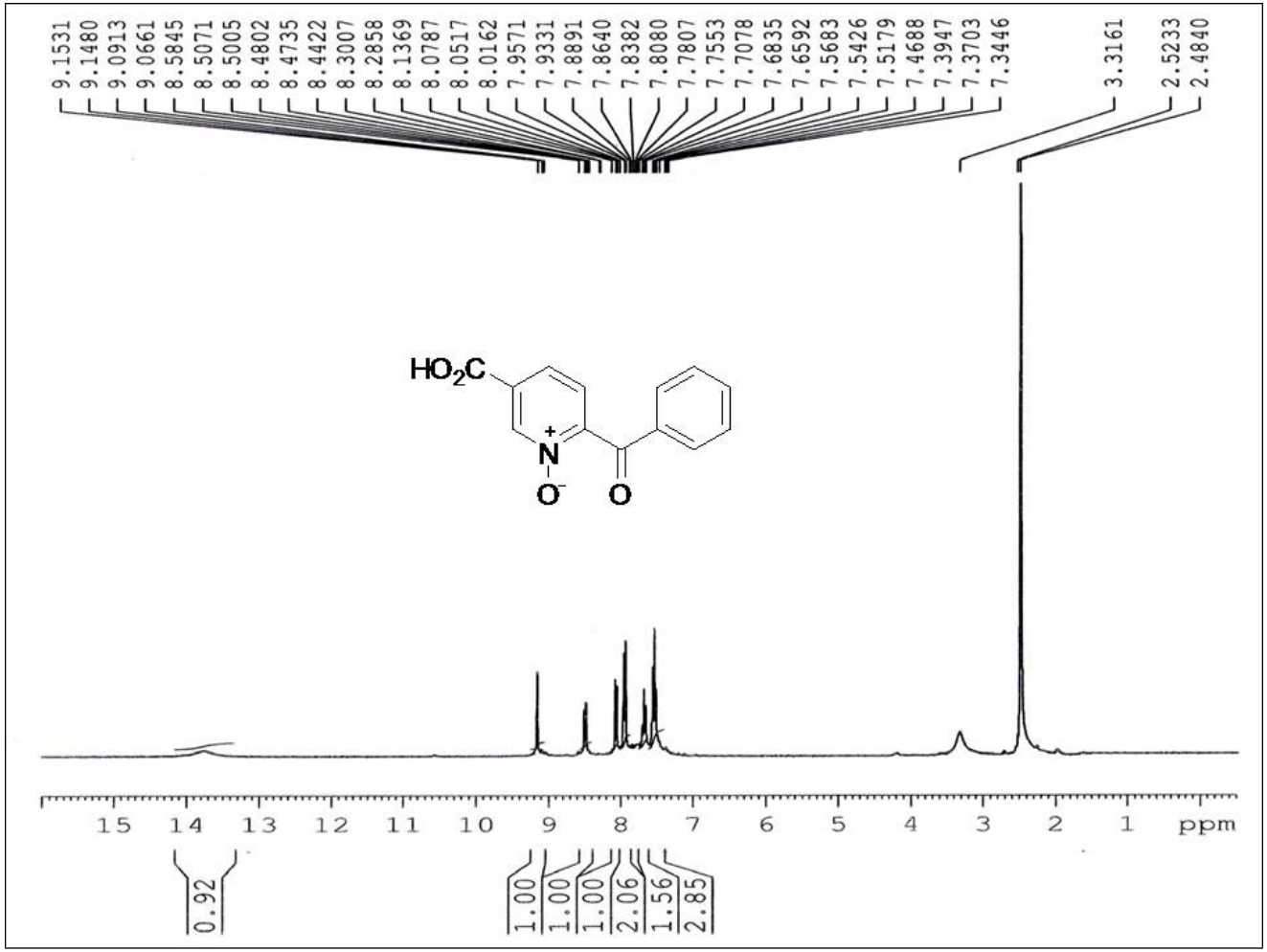


Fig 54. ^{1}H-NMR spectrum of **3r**

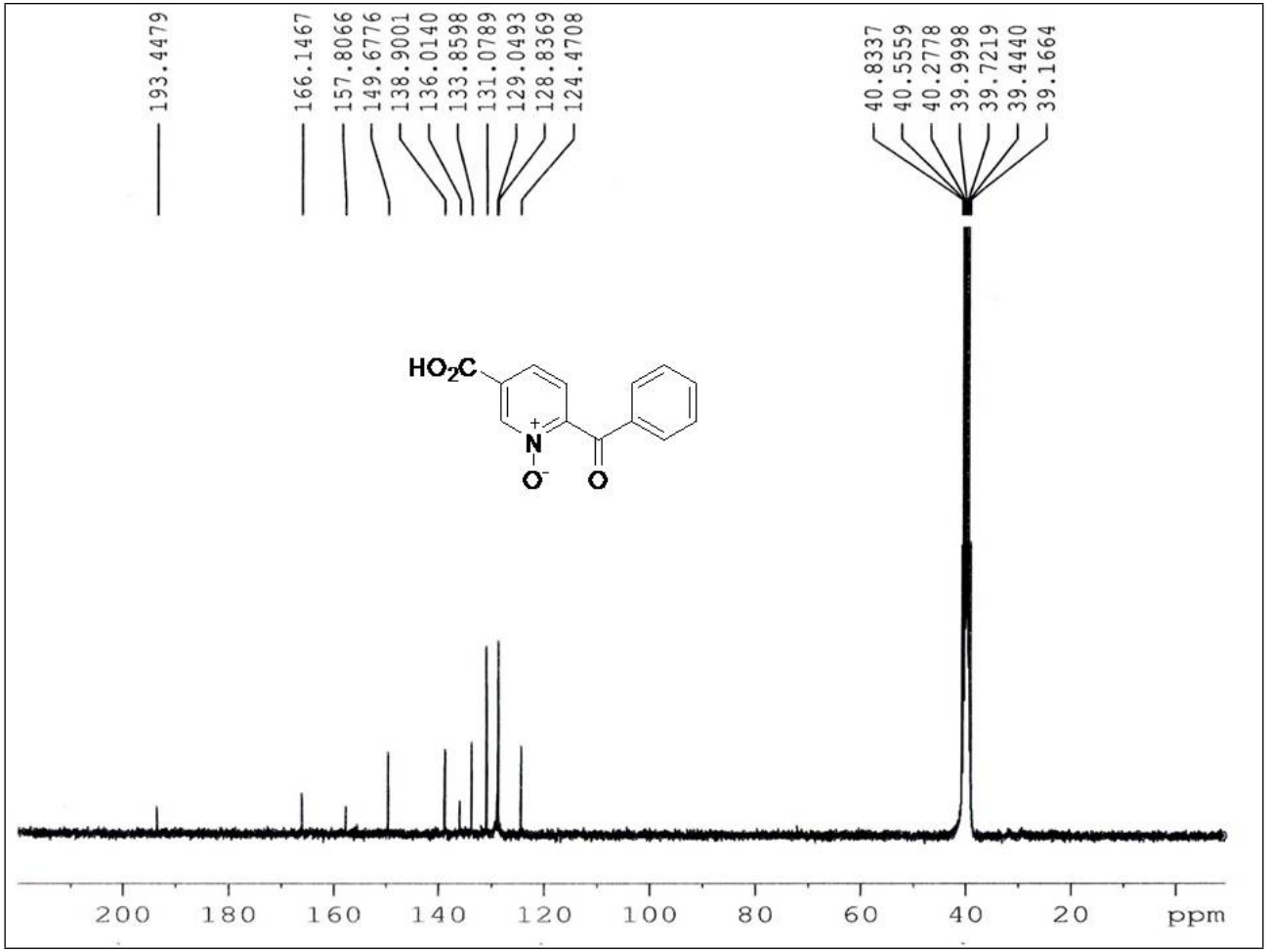


Fig 55. ^{13}C-NMR spectrum of **3r**

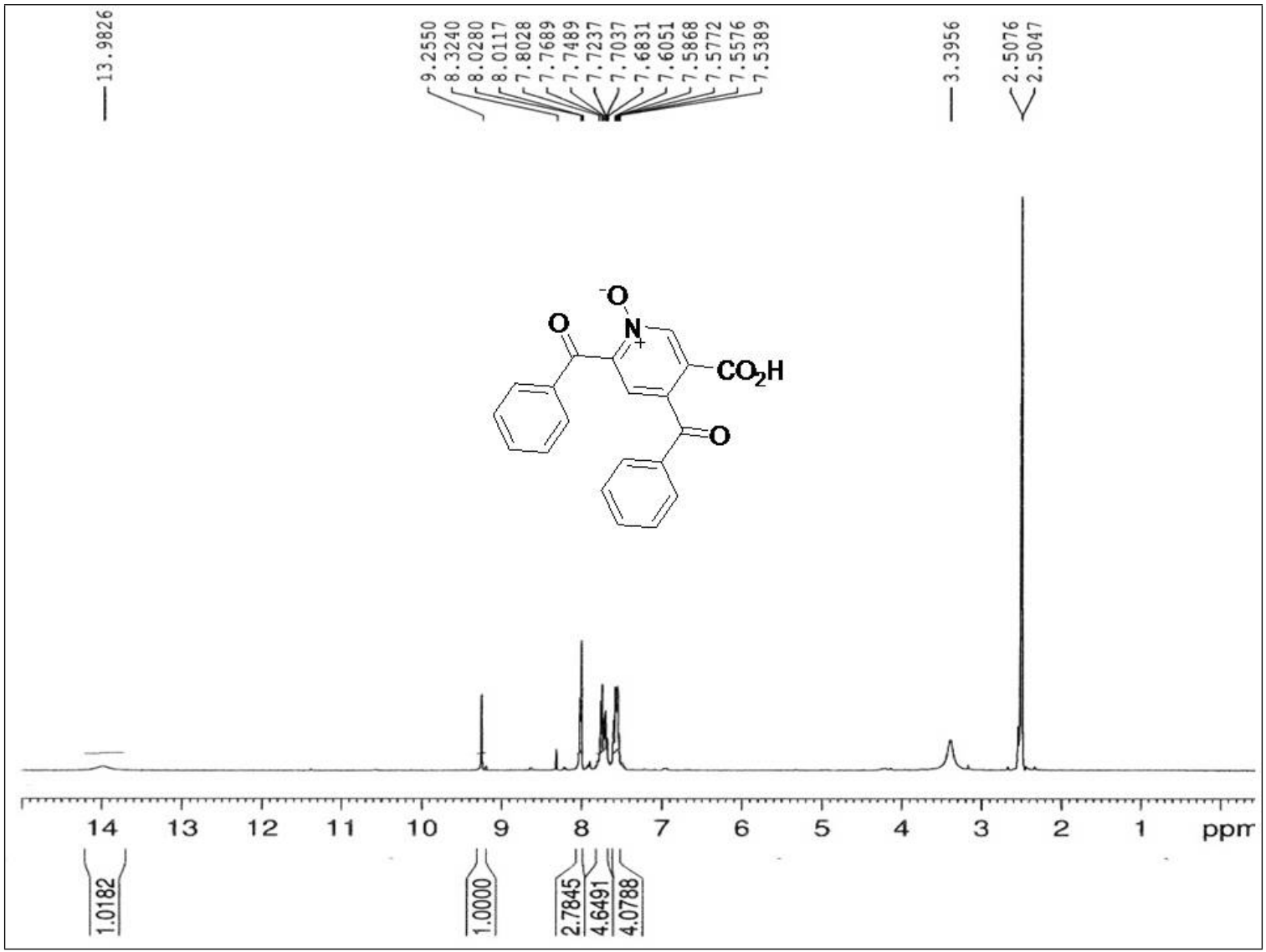


Fig 56. ^{1}H-NMR spectrum of **3r'**

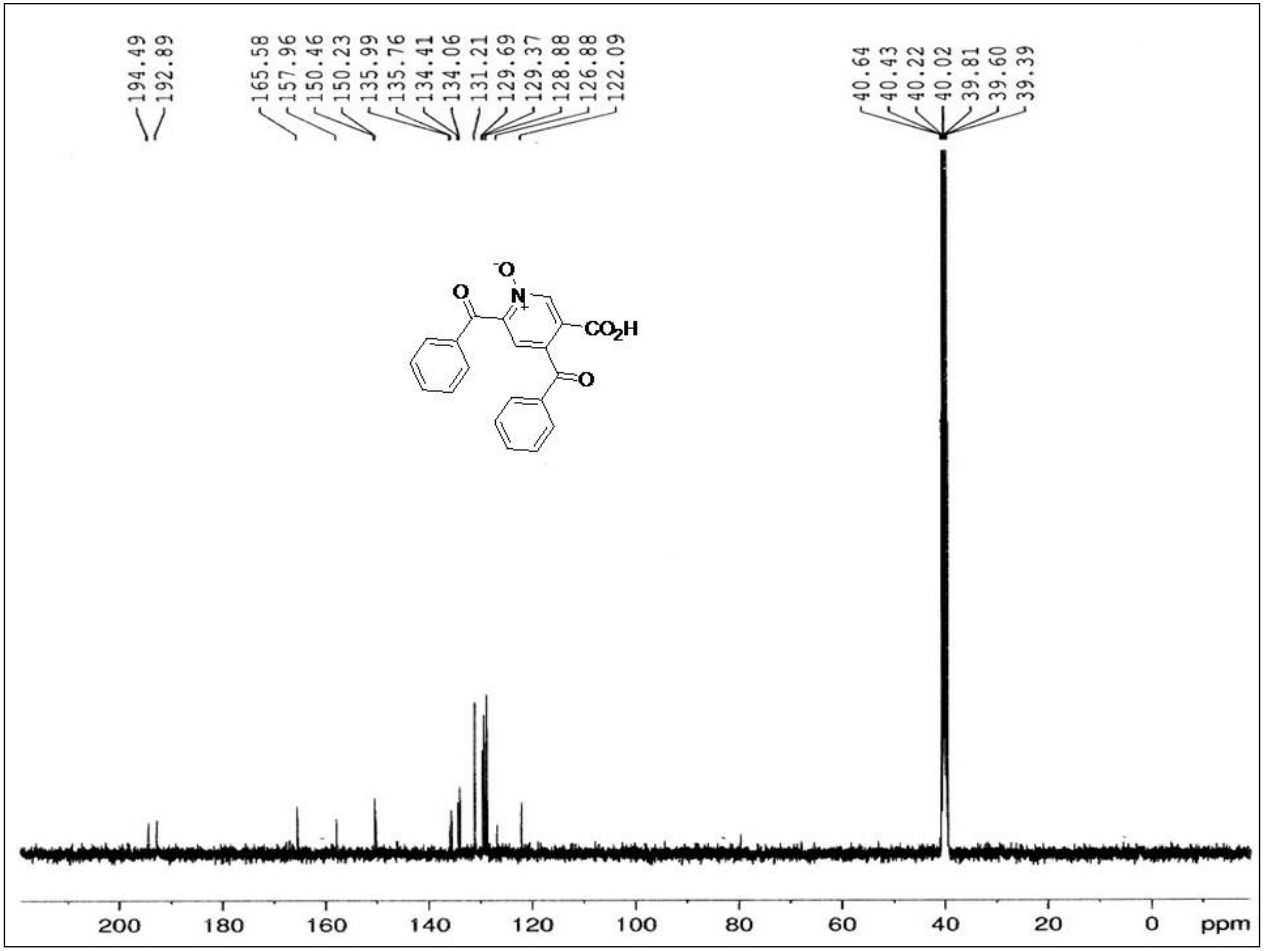


Fig 57. ^{13}C-NMR spectrum of **3r'**

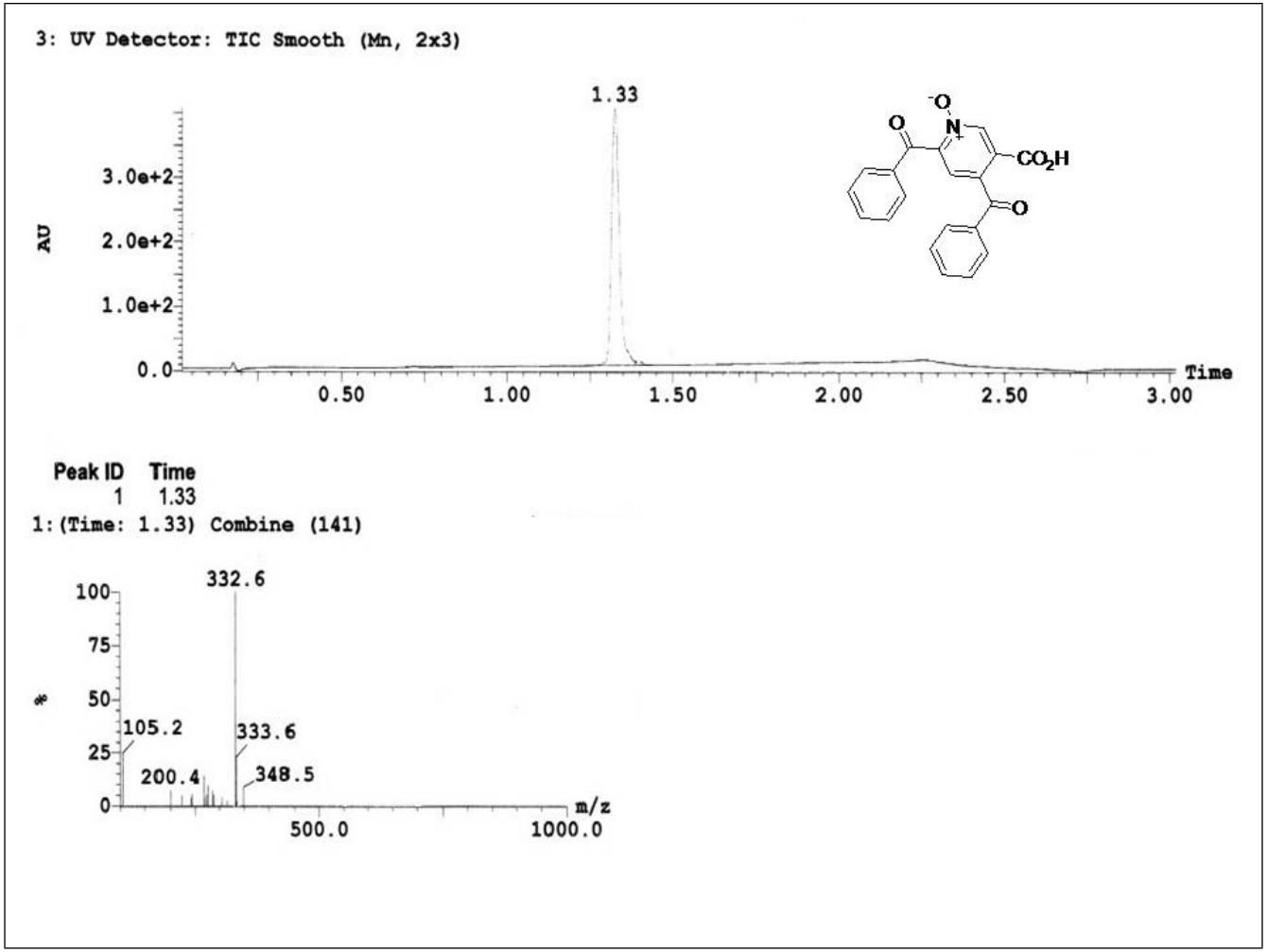


Fig 58. Mass spectrum of **3r'**

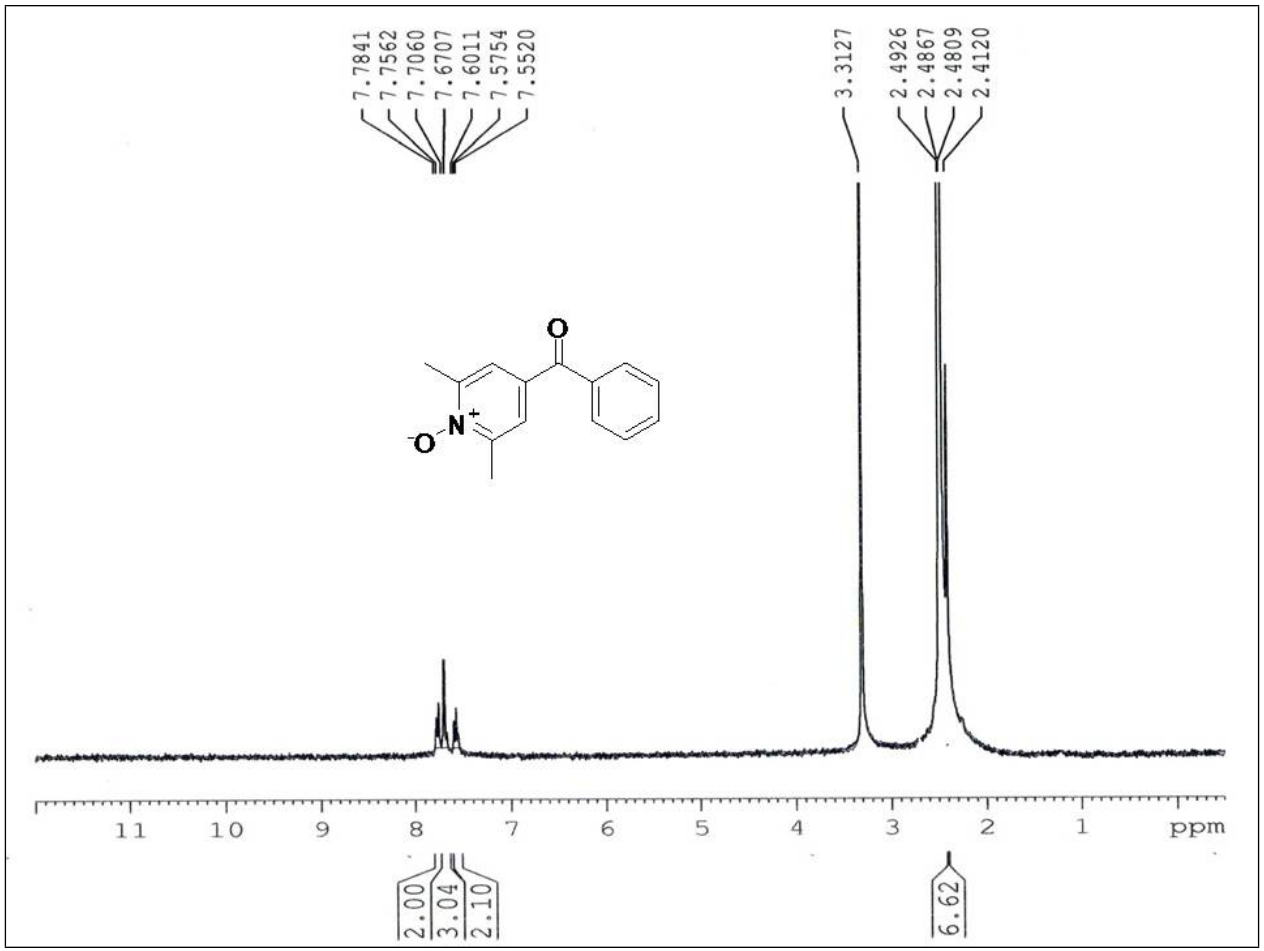


Fig 59. [1]H-NMR spectrum of **3s**

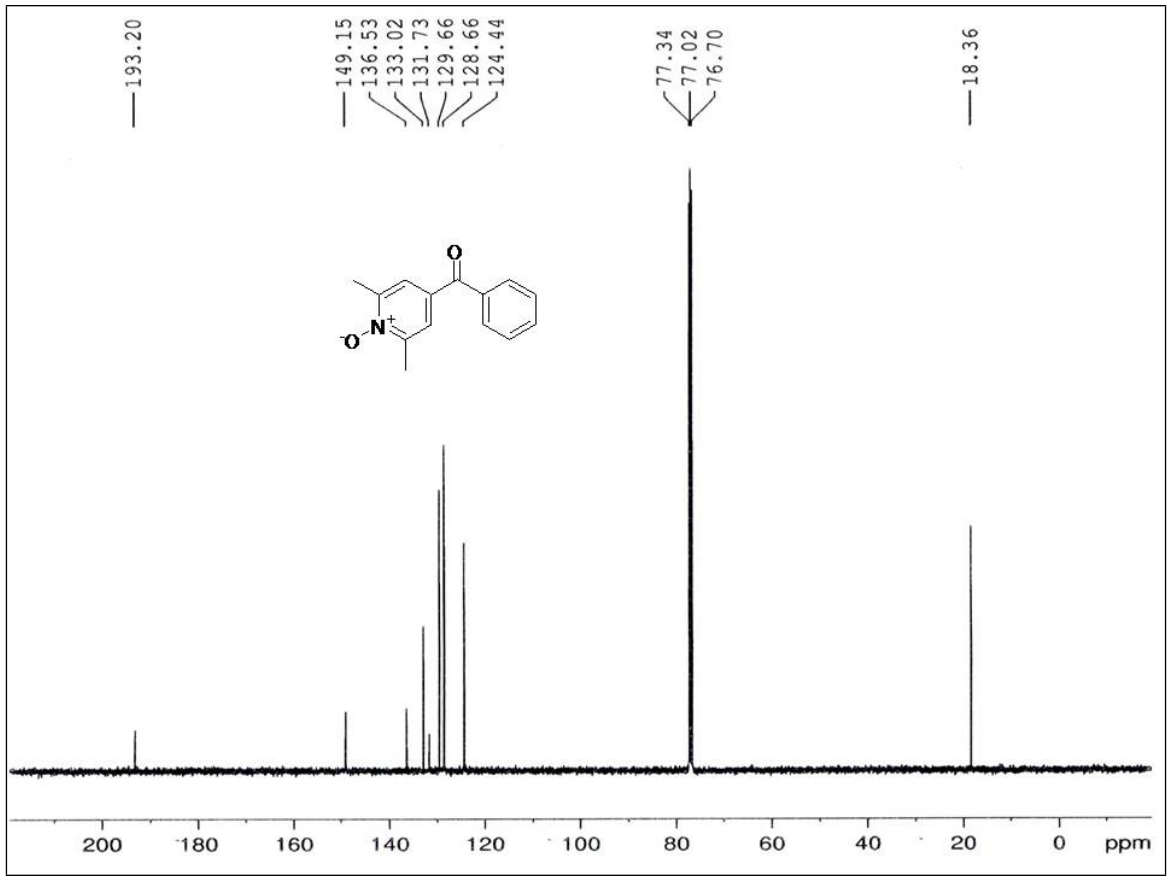


Fig 60. ^{13}C-NMR spectrum of **3s**

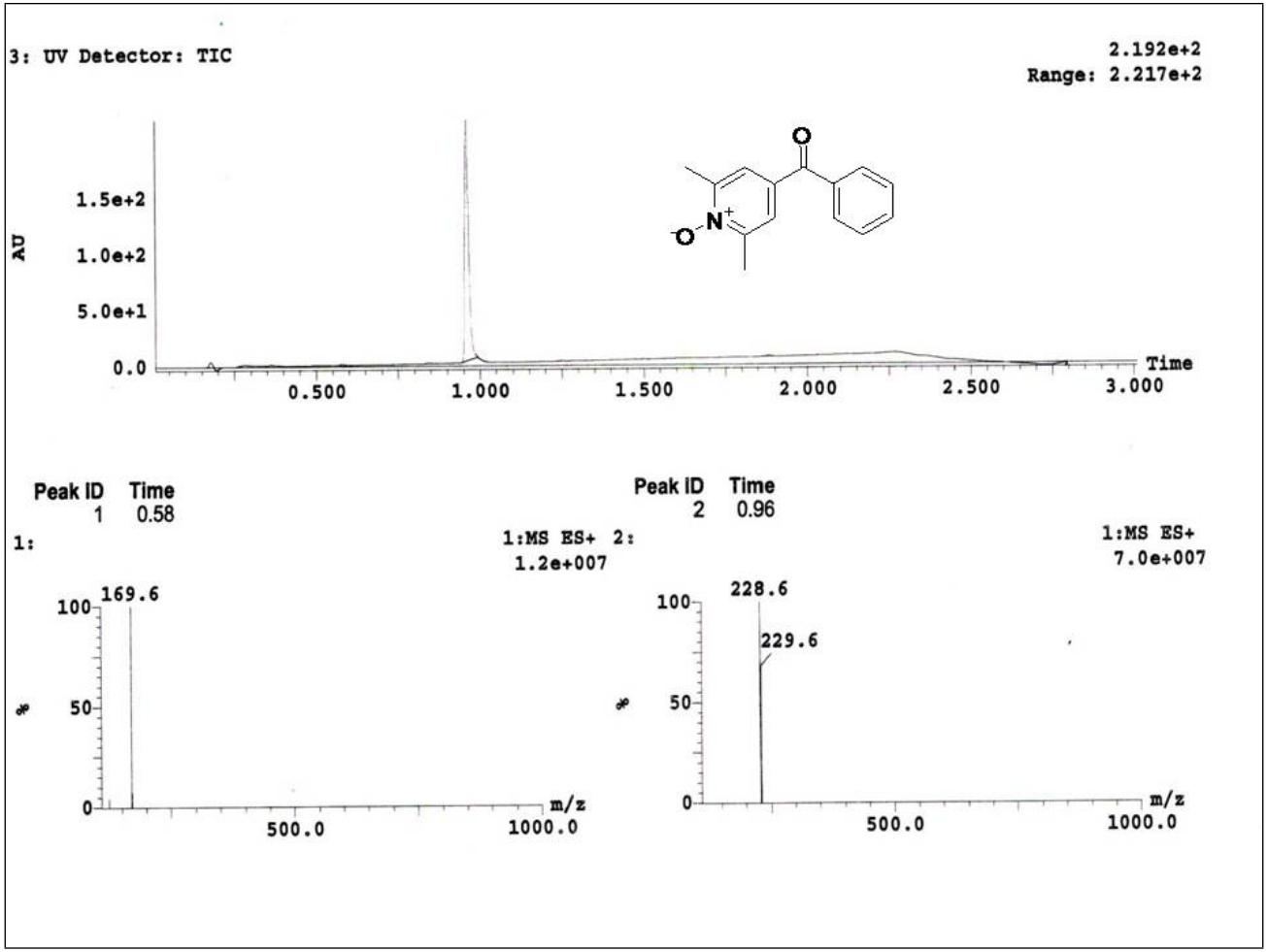


Fig 61. Mass spectrum of **3s**

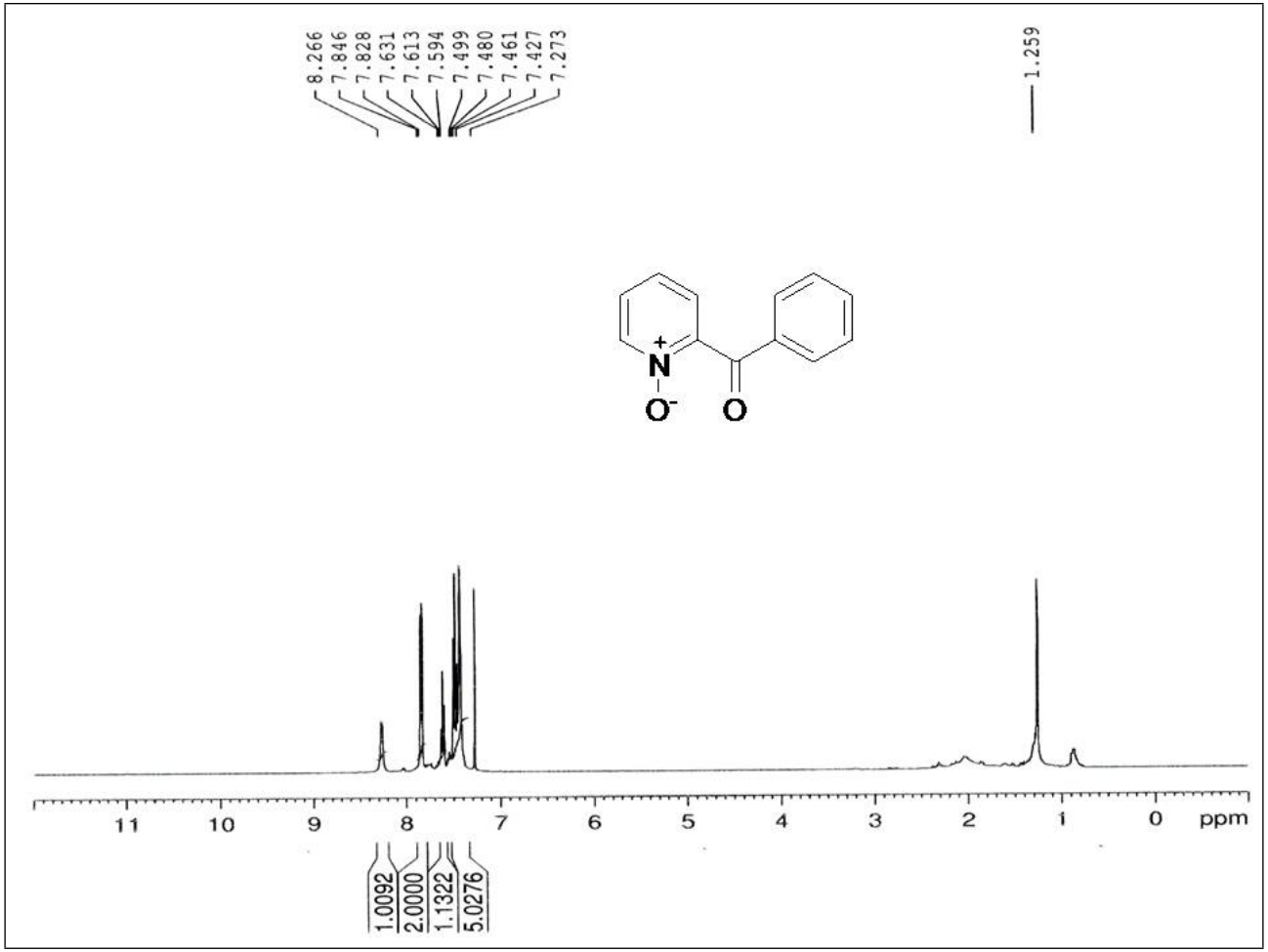


Fig 62. ^{1}H-NMR spectrum of **3t**

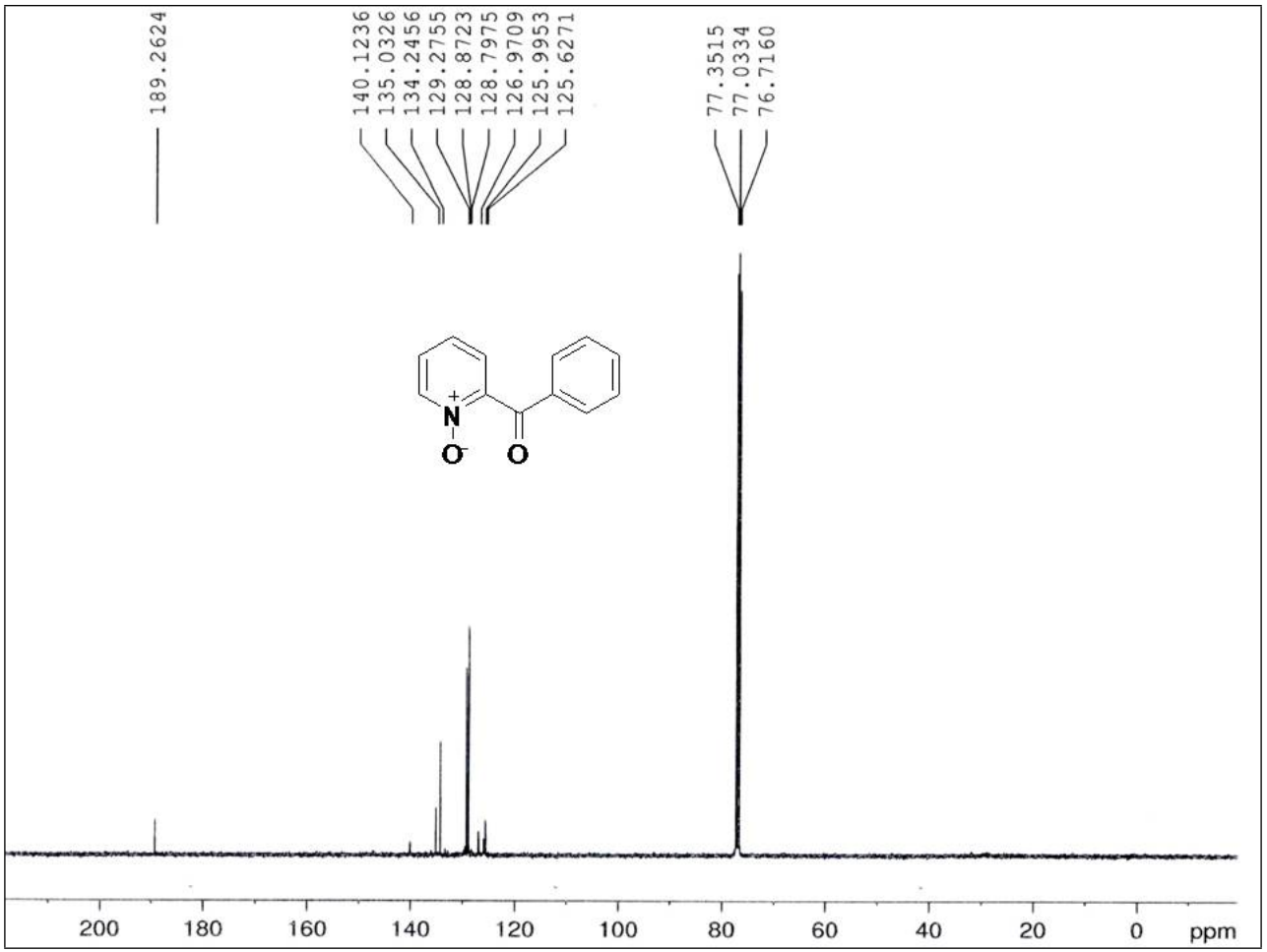


Fig 63. ^{13}C-NMR spectrum of **3t**

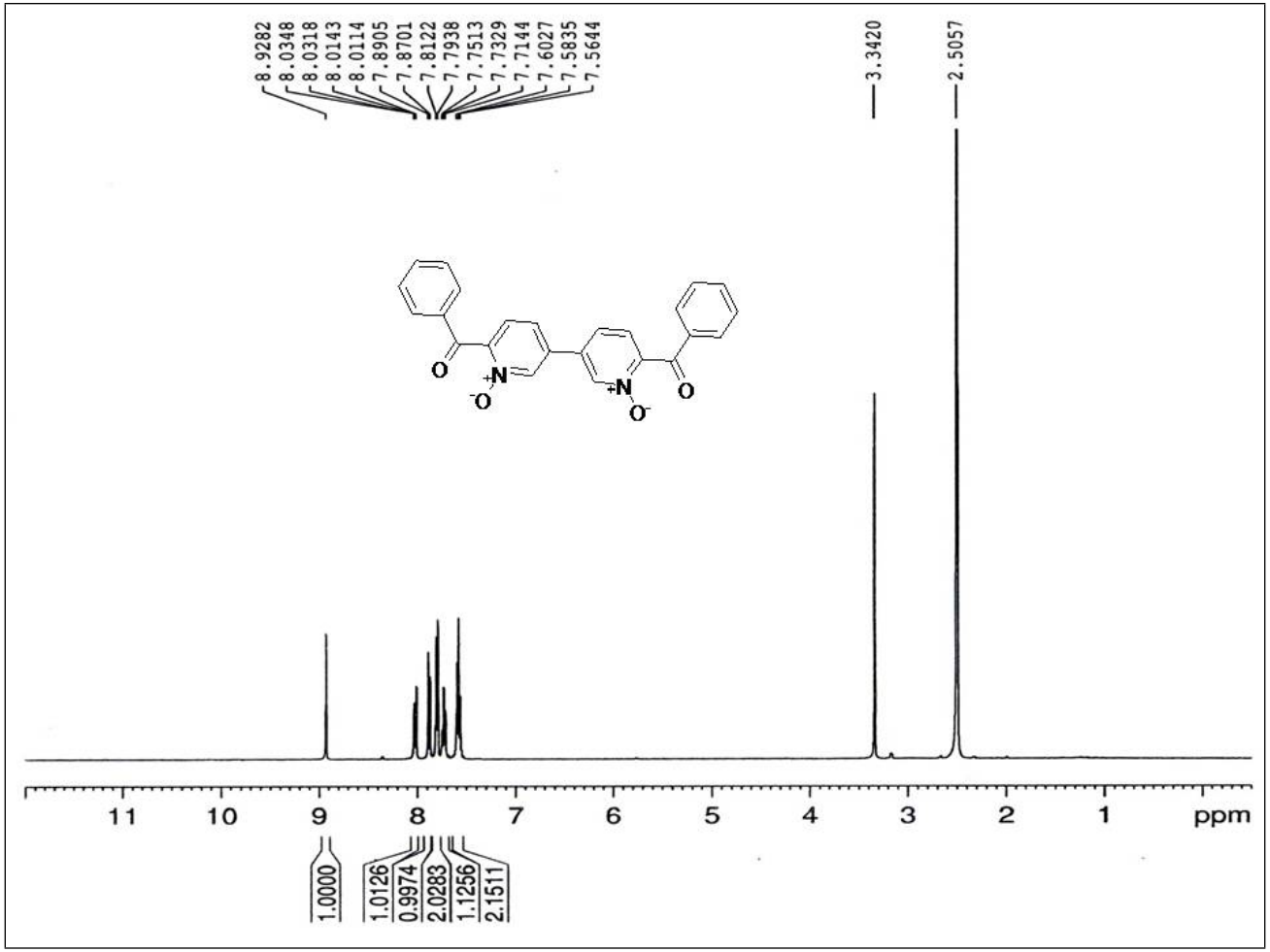


Fig 64. ^{1}H-NMR spectrum of **3t'**

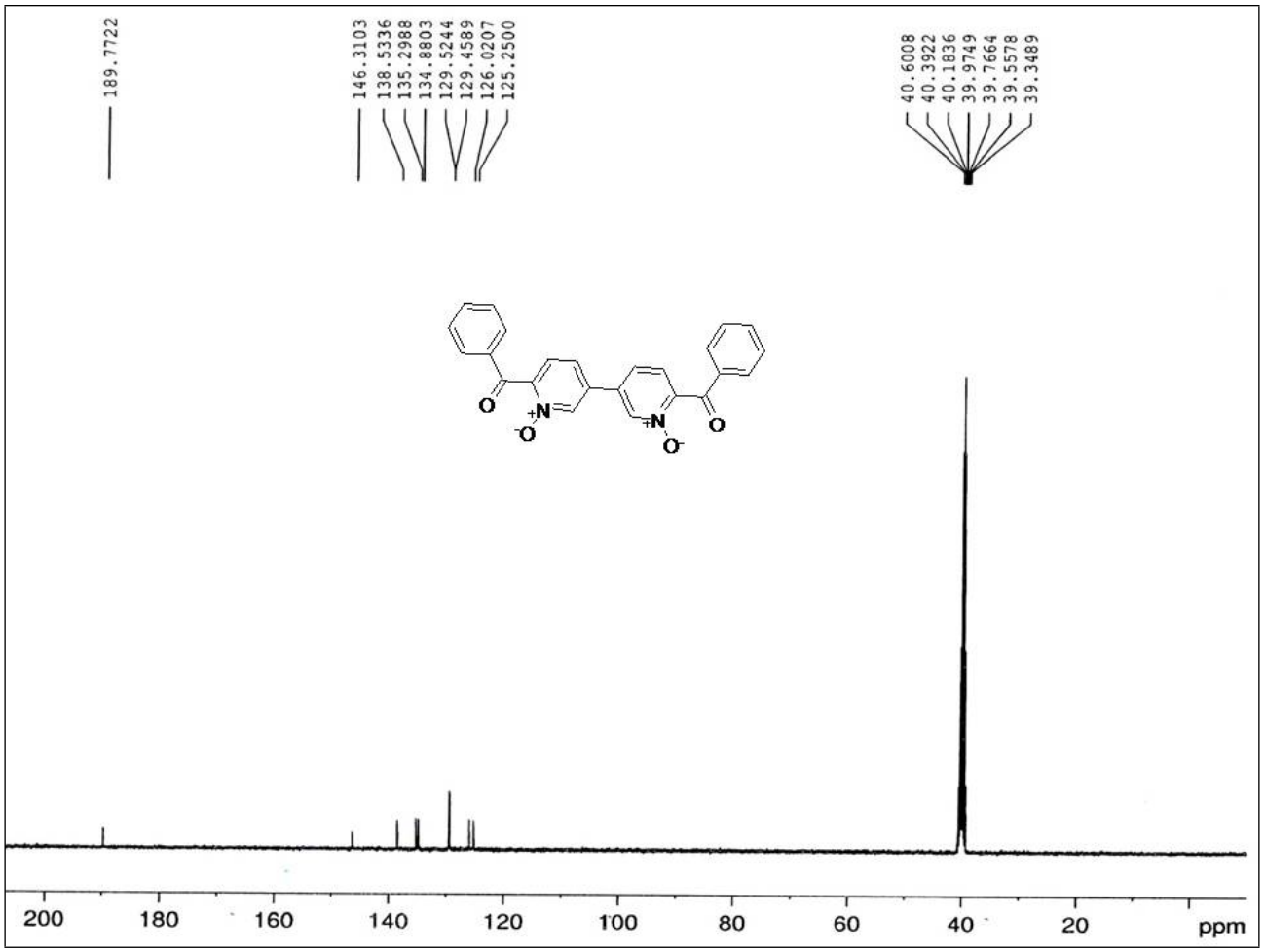


Fig 65. ^{13}C-NMR spectrum of **3t'**

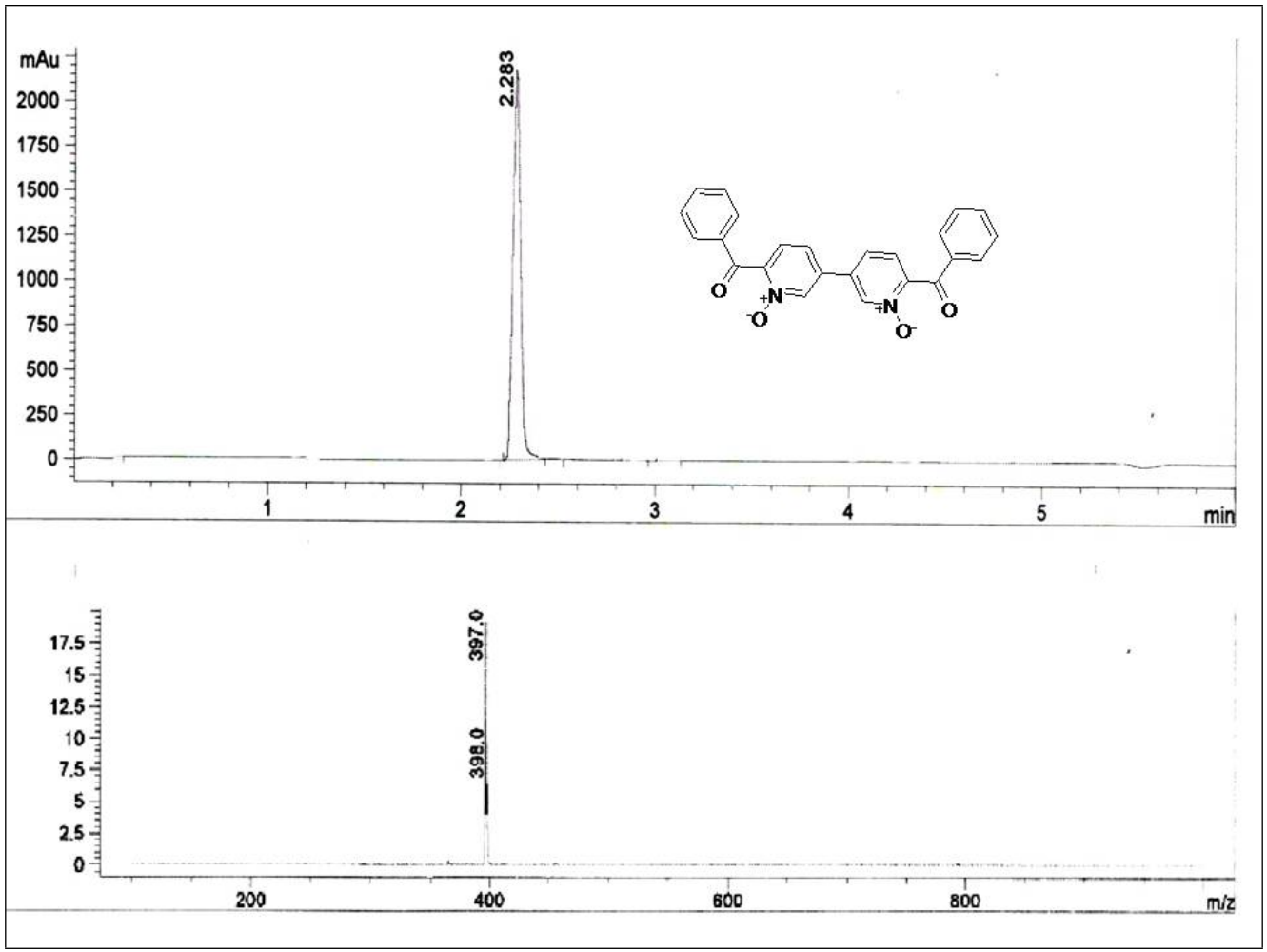


Fig 66. Mass spectrum of **3t'**

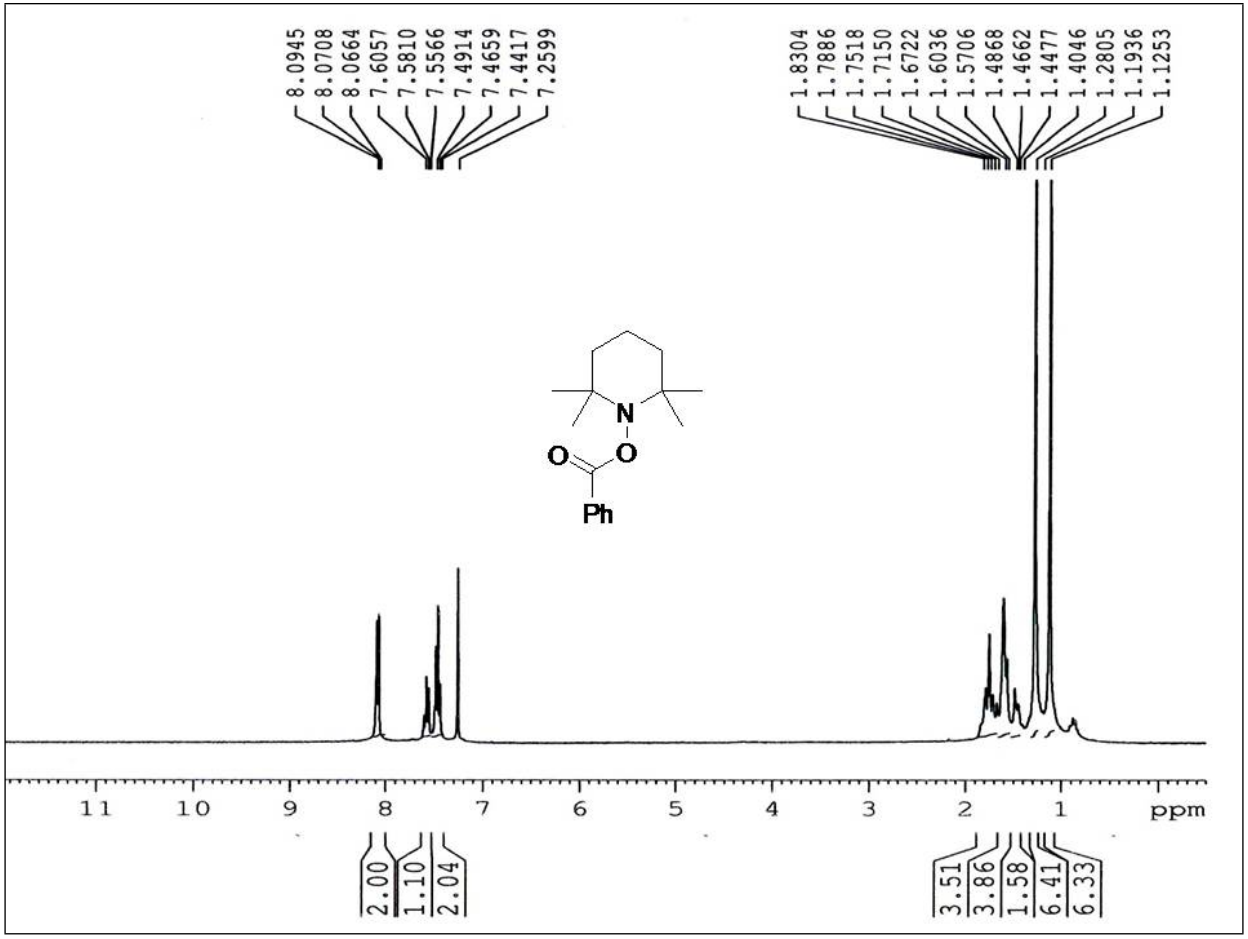


Fig 67. ^{1}H-NMR spectrum of **4**

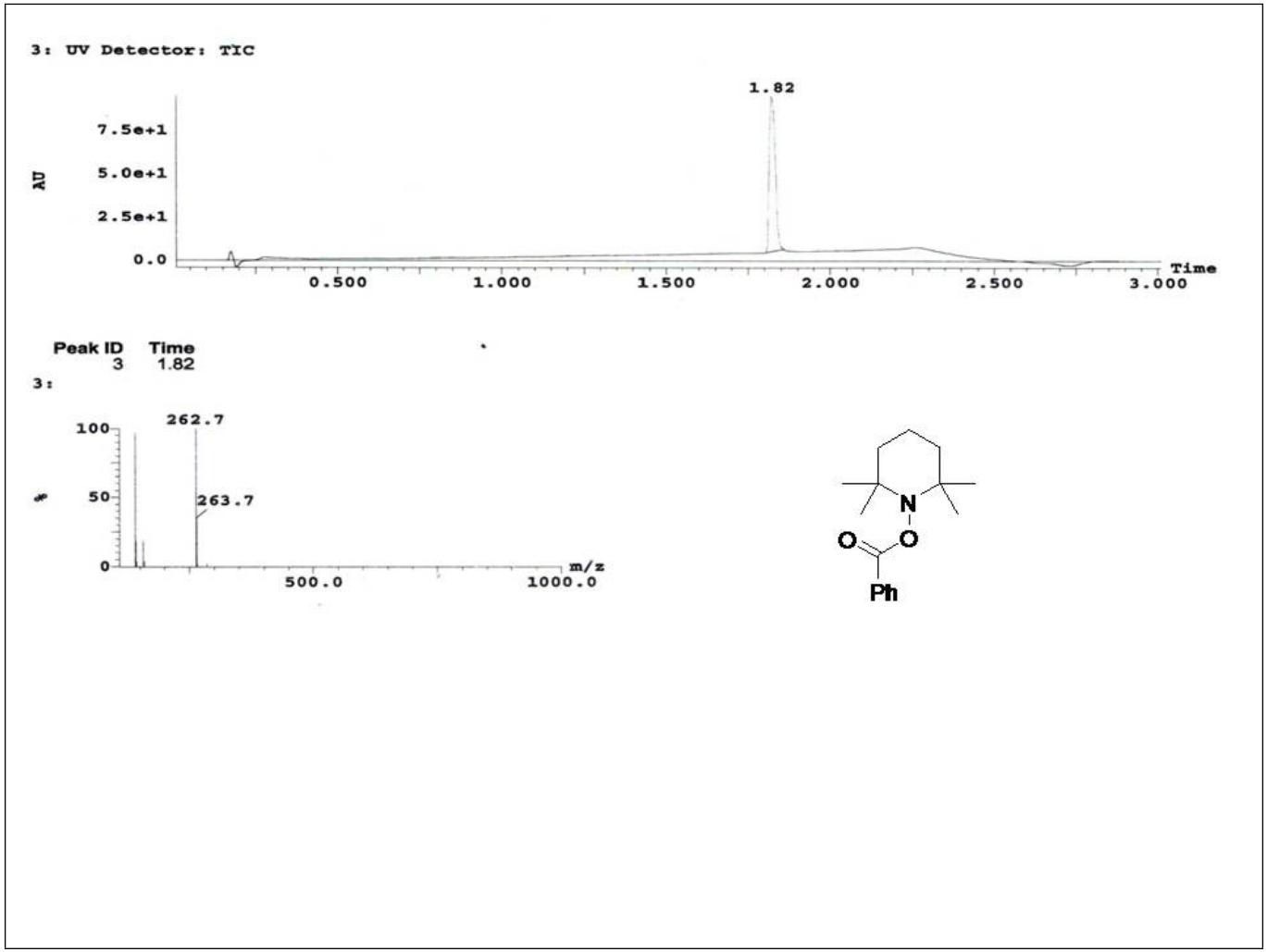


Fig 68. Mass spectrum of **4**