

# Supporting Information

## Diversity Synthesis of *N*-Substituted 2-Amino[1,6]naphthyridine Derivatives under Microwave Irradiation

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### Experimental

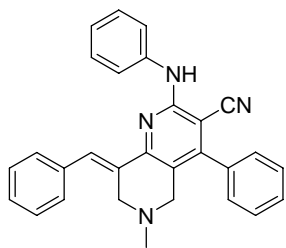
#### General

All reactions were performed in a monomodal Emrys<sup>TM</sup> Creator from Personal Chemistry, Uppsala, Sweden. Melting points were determined in open capillaries and are uncorrected. IR spectra were recorded on an FT-IR-tensor 27 spectrometer. <sup>1</sup>H NMR spectra were measured on a DPX 400 MHz spectrometer using TMS as an internal standard and DMSO-*d*<sub>6</sub> as solvent. HRMS (ESI) was determined by using the micrOTOF-Q II HPLC/MS instrument (BRUKER). X-Ray crystallographic analysis was performed with a Siemens SMART CCD and a Siemens P4 diffractometer.

#### General procedure for the synthesis of compounds **4** with microwave irradiation

**Preparation of compounds 4:** In a 10 mL reaction vial, the 3,5-dibenzylidenepiperidin-4-one **1** (1 mmol), malononitrile **2** (1 mmol), amine **3** (1.1 mmol) and HOAc (2 mL) were mixed and then capped. The mixture was irradiated at 200 W (initial power 150 W, maximum power 200 W) and at 110 °C for a give time. Upon completion, monitored by TLC, the reaction mixture was cooled to room temperature and then poured into cold water (70 mL), filtered to give the crude product, which was further purified by recrystallization from 95% EtOH to give pure product.

**8-Benzylidene-5,6,7,8-tetrahydro-6-methyl-4-phenyl-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4a)**

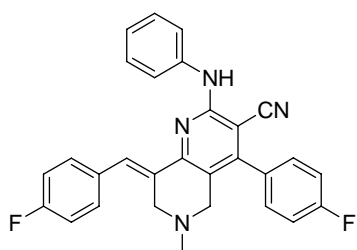


IR (KBr): 3341, 2209, 1597, 1549, 1512, 1440, 1361, 1292, 1261, 1185, 1114, 927, 774, 716, 690  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.04 (s, 1H, NH), 7.85 (s, 1H, =CH), 7.68 (d, 2H,  $J = 8.0$  Hz, ArH), 7.57 (m, 3H, ArH), 7.47-7.44 (m, 4H, ArH), 7.39-7.35 (m, 5H, ArH), 7.05 (t, 1H,  $J = 7.2$  Hz, ArH), 3.62 (s, 2H,  $\text{CH}_2$ ), 3.20 (s, 2H,  $\text{CH}_2$ ), 2.24 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 429.2080  $[\text{M}+\text{H}]^+$ , found: 429.2077.

**(4-Fluorobenzylidene)-4-(4-fluorophenyl)-5,6,7,8-tetrahydro-6-methyl-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4b)**

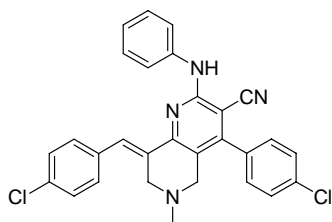


IR (KBr): 3317, 2217, 1601, 1551, 1506, 1442, 1364, 1262, 1230, 1159, 1110, 925, 915, 900, 843, 758, 707, 692  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.06 (s, 1H, NH), 7.81 (s, 1H, =CH), 7.67 (d, 2H,  $J = 8.0$  Hz, ArH), 7.54-7.50 (m, 2H, ArH), 7.44-7.35 (m, 6H, ArH), 7.29 (t, 2H,  $J = 8.4$  Hz, ArH), 7.05 (t, 1H,  $J = 7.2$  Hz, ArH), 3.59 (s, 2H,  $\text{CH}_2$ ), 3.20 (s, 2H,  $\text{CH}_2$ ), 2.25 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 465.1891  $[\text{M}+\text{H}]^+$ , found: 465.1871.

**8-(4-Chlorobenzylidene)-4-(4-chlorophenyl)-5,6,7,8-tetrahydro-6-methyl-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4c)**

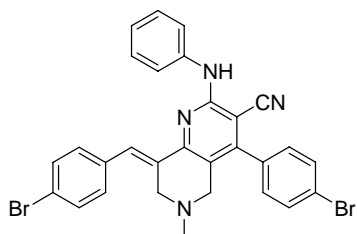


IR (KBr): 3329, 2215, 1601, 1547, 1493, 1441, 1364, 1258, 1183, 1119, 1091, 1013, 913, 838, 745, 690  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.09 (s, 1H, NH), 7.79 (s, 1H, =CH), 7.68-7.64 (m, 4H, ArH), 7.50 (t, 4H,  $J = 6.8$  Hz, ArH), 7.38-7.35 (m, 4H, ArH), 7.05 (t, 1H,  $J = 7.2$  Hz, ArH), 3.59 (s, 2H,  $\text{CH}_2$ ), 3.20 (s, 2H,  $\text{CH}_2$ ), 2.25 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 497.1300  $[\text{M}+\text{H}]^+$ , found: 497.1286.

**8-(4-Bromobenzylidene)-4-(4-bromophenyl)-5,6,7,8-tetrahydro-6-methyl-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4d)**

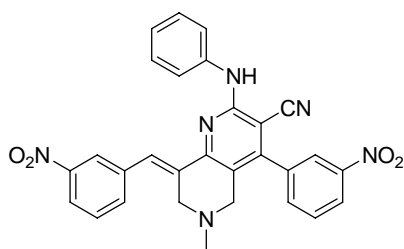


IR (KBr): 3332, 2213, 1599, 1567, 1547, 1494, 1440, 1363, 1257, 1183, 1119, 1073, 1010, 912, 823, 742, 689  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.09 (s, 1H, NH), 7.80 (s, 1H, =CH), 7.78-7.76 (m, 2H, ArH), 7.65 (t, 4H,  $J = 8.4$  Hz, ArH), 7.42 (d, 2H,  $J = 8.0$  Hz, ArH), 7.36 (t, 2H,  $J = 8.0$  Hz, ArH), 7.30 (d, 2H,  $J = 8.4$  Hz, ArH), 7.05 (t, 1H,  $J = 7.2$  Hz, ArH), 3.58 (s, 2H,  $\text{CH}_2$ ), 3.19 (s, 2H,  $\text{CH}_2$ ), 2.25 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 585.0289  $[\text{M}+\text{H}]^+$ , found: 585.0283.

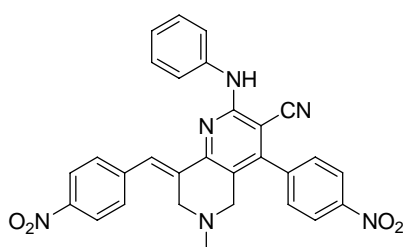
**8-(3-Nitrobenzylidene)-5,6,7,8-tetrahydro-6-methyl-4-(3-nitrophenyl)-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4e)**

**aphthyridine-3-carbonitrile (4e)**

IR (KBr): 3313, 2210, 1607, 1569, 1553, 1495, 1440, 1349, 1254, 1099, 1025, 917, 811, 751, 723, 690  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.18 (s, 1H, NH), 8.41 (d, 1H,  $J = 8.0$  Hz, ArH), 8.37 (s, 1H, ArH), 8.20 (d, 1H,  $J = 8.4$  Hz, ArH), 8.12 (s, 1H, =CH), 7.97 (d, 1H,  $J = 7.6$  Hz, ArH), 7.92-7.89 (m, 2H, ArH), 7.81 (d, 1H,  $J = 7.6$  Hz, ArH), 7.76-7.68 (m, 3H, ArH), 7.38 (t, 2H,  $J = 7.6$  Hz, ArH), 7.07 (t, 1H,  $J = 7.2$  Hz, ArH), 3.63 (s, 2H,  $\text{CH}_2$ ), 3.28-3.19 (m, 2H,  $\text{CH}_2$ ), 2.25 (s, 3H,  $\text{CH}_3$ ).

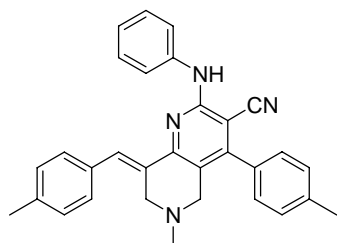
HRMS (ESI):  $m/z$  calcd for: 519.1781  $[\text{M}+\text{H}]^+$ , found: 519.1763.

**8-(4-Nitrobenzylidene)-5,6,7,8-tetrahydro-6-methyl-4-(4-nitrophenyl)-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4f)**

IR (KBr): 3339, 2212, 1598, 1557, 1519, 1442, 1344, 1258, 1181, 1110, 853, 754, 721, 693  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.20 (s, 1H, NH), 8.43 (d, 2H,  $J = 8.8$  Hz, ArH), 8.28 (d, 2H,  $J = 8.8$  Hz, ArH), 7.89 (s, 1H, =CH), 7.80 (d, 2H,  $J = 8.8$  Hz, ArH), 7.68 (d, 2H,  $J = 8.0$  Hz, ArH), 7.63 (d, 2H,  $J = 8.4$  Hz, ArH), 7.39 (t, 2H,  $J = 8.0$  Hz, ArH), 7.09 (t, 1H,  $J = 7.2$  Hz, ArH), 3.64 (s, 2H,  $\text{CH}_2$ ), 3.21 (s, 2H,  $\text{CH}_2$ ), 2.25 (s, 3H,  $\text{CH}_3$ ).

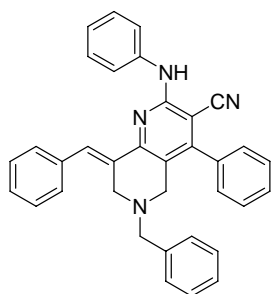
HRMS (ESI):  $m/z$  calcd for: 519.1781  $[\text{M}+\text{H}]^+$ , found: 519.1801.

**8-(4-Methylbenzylidene)-4-(4-methylphenyl)-5,6,7,8-tetrahydro-6-methyl-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4g)**

IR (KBr): 3331, 2209, 1599, 1565, 1542, 1497, 1438, 1360, 1262, 1172, 1116, 1054, 925, 811, 756, 739, 690  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.98 (s, 1H, NH), 7.81 (s, 1H, =CH), 7.67 (d, 2H,  $J = 8.0$  Hz, ArH), 7.38-7.36 (m, 4H, ArH), 7.32 (t, 2H,  $J = 7.6$  Hz, ArH), 7.25-7.23 (m, 4H, ArH), 7.05 (t, 1H,  $J = 7.2$  Hz, ArH), 3.60 (s, 2H,  $\text{CH}_2$ ), 3.20 (s, 2H,  $\text{CH}_2$ ), 2.41 (s, 3H,  $\text{CH}_3$ ), 2.34 (s, 3H,  $\text{CH}_3$ ), 2.24 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 457.2392  $[\text{M}+\text{H}]^+$ , found: 457.2392.

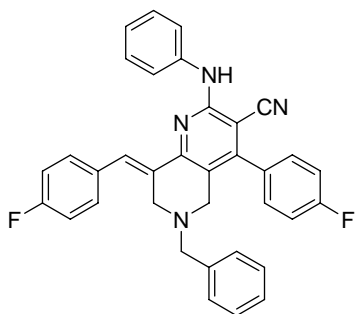
**6-Benzyl-8-benzylidene-5,6,7,8-tetrahydro-4-phenyl-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4h)**

IR (KBr): 3345, 2211, 1598, 1549, 1495, 1439, 1362, 1260, 1213, 1182, 1100, 1072, 1024, 980, 925, 898, 771, 756, 745, 695  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.02 (s, 1H, NH), 7.87 (s, 1H, =CH), 7.70 (d, 2H,  $J = 8.0$  Hz, ArH), 7.51-7.49 (m, 3H, ArH), 7.40-7.35 (m, 6H, ArH), 7.31 (d, 1H,  $J = 7.2$  Hz, ArH), 7.28 (d, 2H,  $J = 7.6$  Hz, ArH), 7.21-7.13 (m, 5H, ArH), 7.06 (t, 1H,  $J = 7.2$  Hz, ArH), 3.74 (s, 2H,  $\text{CH}_2$ ), 3.52 (s, 2H,  $\text{CH}_2$ ), 3.33 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 505.2392  $[\text{M}+\text{H}]^+$ , found: 505.2399.

**8-(4-Fluorobenzylidene)-6-benzyl-4-(4-fluorophenyl)-5,6,7,8-tetrahydro-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4i)**

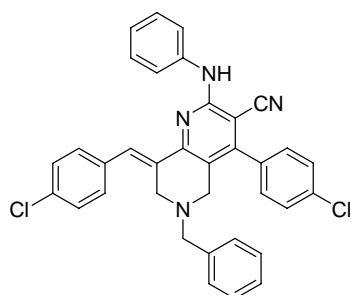


IR (KBr): 3346, 2210, 1598, 1564, 1505, 1439, 1362, 1292, 1230, 1159, 1104, 929, 900, 834, 808, 788, 751, 701  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.05 (s, 1H, NH), 7.82 (s, 1H, =CH), 7.70-7.67 (d, 2H,  $J = 8.0$  Hz, ArH), 7.47-7.43 (m, 2H, ArH), 7.38 (d, 2H,  $J = 7.2$  Hz, ArH), 7.35-7.31 (m, 4H, ArH), 7.23 (d, 2H,  $J = 8.8$  Hz, ArH), 7.20-7.17 (m, 3H, ArH), 7.15-7.13 (m, 2H, ArH), 7.06 (t, 1H,  $J = 7.2$  Hz, ArH), 3.71 (s, 2H,  $\text{CH}_2$ ), 3.55 (s, 2H,  $\text{CH}_2$ ), 3.33 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 541.2204  $[\text{M}+\text{H}]^+$ , found: 541.2206.

**8-(4-Chlorobenzylidene)-6-benzyl-4-(4-chlorophenyl)-5,6,7,8-tetrahydro-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4j)**

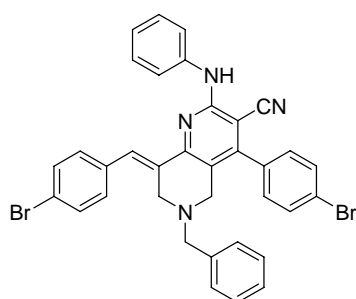


IR (KBr): 3360, 2209, 1597, 1561, 1545, 1492, 1439, 1363, 1258, 1212, 1182, 1092, 1013, 835, 816, 754, 700  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.07 (s, 1H, NH), 7.80 (s, 1H, =CH), 7.68 (d, 2H,  $J = 8.0$  Hz, ArH), 7.58 (d, 2H,  $J = 8.0$  Hz, ArH), 7.45-7.41 (m, 4H, ArH), 7.37 (t, 2H,  $J = 7.6$  Hz, ArH), 7.29 (d, 2H,  $J = 8.0$  Hz, ArH), 7.23-7.18 (m, 3H, ArH), 7.16-7.14 (d, 2H,  $J = 7.6$  Hz, ArH), 7.06 (t, 1H,  $J = 7.2$  Hz, ArH), 3.71 (s, 2H,  $\text{CH}_2$ ), 3.56 (s, 2H,  $\text{CH}_2$ ), 3.33 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 573.1613  $[\text{M}+\text{H}]^+$ , found: 573.1619.

**8-(4-Bromobenzylidene)-6-benzyl-4-(4-bromophenyl)-5,6,7,8-tetrahydro-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4k)**

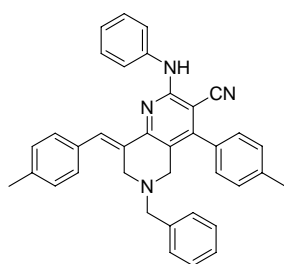


IR (KBr): 3332, 2209, 1598, 1563, 1544, 1490, 1440, 1363, 1289, 1257, 1073, 1010, 914, 899, 832, 817, 754, 698  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.07 (s, 1H, NH), 7.78 (s, 1H, =CH), 7.72 (d, 2H,  $J = 8.0$  Hz, ArH), 7.68 (d, 2H,  $J = 8.0$  Hz, ArH), 7.57 (d, 2H,  $J = 8.0$  Hz, ArH), 7.39-7.34 (m, 4H, ArH), 7.23-7.18 (m, 5H, ArH), 7.16-7.14 (m, 2H, ArH), 7.06 (t, 1H,  $J = 7.2$  Hz, ArH), 3.70 (s, 2H,  $\text{CH}_2$ ), 3.56 (s, 2H,  $\text{CH}_2$ ), 3.33 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 661.0602  $[\text{M}+\text{H}]^+$ , found: 661.0551.

**8-(4-Methylbenzylidene)-6-benzyl-4-(4-methylphenyl)-5,6,7,8-tetrahydro-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4l)**

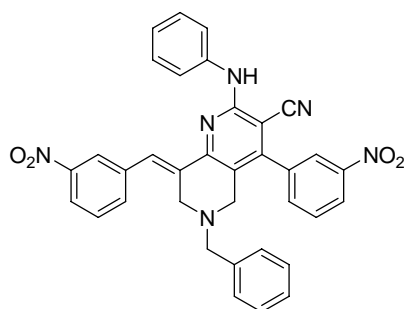


IR (KBr): 3347, 2206, 1599, 1561, 1544, 1498, 1439, 1363, 1260, 1214, 1182, 1098, 1028, 916, 816, 751, 698  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.96 (s, 1H, NH), 7.83 (s, 1H, =CH), 7.69 (d, 2H,  $J = 8.0$  Hz, ArH), 7.37 (t, 2H,  $J = 7.6$  Hz, ArH), 7.31 (d, 2H,  $J = 8.0$  Hz, ArH), 7.25 (d, 2H,  $J = 8.0$  Hz, ArH), 7.21-7.16 (m, 9H, ArH), 7.04 (t, 1H,  $J = 7.2$  Hz, ArH), 3.73 (s, 2H,  $\text{CH}_2$ ), 3.55 (s, 2H,  $\text{CH}_2$ ), 3.36 (s, 2H,  $\text{CH}_2$ ), 2.39 (s, 3H,  $\text{CH}_3$ ), 2.31 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 533.2705  $[M+H]^+$ , found: 533.2699.

**8-(3-Nitrobenzylidene)-6-benzyl-5,6,7,8-tetrahydro-4-(3-nitrophenyl)-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4m)**



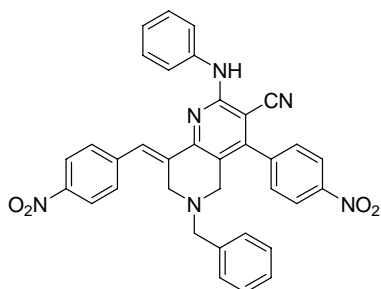
IR (KBr): 3365, 2214, 1609, 1557, 1531, 1495, 1440, 1350, 1253, 1178, 1074, 1030, 912, 807, 749, 723, 699  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.18 (s, 1H, NH), 8.38-8.36 (m, 1H, ArH), 8.31 (s, 1H, ArH), 8.15 (d, 1H,  $J = 8.0$  Hz, ArH), 8.07 (s, 1H, =CH), 7.90 (d, 2H,  $J = 8.0$  Hz, ArH), 7.83 (t, 1H,  $J = 8.0$  Hz, ArH), 7.71-7.64 (m, 4H, ArH), 7.38 (t, 2H,  $J = 8.0$  Hz, ArH), 7.14-7.11 (m, 5H, ArH), 7.08 (t, 1H,  $J = 7.2$  Hz, ArH), 3.74 (s, 2H,  $\text{CH}_2$ ), 3.55 (s, 2H,  $\text{CH}_2$ ), 3.34 (s, 2H,

$\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 595.2094  $[M+H]^+$ , found: 595.2087.

**8-(4-Nitrobenzylidene)-6-benzyl-5,6,7,8-tetrahydro-4-(4-nitrophenyl)-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4n)**



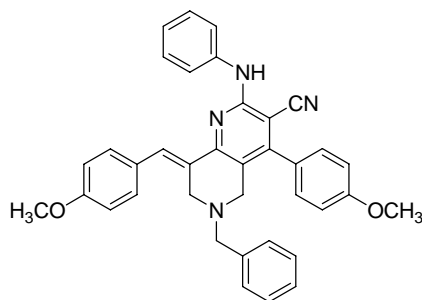
IR (KBr): 3322, 2212, 1598, 1555, 1519, 1495, 1441, 1344, 1256, 1179, 1109, 1014, 900, 852, 751, 722, 695  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.20 (s, 1H, NH), 8.36 (d, 2H,  $J = 8.4$  Hz, ArH), 8.20 (d, 2H,  $J = 8.4$  Hz, ArH), 7.89 (s, 1H, =CH), 7.73 (d, 2H,  $J = 8.8$  Hz, ArH), 7.69 (d, 2H,  $J = 8.0$  Hz, ArH), 7.53 (d, 2H,  $J = 8.8$  Hz, ArH), 7.39 (t, 2H,  $J = 8.0$  Hz, ArH), 7.20-7.17 (m, 3H, ArH), 7.14-7.11 (m, 2H, ArH), 7.07 (t, 1H,  $J = 7.2$  Hz, ArH), 3.75 (s, 2H,  $\text{CH}_2$ ), 3.55 (s, 2H,  $\text{CH}_2$ ), 3.35

(s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 595.2094  $[M+H]^+$ , found: 595.2110.

**8-(4-Methoxybenzylidene)-6-benzyl-5,6,7,8-tetrahydro-4-(4-methoxyphenyl)-2-(phenylamino)-1,6-naphthyridine-3-carbonitrile (4o)**



IR (KBr): 3337, 2217, 1608, 1547, 1510, 1441, 1382, 1298, 1253, 1174, 1030, 830, 756, 697  $\text{cm}^{-1}$ .

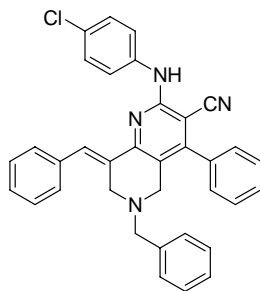
$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.93 (s, 1H, NH), 7.80 (s, 1H, =CH), 7.68 (d, 2H,  $J = 8.0$  Hz, ArH), 7.36 (t, 2H,  $J = 8.0$  Hz, ArH), 7.30 (d, 2H,  $J = 8.8$  Hz, ArH), 7.25-7.15 (m, 7H, ArH), 7.06-7.03 (m, 3H, ArH), 6.95 (d, 2H,  $J = 8.8$  Hz, ArH), 3.82 (s, 3H,  $\text{OCH}_3$ ), 3.77 (s, 3H,  $\text{OCH}_3$ ), 3.73 (s, 2H,  $\text{CH}_2$ ),

3.56 (s, 2H,  $\text{CH}_2$ ), 3.37 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 565.2603  $[M+H]^+$ , found: 565.2619.

**2-(4-Chlorophenylamino)-6-benzyl-8-benzylidene-5,6,7,8-tetrahydro-4-phenyl-1,6-naphthyridine-3-carbonitrile (4p)**

IR (KBr): 3335, 2219, 1607, 1547, 1518, 1490, 1452, 1403, 1367, 1257, 1184, 1091, 1011, 919,

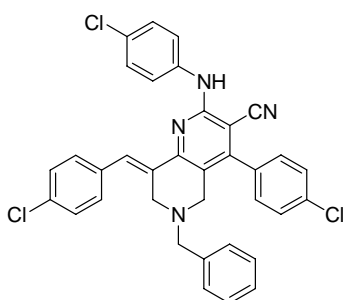


822, 749, 725, 696  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.17 (s, 1H, NH), 7.84 (s, 1H, =CH), 7.73 (d, 2H,  $J = 9.2$  Hz, ArH), 7.51-7.49 (m, 3H, ArH), 7.42 (d, 2H,  $J = 8.8$  Hz, ArH), 7.39 (s, 1H, ArH), 7.37-7.35 (m, 3H, ArH), 7.31-7.29 (m, 3H, ArH), 7.19-7.16 (m, 3H, ArH), 7.15-7.13 (m, 2H, ArH), 3.74 (s, 2H,  $\text{CH}_2$ ), 3.54 (s, 2H,  $\text{CH}_2$ ), 3.31 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 530.2002  $[\text{M}+\text{H}]^+$ , found: 530.2008.

**8-(4-Chlorobenzylidene)-2-(4-chlorophenylamino)-5,6,7,8-tetrahydro-4-(4-chlorophenyl)-6-benzyl-1,6-naphthyridine-3-carbonitrile (4q)**



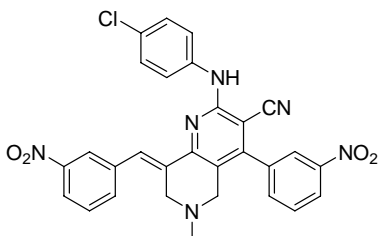
IR (KBr): 3313, 2218, 1599, 1560, 1543, 1490, 1455, 1406, 1362, 1260, 1246, 1179, 1090, 1009, 932, 912, 826, 754, 694  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.21 (s, 1H, NH), 7.78 (s, 1H, =CH), 7.72 (d, 2H,  $J = 8.8$  Hz, ArH), 7.57 (d, 2H,  $J = 8.4$  Hz, ArH), 7.43-7.39 (m, 6H, ArH), 7.31 (d, 2H,  $J = 8.8$  Hz, ArH), 7.22-7.17 (m, 3H, ArH), 7.15-7.13 (m, 2H, ArH), 3.70 (s, 2H,  $\text{CH}_2$ ), 3.54 (s, 2H,  $\text{CH}_2$ ), 3.33 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$   $m/z$  calcd for: 607.1223  $[\text{M}+\text{H}]^+$ , found:

607.1221.

**8-(3-Nitrobenzylidene)-2-(4-chlorophenylamino)-5,6,7,8-tetrahydro-6-methyl-4-(3-nitrophenyl)-1,6-naphthyridine-3-carbonitrile (4r)**

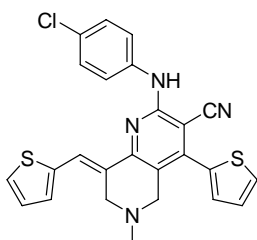


IR (KBr): 3274, 2207, 1606, 1566, 1557, 1536, 1490, 1449, 1404, 1352, 1254, 1174, 1094, 1011, 914, 828, 723, 669  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.29 (s, 1H, NH), 8.41 (d, 1H,  $J = 8.0$  Hz, ArH), 8.36 (s, 1H, ArH), 8.20 (d, 1H,  $J = 8.4$  Hz, ArH), 8.15 (s, 1H, ArH), 7.95 (d, 1H,  $J = 7.6$  Hz, ArH), 7.91 (d, 1H,  $J = 8.0$  Hz, ArH), 7.88 (s, 1H, =CH), 7.84 (d, 1H,  $J = 7.6$  Hz, ArH), 7.76-7.72 (m, 3H, ArH), 7.43 (d, 2H,  $J = 8.8$  Hz, ArH), 3.63 (s, 2H,  $\text{CH}_2$ ), 3.24 (d, 2H,  $J = 7.2$  Hz,  $\text{CH}_2$ ), 2.25 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 553.1393  $[\text{M}+\text{H}]^+$ , found: 553.1394.

**2-(4-Chlorophenylamino)-5,6,7,8-tetrahydro-6-methyl-4-(thiophen-2-yl)-8-((thiophen-2-yl)methylene)-1,6-naphthyridine-3-carbonitrile (4s)**

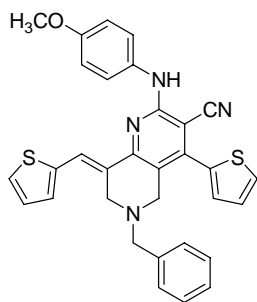


IR (KBr): 3320, 2213, 1601, 1565, 1547, 1490, 1457, 1402, 1360, 1261, 1174, 1089, 821, 699  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.19 (s, 1H, NH), 7.93 (s, 1H, =CH), 7.90-7.89 (m, 1H, ArH), 7.79 (d, 1H,  $J = 4.8$  Hz, ArH), 7.68 (d, 2H,  $J = 9.2$  Hz, ArH), 7.44 (d, 2H,  $J = 8.8$  Hz, ArH), 7.35-7.34 (m, 1H, ArH), 7.32-7.28 (m, 2H, ArH), 7.23-7.21 (m, 1H, ArH), 3.68 (s, 2H,  $\text{CH}_2$ ), 3.37 (s, 2H,  $\text{CH}_2$ ), 2.36 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 475.0818  $[\text{M}+\text{H}]^+$ , found: 475.0807.

**8-((Thiophen-2-yl)methylene)-2-(4-methoxyphenylamino)-5,6,7,8-tetrahydro-4-(thiophen-2-yl)-6-benzyl-1,6-naphthyridine-3-carbonitrile (4t)**

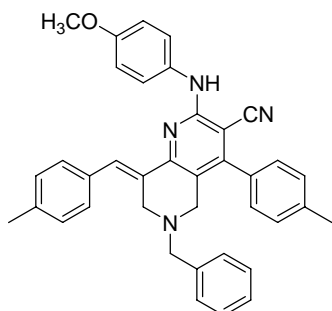


IR (KBr): 3334, 2216, 1607, 1569, 1537, 1454, 1373, 1292, 1235, 1088, 1034, 823, 706  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.86 (s, 1H, NH), 7.91 (s, 1H, =CH), 7.84-7.82 (m, 1H, ArH), 7.72 (d, 1H,  $J = 5.2$  Hz, ArH), 7.54 (d, 2H,  $J = 9.2$  Hz, ArH), 7.28-7.27 (m, 2H, ArH), 7.25-7.21 (m, 5H, ArH), 7.17-7.15 (m, 2H, ArH), 6.97 (d, 2H,  $J = 9.2$  Hz, ArH), 3.78 (s, 3H,  $\text{OCH}_3$ ), 3.71 (s, 2H,  $\text{CH}_2$ ), 3.64 (s, 2H,  $\text{CH}_2$ ), 3.49 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 547.1628  $[\text{M}+\text{H}]^+$ , found: 547.1617.

**8-(4-Methylbenzylidene)-2-(4-methoxyphenylamino)-5,6,7,8-tetrahydro-4-(4-methylphenyl)-6-benzyl-1,6-naphthyridine-3-carbonitrile (4u)**



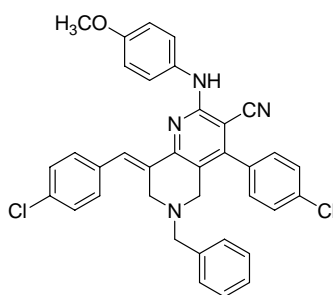
IR (KBr): 3335, 2214, 1604, 1570, 1551, 1509, 1457, 1418, 1381, 1344, 1237, 1182, 1037, 827, 700  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.78 (s, 1H, NH), 7.77 (s, 1H, =CH), 7.57 (d, 2H,  $J = 8.8$  Hz, ArH), 7.30 (d, 2H,  $J = 8.0$  Hz, ArH), 7.23 (d, 2H,  $J = 8.4$  Hz, ArH), 7.20-7.18 (m, 5H, ArH), 7.15 (d, 4H,  $J = 8.0$  Hz, ArH), 6.95 (d, 2H,  $J = 8.8$  Hz, ArH), 3.76 (s, 3H,  $\text{OCH}_3$ ), 3.70 (s, 2H,  $\text{CH}_2$ ), 3.53 (s, 2H,  $\text{CH}_2$ ), 3.33 (s, 2H,  $\text{CH}_2$ ), 2.37 (s, 3H,  $\text{CH}_3$ ), 2.30 (s, 3H,  $\text{CH}_3$ ).

$^{13}\text{C}$  NMR (100 MHz,  $\text{DMSO-}d_6$ , 25  $^\circ\text{C}$ ) ( $\delta$ , ppm): 160.73, 153.90, 153.59, 146.96, 146.81, 146.43, 146.34, 145.17, 137.74, 136.82, 132.89, 130.91, 129.50, 129.17, 128.70, 128.04, 126.95, 124.21, 123.14, 113.45, 112.71, 55.23, 50.43, 49.12, 30.67, 20.88, 20.84.

HRMS (ESI):  $m/z$  calcd for: 563.2811  $[\text{M}+\text{H}]^+$ , found: 563.2813.

**8-(4-Chlorobenzylidene)-2-(4-methoxyphenylamino)-5,6,7,8-tetrahydro-4-(4-chlorophenyl)-6-benzyl-1,6-naphthyridine-3-carbonitrile (4v)**

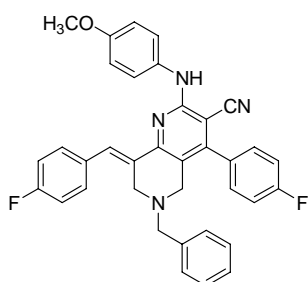


IR (KBr): 3325, 2213, 1599, 1567, 1548, 1501, 1452, 1415, 1364, 1292, 1242, 1179, 1091, 1033, 1014, 912, 823, 772, 751, 695  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.87 (s, 1H, NH), 7.76 (s, 1H, =CH), 7.58-7.55 (m, 4H, ArH), 7.41 (t, 4H,  $J = 8.8$  Hz, ArH), 7.27 (d, 2H,  $J = 8.4$  Hz, ArH), 7.22-7.17 (m, 3H, ArH), 7.15-7.13 (m, 2H, ArH), 6.95 (d, 2H,  $J = 9.2$  Hz, ArH), 3.76 (s, 3H,  $\text{OCH}_3$ ), 3.68 (s, 2H,  $\text{CH}_2$ ), 3.54 (s, 2H,  $\text{CH}_2$ ), 3.31 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 603.1718  $[\text{M}+\text{H}]^+$ , found: 603.1727.

**8-(4-fluorobenzylidene)-2-(4-methoxyphenylamino)-5,6,7,8-tetrahydro-4-(4-fluorophenyl)-6-benzyl-1,6-naphthyridine-3-carbonitrile (4w)**



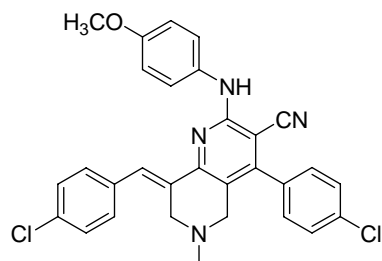
IR (KBr): 3334, 2215, 1605, 1571, 1548, 1509, 1454, 1420, 1361, 1293, 1229, 1160, 1106, 1034, 974, 909, 844, 822, 767, 746, 698, 625  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.84 (s, 1H, NH), 7.78 (s, 1H, =CH), 7.57 (d, 2H,  $J = 9.2$  Hz, ArH), 7.44-7.41 (m, 2H, ArH), 7.34 (d, 2H,  $J = 8.8$  Hz, ArH), 7.32 (d, 2H,  $J = 7.2$  Hz, ArH), 7.22 (d, 2H,  $J = 9.2$  Hz, ArH), 7.20-7.16 (m, 3H, ArH), 7.14-7.12 (m,

2H, ArH), 6.95 (d, 2H,  $J = 9.2$  Hz, ArH), 3.76 (s, 3H, OCH<sub>3</sub>), 3.69 (s, 2H, CH<sub>2</sub>), 3.53 (s, 2H, CH<sub>2</sub>), 3.30 (s, 2H, CH<sub>2</sub>).

HRMS (ESI):  $m/z$  calcd for: 571.2309 [M+H]<sup>+</sup>, found: 571.2307.

**8-(4-Chlorobenzylidene)-2-(4-methoxyphenylamino)-4-(4-chlorophenyl)-5,6,7,8-tetrahydro-6-methyl-1,6-naphthyridine-3-carbonitrile (4x)**



IR (KBr): 3320, 2215, 1597, 1561, 1548, 1509, 1491, 1449, 1412, 1364, 1295, 1245, 1179, 1090, 1013, 826 cm<sup>-1</sup>.

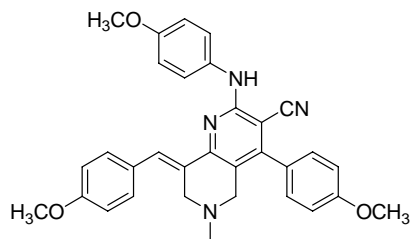
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.88 (s, 1H, NH), 7.75 (s, 1H, =CH), 7.64 (d, 2H,  $J = 8.4$  Hz, ArH), 7.55 (d, 2H,  $J = 8.8$  Hz, ArH), 7.51-7.46 (m, 4H, ArH), 7.36 (d, 2H,  $J = 8.4$  Hz, ArH), 6.95 (d, 2H,  $J = 9.2$  Hz, ArH), 3.76 (s, 3H, OCH<sub>3</sub>), 3.56 (s, 2H, CH<sub>2</sub>), 3.17 (s, 2H, CH<sub>2</sub>), 2.24 (s, 3H,

CH<sub>3</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) ( $\delta$ , ppm): 161.65, 155.23, 151.60, 134.13, 133.80, 133.33, 132.65, 132.50, 131.32, 130.22, 128.92, 128.65, 127.50, 123.26, 122.11, 122.07, 119.40, 113.49, 112.72, 100.57, 55.22, 55.01, 54.32, 45.02.

HRMS (ESI):  $m/z$  calcd for: 527.1405 [M+H]<sup>+</sup>, found: 527.1403.

**8-(4-Methoxybenzylidene)-2-(4-methoxyphenylamino)-5,6,7,8-tetrahydro-4-(4-methoxyphenyl)-6-methyl-1,6-naphthyridine-3-carbonitrile (4y)**



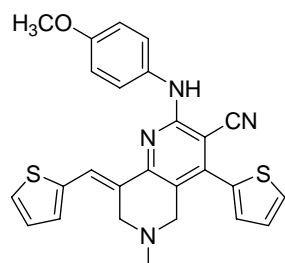
IR (KBr): 3327, 2198, 1605, 1551, 1508, 1451, 1417, 1361, 1290, 1252, 1174, 1115, 1037, 925, 837, 773 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.73 (s, 1H, NH), 7.76 (s, 1H, =CH), 7.57 (d, 2H,  $J = 8.8$  Hz, ArH), 7.34 (d, 2H,  $J = 8.8$  Hz, ArH), 7.29 (d, 2H,  $J = 8.8$  Hz, ArH), 7.10 (d, 2H,  $J = 8.4$  Hz, ArH), 7.01 (d, 2H,  $J = 8.8$  Hz, ArH), 6.95 (d, 2H,

$J = 8.8$  Hz, ArH), 3.85 (s, 3H, OCH<sub>3</sub>), 3.79 (s, 3H, OCH<sub>3</sub>), 3.76 (s, 3H, OCH<sub>3</sub>), 3.58 (s, 2H, CH<sub>2</sub>), 3.19 (s, 2H, CH<sub>2</sub>), 2.24 (s, 3H, CH<sub>3</sub>).

HRMS (ESI):  $m/z$  calcd for: 519.2396 [M+H]<sup>+</sup>, found: 519.2397.

**2-(4-Methoxyphenylamino)-5,6,7,8-tetrahydro-6-methyl-4-(thiophen-2-yl)-8-((thiophen-2-yl)methylene)-1,6-naphthyridine-3-carbonitrile (4z)**



IR (KBr): 3327, 2213, 1601, 1550, 1509, 1459, 1414, 1361, 1290, 1265, 1234, 1176, 1101, 1035, 855, 825, 712 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.86 (s, 1H, NH), 7.90 (s, 1H, =CH), 7.87 (d, 1H,  $J = 4.8$  Hz, ArH), 7.77 (d, 1H,  $J = 4.8$  Hz, ArH), 7.53 (d, 2H,  $J = 8.8$  Hz, ArH), 7.33 (d, 1H,  $J = 2.4$  Hz, ArH), 7.29-7.27 (m, 1H, ArH), 7.24 (d, 1H,  $J = 3.2$  Hz, ArH), 7.22-7.20 (m, 1H, ArH), 6.98 (d, 2H,  $J = 9.2$  Hz, ArH), 3.78 (s, 3H, OCH<sub>3</sub>), 3.65 (s,

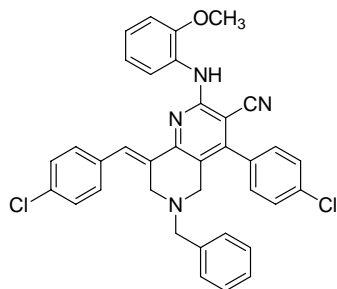
2H, CH<sub>2</sub>), 3.33 (s, 2H, CH<sub>2</sub>), 2.35 (s, 3H, CH<sub>3</sub>).

HRMS (ESI):  $m/z$  calcd for: 471.1308 [M+H]<sup>+</sup>, found: 471.1306.

**8-(4-Chlorobenzylidene)-2-(2-methoxyphenylamino)-6-benzyl-5,6,7,8-tetrahydro-4-(4-chloro**



**phenyl)-1,6-naphthyridine-3-carbonitrile(4aa)**

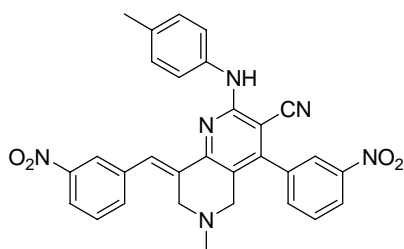


IR (KBr): 3391, 2208, 1606, 1567, 1548, 1520, 1483, 1455, 1382, 1288, 1259, 1213, 1179, 1092, 1027, 921, 829, 746, 696  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.29-8.26 (m, 1H, ArH), 8.07 (s, 1H, NH), 7.83 (s, 1H, =CH), 7.58 (d, 2H,  $J = 8.8$  Hz, ArH), 7.45-7.42 (m, 4H, ArH), 7.31 (d, 2H,  $J = 8.4$  Hz, ArH), 7.20-7.17 (m, 3H, ArH), 7.15-7.12 (m, 3H, ArH), 7.10-7.04 (m, 2H, ArH), 3.89 (s, 3H,  $\text{OCH}_3$ ), 3.70 (s, 2H,  $\text{CH}_2$ ), 3.54 (s, 2H,  $\text{CH}_2$ ), 3.33 (s, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 603.1718  $[\text{M}+\text{H}]^+$ , found: 603.1718.

**8-(3-Nitrobenzylidene)-2-(4-methylphenylamino)-5,6,7,8-tetrahydro-6-methyl-4-(3-nitrophenyl)-1,6-naphthyridine-3-carbonitrile (4ab)**



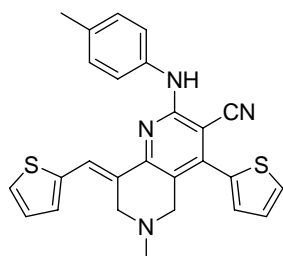
IR (KBr): 3265, 2207, 1611, 1556, 1528, 1454, 1408, 1348, 1299, 1258, 1101, 932, 907, 815, 720, 683  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.03 (s, 1H, NH), 8.42-8.40 (m, 1H, ArH), 8.36 (s, 1H, ArH), 8.21-8.19 (m, 1H, ArH), 8.12 (s, 1H, ArH), 7.95 (d, 1H,  $J = 7.6$  Hz, ArH), 7.90 (d, 1H,  $J = 7.6$  Hz, ArH), 7.89 (s, 1H, =CH), 7.81 (d, 1H,  $J = 8.0$  Hz, ArH), 7.74 (t, 1H,  $J = 8.0$  Hz,

ArH), 7.58 (d, 2H,  $J = 8.4$  Hz, ArH), 7.18 (d, 2H,  $J = 8.4$  Hz, ArH), 3.63 (s, 2H,  $\text{CH}_2$ ), 3.23 (d, 2H,  $J = 7.6$  Hz,  $\text{CH}_2$ ), 2.30 (s, 3H,  $\text{CH}_3$ ), 2.24 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 533.1937  $[\text{M}+\text{H}]^+$ , found: 533.1953.

**2-(4-Methylphenylamino)-5,6,7,8-tetrahydro-6-methyl-4-(thiophen-2-yl)-8-((thiophen-2-yl)methylene)-1,6-naphthyridine-3-carbonitrile (4ac)**



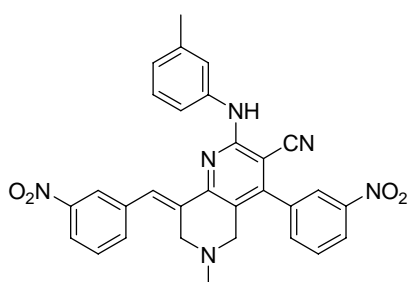
IR (KBr): 3335, 2216, 1614, 1540, 1509, 1464, 1408, 1359, 1287, 1247, 1174, 1129, 1101, 813, 699  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.92 (s, 1H, NH), 7.94 (s, 1H, =CH), 7.89-7.87 (m, 1H, ArH), 7.78 (d, 1H,  $J = 4.8$  Hz, ArH), 7.53 (d, 2H,  $J = 8.4$  Hz, ArH), 7.34-7.33 (m, 1H, ArH), 7.29-7.27 (m, 1H, ArH), 7.25 (d, 1H,  $J = 3.2$  Hz, ArH), 7.23-7.22 (m, 1H, ArH), 7.21-7.18 (m, 2H, ArH), 3.67 (s, 2H,  $\text{CH}_2$ ), 3.36 (s, 2H,  $\text{CH}_2$ ), 2.35

(s, 3H,  $\text{CH}_3$ ), 2.32 (s, 3H,  $\text{CH}_3$ ).

HRMS (ESI):  $m/z$  calcd for: 455.1366  $[\text{M}+\text{H}]^+$ , found: 455.1349.

**8-(3-nitrobenzylidene)-2-(3-methylphenylamino)-5,6,7,8-tetrahydro-6-methyl-4-(3-nitrophenyl)-1,6-naphthyridine-3-carbonitrile (4ad)**



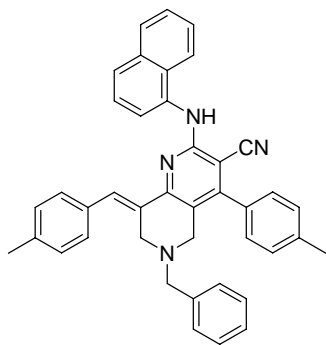
IR (KBr): 3386, 2211, 1595, 1568, 1553, 1523, 1449, 1346, 1257, 1168, 1119, 1097, 920, 822, 783, 718, 687  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 9.06 (s, 1H, NH), 8.43-8.40 (m, 1H, ArH), 8.36 (s, 1H, ArH), 8.21-8.19 (m, 1H, ArH), 8.12 (s, 1H, ArH), 7.95 (d, 1H,  $J = 8.0$  Hz, ArH), 7.92-7.88 (m, 2H, ArH), 7.81 (d, 1H,  $J = 8.0$  Hz, ArH), 7.75 (t, 1H,  $J = 8.0$  Hz, ArH), 7.63 (s, 1H, =CH), 7.46 (d, 1H,  $J = 8.8$  Hz, ArH), 7.25 (t,

1H,  $J = 8.0$  Hz, ArH), 6.90 (d, 1H,  $J = 7.6$  Hz, ArH), 3.66 (s, 2H, CH<sub>2</sub>), 3.25-3.24 (m, 2H, CH<sub>2</sub>), 2.35 (s, 3H, CH<sub>3</sub>), 2.26 (s, 3H, CH<sub>3</sub>).

HRMS (ESI):  $m/z$  calcd for: 533.1937 [M+H]<sup>+</sup>, found: 533.1912.

**6-Benzyl-8-(4-methylbenzylidene)-2-(naphthalen-1-ylamino)-5,6,7,8-tetrahydro-4-(4-methylphenyl)-1,6-naphthyridine-3-carbonitrile (4ae)**



IR (KBr): 3416, 3093, 2204, 1610, 1567, 1561, 1520, 1490, 1452, 1362, 1272, 1213, 1182, 1020, 972, 895, 811, 784, 697 cm<sup>-1</sup>.

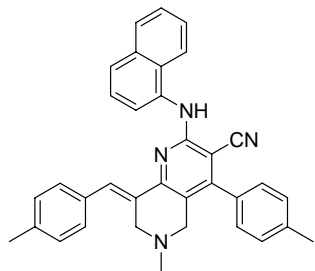
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 9.17 (s, 1H, NH), 7.99 (d, 1H,  $J = 8.0$  Hz, ArH), 7.93 (d, 1H,  $J = 8.4$  Hz, ArH), 7.82 (d, 1H,  $J = 8.0$  Hz, ArH), 7.63 (d, 1H,  $J = 6.8$  Hz, ArH), 7.57-7.47 (m, 3H, ArH), 7.31 (d, 2H,  $J = 8.4$  Hz, ArH), 7.28 (d, 2H,  $J = 8.0$  Hz, ArH), 7.20-7.15 (m, 3H, ArH), 7.13-7.11 (m, 2H, ArH), 7.08 (d, 2H,  $J = 8.0$  Hz, ArH), 6.92 (s, 1H, =CH), 6.77 (d, 2H,  $J = 8.0$  Hz,

ArH), 3.59 (s, 2H, CH<sub>2</sub>), 3.50 (s, 2H, CH<sub>2</sub>), 3.32 (s, 2H, CH<sub>2</sub>), 2.38 (s, 3H, CH<sub>3</sub>), 2.24 (s, 3H, CH<sub>3</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) ( $\delta$ , ppm): 155.52, 153.45, 152.33, 138.59, 137.69, 137.35, 136.20, 133.82, 132.83, 132.07, 131.18, 129.39, 129.20, 129.04, 128.67, 128.17, 128.01, 126.96, 125.68, 125.58, 125.41, 125.28, 123.96, 122.10, 119.16, 116.48, 91.47, 60.39, 52.84, 52.15, 30.67, 20.89, 20.76.

HRMS (ESI):  $m/z$  calcd for: 583.2862 [M+H]<sup>+</sup>, found: 583.2860.

**6-Methyl-8-(4-methylbenzylidene)-2-(naphthalen-1-ylamino)-5,6,7,8-tetrahydro-4-(4-methylphenyl)-1,6-naphthyridine-3-carbonitrile (4af)**



IR (KBr): 3432, 3024, 2205, 1588, 1547, 1527, 1498, 1459, 1404, 1362, 1261, 1181, 1105, 914, 828, 794, 775 cm<sup>-1</sup>.

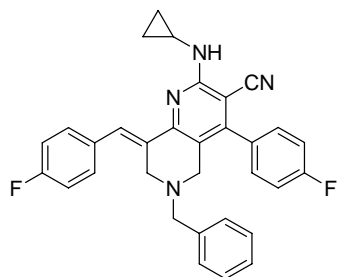
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 9.17 (s, 1H, NH), 7.99 (d, 1H,  $J = 8.0$  Hz, ArH), 7.91 (d, 1H,  $J = 8.0$  Hz, ArH), 7.82 (d, 1H,  $J = 8.0$  Hz, ArH), 7.62 (d, 1H,  $J = 7.2$  Hz, ArH), 7.57-7.47 (m, 3H, ArH), 7.39 (d, 2H,  $J = 8.0$  Hz, ArH), 7.34 (d, 2H,  $J = 8.0$  Hz, ArH), 7.15 (d, 2H,  $J = 8.0$  Hz, ArH), 6.92 (s, 1H, =CH), 6.86 (d, 2H,  $J = 8.4$  Hz, ArH), 3.48 (s, 2H, CH<sub>2</sub>), 3.17 (s, 2H, CH<sub>2</sub>), 2.42 (s, 3H, CH<sub>3</sub>), 2.28 (s, 3H,

CH<sub>3</sub>), 2.20 (s, 3H, CH<sub>3</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) ( $\delta$ , ppm): 155.45, 153.27, 151.87, 138.71, 137.39, 136.19, 133.82, 132.91, 132.17, 131.48, 129.34, 129.30, 129.11, 128.84, 128.22, 128.02, 125.68, 125.59, 125.40, 125.27, 123.95, 122.04, 119.40, 116.47, 91.40, 55.30, 54.50, 45.17, 30.67, 20.92, 20.80.

HRMS (ESI):  $m/z$  calcd for: 507.2550 [M+H]<sup>+</sup>, found: 507.2555.

**8-(4-Fluorobenzylidene)-6-benzyl-2-(cyclopropylamino)-4-(4-fluorophenyl)-5,6,7,8-tetrahydro-1,6-naphthyridine-3-carbonitrile (4ag)**

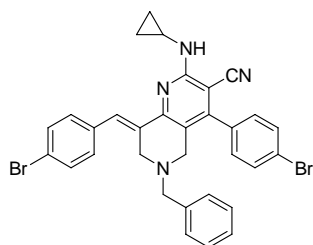


IR (KBr): 3365, 2207, 1644, 1605, 1558, 1493, 1351, 1236, 1159, 1098, 1014, 903, 836, 760, 700  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.07 (s, 1H, NH), 7.40-7.38 (m, 2H, ArH), 7.37-7.35 (m, 2H, ArH), 7.33 (s, 1H, ArH), 7.31 (s, 1H, =CH), 7.24-7.21 (m, 2H, ArH), 7.20-7.17 (m, 4H, ArH), 7.14-7.12 (m, 2H, ArH), 3.69 (s, 2H,  $\text{CH}_2$ ), 3.53 (s, 2H,  $\text{CH}_2$ ), 3.26 (s, 2H,  $\text{CH}_2$ ), 2.92-2.89 (m, 1H, CH), 0.79-0.77 (m, 2H,  $\text{CH}_2$ ), 0.62-0.61 (m, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 505.2204  $[\text{M}+\text{H}]^+$ , found: 505.2209.

**8-(4-bromobenzylidene)-6-benzyl-4-(4-bromophenyl)-2-(cyclopropylamino)-5,6,7,8-tetrahydro-1,6-naphthyridine-3-carbonitrile (4ah)**

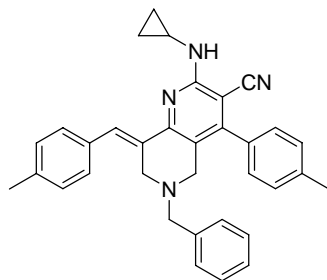


IR (KBr): 3347, 2208, 1591, 1554, 1489, 1343, 1259, 1181, 1074, 1009, 912, 824, 759, 700  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.03 (s, 1H, NH), 7.67 (d, 2H,  $J = 8.4$  Hz, ArH), 7.56 (d, 2H,  $J = 8.4$  Hz, ArH), 7.27 (d, 4H,  $J = 8.4$  Hz, ArH), 7.22-7.17 (m, 4H, ArH and =CH), 7.15-7.13 (m, 2H, ArH), 3.67 (s, 2H,  $\text{CH}_2$ ), 3.53 (s, 2H,  $\text{CH}_2$ ), 3.27 (s, 2H,  $\text{CH}_2$ ), 2.94-2.88 (m, 1H, CH), 0.78-0.76 (m, 2H,  $\text{CH}_2$ ), 0.63-0.59 (m, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 625.0602  $[\text{M}+\text{H}]^+$ , found: 625.0556.

**8-(4-Methylbenzylidene)-6-benzyl-2-(cyclopropylamino)-4-(4-methylphenyl)-5,6,7,8-tetrahydro-1,6-naphthyridine-3-carbonitrile (4ai)**



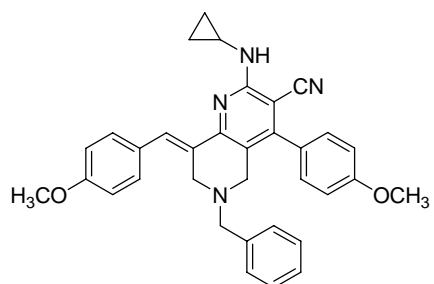
IR (KBr): 3354, 2209, 1640, 1556, 1491, 1453, 1340, 1258, 1179, 1100, 1021, 905, 817, 756, 699  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.06 (s, 1H, NH), 7.27 (d, 2H,  $J = 8.4$  Hz, ArH), 7.23 (s, 1H, ArH), 7.21-7.18 (m, 7H, ArH), 7.16-7.14 (m, 3H, ArH), 7.07 (d, 1H,  $J = 2.8$  Hz, =CH), 3.70 (s, 2H,  $\text{CH}_2$ ), 3.52 (s, 2H,  $\text{CH}_2$ ), 3.29 (s, 2H,  $\text{CH}_2$ ), 2.91-2.88 (m, 1H, CH), 2.35 (s, 3H,  $\text{CH}_3$ ), 2.31 (s, 3H,  $\text{CH}_3$ ), 0.80-0.75 (m, 2H,  $\text{CH}_2$ ),

0.62-0.59 (m, 2H,  $\text{CH}_2$ ).

HRMS (ESI):  $m/z$  calcd for: 497.2705  $[\text{M}+\text{H}]^+$ , found: 497.2736.

**8-(4-Methoxybenzylidene)-6-benzyl-2-(cyclopropylamino)-5,6,7,8-tetrahydro-4-(4-methoxyphenyl)-1,6-naphthyridine-3-carbonitrile (4aj)**



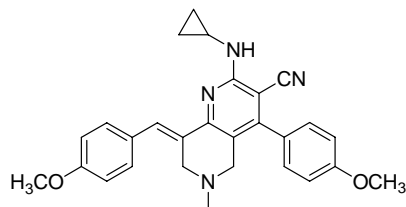
IR (KBr): 3372, 2207, 1605, 1556, 1511, 1497, 1454, 1338, 1292, 1250, 1176, 1089, 1032, 931, 835, 756, 700  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.04 (s, 1H, NH), 7.29 (d, 2H,  $J = 8.8$  Hz, ArH), 7.24-7.20 (m, 3H, ArH), 7.19-7.15 (m, 4H, ArH), 7.04 (d, 1H,  $J = 2.8$  Hz, =CH), 7.00 (d, 2H,  $J = 8.8$  Hz, ArH), 6.95 (d, 2H,  $J = 8.4$  Hz,

ArH), 3.80 (s, 3H, OCH<sub>3</sub>), 3.77 (s, 3H, OCH<sub>3</sub>), 3.70 (s, 2H, CH<sub>2</sub>), 3.53 (s, 2H, CH<sub>2</sub>), 3.30 (s, 2H, CH<sub>2</sub>), 2.92-2.87 (m, 1H, CH), 0.78-0.76 (m, 2H, CH<sub>2</sub>), 0.62-0.60 (m, 2H, CH<sub>2</sub>).

HRMS (ESI): *m/z* calcd for: 529.2603 [M+H]<sup>+</sup>, found: 529.2561.

**8-(4-Methoxybenzylidene)-2-(cyclopropylamino)-5,6,7,8-tetrahydro-4-(4-methoxyphenyl)-6-methyl-1,6-naphthyridine-3-carbonitrile (4ak)**



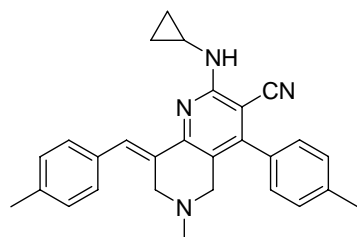
IR (KBr): 3369, 2209, 1608, 1553, 1499, 1460, 1339, 1293, 1249, 1174, 1118, 1029, 928, 838, 818 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.03 (s, 1H, NH), 7.36 (d, 2H, *J* = 8.8 Hz, ArH), 7.29 (d, 2H, *J* = 8.8 Hz, ArH), 7.08 (d, 2H, *J* = 8.8 Hz, ArH), 7.03-7.00 (m, 3H, ArH and =CH), 3.83 (s, 3H, OCH<sub>3</sub>), 3.80 (s, 3H, OCH<sub>3</sub>), 3.58 (s,

2H, CH<sub>2</sub>), 3.16 (s, 2H, CH<sub>2</sub>), 2.92-2.88 (m, 1H, CH), 2.24 (s, 3H, CH<sub>3</sub>), 0.80-0.75 (m, 2H, CH<sub>2</sub>), 0.61-0.60 (m, 2H, CH<sub>2</sub>).

HRMS (ESI): *m/z* calcd for: 453.2290 [M+H]<sup>+</sup>, found: 453.2285.

**8-(4-Methylbenzylidene)-2-(cyclopropylamino)-4-(4-methylphenyl)-5,6,7,8-tetrahydro-6-methyl-1,6-naphthyridine-3-carbonitrile (4al)**



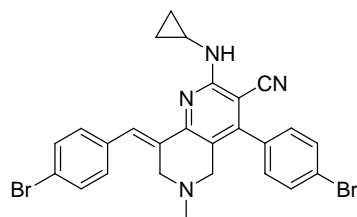
IR (KBr): 3209, 2209, 1557, 1508, 1501, 1450, 1351, 1263, 1179, 1114, 1018, 924, 818, 738 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.05 (s, 1H, NH), 7.35-7.29 (m, 4H, ArH), 7.25 (t, 4H, *J* = 8.0 Hz, ArH), 7.10 (d, 1H, *J* = 2.0 Hz, =CH), 3.57 (s, 2H, CH<sub>2</sub>), 3.14 (s, 2H, CH<sub>2</sub>), 2.94-2.88 (m, 1H, CH), 2.39 (s, 3H, CH<sub>3</sub>), 2.34 (s, 3H, CH<sub>3</sub>), 2.22 (s,

3H, CH<sub>3</sub>), 0.79-0.74 (m, 2H, CH<sub>2</sub>), 0.62-0.58 (m, 2H, CH<sub>2</sub>).

HRMS (ESI): *m/z* calcd for: 421.2392 [M+H]<sup>+</sup>, found: 421.2381.

**8-(4-Bromobenzylidene)-4-(4-bromophenyl)-2-(cyclopropylamino)-5,6,7,8-tetrahydro-6-methyl-1,6-naphthyridine-3-carbonitrile (4am)**



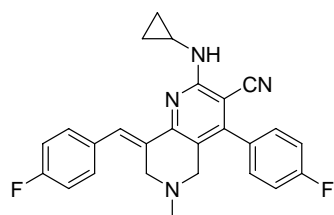
IR (KBr): 3357, 2210, 1592, 1557, 1501, 1488, 1449, 1340, 1261, 1182, 1121, 1072, 1011, 923, 893, 835, 816, 740 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.02 (s, 1H, NH), 7.74 (d, 2H, *J* = 8.4 Hz, ArH), 7.64 (d, 2H, *J* = 8.4 Hz, ArH), 7.35 (t, 4H, *J* = 7.6 Hz, ArH), 7.20 (d, 1H, *J* = 2.8 Hz, =CH), 3.56 (s, 2H, CH<sub>2</sub>), 3.14 (s, 2H, CH<sub>2</sub>), 2.94-2.88 (m, 1H, CH), 2.24 (s,

3H, CH<sub>3</sub>), 0.80-0.75 (m, 2H, CH<sub>2</sub>), 0.63-0.59 (m, 2H, CH<sub>2</sub>).

HRMS (ESI): *m/z* calcd for: 549.0289 [M+H]<sup>+</sup>, found: 549.0307.

**8-(4-Fluorobenzylidene)-2-(cyclopropylamino)-4-(4-fluorophenyl)-5,6,7,8-tetrahydro-6-methyl-1,6-naphthyridine-3-carbonitrile (4an)**



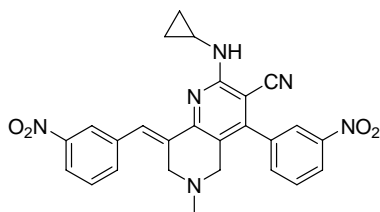
IR (KBr): 3199, 2210, 1605, 1565, 1507, 1452, 1357, 1296, 1223, 1183, 1158, 1117, 1016, 927, 905, 846, 825, 781, 624 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.06 (s, 1H, NH), 7.48-7.42 (m,

4H, ArH), 7.38 (t, 2H,  $J = 8.8$  Hz, ArH), 7.28 (t, 2H,  $J = 8.8$  Hz, ArH), 7.15 (d, 1H,  $J = 2.8$  Hz, =CH), 3.57 (s, 2H, CH<sub>2</sub>), 3.14 (s, 2H, CH<sub>2</sub>), 2.94-2.88 (m, 1H, CH), 2.23 (s, 3H, CH<sub>3</sub>), 0.80-0.75 (m, 2H, CH<sub>2</sub>), 0.63-0.59 (m, 2H, CH<sub>2</sub>).

HRMS (ESI):  $m/z$  calcd for: 429.1891 [M+H]<sup>+</sup>, found: 429.1894.

**8-(3-Nitrobenzylidene)-2-(cyclopropylamino)-5,6,7,8-tetrahydro-6-methyl-4-(3-nitrophenyl)-1,6-naphthyridine-3-carbonitrile (4ao)**



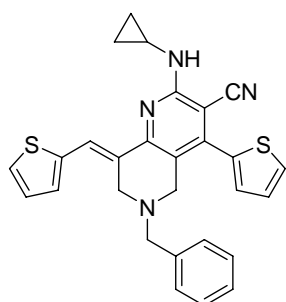
IR (KBr): 3363, 2213, 1557, 1529, 1480, 1454, 1349, 1265, 1099, 918, 815, 740, 722 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.40-8.37 (m, 1H, ArH), 8.28 (s, 1H, ArH), 8.20-8.19 (m, 2H, ArH), 8.16 (s, 1H, NH), 7.91-7.84 (m, 3H, ArH), 7.78-7.73 (m, 1H, ArH), 7.32 (d, 1H,  $J = 2.8$  Hz, =CH), 3.63 (s, 2H, CH<sub>2</sub>), 3.19 (m, 2H,

CH<sub>2</sub>), 3.00-2.96 (m, 1H, CH), 2.24 (s, 3H, CH<sub>3</sub>), 0.81-0.79 (m, 2H, CH<sub>2</sub>), 0.64-0.63 (m, 2H, CH<sub>2</sub>).

HRMS (ESI):  $m/z$  calcd for: 483.1781 [M+H]<sup>+</sup>, found: 483.1765.

**6-Benzyl-2-(cyclopropylamino)-5,6,7,8-tetrahydro-4-(thiophen-2-yl)-8-((thiophen-2-yl)methylene)-1,6-naphthyridine-3-carbonitrile (4ap)**

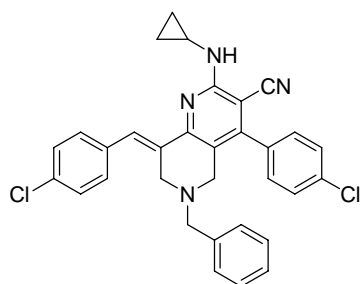


IR (KBr): 3359, 2206, 1559, 1523, 1489, 1449, 1357, 1249, 1144, 1094, 749, 698 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.20 (s, 1H, NH), 7.80-7.79 (m, 1H, ArH), 7.72 (d, 1H,  $J = 5.2$  Hz, ArH), 7.31 (d, 1H,  $J = 3.2$  Hz, =CH), 7.27-7.21 (m, 6H, ArH), 7.20-7.19 (m, 1H, ArH), 7.18-7.16 (m, 2H, ArH), 3.72 (s, 2H, CH<sub>2</sub>), 3.64 (s, 2H, CH<sub>2</sub>), 3.46 (s, 2H, CH<sub>2</sub>), 2.91-2.88 (m, 1H, CH), 0.82-0.78 (m, 2H, CH<sub>2</sub>), 0.63-0.59 (m, 2H, CH<sub>2</sub>).

HRMS (ESI):  $m/z$  calcd for: 481.1520 [M+H]<sup>+</sup>, found: 481.1540.

**8-(4-Chlorobenzylidene)-6-benzyl-4-(4-chlorophenyl)-2-(cyclopropylamino)-5,6,7,8-tetrahydro-1,6-naphthyridine-3-carbonitrile (4aq)**



IR (KBr): 3349, 2209, 1597, 1555, 1491, 1343, 1259, 1180, 1089, 1013, 912, 836, 758, 700 cm<sup>-1</sup>.

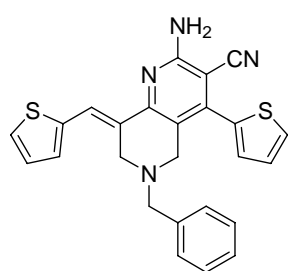
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.05 (s, 1H, NH), 7.54 (d, 2H,  $J = 8.8$  Hz, ArH), 7.43 (d, 2H,  $J = 8.8$  Hz, ArH), 7.35 (d, 4H,  $J = 8.4$  Hz, ArH), 7.21-7.20 (m, 2H, ArH and =CH), 7.18-7.16 (m, 2H, ArH), 7.15-7.14 (m, 2H, ArH), 3.68 (s, 2H, CH<sub>2</sub>), 3.53 (s, 2H, CH<sub>2</sub>), 3.27 (s, 2H, CH<sub>2</sub>), 2.93-2.89 (m, 1H, CH), 0.80-0.76 (m, 2H, CH<sub>2</sub>), 0.63-0.59 (m, 2H, CH<sub>2</sub>).

HRMS (ESI):  $m/z$  calcd for: 537.1613 [M+H]<sup>+</sup>, found: 537.1611.

**2-Amino-6-benzyl-5,6,7,8-tetrahydro-4-(thiophen-2-yl)-8-((thiophen-2-yl)methylene)-1,6-naphthyridine-3-carbonitrile (4ar)**

IR (KBr): 3275, 3201, 2207, 1621, 1556, 1454, 1412, 1352, 1267, 1215, 1088, 1067, 935, 855,

750, 716 cm<sup>-1</sup>.



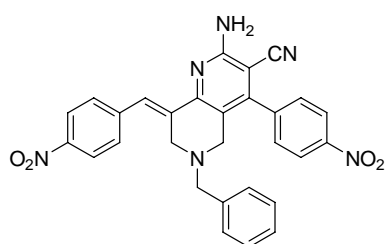
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.07 (s, 1H, =CH), 7.80 (d, 1H, *J* = 5.2 Hz, ArH), 7.73 (d, 1H, *J* = 5.2 Hz, ArH), 7.29-7.25 (m, 5H, ArH), 7.23 (m, 2H, ArH), 7.21-7.16 (m, 2H, ArH), 6.81 (s, 2H, NH<sub>2</sub>), 3.70 (s, 2H, CH<sub>2</sub>), 3.63 (s, 2H, CH<sub>2</sub>), 3.33 (s, 2H, CH<sub>2</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) (δ, ppm): 158.24, 153.04, 145.23, 138.97, 137.55, 134.08, 130.68, 129.57, 129.41, 129.27, 128.94, 128.80, 128.15, 128.11, 127.62, 127.13, 121.38, 118.33,

116.61, 89.28, 60.67, 53.13, 52.20.

HRMS (ESI): *m/z* calcd for: 441.1207 [M+H]<sup>+</sup>, found: 441.1192.

#### 8-(4-Nitrobenzylidene)-2-amino-6-benzyl-5,6,7,8-tetrahydro-4-(4-nitrophenyl)-1,6-naphthyridine-3-carbonitrile (4as)



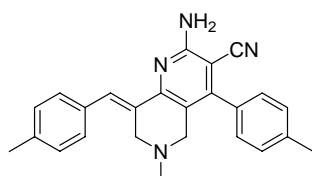
IR (KBr): 3343, 3217, 2211, 1594, 1556, 1513, 1455, 1429, 1342, 1260, 1108, 1014, 894, 851, 750, 703 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.33 (d, 2H, *J* = 8.8, ArH), 8.20 (d, 2H, *J* = 8.8 Hz, ArH), 8.02 (s, 1H, =CH), 7.67 (d, 2H, *J* = 8.8 Hz, ArH), 7.56 (d, 2H, *J* = 8.8 Hz, ArH), 7.20-7.11 (m, 5H, ArH), 6.99 (s, 2H, NH<sub>2</sub>), 3.71 (s, 2H, CH<sub>2</sub>), 3.52 (s, 2H, CH<sub>2</sub>), 3.28 (s, 2H, CH<sub>2</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) (δ, ppm): 157.90, 152.30, 151.25, 147.79, 146.15, 142.71, 141.64, 137.43, 135.55, 130.44, 129.96, 128.80, 128.04, 127.02, 126.78, 123.75, 123.57, 117.30, 116.08, 89.46, 59.77, 52.22, 51.78.

HRMS (ESI): *m/z* calcd for: 519.1781 [M+H]<sup>+</sup>, found: 519.1767.

#### 8-(4-Methylbenzylidene)-2-amino-4-(4-methylphenyl)-5,6,7,8-tetrahydro-6-methyl-1,6-naphthyridine-3-carbonitrile (4at)



IR (KBr): 3330, 3214, 2219, 1626, 1555, 1513, 1449, 1426, 1362, 1265, 1125, 923, 907, 810, 737, 703 cm<sup>-1</sup>.

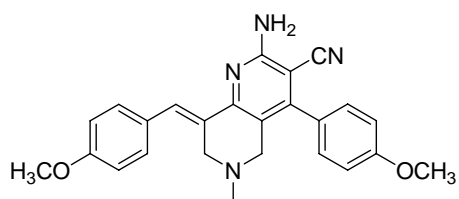
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 7.92 (s, 1H, =CH), 7.34 (d, 2H, *J* = 8.0 Hz, ArH), 7.27-7.24 (m, 6H, ArH), 6.71 (s, 2H, NH<sub>2</sub>), 3.55 (s, 2H, CH<sub>2</sub>), 3.12 (s, 2H, CH<sub>2</sub>), 2.39 (s, 3H, CH<sub>3</sub>), 2.34 (s, 3H, CH<sub>3</sub>),

2.20 (s, 3H, CH<sub>3</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) (δ, ppm): 160.45, 157.94, 139.12, 138.20, 132.51, 131.59, 130.18, 129.79, 129.51, 129.34, 129.27, 129.22, 128.08, 127.62, 116.06, 54.43, 54.08, 44.79, 20.92.

HRMS (ESI): *m/z* calcd for: 381.2079 [M+H]<sup>+</sup>, found: 381.2093.

#### 8-(4-methoxybenzylidene)-2-amino-5,6,7,8-tetrahydro-4-(4-methoxyphenyl)-6-methyl-1,6-naphthyridine-3-carbonitrile (4au)



IR (KBr): 3356, 3226, 2214, 1632, 1605, 1552, 1510, 1460, 1428, 1361, 1262, 1175, 1126, 1027, 927, 901, 838, 787, 744 cm<sup>-1</sup>.

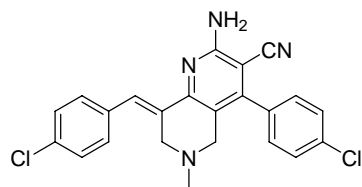
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 7.90 (s, 1H, =CH),

7.35-7.30 (m, 4H,  $J = 8.8$  Hz, ArH), 7.08 (d, 2H,  $J = 8.4$  Hz, ArH), 7.02 (d, 2H,  $J = 8.8$  Hz, ArH), 6.67 (s, 2H, NH<sub>2</sub>), 3.83 (s, 3H, OCH<sub>3</sub>), 3.80 (s, 3H, OCH<sub>3</sub>), 3.56 (s, 2H, CH<sub>2</sub>), 3.15 (s, 2H, CH<sub>2</sub>), 2.22 (s, 3H, CH<sub>3</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) (δ, ppm): 159.64, 158.90, 157.97, 152.64, 152.41, 131.12, 130.72, 129.72, 128.55, 127.96, 117.60, 116.92, 114.13, 114.04, 88.95, 56.02, 55.66, 55.18, 54.78, 45.26.

HRMS (ESI):  $m/z$  calcd for: 413.1978 [M+H]<sup>+</sup>, found: 413.2001.

**8-(4-Chlorobenzylidene)-2-amino-4-(4-chlorophenyl)-5,6,7,8-tetrahydro-6-methyl-1,6-naphthyridine-3-carbonitrile (4av)**



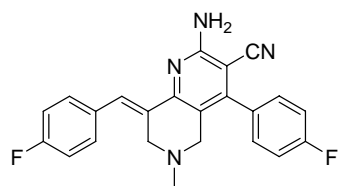
IR (KBr): 3288, 3159, 2210, 1624, 1571, 1555, 1493, 1453, 1422, 1368, 1274, 1260, 1183, 1129, 1089, 1010, 917, 901, 847, 834, 818, 739 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 7.91 (s, 1H, =CH), 7.61 (d, 2H,  $J = 8.4$  Hz, ArH), 7.50 (d, 2H,  $J = 8.4$  Hz, ArH), 7.44-7.39 (m, 4H, ArH), 6.80 (s, 2H, NH<sub>2</sub>), 3.54 (s, 2H, CH<sub>2</sub>), 3.13 (s, 2H, CH<sub>2</sub>), 2.22 (s, 3H, CH<sub>3</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) (δ, ppm): 158.36, 152.46, 134.44, 133.25, 133.10, 131.59, 131.44, 130.25, 129.89, 129.09, 128.84, 115.95, 112.72, 90.17, 52.55, 51.79, 42.37, 30.69.

HRMS (ESI):  $m/z$  calcd for: 421.0987 [M+H]<sup>+</sup>, found: 421.1007.

**8-(4-Fluorobenzylidene)-2-amino-4-(4-fluorophenyl)-5,6,7,8-tetrahydro-6-methyl-1,6-naphthyridine-3-carbonitrile (4aw)**



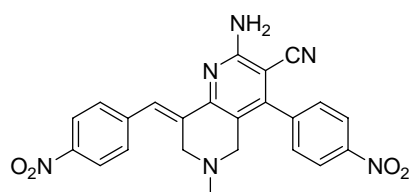
IR (KBr): 3273, 3146, 2215, 1625, 1602, 1558, 1505, 1452, 1423, 1362, 1265, 1224, 1160, 1105, 1012, 917, 848, 829, 798, 736 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 7.92 (s, 1H, =CH), 7.47-7.41 (m, 4H, ArH), 7.38 (t, 2H,  $J = 8.8$  Hz, ArH), 7.28 (t, 2H,  $J = 8.8$  Hz, ArH), 6.76 (s, 2H, NH<sub>2</sub>), 3.54 (s, 2H, CH<sub>2</sub>), 3.13 (s, 2H, CH<sub>2</sub>), 2.22 (s, 3H, CH<sub>3</sub>).

<sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>, 25 °C) (δ, ppm): 163.58, 162.70, 161.14, 160.25, 157.88, 152.30, 151.83, 132.45, 131.66, 131.58, 130.65, 130.57, 127.10, 117.68, 116.56, 115.85, 115.65, 115.44, 89.32, 55.24, 54.58, 45.17.

HRMS (ESI):  $m/z$  calcd for: 389.1578 [M+H]<sup>+</sup>, found: 389.1586.

**8-(4-nitrobenzylidene)-2-amino-5,6,7,8-tetrahydro-6-methyl-4-(4-nitrophenyl)-1,6-naphthyridine-3-carbonitrile (4ax)**

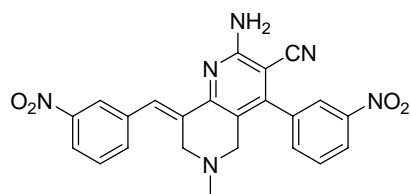


IR (KBr): 3374, 3302, 2210, 1623, 1595, 1558, 1501, 1488, 1451, 1429, 1343, 1266, 1109, 1013, 925, 861, 850, 817, 785, 724 cm<sup>-1</sup>.

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): 8.39 (d, 2H,  $J = 8.4$  Hz, ArH), 8.28 (d, 2H,  $J = 8.8$  Hz, ArH), 8.03 (s, 1H, =CH), 7.74 (d, 2H,  $J = 8.8$  Hz, ArH), 7.66 (d, 2H,  $J = 8.4$  Hz, ArH), 6.95 (s, 2H, NH<sub>2</sub>), 3.60 (s, 2H, CH<sub>2</sub>), 3.15 (s, 2H, CH<sub>2</sub>), 2.23 (s, 3H, CH<sub>3</sub>).

HRMS (ESI):  $m/z$  calcd for: 443.1468 [M+H]<sup>+</sup>, found: 443.1491.

**8-(3-nitrobenzylidene)-2-amino-5,6,7,8-tetrahydro-6-methyl-4-(3-nitrophenyl)-1,6-naphthyridine-3-carbonitrile (4ay)**



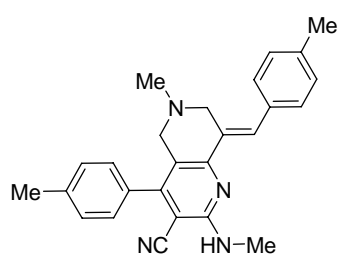
IR (KBr): 3379, 3284, 2212, 1625, 1556, 1526, 1449, 1352, 1264, 1097, 916, 859, 822, 808, 743, 717, 701  $\text{cm}^{-1}$ .

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 8.39 (d, 1H,  $J = 8.0$  Hz, ArH), 8.29 (s, 1H, ArH), 8.20 (d, 1H,  $J = 8.4$  Hz, ArH), 8.15 (s, 1H, ArH), 8.03 (s, 1H, =CH), 7.92-7.84 (m, 3H, ArH), 7.75 (t, 1H,  $J = 8.4$  Hz, ArH), 6.90 (s, 2H,  $\text{NH}_2$ ), 3.61 (s, 2H,  $\text{CH}_2$ ), 3.17 (d, 2H,  $J = 7.2$  Hz,  $\text{CH}_2$ ), 2.23 (s, 3H,  $\text{CH}_3$ ).

$^{13}\text{C}$  NMR (100 MHz,  $\text{DMSO-}d_6$ , 25  $^\circ\text{C}$ ) ( $\delta$ , ppm): 157.85, 151.94, 150.56, 147.95, 147.87, 137.52, 136.60, 135.70, 135.09, 134.90, 130.58, 130.17, 126.02, 124.07, 123.71, 123.23, 122.33, 117.10, 116.25, 89.60, 54.78, 54.23, 44.99.

HRMS (ESI):  $m/z$  calcd for: 443.1468  $[\text{M}+\text{H}]^+$ , found: 443.1460.

**8-(4-Methylbenzylidene)-6-methyl-4-(4-methylphenyl)-2-methylamino-5,6,7,8-tetrahydro-1,6-naphthyridine-3-carbonitrile (4az)**

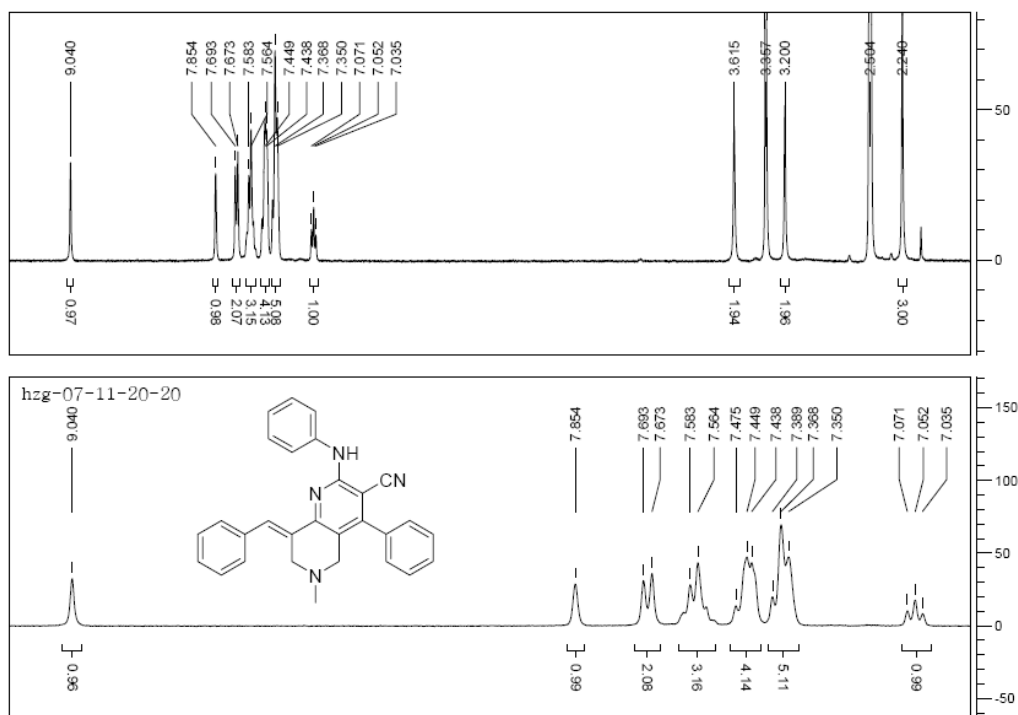


IR (KBr): 3394, 3025, 2879, 2205, 1686, 1658, 1571, 1552, 1454, 1362, 1260, 902, 823  $\text{cm}^{-1}$ .

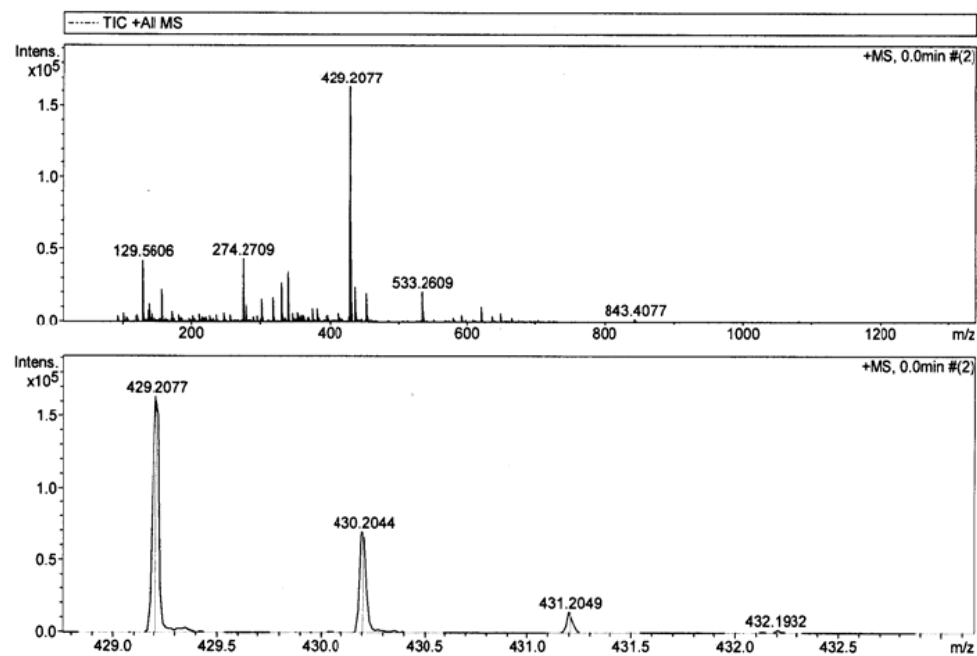
$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ): 7.74 (s, 1H, NH), 7.36 (d, 2H,  $J = 8.0$  Hz, ArH), 7.30-7.29 (m, 7H, ArH, 6H and =CH, 1H), 3.52 (s, 2H,  $\text{CH}_2$ ), 3.30 (s, 3H,  $\text{NCH}_3$ ), 3.04 (s, 2H,  $\text{CH}_2$ ), 2.40 (s, 3H,  $\text{CH}_3$ ), 2.35 (s, 3H,  $\text{CH}_3$ ), 2.20 (s, 3H,  $\text{CH}_3$ ).



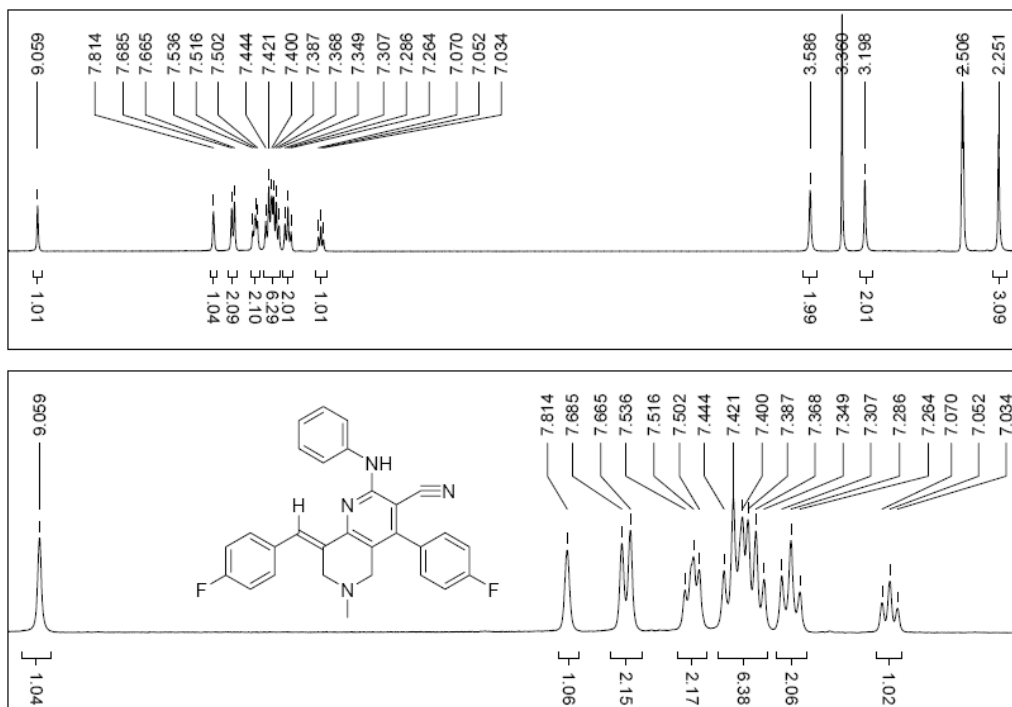
## Copies of spectra for compounds 4



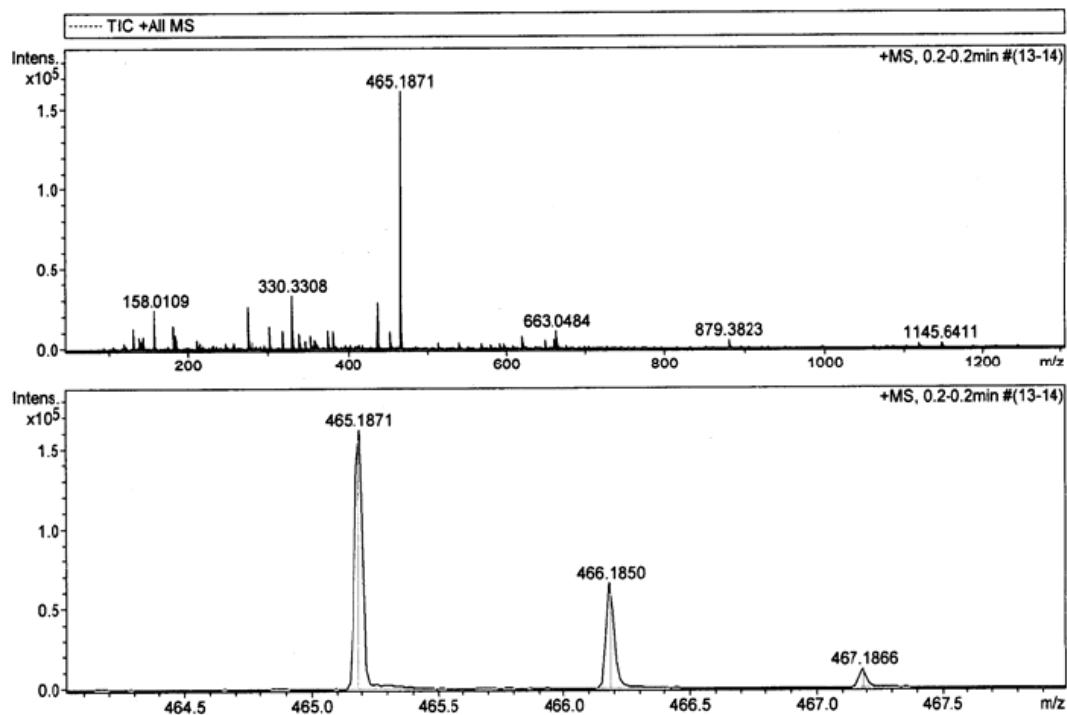
<sup>1</sup>H NMR Spectrum of Compound 4a



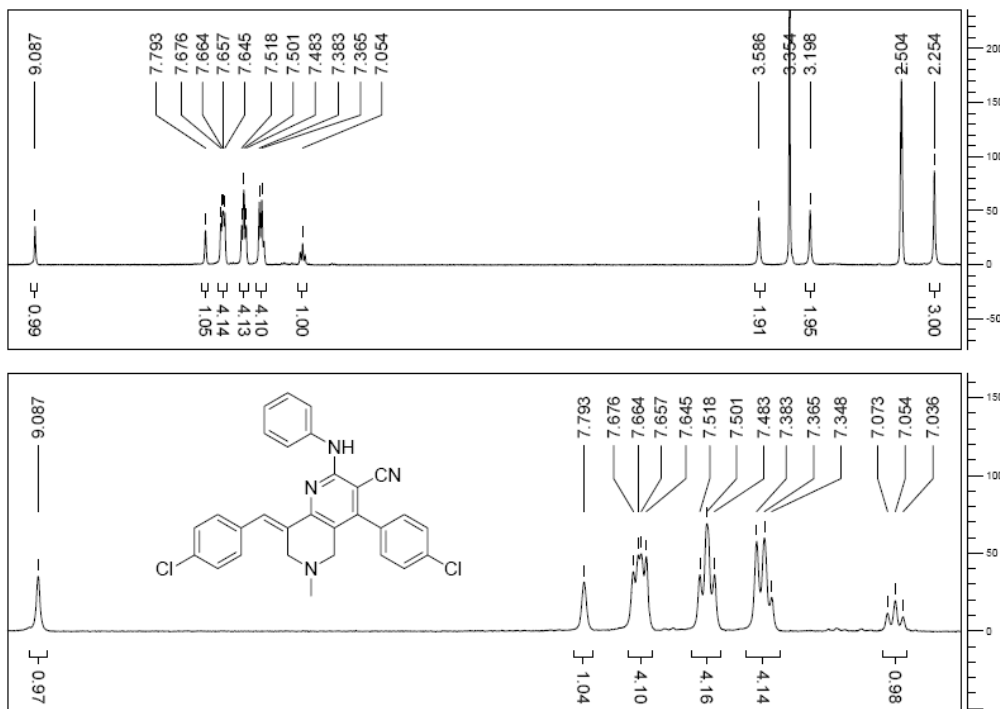
HRMS Spectrum of Compound 4a



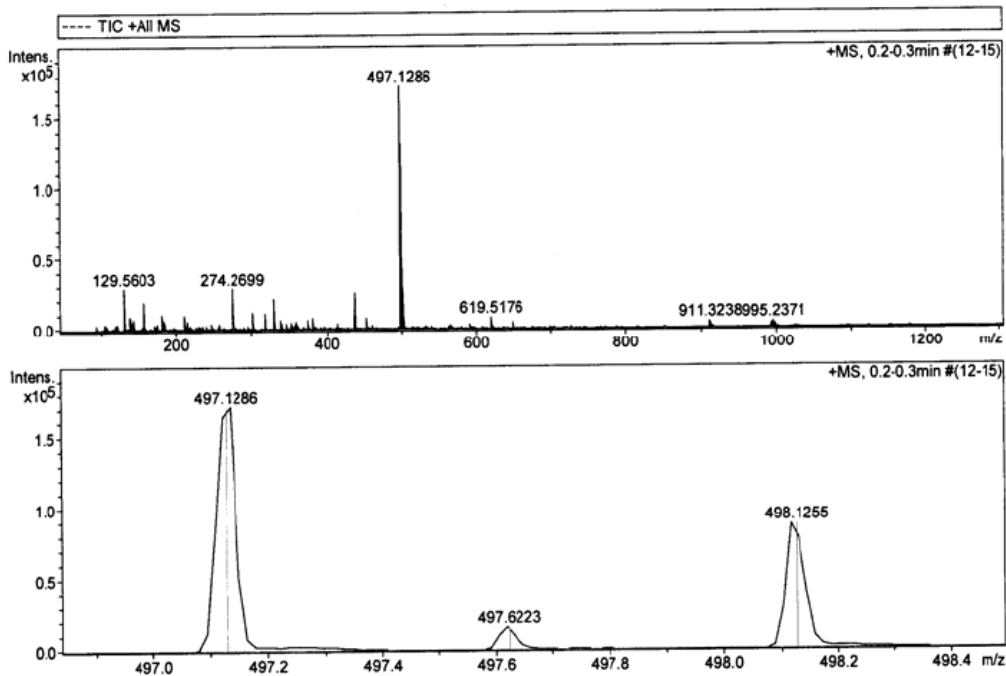
<sup>1</sup>H NMR Spectrum of Compound 4b



HRMS Spectrum of Compound 4b

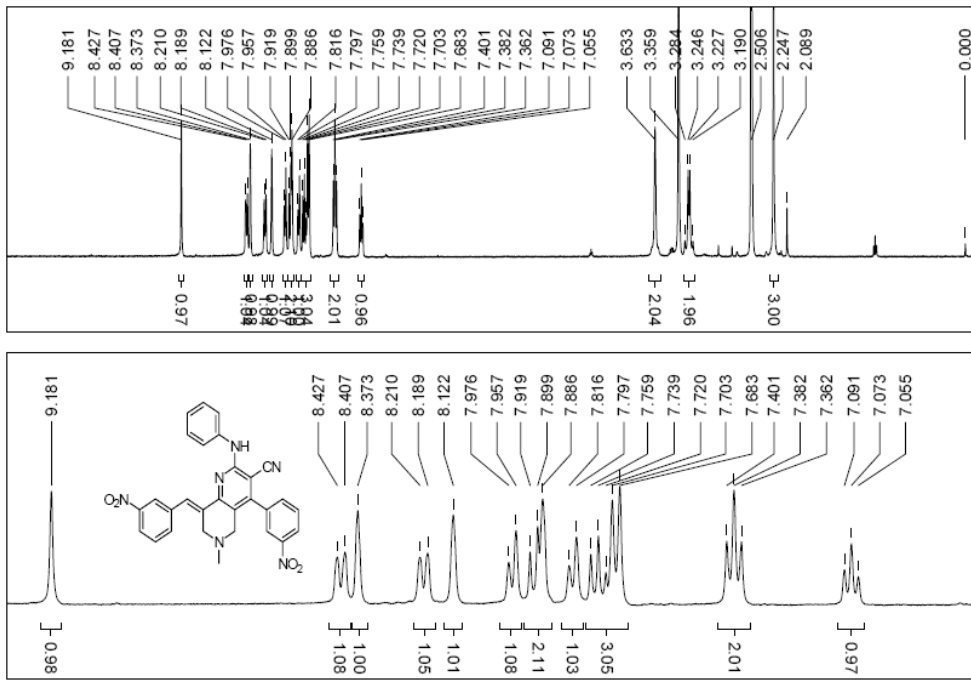


<sup>1</sup>H NMR Spectrum of Compound 4c

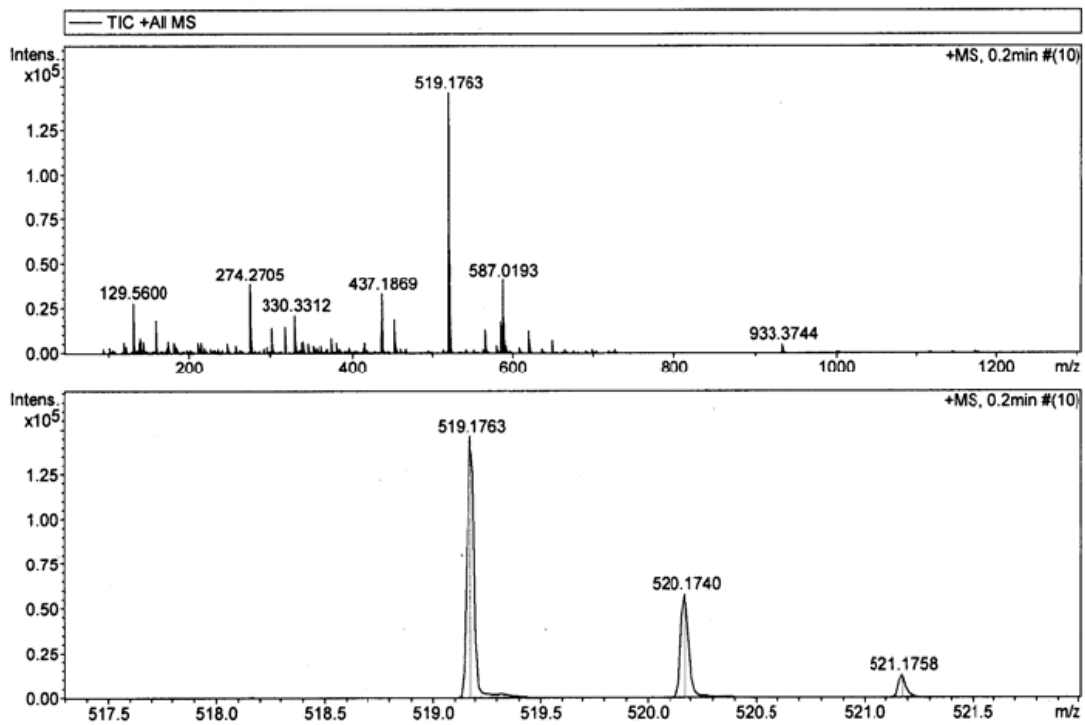


HRMS Spectrum of Compound 4c



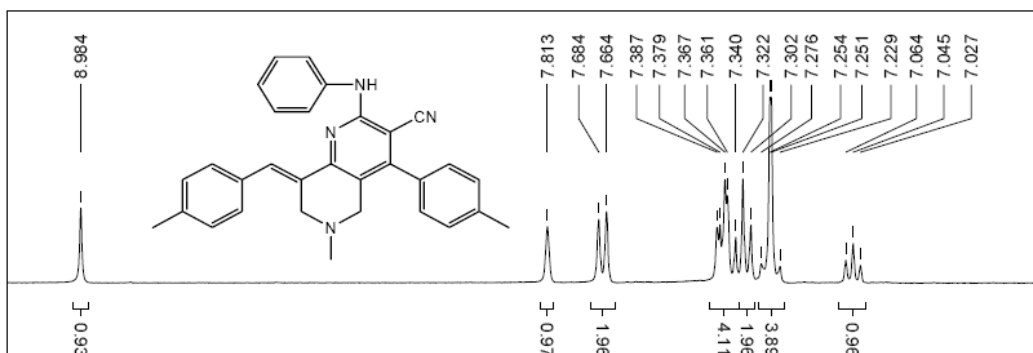
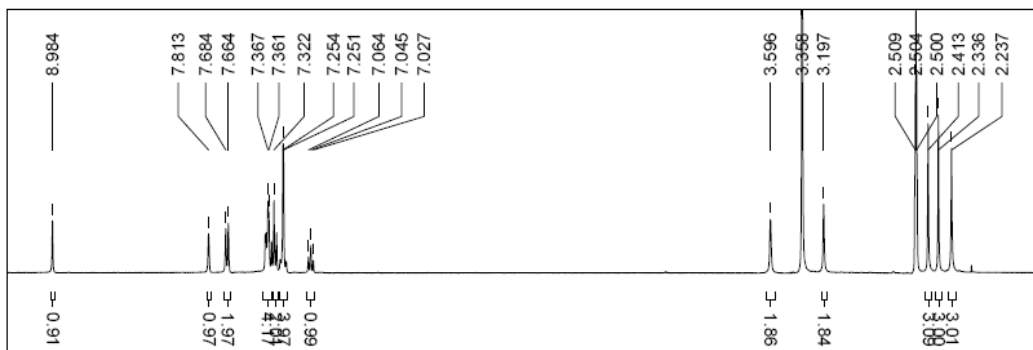


**<sup>1</sup>H NMR Spectrum of Compound 4e**

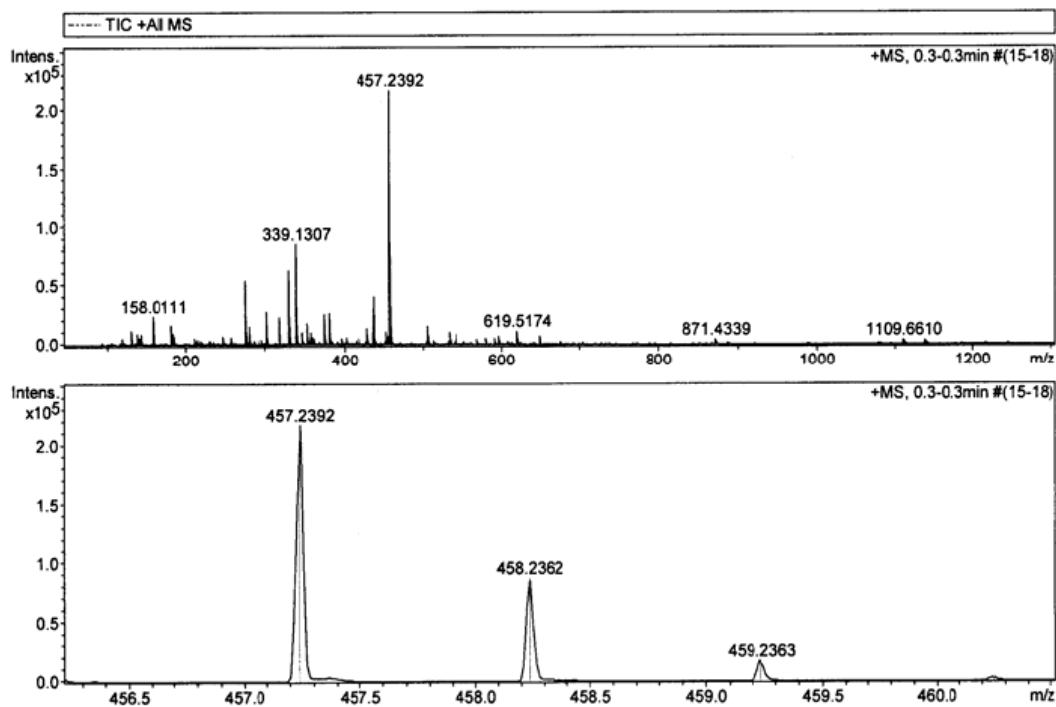


**HRMS Spectrum of Compound 4e**

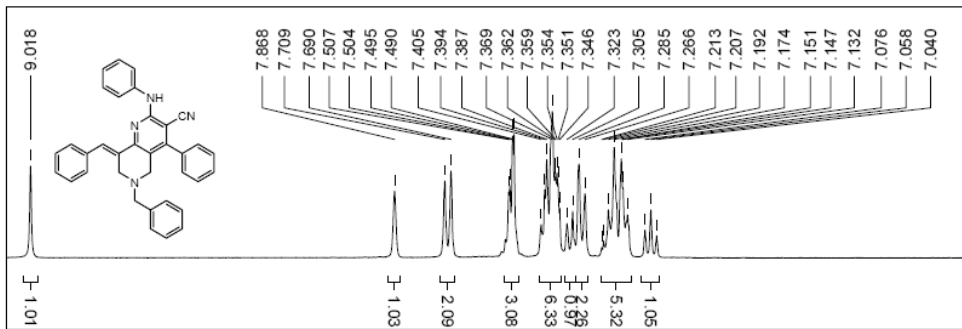
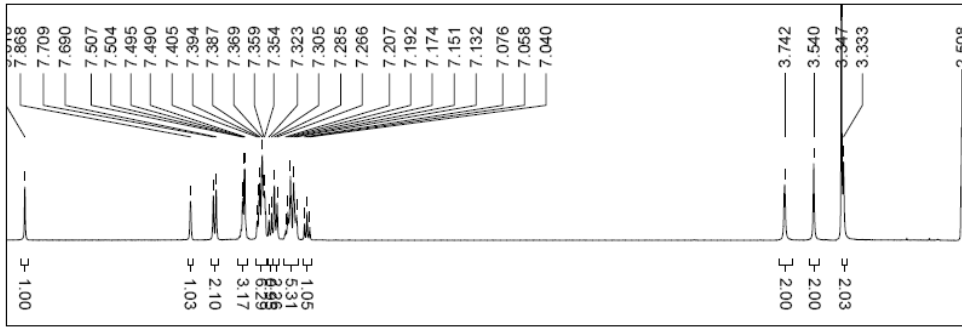




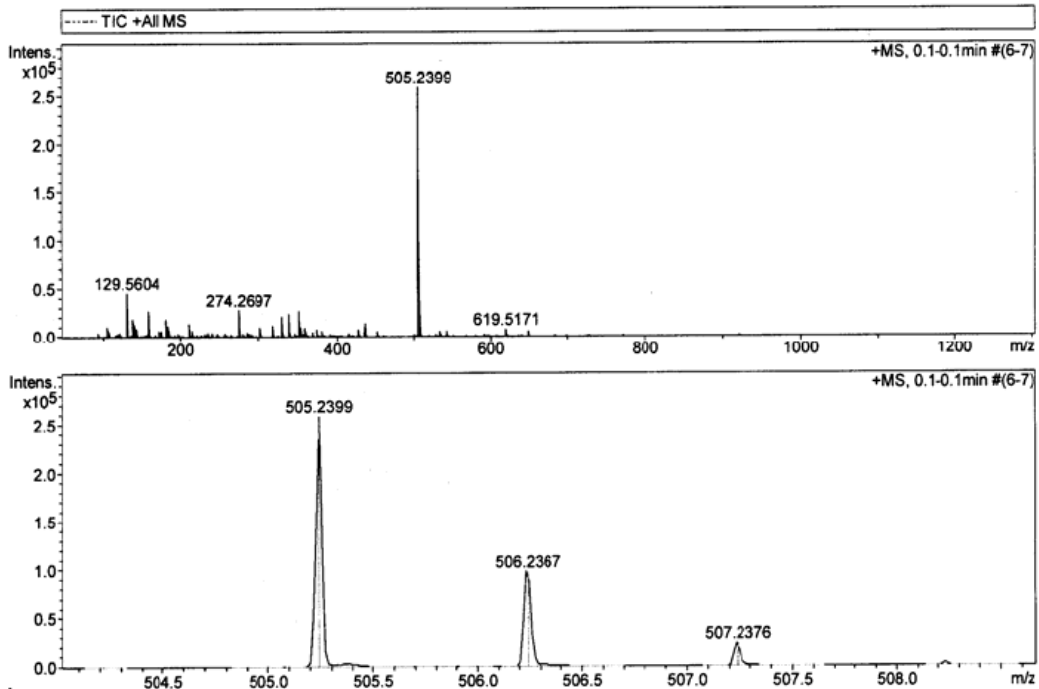
<sup>1</sup>H NMR Spectrum of Compound 4g



HRMS Spectrum of Compound 4g

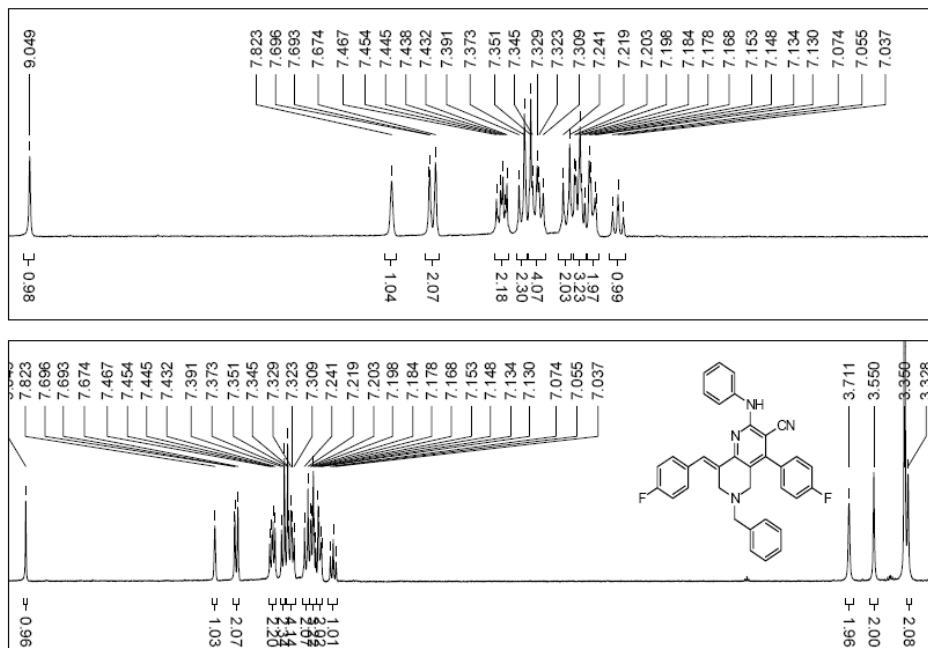


<sup>1</sup>H NMR Spectrum of Compound 4h

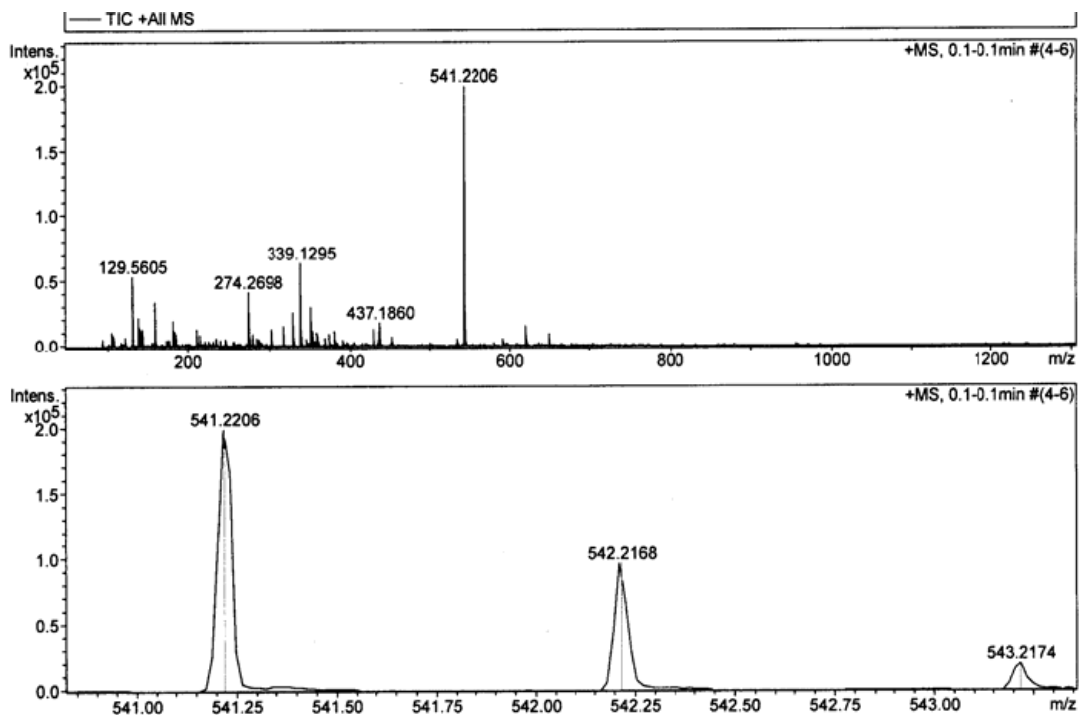


HRMS Spectrum of Compound 4h

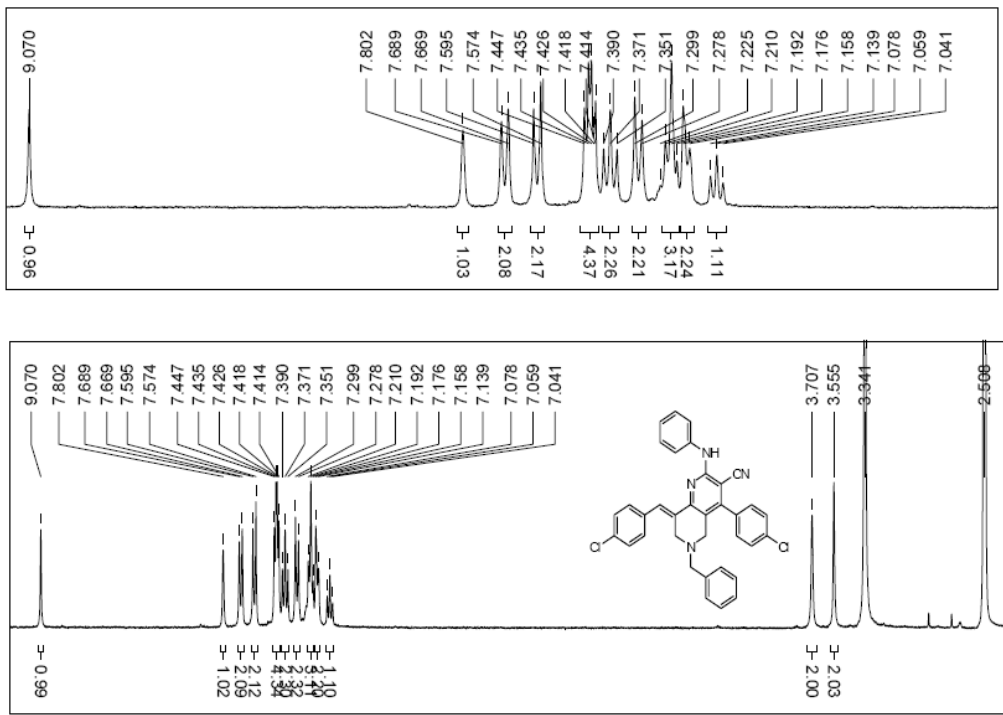




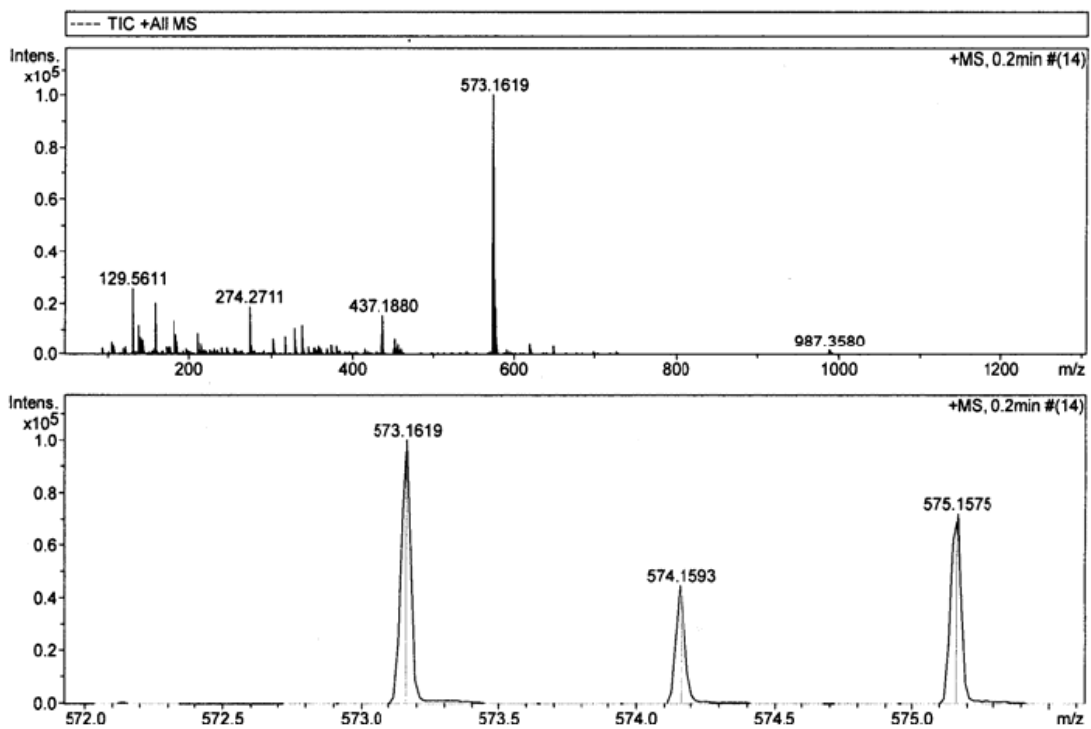
**<sup>1</sup>H NMR Spectrum of Compound 4i**



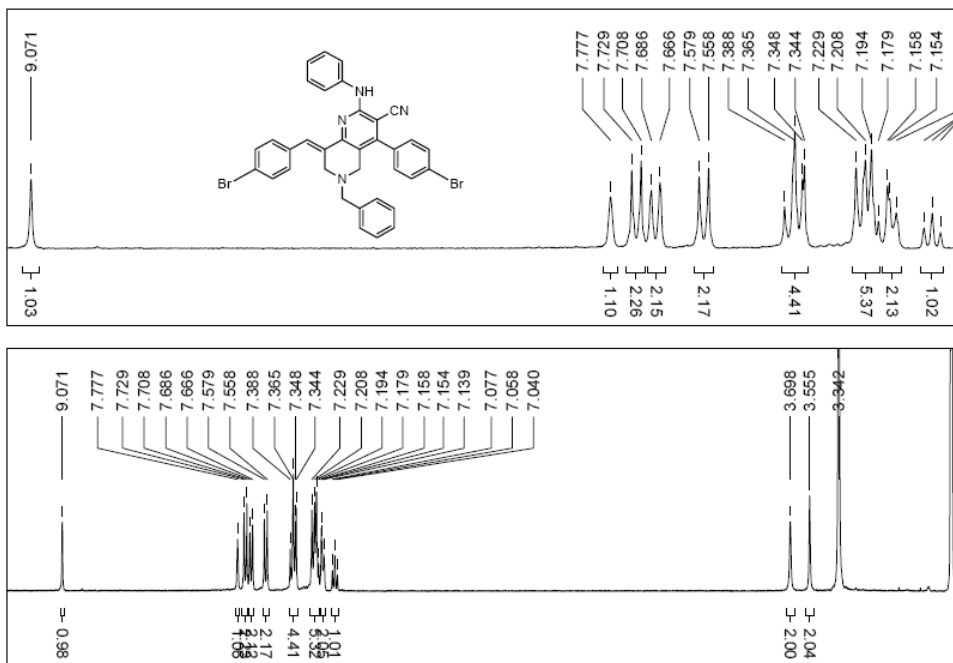
**HRMS Spectrum of Compound 4i**



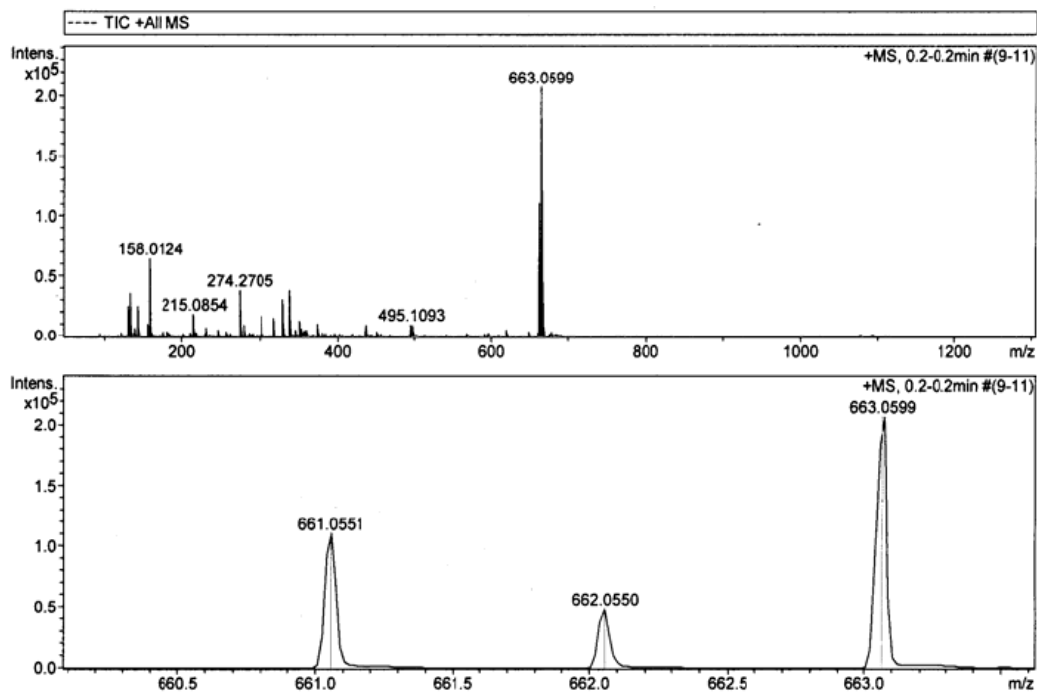
**<sup>1</sup>H NMR Spectrum of Compound 4j**



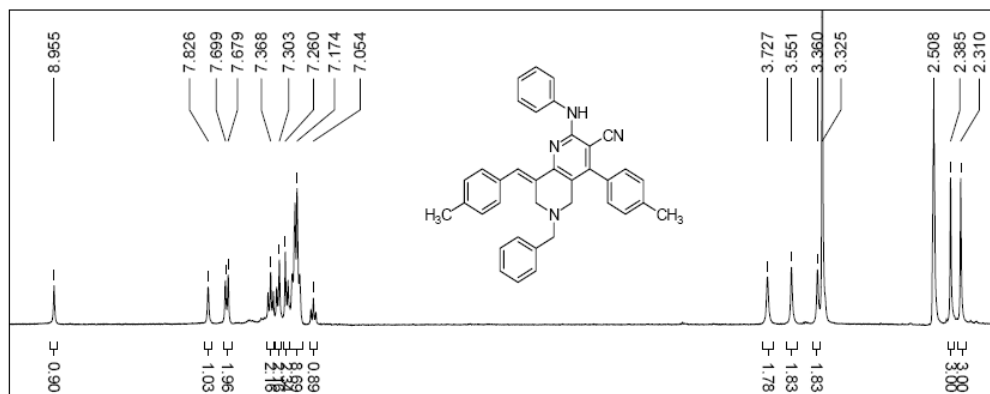
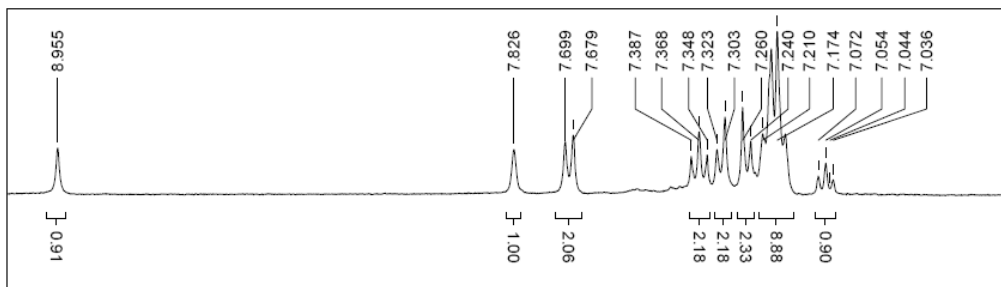
**HRMS Spectrum of Compound 4j**



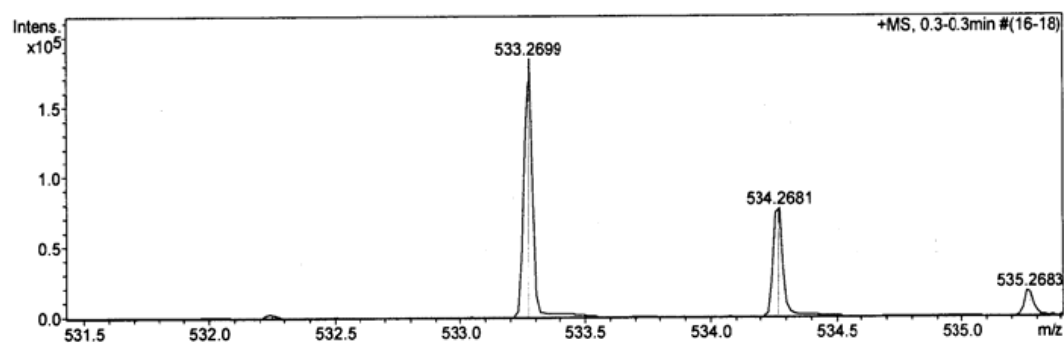
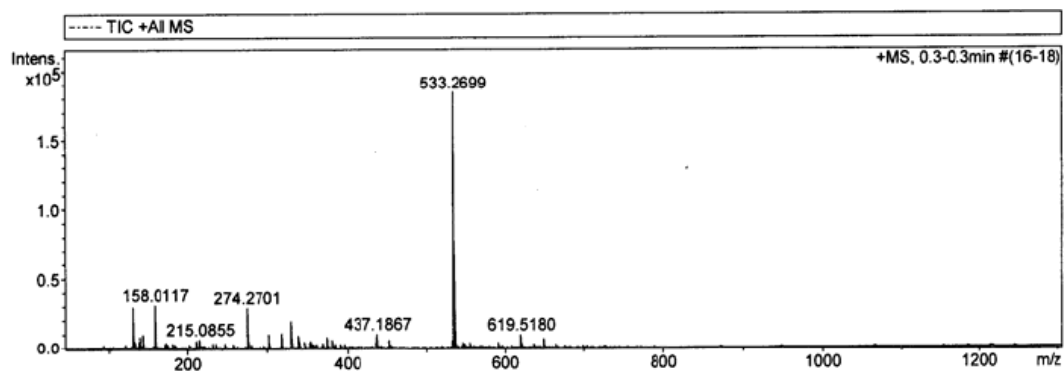
<sup>1</sup>H NMR Spectrum of Compound 4k



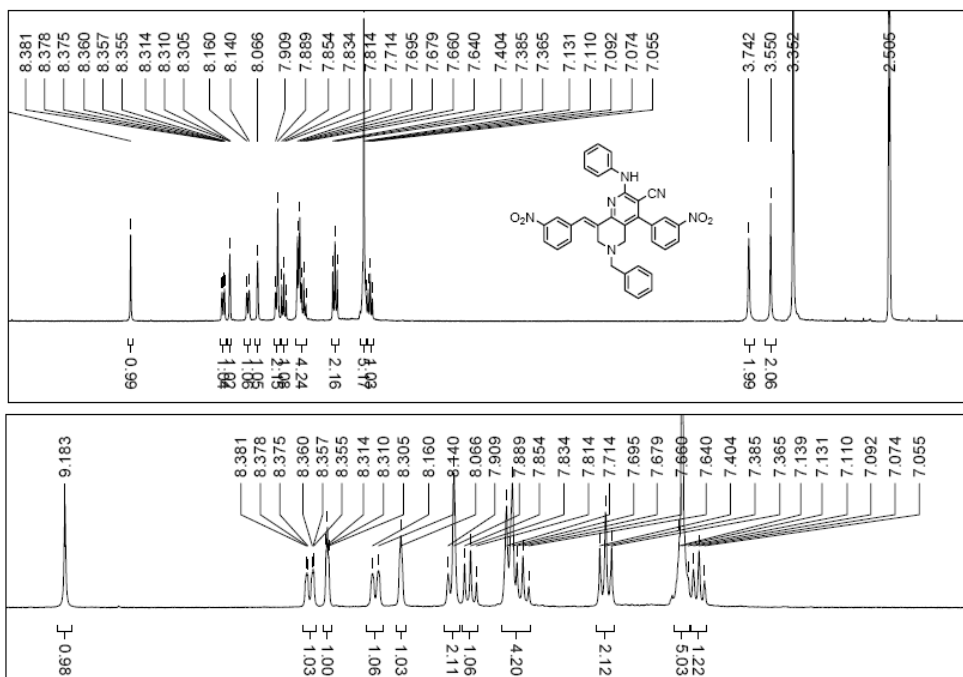
HRMS Spectrum of Compound 4k



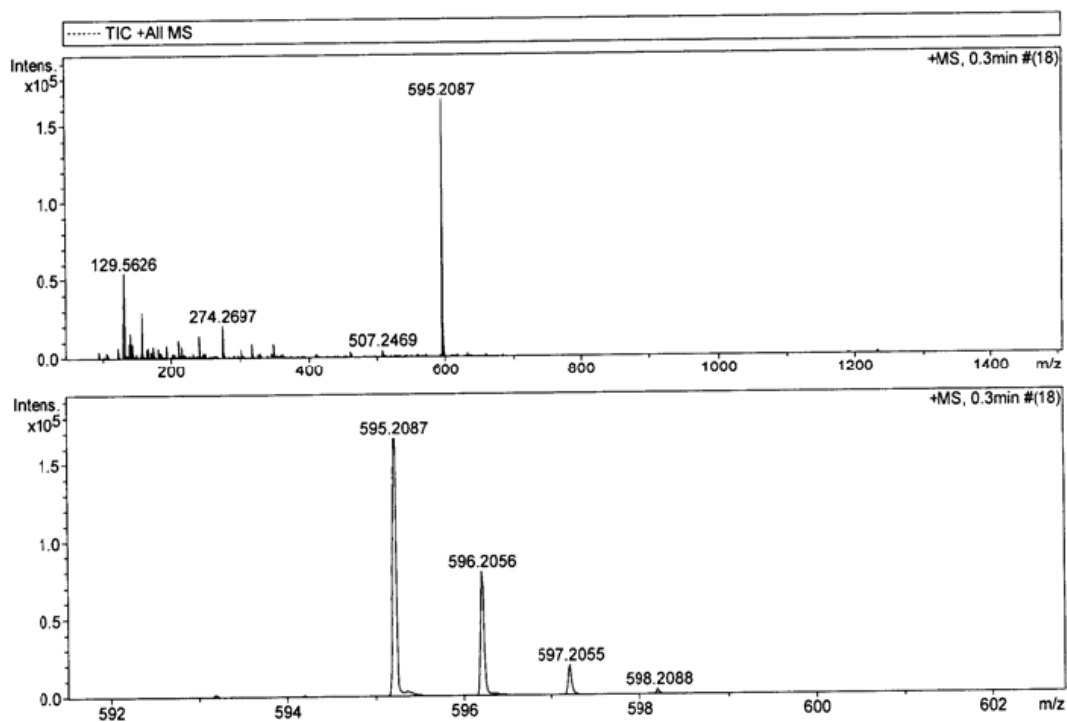
**<sup>1</sup>H NMR Spectrum of Compound 4l**



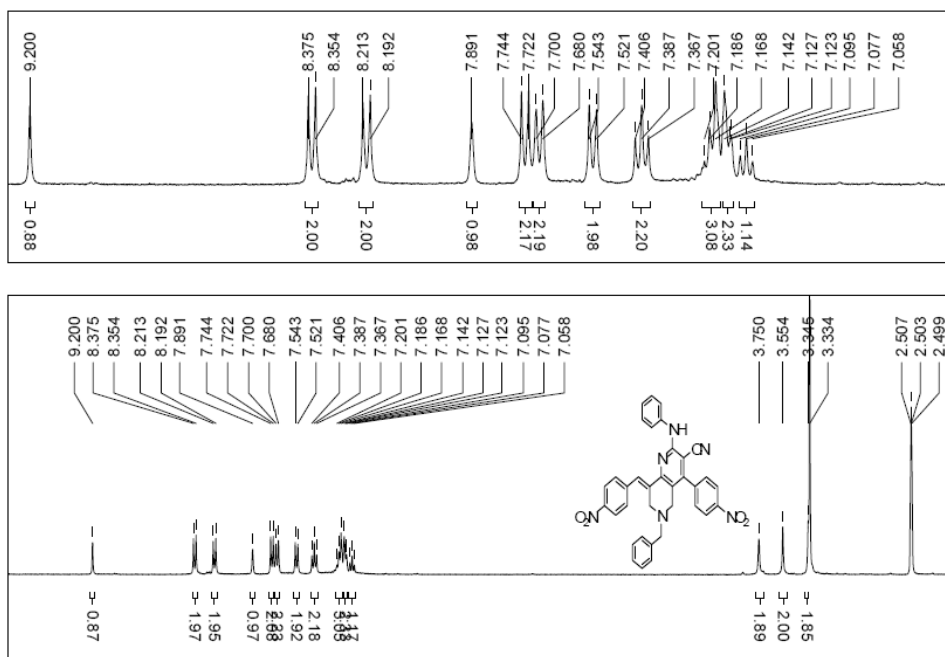
**HRMS Spectrum of Compound 4l**



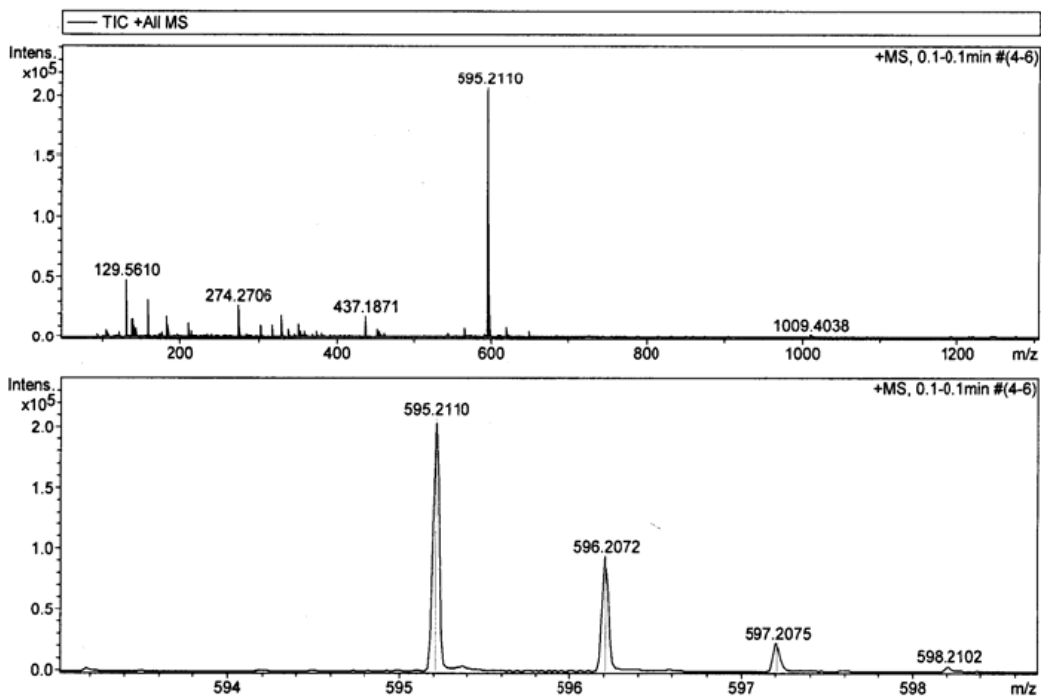
**<sup>1</sup>H NMR Spectrum of Compound 4m**



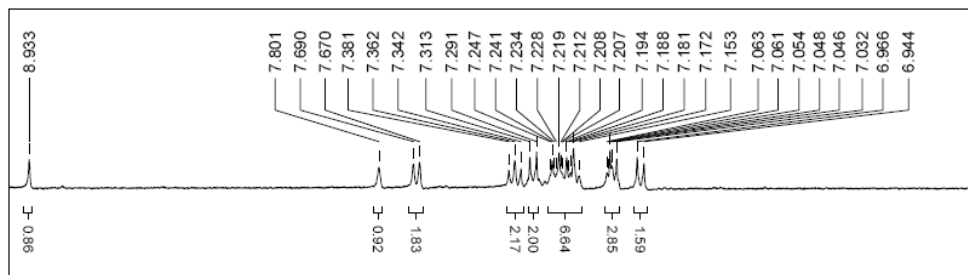
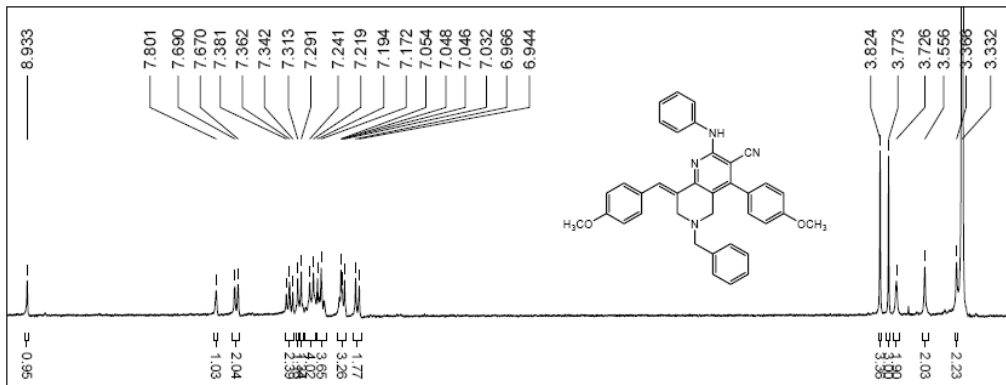
**HRMS Spectrum of Compound 4m**



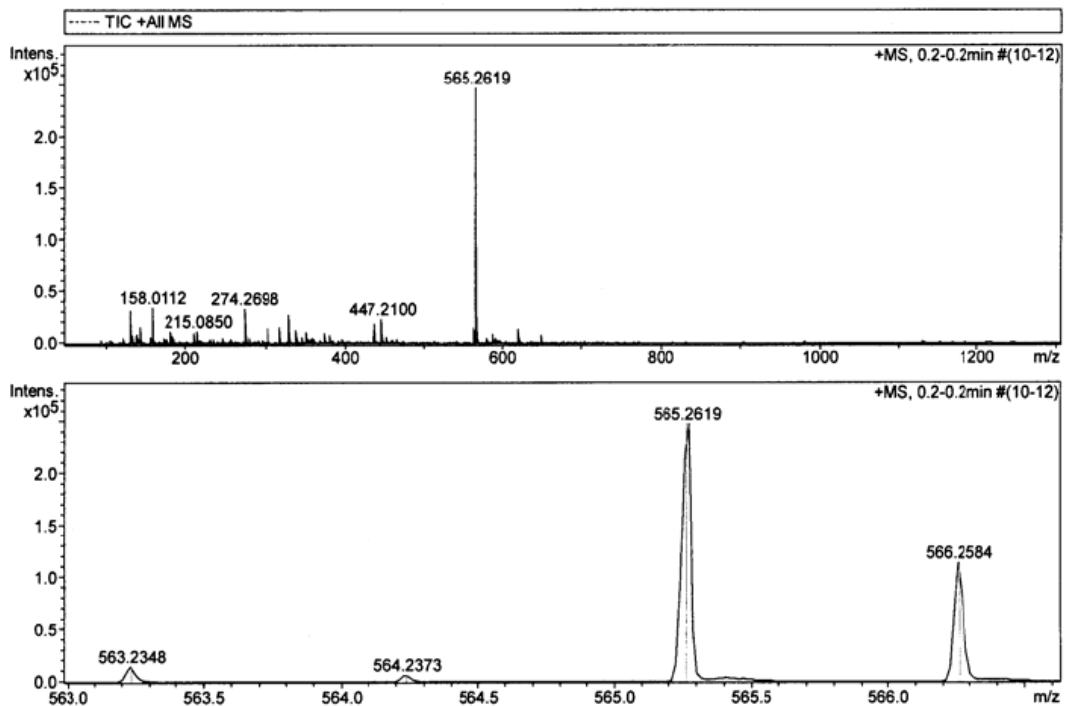
**<sup>1</sup>H NMR Spectrum of Compound 4n**



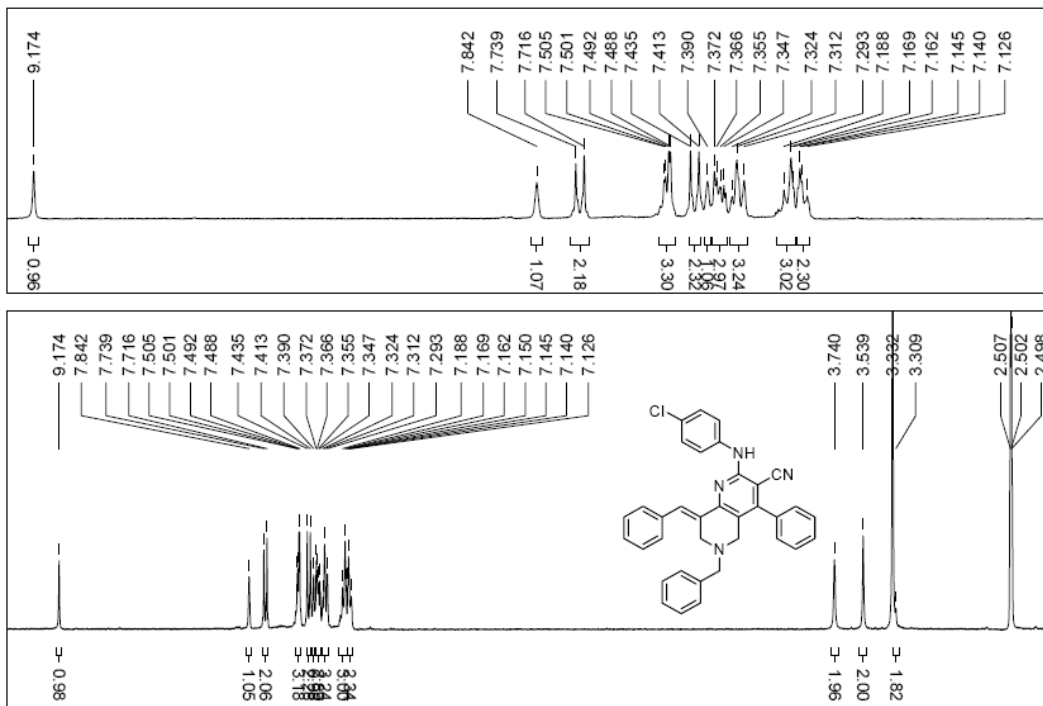
**HRMS Spectrum of Compound 4n**



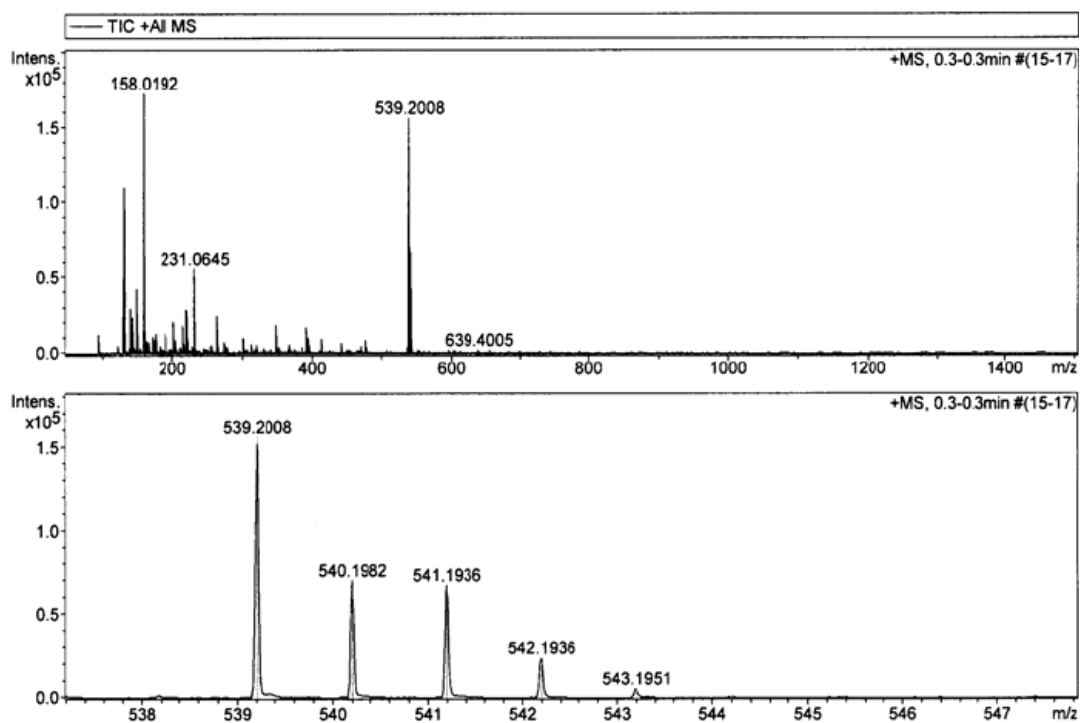
**<sup>1</sup>H NMR Spectrum of Compound 40**



**HRMS Spectrum of Compound 40**

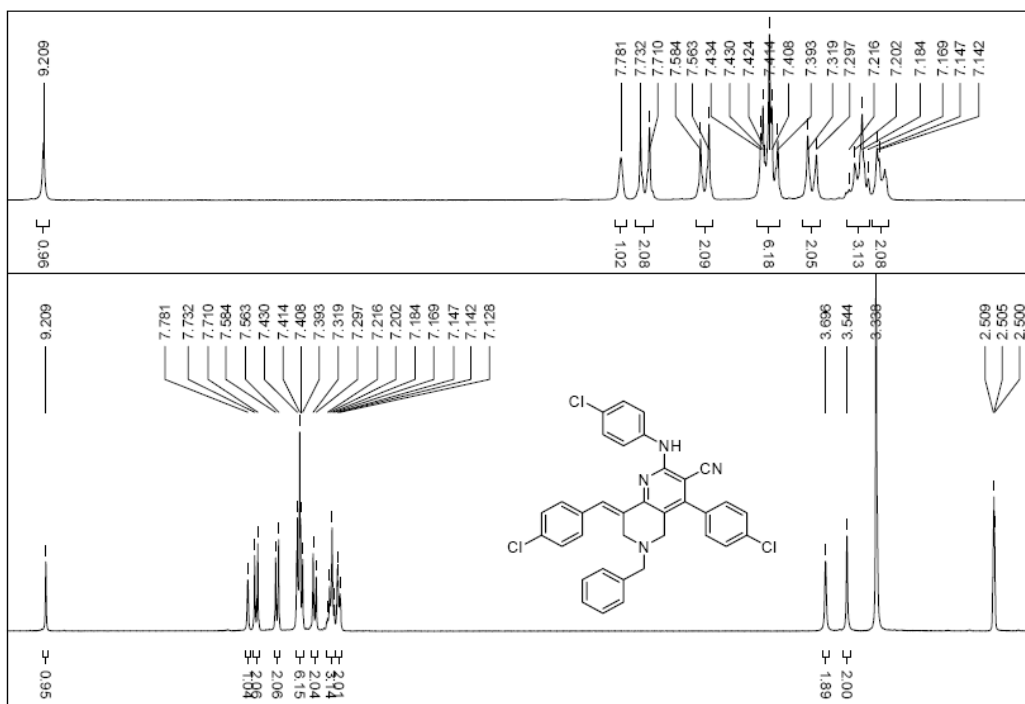


**<sup>1</sup>H NMR Spectrum of Compound 4p**

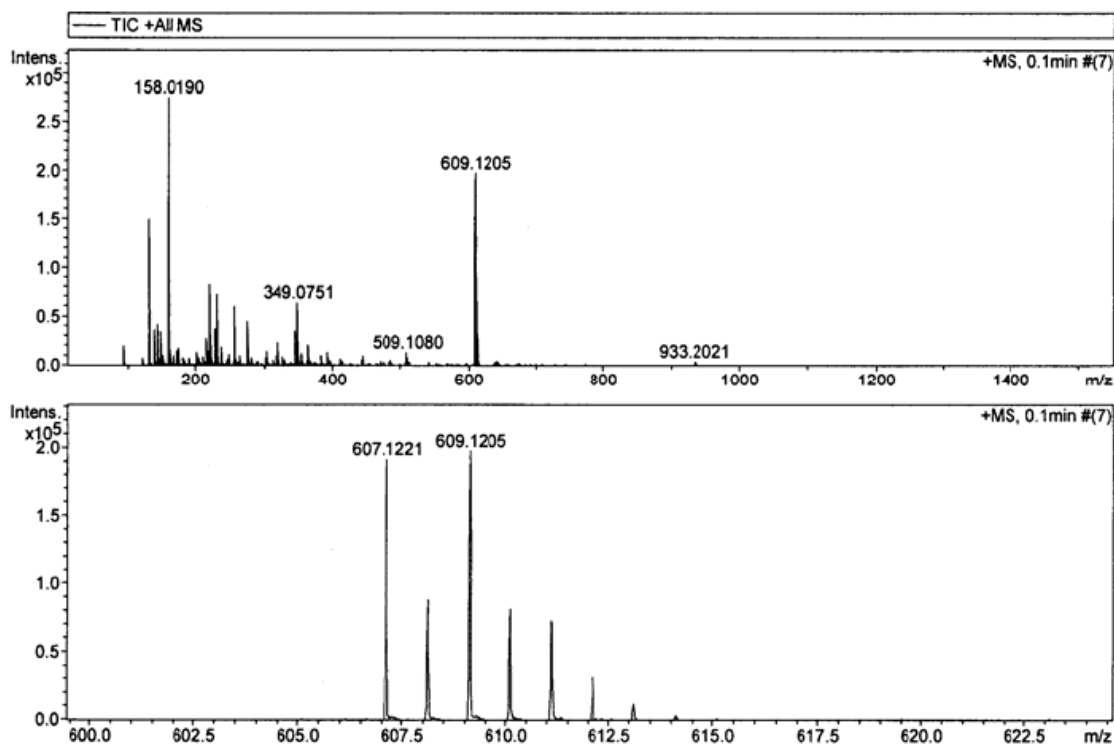


**HRMS Spectrum of Compound 4p**

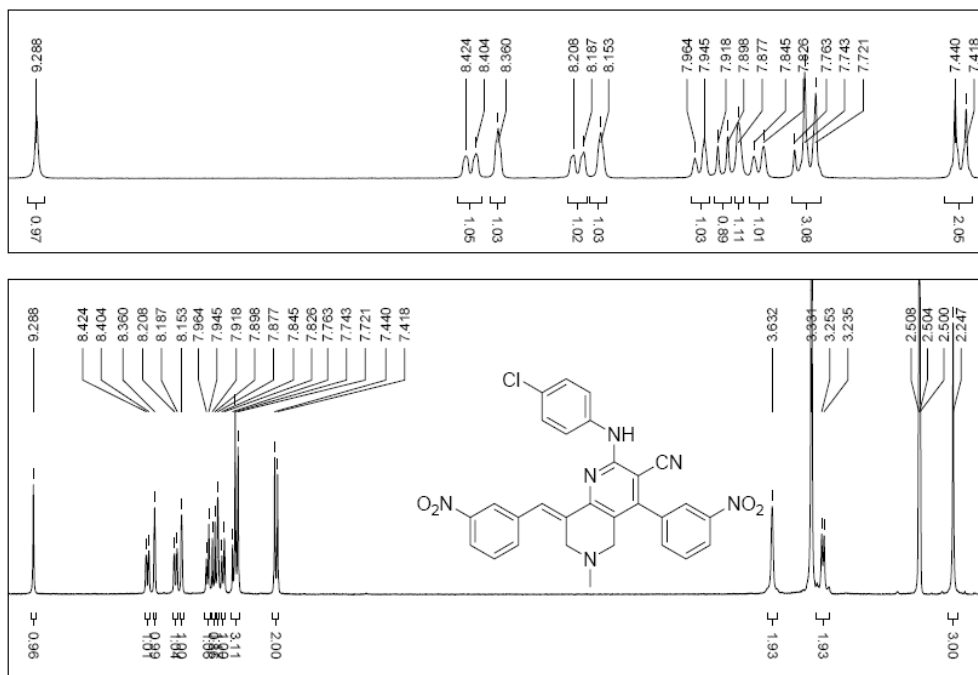




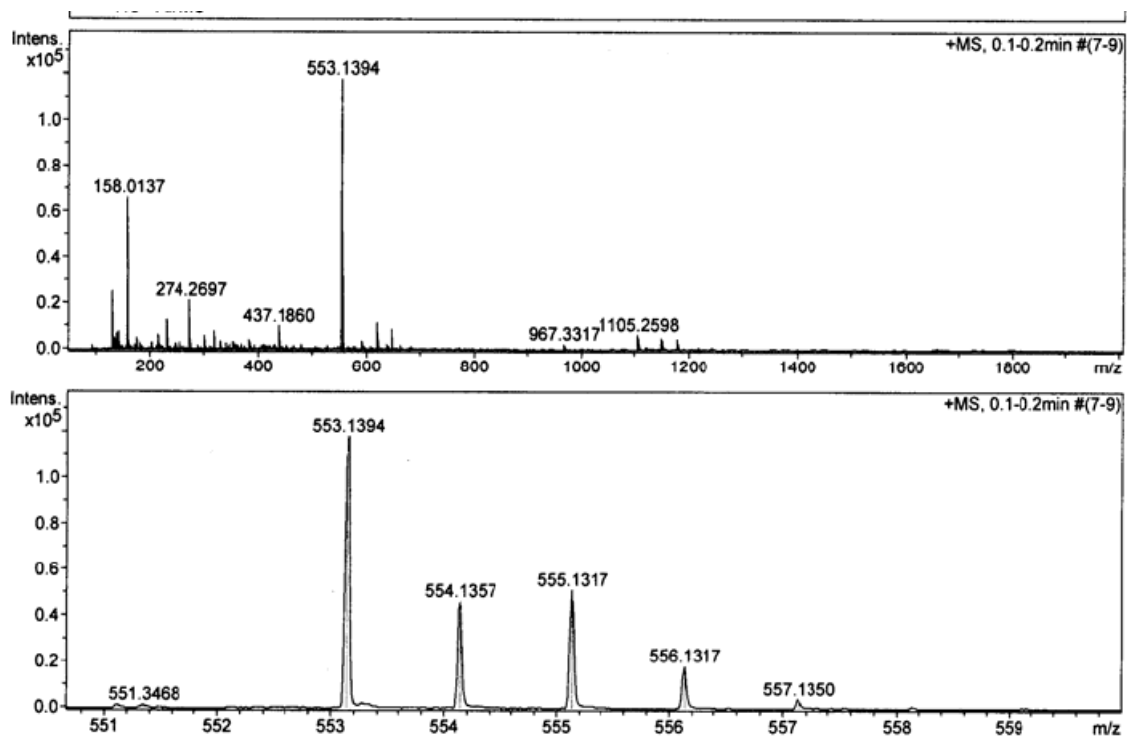
<sup>1</sup>H NMR Spectrum of Compound 4q



HRMS Spectrum of Compound 4q

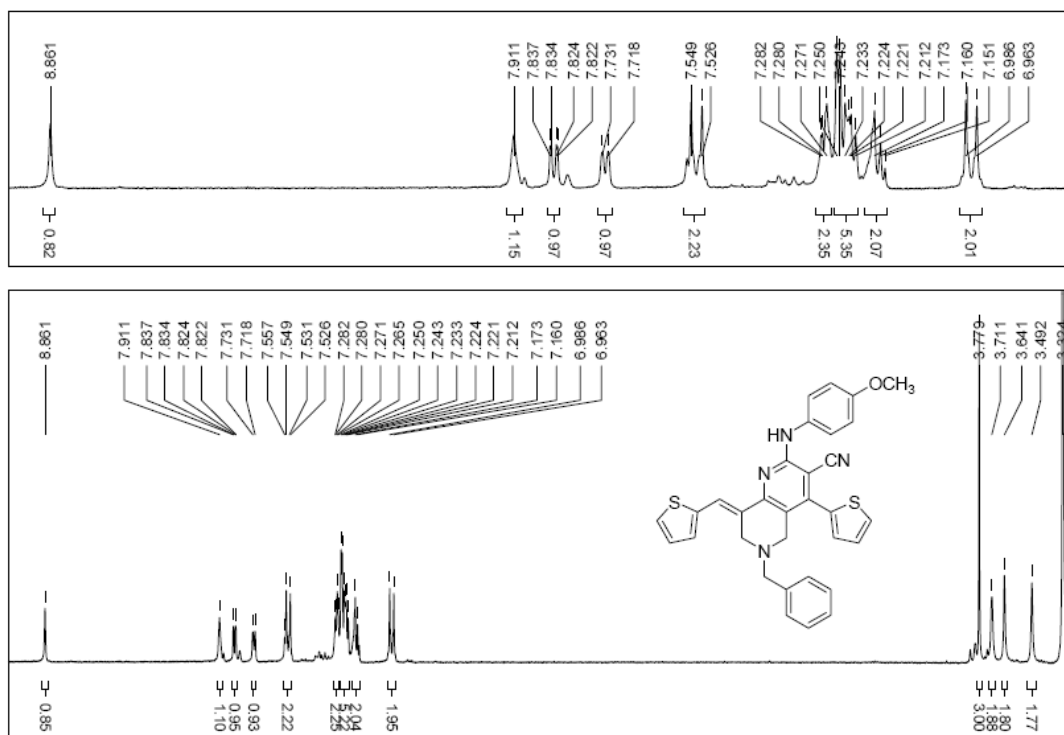


<sup>1</sup>H NMR Spectrum of Compound 4r

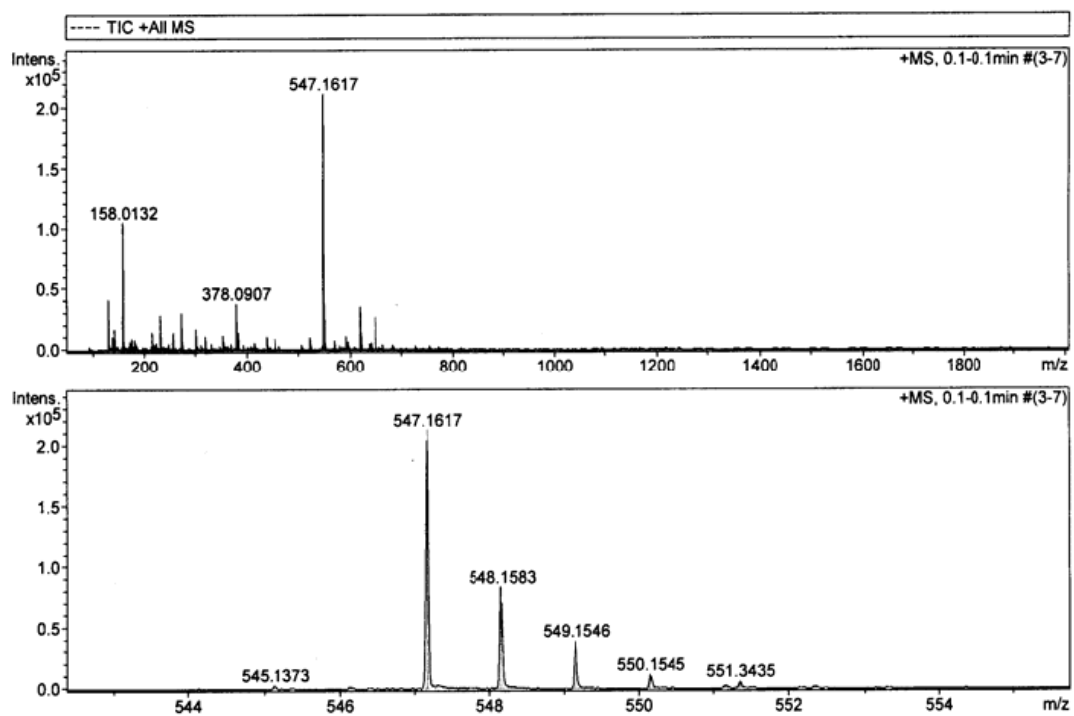


HRMS Spectrum of Compound 4r



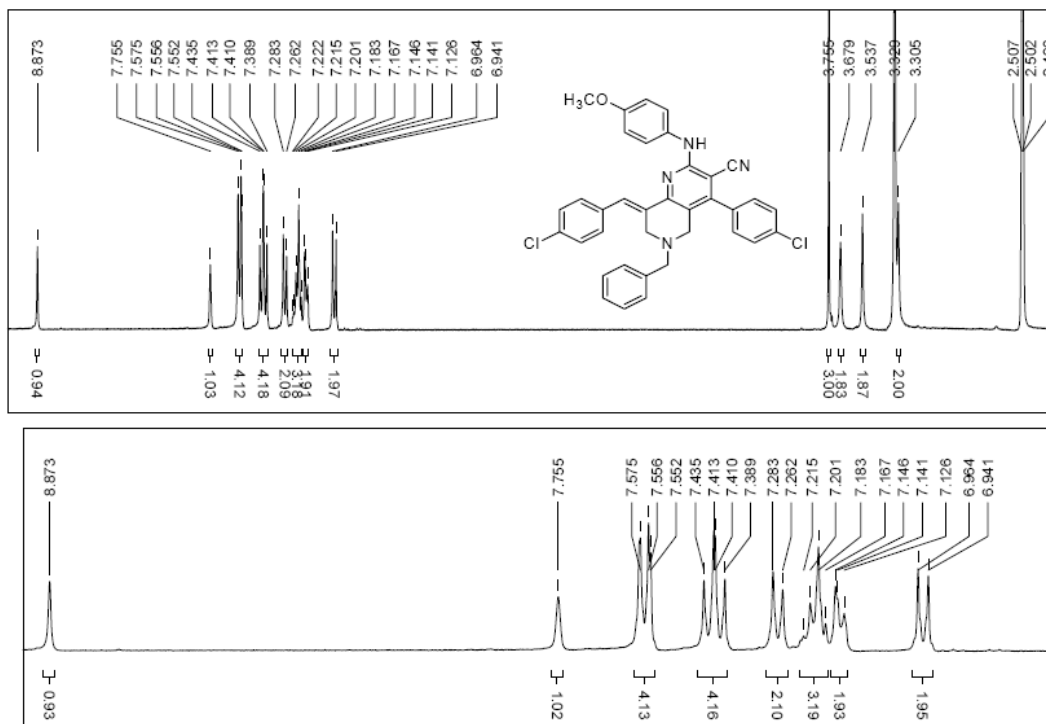


<sup>1</sup>H NMR Spectrum of Compound 4t

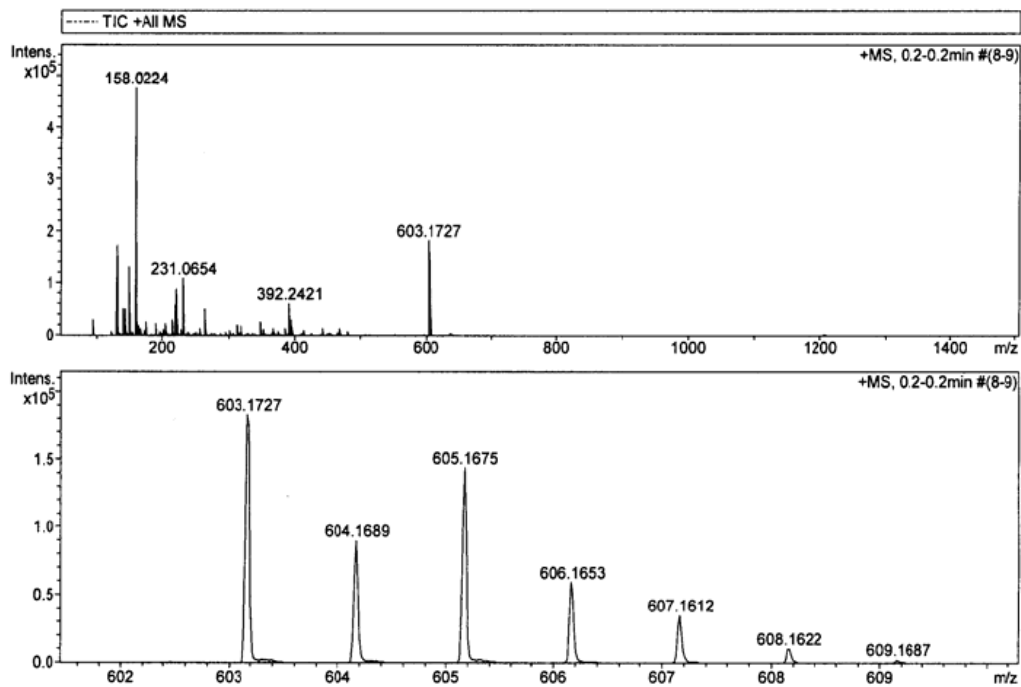


HRMS Spectrum of Compound 4t

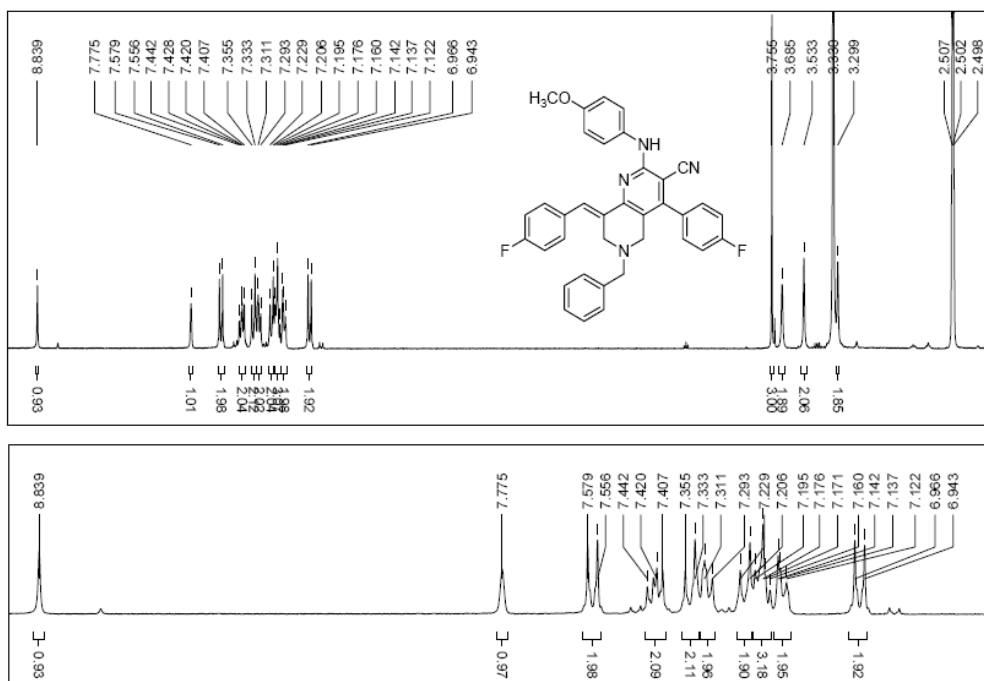




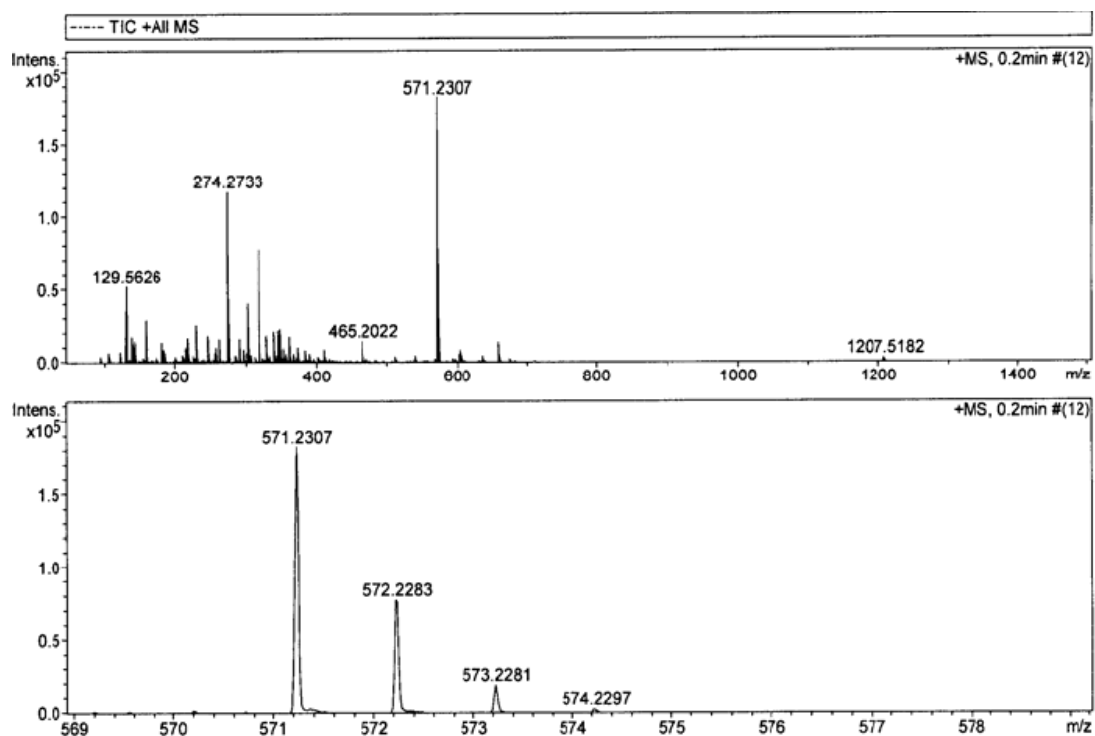
**<sup>1</sup>H NMR Spectrum of Compound 4v**

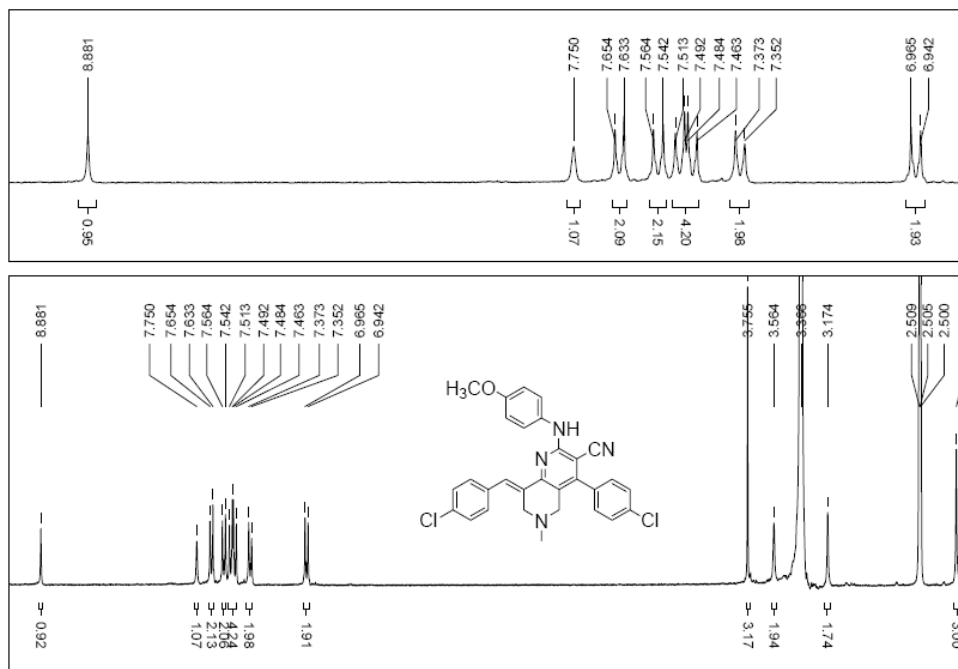


**HRMS Spectrum of Compound 4v**

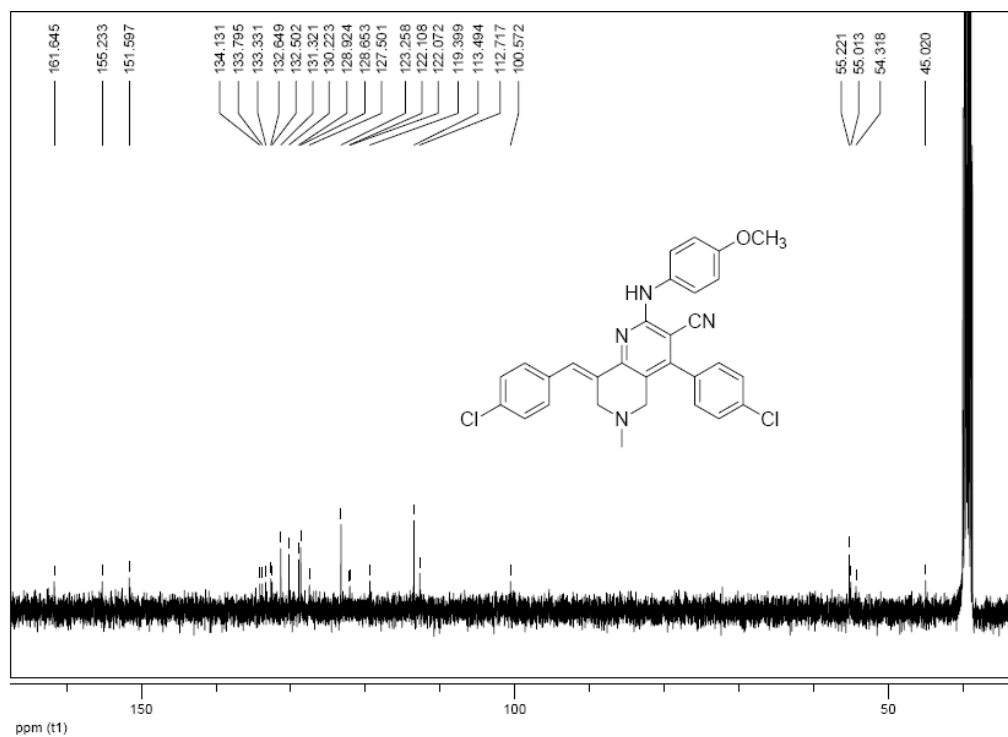


**<sup>1</sup>H NMR Spectrum of Compound 4w**



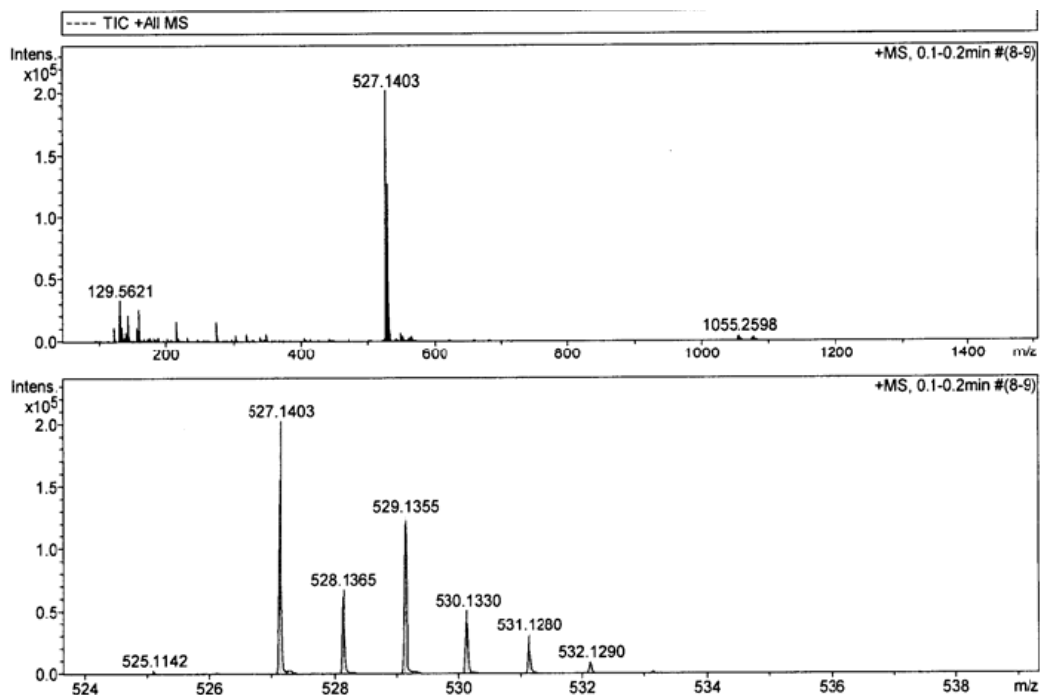


**<sup>1</sup>H NMR Spectrum of Compound 4x**

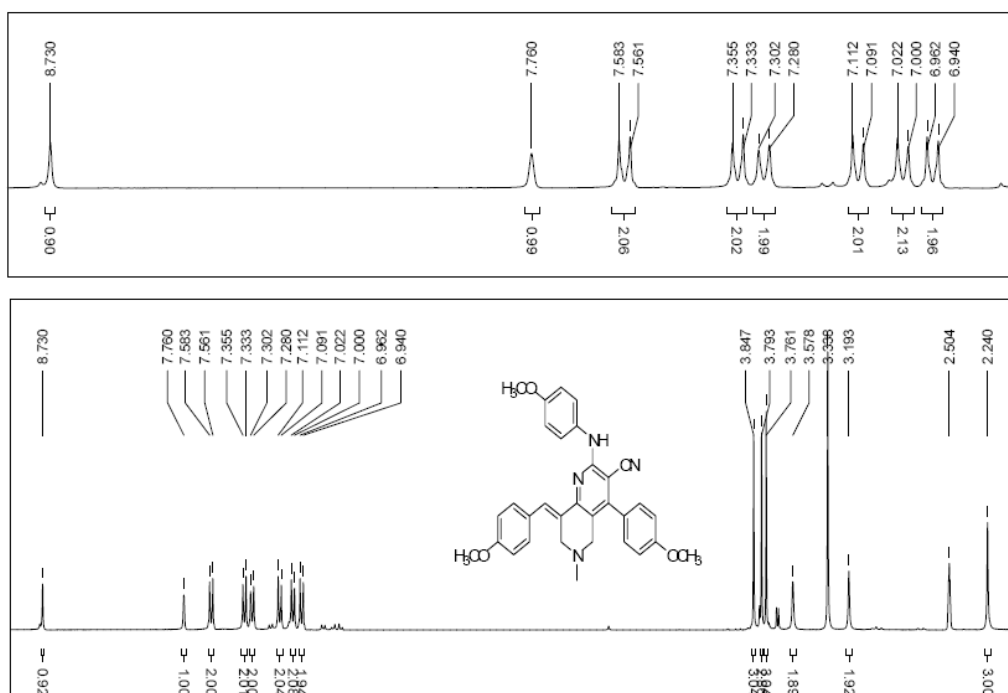


**<sup>13</sup>C NMR Spectrum of Compound 4x**

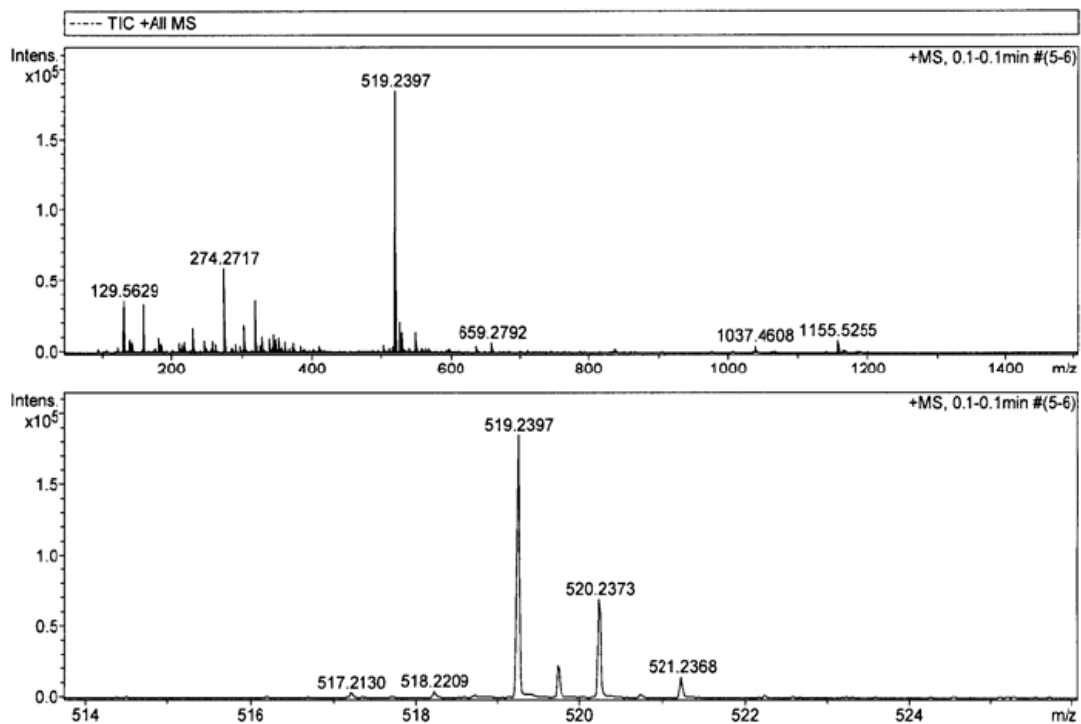




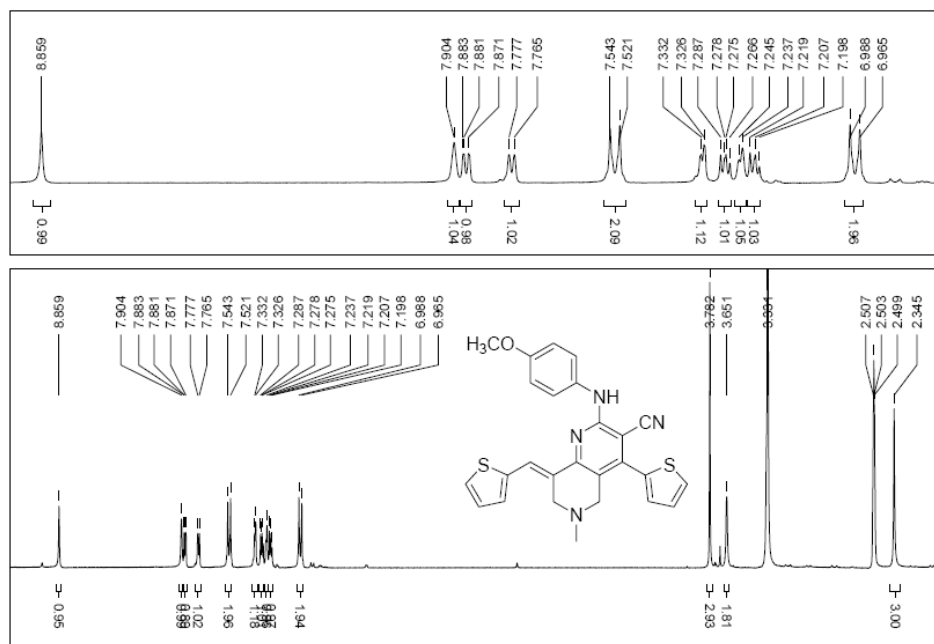
### HRMS Spectrum of Compound 4x



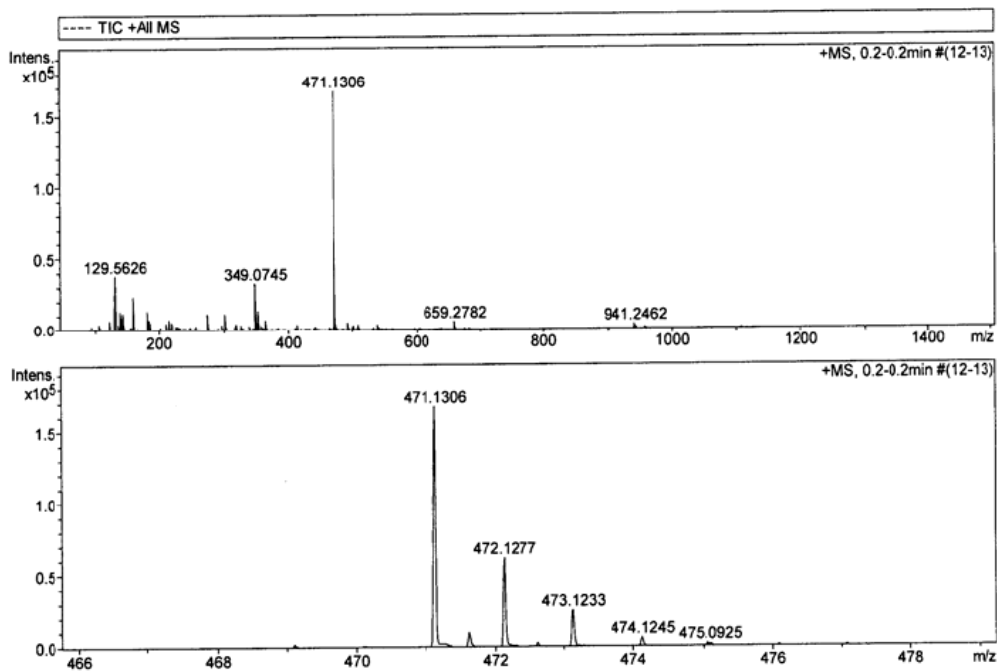
### <sup>1</sup>H NMR Spectrum of Compound 4y



HRMS Spectrum of Compound 4y



<sup>1</sup>H NMR Spectrum of Compound 4z

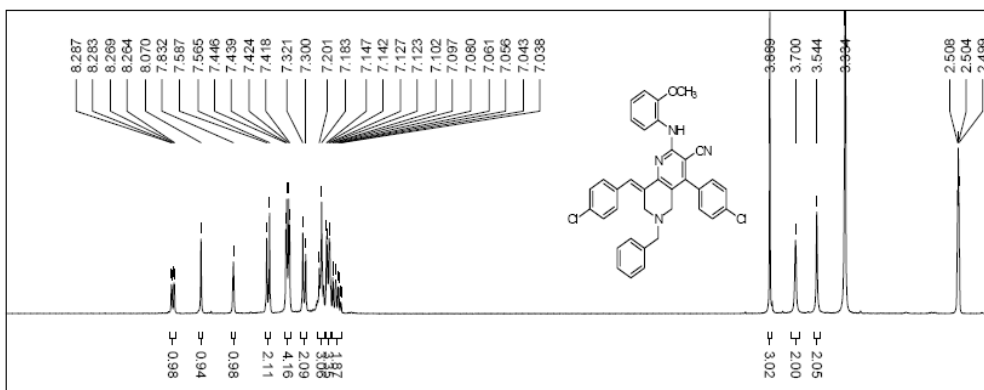
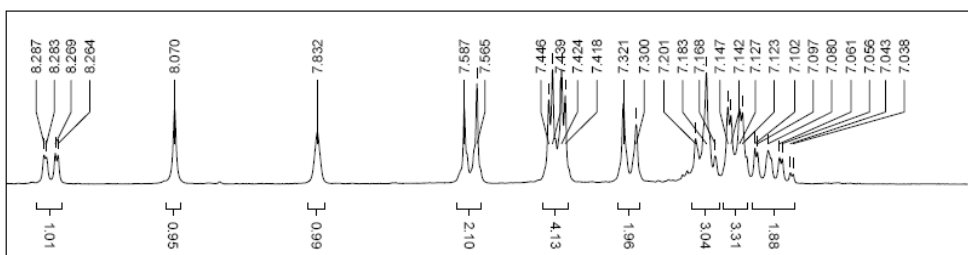


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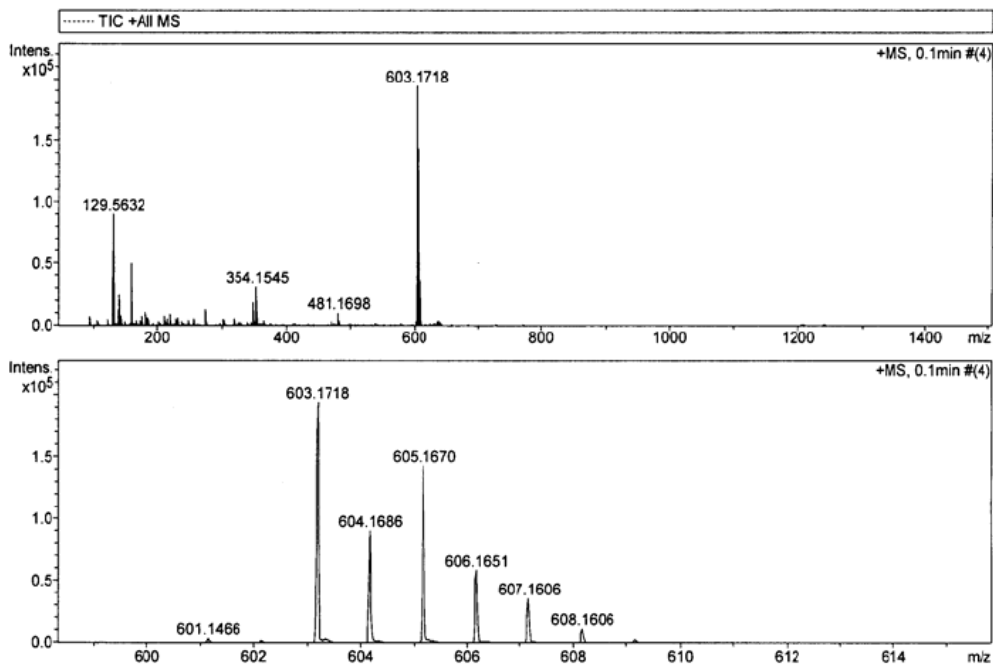
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### HRMS Spectrum of Compound 4z



### <sup>1</sup>H NMR Spectrum of Compound 4aa

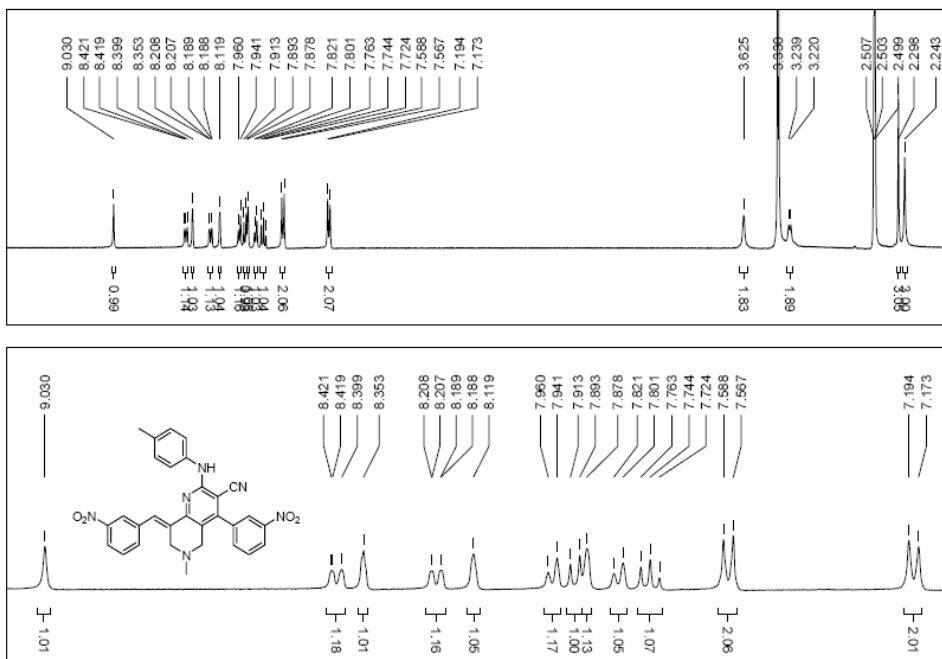


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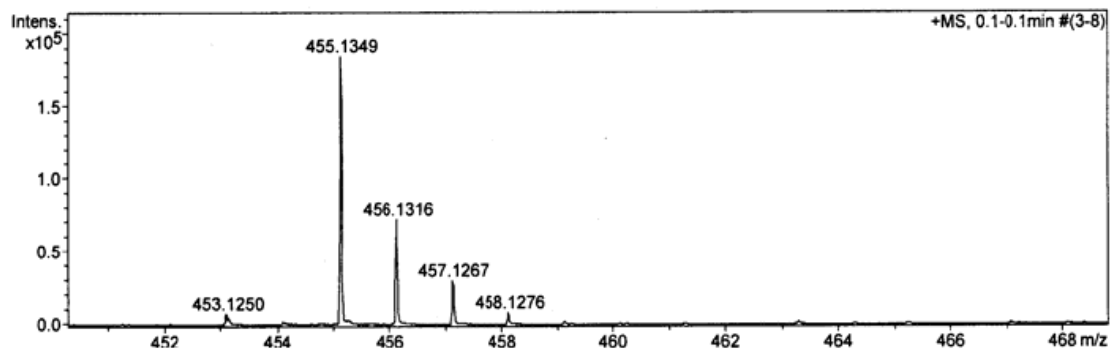
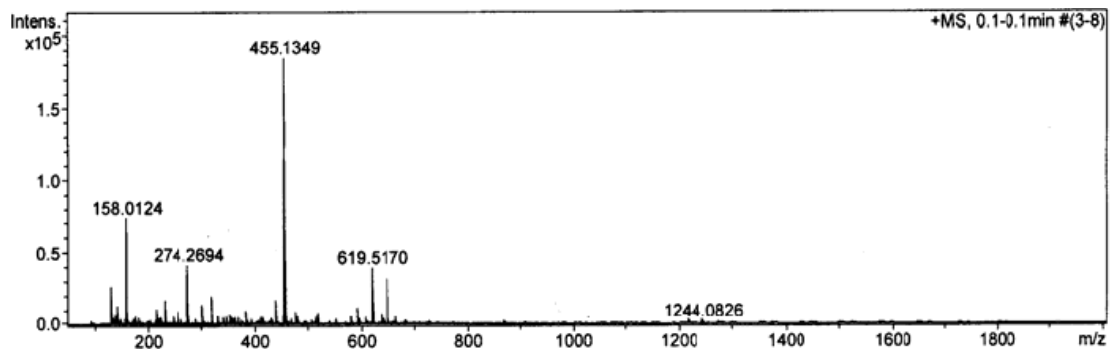
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### HRMS Spectrum of Compound 4aa

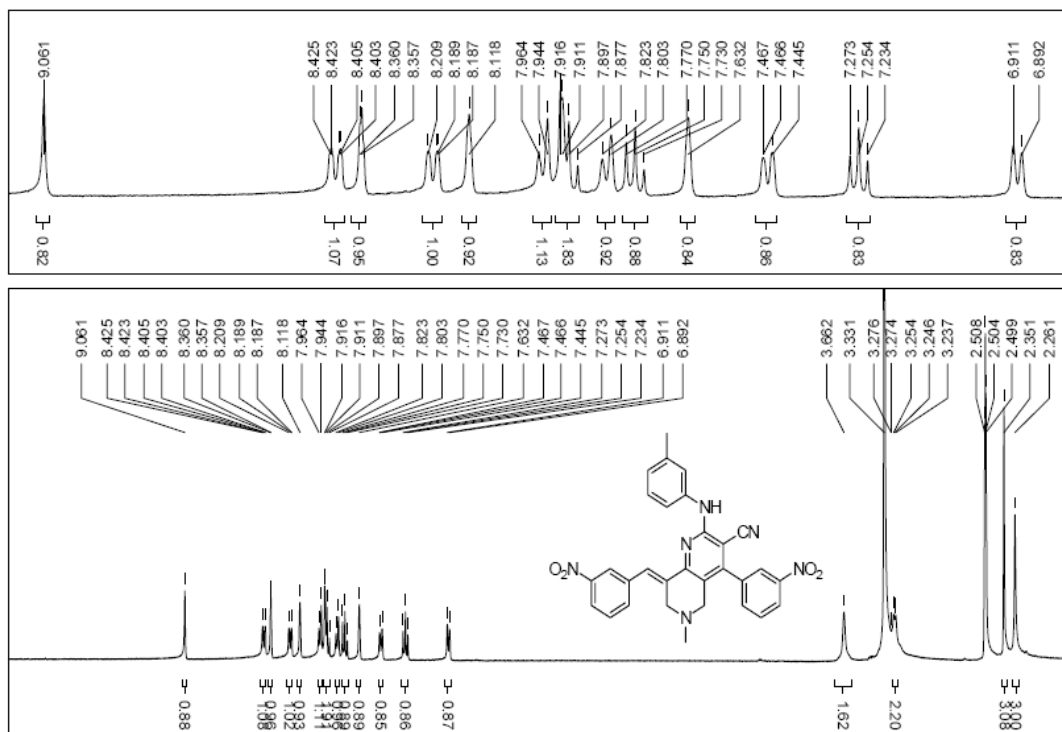


### <sup>1</sup>H NMR Spectrum of Compound 4ab

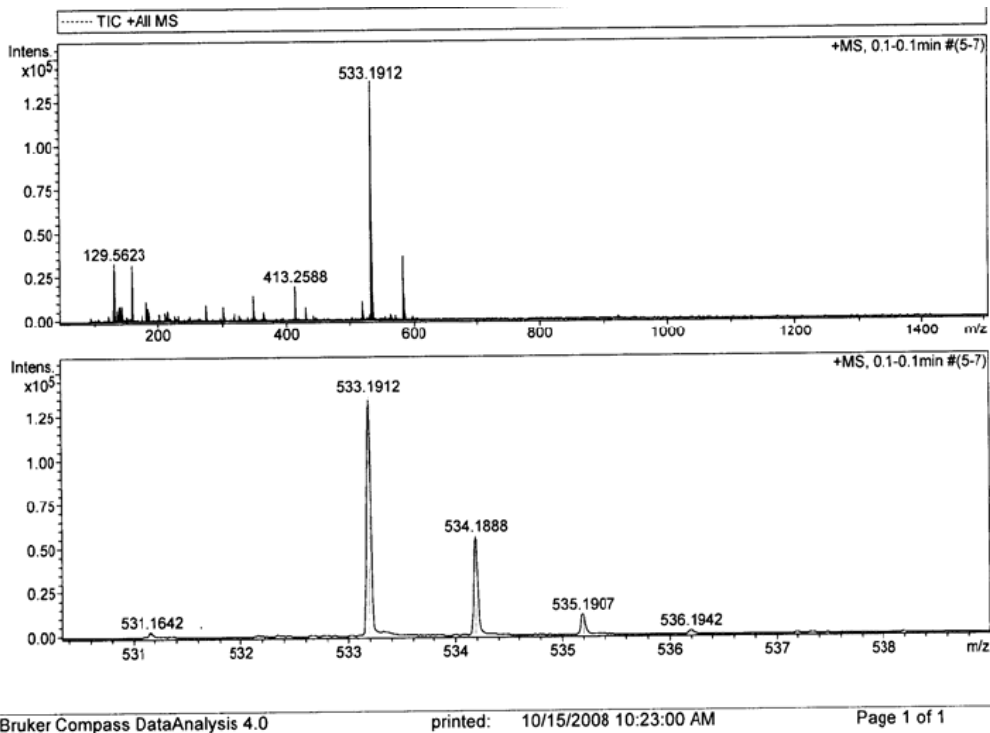




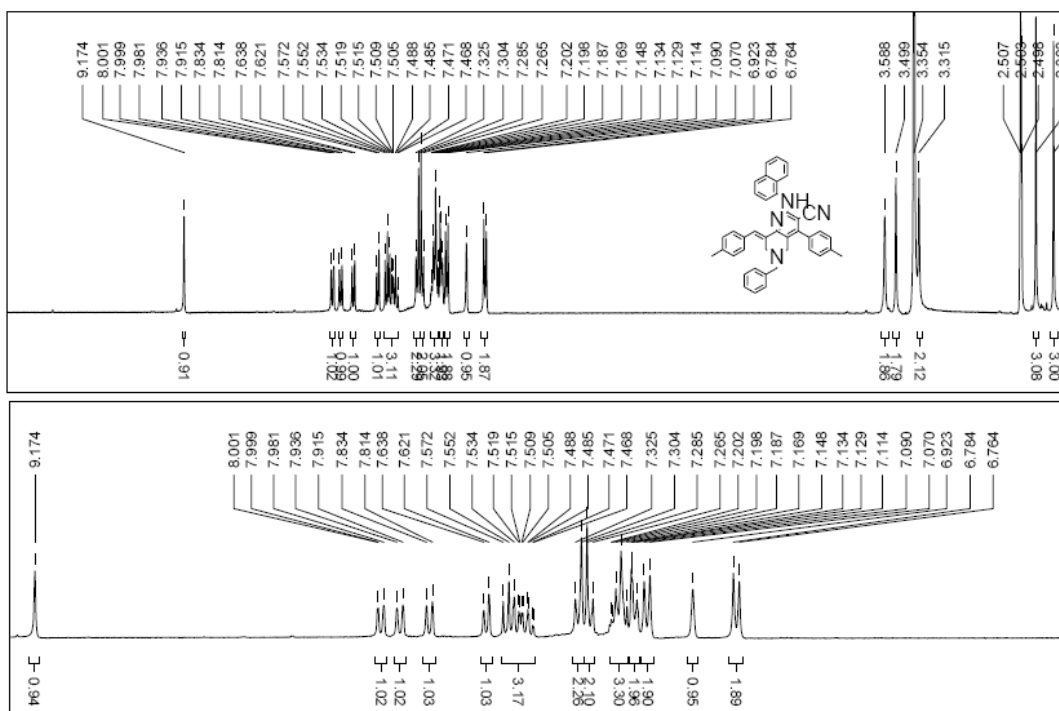
### HRMS Spectrum of Compound 4c



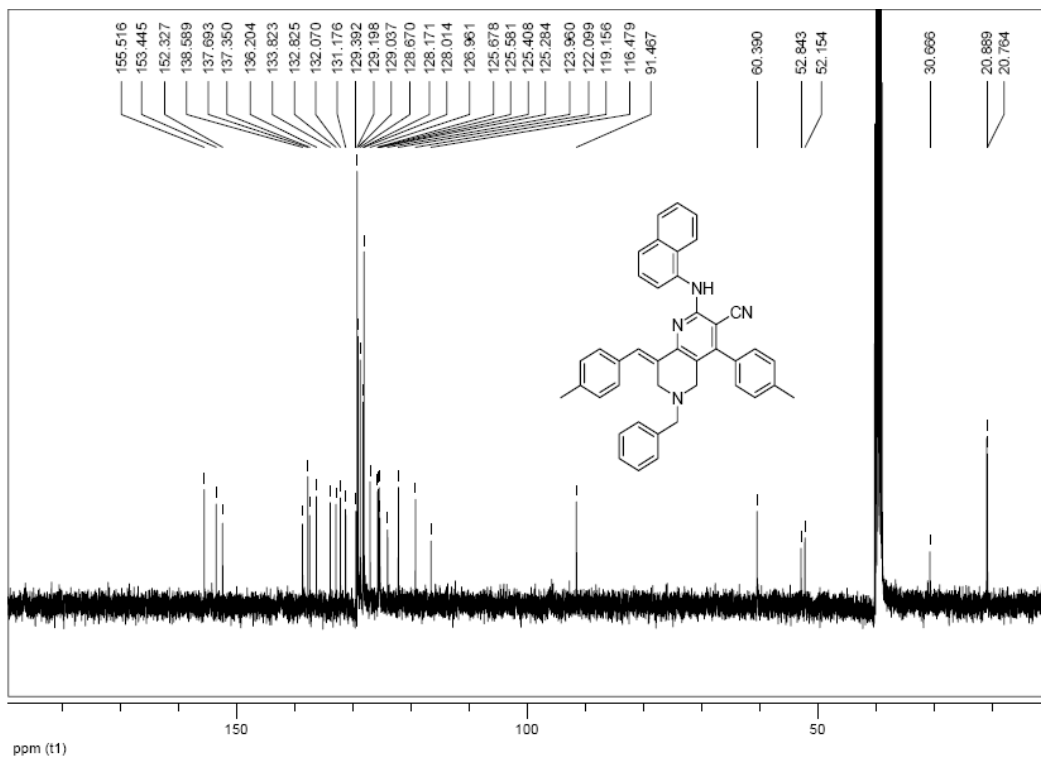
### <sup>1</sup>H NMR Spectrum of Compound 4d



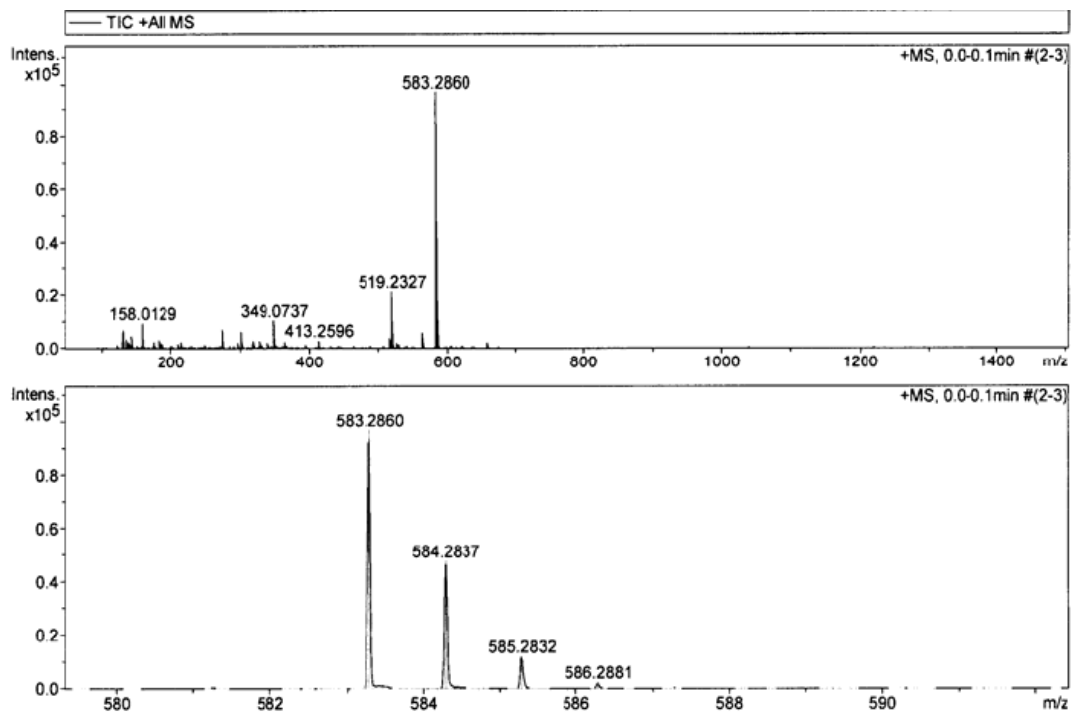
### HRMS Spectrum of Compound 4ad



### <sup>1</sup>H NMR Spectrum of Compound 4ae



<sup>13</sup>C NMR Spectrum of Compound 4ae



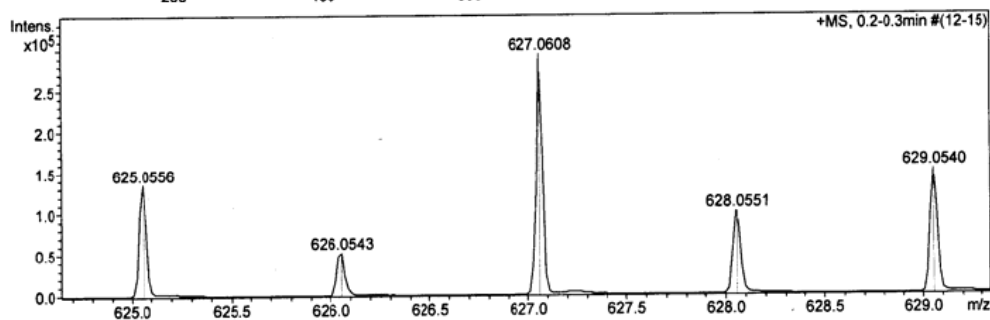
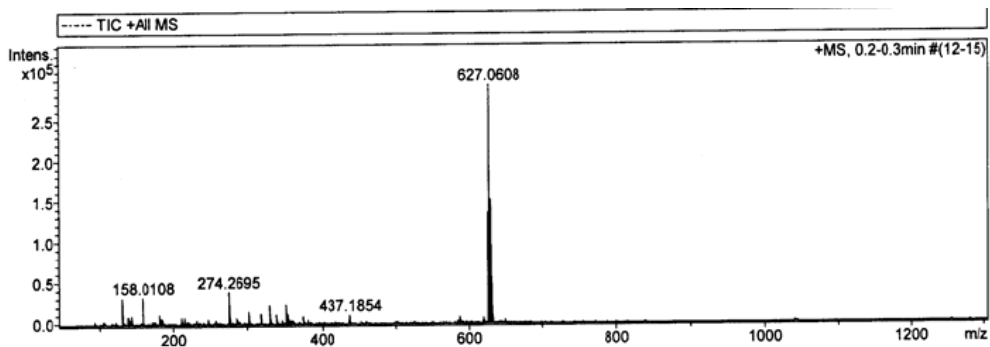
HRMS Spectrum of Compound 4ae









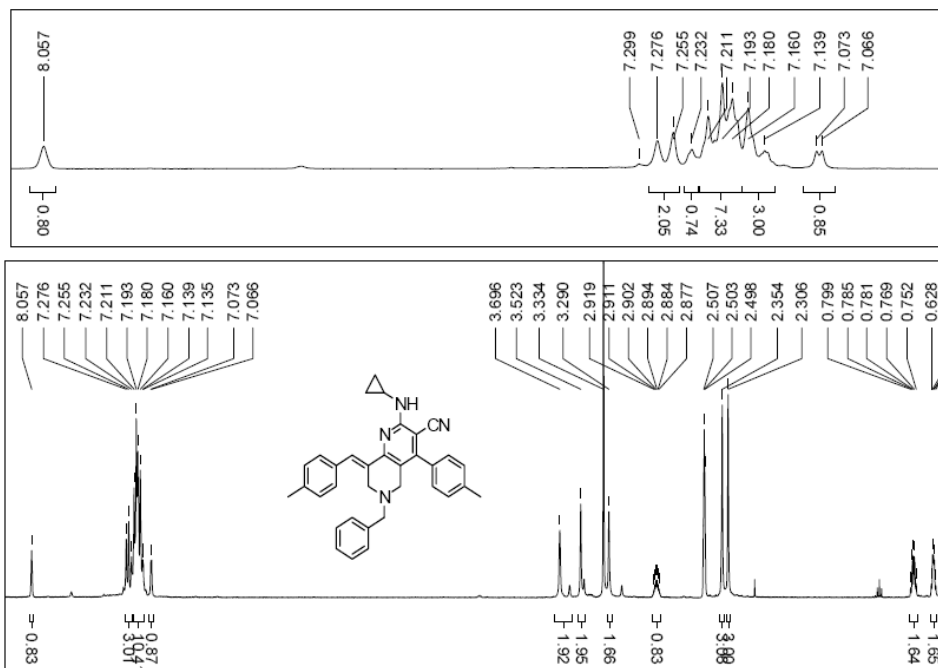


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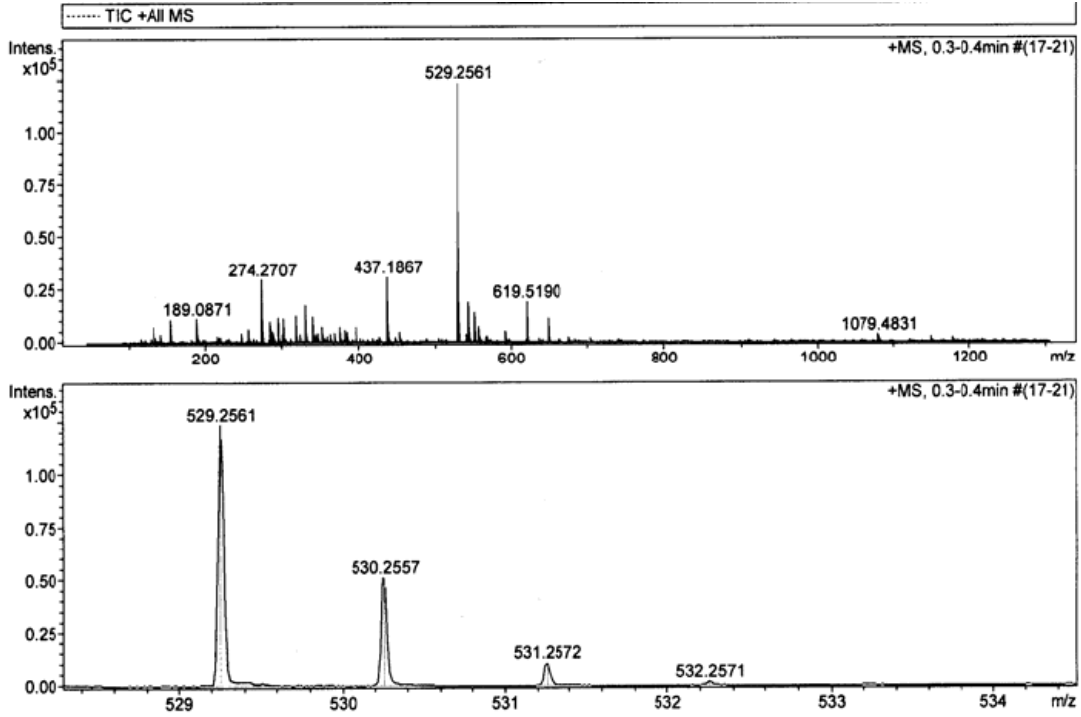
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### HRMS Spectrum of Compound 4ah

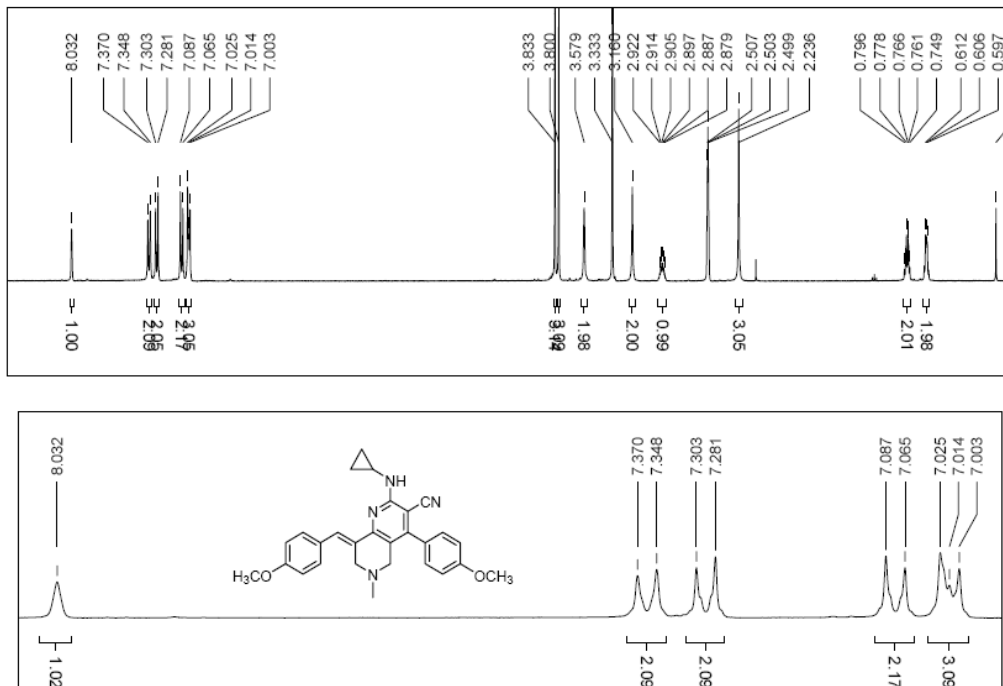


### <sup>1</sup>H NMR Spectrum of Compound 4ai

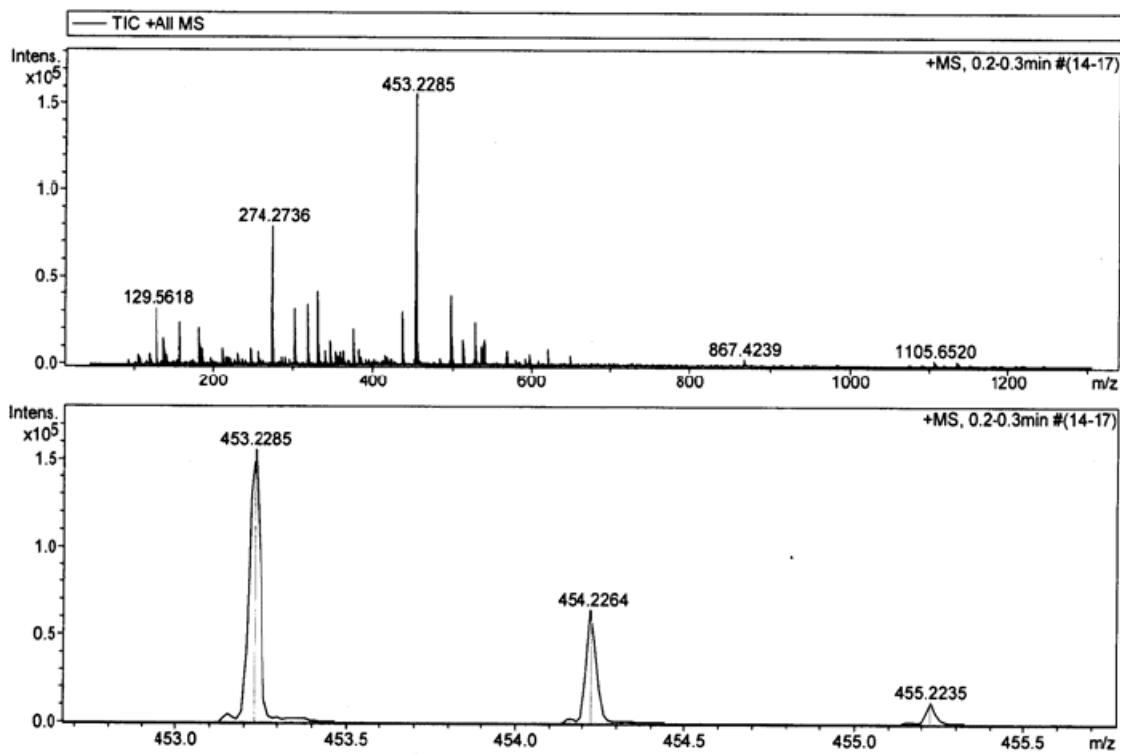




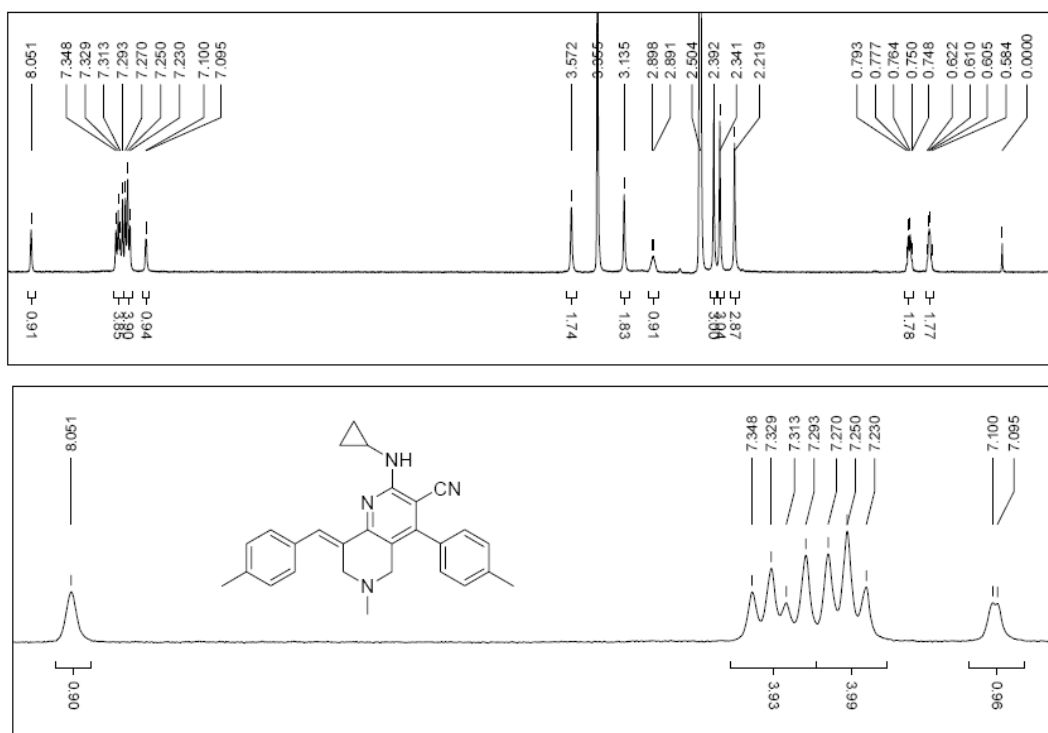
### HRMS Spectrum of Compound 4aj



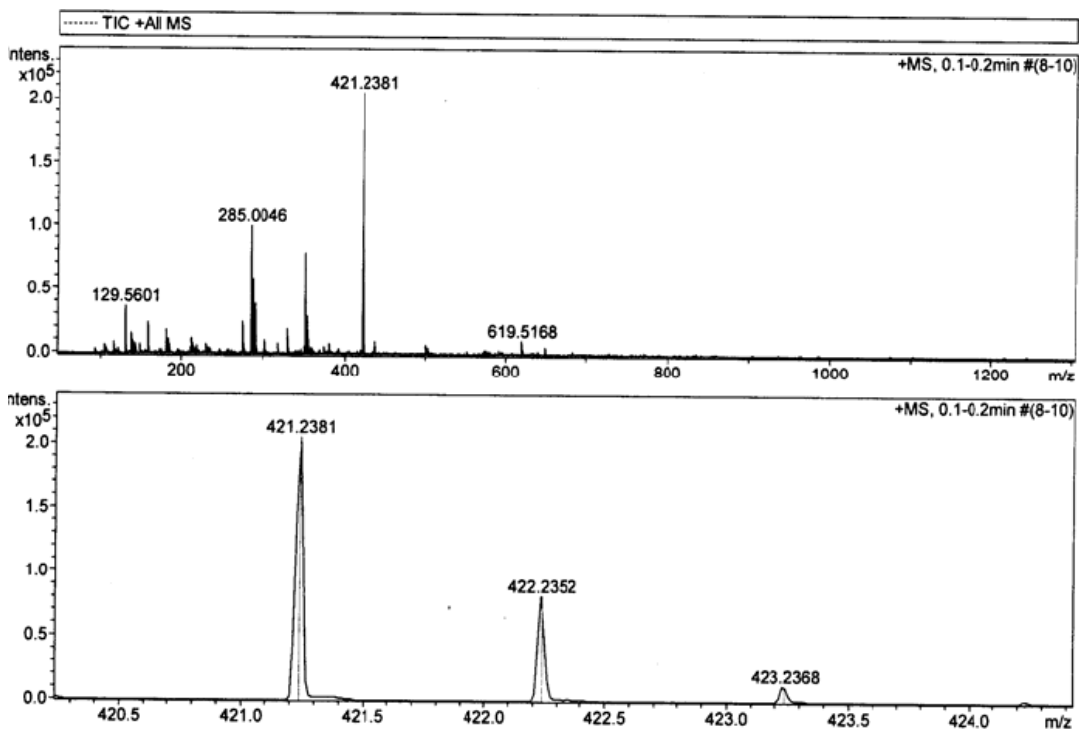
### <sup>1</sup>H NMR Spectrum of Compound 4ak



### HRMS Spectrum of Compound 4ak



### <sup>1</sup>H NMR Spectrum of Compound 4al

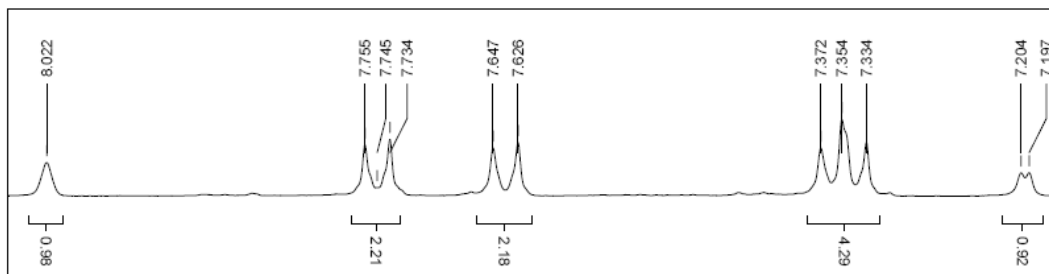


ker Compass DataAnalysis 4.0

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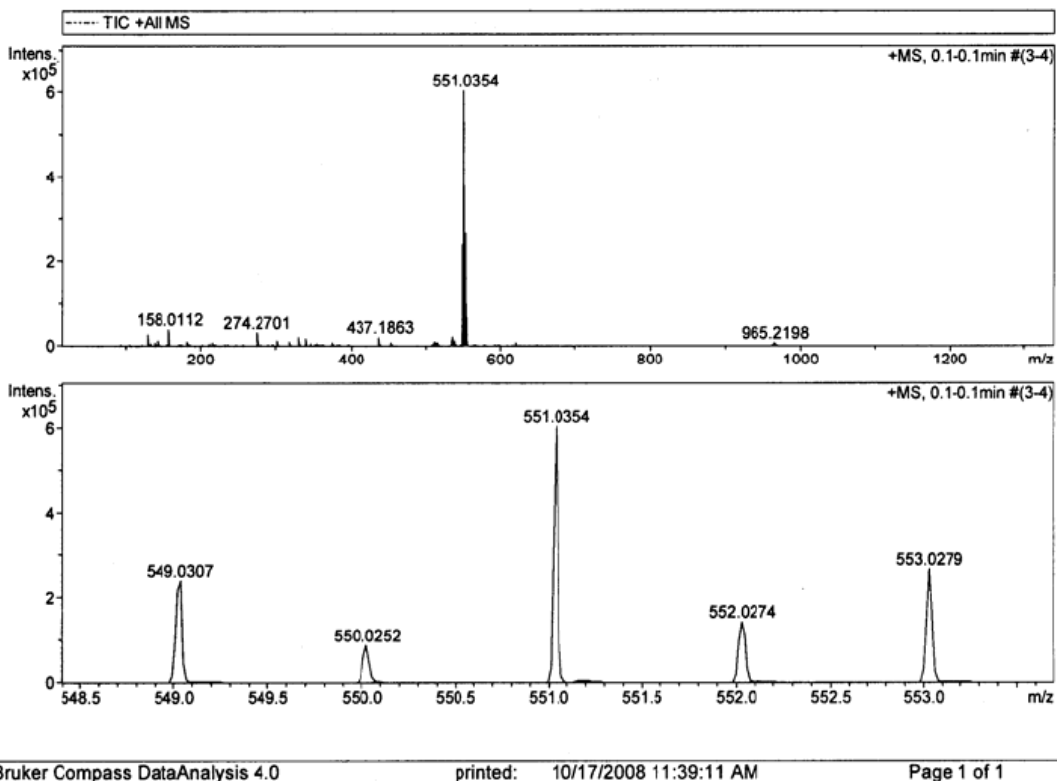
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### HRMS Spectrum of Compound 4a

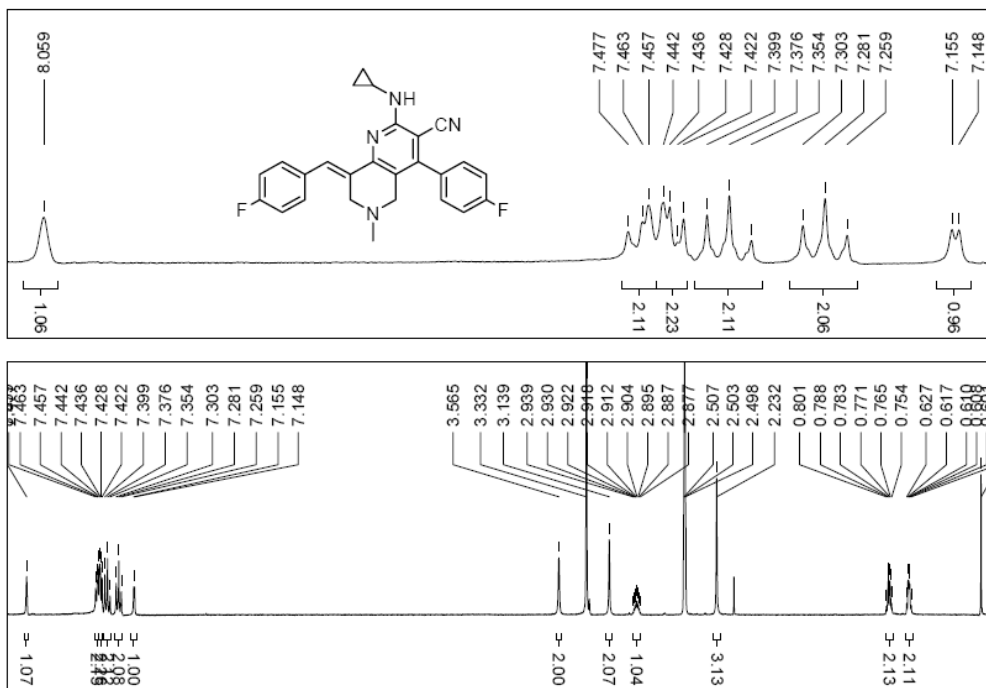


### <sup>1</sup>H NMR Spectrum of Compound 4am



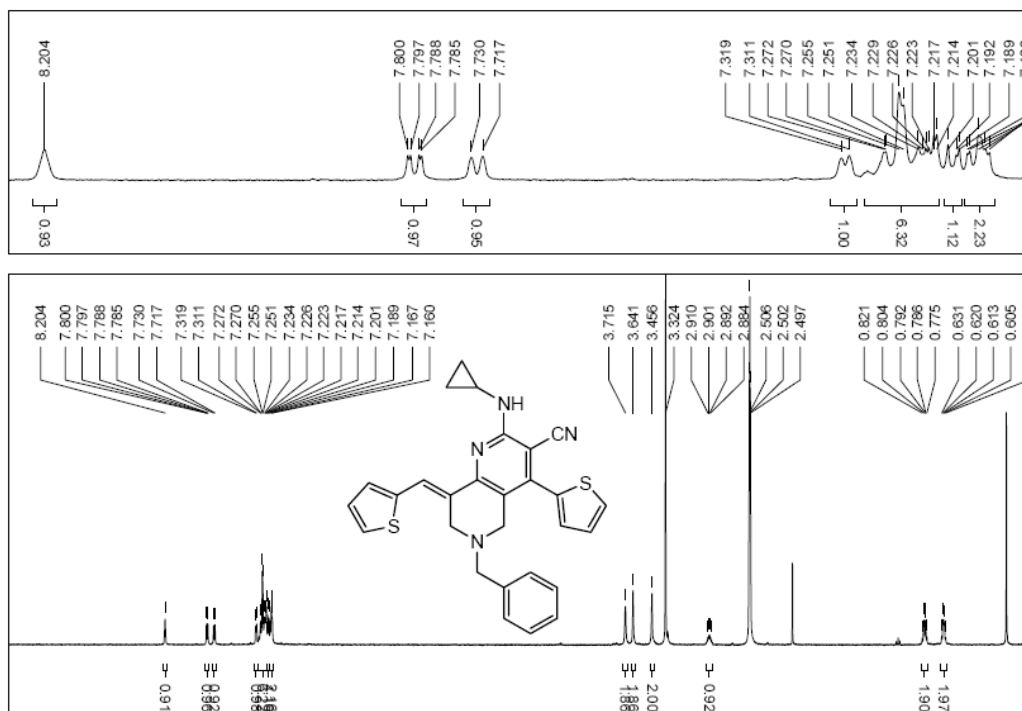


### HRMS Spectrum of Compound 4am

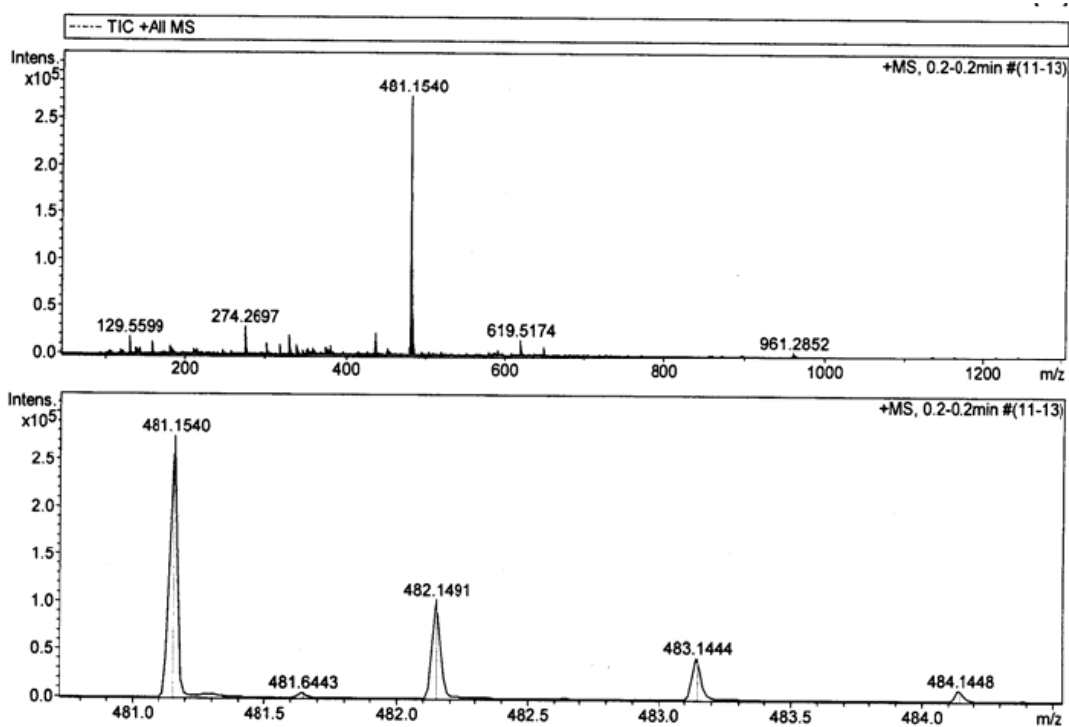


### <sup>1</sup>H NMR Spectrum of Compound 4an

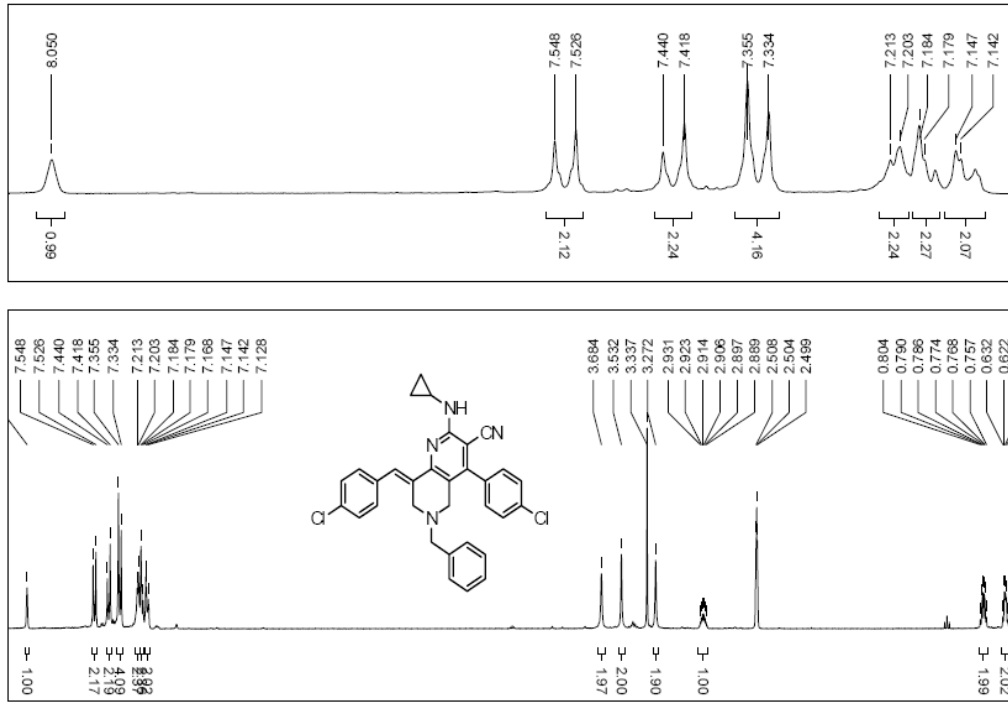




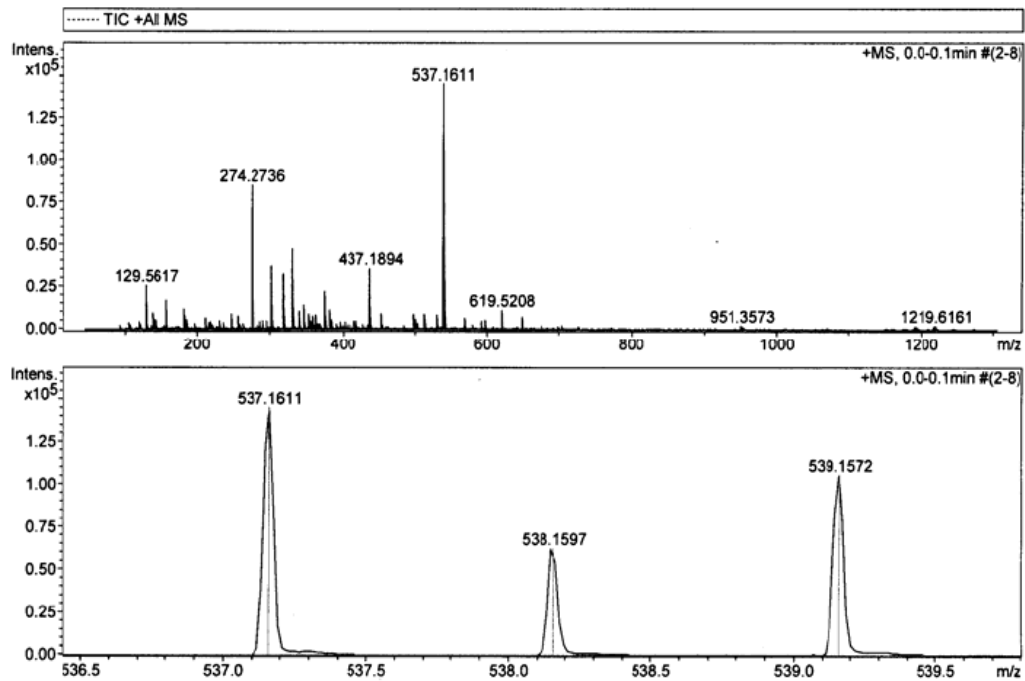
<sup>1</sup>H NMR Spectrum of Compound 4ap



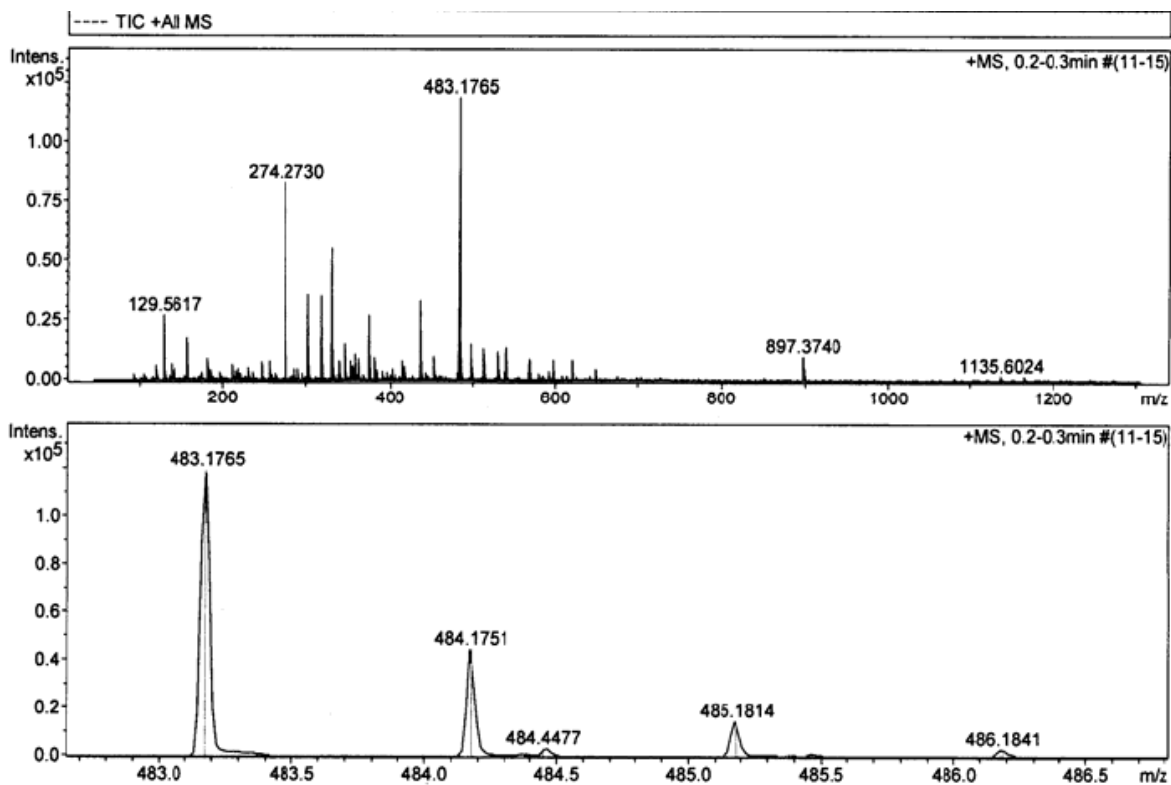
HRMS Spectrum of Compound 4ap



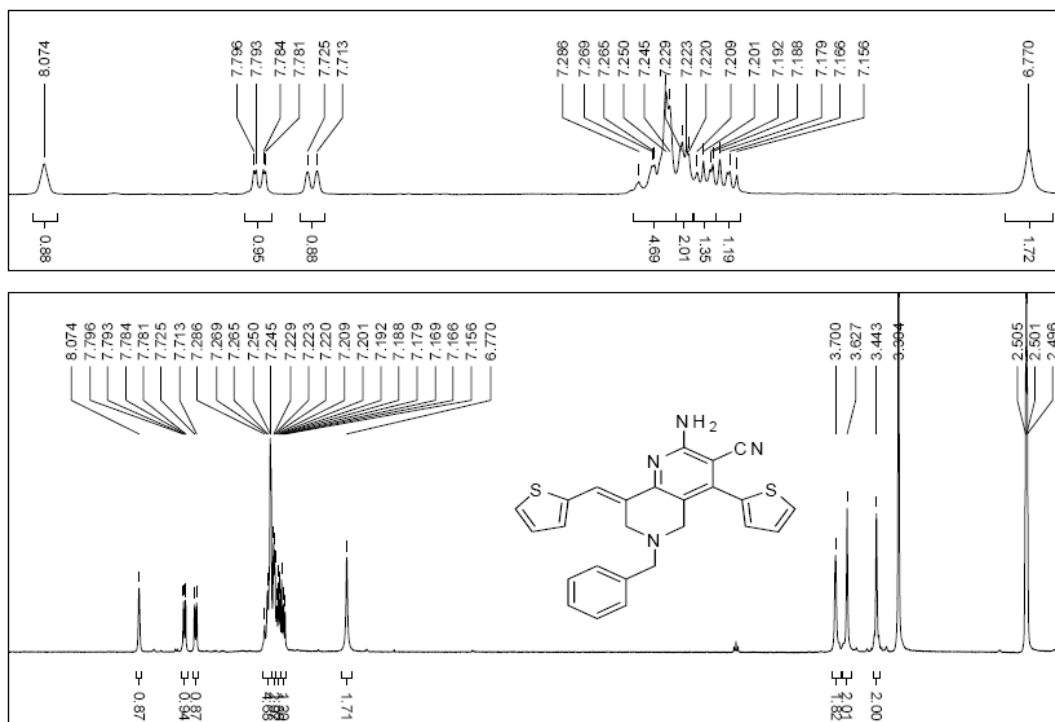
<sup>1</sup>H NMR Spectrum of Compound 4aq



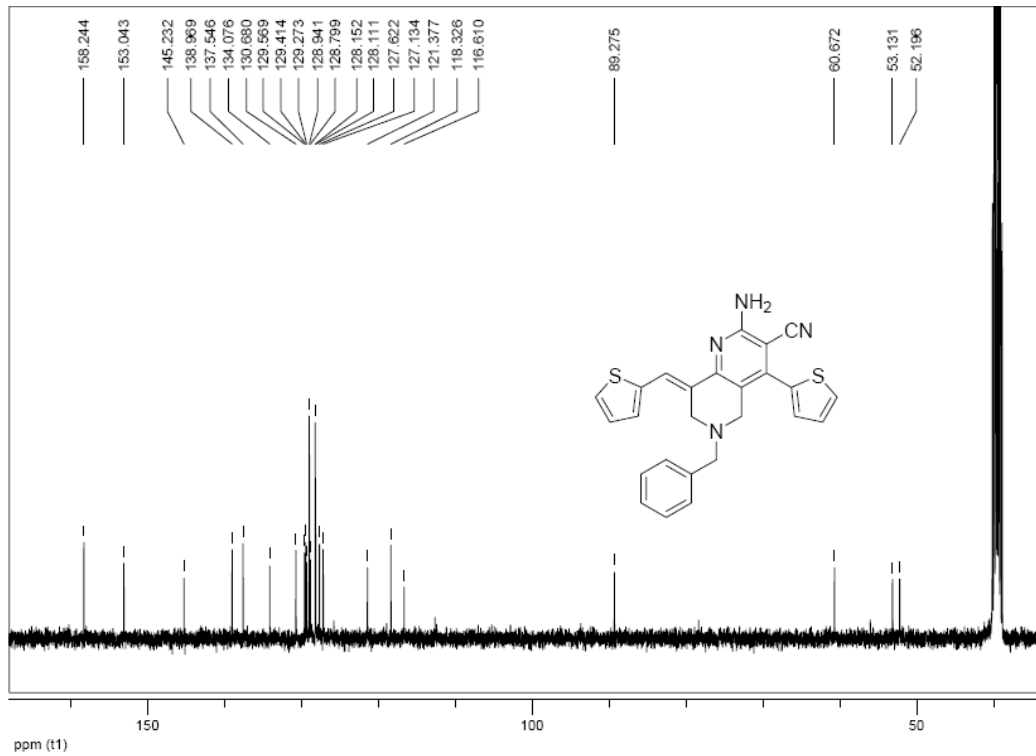
HRMS Spectrum of Compound 4aq



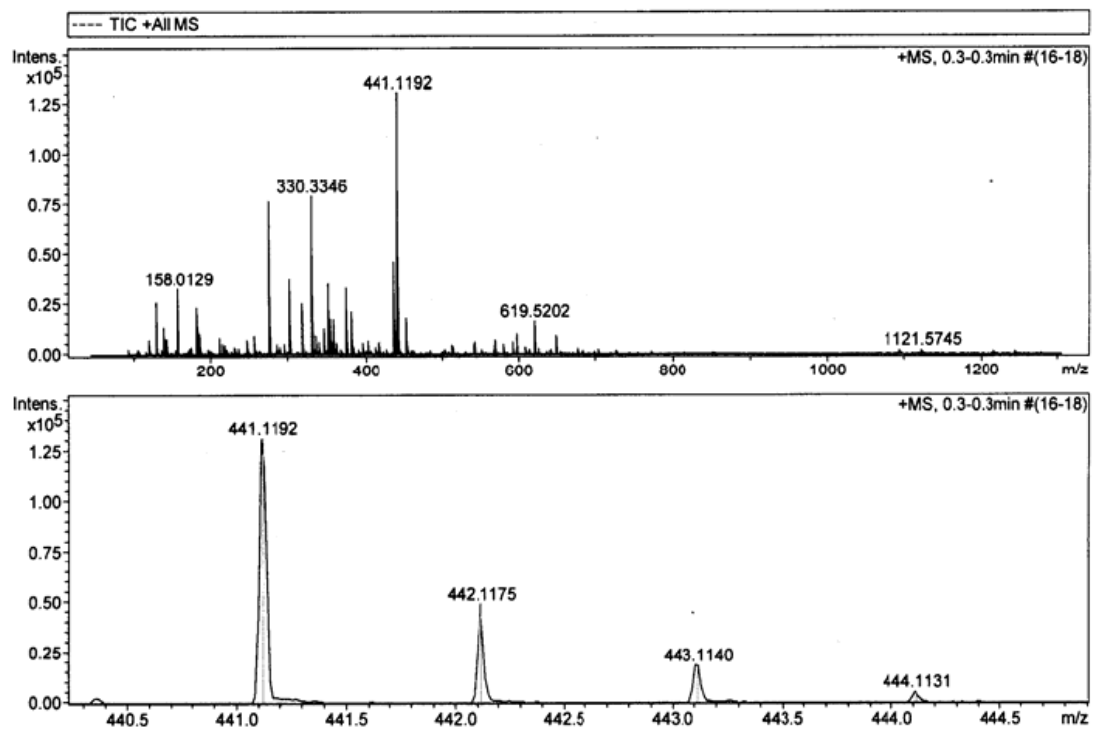
### HRMS Spectrum of Compound 4aq



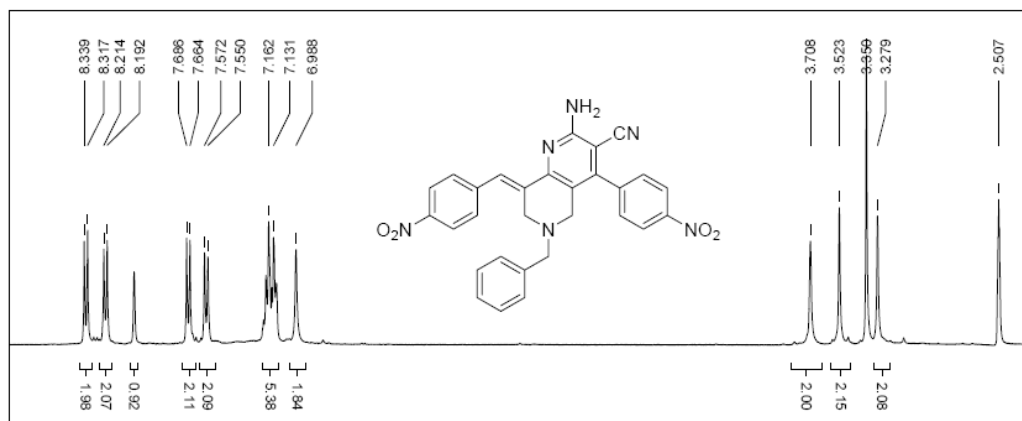
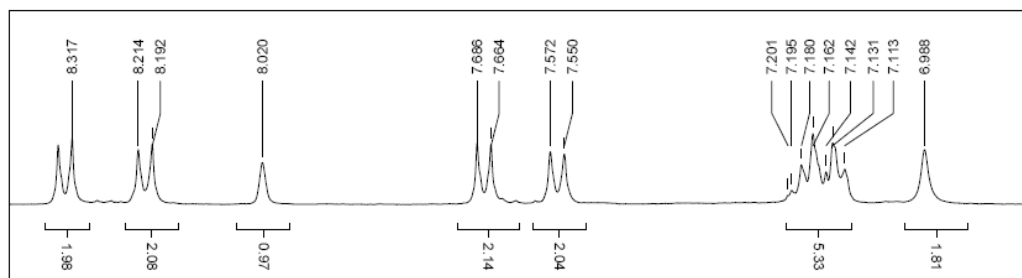
# <sup>1</sup>H NMR Spectrum of Compound 4ar

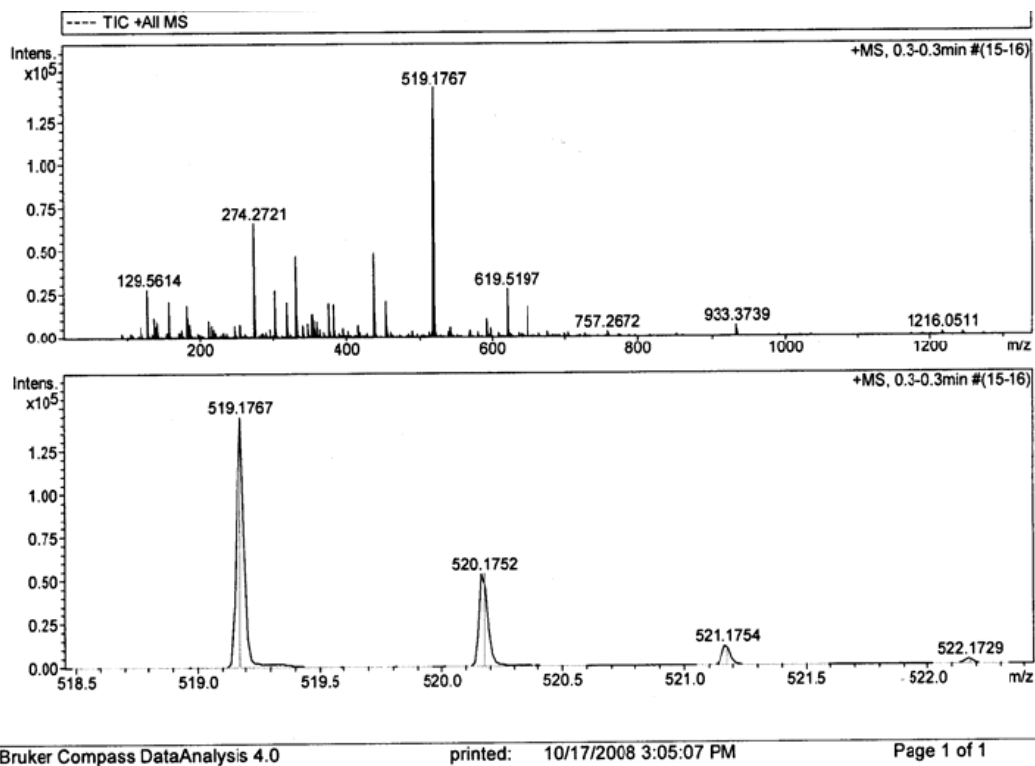


# <sup>13</sup>C NMR Spectrum of Compound 4ar

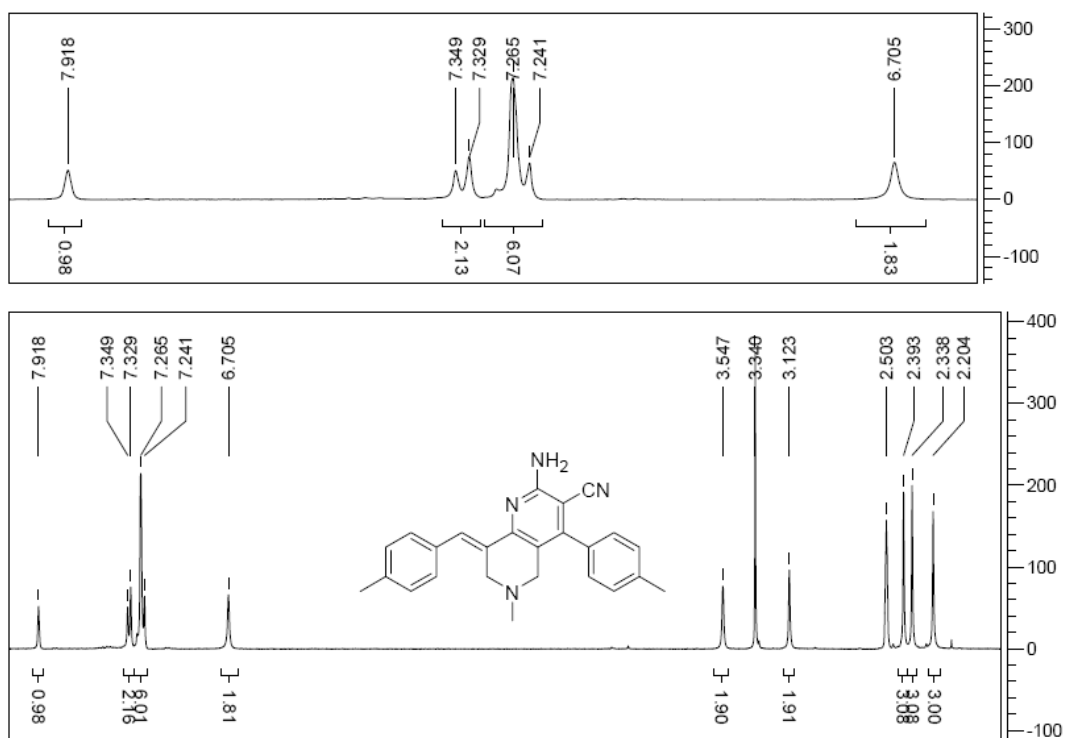


### HRMS Spectrum of Compound 4ar



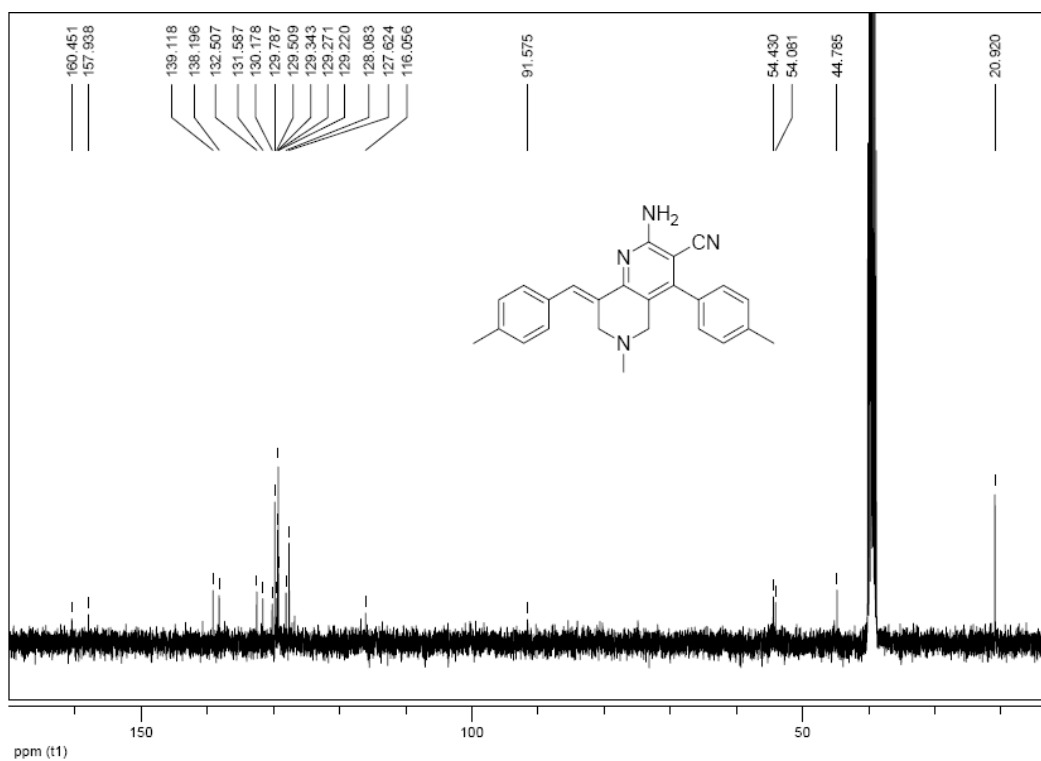


### HRMS Spectrum of Compound 4as

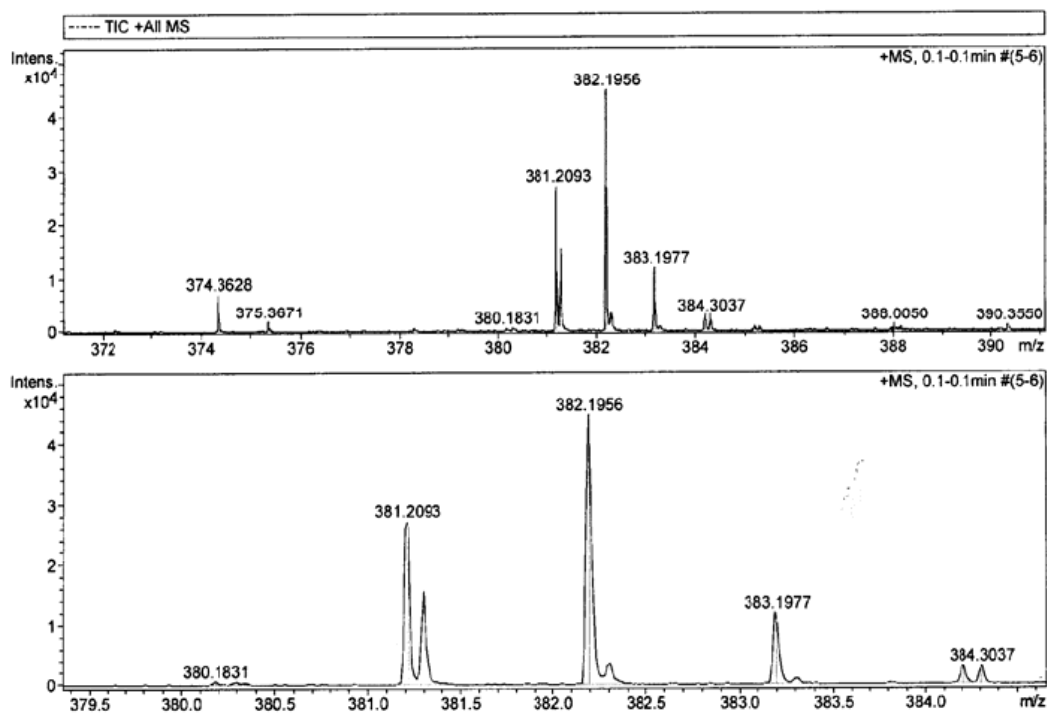


### <sup>1</sup>H NMR Spectrum of Compound 4at

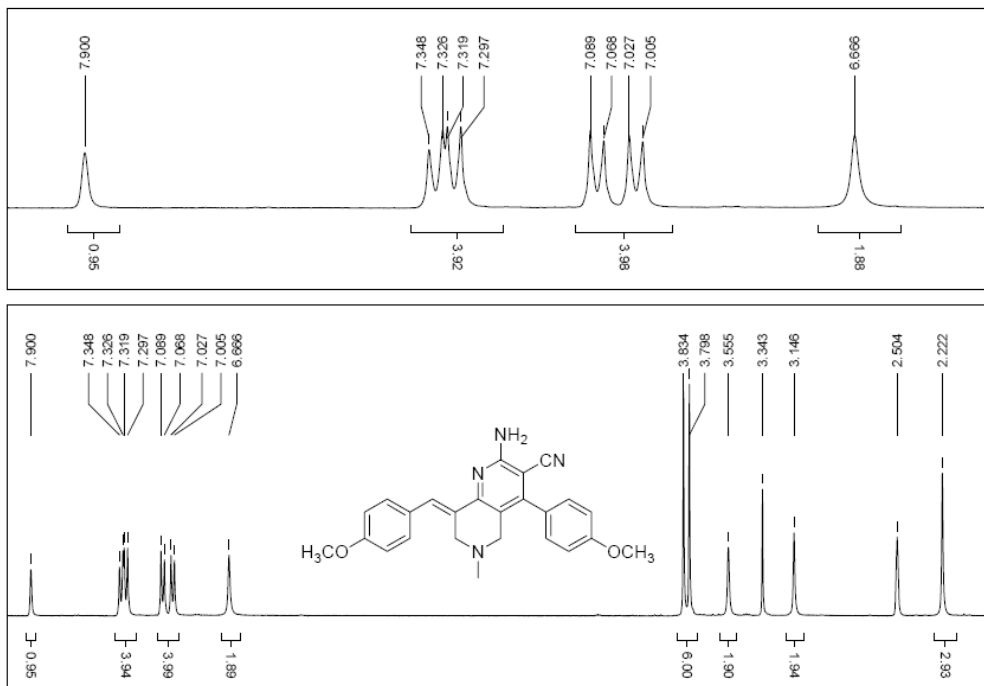




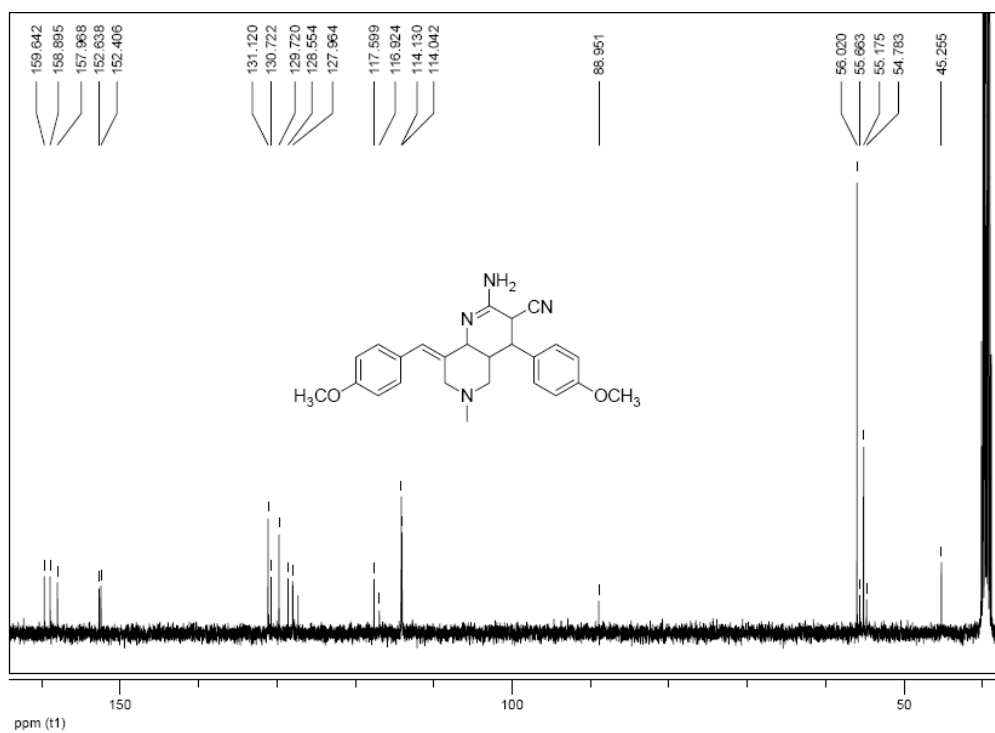
**<sup>13</sup>C NMR Spectrum of Compound 4at**



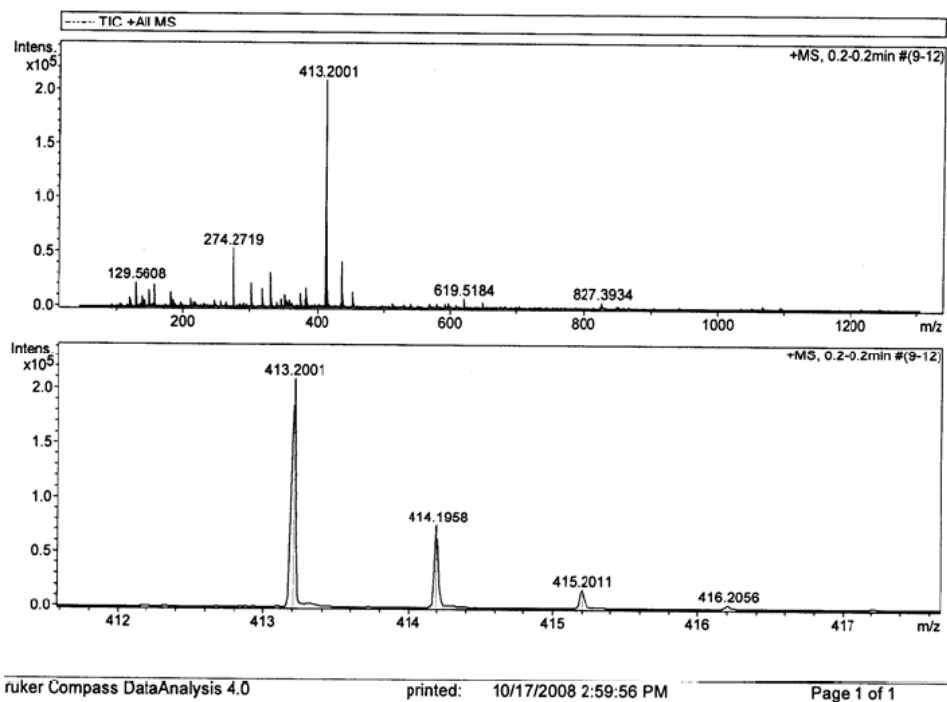
**HRMS Spectrum of Compound 4at**



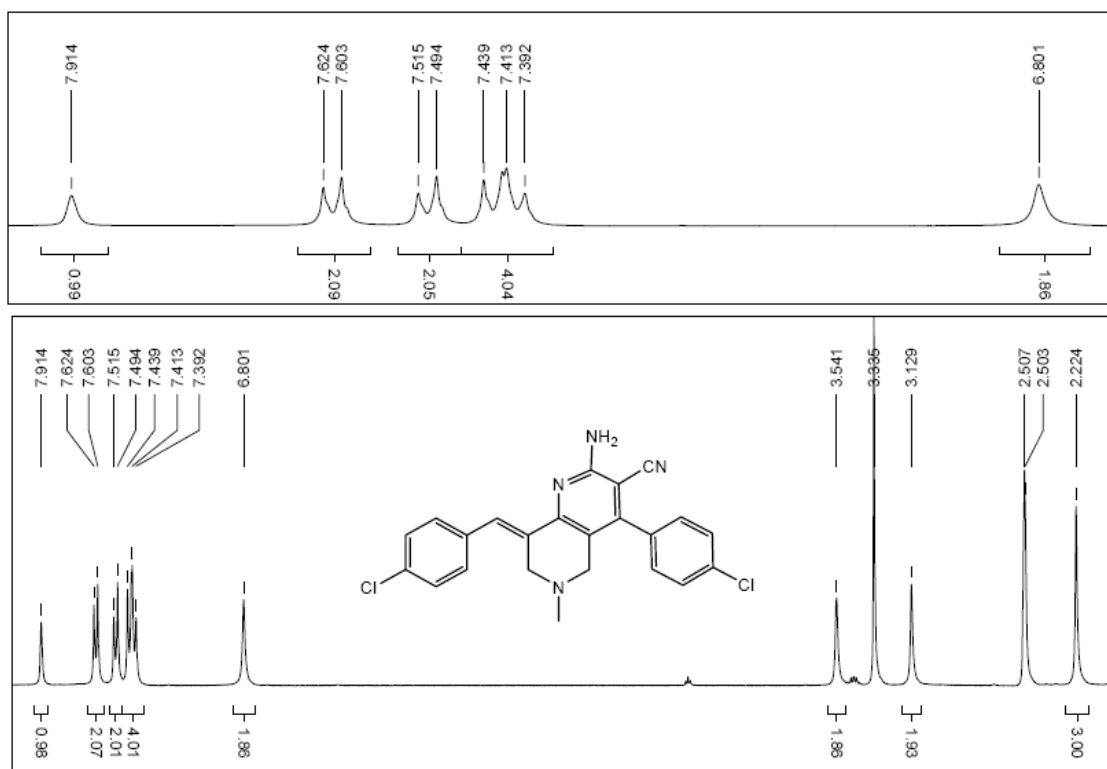
**<sup>1</sup>H NMR Spectrum of Compound 4au**



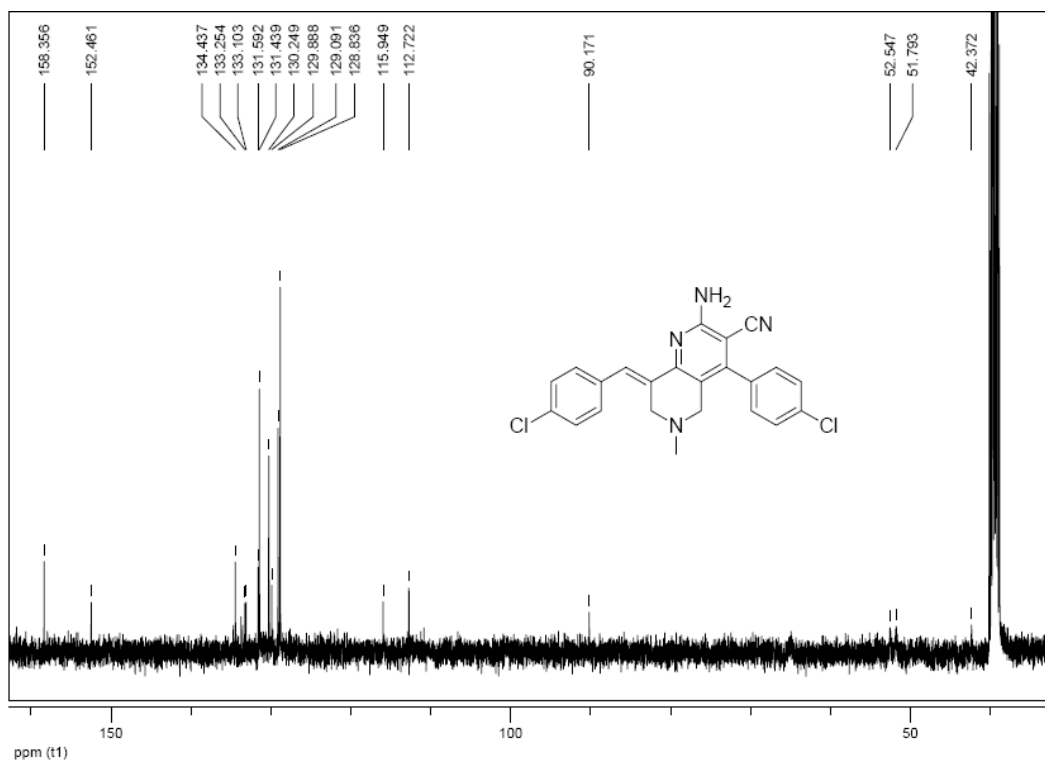
**<sup>13</sup>C NMR Spectrum of Compound 4au**



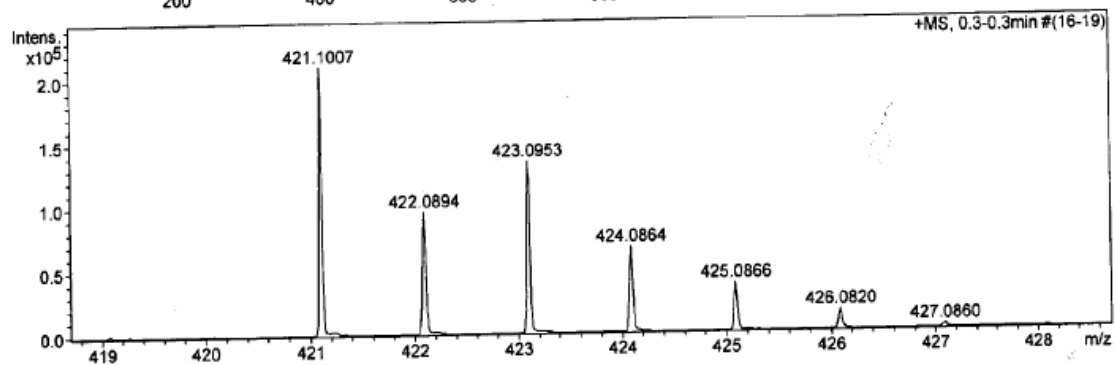
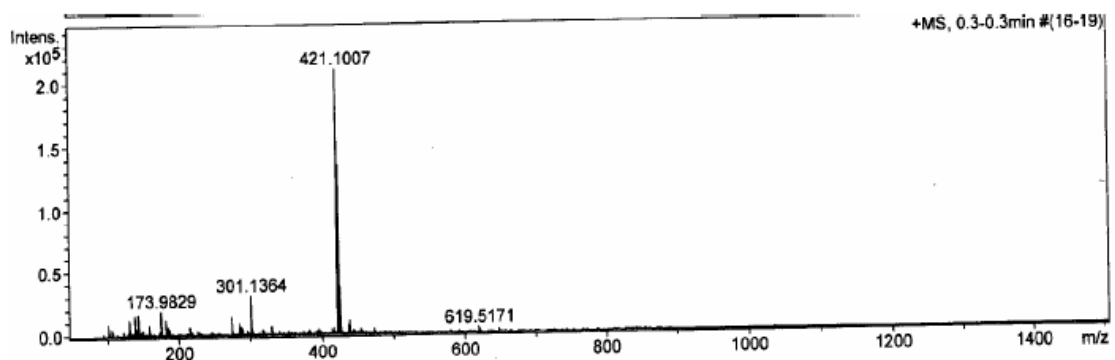
**HRMS Spectrum of Compound 4au**



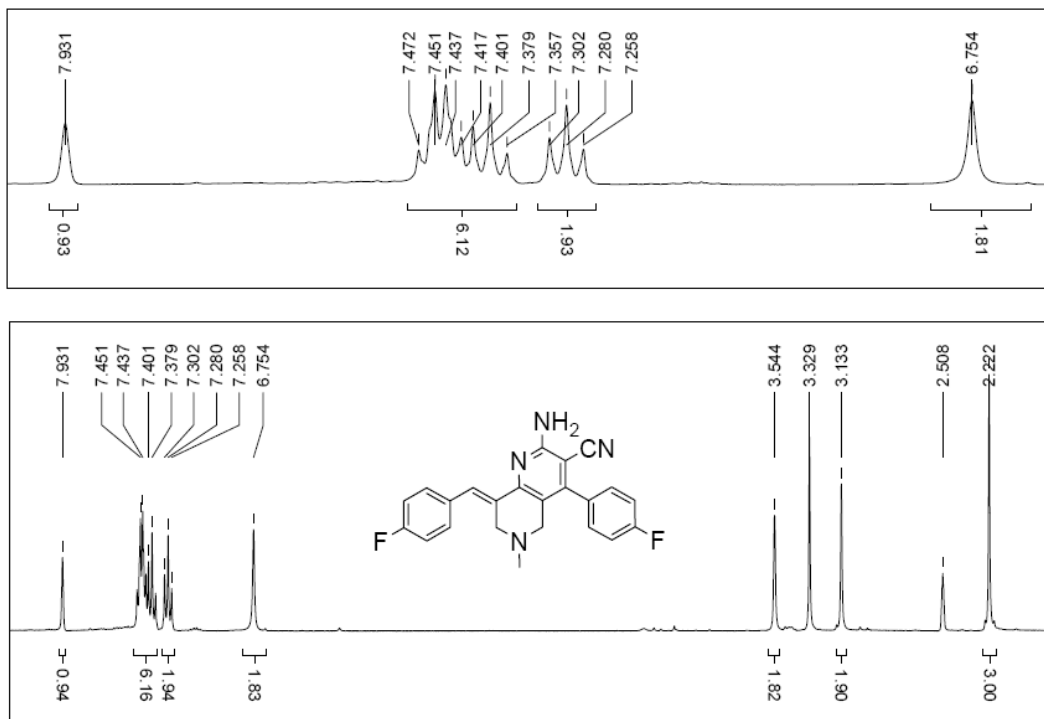
**<sup>1</sup>H NMR Spectrum of Compound 4av**



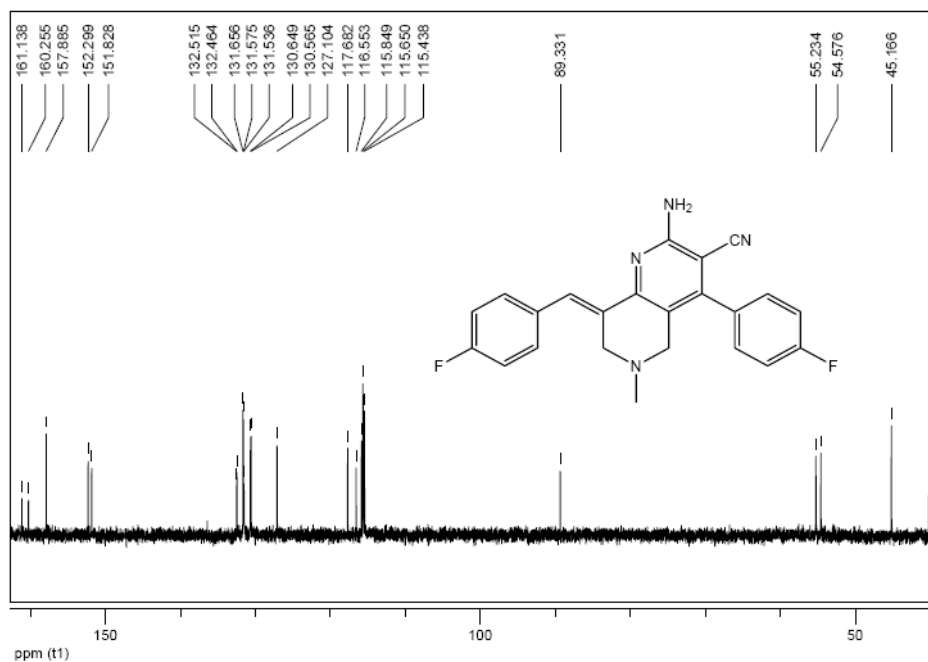
<sup>13</sup>C NMR Spectrum of Compound 4av



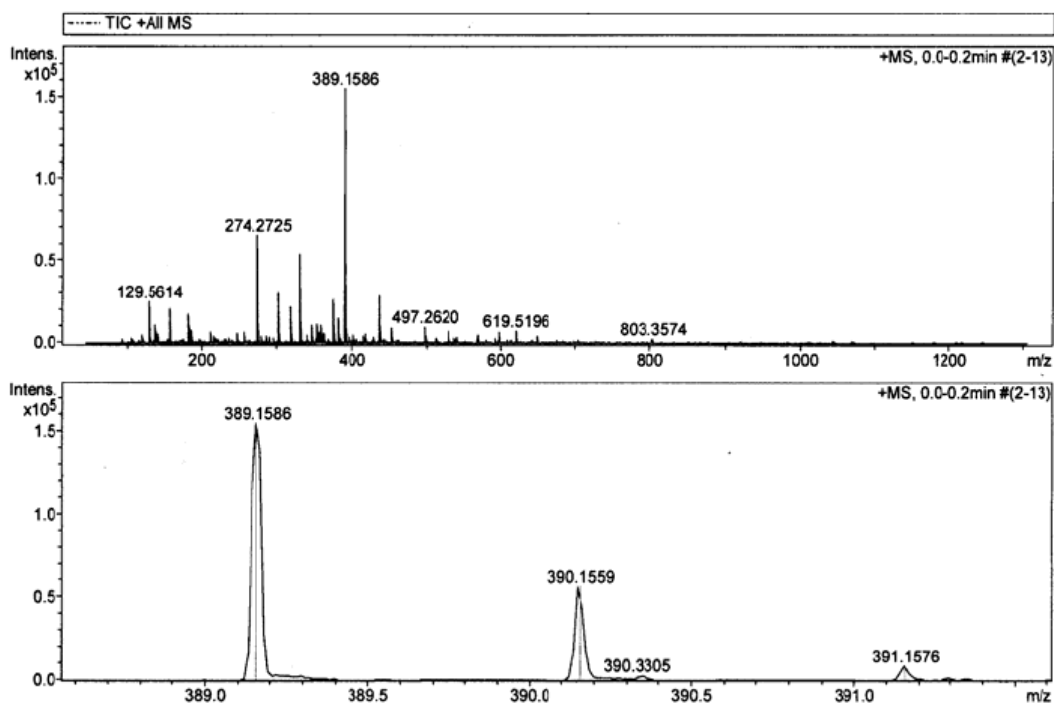
HRMS Spectrum of Compound 4av



**<sup>1</sup>H NMR Spectrum of Compound 4aw**



**<sup>13</sup>C NMR Spectrum of Compound 4aw**

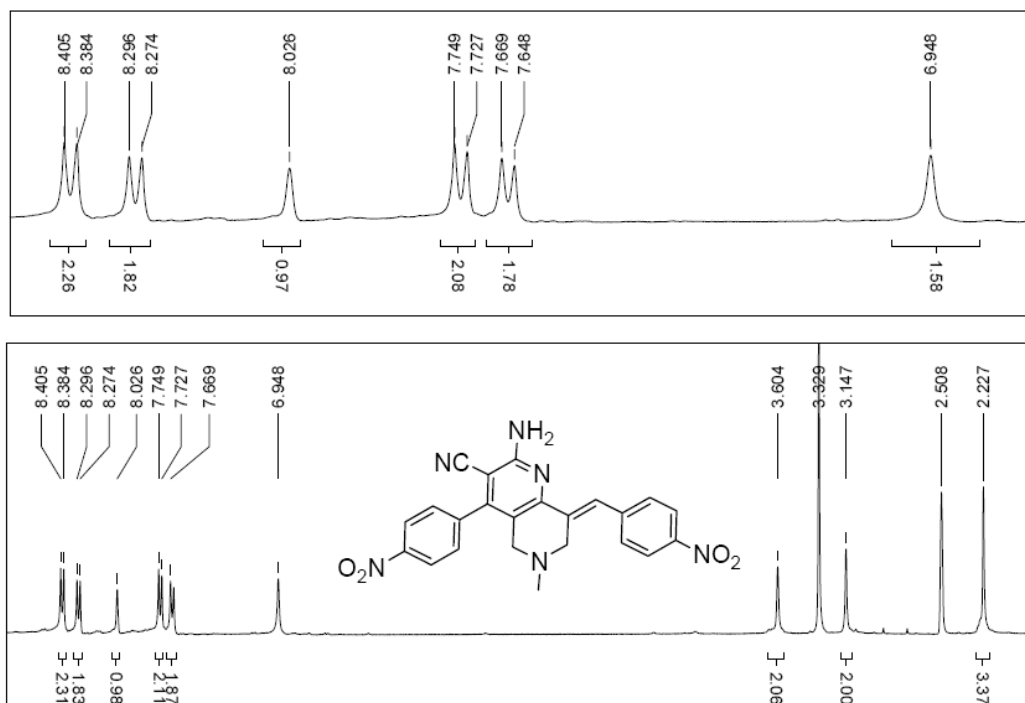


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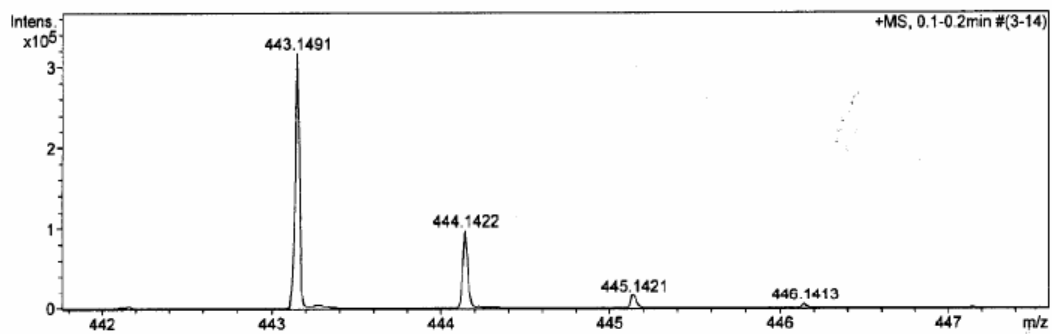
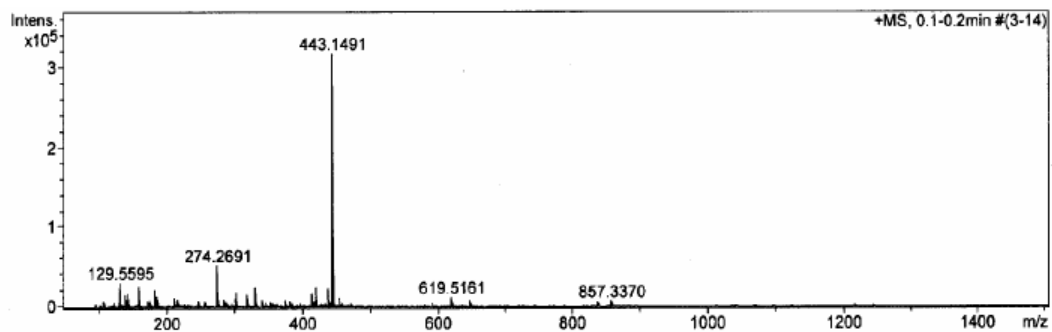
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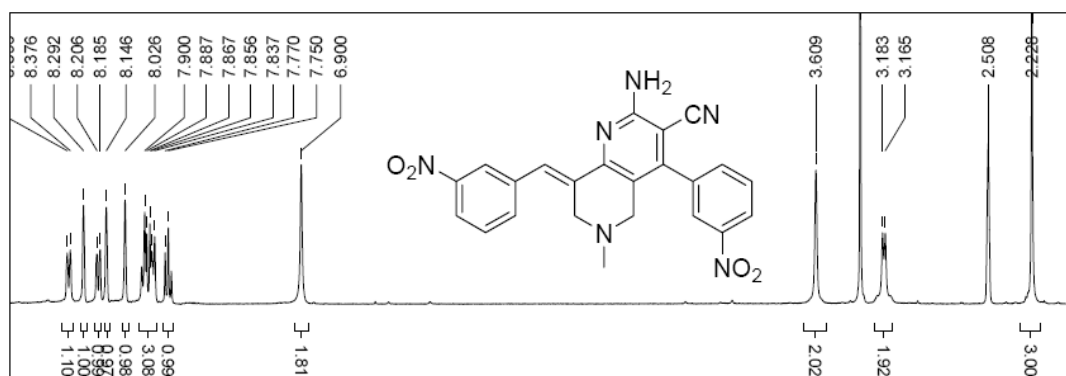
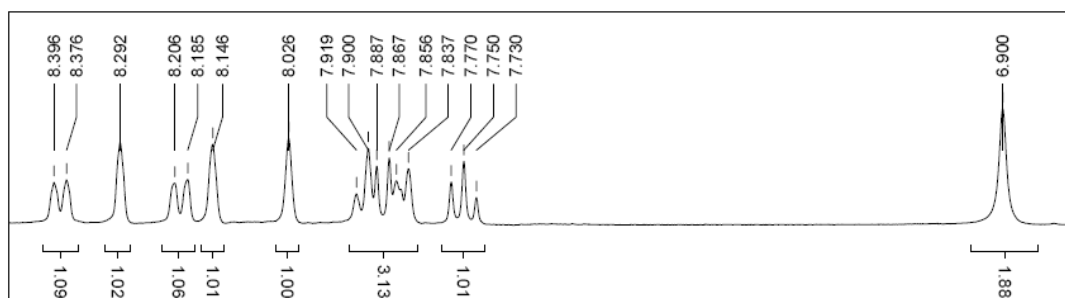
### HRMS Spectrum of Compound 4aw



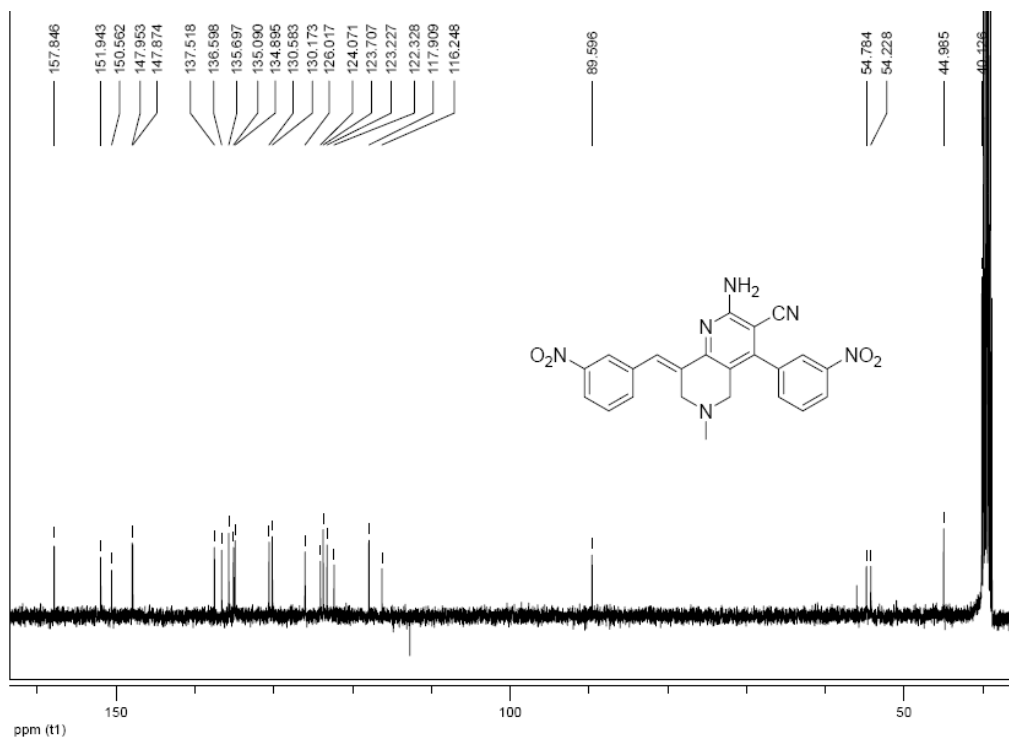
### <sup>1</sup>H NMR Spectrum of Compound 4ax



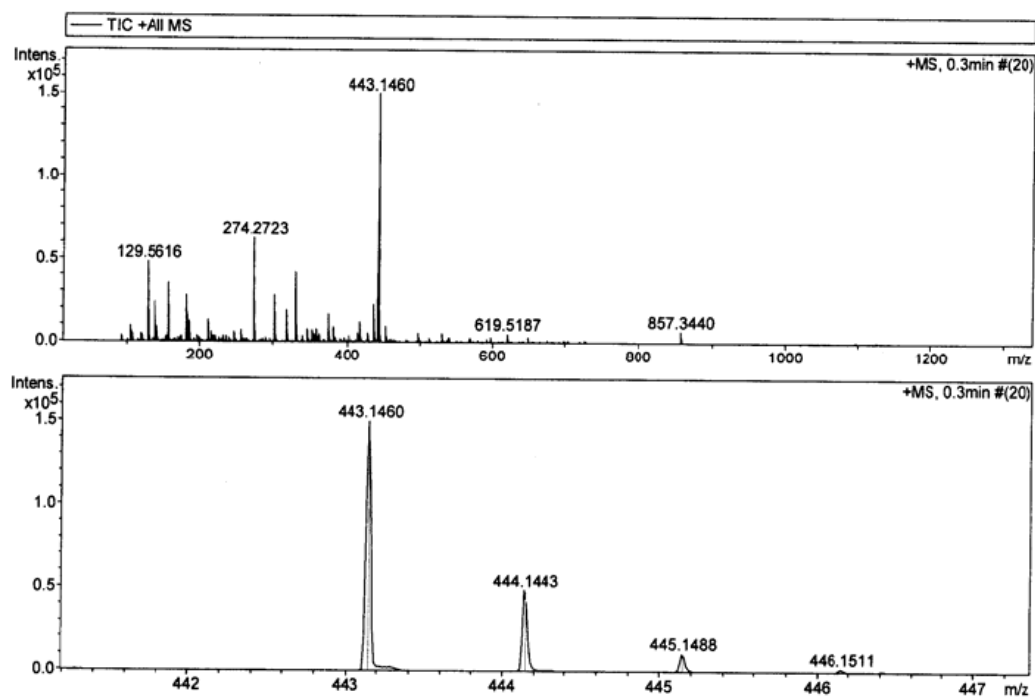
### HRMS Spectrum of Compound 4ax



### <sup>1</sup>H NMR Spectrum of Compound 4ay

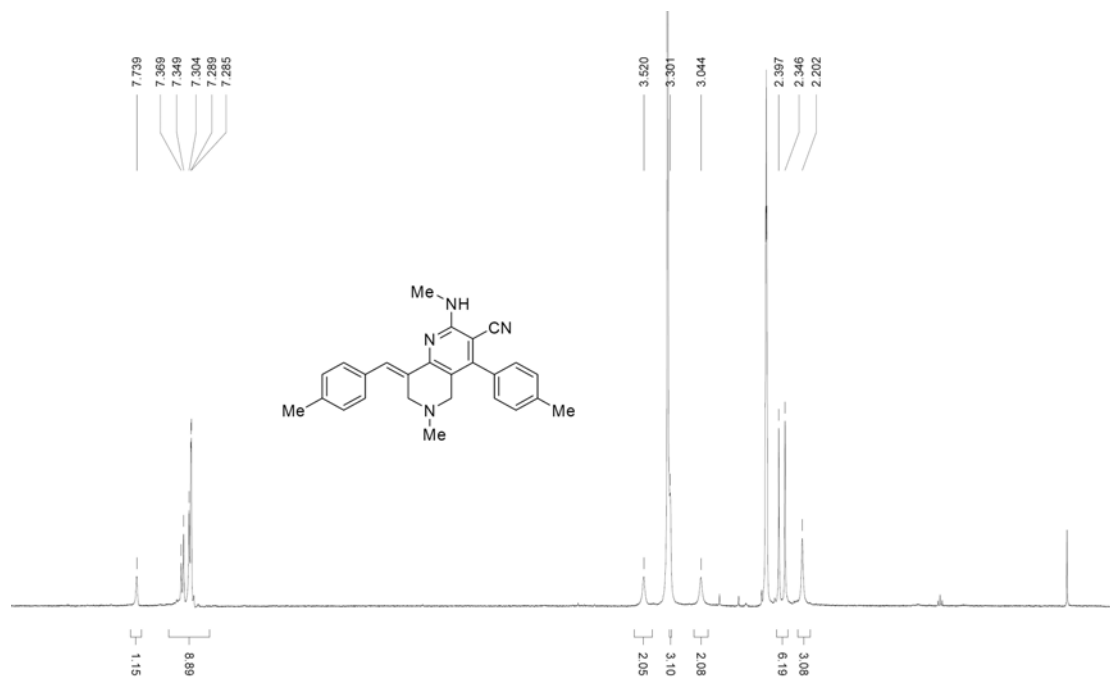


<sup>13</sup>C NMR Spectrum of Compound 4ay



HRMS Spectrum of Compound 4ay





**<sup>1</sup>H NMR Spectrum of Compound 4az**