# Supporting Information

## Base-promoted aerobic oxidative synthesis of fused 1,3,5-triazines under metal-free conditions

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## 1. General experimental information

All reactions were carried out under an atmosphere of oxygen unless otherwise noted. All reported reaction temperatures corresponded to oil bath temperatures. 4 Å molecular sieves were freshly activated in the microwave oven prior to use. Product purifications were performed either with preparative analytical thin-layer chromatography (TLC) or recrystallization. $^{1}$H NMR and $^{13}$C NMR spectra were recorded on Bruker-AV (400 and 100 MHz, respectively) instrument internally referenced to tetramethylsilane (TMS) or chloroform. Mass spectra were measured on Agilent 5975 GC-MS instrument (EI). High-resolution mass spectra (HRMS) were conducted using electrospraying ionization (ESI) and recorded at Beijing Forestry University, Beijing. Melting points were measured with a BÜCHI B-545 melting point instrument and were uncorrected. The structure of known compounds was further corroborated by comparing their $^{1}$H NMR, $^{13}$C NMR data and MS data with those of literature. All reagents were obtained from commercial suppliers and used without further purification unless otherwise noted.

## 2. General procedure

**General procedure for the synthesis of benzimidazo[1,2-a]-1,3,5-triazines.**

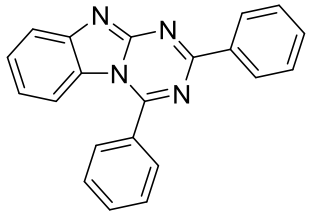
A 10 mL oven-dried reaction vessel was charged with 2-aminobenzimidazoles (0.2 mmol), aromatic aldehyde (0.6 mmol), ammonium iodide (0.3 mmol), sodium hydrogen carbonate (0.2 mmol), freshly activated 4 Å molecular sieves (100 mg), DMSO (0.2 mmol) and chlorobenzene (0.6 mL). The reaction vessel was purged with oxygen for three times and stirred at 140 $^{o}$C for 16 h. After cooling to room temperature, the reaction mixture was diluted with EtOAc or THF and filtered. The filtrate was then concentrated *in vacuo*, and the resulting residue was purified by preparative TLC or recrystallization to afford the corresponding product.

**General Procedure for the Synthesis of benzimidazo[1,2-a]-1,3,5-triazines on 5 mmol Scale.**

To a 25 mL pressure tube with teflon cover was added 2-aminobenzimidazole (665.8 mg, 5 mmol), benzaldehyde (1.53 mL, 15 mmol), ammonium iodide (1.09 mg, 7.5 mmol), $NaHCO_3$ (420.1 mg, 5 mmol), 4Å molecular sieves (2.5 g), DMSO (355.1 μL, 5 mmol) and chlorobenzene (12 mL). The reaction vessel was sealed under argon and stirred at 140 ℃ for 24 h. After the mixture was cooled to room temperature, the volatiles were removed under reduced pressure. The residue was purified by column chromatography on silica gel (PE/EtOAc=5:1) to give the desired product **3aa** as a yellow solid (1.01 g, 63% yield).

## 3. Characterization data of products

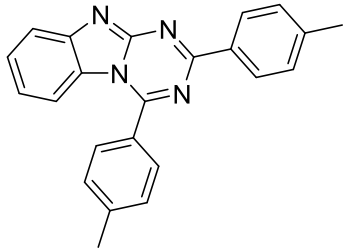
**2,4-Diphenylbenzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3aa)**[1]



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), benzaldehyde (61.2 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3aa** as yellow solid; yield: 50.9 mg (79%), mp 241-243 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.70 (d, *J* = 6.7 Hz, 2H), 7.96 (d, *J* = 8.2 Hz, 1H), 7.90 (d, *J* = 7.0 Hz, 2H), 7.78 (t, *J* = 7.4 Hz, 1H), 7.71 (t, *J* = 7.4 Hz, 2H), 7.58-7.49 (m, 4H), 7.16 (t, *J* = 7.8 Hz, 1H), 7.04 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 162.7, 158.4, 152.1, 145.1, 135.4, 132.3, 132.2, 129.4, 129.2, 128.6, 128.4, 127.2, 126.0, 122.3, 120.4, 114.7.

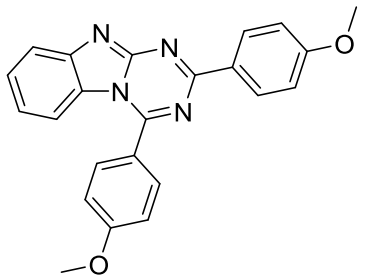
**2,4-Di-p-tolylbenzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ab)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-methylbenzaldehyde (71.1 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ab** as yellow solid; yield: 49.1 mg (70%), mp 250-253 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.58 (d, *J* = 8.2 Hz, 2H), 7.94 (d, *J* = 8.2 Hz, 1H), 7.80 (d, *J* = 8.0 Hz, 2H), 7.49 (d, *J* = 7.8 Hz, 3H), 7.32 (d, *J* = 8.1 Hz, 2H), 7.16-7.12 (m, 2H), 2.57 (s, 3H), 2.45 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 162.8, 158.4, 152.3, 145.1, 143.0, 142.9, 132.8, 129.8, 129.4, 129.4, 129.3, 128.4, 127.0, 126.1, 122.0, 120.3, 114.8, 21.8, 21.7. HRMS (ESI): *m/z* calcd for $C_{23}H_{19}N_4^+$ $[M+H]^+$ 351.1604, found 351.1604.

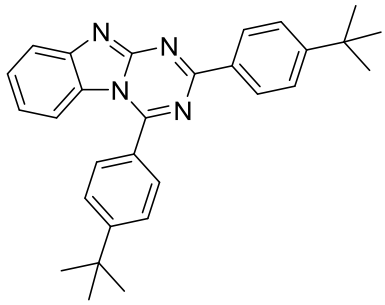
**2,4-Bis(4-methoxyphenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ac)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-methoxybenzaldehyde (72.9 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ac** as yellow solid; yield: 60.9 mg (80%), mp 208-210 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.65 (d, *J* = 8.8 Hz, 2H), 7.91 (t, *J* = 9.3 Hz, 3H), 7.50 (t, *J* = 7.6 Hz, 1H), 7.24 (d, *J* = 8.6 Hz, 1H), 7.20-7.12 (m, 3H), 7.02 (d, *J* = 8.9 Hz, 2H), 3.98 (s, 3H), 3.90 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 163.1, 162.9, 162.4, 158.0, 152.7, 145.1, 131.3, 130.5, 128.2, 126.9, 126.2, 124.4, 121.7, 120.1, 114.7, 114.4, 113.9, 55.6, 55.4. HRMS (ESI): *m/z* calcd for $C_{23}H_{19}N_4O_2^+$ $[M+H]^+$ 383.1503, found 383.1503.

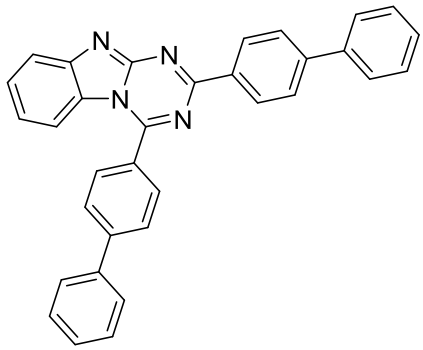
**2,4-Bis(4-(*tert*-butyl)phenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ad)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-(tert-butyl)benzaldehyde (100.3 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ad** as yellow solid; yield: 65.5 mg (75%), mp 273-276 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.62 (d, *J* = 8.5 Hz, 2H), 7.95 (d, *J* = 8.2 Hz, 1H), 7.86 (d, *J* = 8.4 Hz, 2H), 7.70 (d, *J* = 8.4 Hz, 2H), 7.57-7.48 (m, 3H), 7.16 (d, *J* = 4.2 Hz, 2H), 1.47 (s, 9H), 1.38 (s, 9H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 162.8, 158.4, 156.2, 156.0, 152.4, 145.1, 132.8, 129.4, 129.3, 128.3, 127.0, 126.2, 126.1, 125.6, 122.0, 120.3, 114.8, 35.3, 35.1, 31.2, 31.2. HRMS (ESI): *m/z* calcd for $C_{29}H_{31}N_4^+$ $[M+H]^+$ 435.2543, found 435.2543.

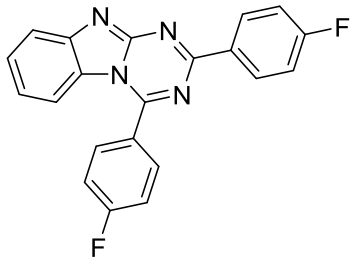
**2,4-Di([1,1'-biphenyl]-4-yl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ae)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-phenylbenzaldehyde (109.3 mg, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ae** as yellow solid; yield: 64.6 mg (68%), mp 227-230 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.78 (d, *J* = 8.3 Hz, 2H), 8.03 (d, *J* = 8.2 Hz, 2H), 7.98 (d, *J* = 8.2 Hz, 1H), 7.94 (d, *J* = 8.2 Hz, 2H), 7.77 (d, *J* = 8.3 Hz, 4H), 7.69 (d, *J* = 7.5 Hz, 2H), 7.55 (q, *J* = 6.8, 6.4 Hz, 3H), 7.50-7.46 (m, 3H), 7.40 (t, *J* = 7.3 Hz, 1H), 7.25 (d, *J* = 7.6 Hz, 1H), 7.18 (t, *J* = 7.7 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 162.4, 158.2, 152.2, 145.3, 145.2, 144.9, 140.1, 139.5, 134.4, 130.8, 129.9, 129.1, 128.8, 128.5, 128.0, 127.7, 127.3, 127.2, 127.2, 126.1, 122.2, 120.4, 114.8. HRMS (ESI): *m/z* calcd for $C_{33}H_{23}N_4^+$ $[M+H]^+$ 475.1917, found 475.1917.

**2,4-Bis(4-fluorophenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3af)**

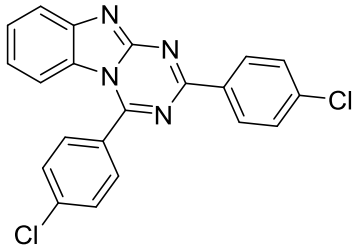


The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-fluorobenzaldehyde (64.4 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by recrystallization from EtOAc to give **3af** as yellow solid; yield: 54.4 mg (76%), mp 292-294 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.68 (dd, *J* = 8.7, 5.6 Hz, 2H), 7.94 (t, *J* = 7.0 Hz, 3H), 7.54 (t, *J* = 7.6 Hz, 1H), 7.41 (t, *J* = 8.5 Hz, 2H), 7.19 (t, *J* = 8.6 Hz, 3H), 7.10 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 165.7 (d, *J* = 252.1 Hz), 165.1 (d, *J* = 253.0 Hz), 161.7, 157.5, 151.9, 145.2, 131.8 (d, *J* = 9.0 Hz), 131.6 (d, *J* = 2.8 Hz), 131.0 (d, *J* = 8.8 Hz), 128.3(d, *J* = 3.4

Hz), 127.4, 125.9, 122.5, 120.6, 116.7 (d, *J* = 22.1 Hz), 115.8(d, *J* = 21.8 Hz), 114.5. HRMS (ESI): *m/z* calcd for $C_{21}H_{13}F_2N_4^+$ $[M+H]^+$ 359.1103, found 359.1103.

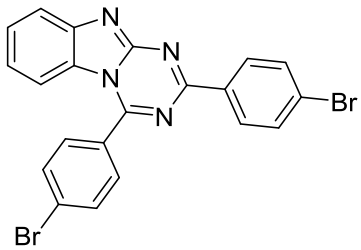
**2,4-Bis(4-chlorophenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ag)**




The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-chlorobenzaldehyde (84.3 mg, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by recrystallization from EtOAc to give **3ag** as yellow solid; yield: 62.7 mg (80%), mp 325-327 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.60 (d, *J* = 8.6 Hz, 2H), 7.96 (d, *J* = 8.2 Hz, 1H), 7.88 (d, *J* = 8.4 Hz, 2H), 7.70 (d, *J* = 8.4 Hz, 2H), 7.54 (t, *J* = 7.7 Hz, 1H), 7.49 (d, *J* = 8.6 Hz, 2H), 7.21 (t, *J* = 7.8 Hz, 1H), 7.12 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 161.7, 157.5, 151.8, 145.3, 139.0, 138.9, 133.9, 130.7, 130.4, 130.0, 129.7, 129.0, 127.6, 125.9, 122.7, 120.8, 114.6. HRMS (ESI): *m/z* calcd for $C_{21}H_{13}Cl_2N_4^+$ $[M+H]^+$ 391.0512, found 391.0513.

**2,4-Bis(4-bromophenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ah)**

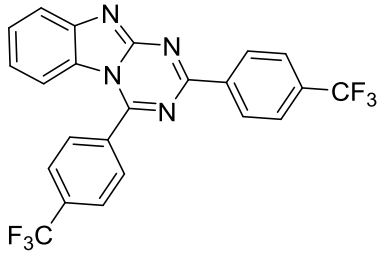



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-bromobenzaldehyde (111.0 mg, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by recrystallization from EtOAc to give **3ah** as yellow solid; yield: 70.8 mg (74%), mp 330-332 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.52 (d, *J* = 8.4 Hz, 2H), 7.96 (d, *J* = 8.2 Hz, 1H), 7.87 (d, *J* = 8.2 Hz, 2H), 7.80 (d, *J* = 8.2 Hz, 2H), 7.65 (d, *J* = 8.4 Hz, 2H), 7.55 (t, *J* = 7.7 Hz, 1H), 7.21 (t, *J* = 7.8 Hz, 1H), 7.12 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 161.8, 157.6, 151.7,

145.3, 134.3, 132.7, 132.0, 130.9, 130.9, 130.1, 127.6, 127.6, 127.4, 125.9, 122.7, 120.8, 114.6. HRMS (ESI): *m/z* calcd for $C_{21}H_{13}Br_2N_4^+$ $[M+H]^+$ 478.9501, found 478.9501.

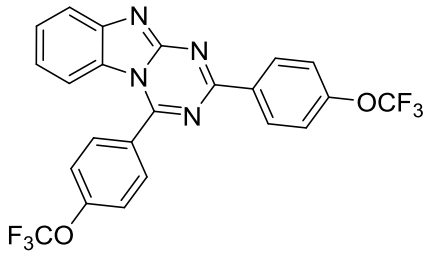
**2,4-Bis(4-(trifluoromethyl)phenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ai)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-trifluoromethylbenzaldehyde (81.9 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ai** as yellow solid; yield: 59.6 mg (65%), mp 284-287 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.76 (d, *J* = 8.2 Hz, 2H), 8.09 (d, *J* = 8.1 Hz, 2H), 8.02 (d, *J* = 8.1 Hz, 2H), 7.98 (d, *J* = 8.1 Hz, 1H), 7.78 (d, *J* = 8.2 Hz, 2H), 7.57 (t, *J* = 7.7 Hz, 1H), 7.23 (t, *J* = 7.9 Hz, 1H), 7.02 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 161.0, 157.3, 151.2, 145.2, 138.4, 135.3, 134.4 (q, *J* = 32.8 Hz), 133.7 (q, *J* = 32.7 Hz), 129.6, 129.1, 127.9, 126.4 (q, *J* = 3.7 Hz), 125.6 (q, *J* = 3.6 Hz), 123.8 (q, *J* = 270.8 Hz), 123.4 (q, *J* = 273.2 Hz), 123.1, 120.9, 114.5. HRMS (ESI): *m/z* calcd for $C_{23}H_{13}F_6N_4^+$ $[M+H]^+$ 459.1039, found 459.1039.

**2,4-Bis(4-(trifluoromethoxy)phenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3aj)**

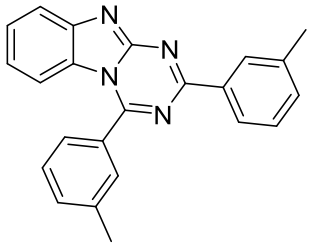


The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-trifluoromethoxybenzaldehyde (85.7 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3aj** as yellow solid; yield: 66.6 mg (68%), mp 216-218 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.71 (d, *J* = 8.8 Hz, 2H), 8.00 (d, *J* = 8.6 Hz, 2H), 7.97 (d, *J* = 8.5 Hz, 1H), 7.56 (d, *J* = 8.1 Hz, 3H), 7.35 (d, *J* = 8.5 Hz, 2H), 7.22 (t, *J* = 7.8 Hz, 1H), 7.09 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 161.2, 157.3, 152.4, 152.2, 151.6, 145.1,

133.6, 131.1, 130.6, 130.3, 127.5, 125.7, 122.7, 121.3, 120.7, 120.5, 120.4 (q, $J$ = 256.9 Hz), 120.3 (q, $J$ = 259.5 Hz), 114.5. HRMS (ESI): *m/z* calcd for $C_{23}H_{13}F_6N_4O_2^+$ $[M+H]^+$ 491.0937, found 491.0938.

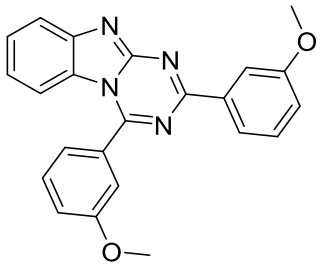
**2,4-Di-*m*-tolylbenzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ak)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 3-methylbenzaldehyde (70.8 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ak** as yellow solid; yield: 43.9 mg (63%), mp 208-211 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.52 (s, 1H), 8.49 (d, $J$ = 7.5 Hz, 1H), 7.96 (d, $J$ = 8.2 Hz, 1H), 7.69 (s, 1H), 7.67 (d, $J$ = 5.0 Hz, 1H), 7.58 (d, $J$ = 5.2 Hz, 2H), 7.52 (t, $J$ = 7.7 Hz, 1H), 7.43-7.36 (m, 2H), 7.15 (t, $J$ = 7.8 Hz, 1H), 7.02 (d, $J$ = 8.4 Hz, 1H), 2.53 (s, 3H), 2.47 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 162.9, 158.5, 152.0, 145.1, 139.3, 138.2, 135.4, 133.1, 133.0, 132.1, 129.9, 129.0, 128.7, 128.4, 127.1, 126.6, 126.1, 125.3, 122.2, 120.3, 114.7, 21.5, 21.4. HRMS (ESI): *m/z* calcd for $C_{23}H_{19}N_4^+$ $[M+H]^+$ 351.1604, found 351.1604.

**2,4-Bis(3-methoxyphenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3al)**

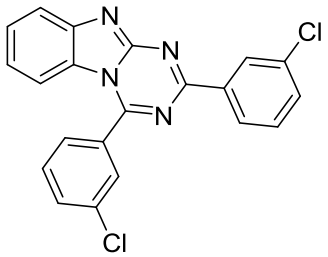


The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 3-methoxybenzaldehyde (73.1 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3al** as yellow solid; yield: 57.2 mg (75%), mp 203-205 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.30 (d, $J$ = 7.7 Hz, 1H), 8.25 (s, 1H), 7.96 (d, $J$ = 8.1 Hz, 1H), 7.61 (t, $J$ = 8.0 Hz, 1H), 7.53 (t, $J$ = 7.7 Hz, 1H), 7.43 (t, $J$ = 8.2 Hz, 2H), 7.37 (s, 1H), 7.30

(d, *J* = 8.3 Hz, 1H), 7.18 (t, *J* = 7.8 Hz, 1H), 7.13 (d, *J* = 8.1 Hz, 1H), 7.08 (d, *J* = 8.3 Hz, 1H), 3.94 (s, 3H), 3.91 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 162.6, 160.1, 159.9, 158.1, 152.0, 145.2, 136.9, 133.3, 130.5, 129.6, 127.3, 126.1, 122.4, 122.2, 120.5, 120.4, 119.4, 118.3, 114.9, 113.5, 113.2, 55.7, 55.6. HRMS (ESI): *m/z* calcd for $C_{23}H_{19}N_4O_2^+$ $[M+H]^+$ 383.1503, found 383.1503.

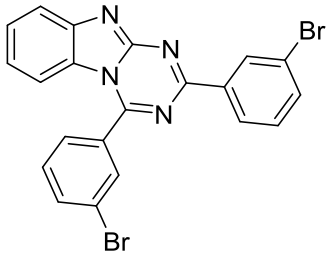
**2,4-Bis(3-chlorophenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3am)**




The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 3-chlorobenzaldehyde (70.3 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by recrystallization from EtOAc to give **3am** as yellow solid; yield: 70.1 mg (90%), mp 288-290 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.65 (s, 1H), 8.56 (d, *J* = 7.8 Hz, 1H), 7.97 (d, *J* = 8.2 Hz, 1H), 7.91 (s, 1H), 7.78 (t, *J* = 8.6 Hz, 2H), 7.67 (t, *J* = 7.9 Hz, 1H), 7.57–7.52 (m, 2H), 7.46 (t, *J* = 7.8 Hz, 1H), 7.22 (t, *J* = 7.8 Hz, 1H), 7.07 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 161.3, 157.1, 151.4, 145.2, 137.1, 135.6, 134.9, 133.6, 132.6, 132.3, 130.6, 129.9, 129.3, 128.6, 127.6, 127.4, 126.5, 125.8, 122.9, 120.8, 114.6. HRMS (ESI): *m/z* calcd for $C_{21}H_{13}Cl_2N_4^+$ $[M+H]^+$ 391.0512, found 391.0513.

**2,4-Bis(3-bromophenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3an)**

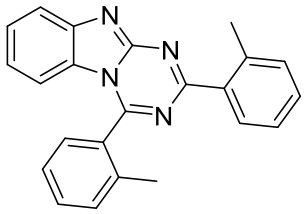



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 3-bromobenzaldehyde (70.0 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by recrystallization from EtOAc to give **3an** as

yellow solid; yield: 53.1 mg (55%), mp 280-283 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.82 (s, 1H), 8.61 (d, *J* = 7.9 Hz, 1H), 8.06 (s, 1H), 7.99 (d, *J* = 8.2 Hz, 1H), 7.93 (d, *J* = 7.9 Hz, 1H), 7.83 (d, *J* = 7.6 Hz, 1H), 7.70 (d, *J* = 7.7 Hz, 1H), 7.60–7.55 (m, 2H), 7.41 (t, *J* = 7.9 Hz, 1H), 7.23 (t, *J* = 7.9 Hz, 1H), 7.07 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 164.3, 161.2, 157.0, 151.5, 145.3, 137.3, 135.5, 135.3, 133.8, 132.3, 131.4, 130.8, 130.2, 127.9, 127.7, 126.9, 125.8, 123.4, 122.9, 120.9, 114.6. HRMS (ESI): *m/z* calcd for $C_{21}H_{13}Br_2N_4^+$ $[M+H]^+$ 478.9501, found 478.9501.

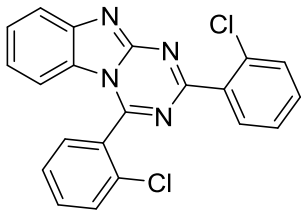
**2,4-Di-o-tolylbenzo[4,5]imidazo[1,2-a][1,3,5]triazine (3ao)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 2-methylbenzaldehyde (69.3 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ao** as yellow solid; yield: 41.7 mg (60%), mp 198-201 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.32 (d, *J* = 7.5 Hz, 1H), 7.97 (d, *J* = 8.2 Hz, 1H), 7.64 (t, *J* = 8.1 Hz, 1H), 7.54–7.48 (d, *J* = 7.5 Hz, 4H), 7.42 (t, *J* = 7.3 Hz, 1H), 7.34 (t, *J* = 7.3 Hz, 2H), 7.13 (t, *J* = 7.8 Hz, 1H), 6.50 (d, *J* = 8.4 Hz, 1H), 2.86 (s, 3H), 2.21 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 165.3, 158.1, 151.1, 144.9, 139.6, 136.0, 135.3, 132.0, 132.0, 131.7, 131.6, 131.2, 131.1, 127.8, 127.3, 126.9, 126.0, 125.8, 123.0, 120.4, 114.0, 22.6, 19.1. HRMS (ESI): *m/z* calcd for $C_{23}H_{19}N_4^+$ $[M+H]^+$ 351.1604, found 351.1604.

**2,4-Bis(2-chlorophenyl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ap)**

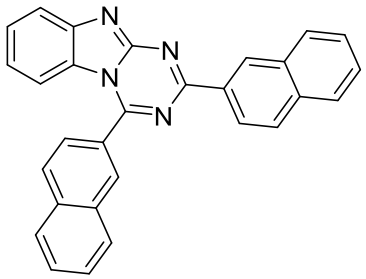


The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 2-chlorobenzaldehyde (67.6 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to

give **3ap** as yellow solid; yield: 28.2 mg (36%), mp 219-221 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.10 (d, *J* = 9.3 Hz, 1H), 8.03 (d, *J* = 8.2 Hz, 1H), 7.76–7.66 (m, 3H), 7.66–7.51 (m, 3H), 7.48–7.39 (m, 2H), 7.21 (t, *J* = 7.8 Hz, 1H), 6.64 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 163.5, 155.9, 150.4, 145.0, 135.5, 133.7, 133.0, 132.8, 132.4, 131.6, 131.4, 131.1, 130.5, 129.9, 128.1, 127.6, 126.9, 125.6, 123.5, 120.8, 113.8. HRMS (ESI): *m/z* calcd for $C_{21}H_{13}Cl_2N_4^+$ $[M+H]^+$ 391.0512, found 391.0513.

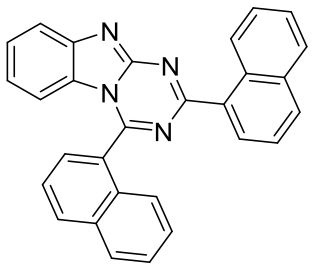
**2,4-Di(naphthalen-2-yl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3aq)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 2-naphthaldehyde (93.7 mg, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3aq** as yellow solid; yield: 52.6 mg (62%), mp 285-287 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 9.27 (s, 1H), 8.78 (d, *J* = 8.7 Hz, 1H), 8.52 (s, 1H), 8.19 (d, *J* = 8.5 Hz, 1H), 8.06 (d, *J* = 8.5 Hz, 2H), 8.02–7.95 (m, 4H), 7.89 (d, *J* = 7.9 Hz, 1H), 7.76–7.67 (m, 2H), 7.59–7.51 (m, 3H), 7.10 (d, *J* = 5.6 Hz, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 162.7, 158.4, 152.2, 145.3, 135.4, 134.9, 133.0, 132.9, 132.7, 130.8, 129.5, 129.4, 129.4, 129.2, 129.0, 128.6, 128.3, 128.1, 127.9, 127.7, 127.5, 127.2, 126.4, 126.2, 125.3, 124.5, 122.3, 120.5, 114.8. HRMS (ESI): *m/z* calcd for $C_{29}H_{19}N_4^+$ $[M+H]^+$ 423.1604, found 423.1605.

**2,4-Di(naphthalen-1-yl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ar)**

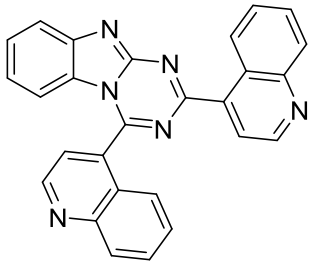


The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 1-naphthaldehyde (81.5 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue

was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ar** as yellow solid; yield: 36.5 mg (43%), mp 140-143 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 9.55 (d, *J* = 8.6 Hz, 1H), 8.68 (d, *J* = 7.3 Hz, 1H), 8.26 (d, *J* = 8.3 Hz, 1H), 8.08 (d, *J* = 8.3 Hz, 1H), 8.04 (d, *J* = 8.0 Hz, 1H), 7.98 (d, *J* = 8.2 Hz, 1H), 7.94 (d, *J* = 8.0 Hz, 1H), 7.89 (d, *J* = 7.0 Hz, 1H), 7.78 (t, *J* = 7.8 Hz, 1H), 7.67–7.55 (m, 5H), 7.47 (t, *J* = 7.7 Hz, 2H), 6.95 (t, *J* = 7.8 Hz, 1H), 6.19 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 164.8, 157.6, 151.1, 144.9, 134.2, 133.5, 132.9, 132.4, 132.0, 131.9, 131.3, 129.7, 129.4, 128.9, 128.6, 128.2, 127.7, 127.3, 127.2, 126.9, 126.5, 126.1, 125.7, 125.4, 124.9, 124.0, 122.8, 120.2, 114.6. HRMS (ESI): m/z calcd for $C_{29}H_{19}N_4^+$ $[M+H]^+$ 423.1604, found 423.1605.

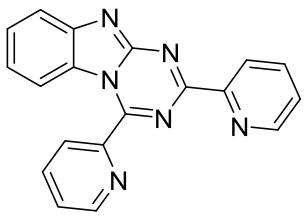
**2,4-Di(quinolin-4-yl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3as)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 4-quinolinecarboxaldehyde (94.3 mg, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 30:1) to give **3as** as yellow solid; yield: 32.3 mg (38%), mp 271-273 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 9.47 (d, *J* = 8.5 Hz, 1H), 9.30 (d, *J* = 4.3 Hz, 1H), 9.07 (d, *J* = 4.6 Hz, 1H), 8.40 (d, *J* = 8.2 Hz, 2H), 8.23 (d, *J* = 8.5 Hz, 1H), 8.03 (d, *J* = 8.5 Hz, 1H), 7.91–7.87 (m, 1H), 7.81–7.76 (m, 2H), 7.69 (t, *J* = 7.6 Hz, 1H), 7.58–7.50 (m, 3H), 7.04 (t, *J* = 7.9 Hz, 1H), 6.30 (d, *J* = 8.4 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 162.6, 155.7, 150.3, 150.0, 149.9, 149.5, 148.7, 145.2, 139.4, 137.0, 131.1, 130.8, 130.2, 129.7, 129.1, 128.2, 128.1, 126.3, 125.4, 125.2, 124.0, 123.9, 123.7, 123.7, 121.0, 120.3, 114.3. HRMS (ESI): *m/z* calcd for $C_{27}H_{17}N_6^+$ $[M+H]^+$ 425.1509, found 425.1509.

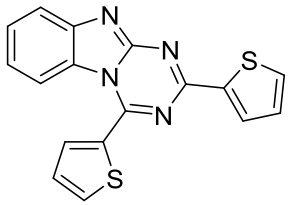
**2,4-Di(pyridin-2-yl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3at)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 2-pyridinecarboxaldehyde (57.1 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 30:1) to give **3at** as yellow solid; yield: 32.1 mg (50%), mp 159-161 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 10.20 (d, *J* = 7.0 Hz, 1H), 8.85 (d, *J* = 4.0 Hz, 1H), 8.67 (d, *J* = 4.1 Hz, 1H), 8.62 (d, *J* = 9.0 Hz, 1H), 8.40 (d, *J* = 8.1 Hz, 2H), 7.91 (t, *J* = 7.4 Hz, 1H), 7.81 (t, *J* = 7.7 Hz, 1H), 7.47 (t, *J* = 5.9 Hz, 1H), 7.39 (t, *J* = 7.8 Hz, 1H), 7.29 (t, *J* = 6.3 Hz, 1H), 7.04 (t, *J* = 6.9 Hz, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 186.5, 156.0, 150.4, 149.4, 148.2, 138.2, 136.7, 136.3, 135.9, 129.8, 129.3, 127.5, 127.2, 125.6, 125.4, 123.0, 122.8, 120.4, 115.7. HRMS (ESI): m/z calcd for $C_{19}H_{13}N_6^+$ $[M+H]^+$ 325.1196, found 325.1197.

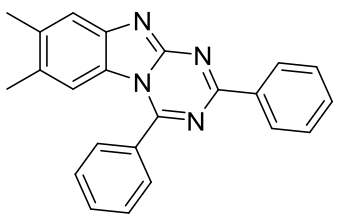
**2,4-Di(thiophen-2-yl)benzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3au)**



The reaction was conducted with 2-aminobenzimidazole (26.6 mg, 0.2 mmol), 2-thiophenecarboxaldehyde (56.1 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3au** as yellow solid; yield: 30.3 mg (45%), mp 258-260 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.23 (d, *J* = 3.0 Hz, 1H), 8.01 (d, *J* = 3.1 Hz, 1H), 7.93 (d, *J* = 7.0 Hz, 1H), 7.84 (d, *J* = 5.0 Hz, 1H), 7.70 (d, *J* = 8.3 Hz, 1H), 7.62 (d, *J* = 4.6 Hz, 1H), 7.53 (t, *J* = 7.4 Hz, 1H), 7.34 (t, *J* = 4.4 Hz, 1H), 7.23 (t, *J* = 8.0 Hz, 1H), 7.19 (t, *J* = 4.2 Hz, 1H).$^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 158.6, 152.9, 145.3, 141.2, 133.3, 132.9, 132.8, 132.2, 131.5, 128.6, 127.9, 127.3, 122.1, 120.5, 114.4. HRMS (ESI): *m/z* calcd for $C_{17}H_{11}N_4S_2^+$ $[M+H]^+$ 335.0420, found 335.0420.

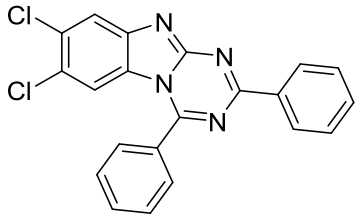
**7,8-Dimethyl-2,4-diphenylbenzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ba)**[1]



The reaction was conducted with 5,6-dimethyl-1*H*-benzo[*d*]imidazol-2-amine (32.2 mg, 0.2 mmol), benzaldehyde (61.2 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ba** as yellow solid; yield: 54.5 mg (78%), mp 291-294 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.68 (d, *J* = 6.4 Hz, 2H), 7.90 (d, *J* = 7.2 Hz, 2H), 7.78 (t, *J* = 7.4 Hz, 1H), 7.74–7.66 (m, 3H), 7.57–7.49 (m, 3H), 6.79 (s, 1H), 2.39 (s, 3H), 2.22 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 161.9, 157.8, 151.5, 143.7, 136.9, 135.6, 132.4, 132.2, 132.0, 131.7, 129.2, 129.1, 128.5, 128.5, 124.3, 120.2, 114.8, 20.7, 20.5.

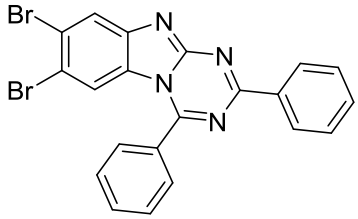
**7,8-Dichloro-2,4-diphenylbenzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3ca)**




The reaction was conducted with 5,6-dichloro-1*H*-benzo[*d*]imidazol-2-amine (40.2 mg, 0.2 mmol), benzaldehyde (61.2 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ca** as yellow solid; yield: 59.5 mg (76%), mp 320-322 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.66 (d, *J* = 7.3 Hz, 2H), 8.03 (s, 1H), 7.89 (d, *J* = 7.0 Hz, 2H), 7.83 (t, *J* = 7.5 Hz, 1H), 7.75 (t, *J* = 7.4 Hz, 2H), 7.58 (t, *J* = 7.2 Hz, 1H), 7.52 (t, *J* = 7.3 Hz, 2H), 7.14 (s, 1H).$^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 163.6, 158.3, 153.2, 144.3, 135.0, 132.9, 132.9, 131.7, 131.4, 129.6, 129.5, 128.7, 128.3, 126.2, 125.0, 121.4, 116.0. HRMS (ESI): *m/z* calcd for $C_{21}H_{13}C_{l2}N_4^+$ $[M+H]^+$ 391.0512, found 391.0512.

**7,8-Dibromo-2,4-diphenylbenzo[4,5]imidazo[1,2-*a*][1,3,5]triazine (3da)**

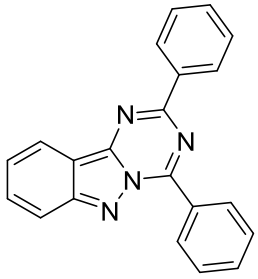



The reaction was conducted with 5,6-dibromo-1*H*-benzo[*d*]imidazol-2-amine (57.8 mg, 0.2 mmol), benzaldehyde (61.2 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6

mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3da** as yellow solid; yield: 80.1 mg (83%), mp 349-351 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 8.67 (d, *J* = 7.6 Hz, 2H), 8.21 (s, 1H), 7.89 (d, *J* = 7.5 Hz, 2H), 7.83 (t, *J* = 7.2 Hz, 1H), 7.75 (t, *J* = 7.5 Hz, 2H), 7.59 (t, *J* = 6.8 Hz, 1H), 7.53 (t, *J* = 7.4 Hz, 2H), 7.32 (s, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 163.8, 158.4, 153.1, 145.2, 135.1, 133.0, 132.9, 131.5, 129.7, 129.5, 128.8, 128.3, 126.0, 124.7, 123.3, 119.1, 117.4. HRMS (ESI): *m/z* calcd for $C_{21}H_{13}Br_2N_4^+$ $[M+H]^+$ 478.9501, found 478.9501.

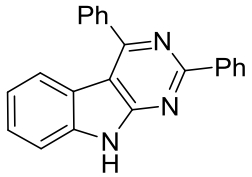
**2,4-Diphenyl-[1,3,5]triazino[1,2-*b*]indazole (3ea)**



The reaction was conducted with 1*H*-indazol-3-amine (26.6 mg, 0.2 mmol), benzaldehyde (61.2 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC ($CH_2Cl_2$/MeOH = 100:1) to give **3ga** as yellow solid; yield: 36.2 mg (56%), mp 225-227 ℃.

$^1$H NMR (400 MHz, $CDCl_3$, ppm) δ 9.11 (d, *J* = 7.9 Hz, 2H), 8.75 (d, *J* = 7.7 Hz, 2H), 8.40 (d, *J* = 8.4 Hz, 1H), 7.91 (d, *J* = 8.8 Hz, 1H), 7.70 (d, *J* = 6.9 Hz, 4H), 7.58 (d, *J* = 7.1 Hz, 3H), 7.33 (t, *J* = 7.4 Hz,, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$, ppm) δ 155.3, 152.7, 152.3, 147.5, 136.2, 133.0, 131.8, 131.4, 131.3, 131.0, 128.7, 128.5, 121.7, 121.6, 117.2, 112.5. HRMS (ESI): *m/z* calcd for $C_{21}H_{15}N_4^+$ $[M+H]^+$ 323.1291, found 323.1293.

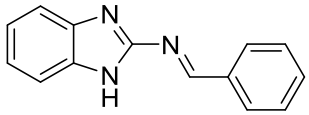
**2,4-Diphenyl-9*H*-pyrimido[4,5-b]indole (3fa)**[2]



The reaction was conducted with 2-aminoindole (26.4 mg, 0.2 mmol), benzaldehyde (61.2 μL, 0.6 mmol), ammonium iodide (43.5 mg, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL). The residue was purified by preparative TLC (PE/EA = 20:1) to give **3fa** as white solid; yield: 36.4 mg (57%), mp 294-296 ℃.

$^1$H NMR (400 MHz, DMSO-$d_6$, ppm) δ 12.56 (s, 1H), 8.56 (d, $J$ = 6.6 Hz, 2H), 8.03 (d, $J$ = 6.4 Hz, 2H), 7.82 (d, $J$ = 8.0 Hz, 1H), 7.67 (q, $J$ = 7.2 Hz, 3H), 7.60-7.49 (m, 5H), 7.20 (t, $J$ = 7.5 Hz, 1H). $^{13}$C NMR (100 MHz, DMSO-$d_6$, ppm) δ 160.4, 160.2, 158.3, 140.2, 139.6, 139.1, 131.4, 131.1, 129.9, 129.8, 129.7, 128.9, 128.6, 122.9, 121.8, 119.9, 113.0, 110.0. HRMS (ESI): *m/z* calcd for $C_{22}H_{16}N_3^+$ $[M+H]^+$ 322.1339, found 322.1338.

**_N_-Benzylidene-1H-benzo[d]imidazol-2-amine (4a)**[3]



The reaction was conducted with 2-aminoindole (26.4 mg, 0.2 mmol), benzaldehyde (30.6 μL, 0.3 mmol), $NaHCO_3$ (16.8 mg, 0.2 mmol), 4Å molecular sieves (100 mg), DMSO (14.2 μL, 0.2 mmol) and chlorobenzene (0.6 mL) stirred at 140 $^o$C for 16 h under oxygen atmosphere. The residue was purified by column chromatography on silica gel (PE/EA = 3:1) to give **4a** as yellow solid; yield: 31.1 mg (70%).

$^1$H NMR (400 MHz, DMSO-$d_6$) δ 12.76 (s, 1H), 9.51 (s, 1H), 8.12 (d, $J$ = 7.0 Hz, 2H), 7.69-7.60 (m, 4H), 7.49 (s, 1H), 7.25-7.20 (m, 2H). $^{13}$C NMR (100 MHz, DMSO-$d_6$) δ 165.4, 155.7, 142.4, 135.1, 134.3, 132.8, 129.5, 129.1, 122.2, 121.9, 118.7, 111.2.

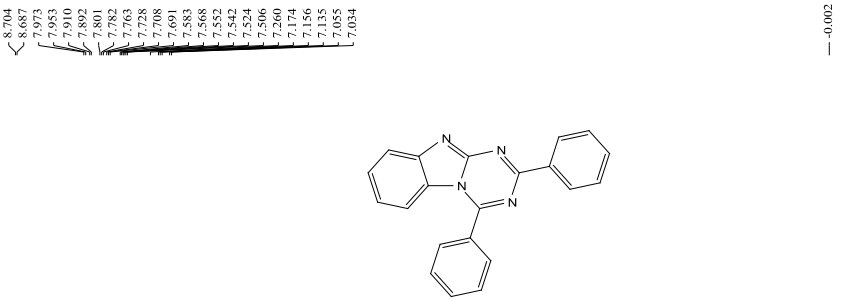
## 4. References


[1] M. M. Wang, Y. G. Meng, W. Wei, J. Wu, W. Q. Yu and J. B. Chang, *Adv. Synth. Catal.*, 2018, **360**, 86;

[2] B. Li, S. H. Guo, J. Zhang, X. Y. Zhang and X. S. Fan, *J. Org. Chem.*, 2015, **80**, 5444.

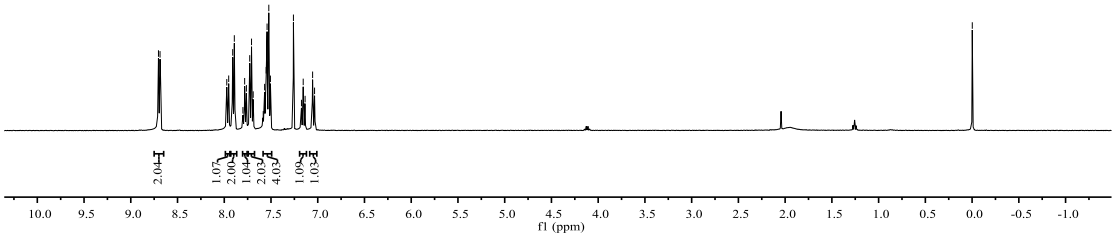
[3] F. Li, Q. K. Kang, H. X. Shan, L. Chen and J. J. Xie, *Eur. J. Org. Chem.*, 2012, 5085.

# 5. $^{1}$H and $^{13}$C NMR spectra of all products

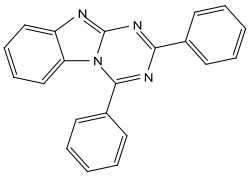




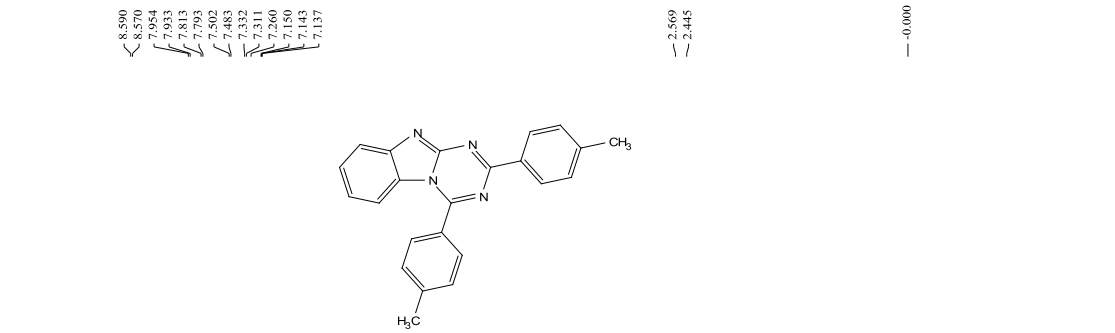

**3aa**



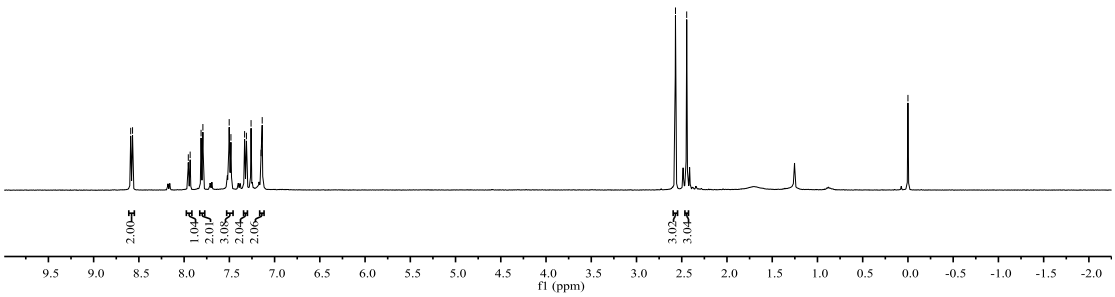


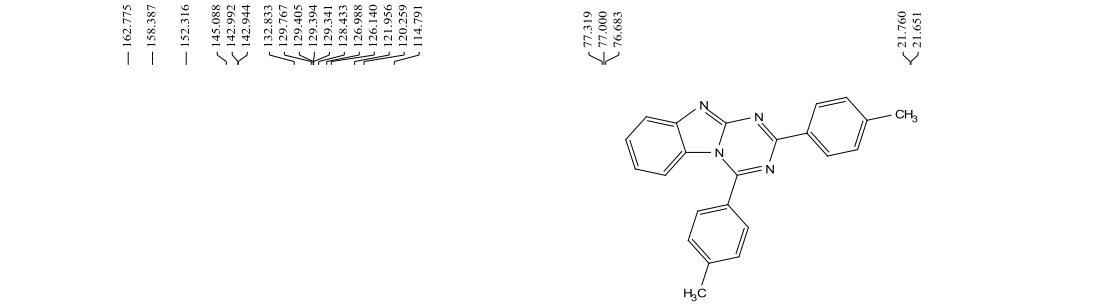


**3aa**


8.590
8.570
7.954
7.933
7.813
7.793
7.502
7.483
7.332
7.311
7.260
7.150
7.143
7.137
2.569
2.445
-0.000
N
N
N
N
CH3
H3C

**3ab**


2.00
1.04
2.01
3.08
2.04
2.06
3.02
3.04
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0 -1.5 -2.0
f1 (ppm)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158.387
152.316
145.088
142.992
142.944
132.833
129.767
129.405
129.394
129.341
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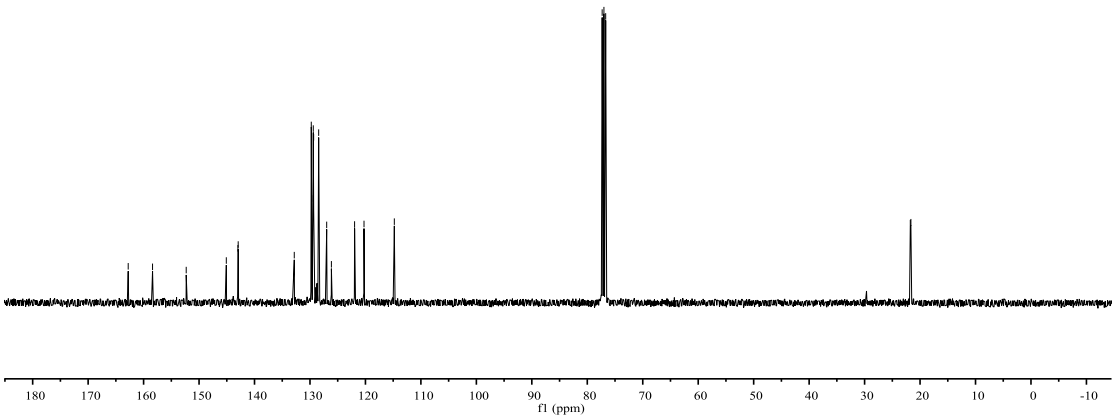
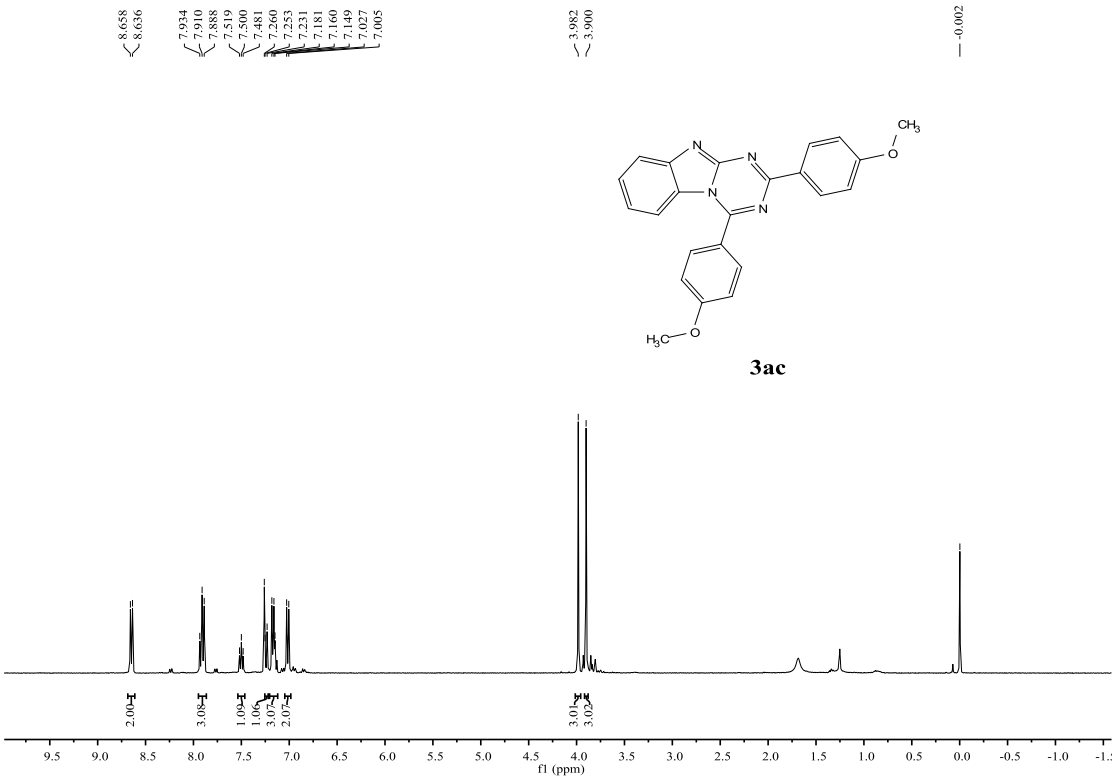
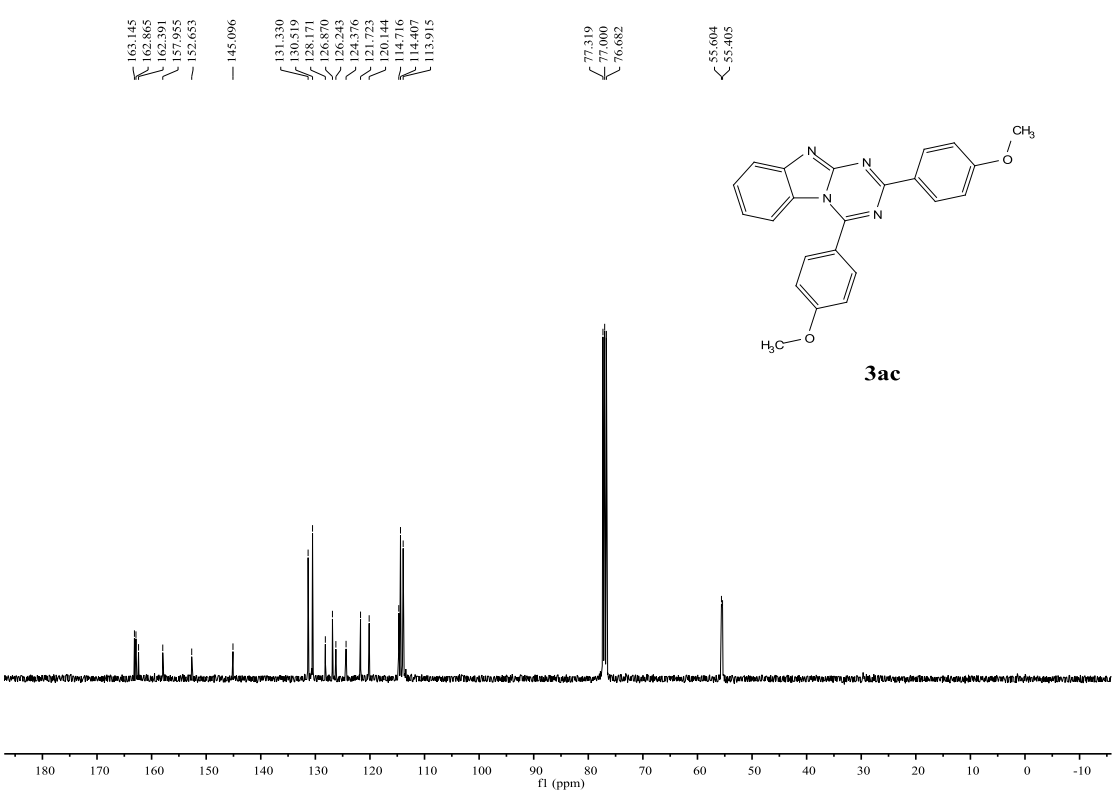
126.988
126.140
121.956
120.259
114.791
77.319
77.000
76.683
21.760
21.651
N
N
N
N
CH3
H3C

**3ab**

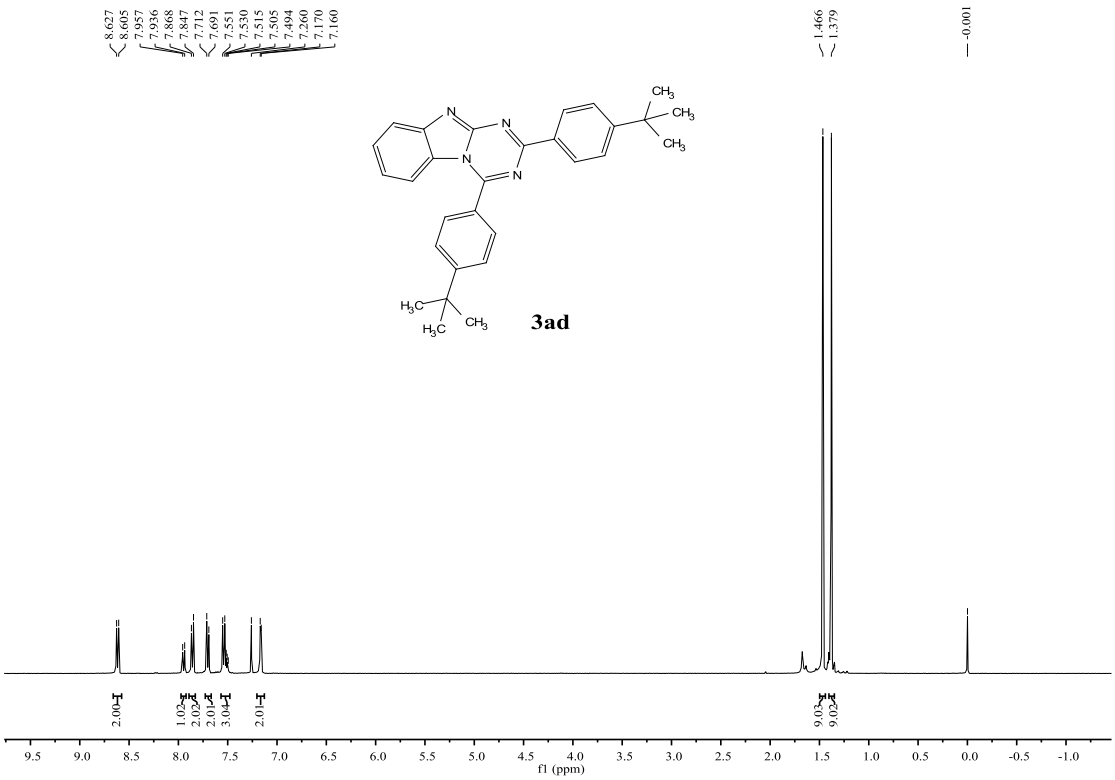

180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


8.658
8.636
7.934
7.910
7.888
7.519
7.500
7.481
7.260
7.253
7.231
7.181
7.160
7.149
7.027
7.005
3.982
3.900
-0.002
2.00
3.08
1.09
1.06
3.07
2.07
3.01
3.02
f1 (ppm)

**3ac**


163.145
162.865
162.391
157.955
152.653
145.096
131.330
130.519
128.171
126.870
126.243
124.376
121.723
120.144
114.716
114.407
113.915
77.319
77.000
76.682
55.604
55.405
f1 (ppm)

**3ac**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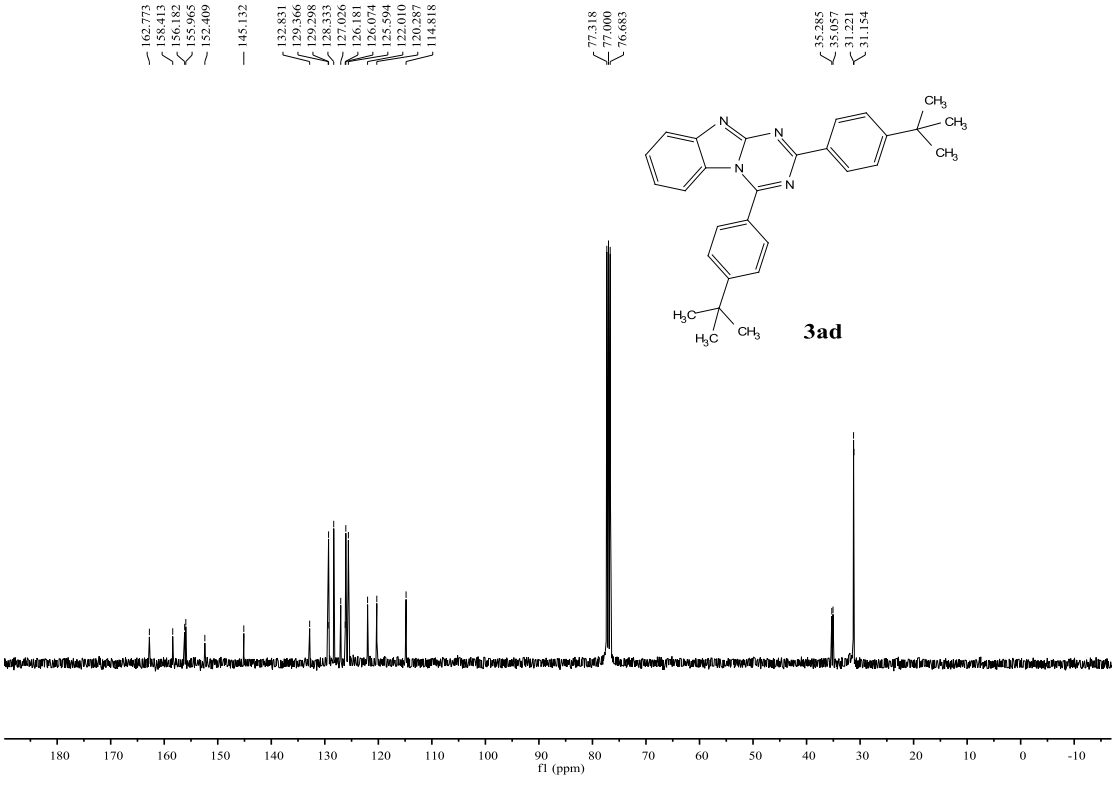
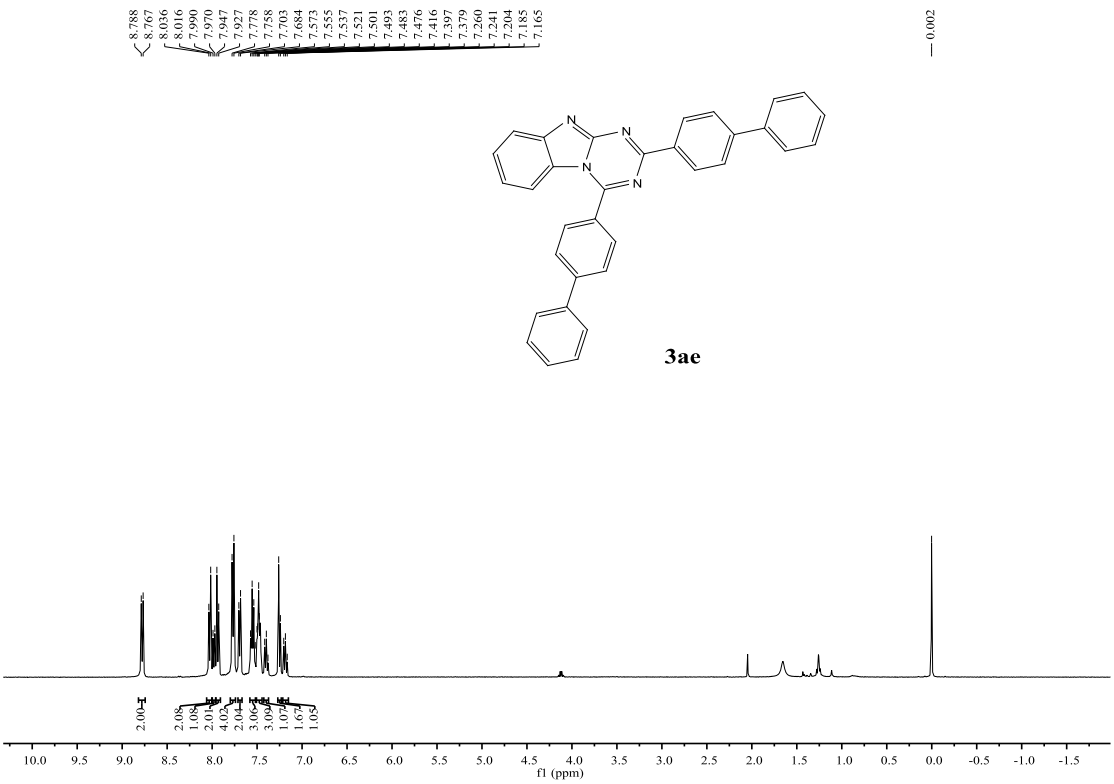
8.605
7.957
7.936
7.868
7.847
7.712
7.691
7.551
7.530
7.515
7.505
7.494
7.260
7.170
7.160
1.466
1.379
-0.001
3ad
2.00
1.02
2.02
2.01
3.04
2.01
9.03
9.02
f1 (ppm)


162.773
158.413
156.182
155.965
152.409
145.132
132.831
129.366
129.298
128.333
127.026
126.181
126.074
125.594
122.010
120.287
114.818
77.318
77.000
76.683
35.285
35.057
31.221
31.154
3ad
f1 (ppm)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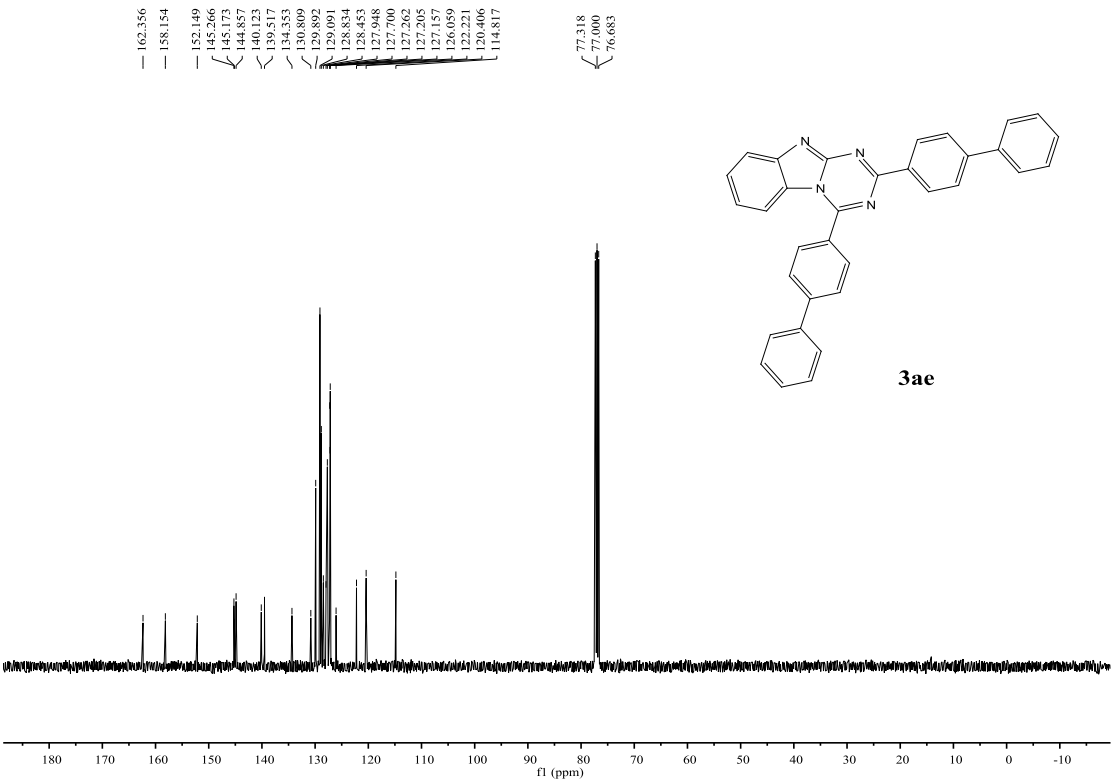
8.767
8.036
8.016
7.990
7.970
7.947
7.927
7.778
7.758
7.703
7.684
7.573
7.555
7.537
7.521
7.501
7.493
7.483
7.476
7.416
7.397
7.379
7.260
7.241
7.204
7.185
7.165
0.002
3ae
2.00
2.08
1.03
2.01
4.02
2.04
3.06
3.09
1.07
1.67
1.05
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0 -1.5
f1 (ppm)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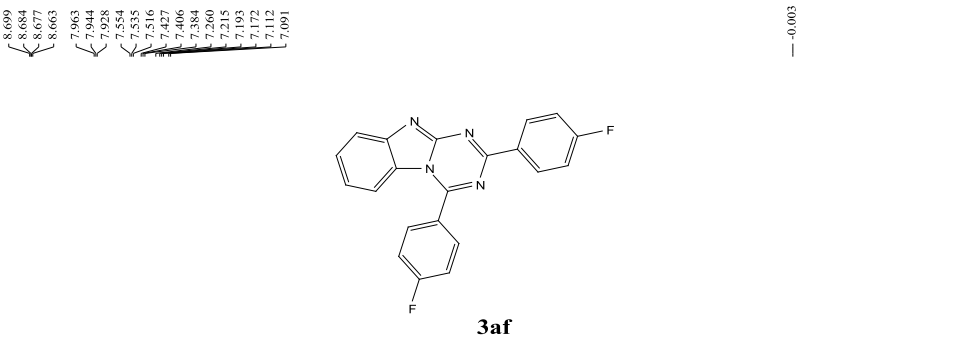
152.149
145.266
145.173
144.857
140.123
139.517
134.353
130.809
129.892
129.091
128.834
128.453
127.948
127.700
127.262
127.205
127.157
126.059
122.221
120.406
114.817
77.318
77.000
76.683
3ae
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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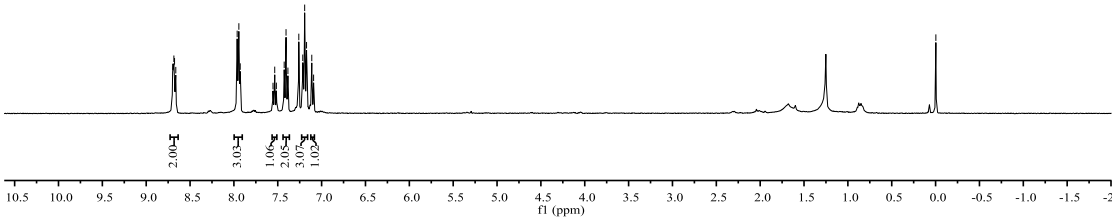
8.684
8.677
8.663
7.963
7.944
7.928
7.554
7.535
7.516
7.427
7.406
7.384
7.260
7.215
7.193
7.172
7.112
7.091
-0.003
N
N
N
N
F
F

**3af**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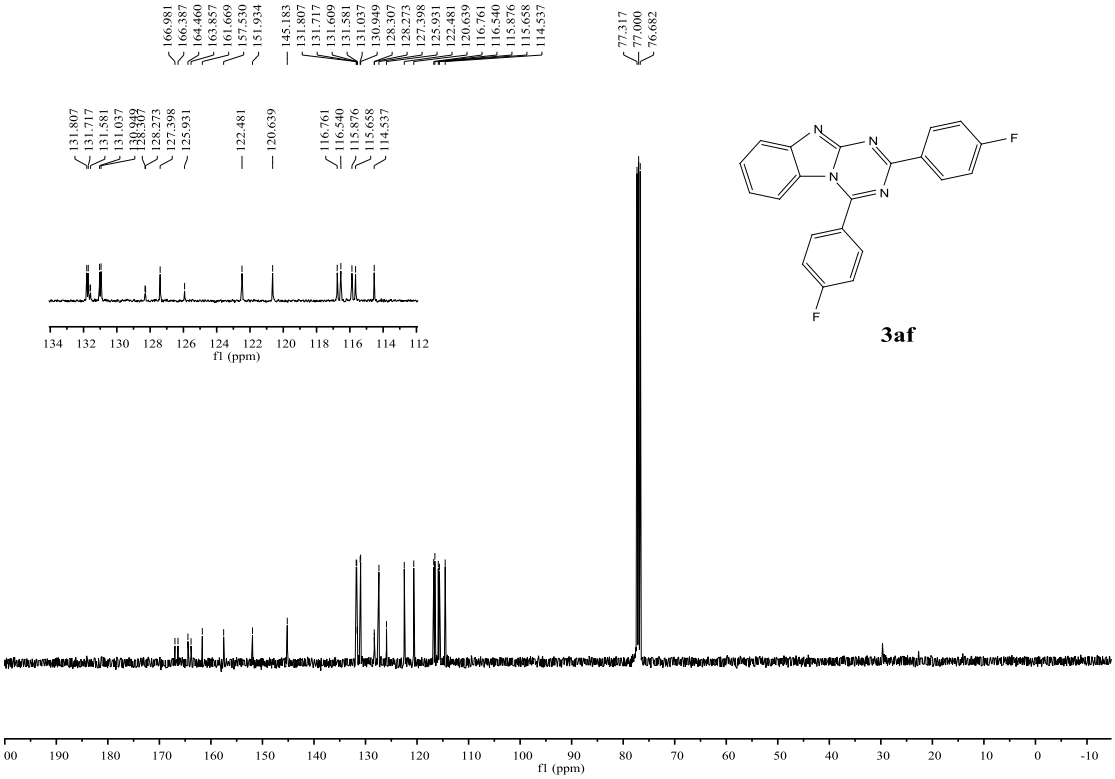
3.03
1.06
2.05
3.07
1.02
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0 -1.5
f1 (ppm)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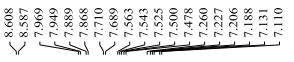
166.387
164.460
163.857
161.669
157.530
151.934
145.183
131.807
131.717
131.609
131.581
131.037
130.949
128.307
128.273
127.398
125.931
122.481
120.639
116.761
116.540
115.876
115.658
114.537
77.317
77.000
76.682
134 132 130 128 126 124 122 120 118 116 114 112
f1 (ppm)
00 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)
N
N
N
N
F
F

**3af**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8.587
7.969
7.949
7.889
7.868
7.710
7.689
7.563
7.543
7.525
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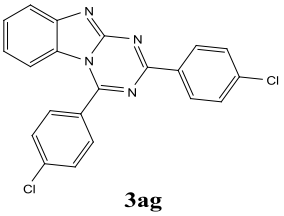
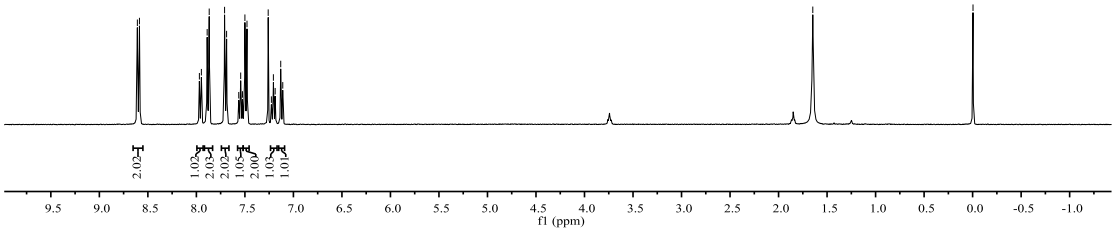
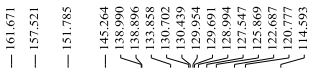
7.188
7.131
7.110
1.647
-0.004


N
N
N
N
Cl
Cl

**3ag**


2.02
1.02
2.03
2.02
1.05
2.00
1.03
1.01
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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138.990
138.896
133.858
130.702
130.439
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129.691
128.994
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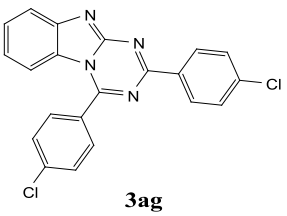
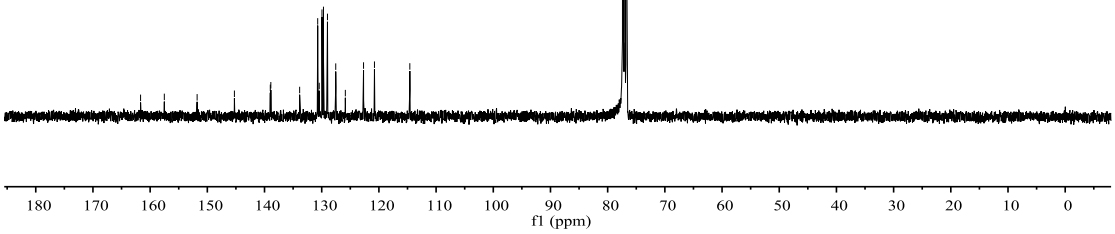
122.687
120.777
114.593
77.317
76.683


N
N
N
N
Cl
Cl

**3ag**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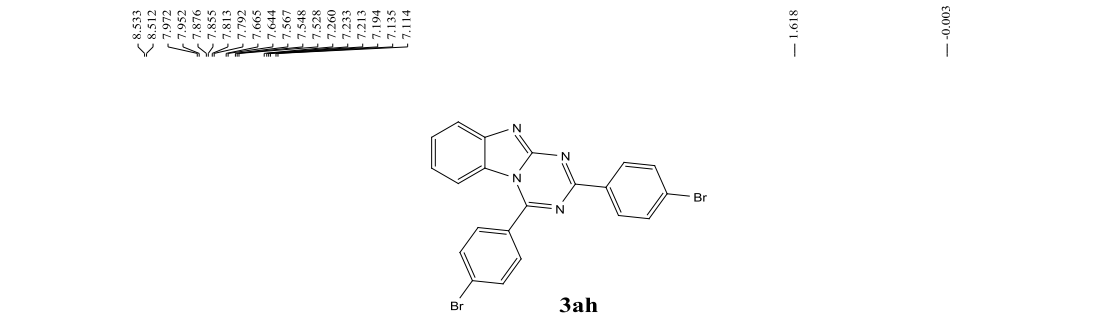
f1 (ppm)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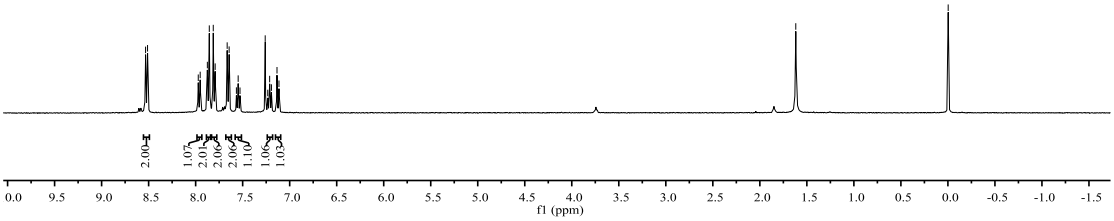
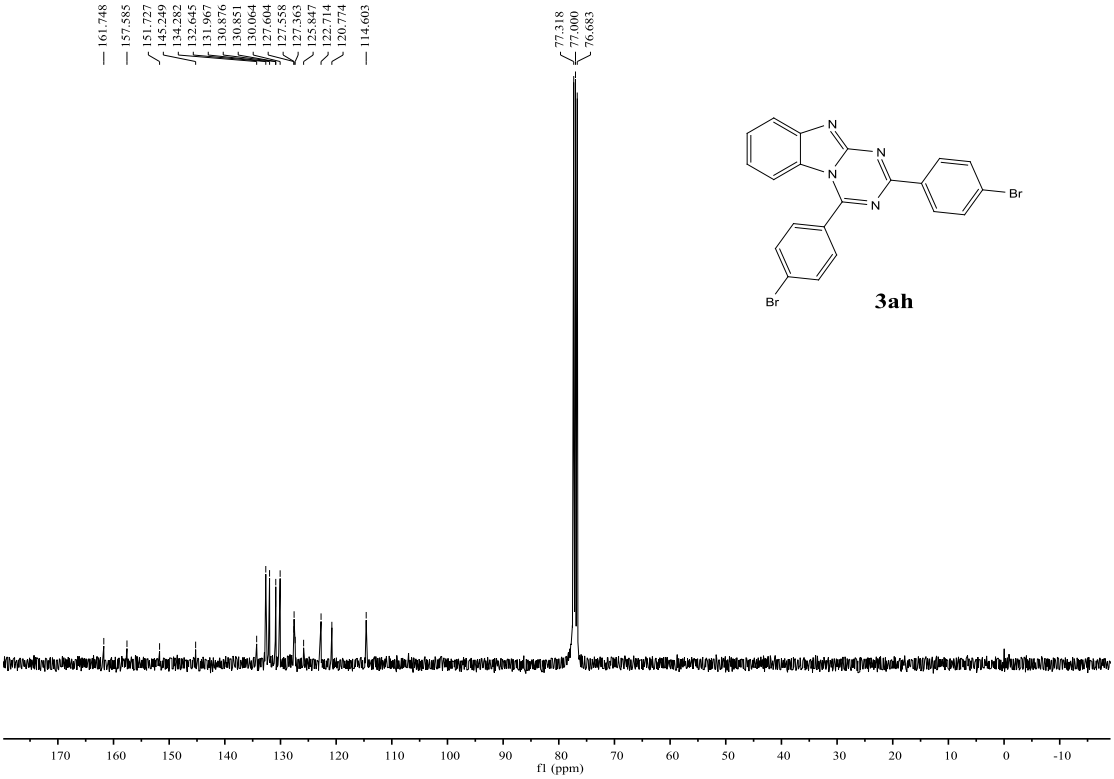
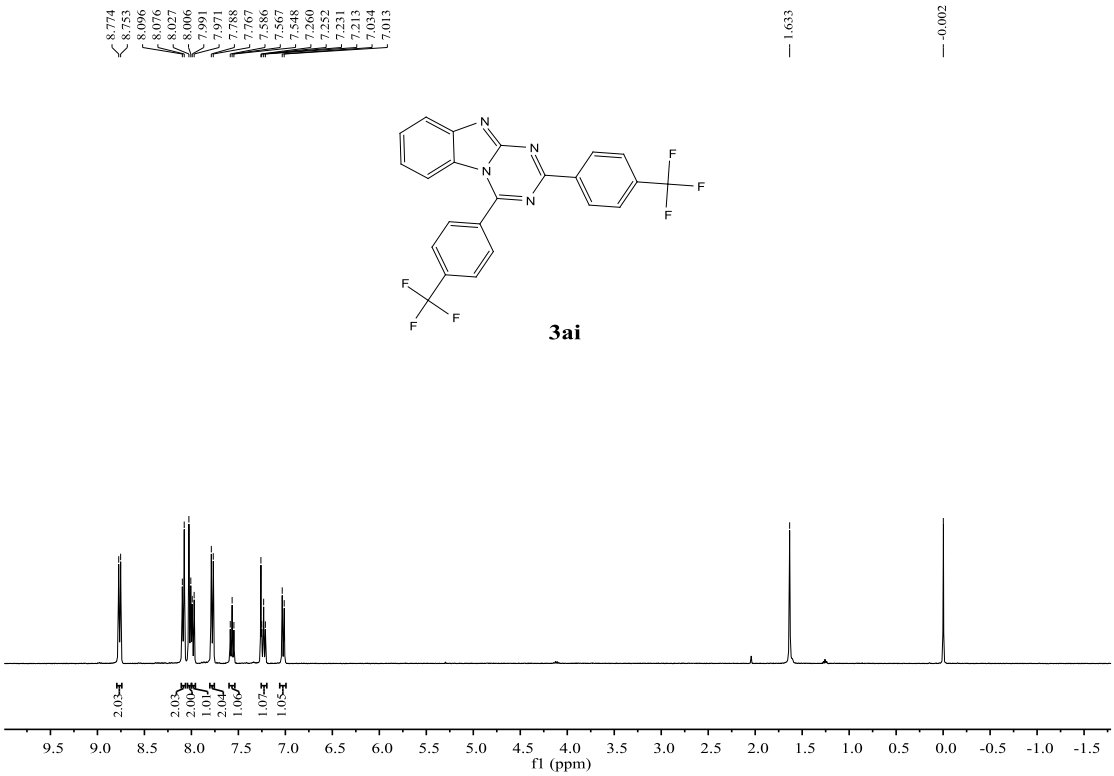
7.876
7.855
7.813
7.792
7.665
7.644
7.567
7.548
7.528
7.260
7.233
7.213
7.194
7.135
7.114
1.618
-0.003
N
N
N
N
Br
Br

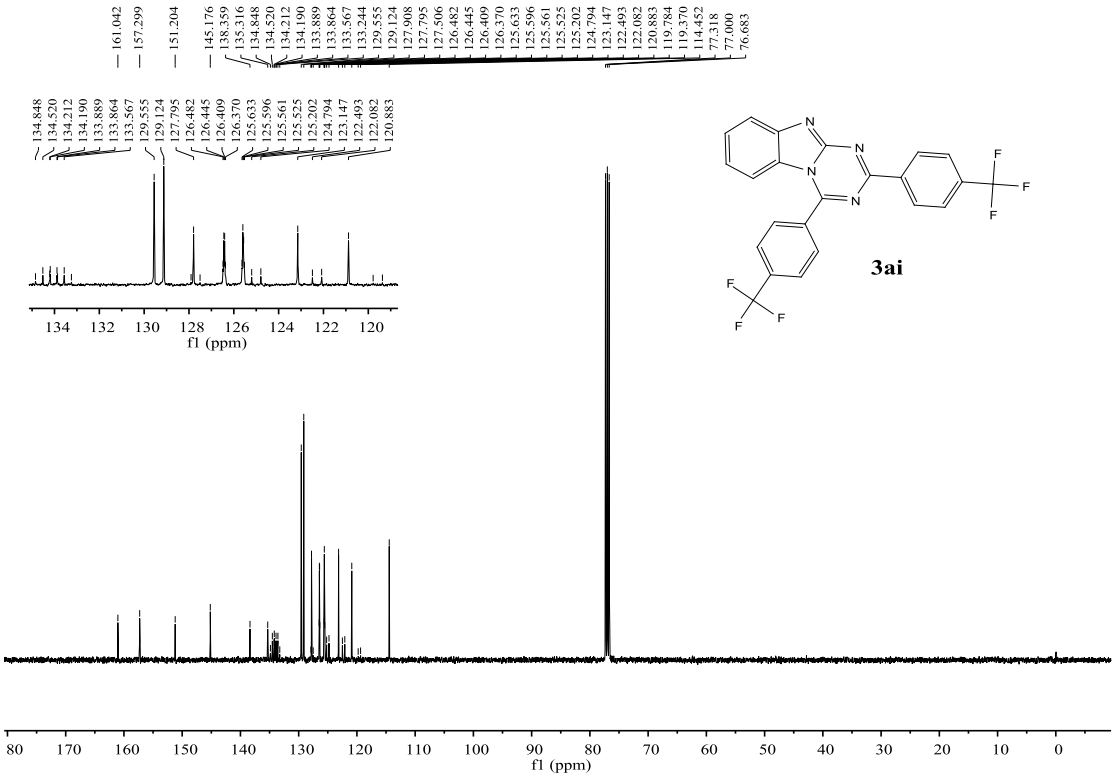
**3ah**

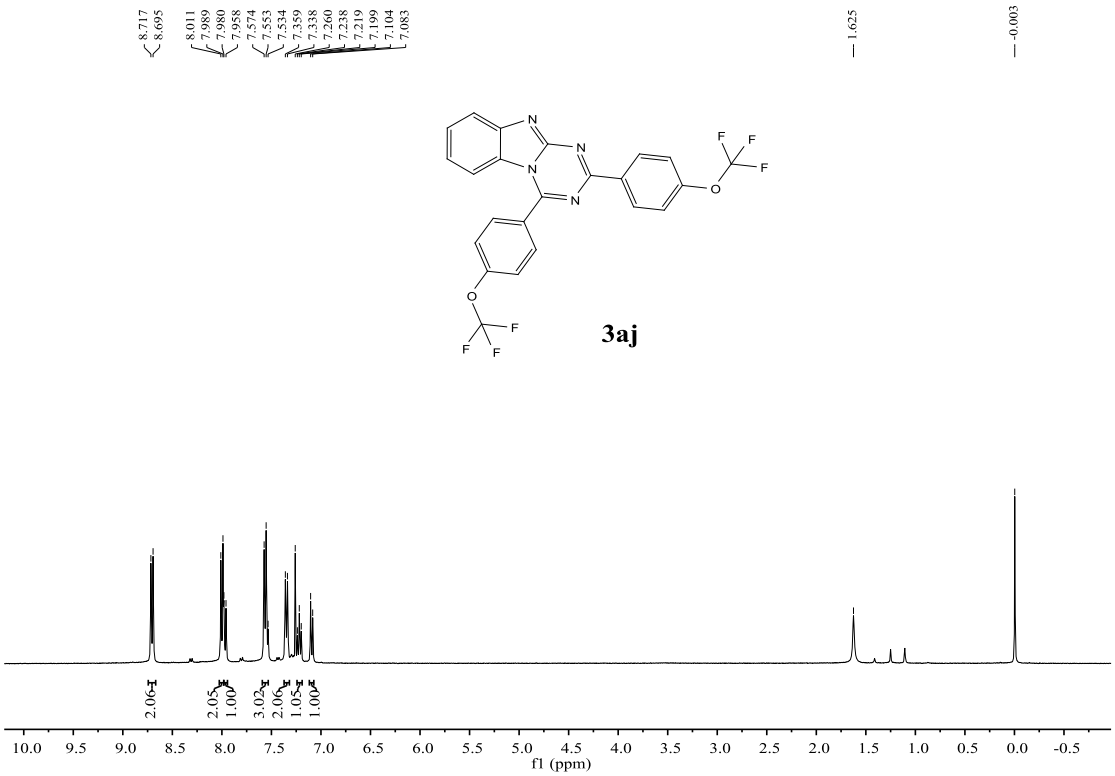

2.00
1.07
2.01
2.00
2.00
1.10
1.06
1.03
0.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
-1.0
-1.5
f1 (ppm)

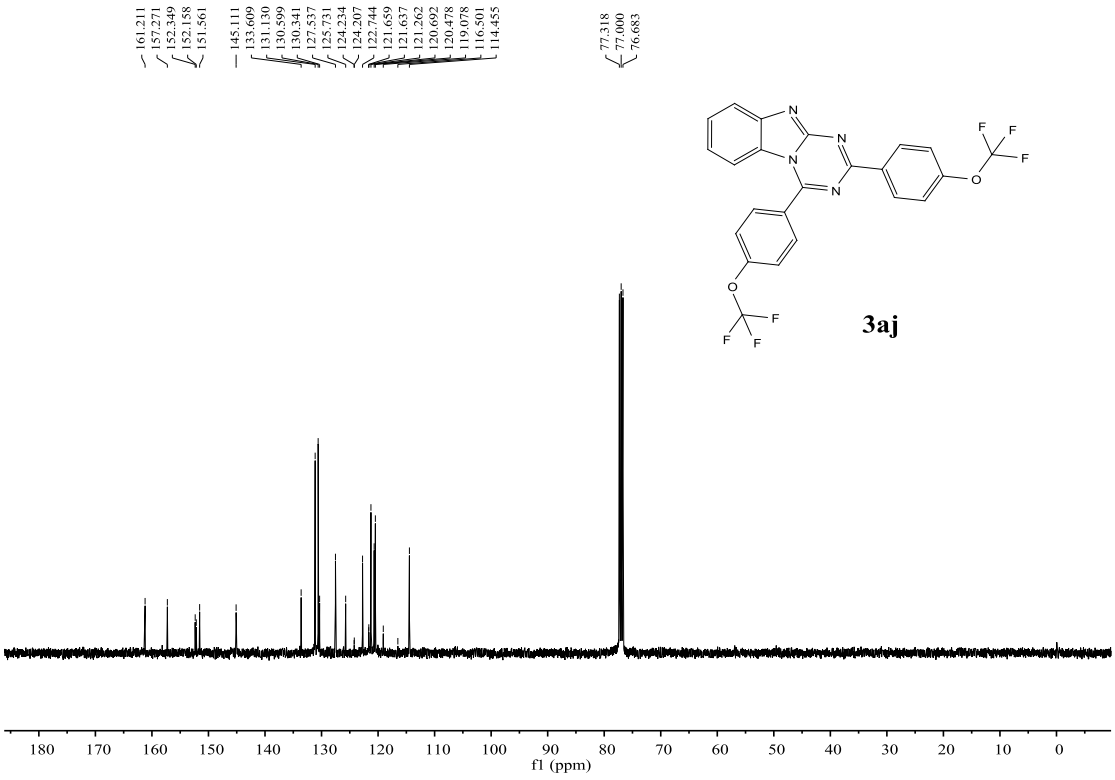

161.748
157.585
151.727
145.249
134.282
132.645
131.967
130.876
130.851
130.064
127.604
127.558
127.363
125.847
122.714
120.774
114.603
77.318
77.000
76.683
N
N
N
N
Br
Br
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)

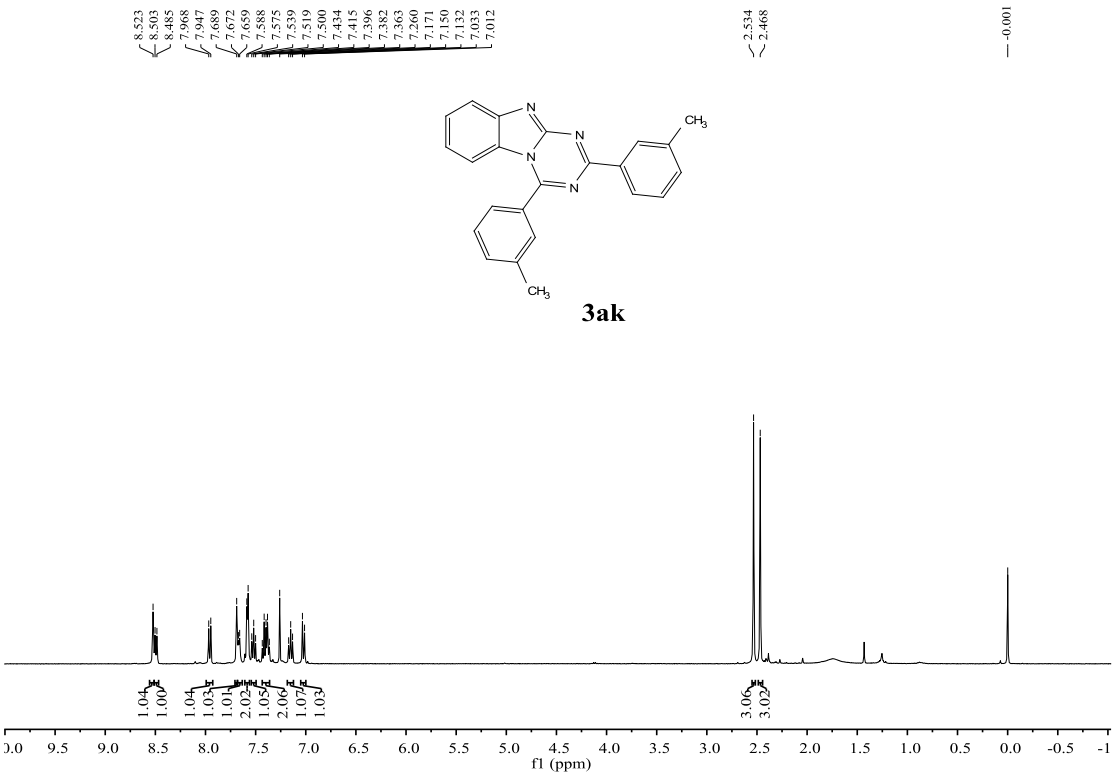
**3ah**


8.774
8.753
8.096
8.076
8.027
8.006
7.991
7.971
7.788
7.767
7.586
7.567
7.548
7.260
7.252
7.231
7.213
7.034
7.013
1.633
-0.002
3ai
2.03
2.03
2.00
1.01
2.04
1.06
1.07
1.05
f1 (ppm)

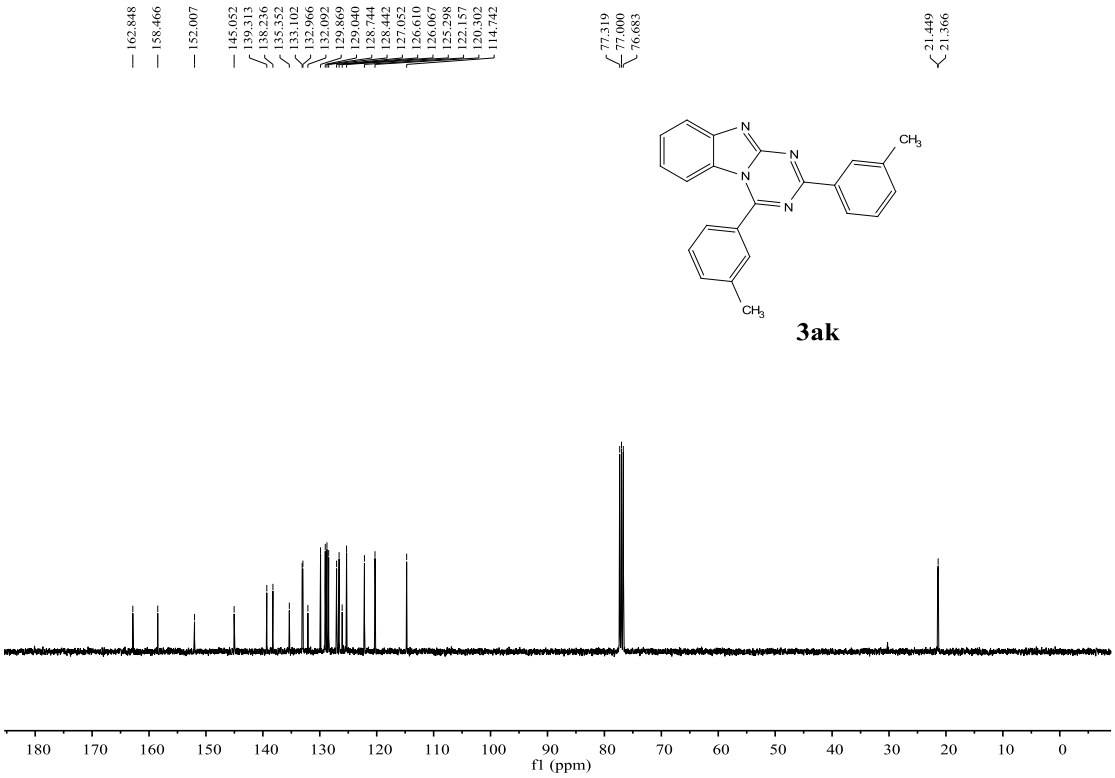

161.042
157.299
151.204
145.176
138.359
135.316
134.848
134.520
134.212
134.190
133.889
133.864
133.567
133.244
129.555
129.124
127.908
127.795
127.506
126.482
126.445
126.409
126.370
125.633
125.596
125.561
125.525
125.202
124.794
123.147
122.493
122.082
120.883
119.784
119.370
114.452
77.318
77.000
76.683
3ai
f1 (ppm)
f1 (ppm)


8.717
8.695
8.011
7.989
7.980
7.958
7.574
7.553
7.534
7.359
7.338
7.260
7.238
7.219
7.199
7.104
7.083
1.625
-0.003
3aj
2.06
2.05
1.00
3.02
2.06
1.05
1.00
f1 (ppm)

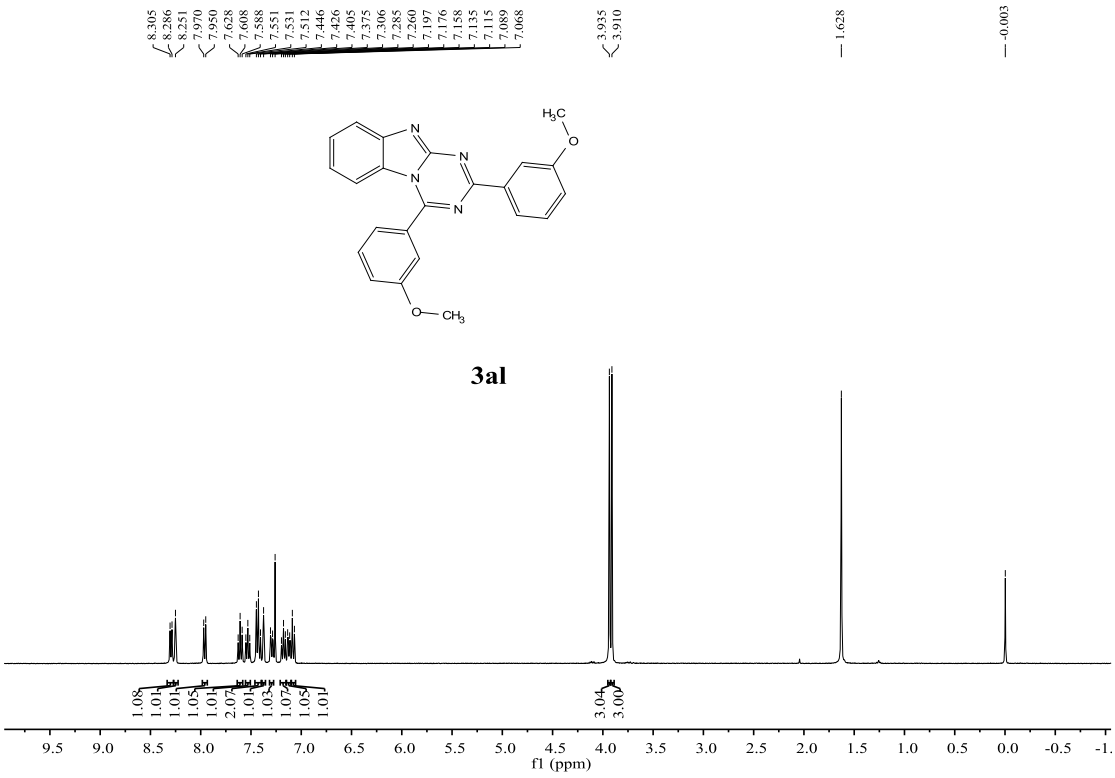

161.211
157.271
152.349
152.158
151.561
145.111
133.609
131.130
130.599
130.341
127.537
125.731
124.234
124.207
122.744
121.659
121.637
121.262
120.692
120.478
119.078
116.501
114.455
77.318
77.000
76.683
3aj
f1 (ppm)


8.523
8.503
8.485
7.968
7.947
7.689
7.672
7.659
7.588
7.575
7.539
7.519
7.500
7.434
7.415
7.396
7.382
7.363
7.260
7.171
7.150
7.132
7.033
7.012
2.534
2.468
-0.001
1.04
1.00
1.04
1.03
1.01
2.02
1.05
2.06
1.07
1.03
3.06
3.02
f1 (ppm)

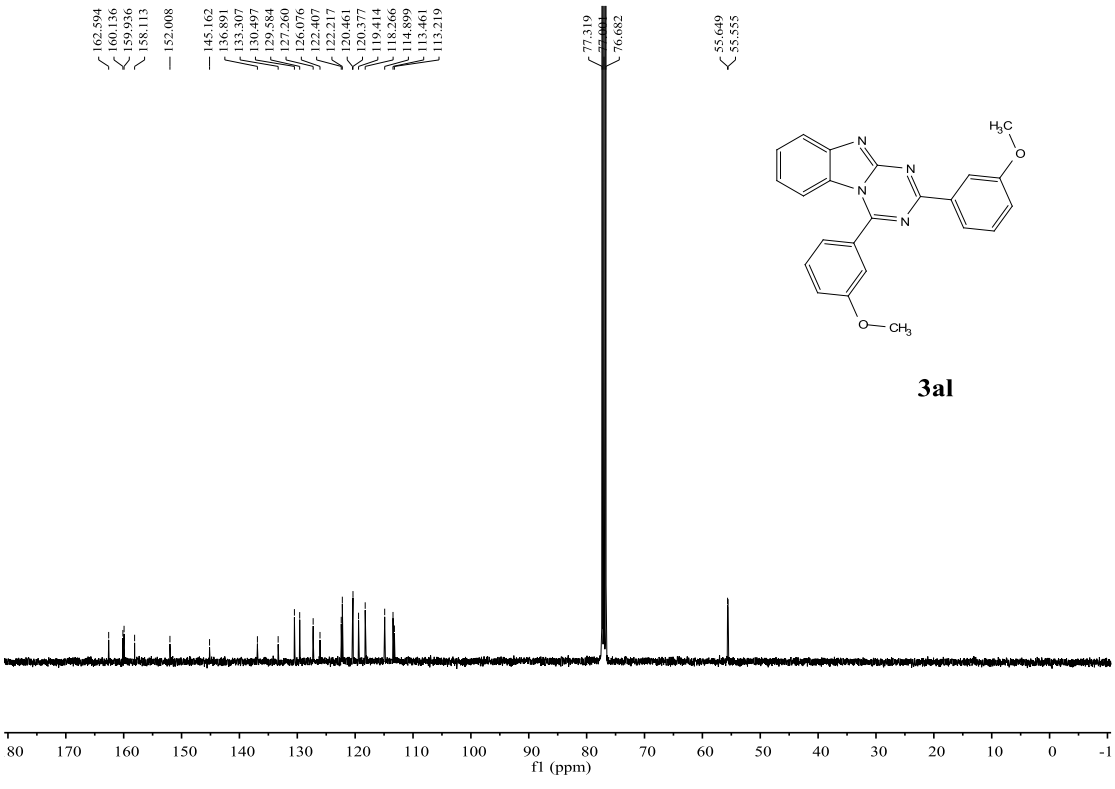
**3ak**


162.848
158.466
152.007
145.052
139.313
138.236
135.352
133.102
132.966
132.092
129.869
129.040
128.744
128.442
127.052
126.610
126.067
125.298
122.157
120.302
114.742
77.319
77.000
76.683
21.449
21.366
f1 (ppm)

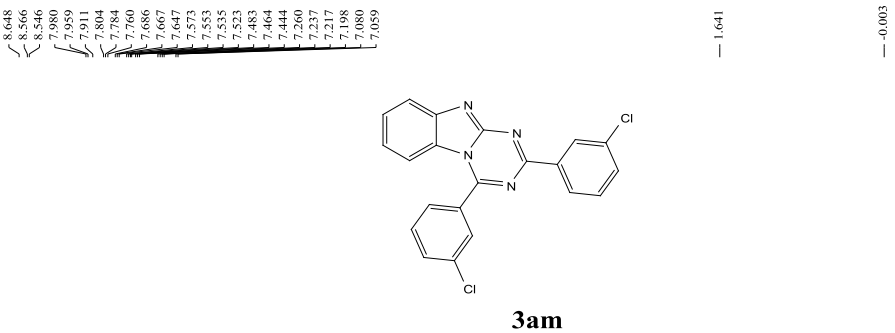
**3ak**


8.305
8.286
8.251
7.970
7.950
7.628
7.608
7.588
7.551
7.531
7.512
7.446
7.426
7.405
7.375
7.306
7.285
7.260
7.197
7.176
7.158
7.135
7.115
7.089
7.068
3.935
3.910
1.628
-0.003
H3C
O
N
N
N
N
O
CH3
1.08
1.01
1.01
1.05
1.01
2.07
1.01
1.03
1.07
1.05
1.01
3.04
3.00
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.
f1 (ppm)

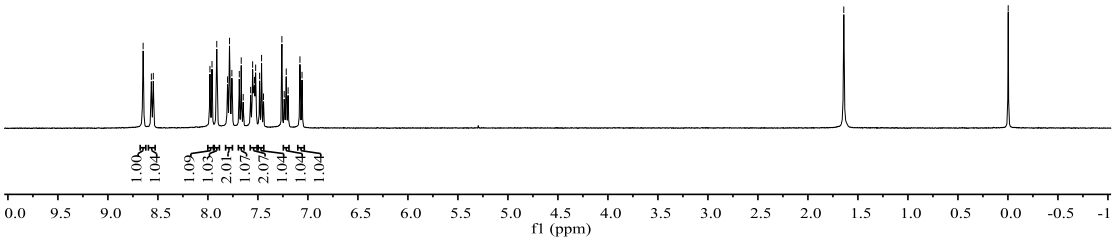
**3al**

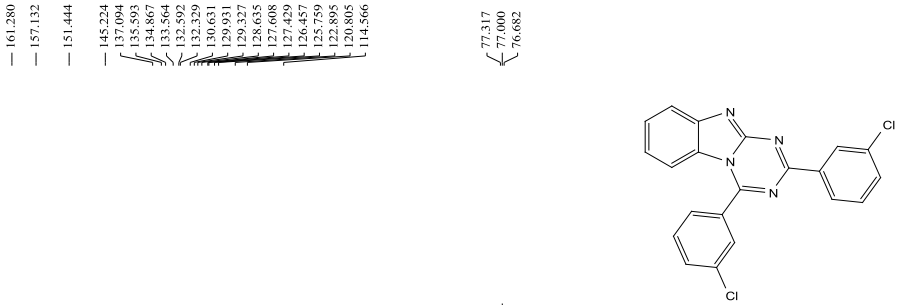

162.594
160.136
159.936
158.113
152.008
145.162
136.891
133.307
130.497
129.584
127.260
126.076
122.407
122.217
120.461
120.377
119.414
118.266
114.899
113.461
113.219
77.319
77.001
76.682
55.649
55.555
H3C
O
N
N
N
N
O
CH3
80 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -1
f1 (ppm)

**3al**

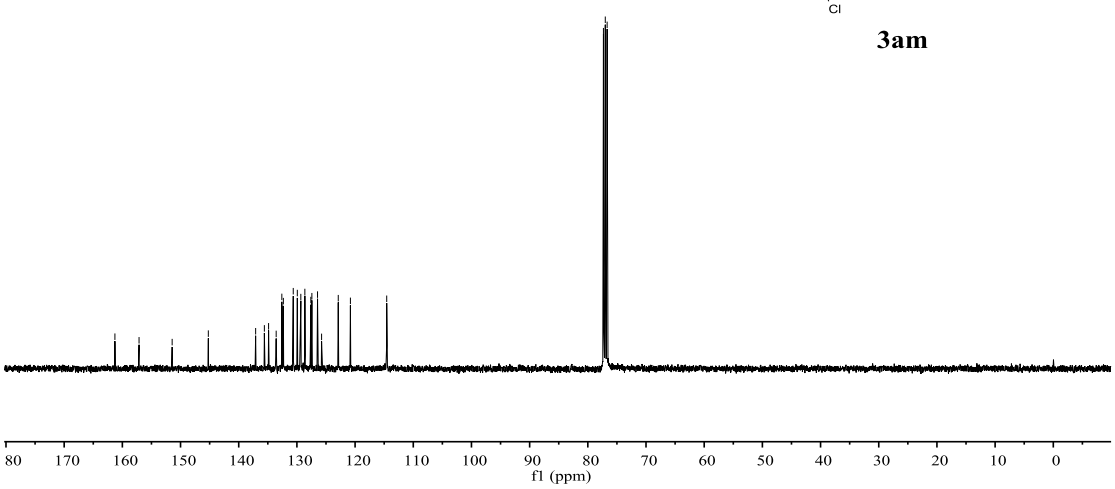

8.648
8.566
8.546
7.980
7.959
7.911
7.804
7.784
7.760
7.686
7.667
7.647
7.573
7.553
7.535
7.523
7.483
7.464
7.444
7.260
7.237
7.217
7.198
7.080
7.059
1.641
-0.003
N
N
N
N
Cl
Cl

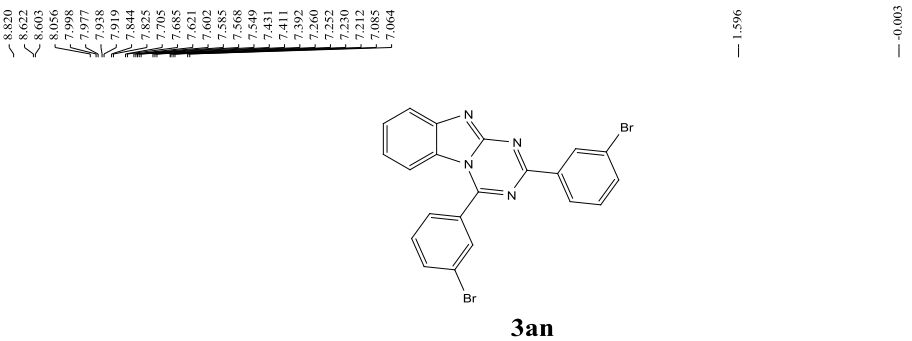
**3am**


1.00
1.04
1.09
1.03
2.01
1.07
2.07
1.04
1.04
1.04
0.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1
f1 (ppm)

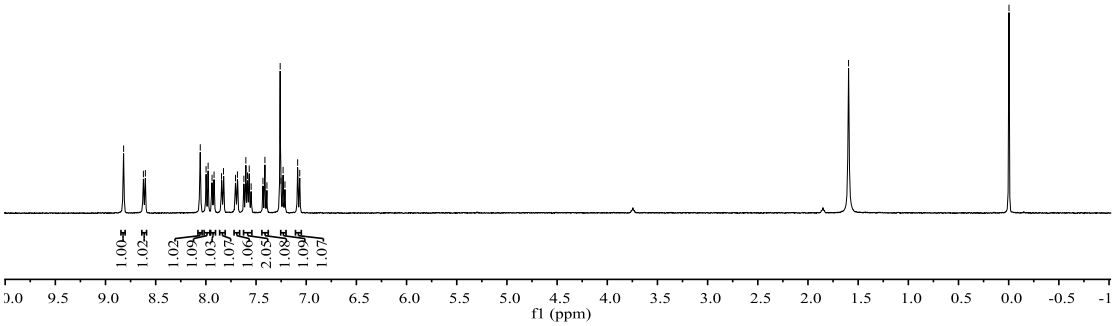

161.280
157.132
151.444
145.224
137.094
135.593
134.867
133.564
132.592
132.329
130.631
129.931
129.327
128.635
127.608
127.429
126.457
125.759
122.895
120.805
114.566
77.317
77.000
76.682
N
N
N
N
Cl
Cl

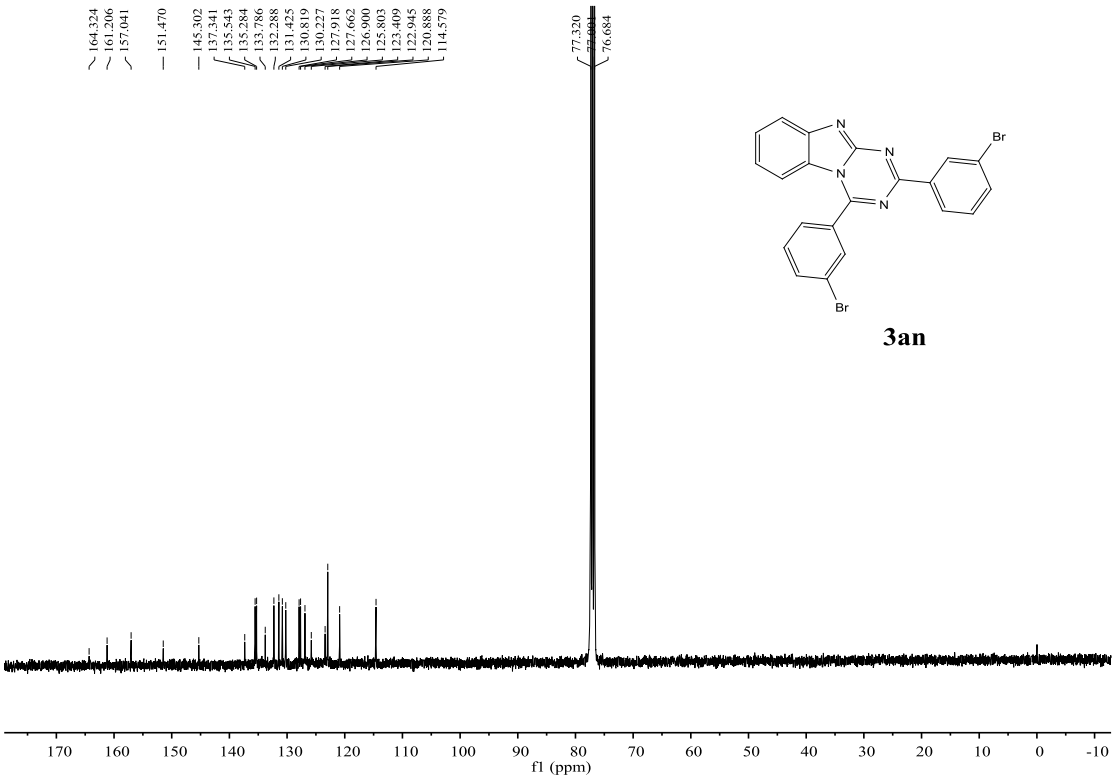
**3am**


80 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

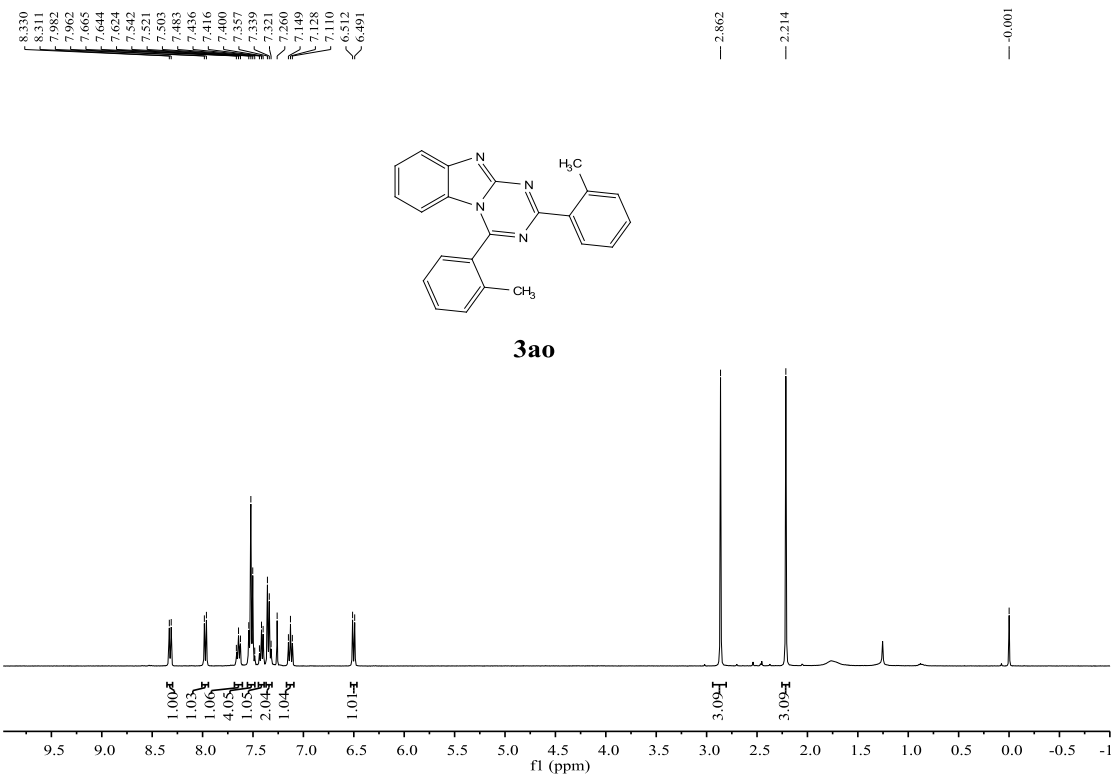

8.820
8.622
8.603
8.056
7.998
7.977
7.938
7.919
7.844
7.825
7.705
7.685
7.621
7.602
7.585
7.568
7.549
7.431
7.411
7.392
7.260
7.252
7.230
7.212
7.085
7.064
1.596
-0.003
N
N
N
N
Br
Br

**3an**

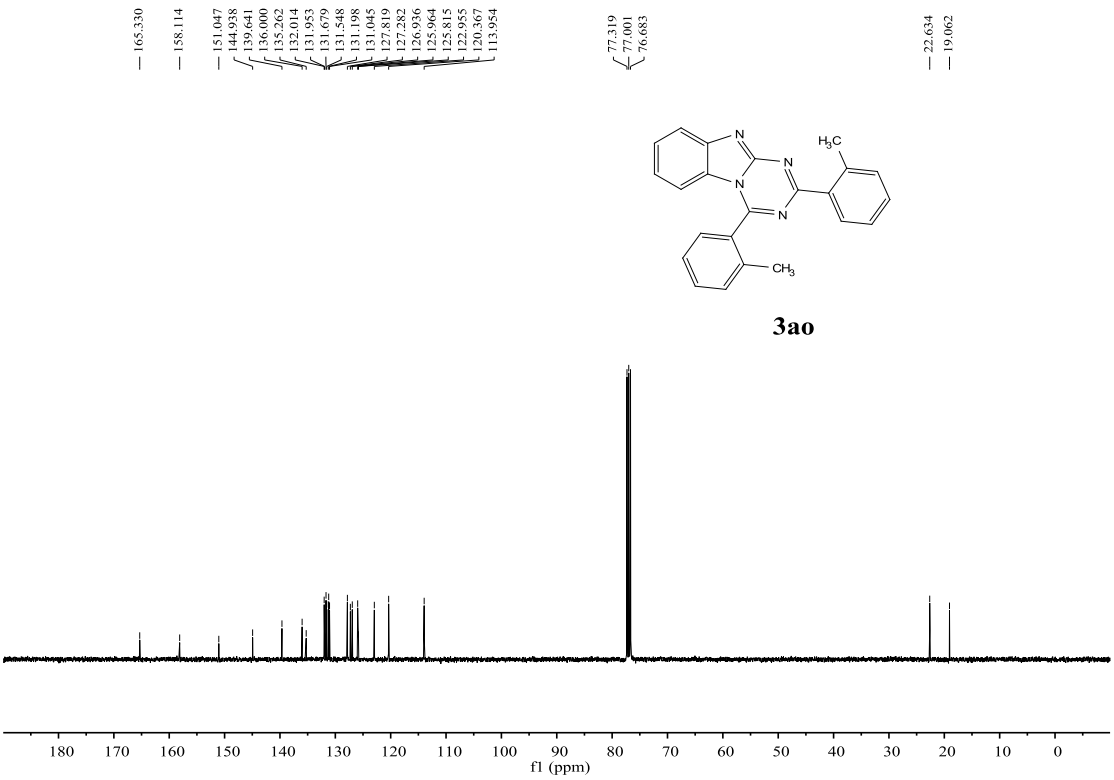

1.00
1.02
1.02
1.09
1.09
1.03
1.07
1.06
2.05
1.08
1.09
1.07
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
f1 (ppm)


164.324
161.206
157.041
151.470
145.302
137.341
135.543
135.284
133.786
132.288
131.425
130.819
130.227
127.918
127.662
126.900
125.803
123.409
122.945
120.888
114.579
77.320
77.004
76.684
N
N
N
N
Br
Br
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)

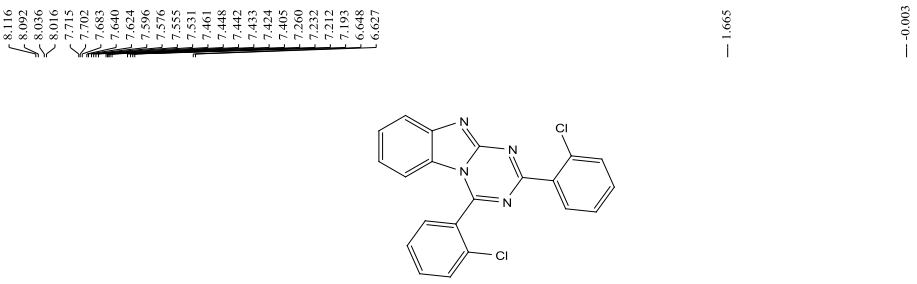
**3an**


8.330
8.311
7.982
7.962
7.665
7.644
7.624
7.542
7.521
7.503
7.483
7.436
7.416
7.400
7.357
7.339
7.321
7.260
7.149
7.128
7.110
6.512
6.491
2.862
2.214
-0.001
N
N
N
N
H3C
CH3
1.00
1.03
1.06
4.05
1.05
2.04
1.04
1.01
3.09
3.09
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1
f1 (ppm)

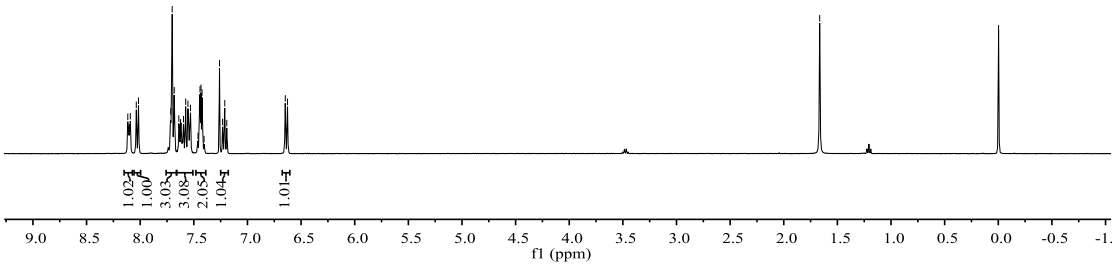
**3ao**

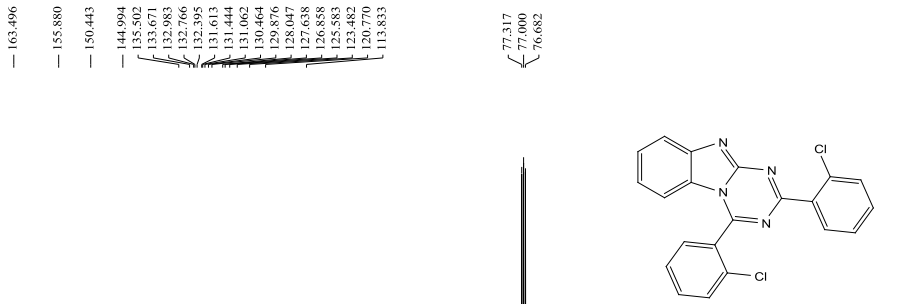

165.330
158.114
151.047
144.938
139.641
136.000
135.262
132.014
131.953
131.679
131.548
131.198
131.045
127.819
127.282
126.936
125.964
125.815
122.955
120.367
113.954
77.319
77.001
76.683
22.634
19.062
N
N
N
N
H3C
CH3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

**3ao**


8.116
8.092
8.036
8.016
7.715
7.702
7.683
7.640
7.624
7.596
7.576
7.555
7.531
7.461
7.448
7.442
7.433
7.424
7.405
7.260
7.232
7.212
7.193
6.648
6.627
1.665
-0.003
N
N
N
N
Cl
Cl

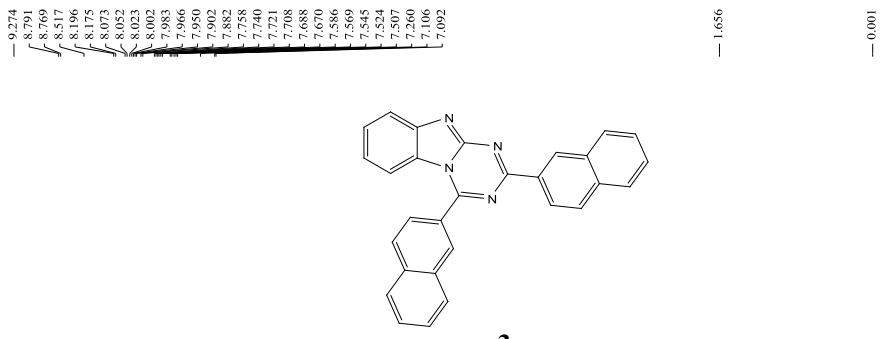
**3ap**


1.02
1.00
3.03
3.08
2.05
1.04
1.01
9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.
f1 (ppm)

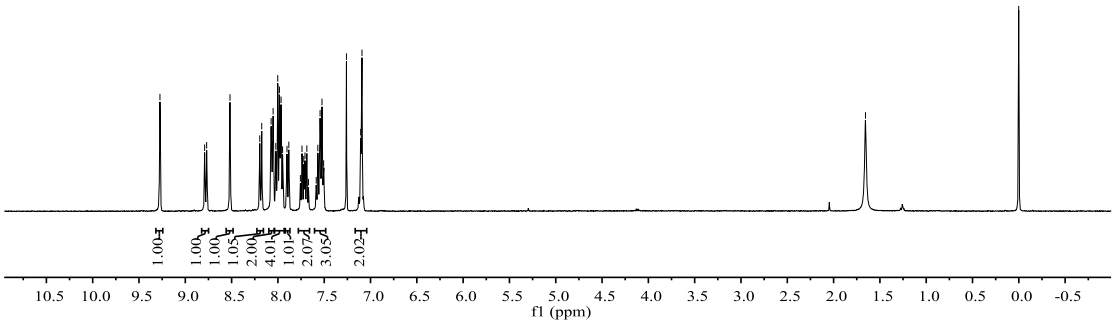

163.496
155.880
150.443
144.994
135.502
133.671
132.983
132.766
132.395
131.613
131.444
131.062
130.464
129.876
128.047
127.638
126.858
125.583
123.482
120.770
113.833
77.317
77.000
76.682
N
N
N
N
Cl
Cl

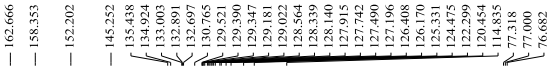
**3ap**

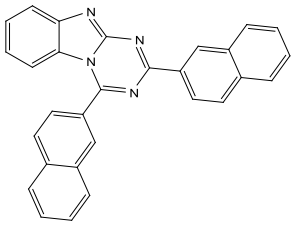

180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


9.274
8.791
8.769
8.517
8.196
8.175
8.073
8.052
8.023
8.002
7.983
7.966
7.950
7.902
7.882
7.758
7.740
7.721
7.708
7.688
7.670
7.586
7.569
7.545
7.524
7.507
7.260
7.106
7.092
1.656
0.001
N
N
N
N

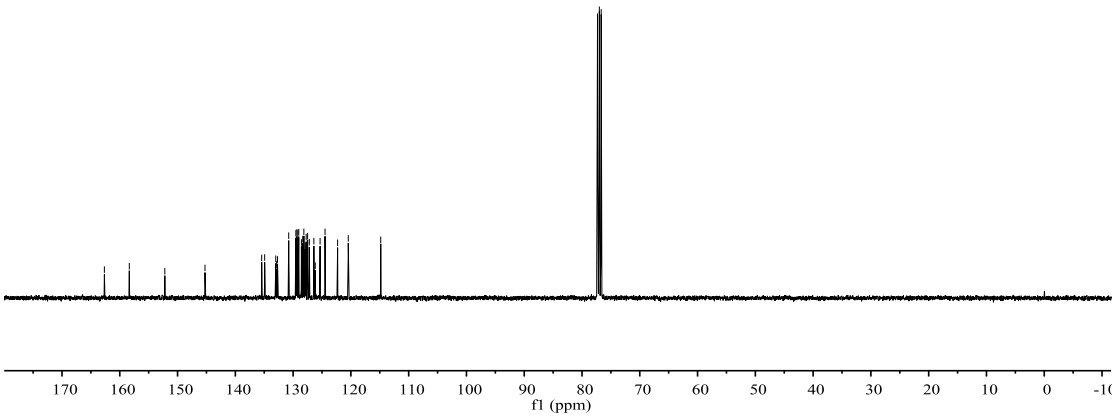
**3aq**

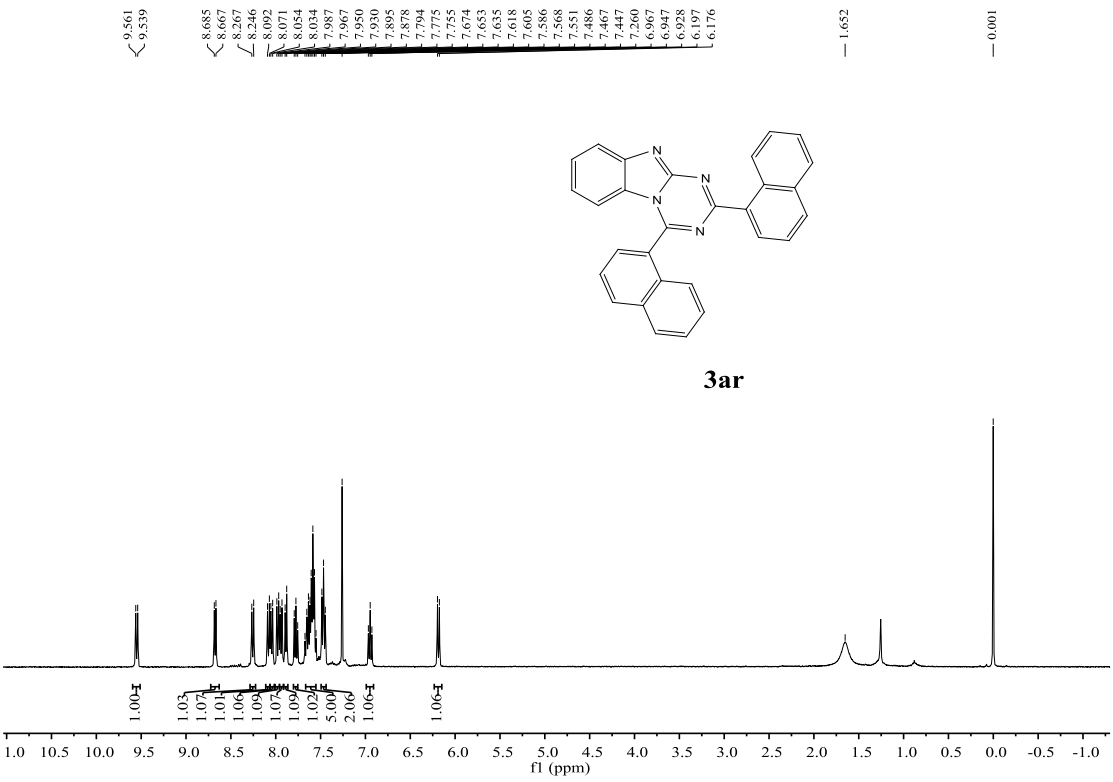

1.00
1.00
1.00
1.05
2.00
4.01
1.01
2.07
3.05
2.02
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)


162.666
158.353
152.202
145.252
135.438
134.924
133.003
132.891
132.697
130.765
129.521
129.390
129.347
129.181
129.022
128.564
128.339
128.140
127.915
127.742
127.490
127.196
126.408
126.170
125.331
124.475
122.299
120.454
114.835
77.318
77.000
76.682

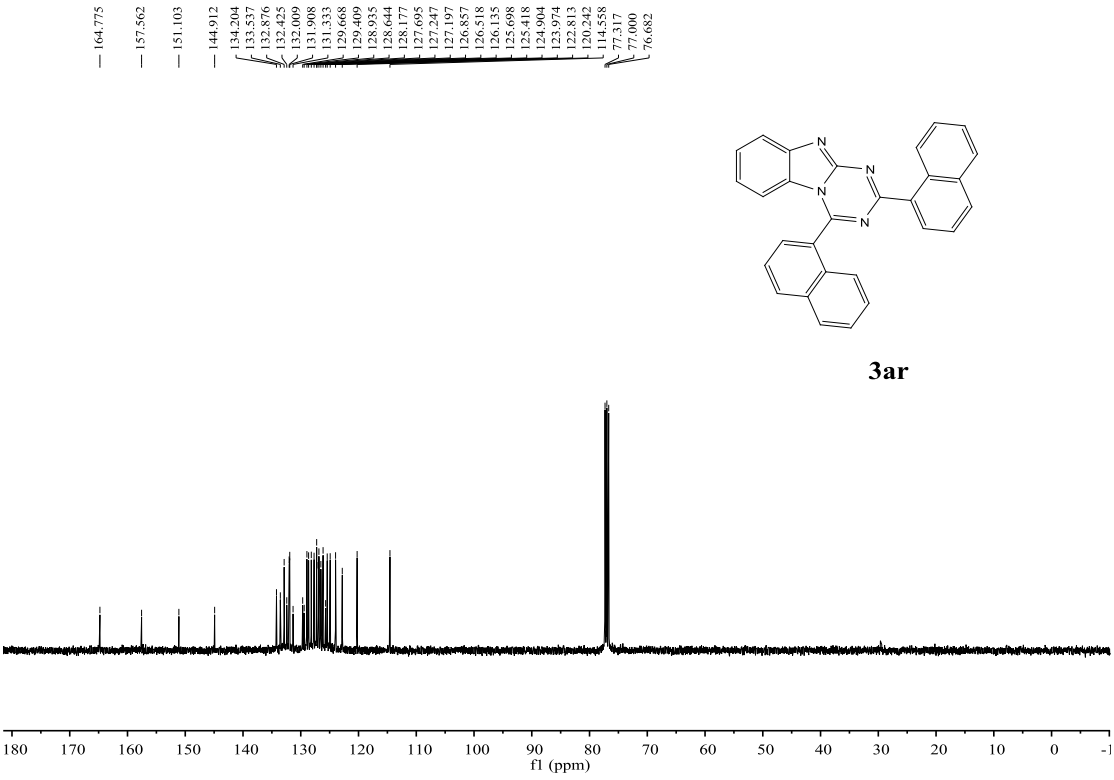

N
N
N
N

**3aq**

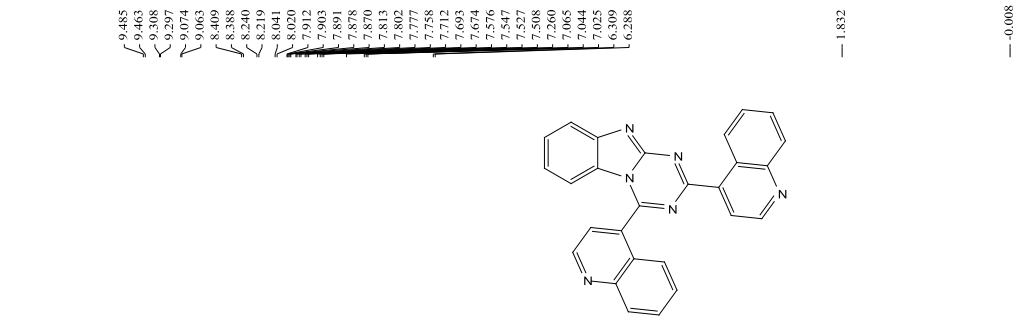

170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


9.561
9.539
8.685
8.667
8.267
8.246
8.092
8.071
8.054
8.034
7.987
7.967
7.950
7.930
7.895
7.878
7.794
7.775
7.755
7.674
7.653
7.635
7.618
7.605
7.586
7.568
7.551
7.486
7.467
7.447
7.260
6.967
6.947
6.928
6.197
6.176
1.652
0.001
N
N
N
N
1.00
1.03
1.07
1.01
1.06
1.09
1.07
1.09
1.02
5.00
2.06
1.06
1.06
1.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

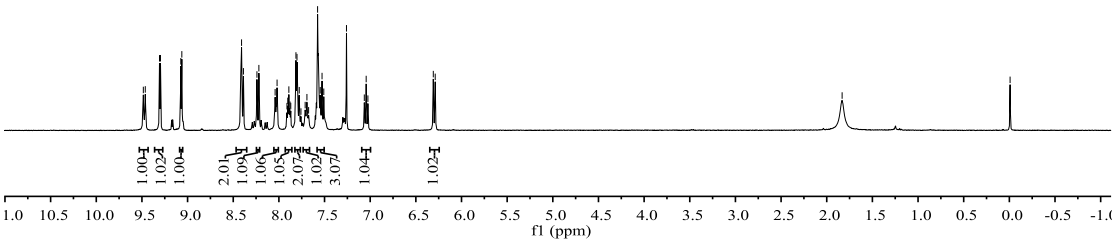
**3ar**

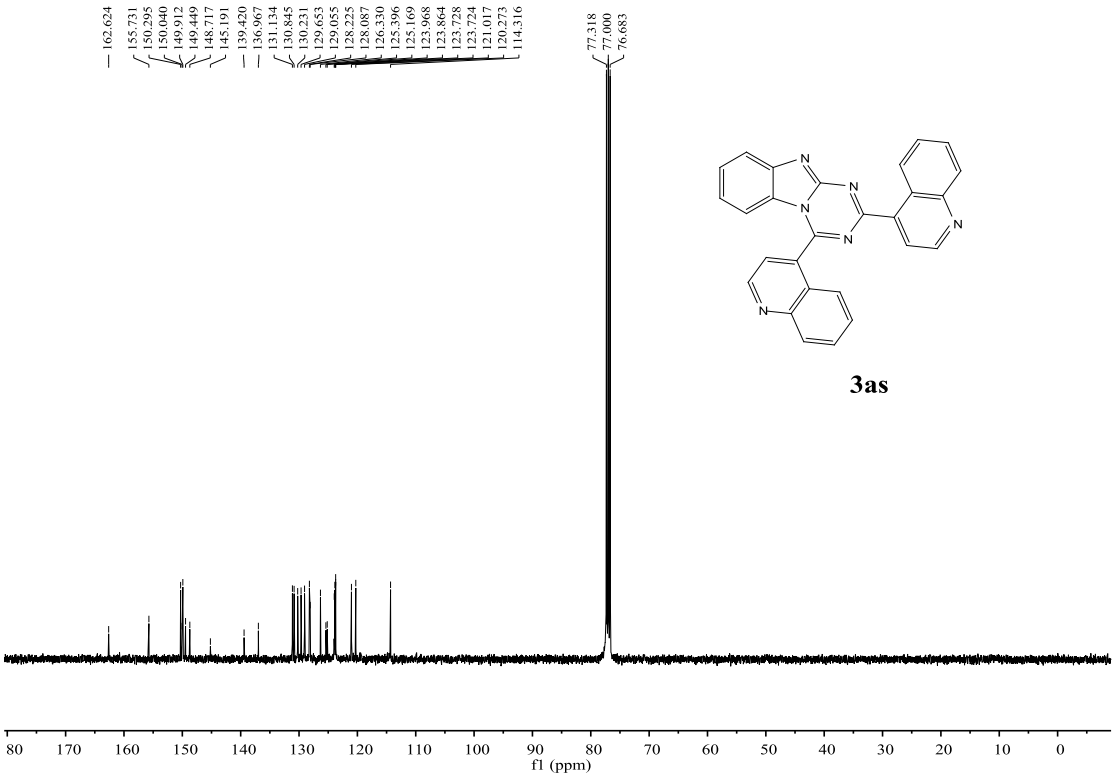

164.775
157.562
151.103
144.912
134.204
133.537
132.876
132.425
132.009
131.908
131.333
129.668
129.409
128.935
128.644
128.177
127.695
127.247
127.197
126.857
126.518
126.135
125.698
125.418
124.904
123.974
122.813
120.242
114.558
77.317
77.000
76.682
N
N
N
N
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -1
f1 (ppm)

**3ar**

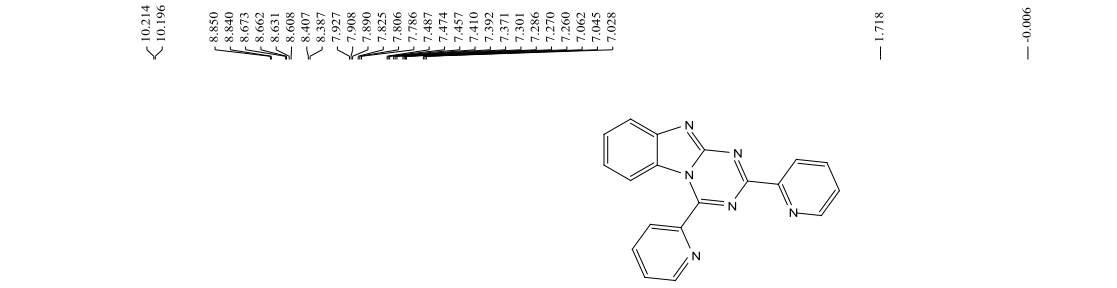

9.485
9.463
9.308
9.297
9.074
9.063
8.409
8.388
8.240
8.219
8.041
8.020
7.912
7.903
7.891
7.878
7.870
7.813
7.802
7.777
7.758
7.712
7.693
7.674
7.576
7.547
7.527
7.508
7.260
7.065
7.044
7.025
6.309
6.288
1.832
-0.008

**3as**

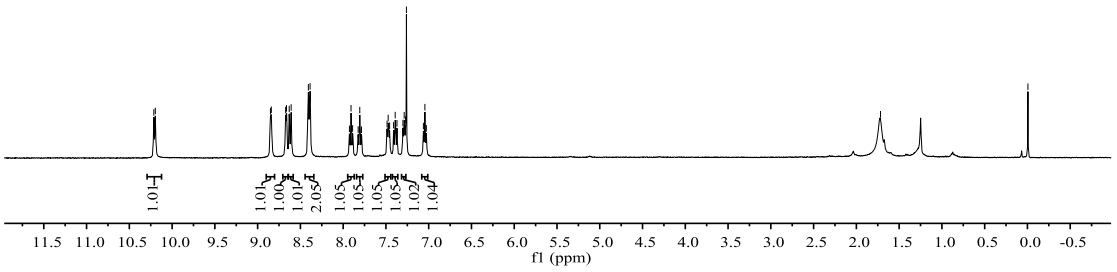

1.00
1.02
1.00
2.01
1.09
1.06
1.05
2.07
1.02
3.07
1.04
1.02
fl (ppm)

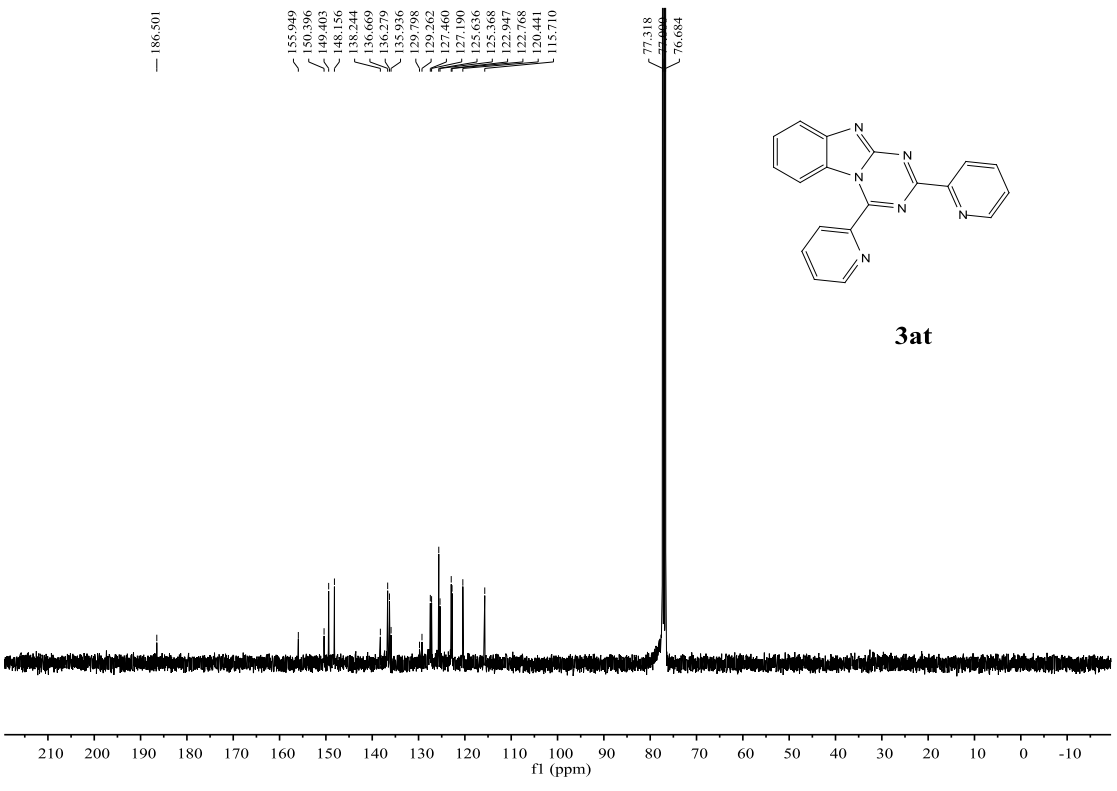

162.624
155.731
150.295
150.040
149.912
149.449
148.717
145.191
139.420
136.967
131.134
130.845
130.231
129.653
129.055
128.225
128.087
126.330
125.396
125.169
123.968
123.864
123.728
123.724
121.017
120.273
114.316
77.318
77.000
76.683
fl (ppm)

**3as**

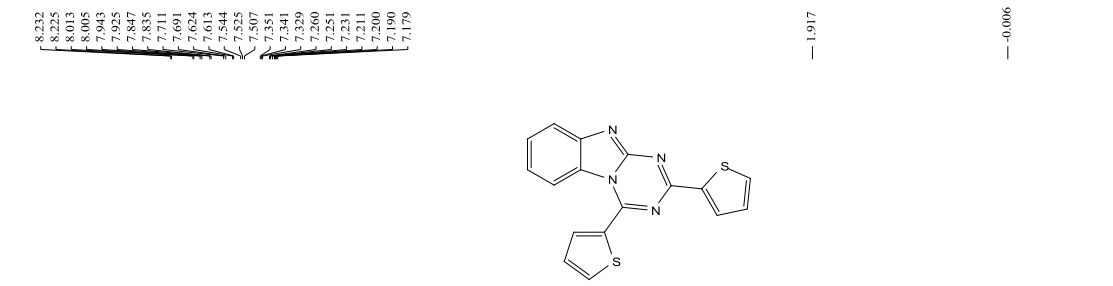

10.214
10.196
8.850
8.840
8.673
8.662
8.631
8.608
8.407
8.387
7.927
7.908
7.890
7.825
7.806
7.786
7.487
7.474
7.457
7.410
7.392
7.371
7.301
7.286
7.270
7.260
7.062
7.045
7.028
1.718
-0.006

**3at**


1.01
1.01
1.00
1.01
2.05
1.05
1.05
1.05
1.03
1.02
1.04
11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)

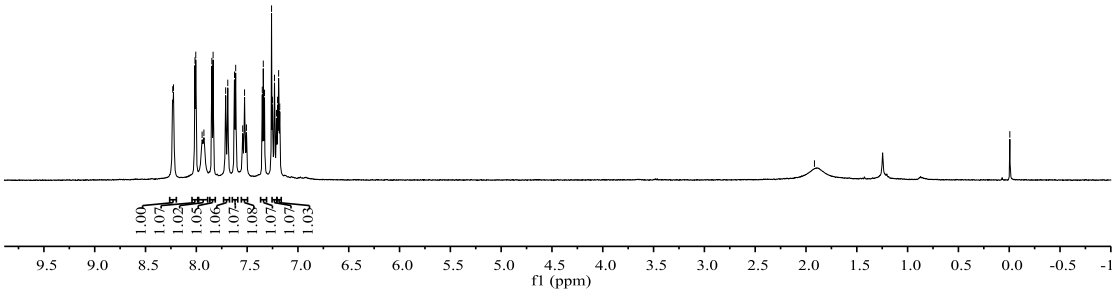

186.501
155.949
150.396
149.403
148.156
138.244
136.669
136.279
135.936
129.798
129.262
127.460
127.190
125.636
125.368
122.947
122.768
120.441
115.710
77.318
76.684
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

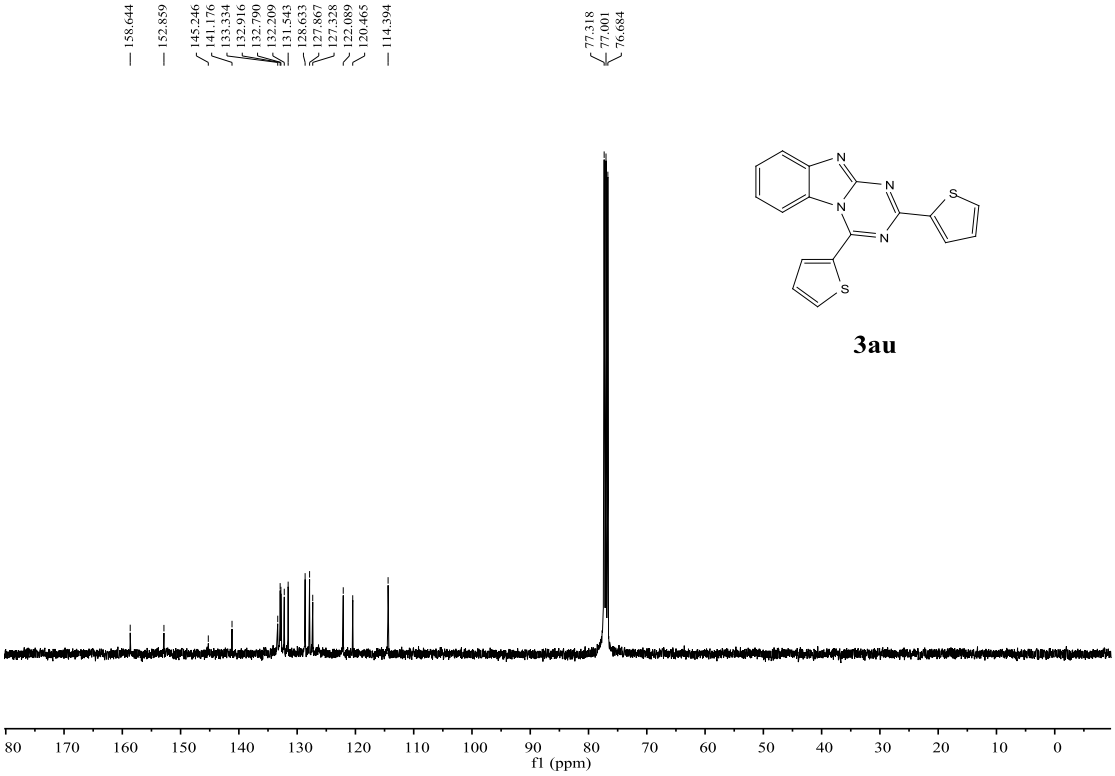
**3at**


8.232
8.225
8.013
8.005
7.943
7.925
7.847
7.835
7.711
7.691
7.624
7.613
7.544
7.525
7.507
7.351
7.341
7.329
7.260
7.251
7.231
7.211
7.200
7.190
7.179
1.917
-0.006



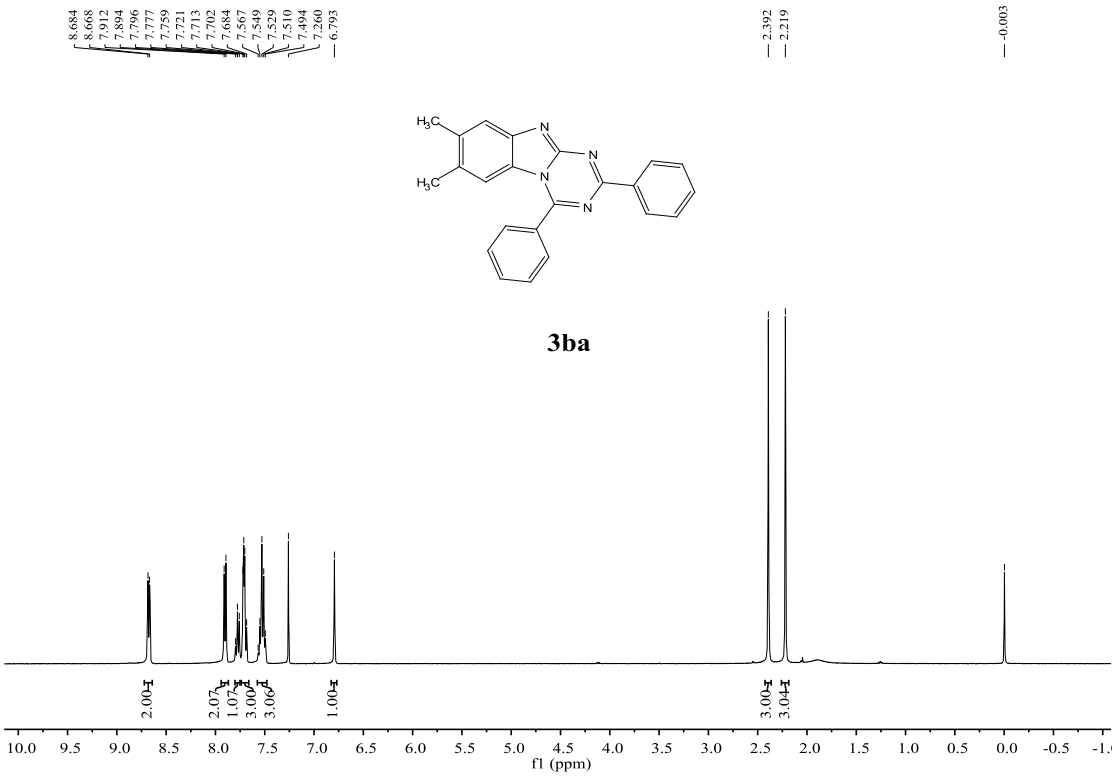
**3au**

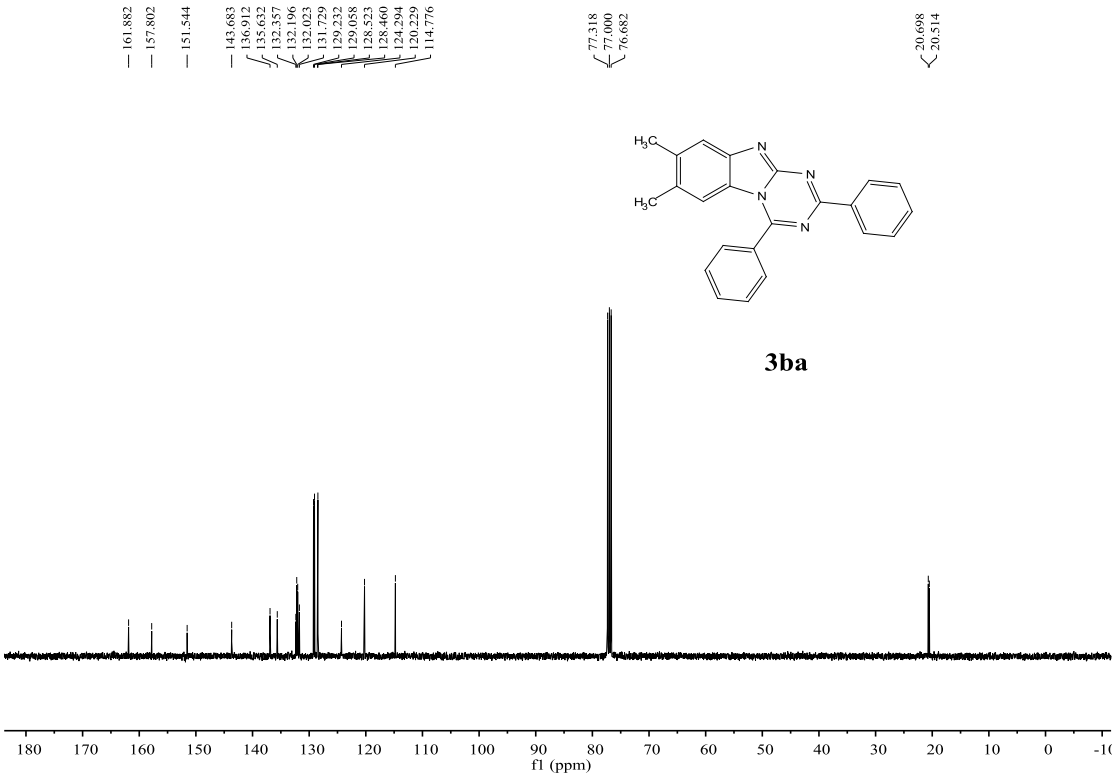

1.00
1.07
1.02
1.03
1.06
1.07
1.08
1.07
1.07
1.03
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1
f1 (ppm)

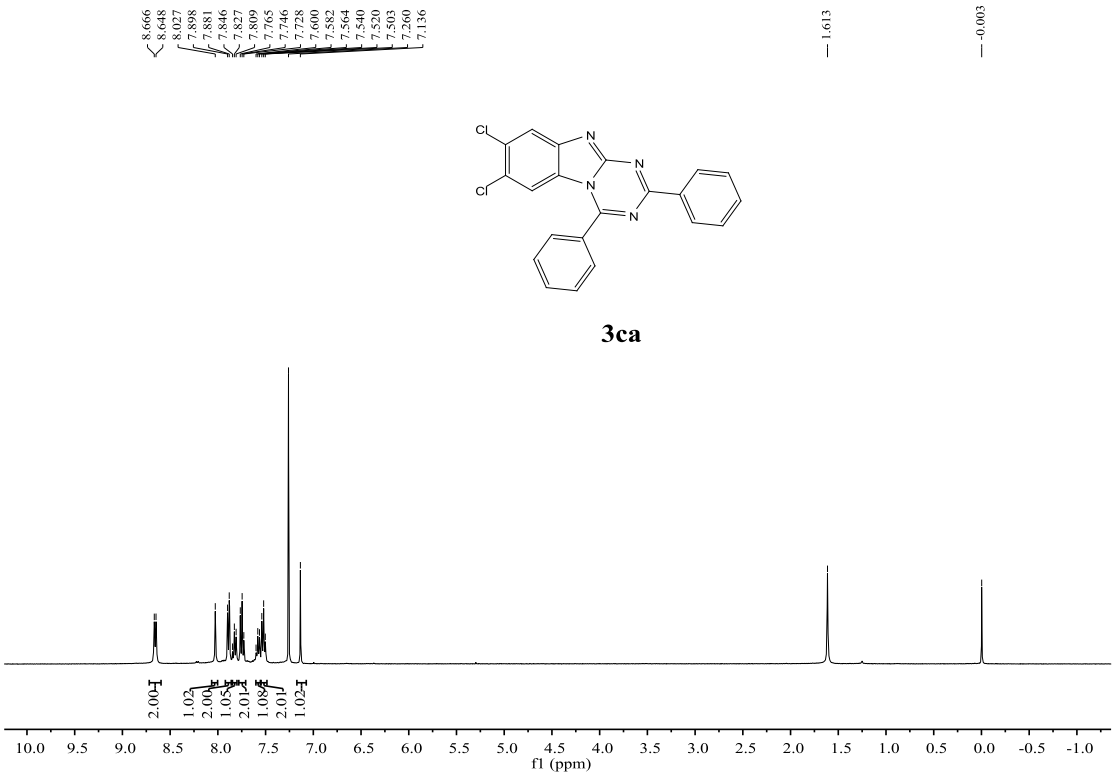

158.644
152.859
145.246
141.176
133.334
132.916
132.790
132.209
131.543
128.633
127.867
127.328
122.089
120.465
114.394
77.318
77.001
76.684
80 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)



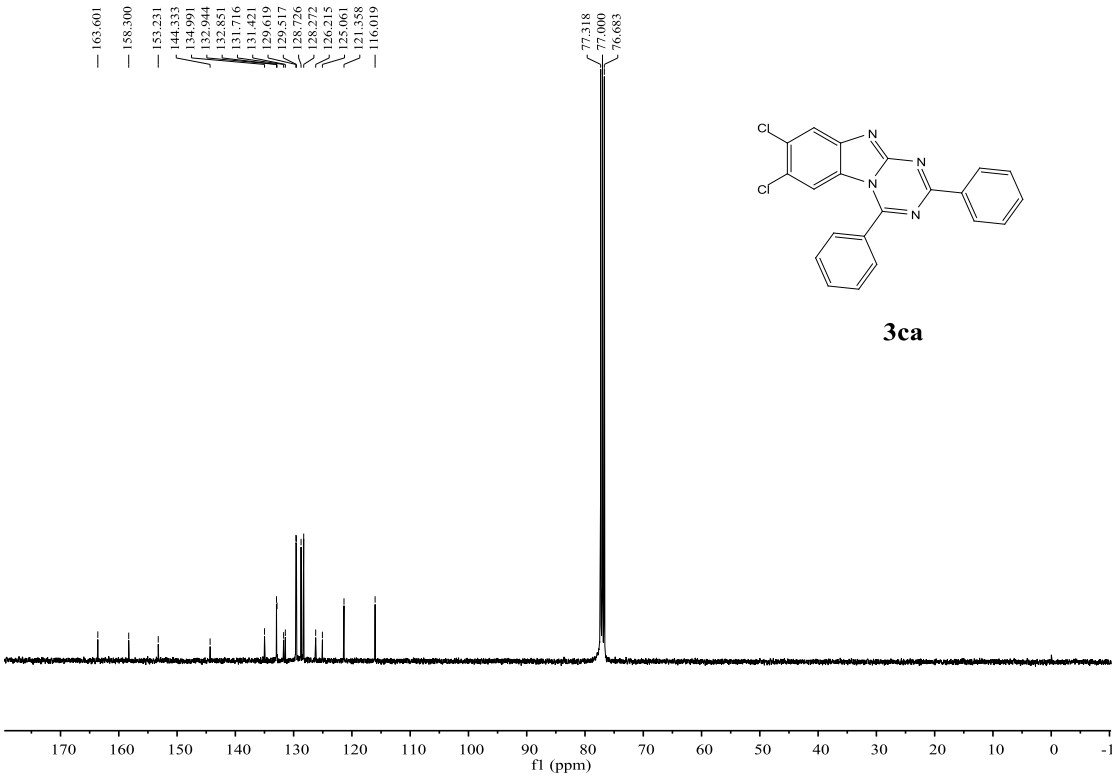
**3au**


8.684
8.668
7.912
7.894
7.796
7.777
7.759
7.721
7.713
7.702
7.684
7.567
7.549
7.529
7.510
7.494
7.260
6.793
2.392
2.219
-0.003
3ba
2.00
2.07
1.07
3.00
3.06
1.00
3.00
3.04
f1 (ppm)

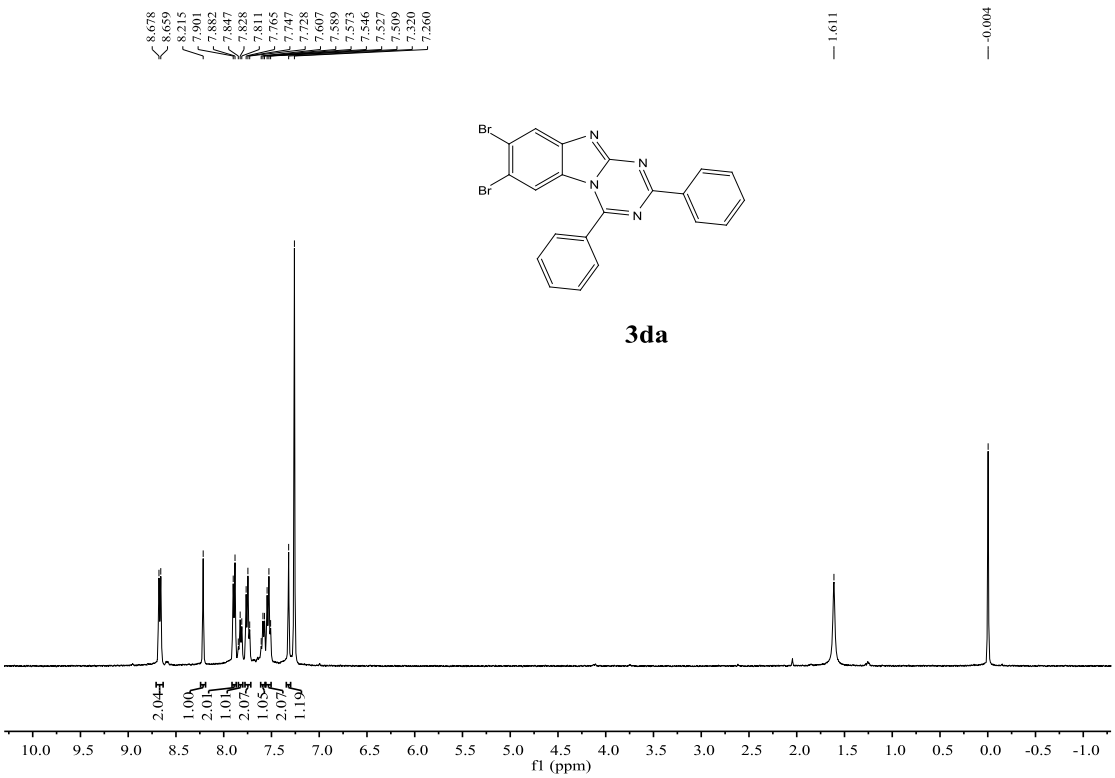

161.882
157.802
151.544
143.683
136.912
135.632
132.357
132.196
132.023
131.729
129.232
129.058
128.523
128.460
124.294
120.229
114.776
77.318
77.000
76.682
20.698
20.514
3ba
f1 (ppm)


8.666
8.648
8.027
7.898
7.881
7.846
7.827
7.809
7.765
7.746
7.728
7.600
7.582
7.564
7.540
7.520
7.503
7.260
7.136
1.613
-0.003
Cl
Cl
N
N
N
N
2.00
1.02
2.00
1.05
2.01
1.08
2.01
1.02
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

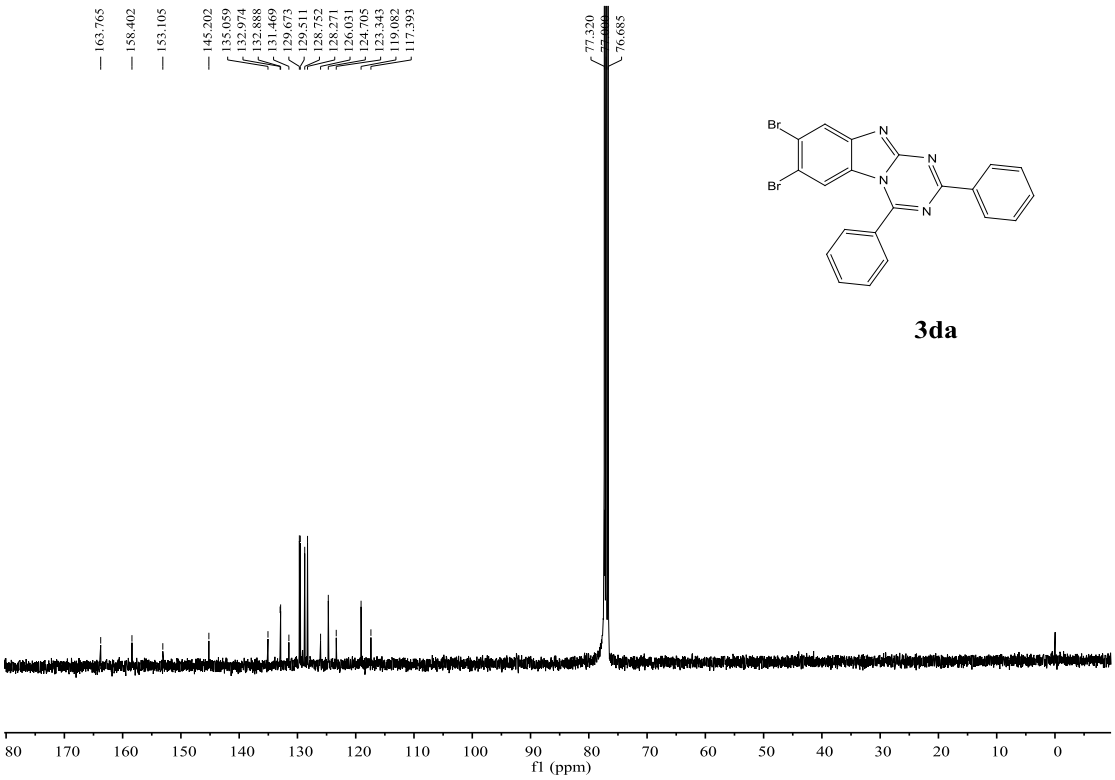
**3ca**


163.601
158.300
153.231
144.333
134.991
132.944
132.851
131.716
131.421
129.619
129.517
128.726
128.272
126.215
125.061
121.358
116.019
77.318
77.000
76.683
Cl
Cl
N
N
N
N
170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -1
f1 (ppm)

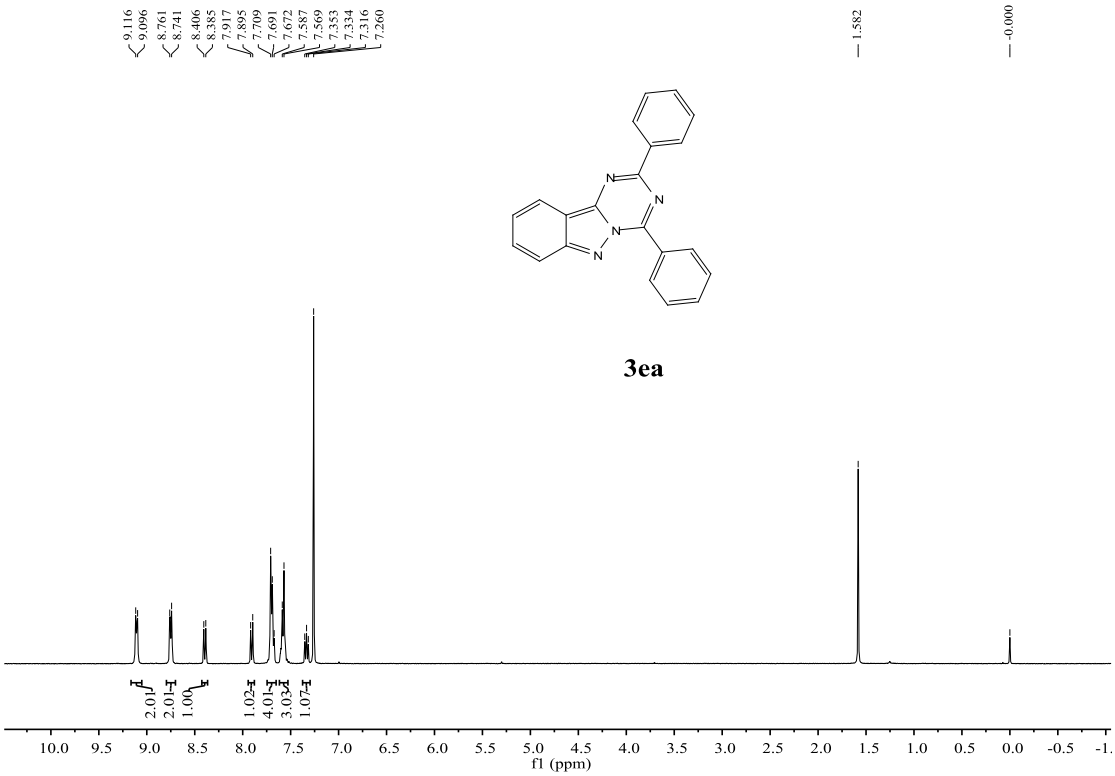
**3ca**


8.678
8.659
8.215
7.901
7.882
7.847
7.828
7.811
7.765
7.747
7.728
7.607
7.589
7.573
7.546
7.527
7.509
7.320
7.260
1.611
0.004
Br
Br
N
N
N
N
2.04
1.00
2.01
1.01
2.07
1.05
2.07
1.19
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

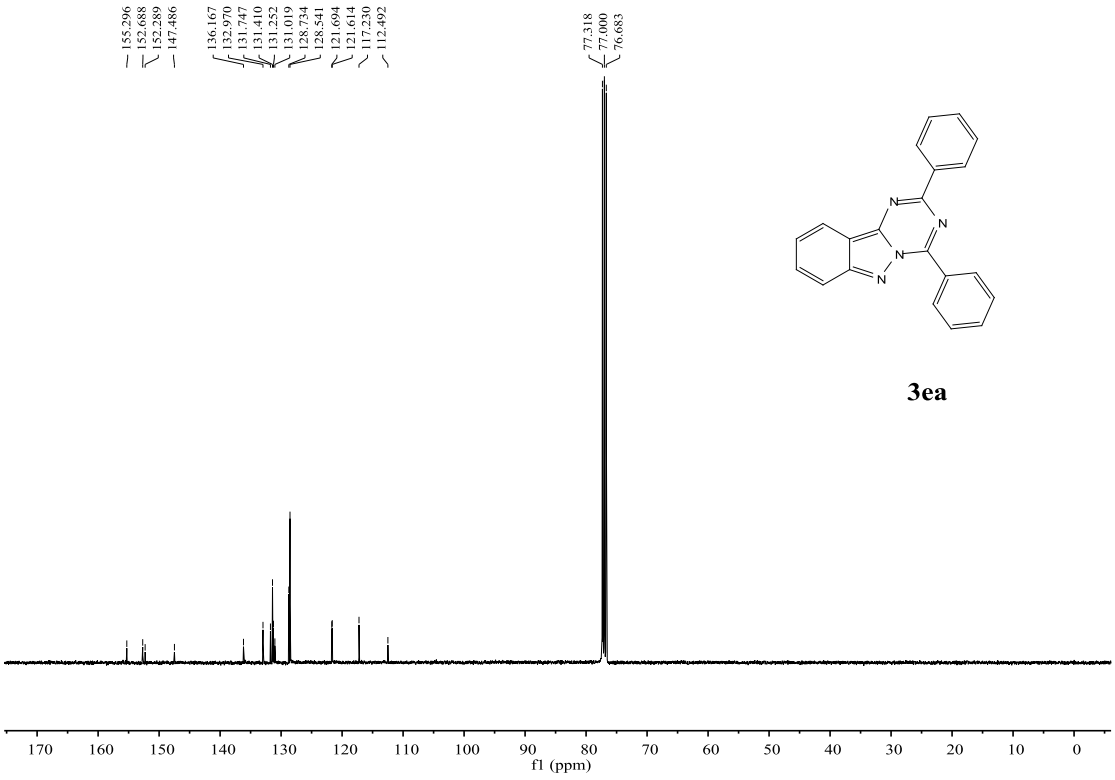
**3da**


163.765
158.402
153.105
145.202
135.059
132.974
132.888
131.469
129.673
129.511
128.752
128.271
126.031
124.705
123.343
119.082
117.393
77.320
76.685
Br
Br
N
N
N
N
80 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

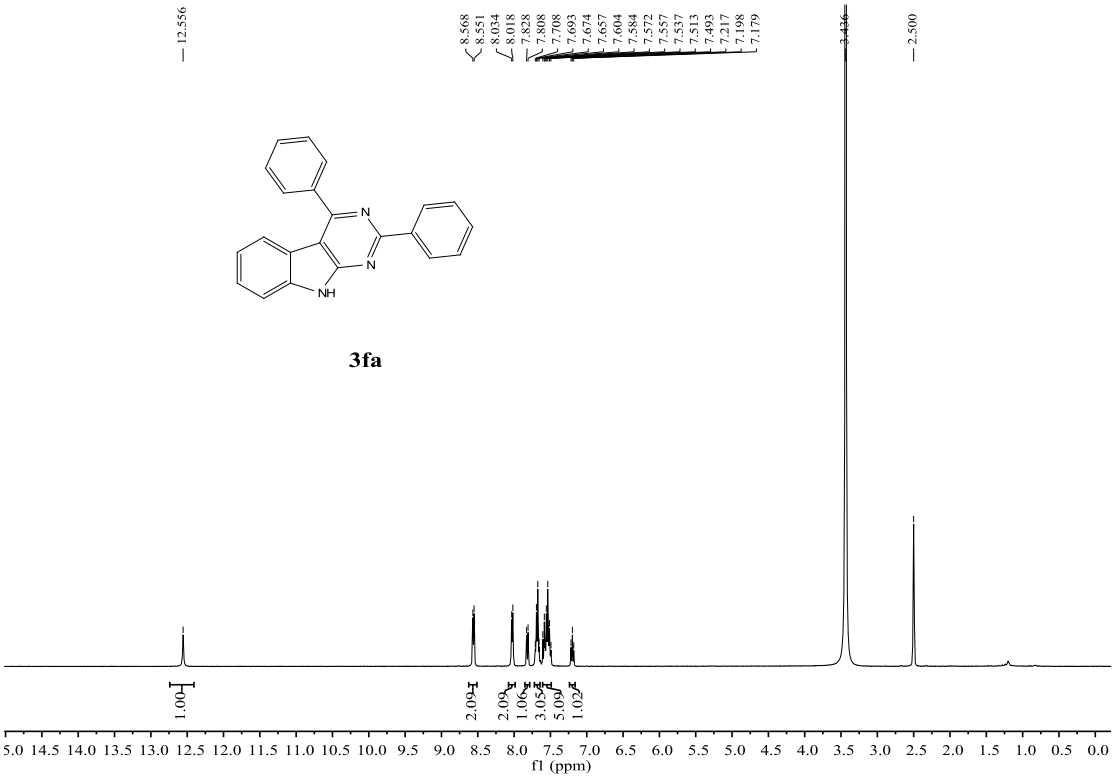
**3da**


9.116
9.096
8.761
8.741
8.406
8.385
7.917
7.895
7.709
7.691
7.672
7.587
7.569
7.353
7.334
7.316
7.260
1.582
-0.000
2.01
2.01
1.00
1.02
4.01
3.03
1.07
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)

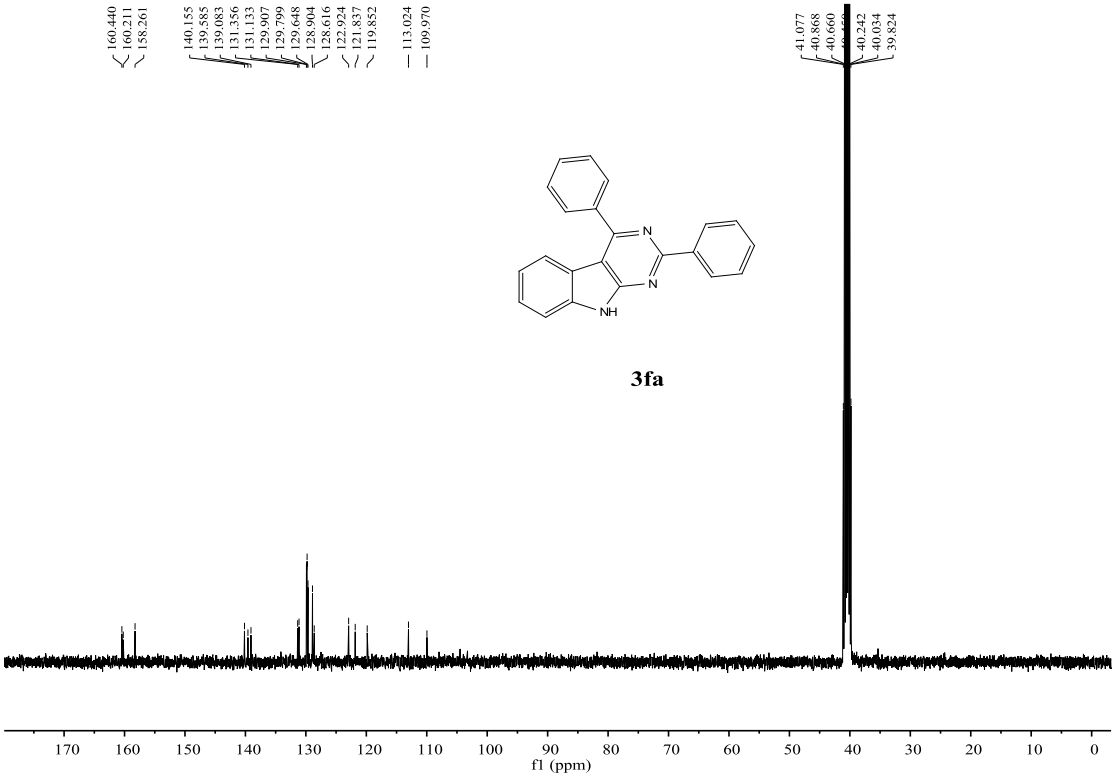
**3ea**


155.296
152.688
152.289
147.486
136.167
132.970
131.747
131.410
131.252
131.019
128.734
128.541
121.694
121.614
117.230
112.492
77.318
77.000
76.683
170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

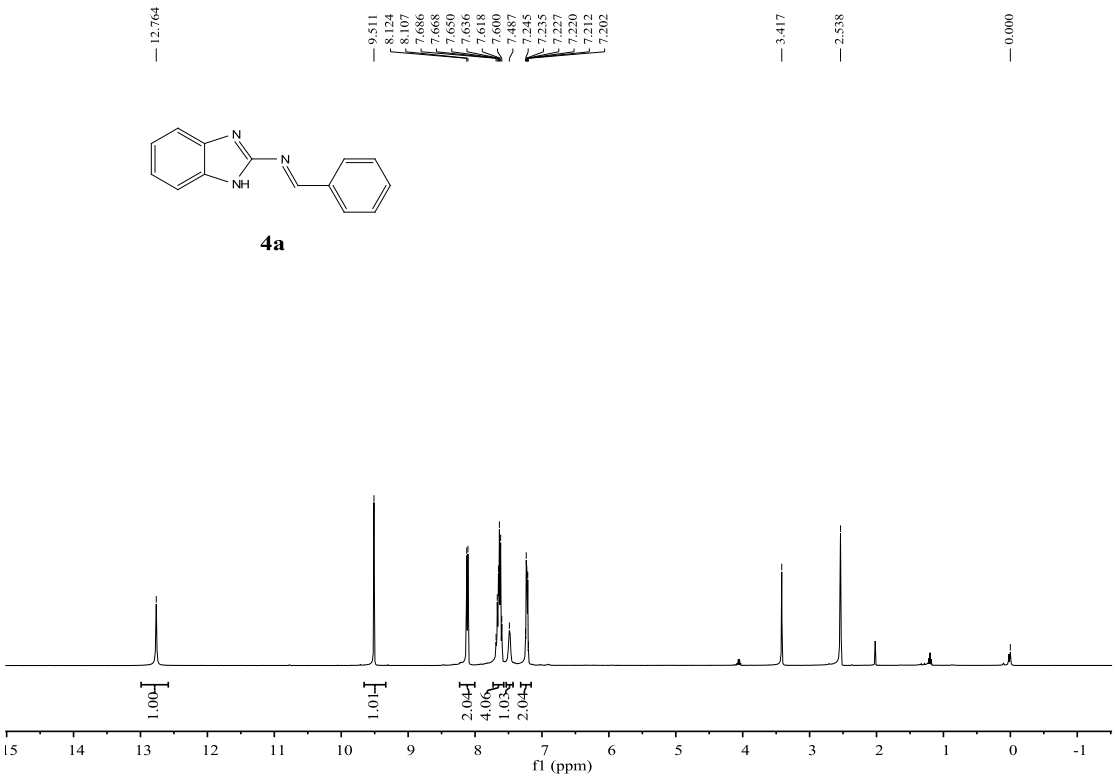
**3ea**


12.556
8.568
8.551
8.034
8.018
7.828
7.808
7.708
7.693
7.674
7.657
7.604
7.584
7.572
7.557
7.537
7.513
7.493
7.217
7.198
7.179
3.436
2.500
N
N
NH
1.00
2.09
2.09
1.06
3.05
5.09
1.02
f1 (ppm)

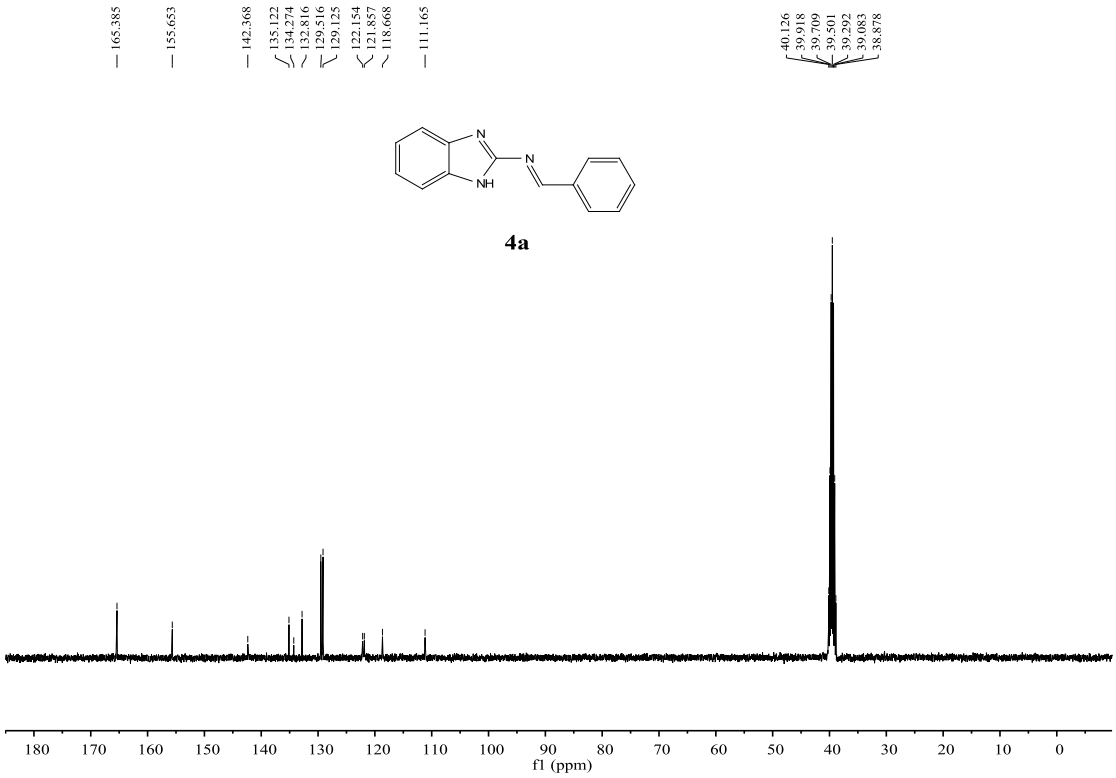
**3fa**


160.440
160.211
158.261
140.155
139.585
139.083
131.356
131.133
129.907
129.799
129.648
128.904
128.616
122.924
121.837
119.852
113.024
109.970
41.077
40.868
40.660
40.242
40.034
39.824
N
N
NH
f1 (ppm)

**3fa**


12.764
9.511
8.124
8.107
7.686
7.668
7.650
7.636
7.618
7.600
7.487
7.245
7.235
7.227
7.220
7.212
7.202
3.417
2.538
0.000
N
N
NH
1.00
1.01
2.04
4.06
1.03
2.04
15 14 13 12 11 10 9 8 7 6 5 4 3 2 1 0 -1
f1 (ppm)

**4a**


165.385
155.653
142.368
135.122
134.274
132.816
129.516
129.125
122.154
121.857
118.668
111.165
40.126
39.918
39.709
39.501
39.292
39.083
38.878
N
N
NH
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

**4a**