

Iron-catalyzed [3+2+1] annulation of 2-aminobenzimidazoles/3-aminopyrazoles and aromatic alkynes using *N,N*-dimethylaminoethanol as one carbon synthon for the synthesis of pyrimido[1,2-*a*]benzimidazoles and pyrimido[1,2-*b*]indazoles

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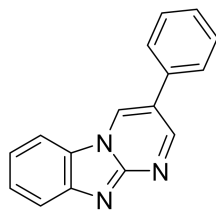
General methods. Starting materials, reagents and solvents were purchased commercial sources and used as received. ¹H and ¹³C NMR spectra were recorded at 400 and 101 MHz, respectively. High-resolution mass spectra (HRMS) were performed on a Waters SYNAPT G2-Si mass spectrometer. HPLC analyses were carried out on an Agilent 1260 Infinity II instrument. Melting points were determined using a X-4 digital micro melting point apparatus. All reactions were monitored by thin-layer chromatography (TLC) using silica gel plates (silica gel 60 F₂₅₄).

General Procedure for the synthesis of 3-substituted pyrimido[1,2-a]benzimidazoles and pyrimido[1,2-b]indazoles 3a-3g' from alkynes The reaction was carried out with **1** (1 mmol), **2** (1 mmol), FeCl₃ (20 mol%) in DMEA (2 mL) at 140 °C for 36 h in a sealed tube under air atmosphere. After being cooled to room temperature, the reaction was extracted with EtOAc. The combine organic layer was washed with H₂O and brine, and then dried over anhydrous Na₂SO₄. The solvent was removed under reduced pressure and the residue was purified by chromatography on silica gel to afford the desired product.

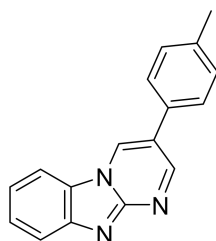
General Procedure for the synthesis of pyrimido[1,2-a]benzimidazoles (3a, 3c) and pyrimido[1,2-b]indazole 3b' from acetals The reaction was carried out with **1** (1 mmol), **6** (1 mmol), FeCl₃ (20 mol%) in DMEA (2 mL) at 140 °C for 36 h in a sealed tube under air atmosphere. After being cooled to room temperature, the reaction was extracted with EtOAc. The combine organic layer was washed with H₂O and brine, and then dried over anhydrous Na₂SO₄. The solvent was removed under reduced pressure and the residue was purified by chromatography on silica gel to afford the desired product.

General Procedure for the synthesis of 2-substituted pyrimido[1,2-a]benzimidazoles 4a-4k The reaction was carried out with **1a** (1 mmol), **2** (1 mmol), TfOH (2 mmol), FeCl₃ (20 mol%) in DMEA (2 mL) at 140 °C for 36 h in a sealed tube under air atmosphere. After being cooled to room temperature, the reaction was extracted with EtOAc. The combine organic layer was washed with H₂O and brine, and then dried over anhydrous Na₂SO₄. The solvent was removed under reduced pressure and the residue was purified by chromatography on silica gel to afford the desired product.

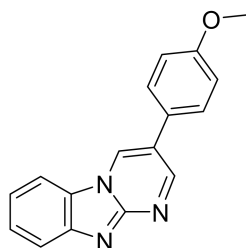
Gram-scale synthesis of pyrimido[1,2-a]benzimidazole (3a) The reaction was carried out with **1a** (10 mmol), **2a** (10 mmol), FeCl₃ (2 mmol) in DMEA (20 mL) at 140 °C for 36 h in a round flask under air atmosphere. After being cooled to room temperature, water (20 mL) was added and the reaction mixture was extracted with EtOAc (30 mL x2). The combine organic layer was washed with H₂O and brine, and then dried over anhydrous Na₂SO₄. The solvent was removed under reduced pressure and the residue was purified by chromatography on silica gel to afford the desired product.



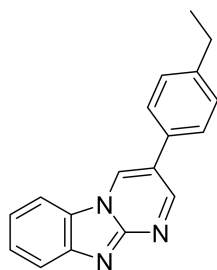
3-Phenylbenzo[4,5]imidazo[1,2-*a*]pyrimidine (3a)¹ Yellow solid, (152 mg, 65%); mp 205.4–207.6 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.05 (d, *J* = 2.4 Hz, 1H), 8.85 (d, *J* = 2.4 Hz, 1H), 8.02 (d, *J* = 8.4 Hz, 1H), 7.93 (d, *J* = 8.4 Hz, 1H), 7.63 (d, *J* = 7.2 Hz, 2H), 7.58 (t, *J* = 7.2 Hz, 1H), 7.55 (t, *J* = 7.2 Hz, 2H), 7.47 (t, *J* = 7.2 Hz, 1H), 7.43 (t, *J* = 7.2 Hz, 1H). ¹³C NMR (101 MHz, CDCl₃) δ 155.7, 150.0, 144.7, 133.9, 129.8, 129.6, 128.7, 127.0, 126.9, 126.6, 122.4, 121.1, 120.8, 110.8. ESI-MS: *m/z* [M+H]⁺ 246.



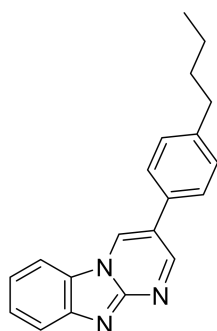
3-(4-Tolyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3b) Yellow solid, (155 mg, 60%); mp 196.6–198.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.06 (d, *J* = 2.4 Hz, 1H), 8.83 (d, *J* = 2.4 Hz, 1H), 8.03 (d, *J* = 8.4 Hz, 1H), 7.94 (d, *J* = 8.0 Hz, 1H), 7.59 (t, *J* = 7.6 Hz, 1H), 7.53 (d, *J* = 8.0 Hz, 2H), 7.44 (t, *J* = 7.6 Hz, 1H), 7.36 (d, *J* = 8.0 Hz, 2H), 2.45 (s, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 155.8, 150.0, 144.6, 138.7, 130.9, 130.2, 129.3, 126.9, 126.7, 126.6, 122.2, 121.2, 120.8, 110.7, 21.2. ESI-MS: *m/z* [M+H]⁺ 314. HRMS (ESI): calcd. for C₁₇H₁₄N₃⁺ [M+H]⁺ 260.1182; found 260.1180.



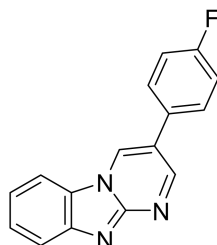
3-(4-Methoxyphenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3c) Yellow solid, (173 mg, 63%); mp 201.3–205.8 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.04 (d, *J* = 2.4 Hz, 1H), 8.79 (d, *J* = 2.4 Hz, 1H), 8.03 (d, *J* = 8.4 Hz, 1H), 7.93 (d, *J* = 8.4 Hz, 1H), 7.59 (t, *J* = 7.6 Hz, 1H), 7.56 (d, *J* = 8.8 Hz, 2H), 7.44 (t, *J* = 7.6 Hz, 1H), 7.08 (d, *J* = 8.8 Hz, 2H), 3.89 (s, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 160.1, 155.8, 149.9, 144.6, 128.9, 128.1, 127.0, 126.5, 126.1, 122.1, 121.0, 120.8, 115.0, 110.7, 55.5. HRMS (ESI): calcd. for C₁₇H₁₄N₃O⁺ [M+H]⁺ 276.1131; found 276.1130.



3-(4-Ethylphenyl)benzo[4,5]imidazo[1,2-a]pyrimidine (3d) Yellow solid, (169 mg, 62%); mp 200.3–204.8 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.05 (d, *J* = 2.6 Hz, 1H), 8.82 (d, *J* = 2.6 Hz, 1H), 8.02 (d, *J* = 8.3 Hz, 1H), 7.93 (d, *J* = 8.2 Hz, 1H), 7.58 (t, *J* = 8.0 Hz, 1H), 7.55 (d, *J* = 8.0 Hz, 2H), 7.43 (t, *J* = 8.0 Hz, 1H), 7.38 (d, *J* = 8.0 Hz, 2H), 2.74 (q, *J* = 7.6 Hz, 2H), 1.30 (t, *J* = 7.6 Hz, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 155.8, 150.0, 145.1, 144.7, 131.1, 129.4, 129.1, 126.9, 126.8, 126.5, 122.2, 121.2, 120.8, 110.7, 28.6, 15.6. HRMS (ESI): calcd. for C₁₈H₁₆N₃⁺ [M+H]⁺ 274.1339; found 274.1336.

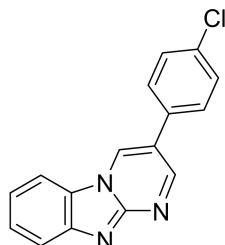


3-(4-Butylphenyl)benzo[4,5]imidazo[1,2-a]pyrimidine (3e) Yellow solid, (211 mg, 70%); mp 205.3–207.3 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.02 (d, *J* = 2.4 Hz, 1H), 8.81 (d, *J* = 2.4 Hz, 1H), 7.99 (d, *J* = 8.4 Hz, 1H), 7.91 (d, *J* = 8.4 Hz, 1H), 7.55 (t, *J* = 8.0 Hz, 1H), 7.52 (t, *J* = 8.0 Hz, 2H), 7.40 (t, *J* = 7.6 Hz, 1H), 7.34 (d, *J* = 8.0 Hz, 2H), 2.69 (t, *J* = 7.6 Hz, 2H), 1.72–1.56 (m, 2H), 1.47–1.31 (m, 2H), 0.96 (t, *J* = 7.2 Hz, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 155.8, 149.9, 144.6, 143.7, 131.0, 129.6, 129.4, 126.9, 126.7, 126.5, 122.1, 121.1, 120.7, 110.7, 35.3, 33.5, 22.3, 13.9. HRMS (ESI): calcd. for C₂₀H₂₀N₃⁺ [M+H]⁺ 302.1652; found 302.1650.

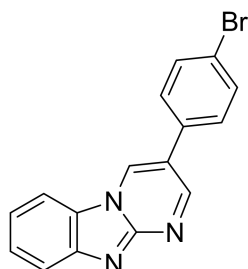


3-(4-Fluorophenyl)benzo[4,5]imidazo[1,2-a]pyrimidine (3f) Yellow solid, (147 mg, 56%); mp 198.3–200.6 °C. ¹H NMR (400 MHz, DMSO) δ 9.77 (d, *J* = 2.4 Hz, 1H), 9.16 (d, *J* = 2.4 Hz, 1H), 8.38 (d, *J* = 8.0 Hz, 1H), 7.91 (dd, *J* = 8.8, 5.2 Hz, 2H), 7.85

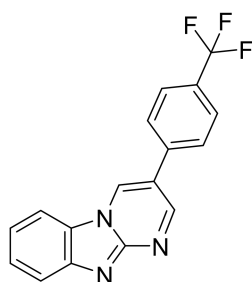
(d, $J = 8.0$ Hz, 1H), 7.56 (t, $J = 7.6$ Hz, 1H), 7.44 (t, $J = 8.0$ Hz, 1H), 7.38 (t, $J = 8.8$ Hz, 2H). ^{13}C NMR (101 MHz, DMSO) δ 162.7 (d, $J = 244$ Hz), 156.1, 149.8, 144.2, 132.8, 130.6 (d, $J = 3.1$ Hz), 129.3 (d, $J = 8.3$ Hz), 127.6, 126.8, 122.1, 119.7, 119.31, 116.5 (d, $J = 21.3$ Hz), 113.4. HRMS (ESI): calcd. for $\text{C}_{16}\text{H}_{11}\text{FN}_3^+$ $[\text{M}+\text{H}]^+$ 264.0932; found 264.0930.



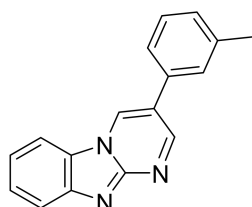
3-(4-Chlorophenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3g) Yellow solid, (212 mg, 76%); mp 201.6–203.5 °C. ^1H NMR (400 MHz, CDCl_3) δ 9.02 (d, $J = 2.4$ Hz, 1H), 8.86 (d, $J = 2.4$ Hz, 1H), 8.04 (d, $J = 8.4$ Hz, 1H), 7.95 (d, $J = 8.4$ Hz, 1H), 7.65–7.51 (m, 5H), 7.47 (t, $J = 7.6$ Hz, 1H). ^{13}C NMR (101 MHz, CDCl_3) δ 155.3, 149.8, 144.7, 135.0, 132.3, 129.8, 129.8, 128.2, 126.9, 126.8, 122.5, 120.9, 120.1, 110.7. HRMS (ESI): calcd. for $\text{C}_{16}\text{H}_{11}\text{ClN}_3^+$ $[\text{M}+\text{H}]^+$ 280.0636; found 280.0639.



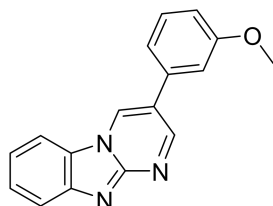
3-(4-Bromophenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3h) Yellow solid, (210 mg, 65%); mp 198.3–201.5 °C. ^1H NMR (400 MHz, CDCl_3) δ 9.03 (s, 1H), 8.87 (s, 1H), 8.06 (d, $J = 8.0$ Hz, 1H), 7.96 (d, $J = 8.0$ Hz, 1H), 7.70 (d, $J = 8.0$ Hz, 2H), 7.63 (t, $J = 7.6$ Hz, 1H), 7.52 (d, $J = 8.0$ Hz, 2H), 7.48 (t, $J = 7.6$ Hz, 1H). ^{13}C NMR (101 MHz, CDCl_3) δ 155.22, 149.87, 144.73, 132.80, 129.69, 128.46, 126.86, 123.13, 122.50, 120.99, 120.21, 120.13, 110.74. HRMS (ESI): calcd. for $\text{C}_{16}\text{H}_{11}\text{BrN}_3^+$ $[\text{M}+\text{H}]^+$ 324.0131; found 324.0134.



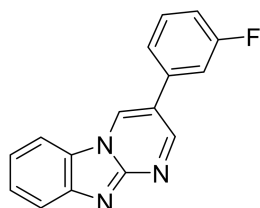
3-(4-(Trifluoromethyl)phenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3i) Yellow solid,(225 mg, 72%); mp 197.8.3–199.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.08 (s, 1H), 8.93 (s, 1H), 8.07 (d, *J* = 8.0 Hz, 1H), 7.97 (d, *J* = 8.0 Hz, 1H), 7.83 (d, *J* = 8.0 Hz, 2H), 7.78 (d, *J* = 8.0 Hz, 2H), 7.64 (t, *J* = 7.6 Hz, 1H), 7.50 (t, *J* = 7.6 Hz, 1H). ¹³C NMR (101 MHz, DMSO) δ 155.9, 149.9, 144.5, 138.4, 134.2, 127.9, 127.8, 126.9, 126.5 (q, *J* = 3.6 Hz), 124.8 (q, *J* = 270 Hz), 122.2, 119.8, 118.5, 113.6. HRMS (ESI): calcd. for C₁₇H₁₁F₃N₃⁺ [M+H]⁺ 314.0900; found 314.0904.



3-(3-Tolyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3j) Yellow solid,(166 mg, 64%); mp 198.7-200.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.03 (d, *J* = 2.4 Hz, 1H), 8.83 (d, *J* = 2.4 Hz, 1H), 8.01 (d, *J* = 8.4 Hz, 1H), 7.93 (d, *J* = 8.0 Hz, 1H), 7.57 (t, *J* = 7.6 Hz, 1H), 7.48–7.35(m, 4H), 7.32–7.26 (m, 1H), 2.47 (s, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 155.8, 150.0, 144.6, 139.4, 133.7, 129.7, 129.4, 129.4, 127.6, 126.9, 126.6, 123.9, 122.2, 121.2, 120.7, 110.78, 21.5. HRMS (ESI): calcd. for C₁₇H₁₄N₃⁺ [M+H]⁺ 260.1182; found 260.1178.

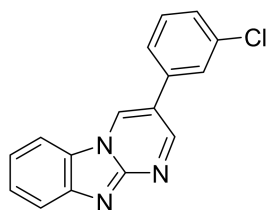


3-(3-Methoxyphenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3k) Yellow solid, (171 mg, 62%); mp 198.3–200.8 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.05 (d, *J* = 2.4 Hz, 1H), 8.85 (d, *J* = 2.4 Hz, 1H), 8.03 (d, *J* = 8.4 Hz, 1H), 7.94 (d, *J* = 8.4 Hz, 1H), 7.60 (t, *J* = 8.0 Hz, 1H), 7.48–7.42 (m, 2H), 7.20 (d, *J* = 7.6 Hz, 1H), 7.14 (d, *J* = 2.4 Hz, 1H), 7.01 (dd, *J* = 8.4, 2.4 Hz, 1H). ¹³C NMR (101 MHz, CDCl₃) δ 160.4, 155.7, 150.0, 144.7, 135.2, 130.7, 129.8, 126.9, 126.7, 122.3, 121.0, 120.8, 119.2, 113.7, 112.9, 110.7, 55.5. HRMS (ESI): calcd. for C₁₇H₁₄N₃O⁺ [M+H]⁺ 276.1131; found 276.1130.

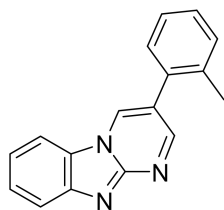


3-(3-Fluorophenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3l) Yellow solid, (147 mg, 56%); mp 198.3–200.8 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.00 (d, *J* = 2.4 Hz, 1H), 8.87 (d, *J* = 2.4 Hz, 1H), 8.01 (d, *J* = 8.0 Hz, 1H), 7.94 (d, *J* = 8.0 Hz, 1H), 7.58 (t, *J* =

8.0 Hz, 1H), 7.55-7.50 (m, 1H), 7.46-7.40 (m, 2H), 7.34 (d, $J = 8.4$ Hz, 1H), 7.17 (td, $J = 8.4, 2.4$ Hz, 1H). ^{13}C NMR (101 MHz, CDCl_3) δ 163.4 (d, $J = 246$ Hz), 155.2, 149.8, 144.7, 136.0 (d, $J = 7.9$ Hz), 131.3 (d, $J = 8.5$ Hz), 130.1, 126.9, 126.8, 122.6 (d, $J = 2.8$ Hz), 122.4, 120.8, 119.9, 115.6 (d, $J = 21.0$ Hz), 113.9 (d, $J = 22.5$ Hz), 110.8. HRMS (ESI): calcd. for $\text{C}_{16}\text{H}_{11}\text{FN}_3^+$ $[\text{M}+\text{H}]^+$ 264.0930; found 264.0935.

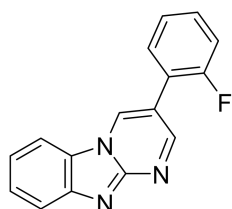


3-(4-Chlorophenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3m) Yellow solid, (167 mg, 60%); mp 200.3–203.4 °C. ^1H NMR (400 MHz, CDCl_3) δ 9.00 (d, $J = 2.4$ Hz, 1H), 8.88 (d, $J = 2.4$ Hz, 1H), 8.02 (d, $J = 8.4$ Hz, 1H), 7.96 (d, $J = 8.4$ Hz, 1H), 7.62 (s, 1H), 7.60 (s, $J = 8.0$ Hz, 1H), 7.53–7.44 (m, 4H). ^{13}C NMR (101 MHz, CDCl_3) δ 155.1, 149.8, 144.7, 135.7, 135.5, 130.8, 130.1, 128.7, 126.9, 126.9, 126.8, 125.1, 122.5, 120.9, 119.8, 110.8. HRMS (ESI): calcd. for $\text{C}_{16}\text{H}_{11}\text{ClN}_3^+$ $[\text{M}+\text{H}]^+$ 280.0636; found 280.0630.



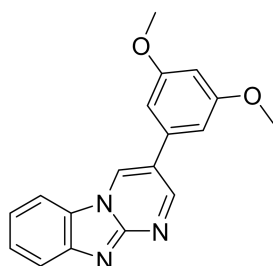
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3-(2-Tolyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3n) Yellow solid, (142 mg, 55%); mp 204.3–206.7 °C. ^1H NMR (400 MHz, CDCl_3) δ 8.80 (d, $J = 2.0$ Hz, 1H), 8.70 (d, $J = 2.4$ Hz, 1H), 8.03 (d, $J = 8.4$ Hz, 1H), 7.90 (d, $J = 8.4$ Hz, 1H), 7.59 (t, $J = 7.6$ Hz, 1H), 7.43 (t, $J = 7.6$ Hz, 1H), 7.40–7.28 (m, 4H), 2.37 (s, 3H). ^{13}C NMR (101 MHz, CDCl_3) δ 157.1, 149.8, 144.6, 136.3, 133.5, 131.4, 130.9, 130.3, 129.1, 126.9, 126.6, 126.6, 122.2, 121.4, 120.7, 110.7, 20.5. HRMS (ESI): calcd. for $\text{C}_{17}\text{H}_{14}\text{N}_3^+$ $[\text{M}+\text{H}]^+$ 260.1182; found 260.1180.

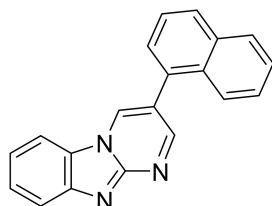


3-(2-Fluorophenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3o) Yellow solid, (158 mg, 60%); mp 198.6–201.5 °C. ^1H NMR (400 MHz, CDCl_3) δ 8.97 (s, 1H), 8.93 (s, 1H), 8.00 (d, $J = 8.4$ Hz, 1H), 7.91 (d, $J = 8.4$ Hz, 1H), 7.61–7.50 (m, 2H), 7.49–7.38 (m,

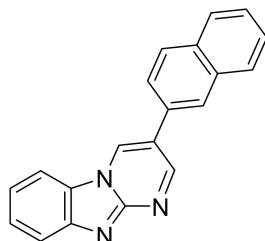
2H), 7.32 (t, $J = 7.6$ Hz, 1H), 7.24 (t, $J = 9.2$ Hz, 1H). ^{13}C NMR (101 MHz, CDCl_3) δ 159.8 (d, $J = 245$ Hz), 156.1 (d, $J = 2.7$ Hz), 149.6, 144.6, 132.0 (d, $J = 5.3$ Hz), 130.6 (d, $J = 8.4$ Hz), 129.9 (d, $J = 2.8$ Hz), 126.9, 126.7, 125.2 (d, $J = 3.7$ Hz), 122.3, 121.5 (d, $J = 13.2$ Hz), 120.7, 116.6 (d, $J = 21.9$ Hz), 115.4, 110.8. HRMS (ESI): calcd. for $\text{C}_{16}\text{H}_{11}\text{FN}_3^+ [\text{M}+\text{H}]^+$ 264.0932; found 264.0930.



3-(3,5-Dimethoxyphenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3p) Yellow solid, (190 mg, 62%); mp 200.5–203.6 °C. ^1H NMR (400 MHz, CDCl_3) δ 9.02 (s, 1H), 8.83 (d, $J = 2.4$ Hz, 1H), 8.02 (d, $J = 8.4$ Hz, 1H), 7.93 (d, $J = 8.4$ Hz, 1H), 7.59 (t, $J = 7.6$ Hz, 1H), 7.44 (t, $J = 7.6$ Hz, 1H), 6.72 (d, $J = 1.2$ Hz, 2H), 6.54 (s, 1H), 3.88 (s, 6H). ^{13}C NMR (101 MHz, CDCl_3) δ 161.7, 155.6, 150.0, 144.7, 135.8, 129.9, 126.9, 126.6, 122.3, 121.1, 120.8, 110.8, 105.3, 100.0, 55.6. HRMS (ESI): calcd. for $\text{C}_{18}\text{H}_{16}\text{N}_3\text{O}_2^+ [\text{M}+\text{H}]^+$ 306.1237; found 306.1235.

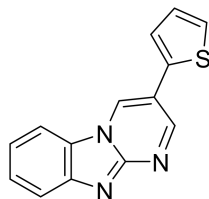


3-(Naphthalen-1-yl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3q)² Yellow solid, (194 mg, 66%); mp 202.3–204.3 °C. ^1H NMR (400 MHz, CDCl_3) δ 8.92 (d, $J = 2.4$ Hz, 1H), 8.84 (d, $J = 2.4$ Hz, 1H), 8.05 (d, $J = 8.4$ Hz, 1H), 8.02–7.94 (m, 2H), 7.90 (d, $J = 8.4$ Hz, 1H), 7.83 (d, $J = 8.4$ Hz, 1H), 7.63–7.48 (m, 5H), 7.43 (t, $J = 7.6$ Hz, 1H). ^{13}C NMR (101 MHz, CDCl_3) δ 157.5, 149.9, 144.6, 133.9, 132.1, 131.7, 131.6, 129.5, 128.8, 128.2, 127.2, 126.9, 126.6, 126.5, 125.5, 124.6, 122.3, 120.8, 120.3, 110.8. HRMS (ESI): calcd. for $\text{C}_{20}\text{H}_{14}\text{N}_3^+ [\text{M}+\text{H}]^+$ 296.1182; found 296.1178.

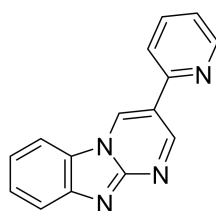


3-(Naphthalen-2-yl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (3r)¹ Yellow solid, (215 mg, 73%); mp 203.5–205.4 °C. ^1H NMR (400 MHz, CDCl_3) δ 9.20 (d, $J = 2.4$ Hz, 1H), 8.97 (d, $J = 2.4$ Hz, 1H), 8.12–7.89 (m, 6H), 7.73 (dd, $J = 8.4, 1.6$ Hz, 1H), 7.66–7.52 (m, 3H), 7.47 (t, $J = 7.6$ Hz, 1H). ^{13}C NMR (101 MHz, CDCl_3) δ 155.8,

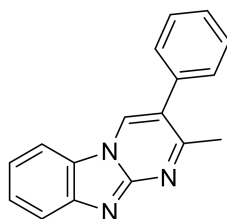
145.6, 133.6, 132.9, 131.1, 129.9, 129.5, 128.2, 127.8, 127.1, 126.9, 126.7, 126.0, 124.3, 122.3, 121.1, 120.9, 110.8. HRMS (ESI): calcd. for $C_{20}H_{14}N_3^+$ $[M+H]^+$ 296.1182; found 296.1180.



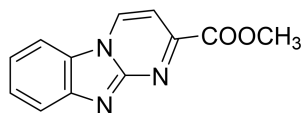
3-(Thiophen-2-yl)benzo[4,5]imidazo[1,2-a]pyrimidine (3s) Yellow solid, (108 mg, 43%); mp 200.3–204.3 °C. 1H NMR (400 MHz, $CDCl_3$) δ 9.07 (d, $J = 2.4$ Hz, 1H), 8.86 (d, $J = 2.4$ Hz, 1H), 8.03 (d, $J = 8.4$ Hz, 1H), 7.94 (d, $J = 8.4$ Hz, 1H), 7.60 (t, $J = 7.6$ Hz, 1H), 7.46 (t, $J = 7.6$ Hz, 1H), 7.43 (d, $J = 4.8$ Hz, 1H), 7.40 (d, $J = 3.2$ Hz, 1H), 7.19 (dd, $J = 4.0, 3.6$ Hz, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 154.7, 149.7, 144.6, 135.9, 128.6, 128.4, 126.9, 126.7, 126.3, 124.9, 122.4, 120.9, 115.6, 110.8. HRMS (ESI): calcd. for $C_{14}H_{10}N_3S^+$ $[M+H]^+$ 252.0590; found 252.0586.



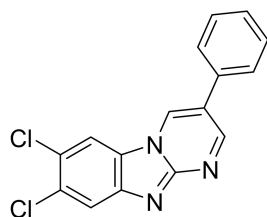
3-(Pyridin-2-yl)benzo[4,5]imidazo[1,2-a]pyrimidine (3t) Yellow solid, (79 mg, 32%); mp 202.3–204.3 °C. 1H NMR (400 MHz, $CDCl_3$) δ 9.55 (d, $J = 2.4$ Hz, 1H), 9.38 (d, $J = 2.4$ Hz, 1H), 8.74 (d, $J = 4.4$ Hz, 1H), 8.03 (t, $J = 8.8$ Hz, 2H), 7.84–7.87 (m, 2H), 7.61 (t, $J = 8.4$ Hz, 1H), 7.47 (t, $J = 8.0$ Hz, 1H), 7.35 (t, $J = 6.8$ Hz, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 154.2, 151.4, 150.3, 144.9, 137.5, 131.7, 127.2, 126.9, 123.2, 122.4, 120.8, 119.5, 119.2, 111.1. HRMS (ESI): calcd. for $C_{15}H_{11}N_4^+$ $[M+H]^+$ 247.0978; found 247.0975.



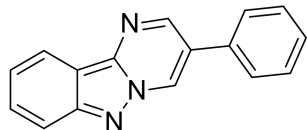
2-Methyl-3-phenylbenzo[4,5]imidazo[1,2-a]pyrimidine (3u) Yellow solid, (18mg, 65%); mp 200.3–204.8 °C. 1H NMR (400 MHz, $CDCl_3$) δ 8.59 (s, 1H), 8.00 (d, $J = 8.4$ Hz, 1H), 7.87 (d, $J = 8.0$ Hz, 1H), 7.76–7.72 (m, 2H), 7.62–7.45 (m, 4H), 7.40 (t, $J = 7.7$ Hz, 1H), 2.46 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.6, 150.4, 144.9, 138.4, 131.8, 129.7, 129.0, 128.4, 126.5, 126.1, 121.7, 120.5, 115.0, 110.5, 18.0. HRMS (ESI): calcd. for $C_{17}H_{14}N_3^+$ $[M+H]^+$ 260.1182; found 260.1186.



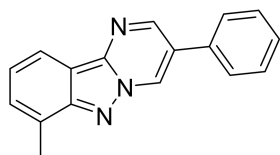
Methyl benzo[4,5]imidazo[1,2-a]pyrimidine-2-carboxylate (3v) Yellow solid, (68 mg, 30%); mp 196.5–199.8 °C. ¹H NMR (400 MHz, CDCl₃) δ 8.09 (d, *J* = 8.0 Hz, 1H), 7.73 (d, *J* = 8.0 Hz, 1H), 7.53 (d, *J* = 8.0 Hz, 1H), 7.40 (t, *J* = 8.0 Hz, 1H), 7.31 (t, *J* = 7.6 Hz, 1H), 6.23 (d, *J* = 8.0 Hz, 1H), 3.76 (s, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 177.6, 160.5, 147.9, 142.4, 131.8, 129.3, 125.0, 122.0, 119.0, 108.7, 105.5, 58.5. HRMS (ESI): calcd. for C₁₂H₁₀N₃O₂⁺ [M+H]⁺: 228.0768; found 228.0766.



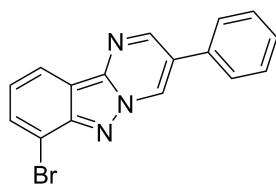
7,8-Dichloro-3-phenylbenzo[4,5]imidazo[1,2-a]pyrimidine (3a') Yellow solid, (163 mg, 52%); mp 200.3–203.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.11 (d, *J* = 2.4 Hz, 1H), 8.79 (d, *J* = 2.4 Hz, 1H), 8.13 (s, 1H), 8.09 (s, 1H), 7.64–7.49 (m, 5H). ¹³C NMR (101 MHz, CDCl₃) δ 156.8, 151.0, 143.7, 133.2, 131.1, 129.7 (2), 129.3, 129.1, 128.1, 126.9, 122.2, 121.9, 112.4. HRMS (ESI): calcd. for C₁₆H₁₀Cl₂N₃⁺ [M+H]⁺ 314.0246; found 314.0243.



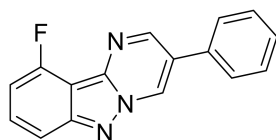
3-Phenylpyrimido[1,2-b]indazole (3b ')⁴ Yellow solid, (161 mg, 62%); mp 197.8–200.6 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.17 (d, *J* = 2.0 Hz, 1H), 8.86 (d, *J* = 2.0 Hz, 1H), 8.15 (d, *J* = 8.4 Hz, 1H), 7.87 (d, *J* = 8.8 Hz, 1H), 7.70 (d, *J* = 7.2 Hz, 2H), 7.67 (t, *J* = 7.2 Hz, 1H), 7.58 (t, *J* = 7.6 Hz, 2H), 7.51 (t, *J* = 7.2 Hz, 1H), 7.36 (t, *J* = 7.6 Hz, 1H). ¹³C NMR (101 MHz, CDCl₃) δ 151.8, 145.2, 142.7, 133.9, 130.7, 130.0, 129.6, 129.1, 127.4, 126.3, 121.3, 120.7, 116.2, 113.4. ESI-MS: *m/z* [M+H]⁺ 246.



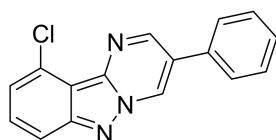
7-Methyl-3-phenylpyrimido[1,2-b]indazole (3c') Yellow solid, (161 mg, 62%); mp 197.8–200.6 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.17 (d, *J* = 2.0 Hz, 1H), 8.86 (d, *J* = 2.0 Hz, 1H), 8.15 (d, *J* = 8.4 Hz, 1H), 7.66 (d, *J* = 7.2 Hz, 2H), 7.55 (t, *J* = 7.2 Hz, 2H), 7.48 (t, *J* = 7.2 Hz, 1H), 7.41 (d, *J* = 6.8 Hz, 1H), 7.24 (t, *J* = 7.2 Hz, 1H), 2.75 (s, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 155.2, 149.9, 147.7, 144.8, 132.9, 132.8, 129.7, 128.4, 126.8, 123.1, 122.5, 121.0, 120.1, 110.7, 18.5. HRMS (ESI): calcd. for C₁₇H₁₄N₃⁺ [M+H]⁺ 260.1182; found 260.1180.



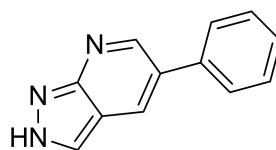
7-Bromo-2-phenylpyrimido[1,2-*b*]indazole (3d') Yellow solid, (149 mg, 46%); mp 197.0–199.0 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.28 (d, *J* = 2.0 Hz, 1H), 8.96 (d, *J* = 2.0 Hz, 1H), 8.31 (d, *J* = 8.0 Hz, 1H), 7.88 (d, *J* = 6.8 Hz, 1H), 7.69 (d, *J* = 7.2 Hz, 2H), 7.59 (t, *J* = 7.2 Hz, 2H), 7.53 (t, *J* = 7.2 Hz, 1H), 7.23 (t, *J* = 8.0 Hz, 1H). ¹³C NMR (101 MHz, CDCl₃) δ 157.5, 150.0, 146.3, 133.5, 132.6, 131.2, 129.7, 129.3, 127.3, 126.9, 121.9, 120.1, 114.6, 109.6. HRMS (ESI): calcd. for C₁₆H₁₁BrN₃⁺ [M+H]⁺ 324.0131; found 324.0136.



10-Fluoro-3-phenylpyrimido[1,2-*b*]indazole (3e') Yellow solid, (158 mg, 60%); mp 197.8–200.6 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.15 (d, *J* = 2.0 Hz, 1H), 8.99 (d, *J* = 2.0 Hz, 1H), 7.69 (d, *J* = 7.2 Hz, 2H), 7.66–7.50 (m, 5H), 6.98 (dd, *J* = 10.0, 7.6 Hz, 1H). ¹³C NMR (101 MHz, CDCl₃) δ 157.1 (d, *J* = 255 Hz), 153.5 (d, *J* = 4.8 Hz), 146.4, 141.1 (d, *J* = 5.3 Hz), 133.5, 130.6, 130.4 (d, *J* = 7.8 Hz), 129.7, 129.3, 127.4, 126.6, 112.2 (d, *J* = 4.5 Hz), 105.2 (d, *J* = 17.6 Hz). HRMS (ESI): calcd. for C₁₆H₁₁FN₃⁺ [M+H]⁺ 264.0932; found 264.0930.

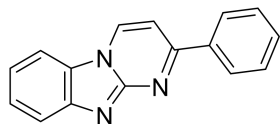


10-Chloro-3-phenylpyrimido[1,2-*b*]indazole (3f') Yellow solid, (134 mg, 48%); mp 203.8–206.5 °C. ¹H NMR (400 MHz, CDCl₃) δ 9.14 (d, *J* = 1.6 Hz, 1H), 9.03 (d, *J* = 1.6 Hz, 1H), 7.76 (d, *J* = 8.4 Hz, 1H), 7.68 (d, *J* = 7.2 Hz, 2H), 7.62–7.47 (m, 4H), 7.32 (d, *J* = 7.2 Hz, 1H). ¹³C NMR (101 MHz, CDCl₃) δ 152.4, 146.0, 142.0, 133.5, 130.7, 129.9, 129.7, 129.2, 127.4, 127.3, 126.6, 121.5, 114.8, 111.3. HRMS (ESI): calcd. for C₁₆H₁₁ClN₃⁺ [M+H]⁺ 280.0636; found 280.0631.

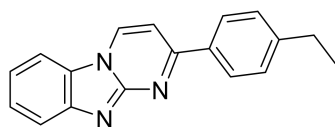


5-Phenyl-2H-pyrazolo[3,4-*b*]pyridine (3g') Yellow solid, (117 mg, 60%); mp 196.3–198.5 °C. ¹H NMR (400 MHz, CDCl₃) δ 12.38 (brs, 1H), 8.90 (d, *J* = 1.6 Hz,

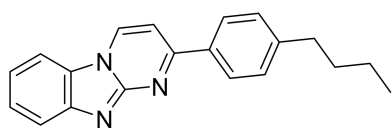
1H), 8.30 (d, $J = 2.0$ Hz, 1H), 8.19 (s, 1H), 7.65 (d, $J = 7.2$ Hz, 2H), 7.52 (t, $J = 7.6$ Hz, 2H), 7.43 (t, $J = 7.2$ Hz, 1H). ^{13}C NMR (101 MHz, CDCl_3) δ 151.3, 148.8, 138.3, 134.3, 131.1, 129.2, 128.3, 127.7, 127.5, 115.3. HRMS (ESI): calcd. for $\text{C}_{12}\text{H}_{11}\text{N}_3^+$ $[\text{M}+\text{H}]^+$ 196.0869; found 196.0862.



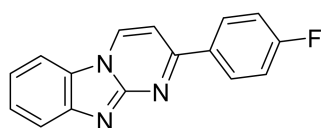
2-Phenylbenzo[4,5]imidazo[1,2-*a*]pyrimidine (4a)³ Yellow solid, (105 mg, 43%); mp 200.3–204.3 °C. ^1H NMR (400 MHz, CDCl_3) δ 8.78 (d, $J = 7.2$ Hz, 1H), 8.34–8.26 (m, 2H), 8.01 (d, $J = 8.0$ Hz, 1H), 7.89 (d, $J = 8.0$ Hz, 1H), 7.62–7.49 (m, 4H), 7.44 (d, $J = 7.2$ Hz, 1H), 7.42 (t, $J = 7.6$ Hz, 1H). ^{13}C NMR (101 MHz, CDCl_3) δ 161.6, 151.1, 145.1, 136.7, 133.1, 131.4, 129.0, 127.8, 127.0, 126.4, 121.9, 120.5, 110.4, 104.0. ESI-MS: m/z $[\text{M}+\text{H}]^+$ 246.



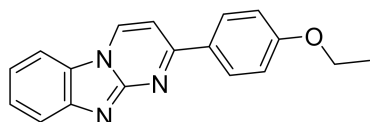
2-(4-Ethylphenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (4b) Yellow solid, (123 mg, 45%); mp 197.3–199.5 °C. ^1H NMR (400 MHz, CDCl_3) δ 8.72 (d, $J = 7.2$ Hz, 1H), 8.21 (d, $J = 8.0$ Hz, 2H), 7.99 (d, $J = 8.0$ Hz, 1H), 7.84 (d, $J = 8.0$ Hz, 1H), 7.56 (t, $J = 7.6$ Hz, 1H), 7.41–7.33 (m, 4H), 2.74 (q, $J = 7.6$ Hz, 2H), 1.30 (t, $J = 7.6$ Hz, 3H). ^{13}C NMR (101 MHz, CDCl_3) δ 161.6, 151.1, 148.3, 144.9, 134.1, 132.9, 128.5, 127.9, 127.0, 126.3, 121.7, 120.3, 110.3, 103.9, 28.8, 15.3. HRMS (ESI): calcd. for $\text{C}_{17}\text{H}_{14}\text{N}_3^+$ $[\text{M}+\text{H}]^+$ 274.1339; found 274.1336.



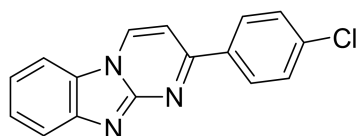
2-(4-Butylphenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (4c) Yellow solid, (141 mg, 47%); mp 201.3–203.5 °C. ^1H NMR (400 MHz, CDCl_3) δ 8.68 (d, $J = 7.2$ Hz, 1H), 8.16 (d, $J = 8.0$ Hz, 2H), 7.97 (d, $J = 8.0$ Hz, 1H), 7.80 (d, $J = 8.0$ Hz, 1H), 7.53 (t, $J = 7.6$ Hz, 1H), 7.39–7.30 (m, 4H), 2.68 (t, $J = 7.6$ Hz, 2H), 1.70–1.58 (m, 2H), 1.45–1.35 (m, 2H), 0.95 (t, $J = 7.2$ Hz, 3H). ^{13}C NMR (101 MHz, CDCl_3) δ 161.6, 151.1, 147.0, 144.9, 134.1, 132.9, 129.1, 127.8, 127.0, 126.2, 121.7, 120.3, 110.4, 103.9, 35.6, 33.4, 29.7, 22.4, 14.0. HRMS (ESI): calcd. for $\text{C}_{20}\text{H}_{20}\text{N}_3^+$ $[\text{M}+\text{H}]^+$ 302.1652; found 302.1650.



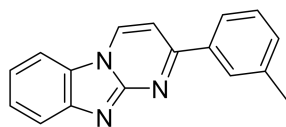
2-(4-Fluorophenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (4d) Yellow solid, (113 mg, 43%); mp199.8–202.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 8.77 (d, *J* = 7.2 Hz, 1H), 8.32-8.27 (m, 2H), 8.00 (d, *J* = 8.4 Hz, 1H), 7.88 (d, *J* = 8.4 Hz, 1H), 7.58 (t, *J* = 7.6 Hz, 1H), 7.42 (t, *J* = 7.2 Hz, 1H), 7.38 (d, *J* = 7.2 Hz, 1H), 7.23 (t, *J* = 8.4 Hz, 2H). ¹³C NMR (101 MHz, DMSO) δ 163.46, 160.49, 150.69, 144.64, 136.57, 133.41, 130.68, 130.59, 127.47, 126.56, 121.88, 119.42, 116.61, 116.39, 112.87, 104.24. HRMS (ESI): calcd. for C₁₆H₁₁FN₃⁺ [M+H]⁺ 264.0932; found 264.0936.



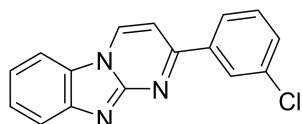
2-(4-Ethoxyphenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (4e) Yellow solid, (101 mg, 35%); mp202.1–205.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 8.68 (d, *J* = 7.2 Hz, 1H), 8.25 (d, *J* = 8.8 Hz, 2H), 7.97 (d, *J* = 8.4 Hz, 1H), 7.83 (d, *J* = 8.0 Hz, 1H), 7.55 (t, *J* = 7.6 Hz, 1H), 7.39 (t, *J* = 7.2 Hz, 1H), 7.35 (d, *J* = 7.2 Hz, 1H), 7.02 (d, *J* = 8.8 Hz, 2H), 4.13 (q, *J* = 7.2 Hz, 2H), 1.47 (t, *J* = 7.2 Hz, 4H). ¹³C NMR (101 MHz, CDCl₃) δ 161.9, 151.2, 144.9, 132.7, 129.7, 129.6, 128.9, 127.1, 126.2, 121.6, 120.3, 114.8, 110.2, 103.6, 63.7, 14.8. HRMS (ESI): calcd. for C₁₈H₁₆N₃O⁺ [M+H]⁺ 290.1288; found 290.1284.



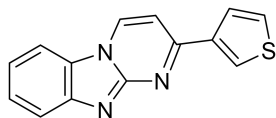
2-(4-Chlorophenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (4f)⁵ Yellow solid, (103 mg, 37%); mp197.2–200.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 8.77 (d, *J* = 7.2 Hz, 1H), 8.23 (d, *J* = 8.8 Hz, 1H), 8.01 (d, *J* = 8.4 Hz, 1H), 7.89 (d, *J* = 8.0 Hz, 1H), 7.59 (t, *J* = 7.6 Hz, 1H), 7.52 (d, *J* = 8.4 Hz, 2H), 7.43 (t, *J* = 7.6 Hz, 1H), 7.39 (d, *J* = 7.2 Hz, 1H). ¹³C NMR (101 MHz, CDCl₃) δ 160.4, 144.6, 138.1, 135.2, 133.4, 131.4, 130.3, 127.8, 126.9, 125.8, 122.4, 120.3, 110.6, 104.0. ESI-MS: m/z [M+H]⁺ 280.



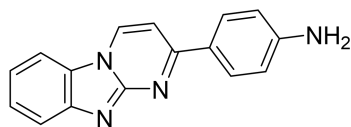
2-(3-Tolyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (4g) Yellow solid, (106 mg, 41%); mp200.2–203.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 8.73 (d, *J* = 7.2 Hz, 1H), 8.14 (s, 1H), 8.02 (d, *J* = 7.6 Hz, 1H), 7.99 (d, *J* = 8.4 Hz, 1H), 7.85 (d, *J* = 8.4 Hz, 1H), 7.56 (t, *J* = 7.6 Hz, 1H), 7.44-7.34 (m, 4H), 2.47 (s, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 161.7, 151.0, 144.9, 138.7, 136.5, 132.9, 132.2, 128.8, 128.5, 126.9, 126.3, 124.9, 121.8, 120.4, 110.4, 104.1, 21.5. HRMS (ESI): calcd. for C₁₇H₁₄N₃⁺ [M+H]⁺ 260.1182; found 260.1182.



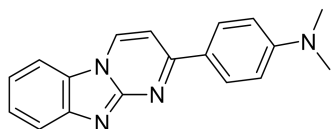
2-(3-Chlorophenyl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (4h)⁵ Yellow solid, (103 mg, 37%); mp 197.3–200.3 °C. ¹H NMR (400 MHz, CDCl₃) δ 8.77 (d, *J* = 7.2 Hz, 1H), 8.32–8.25 (m, 2H), 8.00 (d, *J* = 8.0 Hz, 1H), 7.87 (d, *J* = 8.0 Hz, 1H), 7.61–7.51 (m, 4H), 7.43–7.38 (m, 2H). ¹³C NMR (101 MHz, CDCl₃) δ 161.6, 151.0, 145.0, 136.6, 133.1, 131.4, 129.0, 127.8, 127.0, 126.4, 121.90, 120.5, 110.4, 104.0. ESI-MS: *m/z* [M+H]⁺ 280.



2-(Thiophen-3-yl)benzo[4,5]imidazo[1,2-*a*]pyrimidine (4i) Yellow solid, (100 mg, 40%); mp 196.5–198.4 °C. ¹H NMR (400 MHz, CDCl₃) δ 8.72 (d, *J* = 7.2 Hz, 1H), 8.23 (d, *J* = 1.6 Hz, 1H), 7.99 (d, *J* = 8.0 Hz, 1H), 7.95 (d, *J* = 5.2 Hz, 1H), 7.86 (d, *J* = 8.0 Hz, 1H), 7.57 (t, *J* = 7.6 Hz, 1H), 7.47 (dd, *J* = 5.2, 3.2 Hz, 1H), 7.40 (t, *J* = 7.6 Hz, 1H), 7.28 (d, *J* = 7.2 Hz, 1H). ¹³C NMR (101 MHz, DMSO) δ 157.72, 150.81, 144.57, 140.84, 136.30, 129.90, 128.45, 127.71, 127.07, 126.37, 121.70, 119.34, 112.77, 105.07. HRMS (ESI): calcd. for C₁₄H₁₀N₃S⁺ [M+H]⁺ 252.0590; found 252.0595.



4-(Benzo[4,5]imidazo[1,2-*a*]pyrimidin-2-yl)aniline (4j)⁶ Yellow solid, (176 mg, 72%); mp 201.2–205.4 °C. ¹H NMR (400 MHz, DMSO) δ 9.30 (d, *J* = 7.2 Hz, 1H), 8.19 (d, *J* = 8.0 Hz, 1H), 8.08 (d, *J* = 8.4 Hz, 2H), 7.76 (d, *J* = 8.0 Hz, 1H), 7.61 (d, *J* = 7.2 Hz, 1H), 7.48 (t, *J* = 7.6 Hz, 1H), 7.34 (t, *J* = 7.6 Hz, 1H), 6.71 (d, *J* = 8.4 Hz, 2H), 5.97 (s, 2H). ¹³C NMR (101 MHz, DMSO) δ 161.8, 153.0, 151.4, 144.4, 135.1, 129.9, 127.6, 126.1, 123.5, 121.3, 118.9, 114.1, 112.2, 103.7. ESI-MS: *m/z* [M+H]⁺ 261.

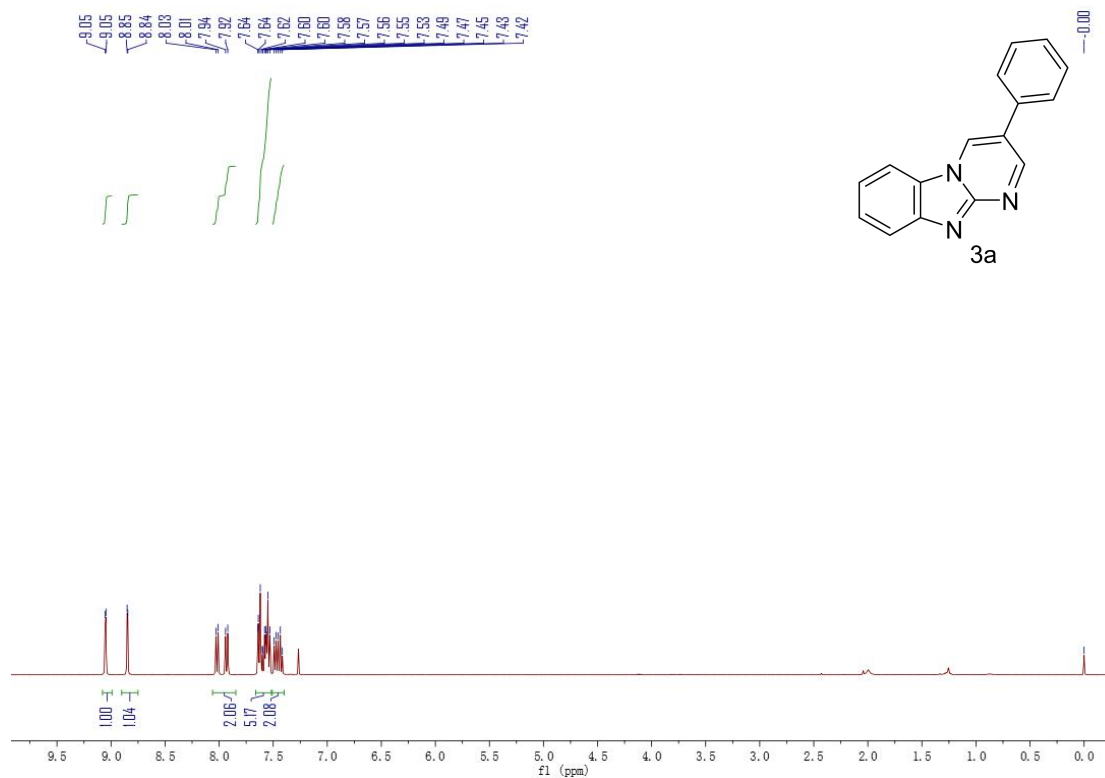


4-Benzo[4,5]imidazo[1,2-*a*]pyrimidin-2-yl)-N,N-dimethylaniline (4k)⁶ Yellow solid, (190 mg, 66%); mp 200.3–204.1 °C. ¹H NMR (400 MHz, CDCl₃) δ 8.58 (d, *J* = 7.2 Hz, 1H), 8.21 (d, *J* = 8.8 Hz, 2H), 7.93 (d, *J* = 8.0 Hz, 1H), 7.78 (d, *J* = 8.0 Hz, 1H), 7.51 (t, *J* = 7.6 Hz, 1H), 7.35 (t, *J* = 8.0 Hz, 1H), 7.31 (t, *J* = 7.2 Hz, 1H), 6.78 (d, *J* = 8.8 Hz, 2H), 3.08 (s, 6H). ¹³C NMR (101 MHz, CDCl₃) δ 161.4, 152.6, 151.7, 145.0, 132.1, 129.4, 127.3, 125.8, 123.8, 121.1, 120.0, 111.7, 110.0, 103.4, 40.2. ESI-MS: *m/z* [M+H]⁺ 289.

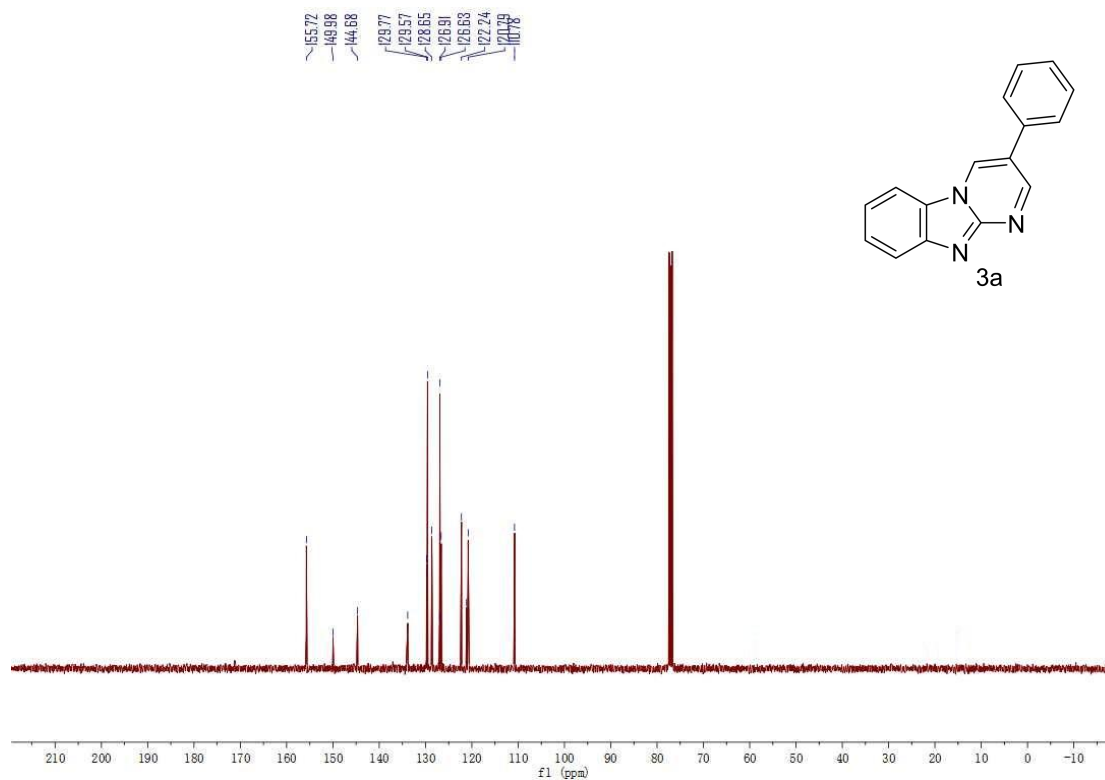
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2. A. M. Mehranpour and M. Zahiri, *Tetrahedron Lett.*, 2014, **55**, 3969-3971.
3. J. Wu, H. Luo, T. Wang, H. Sun, Q. Zhang and Y. Chai, *Tetrahedron*, 2019, **75**, 1052-1063.
4. Y. Gu, F. Wu and J. Yang, *Adv. Synth. Catal.*, 2018, **360**, 2727-2741.
5. S.-S. Tseng, J. W. Epstein, H. J. Brabander and G. Francisco, *J. Heterocyclic Chem.*, 1987, **24**, 837-843.
6. A. K. Szardenings, W. Zhang, H. C. Kolb, D. K. Cashion, G. Chen, D. Kasi, C. Liu, A. Sinha, E. Wang, C. Yu, U. B. Gangadharmath, J. C. Walsh, Siemens Medical Solutions USA, Inc., USA; Eli Lilly and Company. **2011**, pp. 165 pp., Cont.-in-part of U.S. Ser. No. 661777.

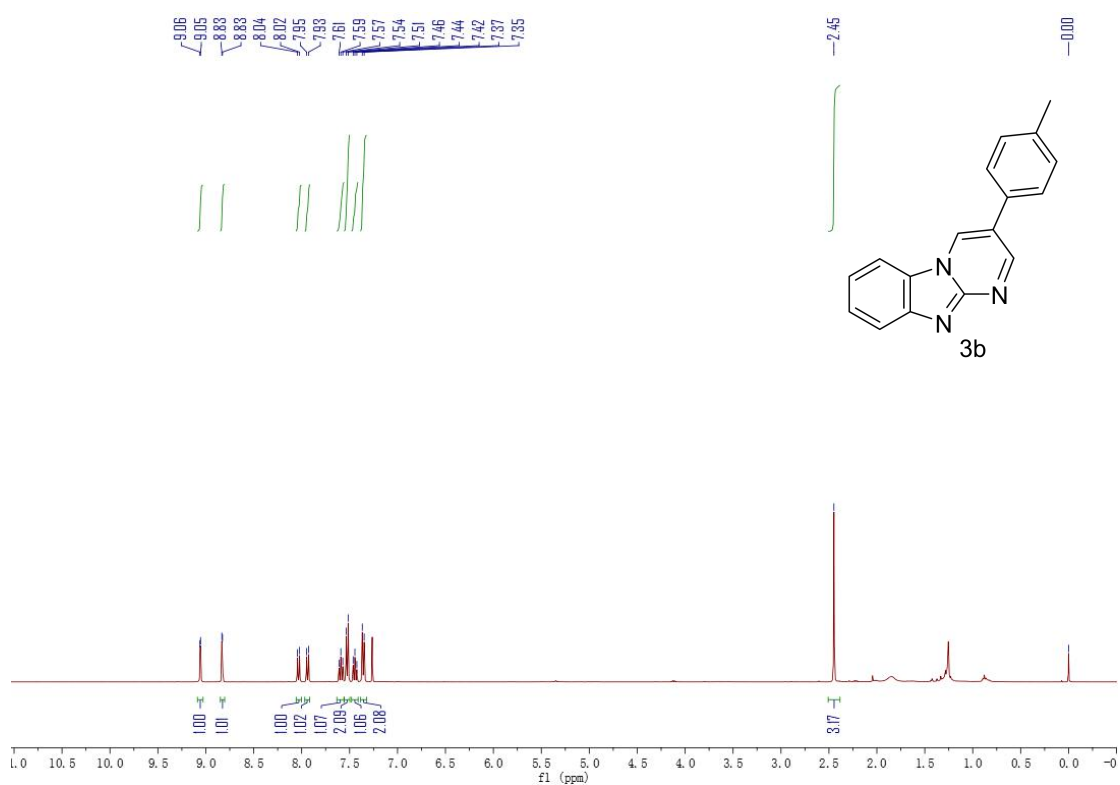
¹H NMR of Compound 3a (400 MHz, CDCl₃)



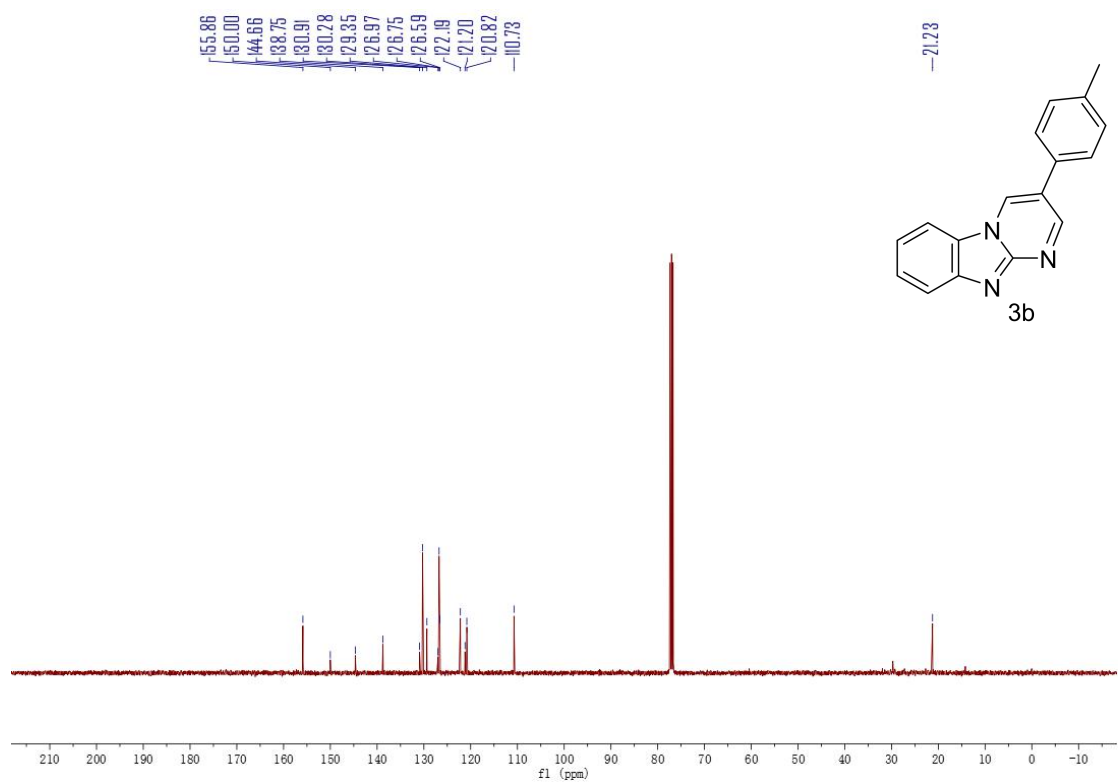
¹³C NMR of Compound 3a (400 MHz, CDCl₃)



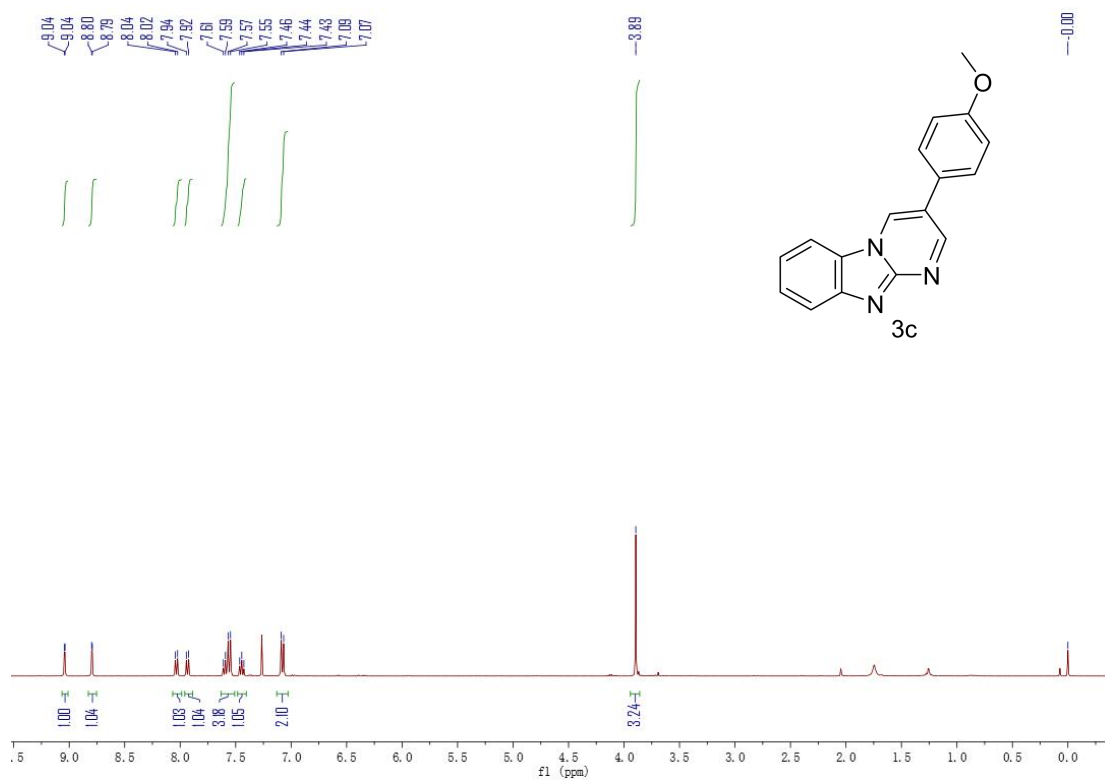
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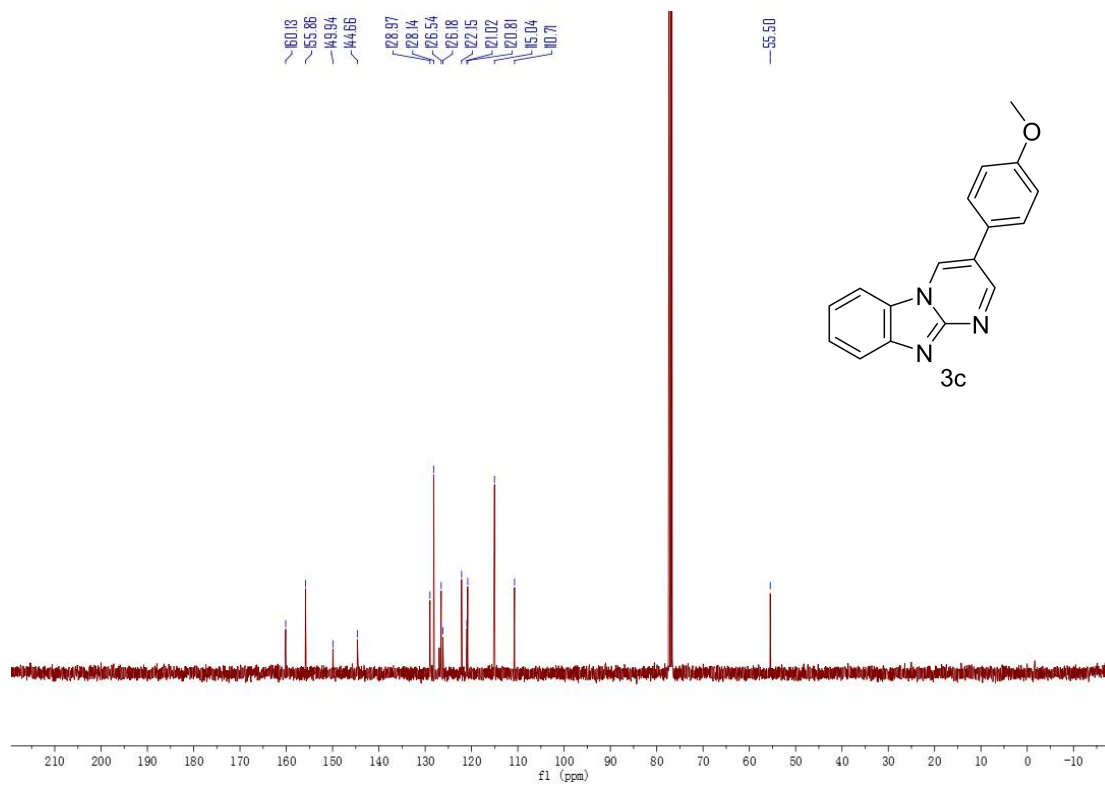
¹³C NMR of Compound 3b (400 MHz, CDCl₃)



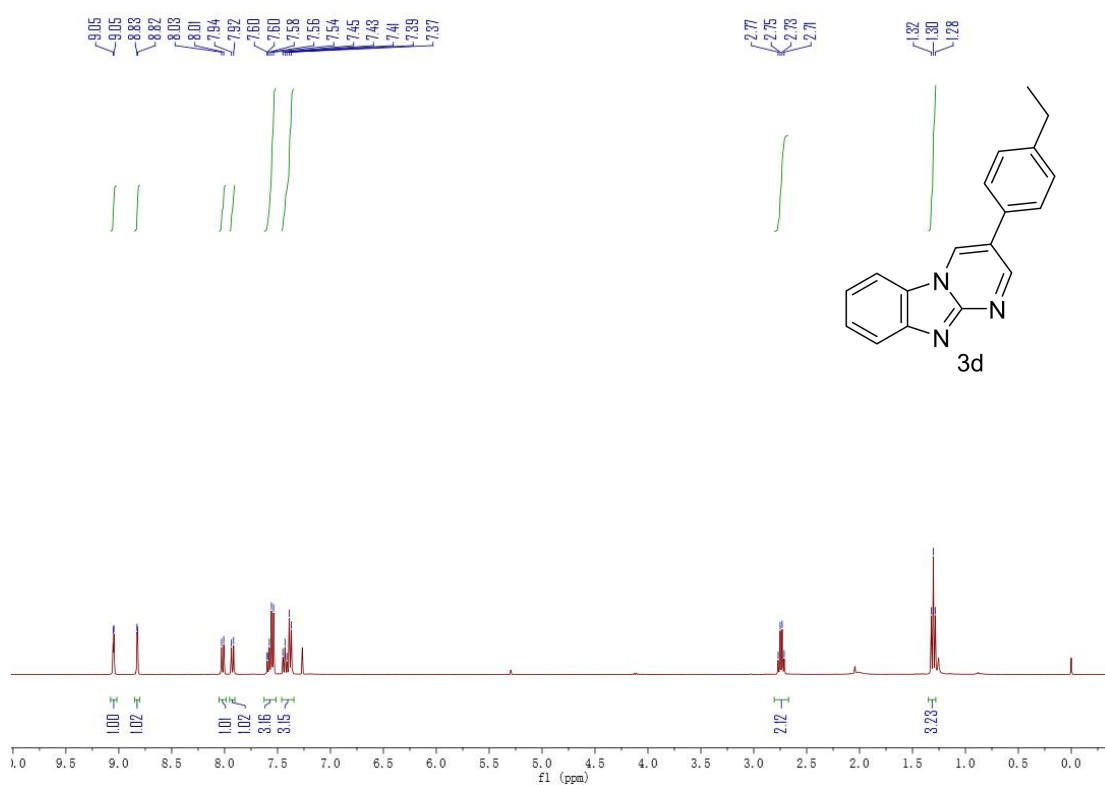
¹H NMR of Compound 3c (400 MHz, CDCl₃)



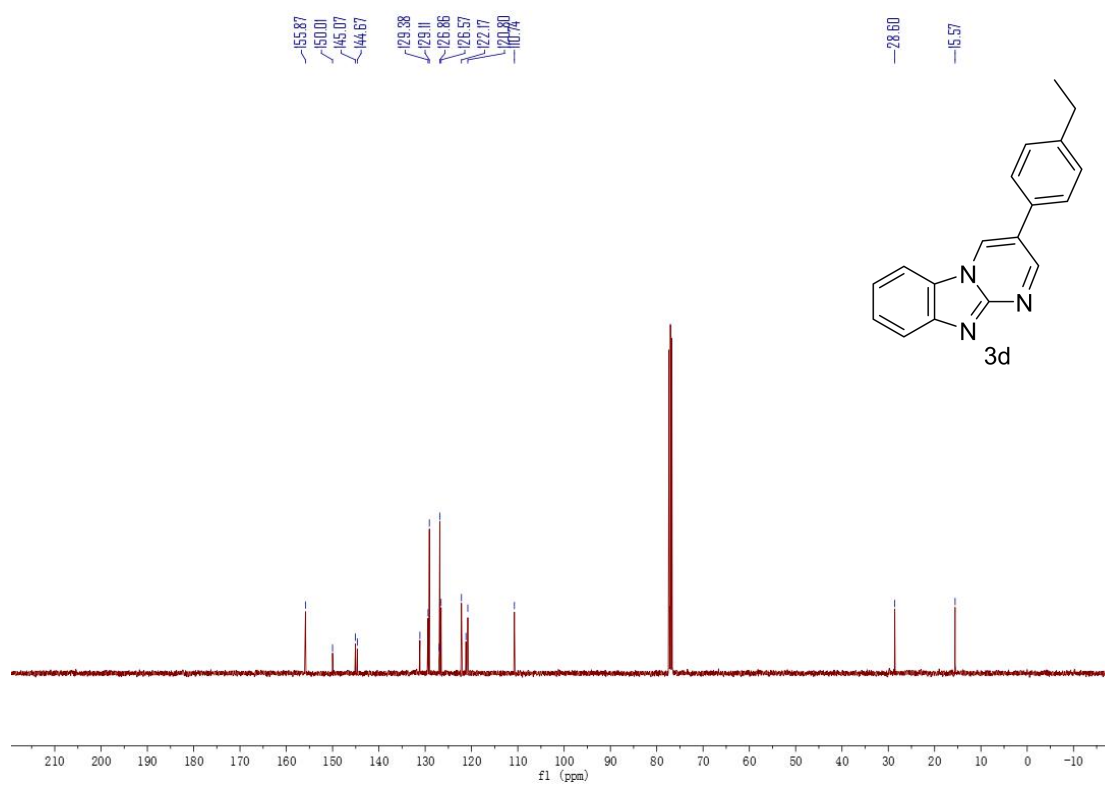
¹³C NMR of Compound 3c (400 MHz, CDCl₃)



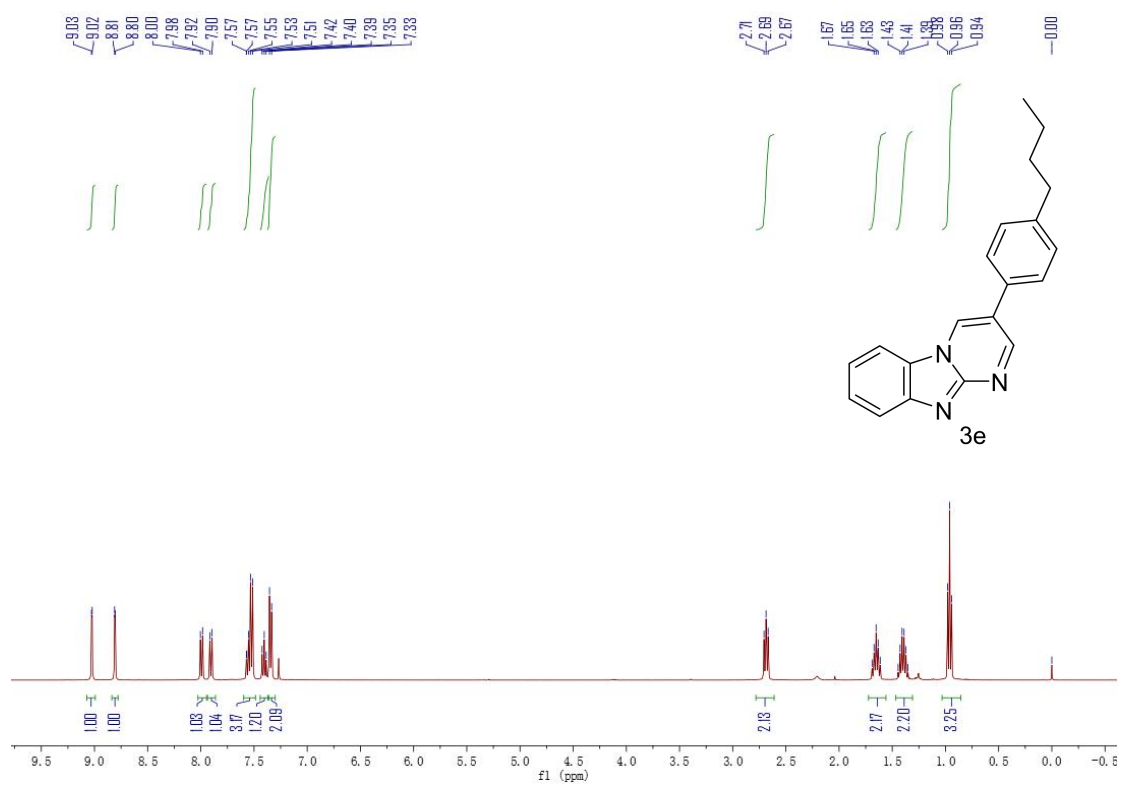
¹H NMR of Compound 3d (400 MHz, CDCl₃)



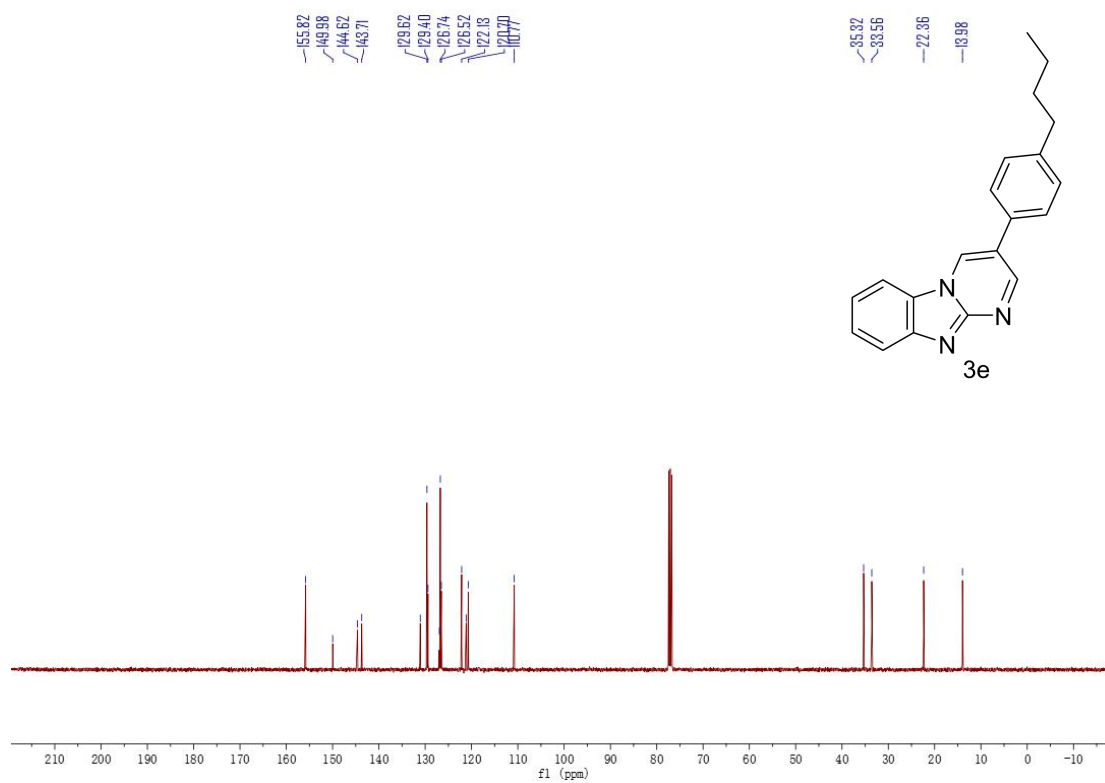
¹³C NMR of Compound 3d (400 MHz, CDCl₃)



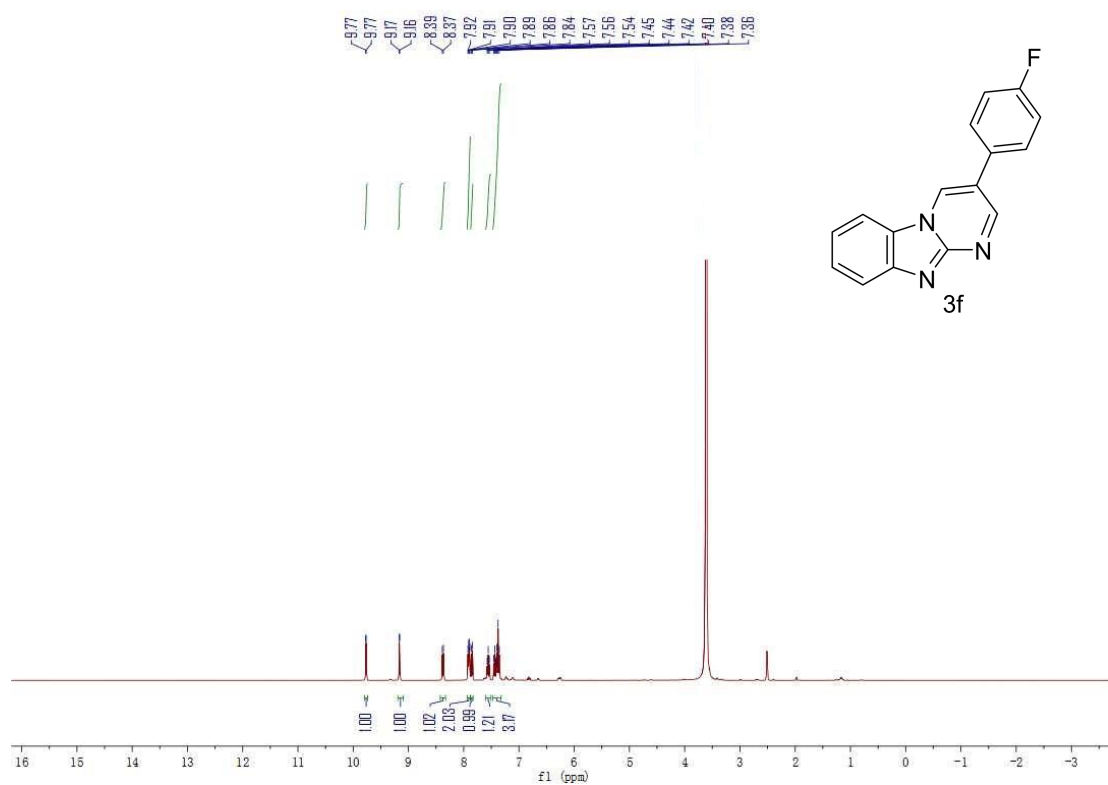
¹H NMR of Compound 3e (400 MHz, CDCl₃)



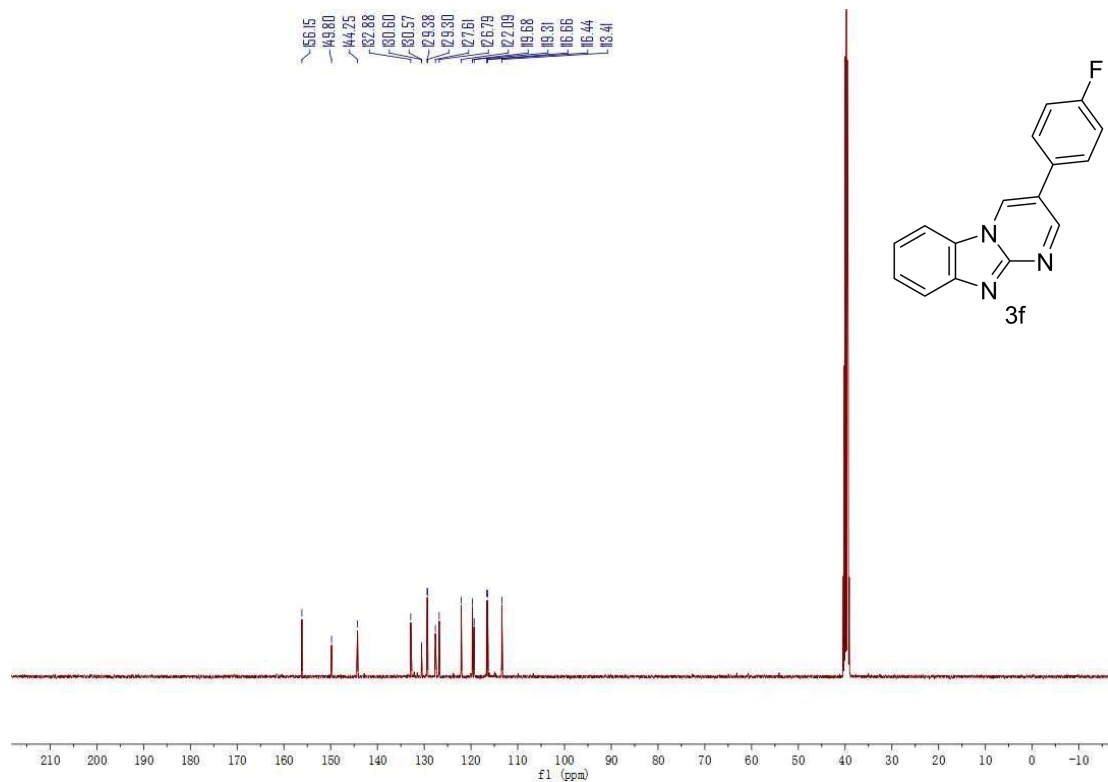
¹³C NMR of Compound 3e (400 MHz, CDCl₃)



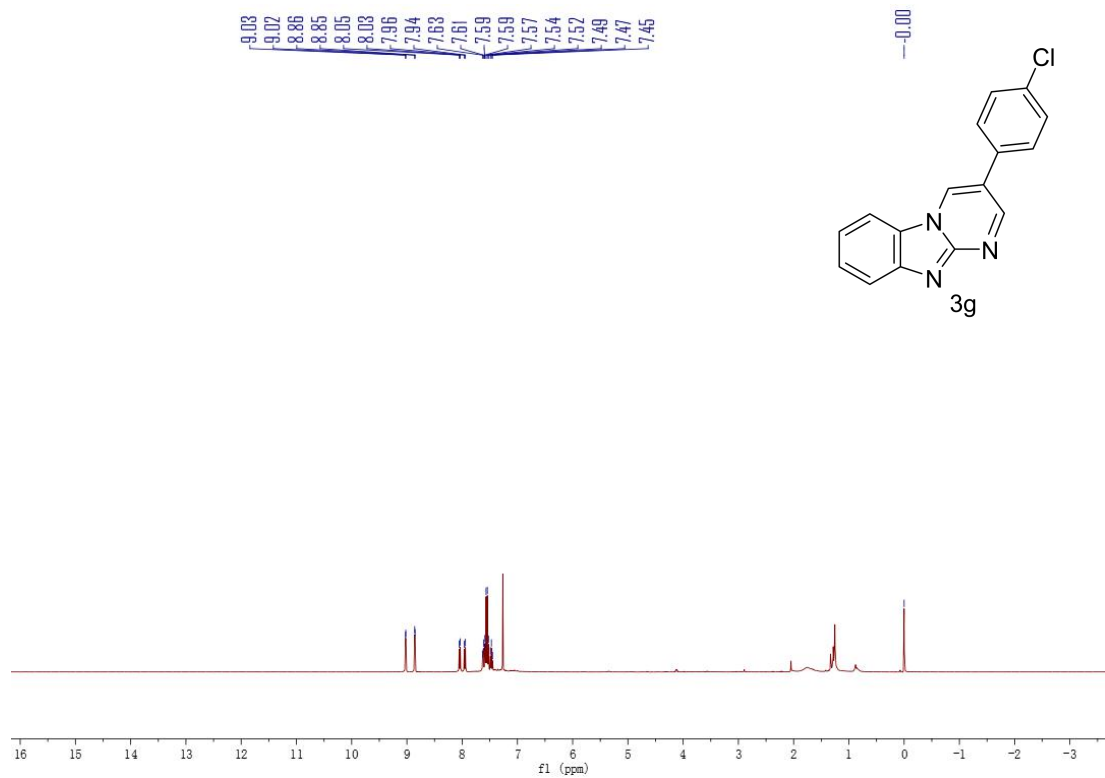
¹H NMR of Compound 3f (400 MHz, DMSO-d₆)



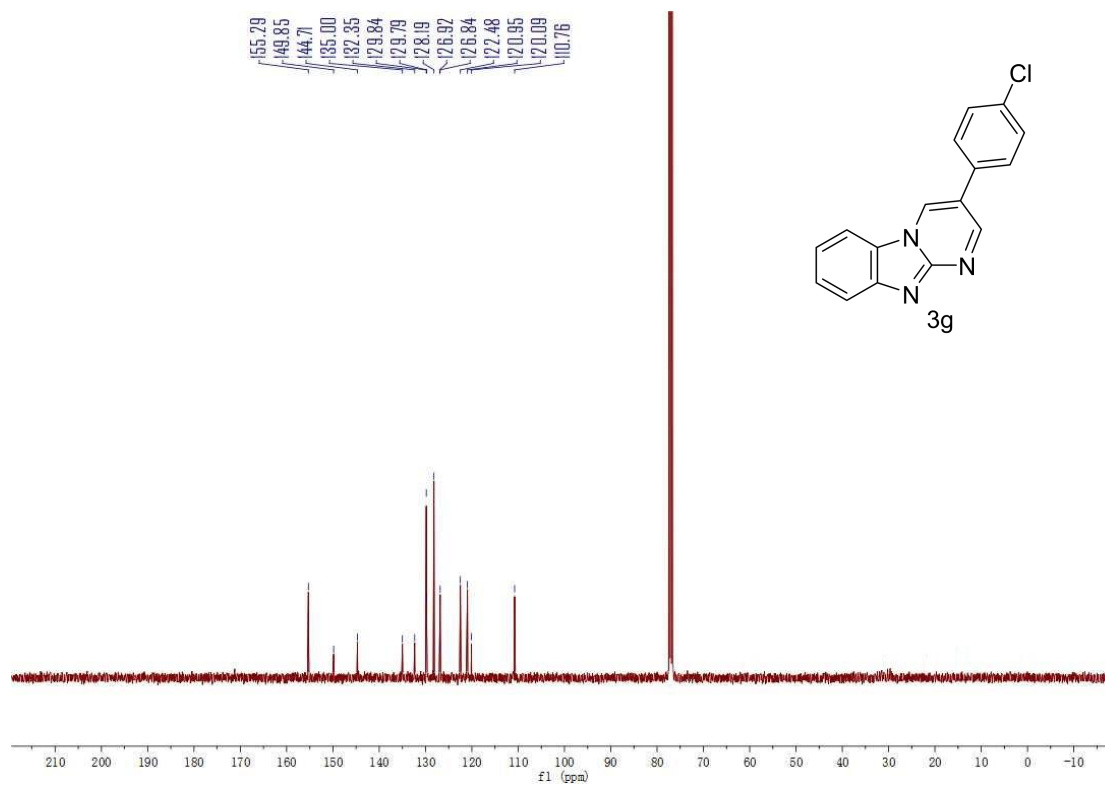
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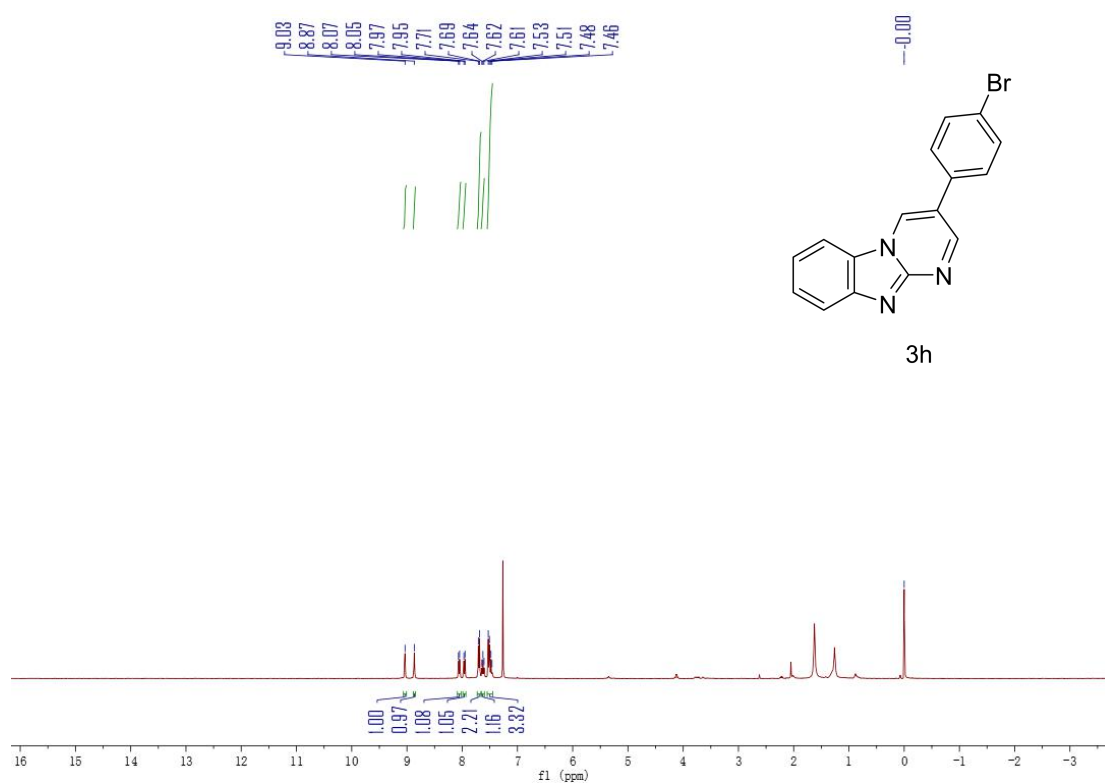
¹H NMR of Compound 3g (400 MHz, CDCl₃)



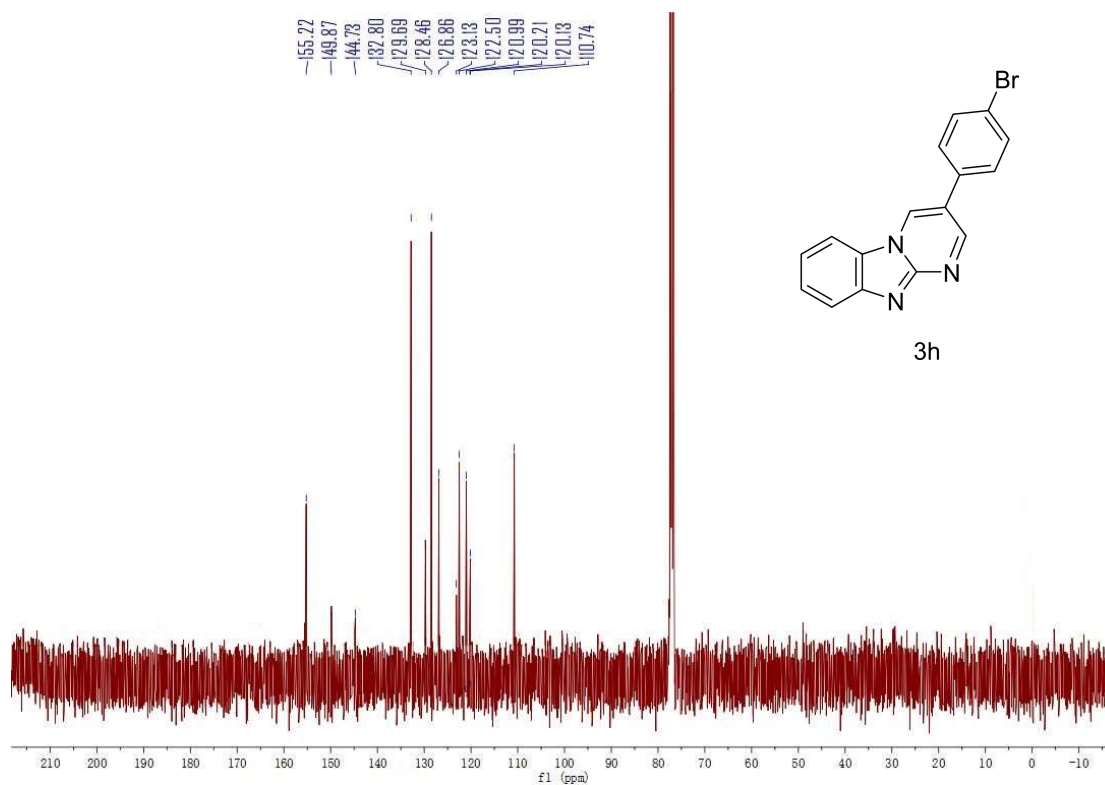
¹³C NMR of Compound 3g (400 MHz, CDCl₃)



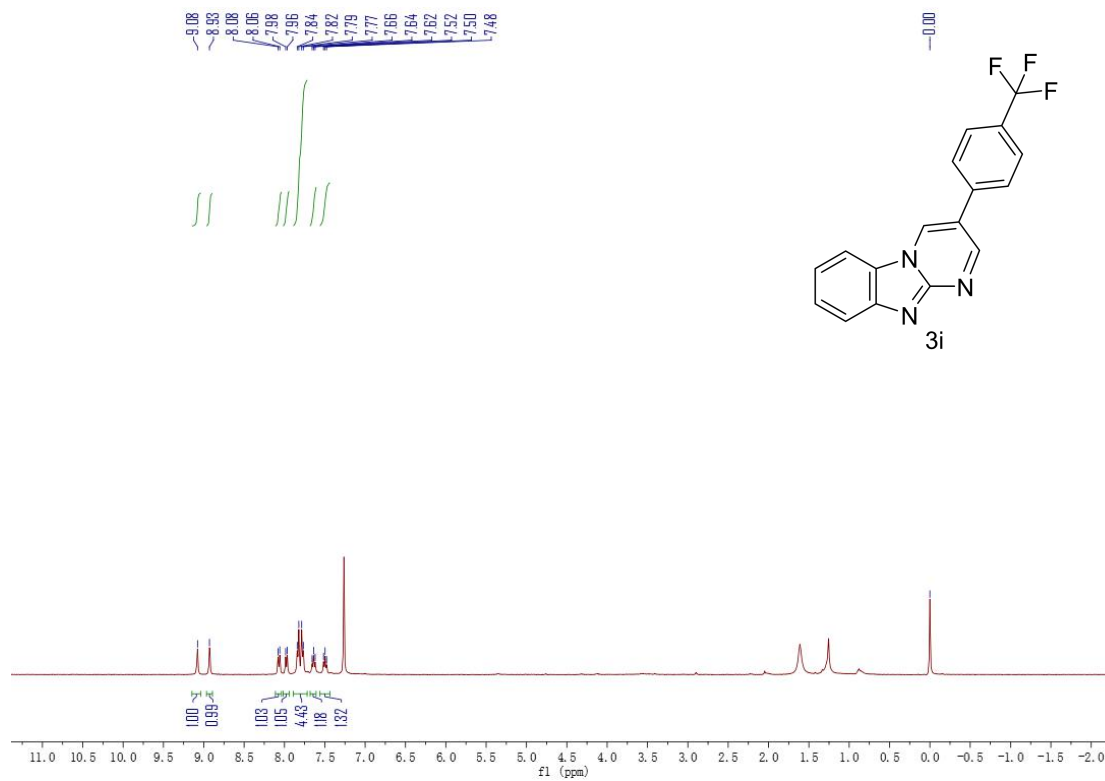
¹H NMR of Compound 3h (400 MHz, CDCl₃)



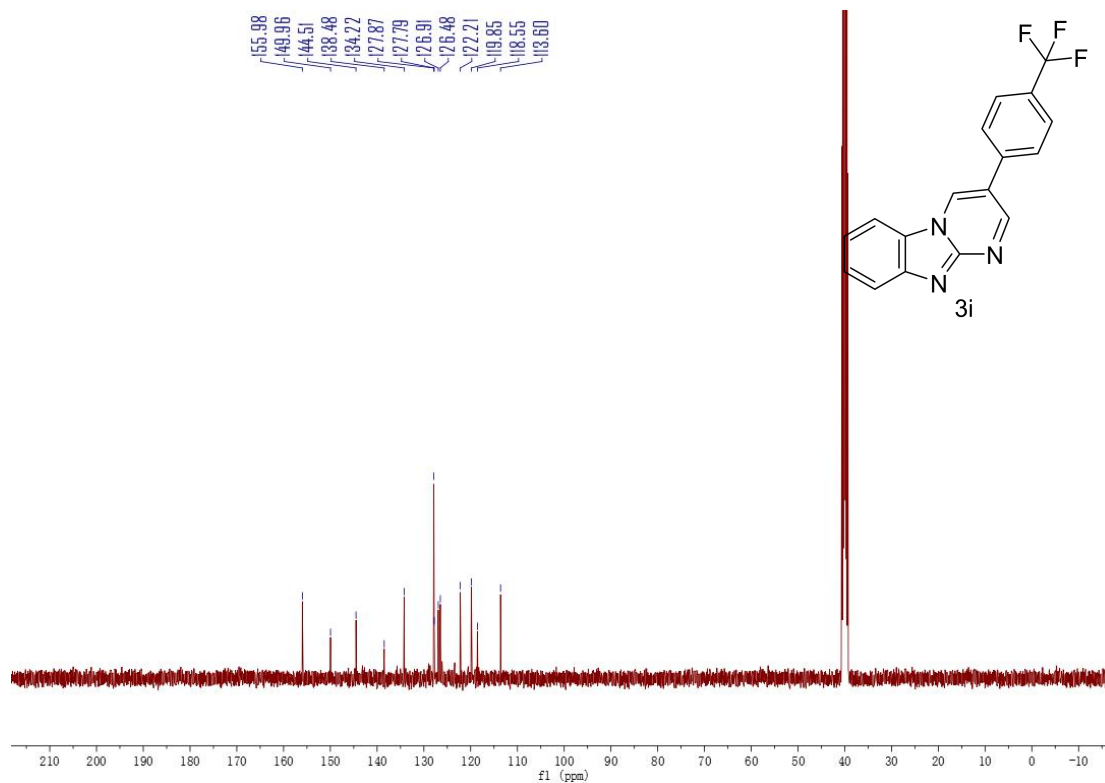
¹³C NMR of Compound 3h (400 MHz, CDCl₃)



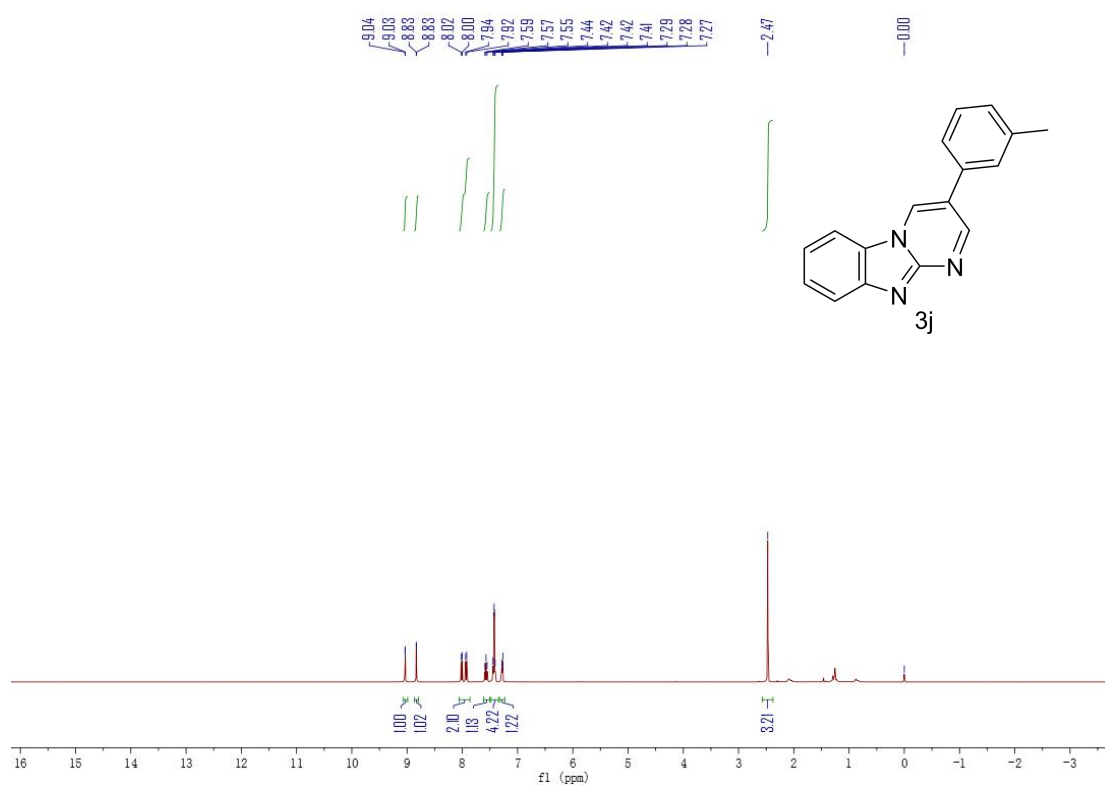
¹H NMR of Compound 3i (400 MHz, CDCl₃)



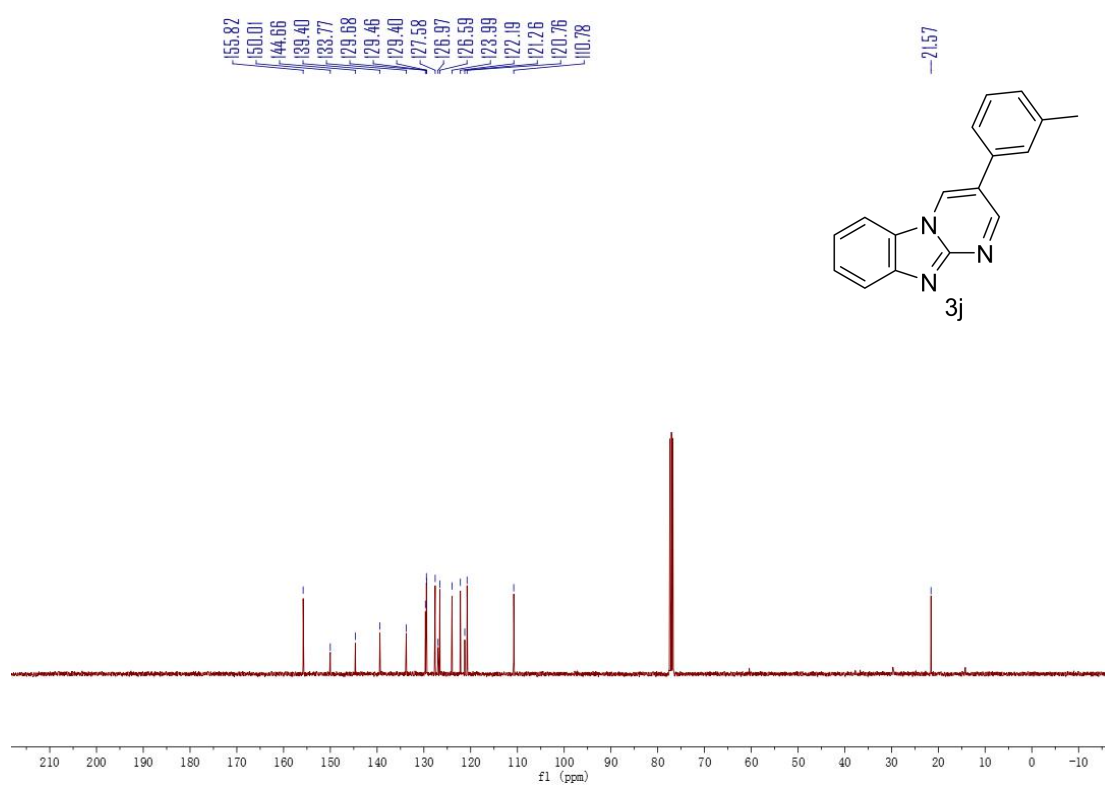
¹³C NMR of Compound 3i (400 MHz, DMSO-*d*₆)



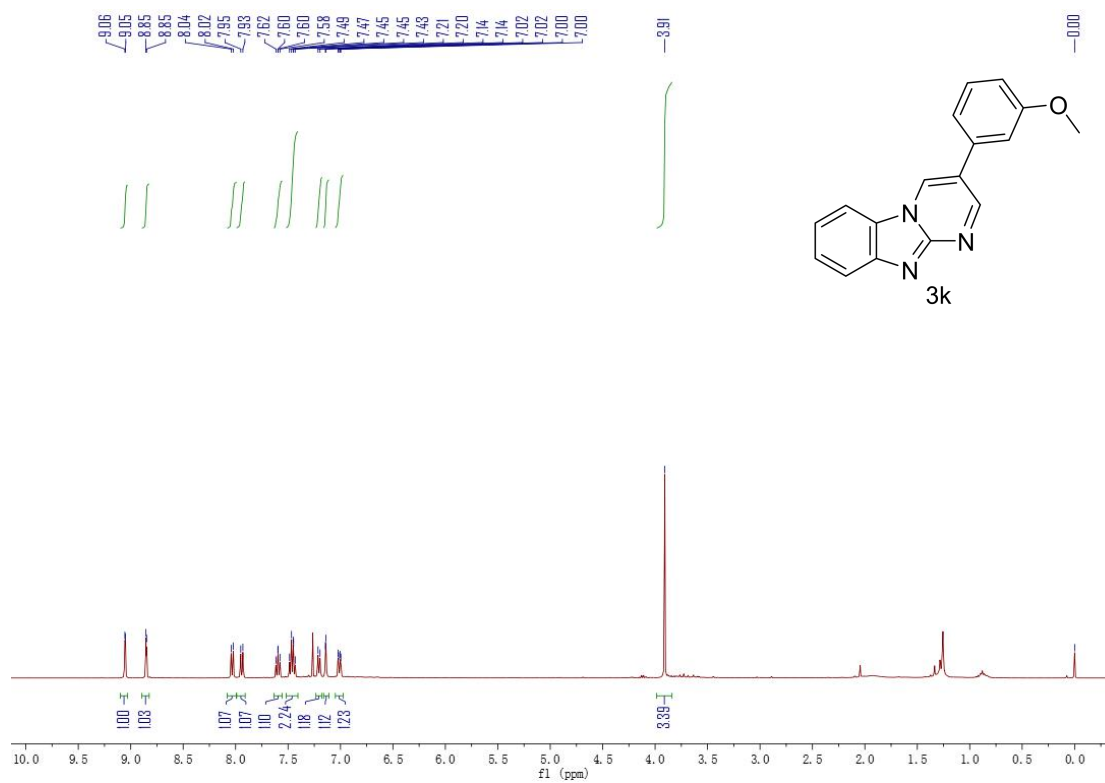
¹H NMR of Compound 3j (400 MHz, CDCl₃)



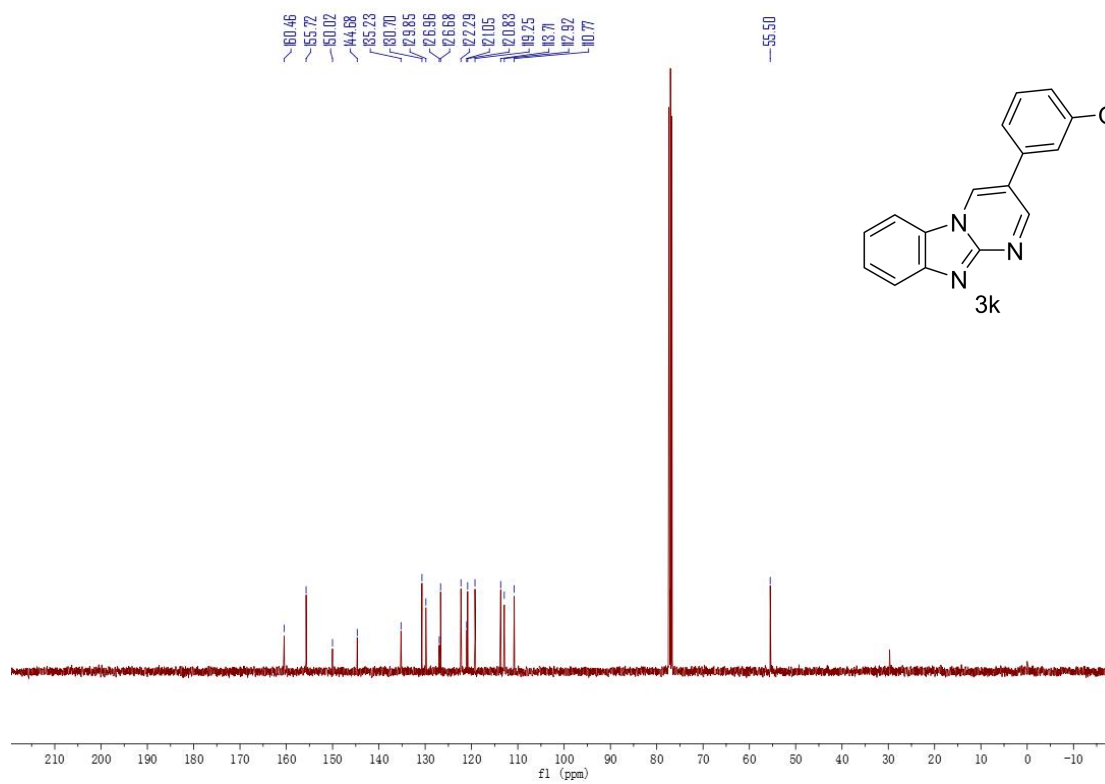
¹³C NMR of Compound 3j (400 MHz, CDCl₃)



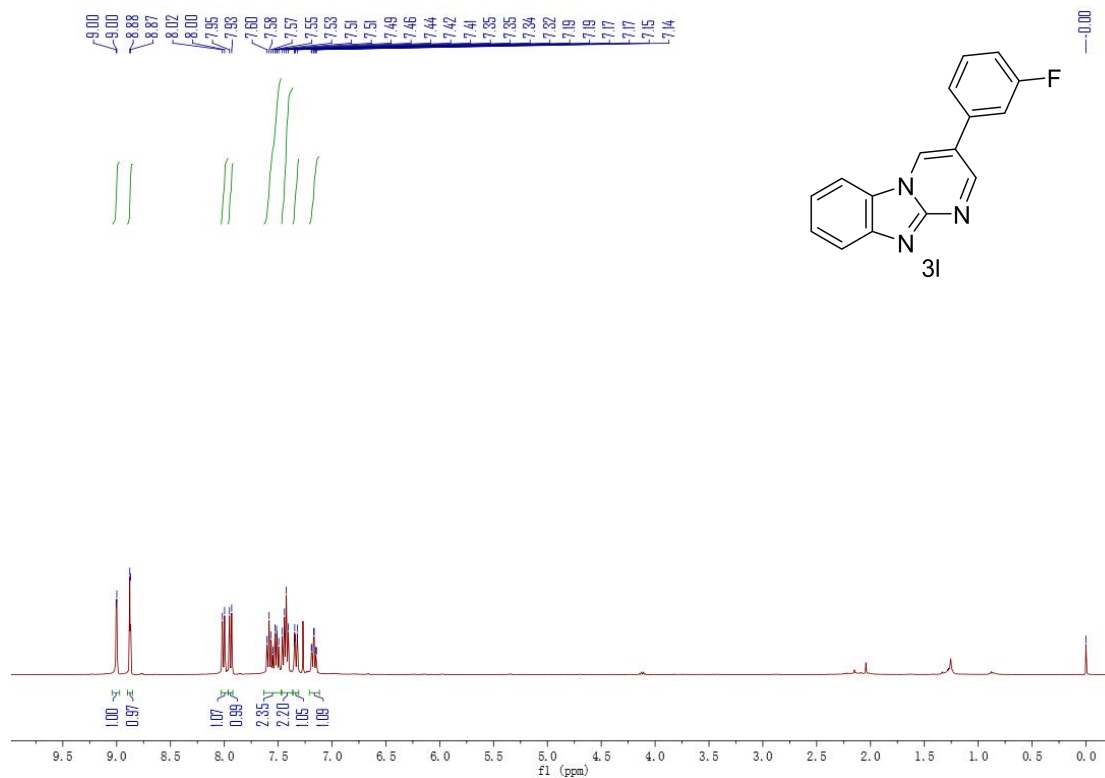
¹H NMR of Compound 3k (400 MHz, CDCl₃)



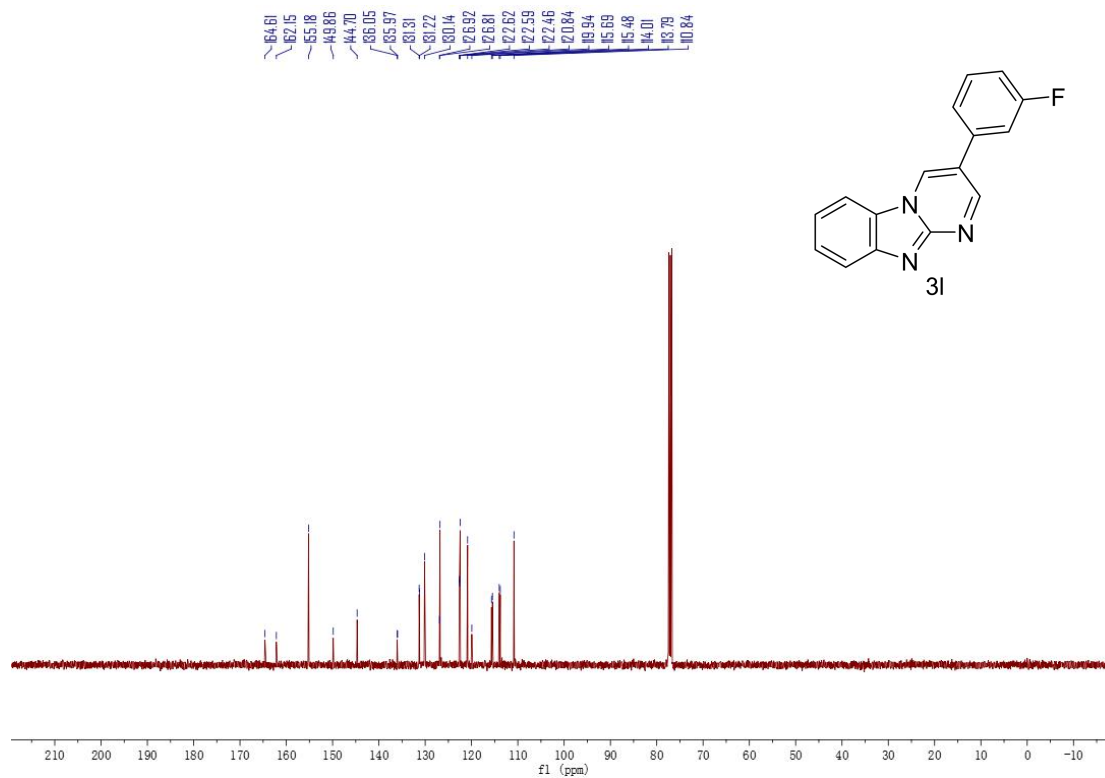
¹³C NMR of Compound 3k (400 MHz, CDCl₃)



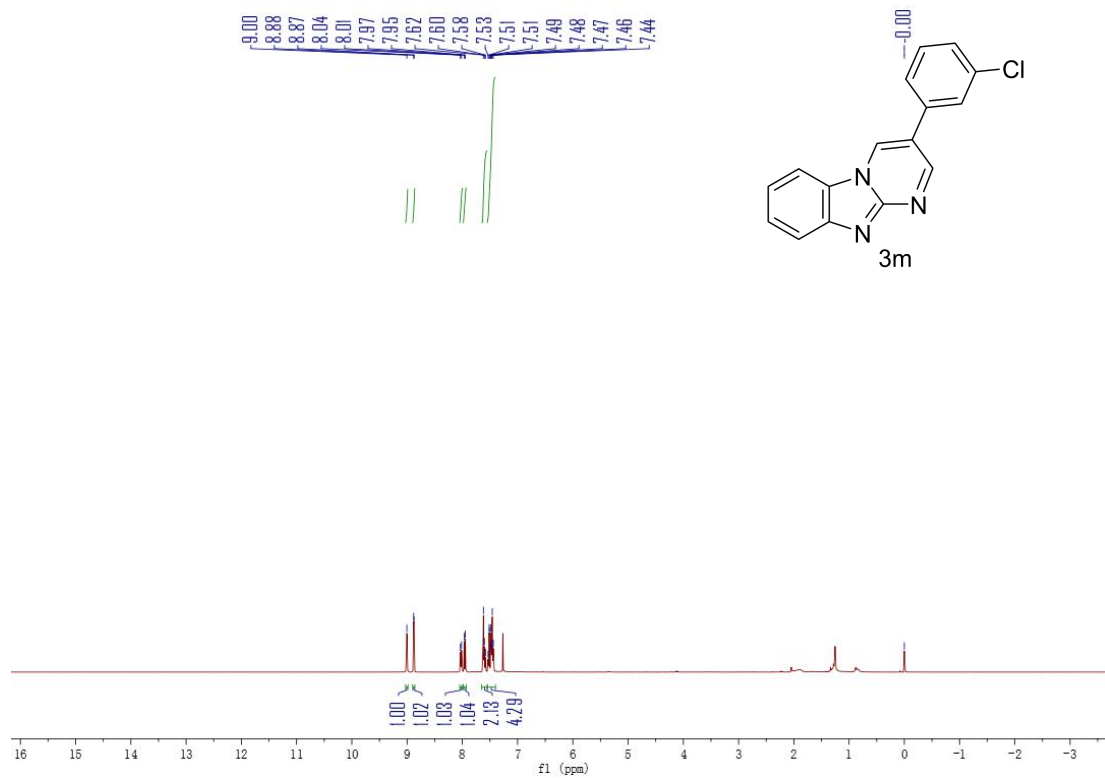
¹H NMR of Compound 31 (400 MHz, CDCl₃)



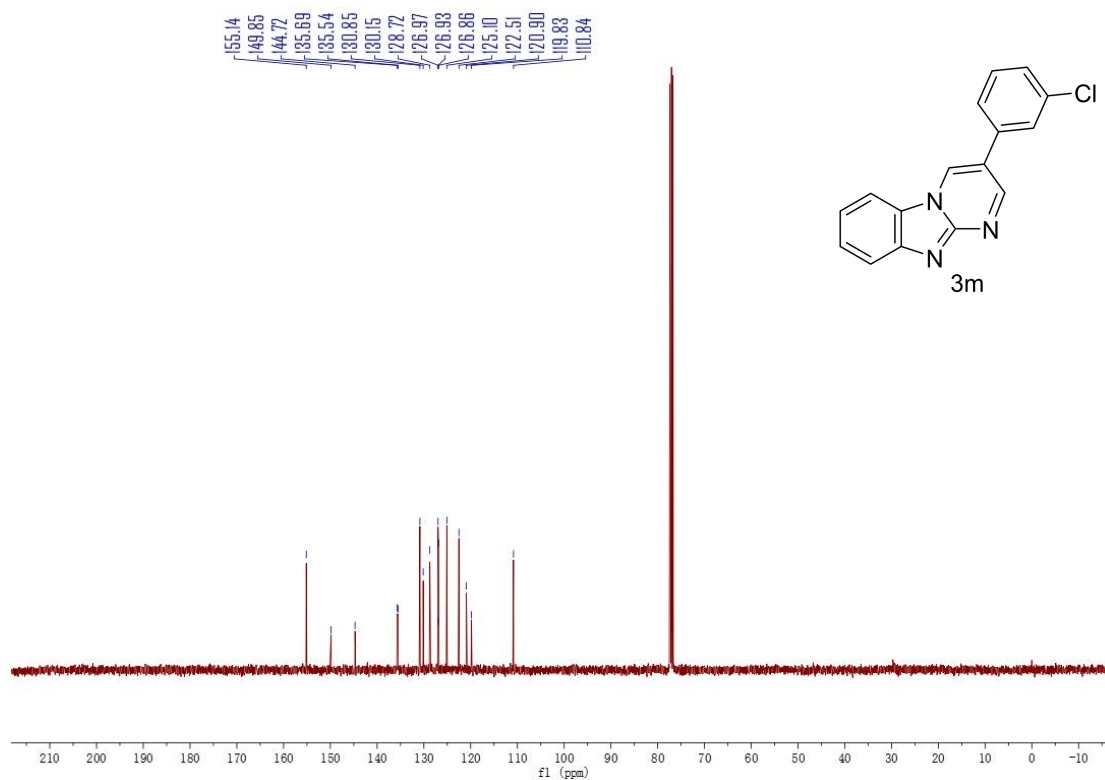
¹³C NMR of Compound 31 (400 MHz, CDCl₃)



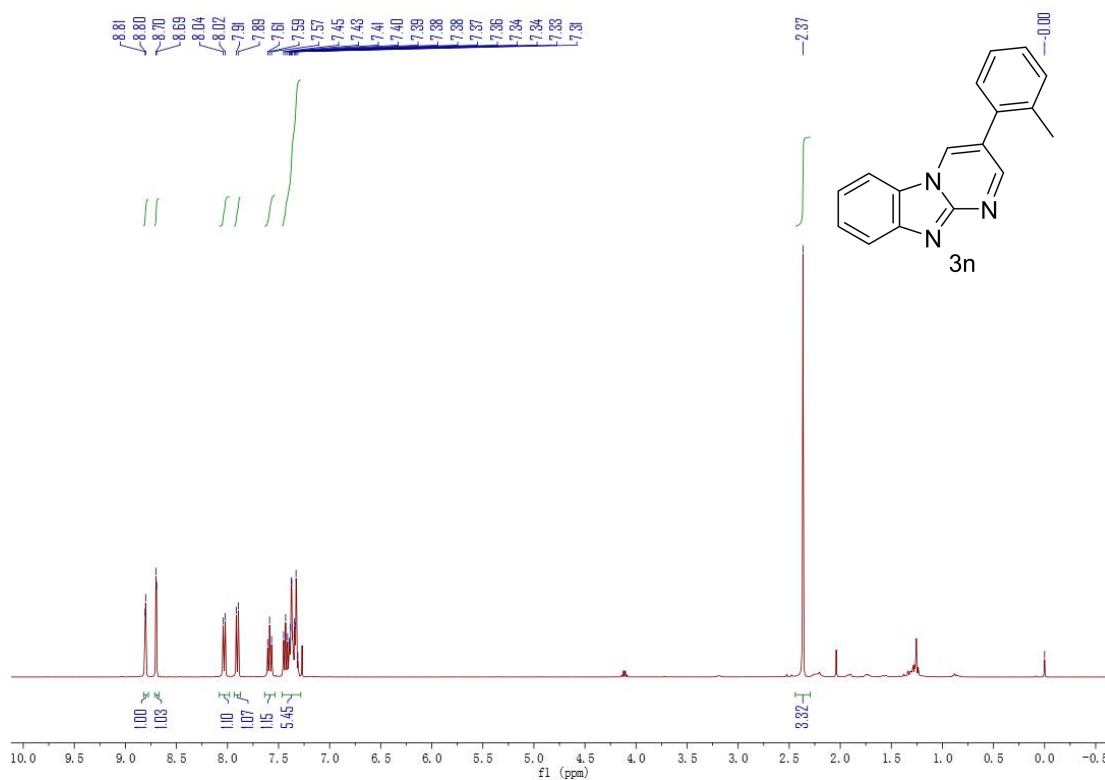
¹H NMR of Compound **3m** (400 MHz, CDCl₃)



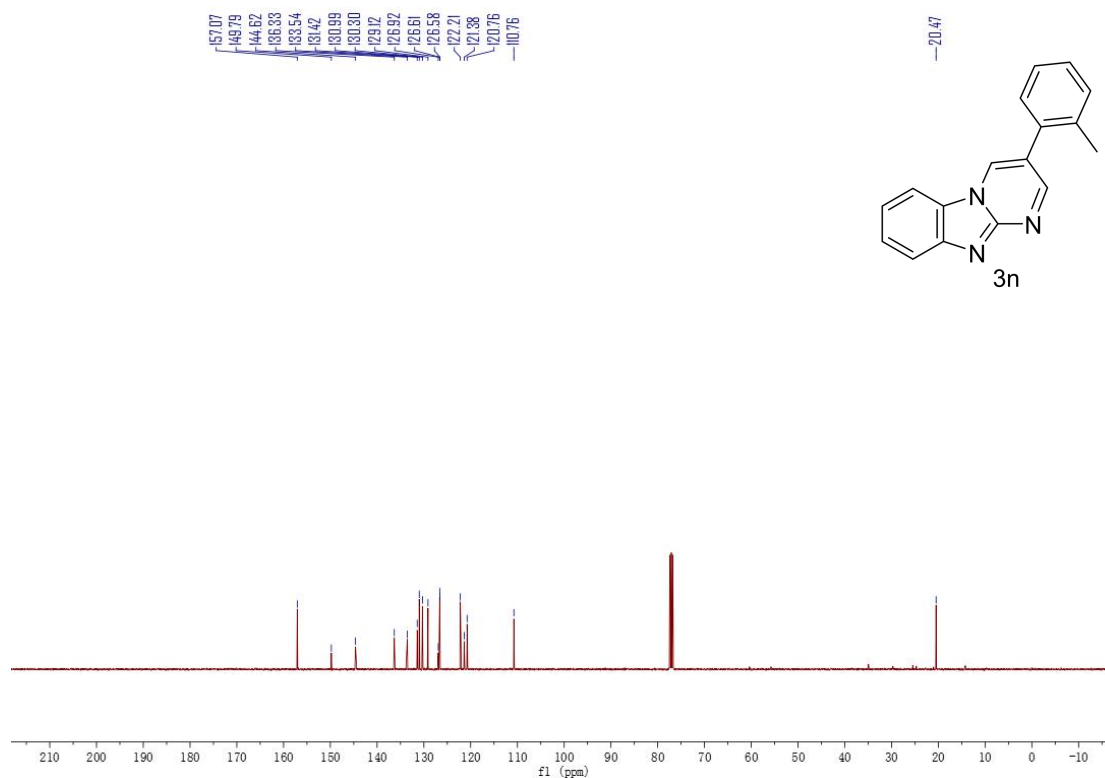
¹³C NMR of Compound **3m** (400 MHz, CDCl₃)



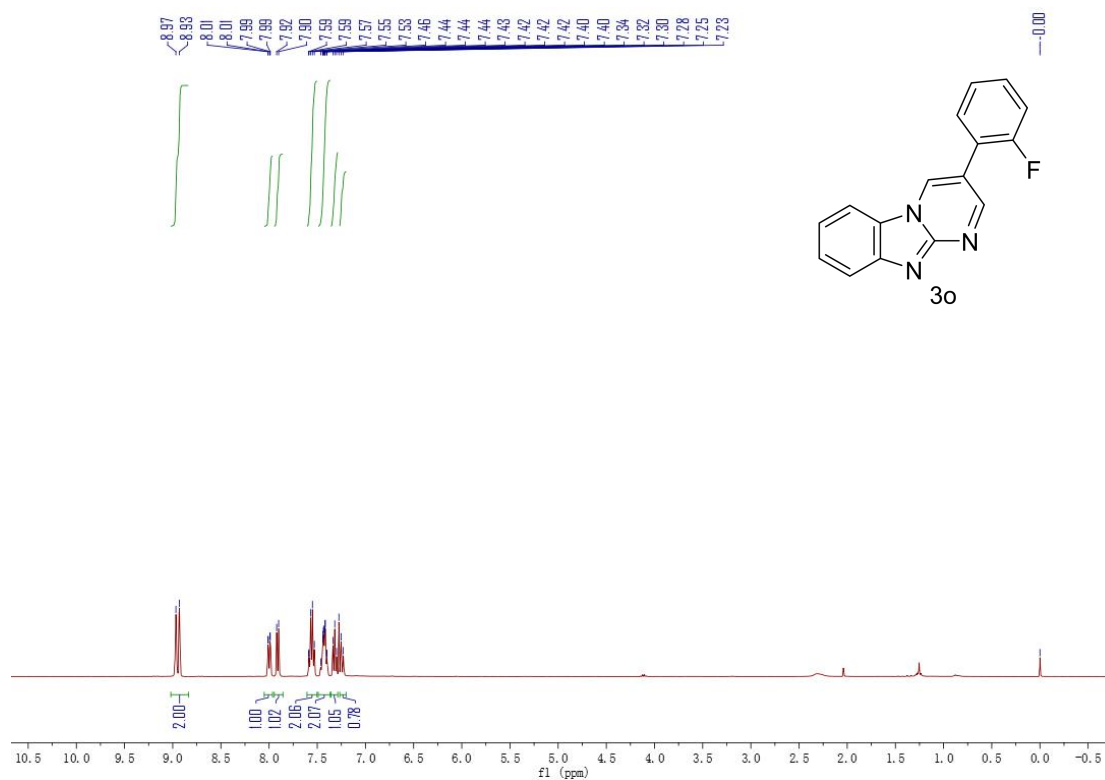
¹H NMR of Compound 3n (400 MHz, CDCl₃)



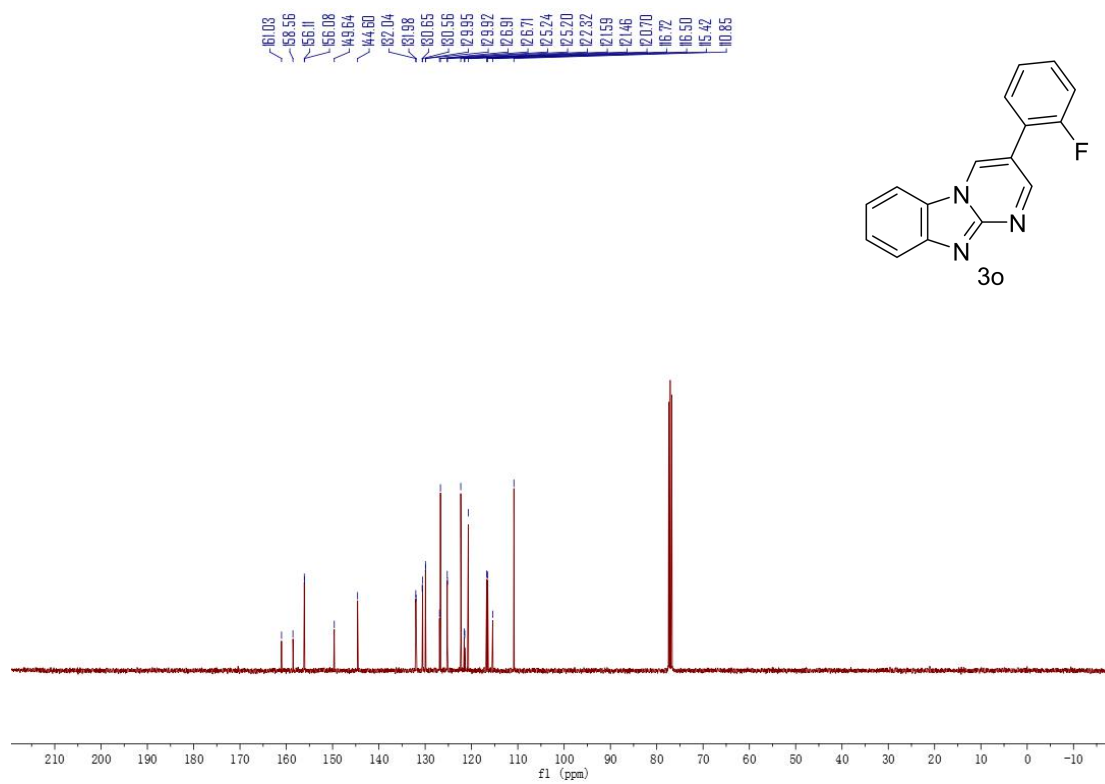
¹³C NMR of Compound 3n (400 MHz, CDCl₃)



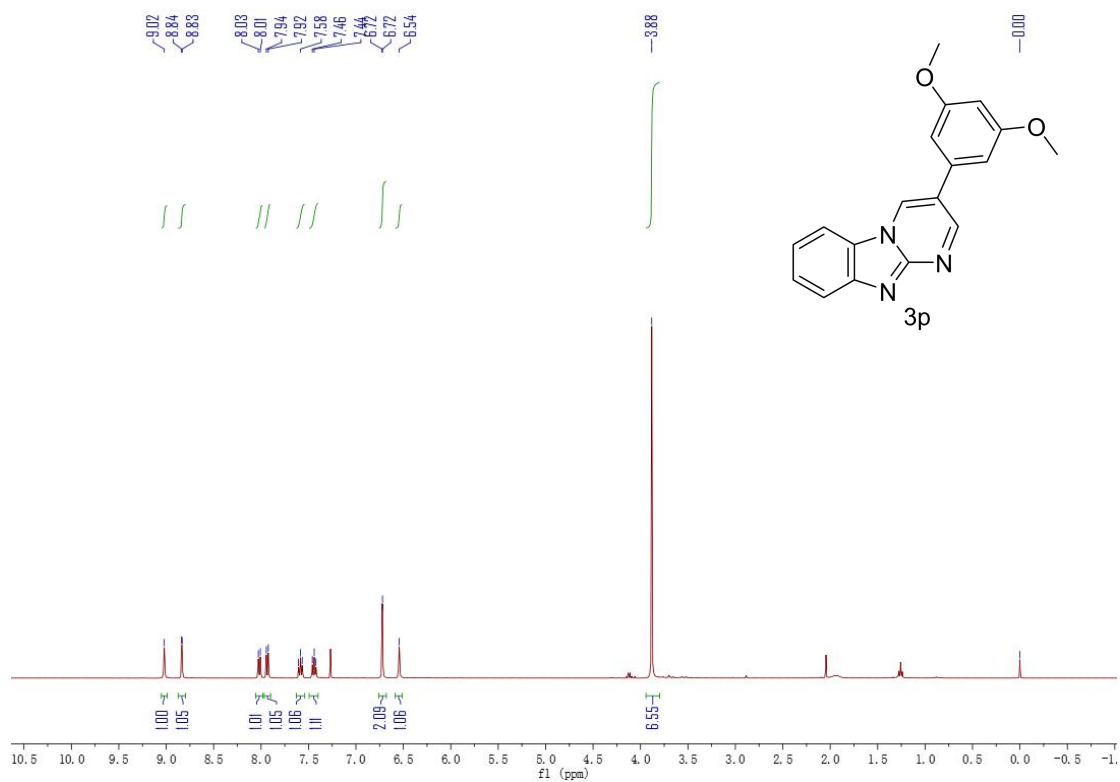
¹H NMR of Compound 3o (400 MHz, CDCl₃)



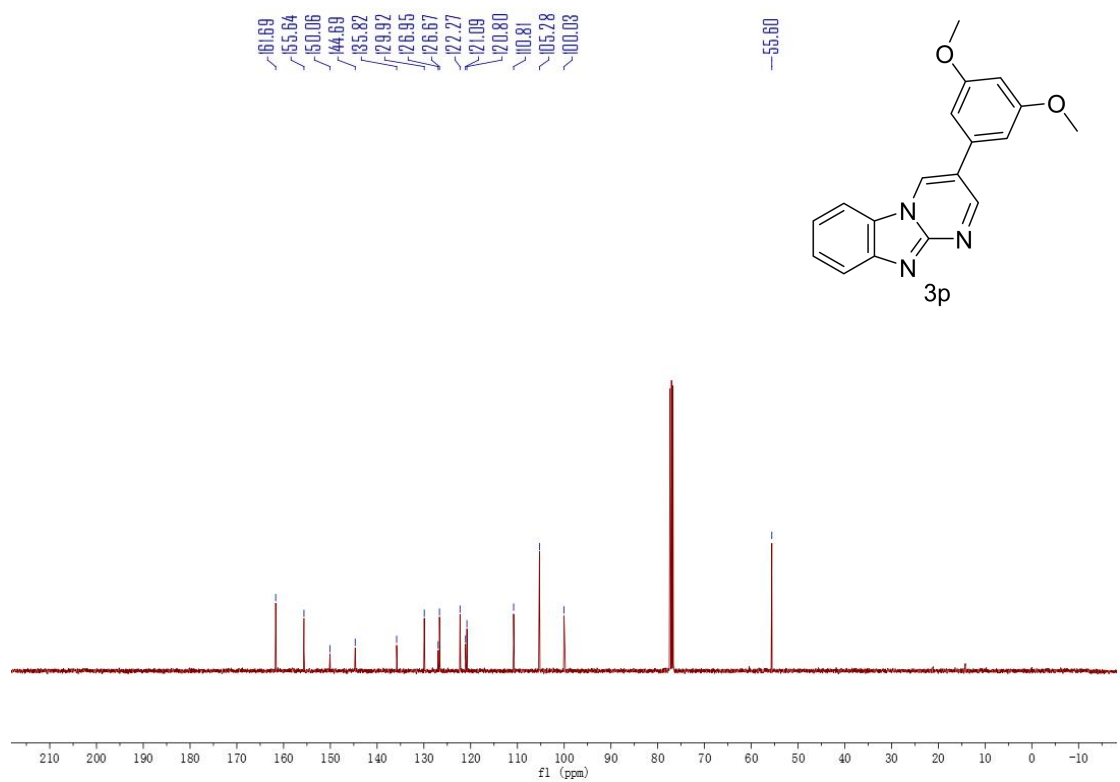
¹³C NMR of Compound 4b (400 MHz, CDCl₃)



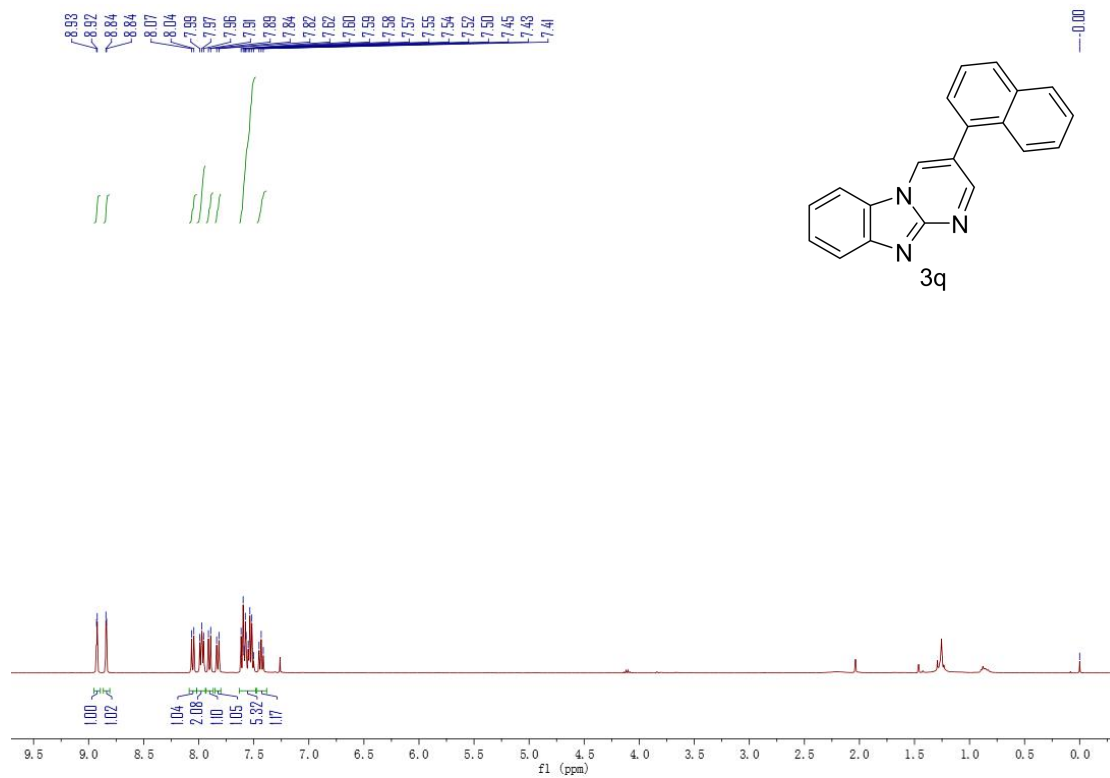
¹H NMR of Compound 3p (400 MHz, CDCl₃)



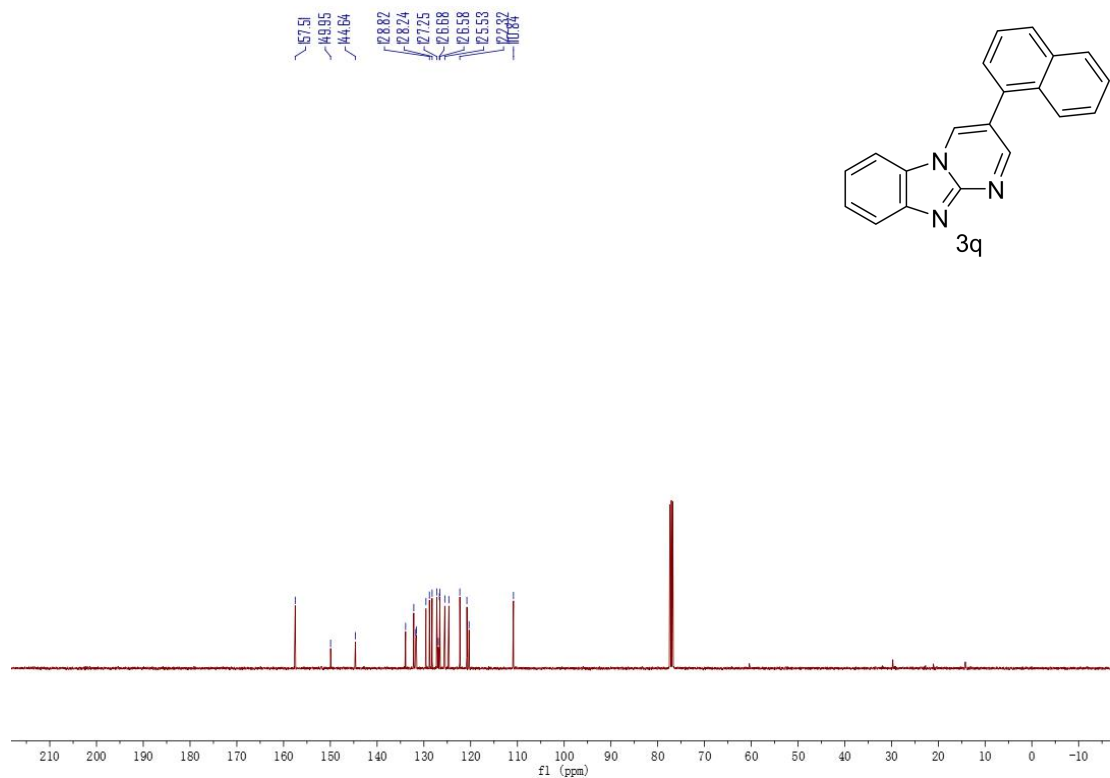
¹³C NMR of Compound 3p (400 MHz, CDCl₃)



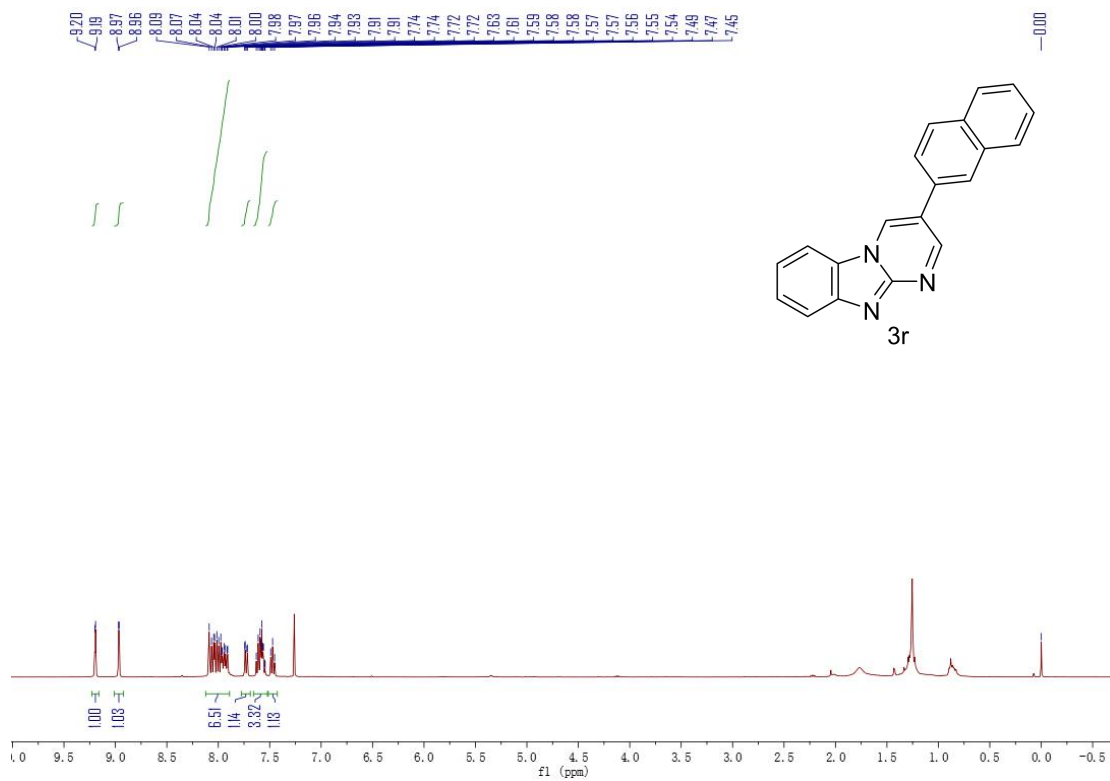
¹H NMR of Compound 3q (400 MHz, CDCl₃)



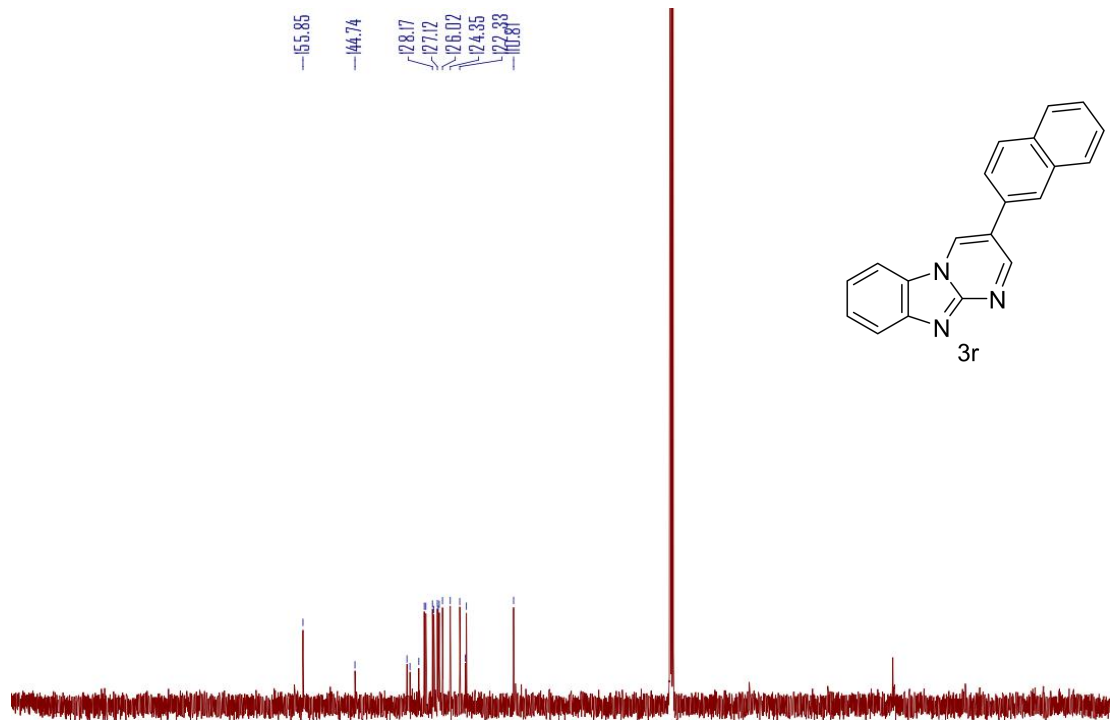
¹³C NMR of Compound 3q (400 MHz, CDCl₃)



¹H NMR of Compound 3r (400 MHz, CDCl₃)

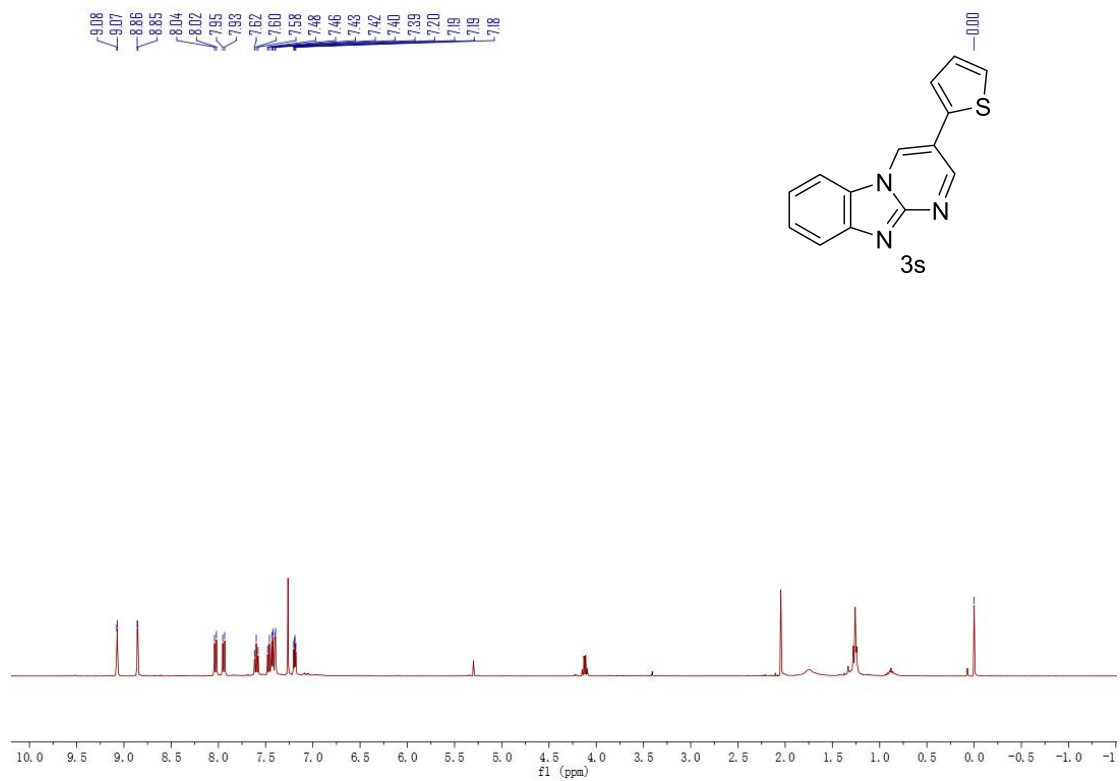


¹³C NMR of Compound 3r (400 MHz, CDCl₃)

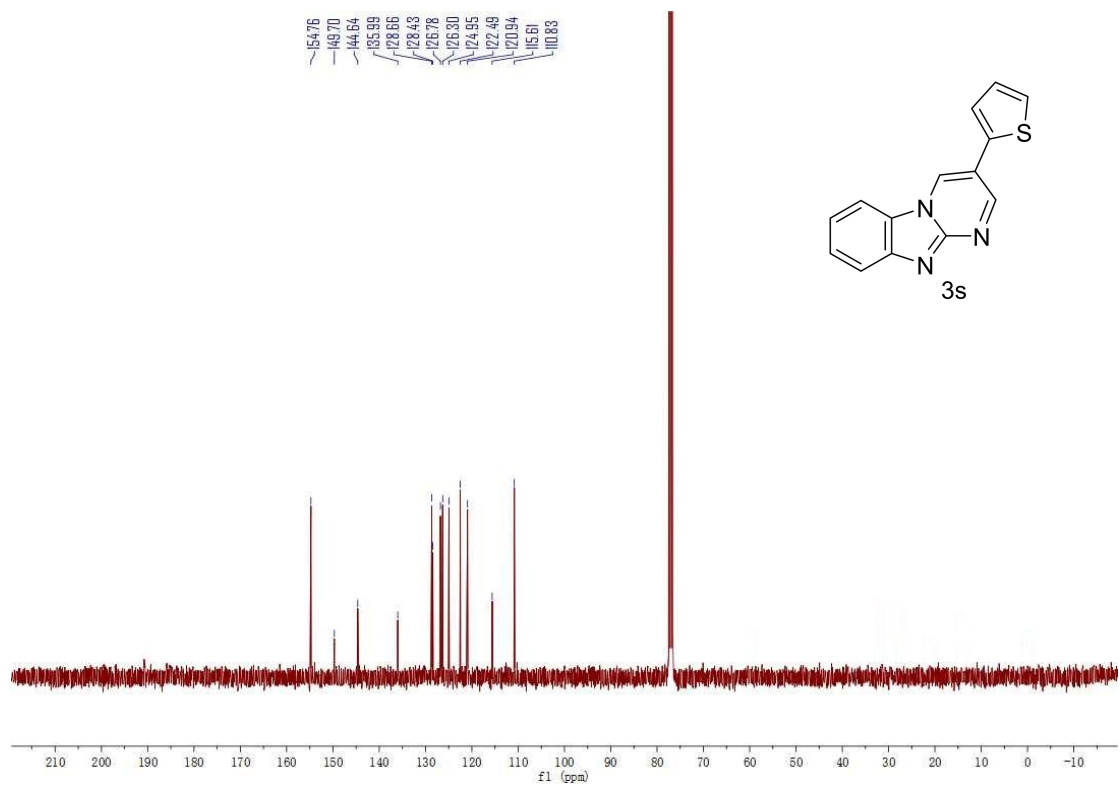


¹H NMR of Compound 3s (400 MHz, CDCl₃)

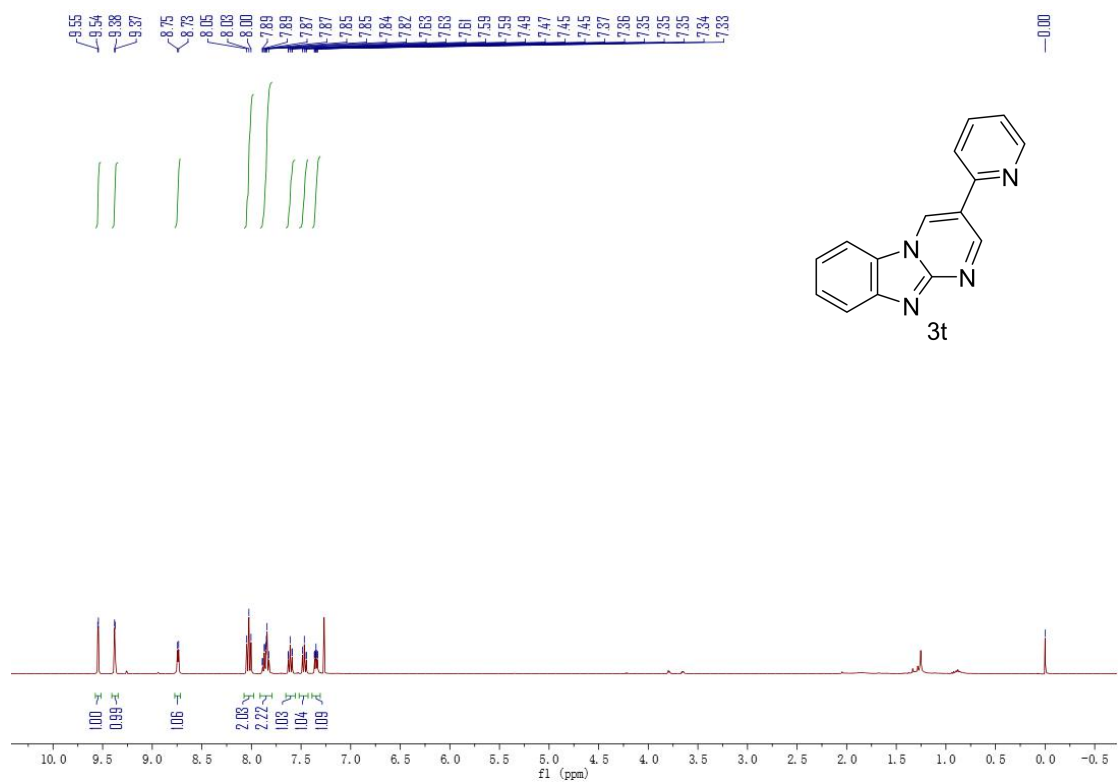




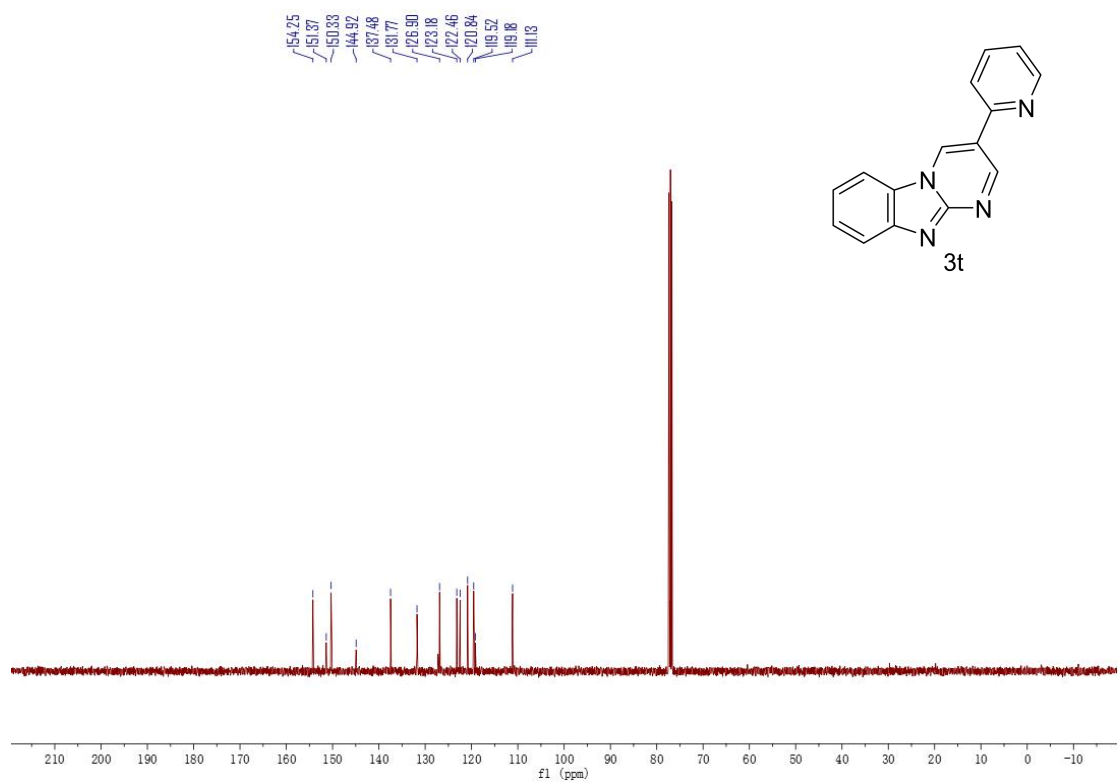
¹³C NMR of Compound **3s** (400 MHz, CDCl₃)



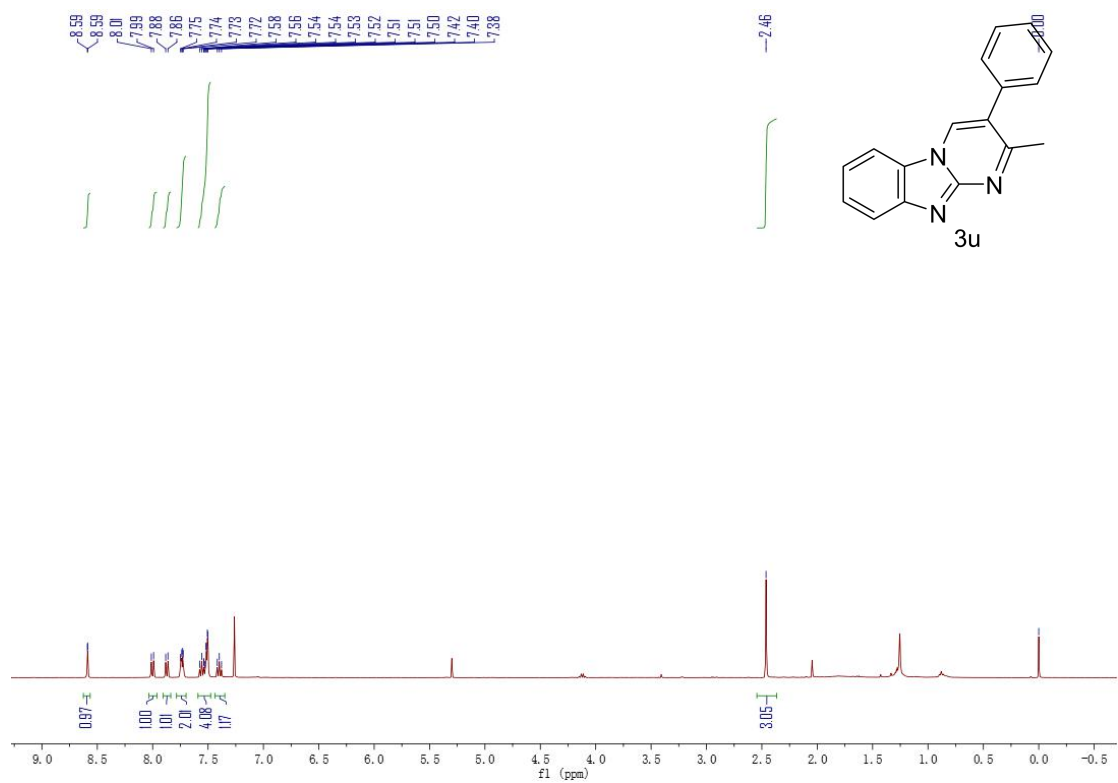
¹H NMR of Compound **3t** (400 MHz, CDCl₃)



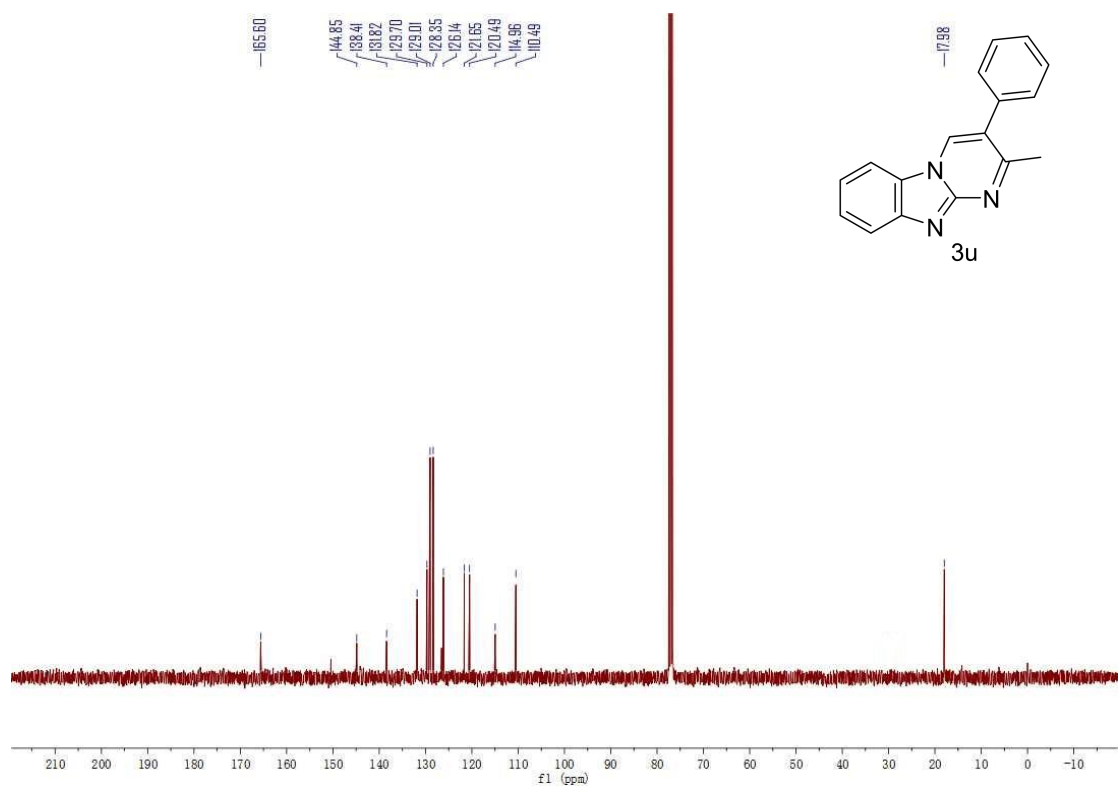
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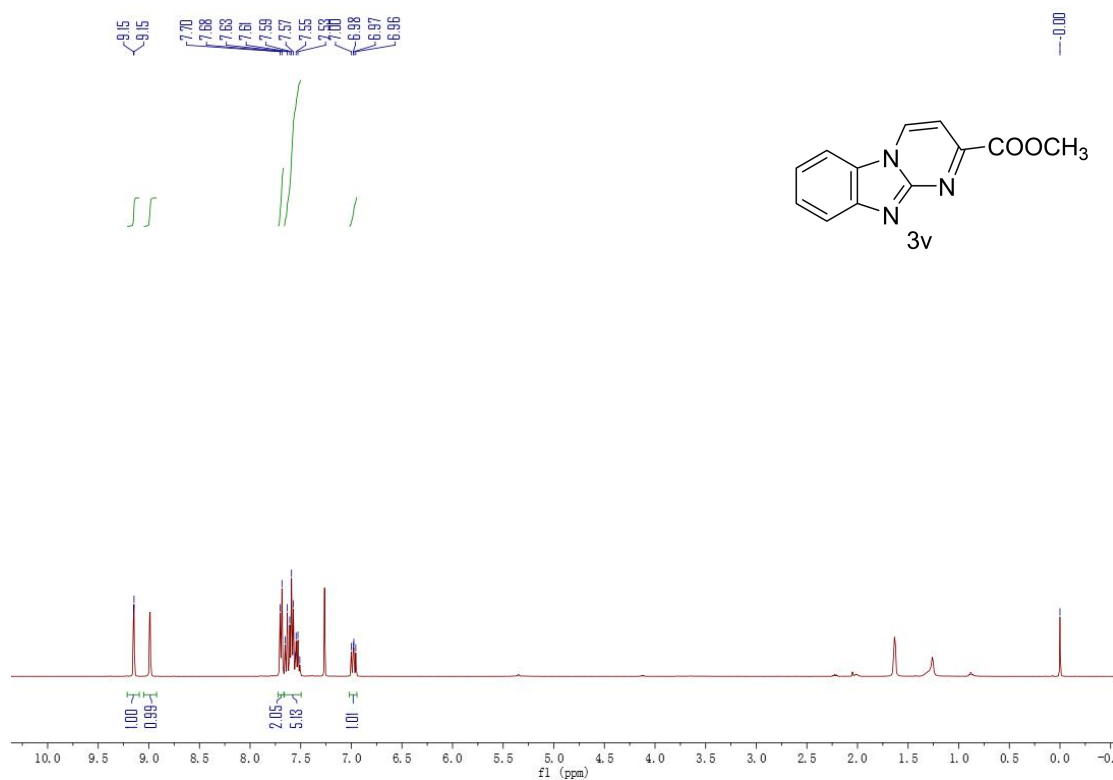
¹H NMR of Compound 3u (400 MHz, CDCl₃)



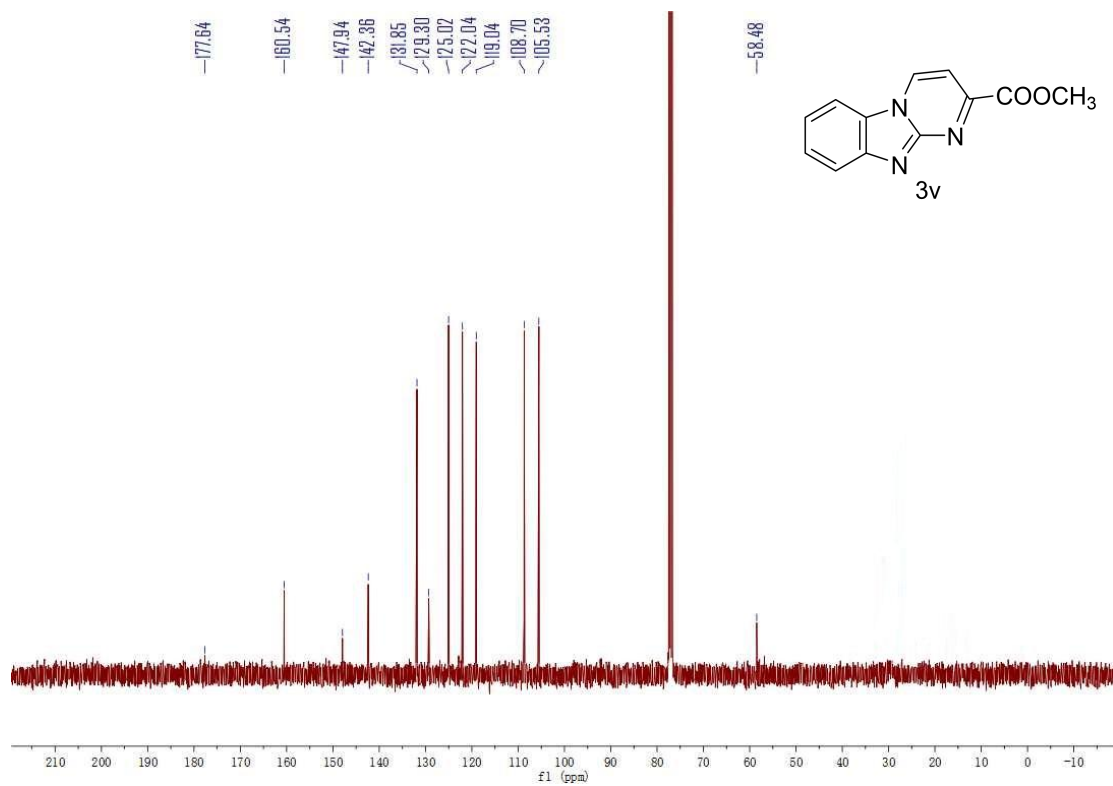
¹³C NMR of Compound 3u (400 MHz, CDCl₃)



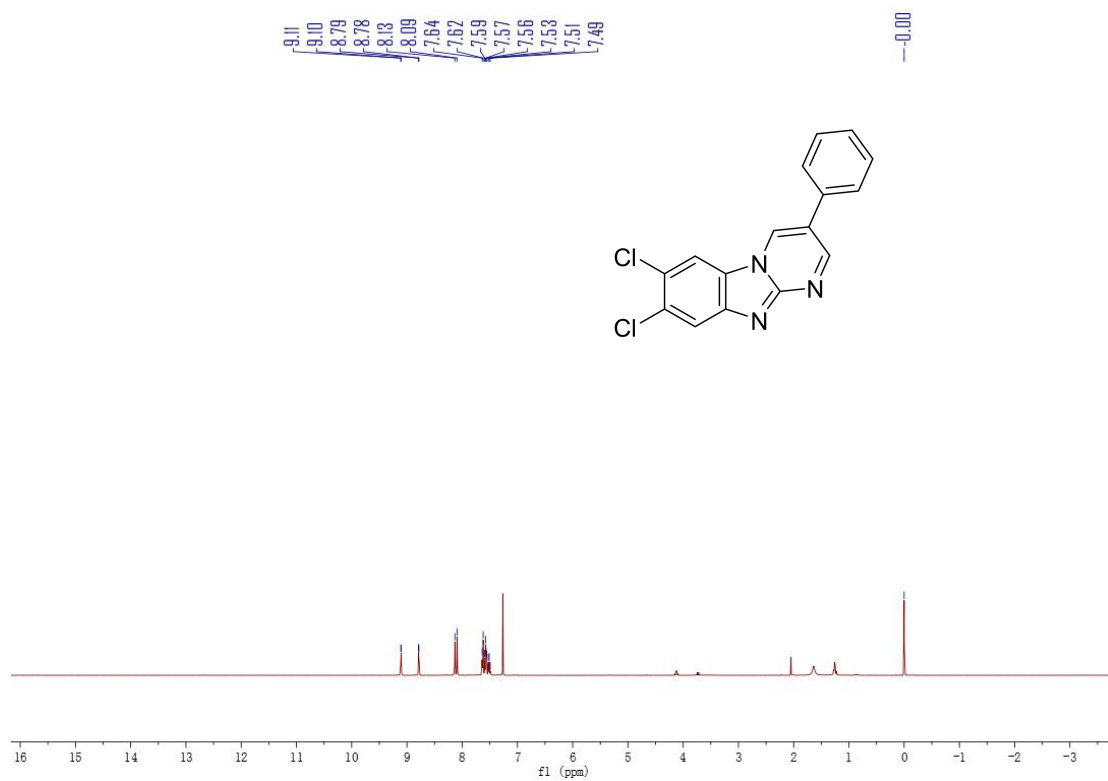
¹H NMR of Compound 3v (400 MHz, CDCl₃)



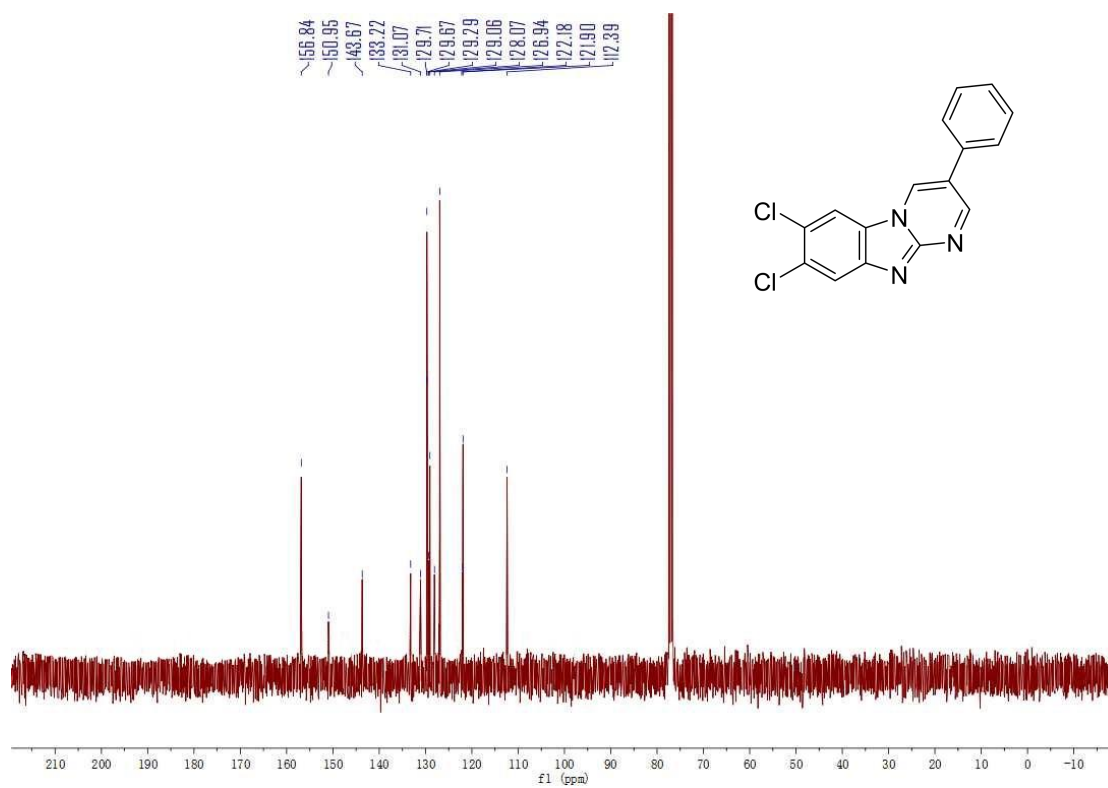
¹³C NMR of Compound **3v** (400 MHz, CDCl₃)



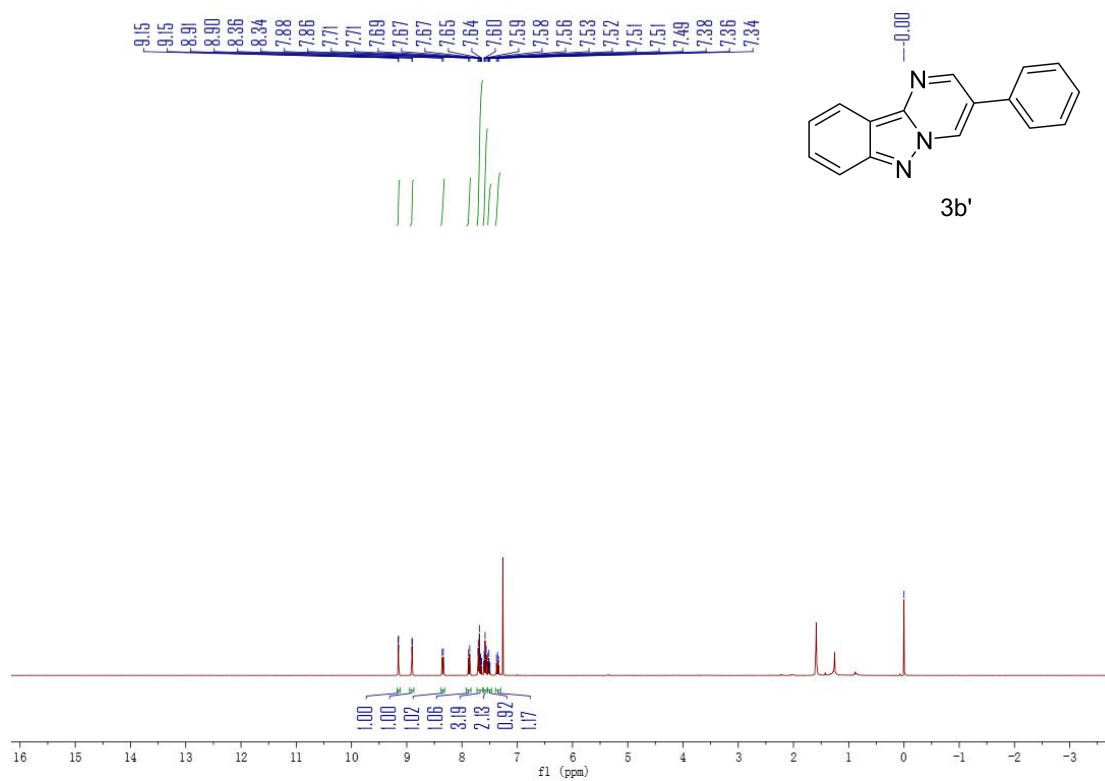
¹H NMR of Compound **3a'** (400 MHz, CDCl₃)



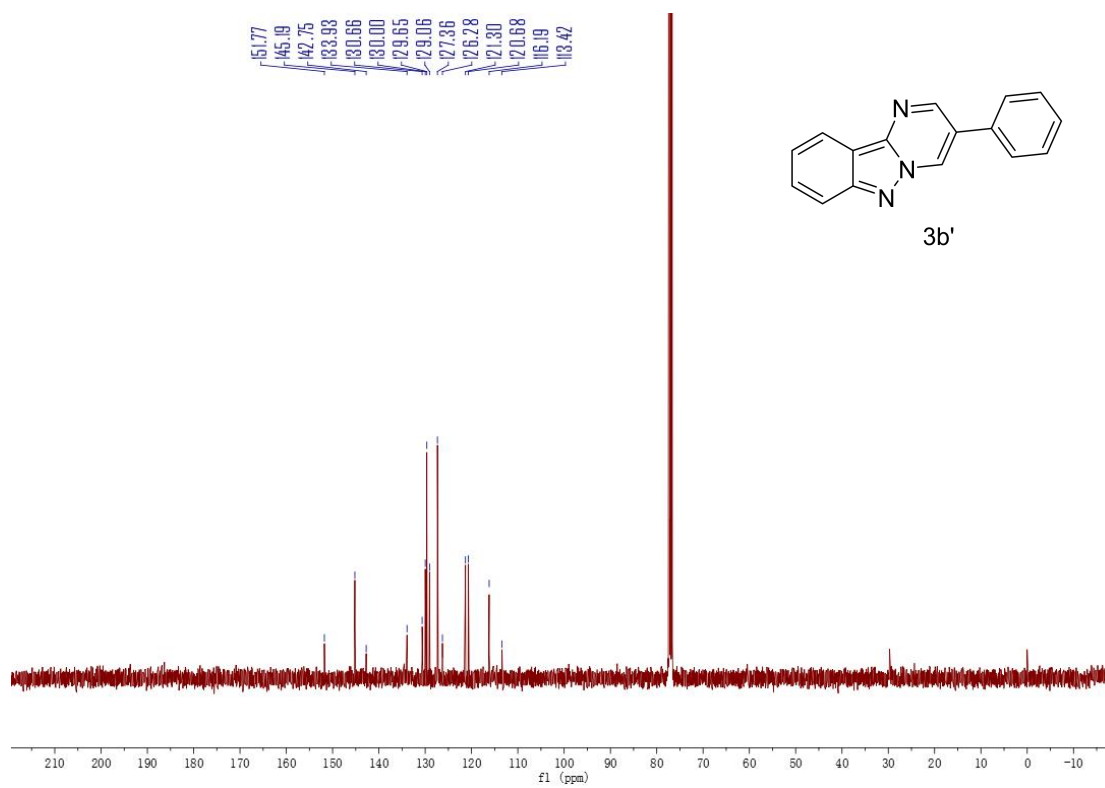
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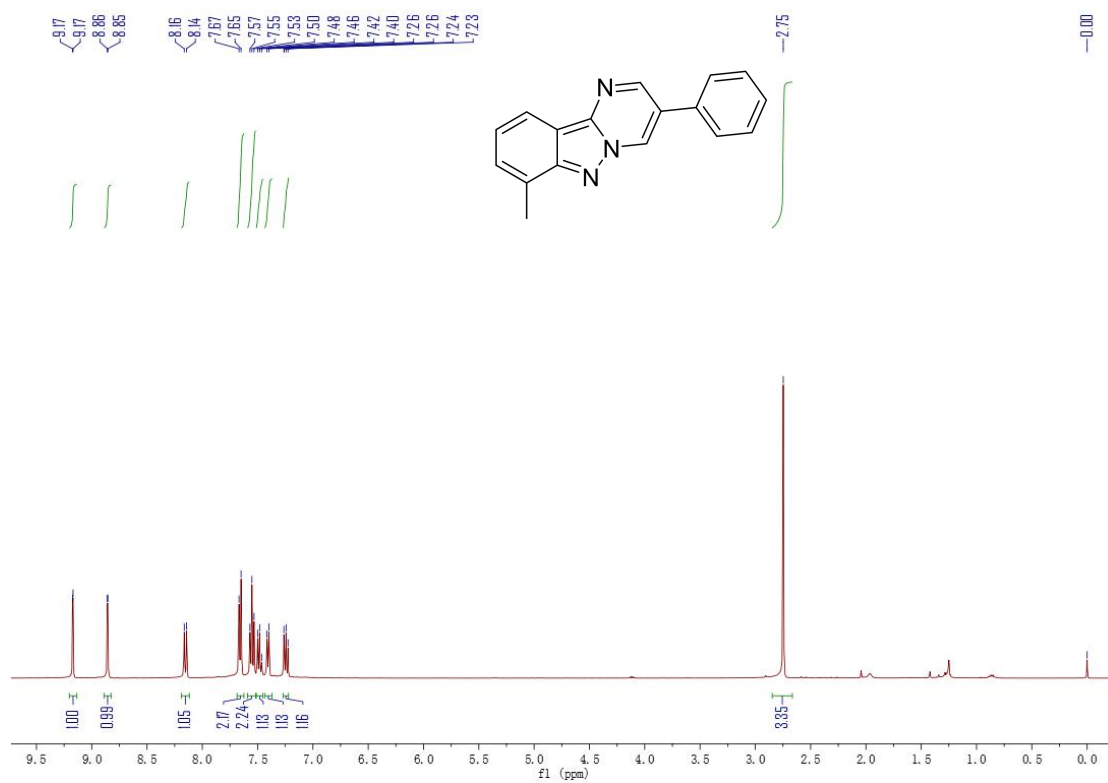
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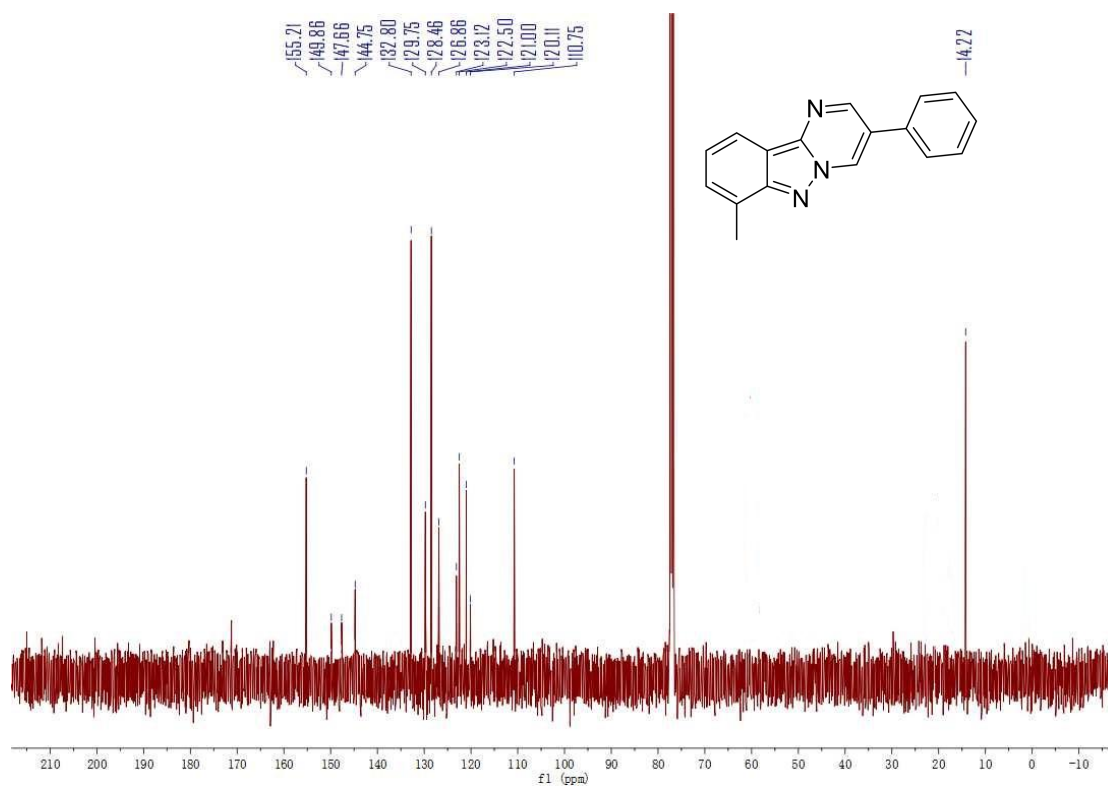
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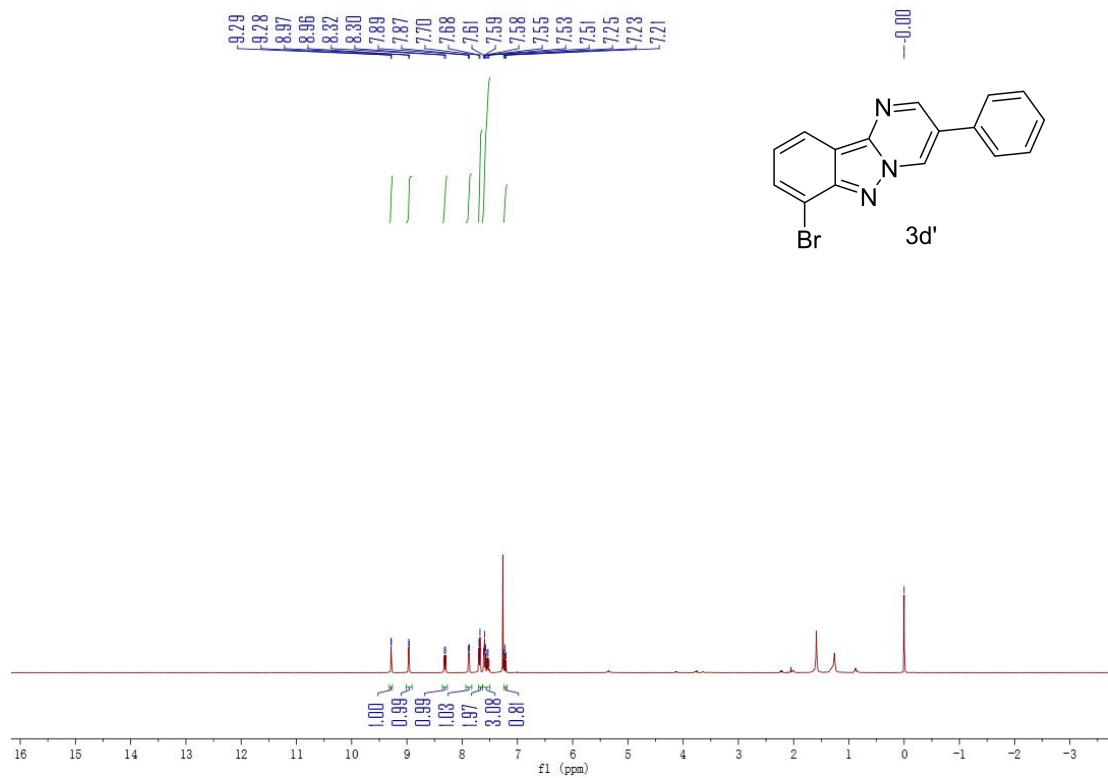
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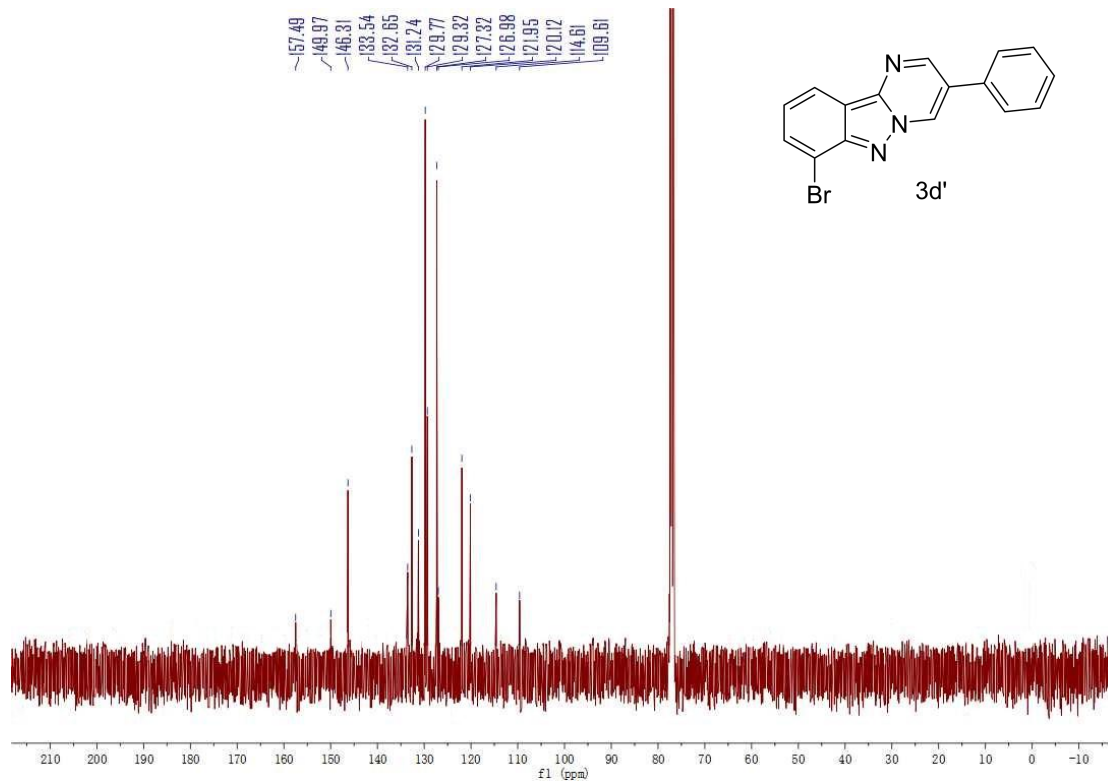
¹³C NMR of Compound 3c' (400 MHz, CDCl₃)



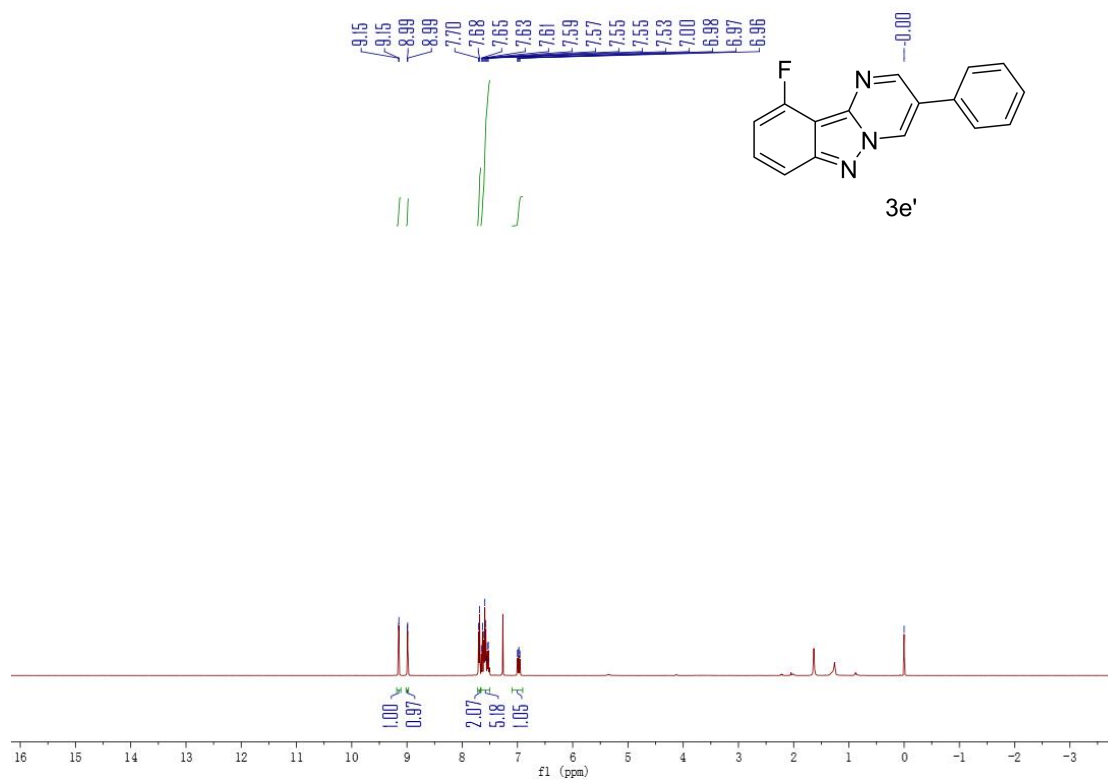
¹H NMR of Compound 3d' (400 MHz, CDCl₃)



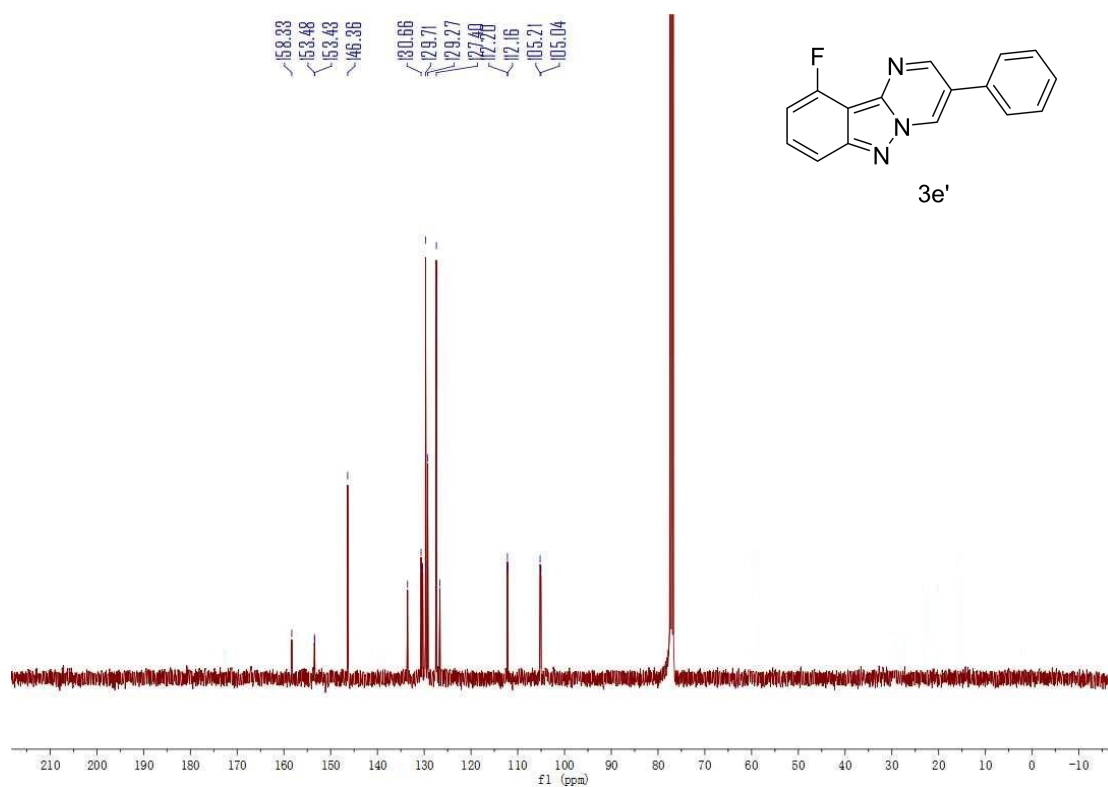
¹³C NMR of Compound 3d' (400 MHz, CDCl₃)



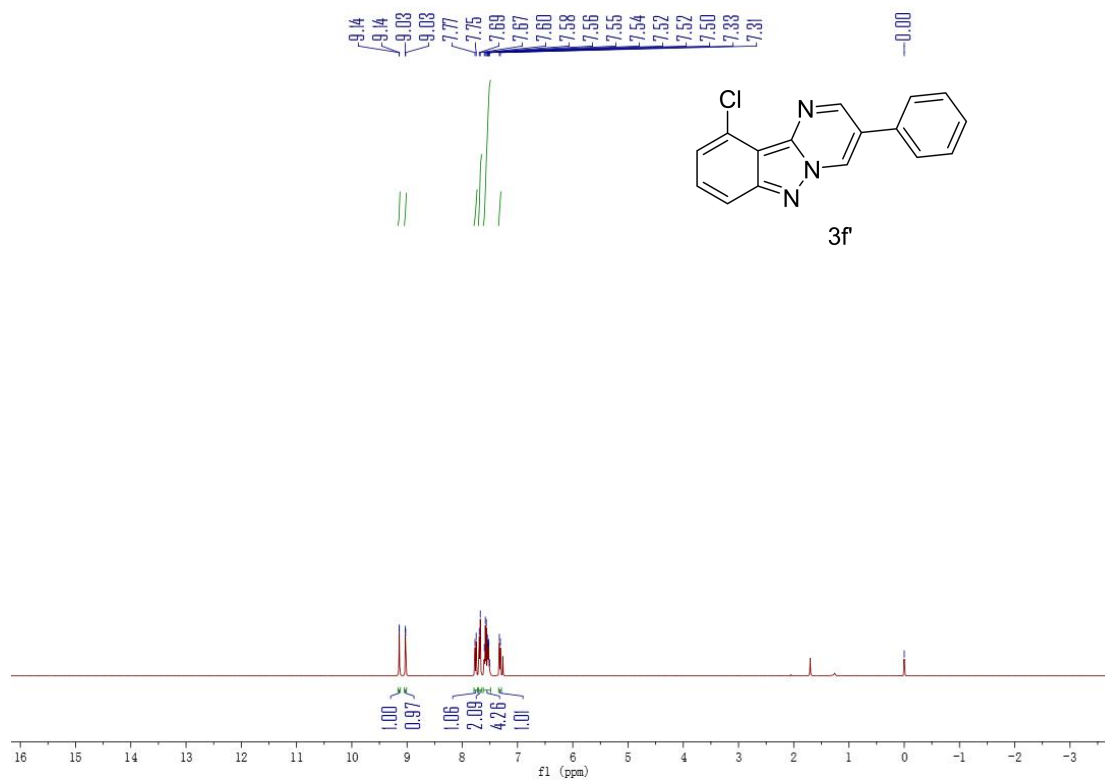
¹H NMR of Compound 3e' (400 MHz, CDCl₃)



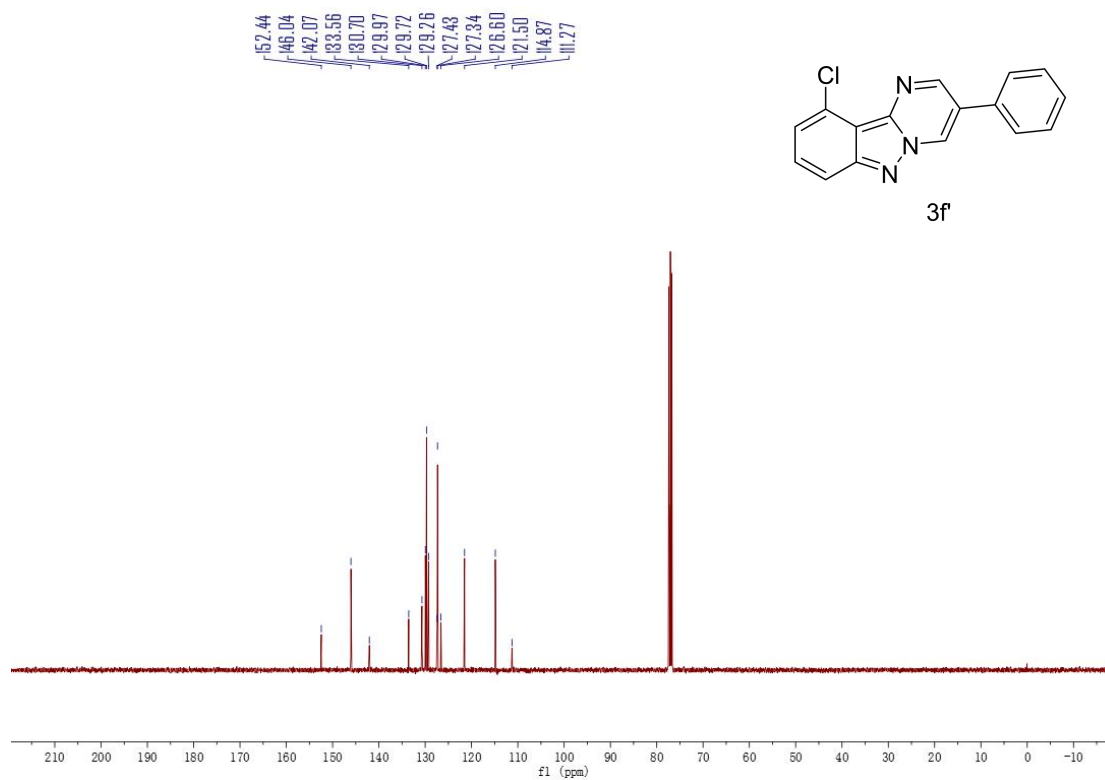
¹³C NMR of Compound 3e' (400 MHz, CDCl₃)



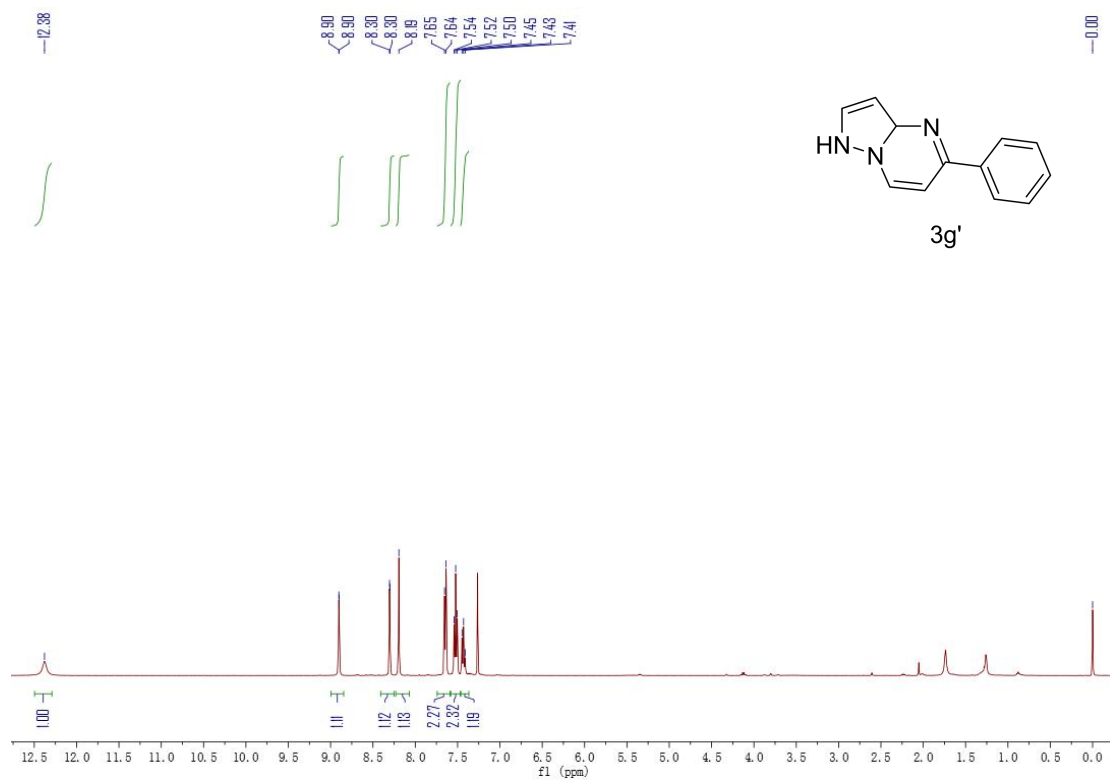
¹H NMR of Compound 3f' (400 MHz, CDCl₃)



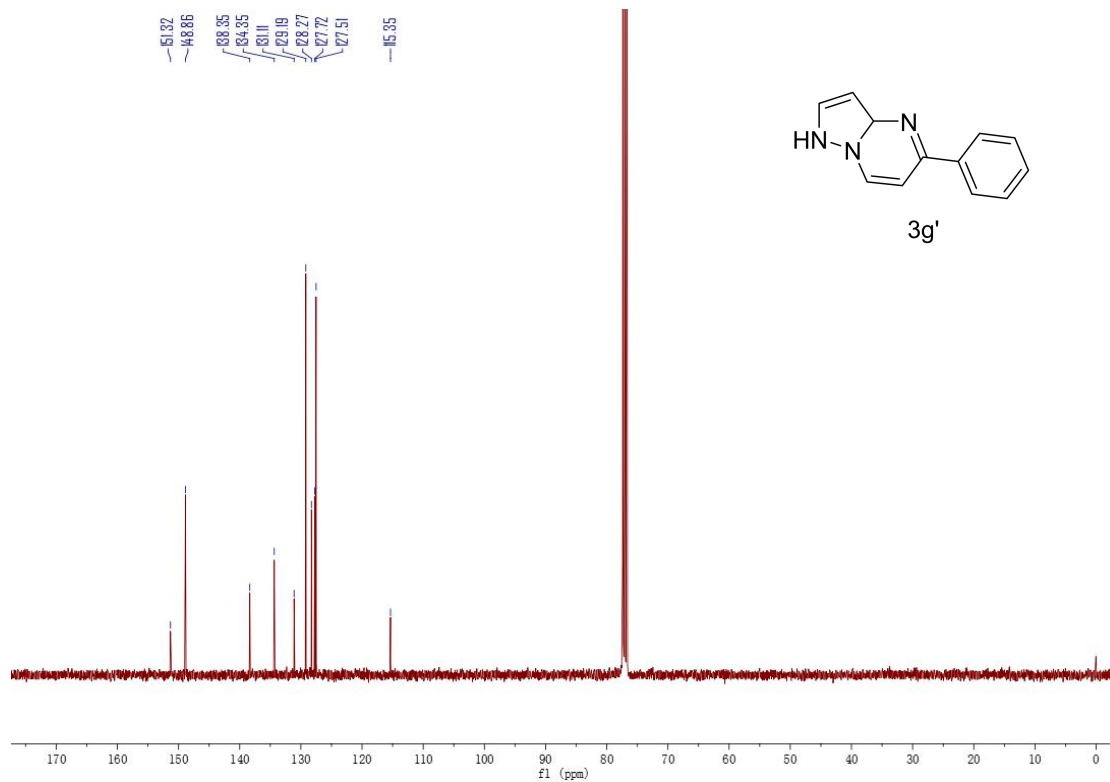
¹³C NMR of Compound **3f** (400 MHz, CDCl₃)



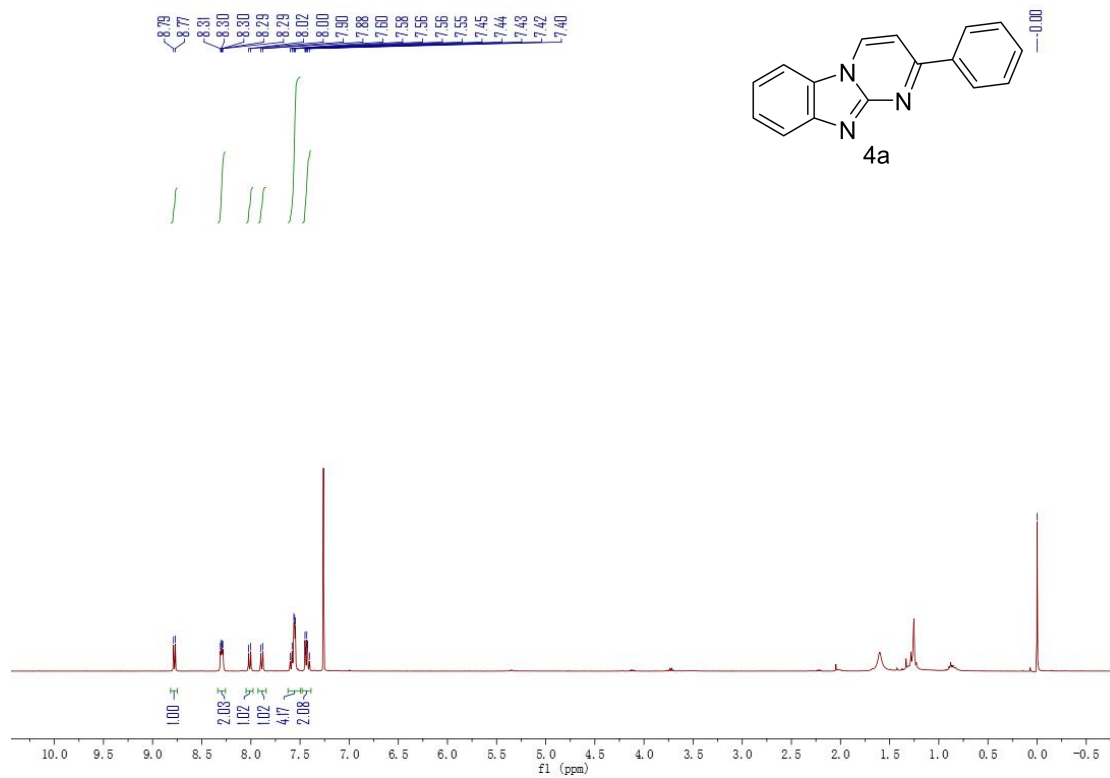
¹H NMR of Compound **3g'** (400 MHz, CDCl₃)



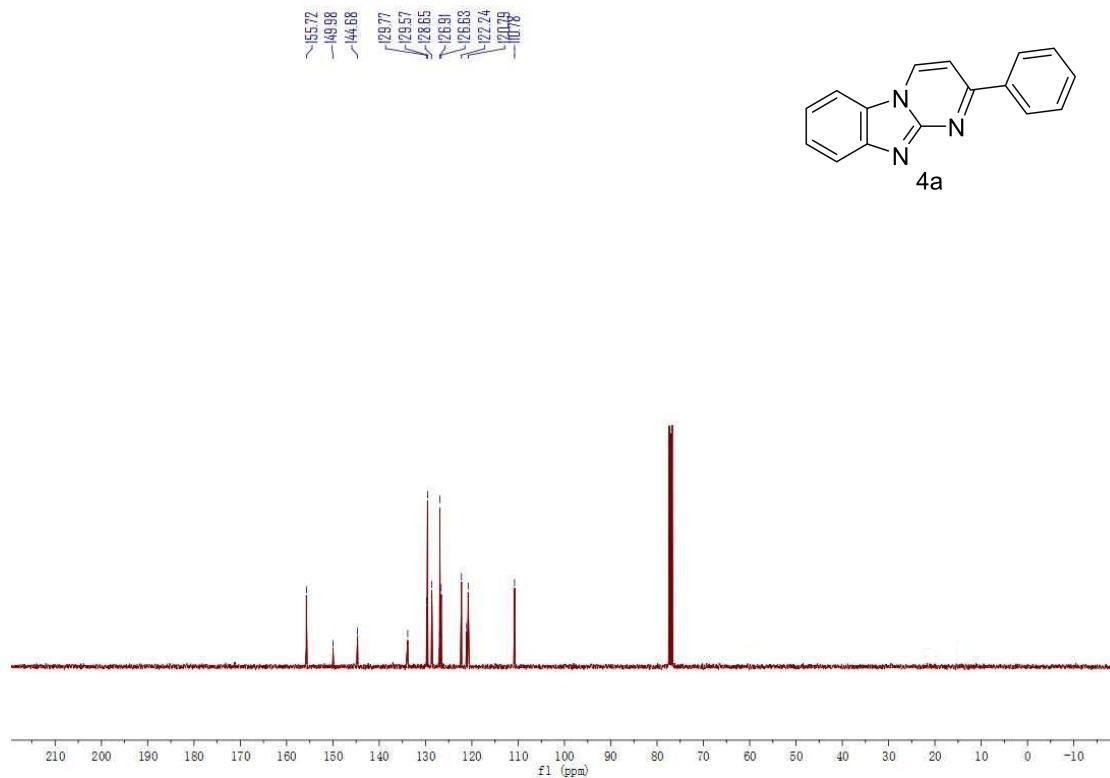
¹³C NMR of Compound **3g'** (400 MHz, CDCl₃)



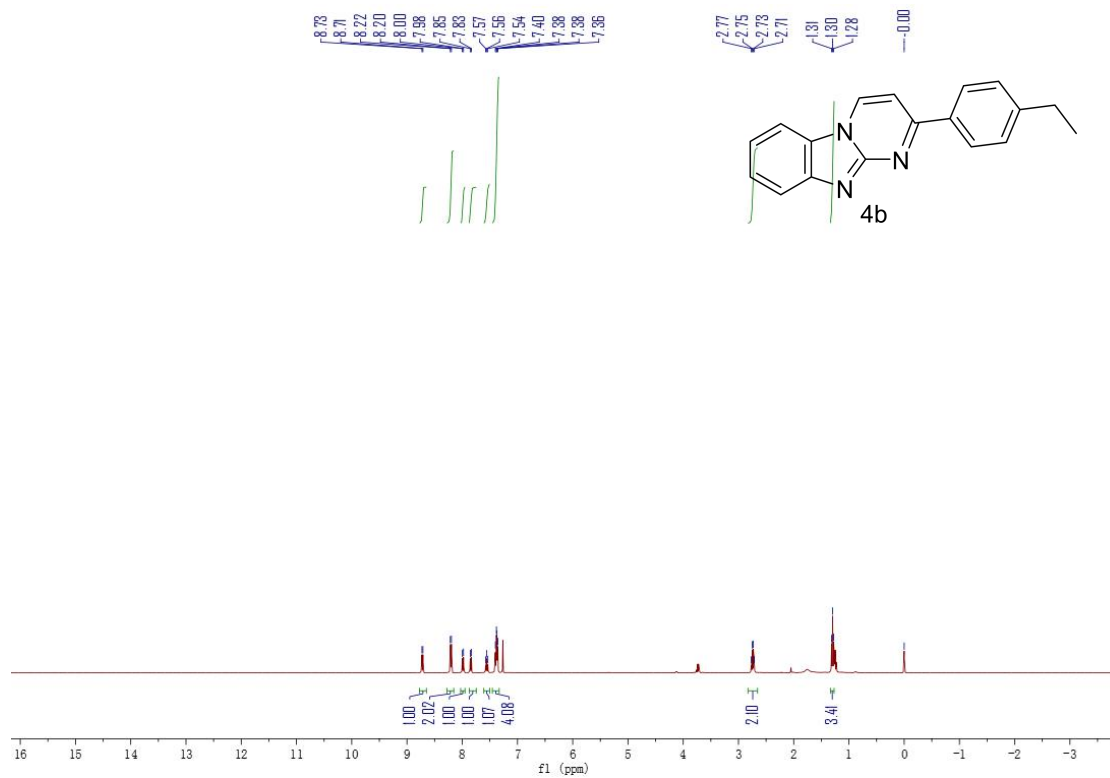
¹H NMR of Compound **4a** (400 MHz, CDCl₃)



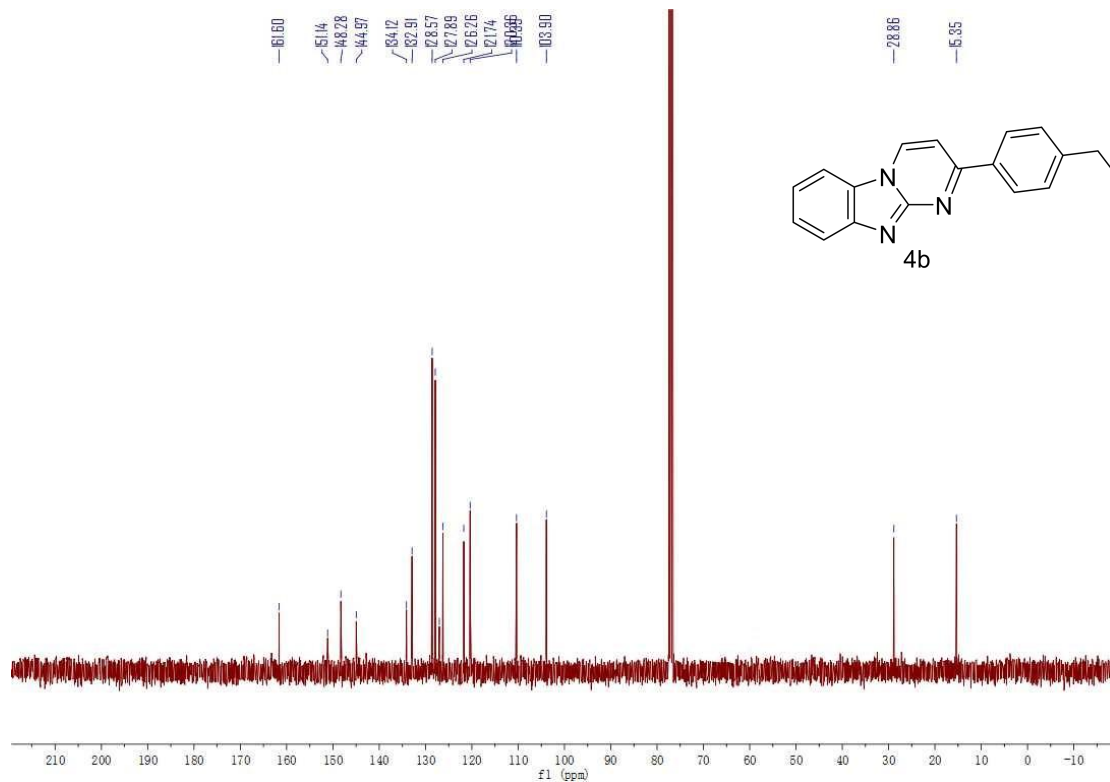
¹³C NMR of Compound 4a (400 MHz, CDCl₃)



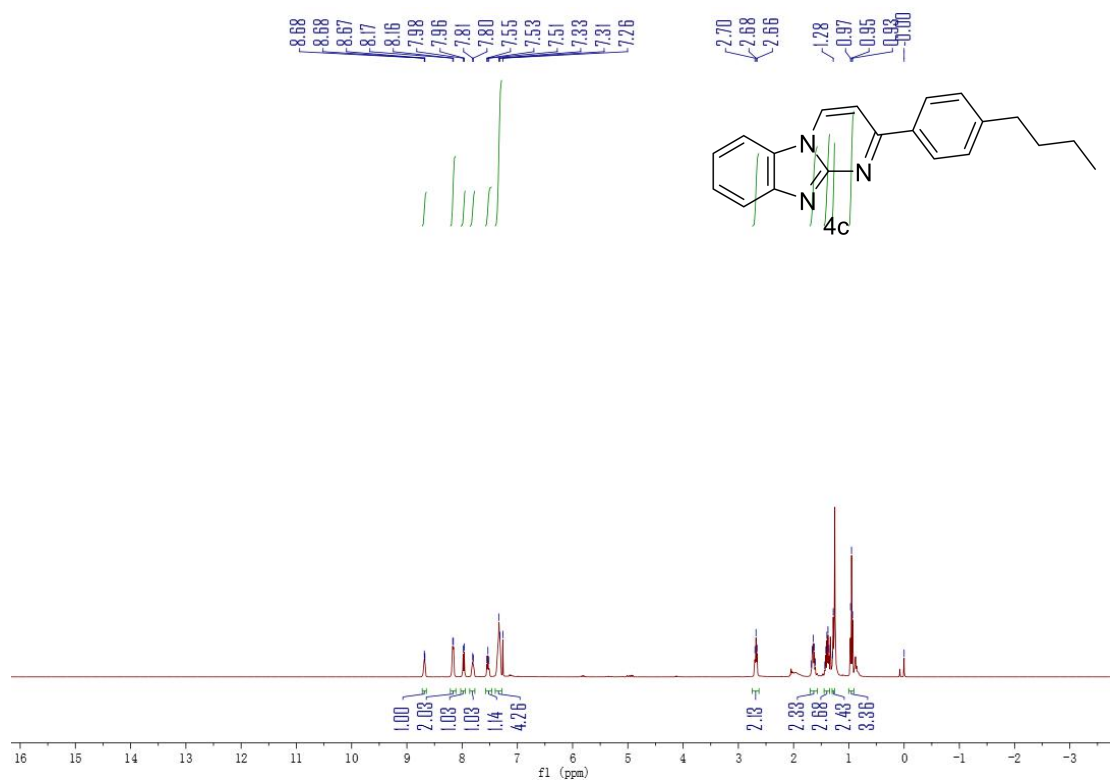
¹H NMR of Compound 4b (400 MHz, CDCl₃)



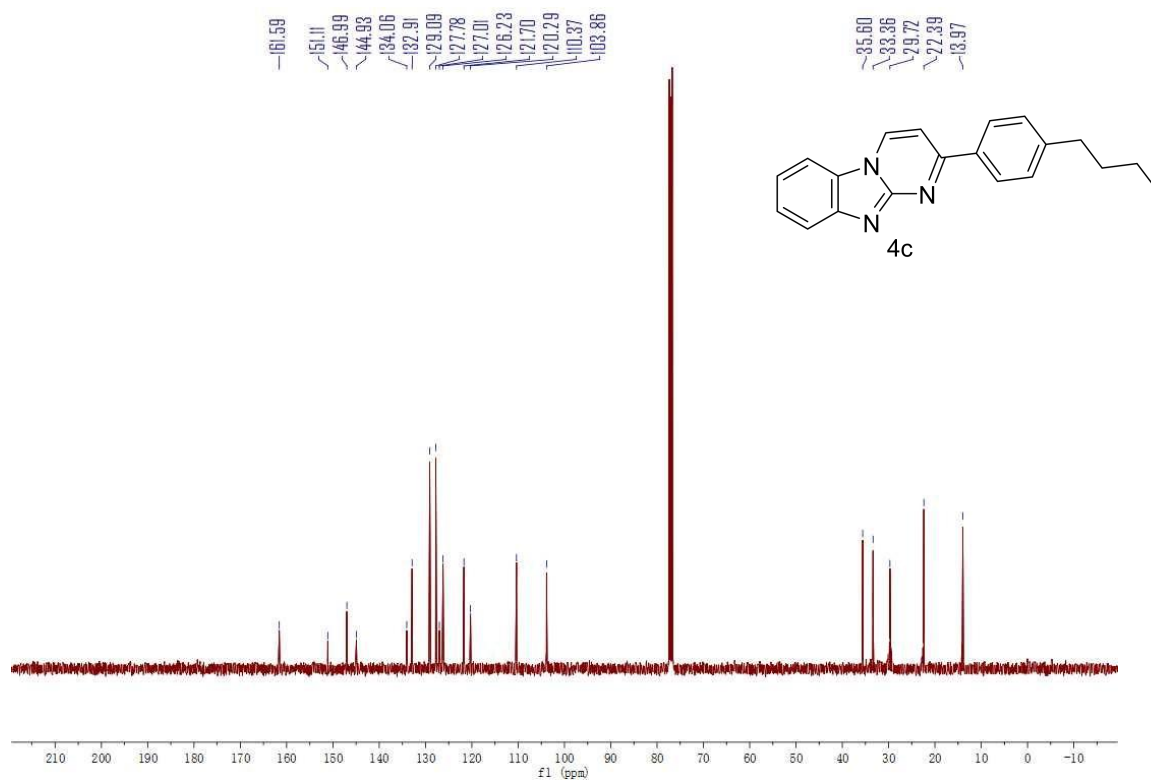
¹³C NMR of Compound 4b (400 MHz, CDCl₃)



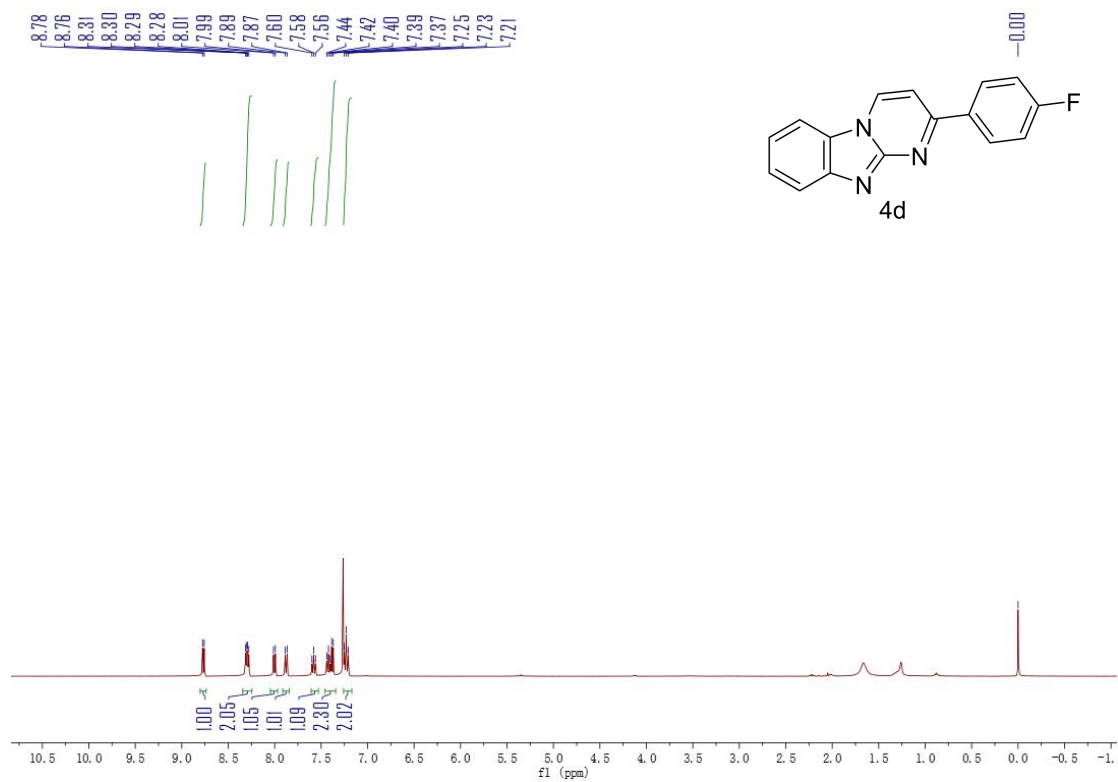
¹H NMR of Compound 4c (400 MHz, CDCl₃)



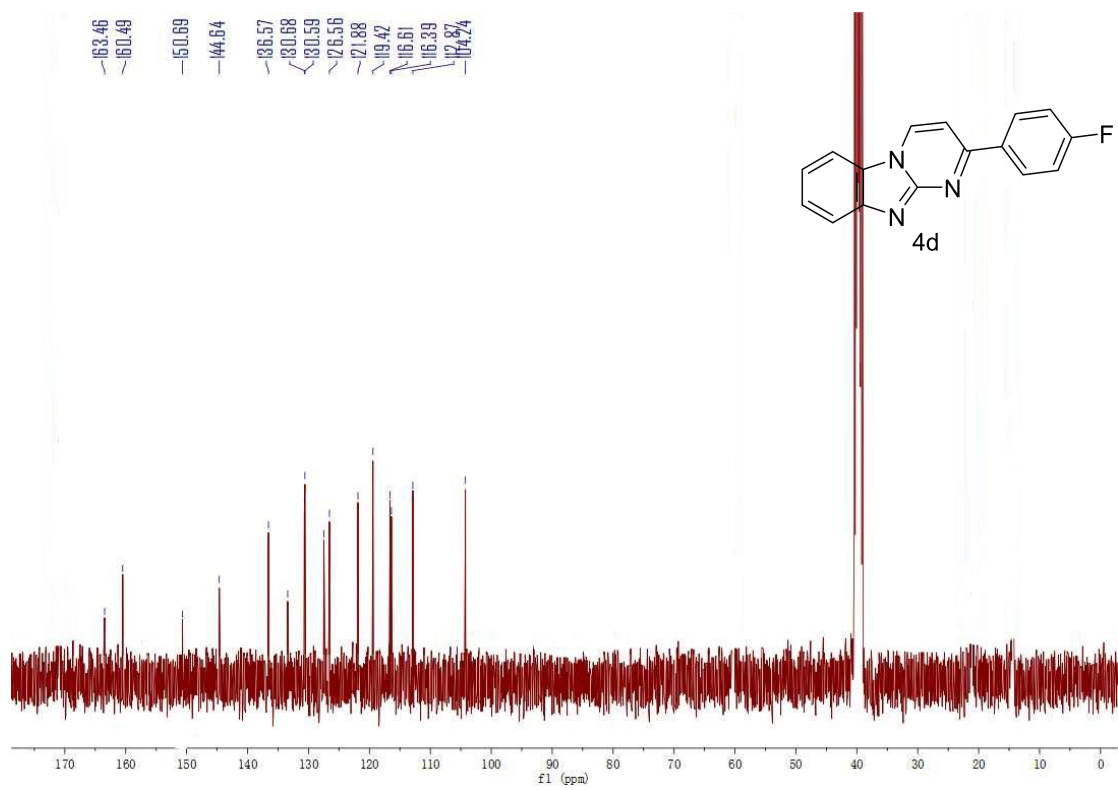
¹³C NMR of Compound 4c (400 MHz, CDCl₃)



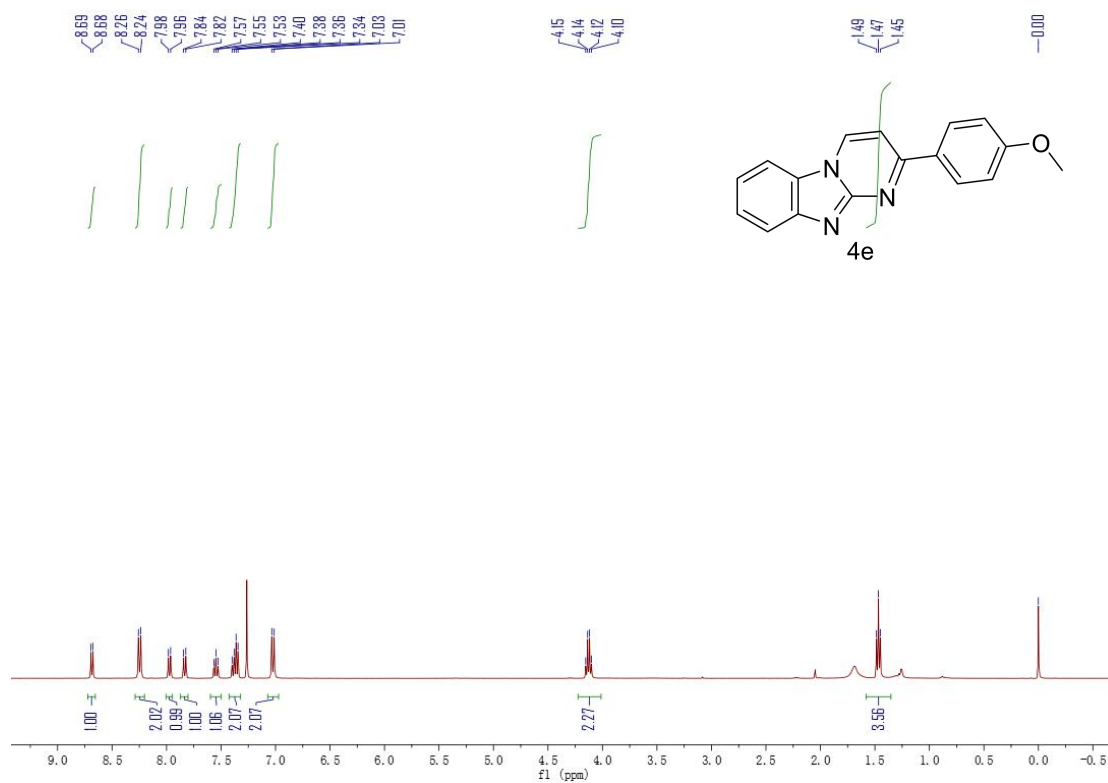
¹H NMR of Compound 4d (400 MHz, CDCl₃)



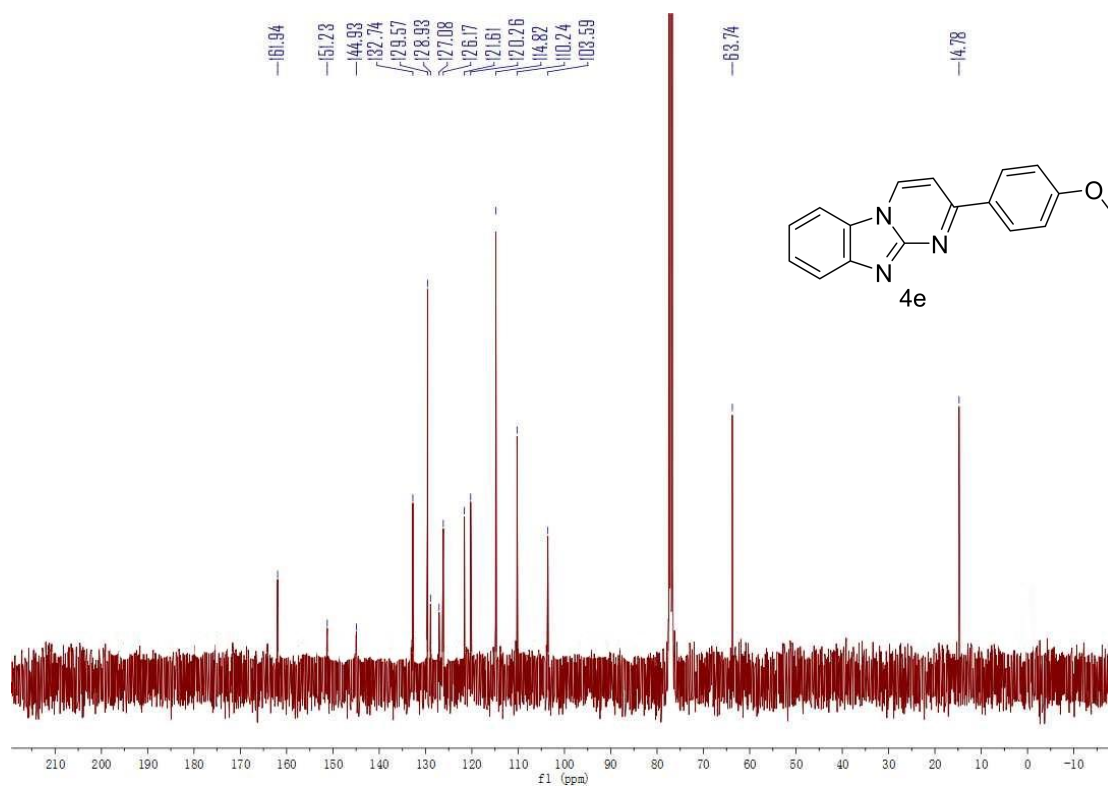
¹³C NMR of Compound 4d (400 MHz, CDCl₃)



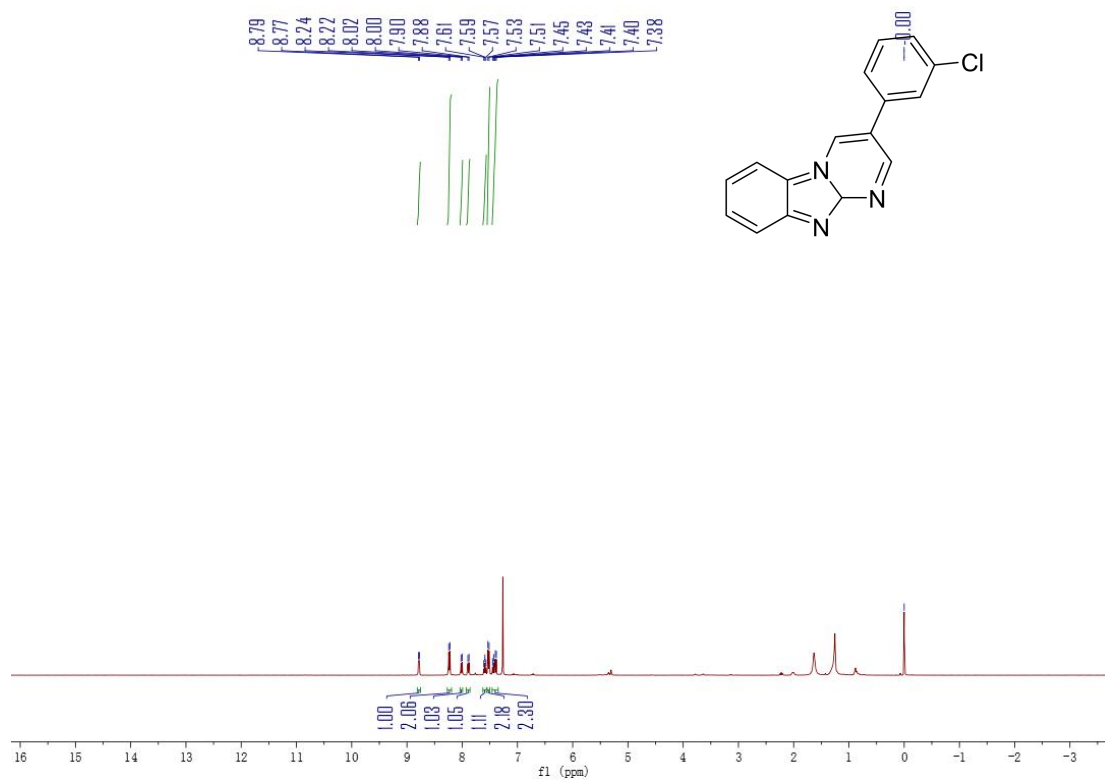
¹H NMR of Compound 4e (400 MHz, CDCl₃)



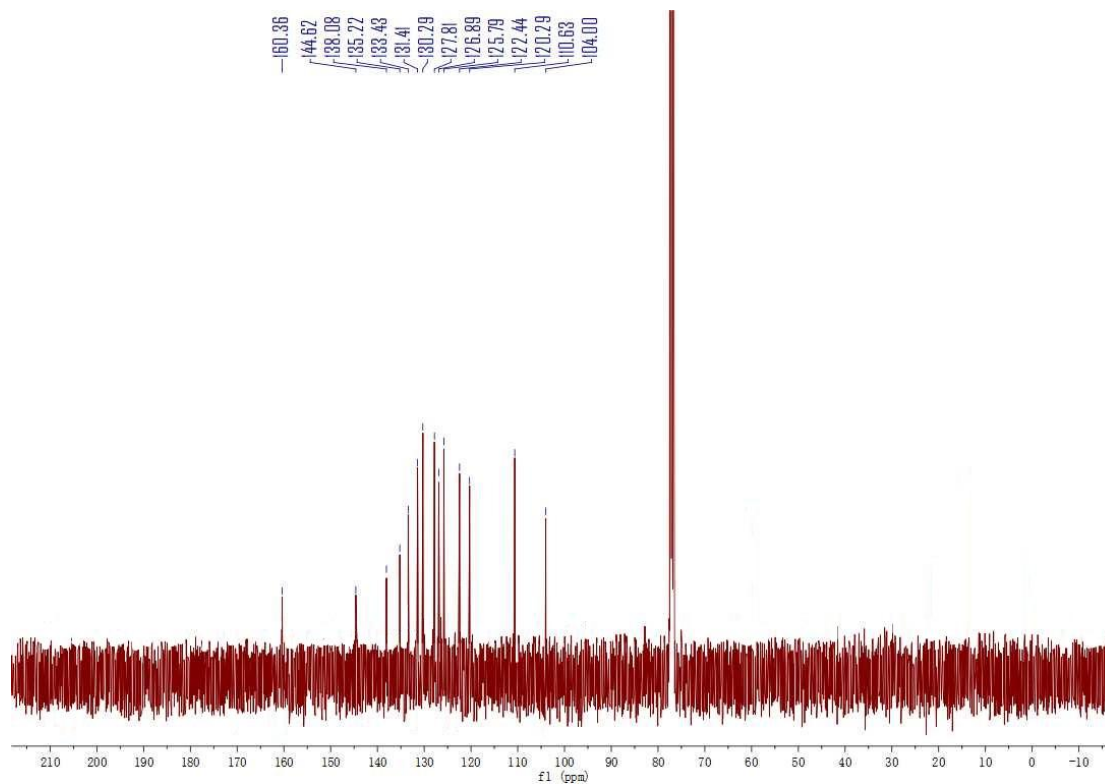
¹³C NMR of Compound 4e (400 MHz, CDCl₃)



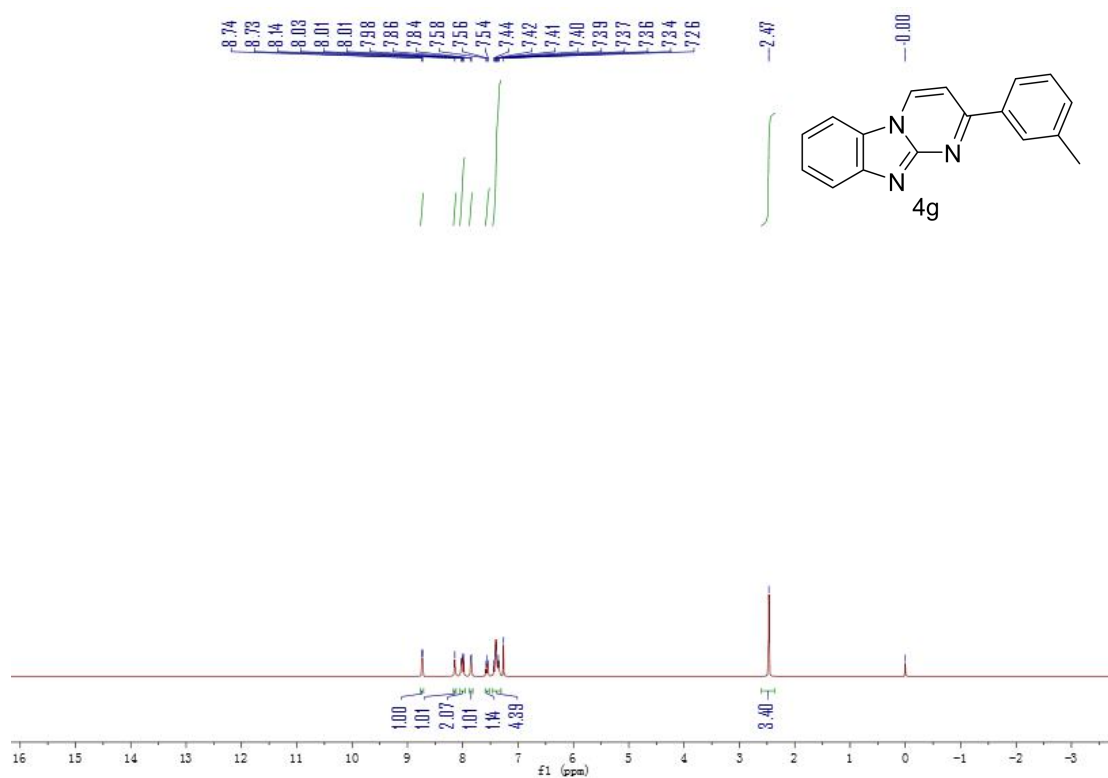
¹H NMR of Compound 4f (400 MHz, CDCl₃)



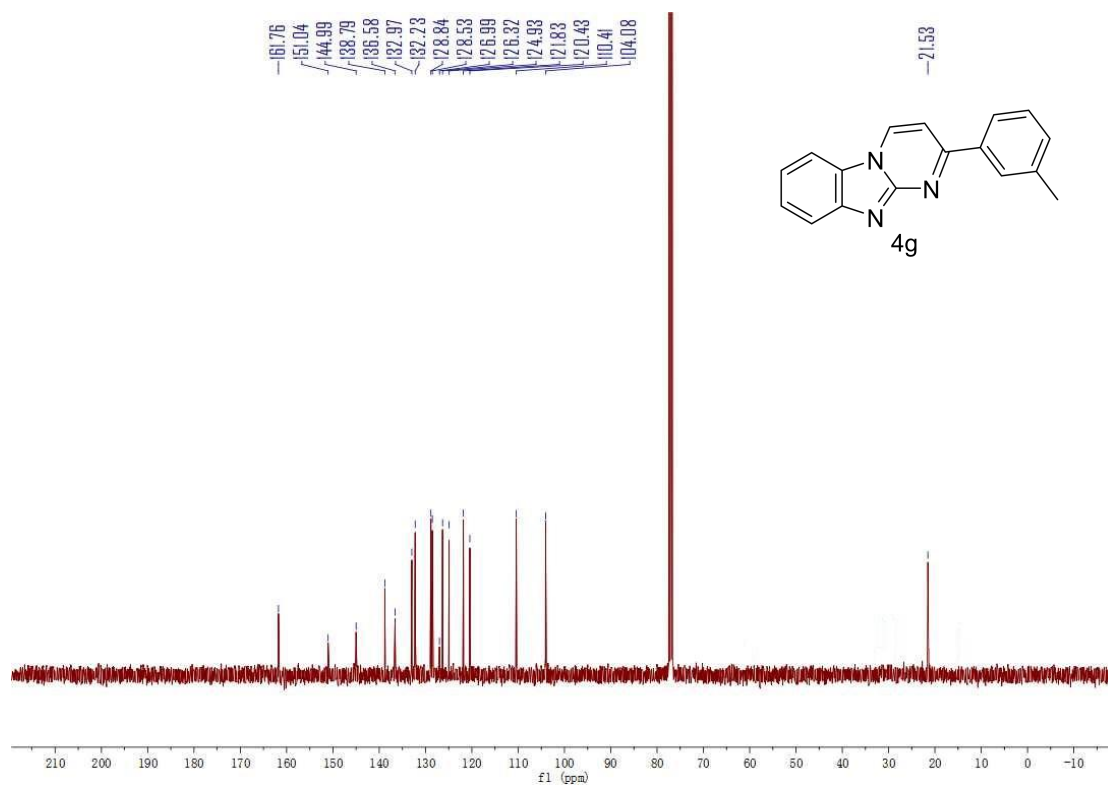
¹³C NMR of Compound 4f (400 MHz, CDCl₃)



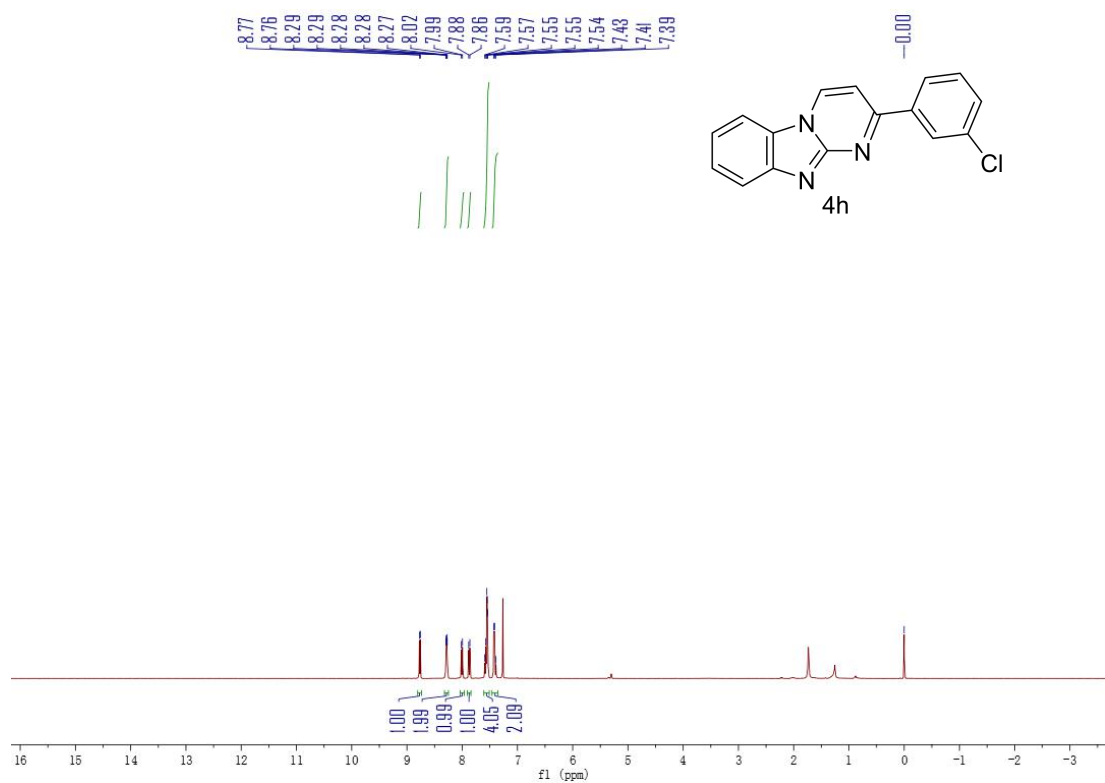
¹H NMR of Compound 4g (400 MHz, CDCl₃)



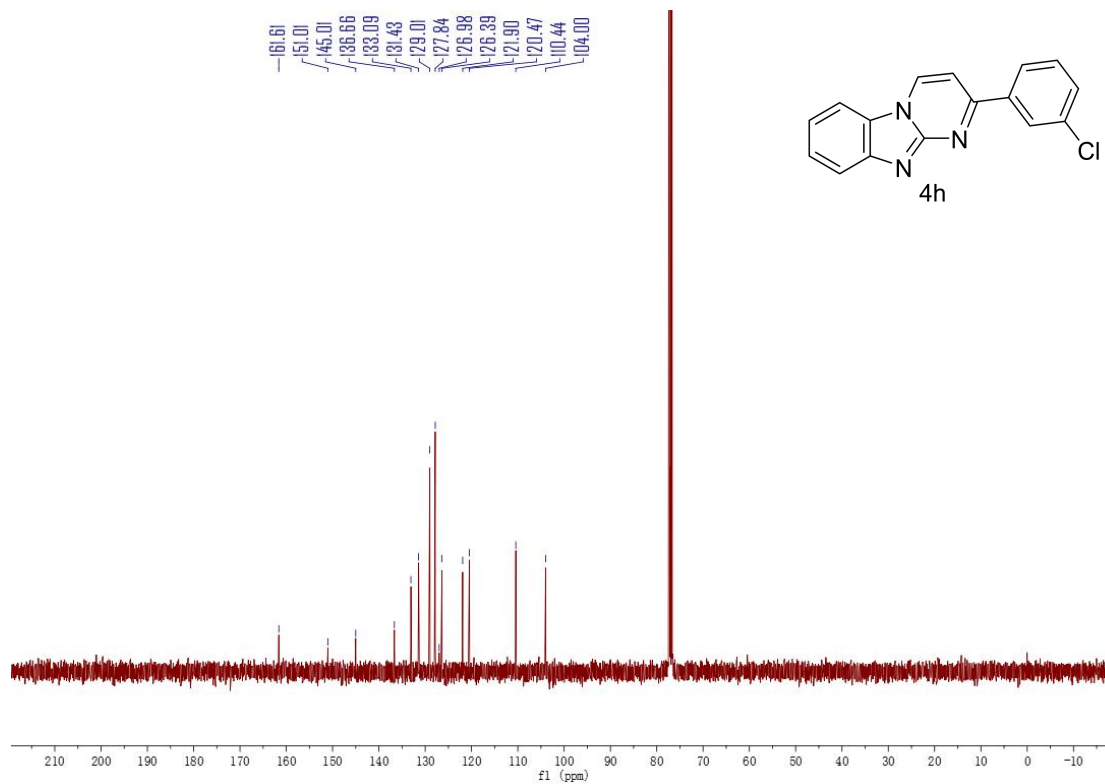
¹³C NMR of Compound 4g (400 MHz, CDCl₃)



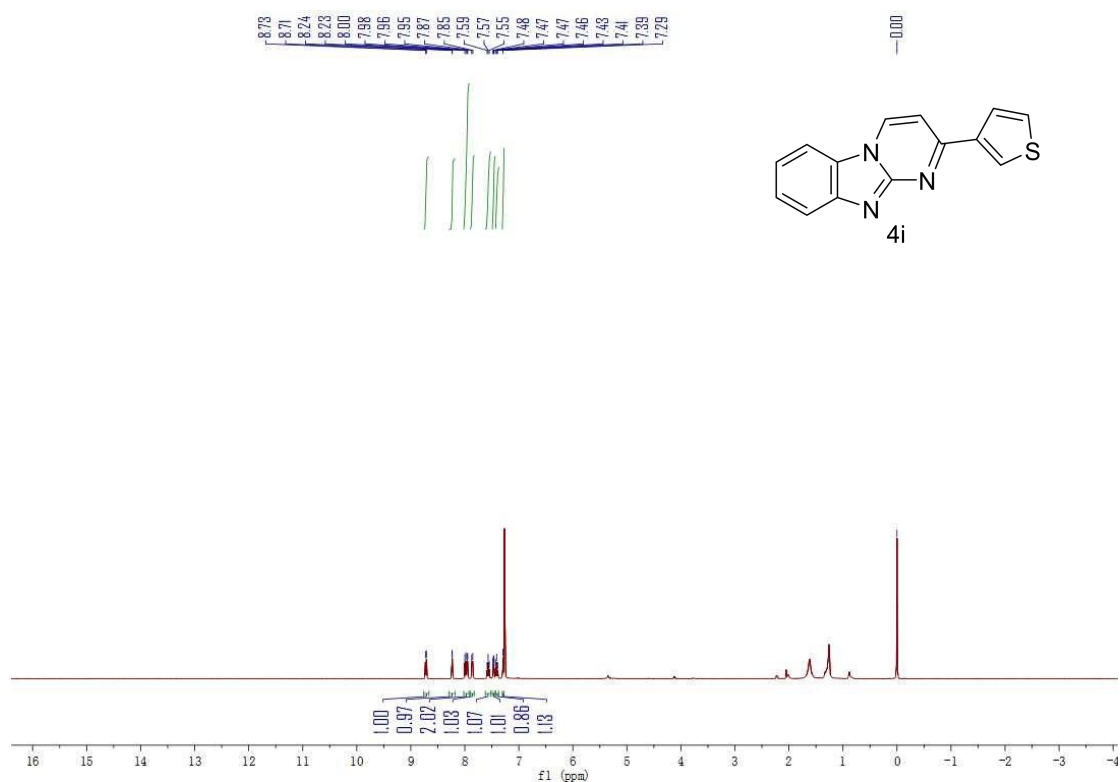
¹H NMR of Compound 4h (400 MHz, CDCl₃)



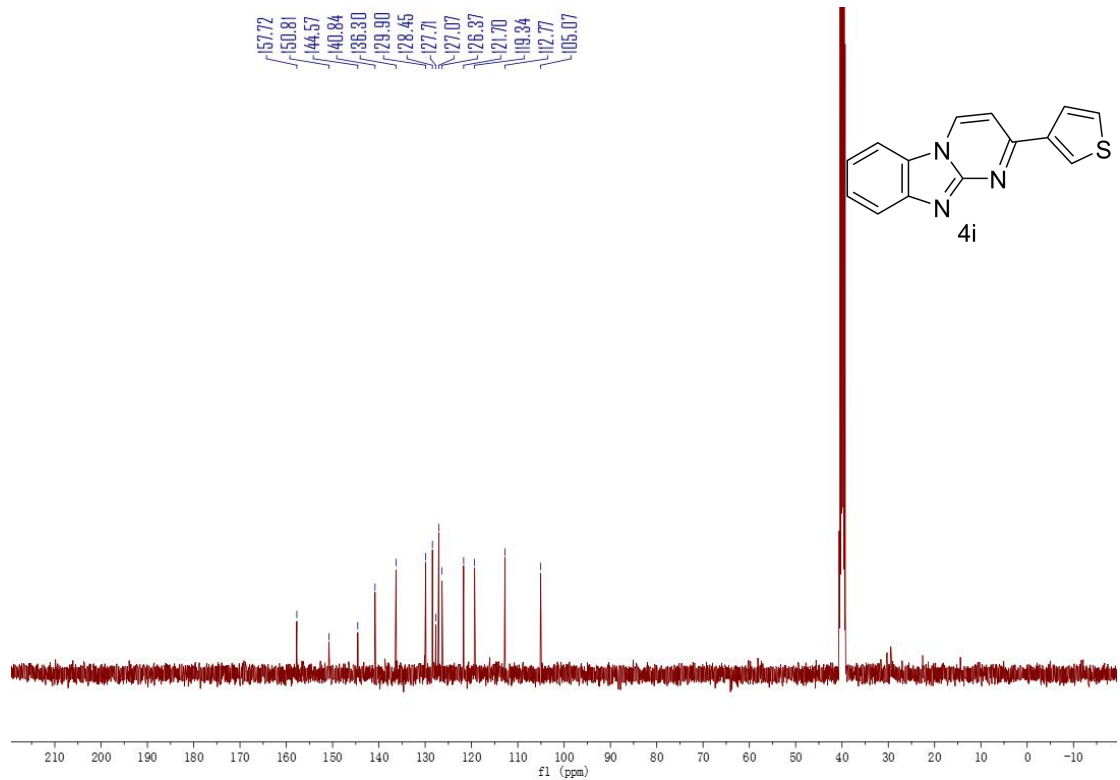
¹³C NMR of Compound 4h (400 MHz, CDCl₃)



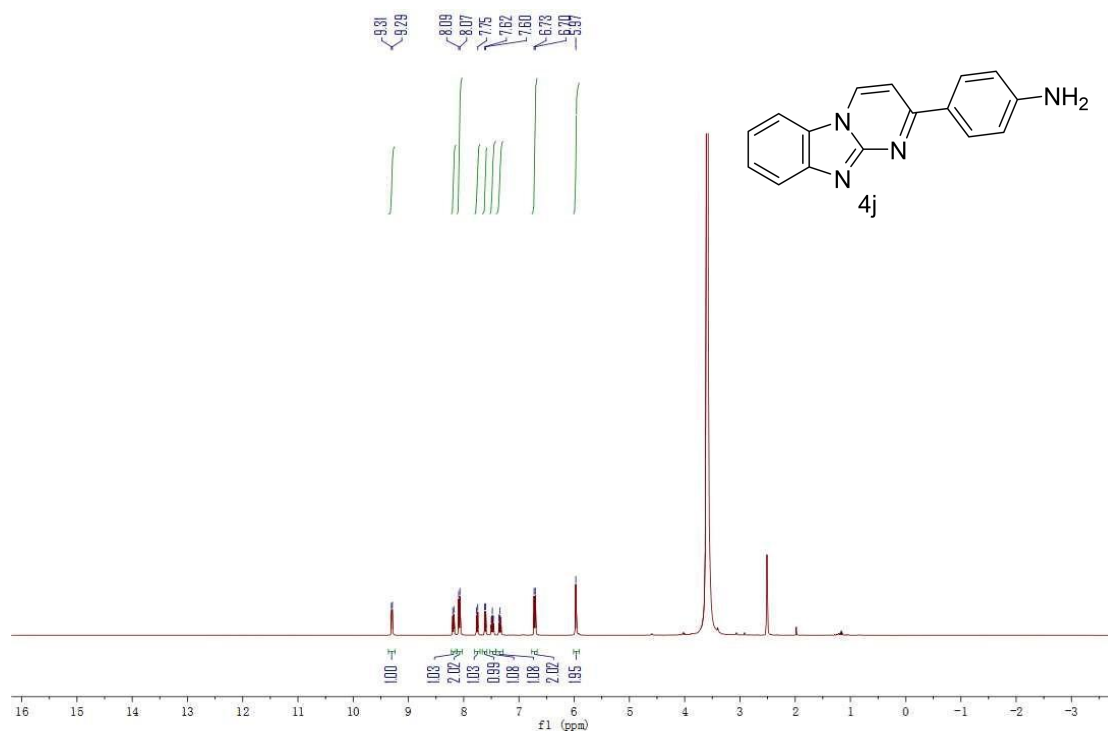
¹H NMR of Compound 4i (400 MHz, CDCl₃)



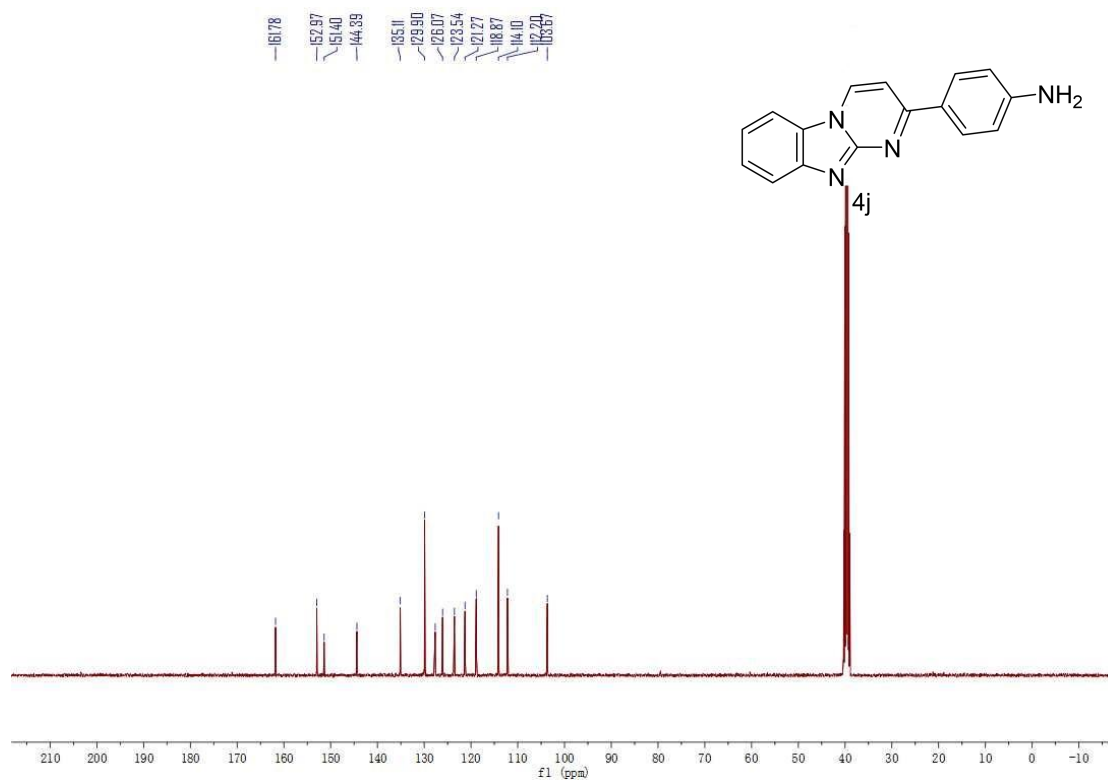
¹³C NMR of Compound 4i (400 MHz, DMSO-*d*₆)



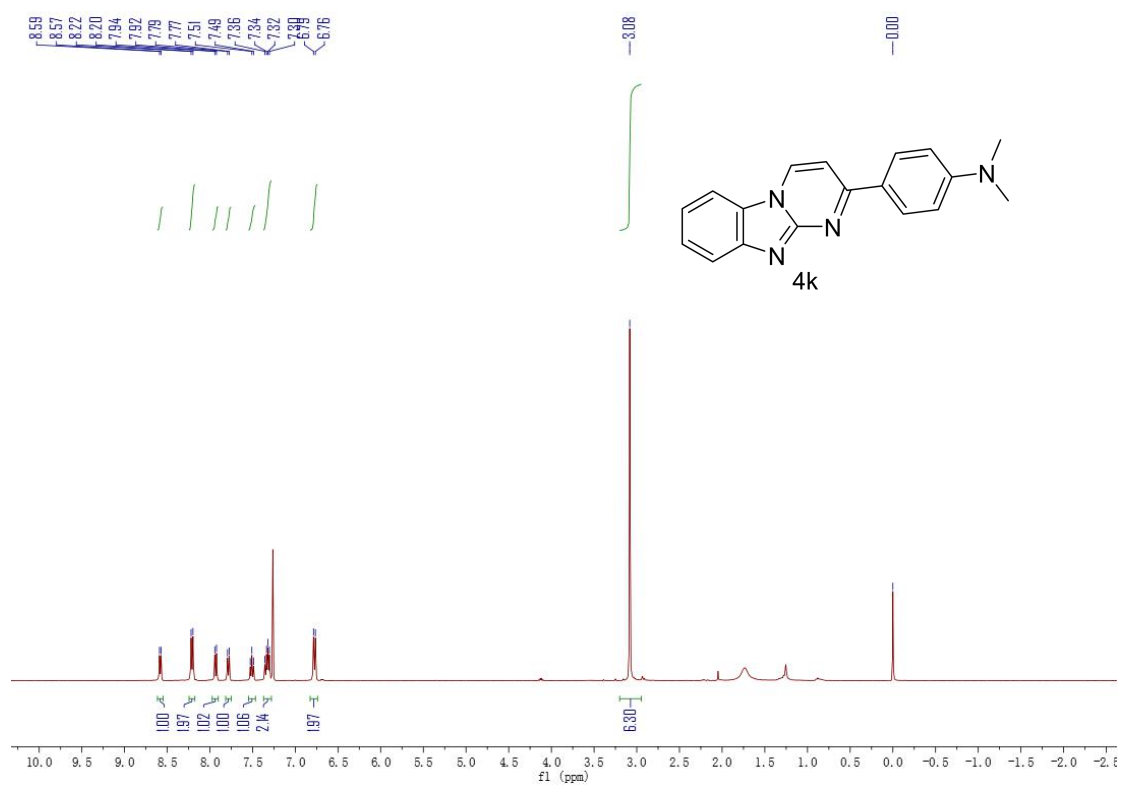
¹H NMR of Compound 4j (400 MHz, DMSO-*d*₆)



¹³C NMR of Compound 4j (400 MHz, DMSO-*d*₆)



¹H NMR of Compound 4k (400 MHz, CDCl₃)



¹³C NMR of Compound 4k (400 MHz, CDCl₃)

