

Supporting Information

Synthesis of Quinazolines via CuO nanoparticles Catalyzed Aerobic Oxidative Coupling of Aromatic Alcohols and Amidines

Wu Zhang,* Fei Guo, Fei Wang, Na Zhao, Liang Liu, Jia Li, Zhenghua Wang

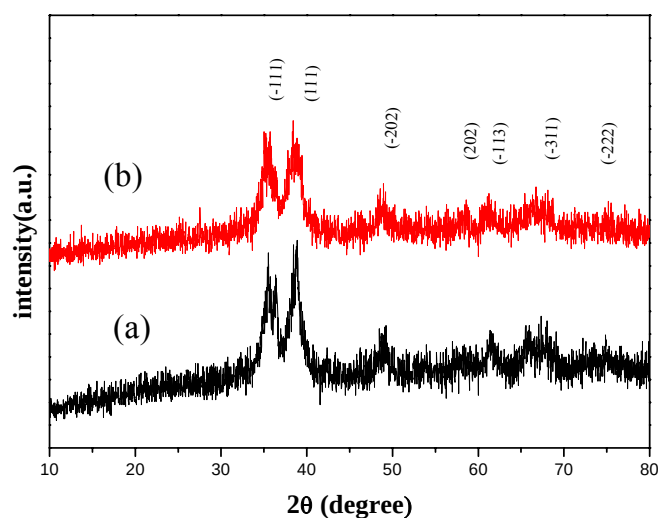
Key Laboratory of Functional Molecular Solids, Ministry of Education, Anhui
Laboratory of Molecule-Based Materials, College of Chemistry and Materials Science,
Anhui Normal University, Wuhu 241000, China. E-mail: zhangwu@mail.ahnu.edu.cn

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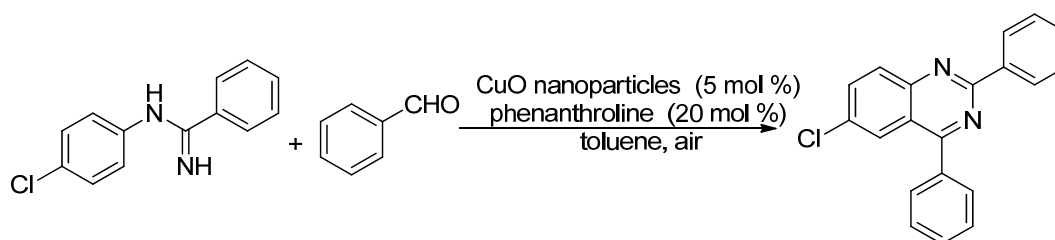
Catalyst Characterization

The obtained CuO nanoparticles were characterized by X-ray powder diffraction analysis (XRD). Figure S1a presents a typical XRD pattern of the as-prepared products. All of the detectable diffraction peaks can be indexed to (-111), (111), (-202), (202), (-113), (-311) and (-222) reflections of the cubic phase CuO (PCPDF card NO.80-1916).



Successive Test of Recycled Catalyst

Table S1.



test	1	2	3
yield (%)	88	83	79

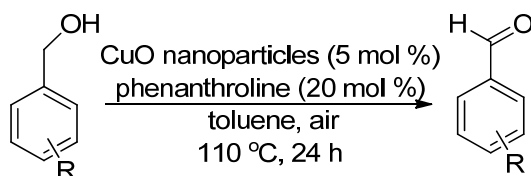
^aReaction condition: N-(chlorophenyl)benzimidamide (1 equiv), benzaldehyde (1.5 equiv), 5 mol % of CuO nanoparticles, 20 mol % phenanthroline, toluene (2 mL), under reflux in air for 24 h.

Recycling Procedure of the Catalysts: The separated precipitates in the above procedure were washed sufficiently with deionized water and ethanol three times each

and then dried under vacuum at 60 °C for 6 h, and then the CuO nanoparticles were recovered. The recyclability of the CuO nanoparticles was also tested as shown in Table. No significant decrease in the catalytic activity for the arylation of N-(chlorophenyl) benzimidamide with benzaldehyde was observed for the recovered CuO nanoparticles. The slight decrease in activity of the catalyst was due to the loss of catalyst during the separating process. XRD of the catalysts after the reaction demonstrated that the catalysts did not change during the reaction process.

Aerobic Oxidation of Alcohols to Aldehydes

Table S2 Aerobic oxidation of alcohols to aldehydes using CuO nanoparticles as catalyst.



entry	R	yield
1	R=H	95%
2	R=4-Cl	98%
3	R=4-OMe	90%
4	R=4-CH ₃	96%
5	R=4-NO ₂	85%
6	R=3-NO ₂	78%
7	R=2-NO ₂	76%

Synthesis of [(phen)Cu(II)] complexes¹

199 mg of copper acetate (1 mmol) and 360 mg of phenanthroline (2 mmol) were added into the 50 mL of ethanol, the mixture was stirred at 80 °C for 3 h. The solution was cooled to 0 °C, and a blue powder was got, which was recrystallized from ethanol to give blue crystals after 24 h. Melting point is 213 °C.

Characterization data for the products

Compounds 3ca, 3cb, 3cg, 3cf, 3cd, 3da, 3ja, 3fa, 3ga (from Table 3) provided spectral data that are agreement with the corresponding products from Table 2.

6-chloro- 2,4-diphenylquinazoline² (3aa)

White solid; yield 88%; m.p.191-193°C; Lit.m.p:185-186°C, ¹H NMR (300 MHz,

CDCl₃) δ 8.64-8.61 (m, 2H), 8.12-8.03 (m, 2H), 7.81-7.75 (m, 3H), 7.57-7.45 (m, 6H); ¹³C NMR (75 MHz, CDCl₃) δ 167.6, 160.4, 150.4, 137.7, 137.1, 134.6, 132.7, 130.8, 130.8, 130.3, 130.1, 128.8, 128.7, 128.6, 125.8, 122.2.

6-fluoro-2,4-diphenylquinazoline³ (3ba)

White solid; yield 86%; m.p.179-180°C; Lit.m.p:172-173°C, ¹H NMR (500 MHz, CDCl₃) δ 8.69-8.67 (m, 2H), 8.21-8.19 (m, 1H), 7.89-7.87 (m, 2H), 7.77-7.75 (m, 1H), 7.70-7.61 (m, 4H), 7.56-7.50 (m, 3H); ¹³C NMR (125 MHz, CDCl₃) δ 167.9 (d, *J* = 5.4 Hz), 161.4, 159.8 (d, *J* = 2.1 Hz), 159.4, 149.0, 137.8, 137.3, 131.7 (d, *J* = 8.5 Hz), 130.7, 130.2, 130.0, 128.7, 128.6, 124.0 (d, *J* = 25.5 Hz), 122.1 (d, *J* = 8.9 Hz), 110.4 (d, *J* = 23.1Hz).

2,4-diphenylquinazoline⁴ (3ca)

Yellow solid; yield 84%; m.p.116 °C; Lit.m.p:118-120°C, ¹H NMR (300 MHz, CDCl₃) δ 8.75-8.73 (m, 2H), 8.32-8.31 (m, 1H), 8.16 (d, *J* = 8.1 Hz, 1H), 7.96-7.90 (m, 3H), 7.63-7.54 (m, 7H); ¹³C NMR (75 MHz, CDCl₃) δ 168.3, 160.2, 152.0, 138.2, 137.7, 133.5, 130.5, 130.2, 129.9, 129.2, 128.7, 128.5, 127.0, 121.7.

6-methyl-2,4-diphenylquinazoline⁵ (3da)

White solid; yield 98%; m.p.178-180 °C; Lit.m.p:226-227°C, ¹H NMR (300 MHz, CDCl₃) δ 8.68 (d, *J* = 6.3 Hz, 2H), 8.11 (d, *J* = 7.6 Hz, 1H), 7.88-7.87 (m, 3H), 7.74 (d, *J* = 7.9 Hz, 1H), 7.62-7.51 (m, 6H), 2.52(s, 3H); ¹³C NMR (75 MHz, CDCl₃) δ 167.6, 159.5, 150.4, 138.2, 137.8, 137.2, 135.9, 130.4, 130.1, 129.8, 128.8, 128.6, 128.5, 125.6, 121.6, 21.9.

8-methyl-2,4-diphenylquinazoline (3ea)

Red solid; yield 94%; m.p.119-120 °C; ¹H NMR (300 MHz, CDCl₃) δ 8.73-8.66 (m, 2H), 7.88 (d, *J* = 8.3 Hz, 1H), 7.81-7.78 (m, 2H), 7.66 (d, *J* = 7.2 Hz, 1H), 7.52-7.33 (m, 7H), 2.85(s, 3H); ¹³C NMR (75 MHz, CDCl₃) δ 168.4, 159.0, 151.0, 138.6, 138.1, 137.5, 133.3, 130.4, 130.2, 129.7, 129.0, 128.6, 128.4, 126.4, 124.7, 121.6, 17.6. HRMS (ESI) calcd for C₂₁H₁₇N₂ ([M+H]⁺) 297.1392, found 297.1386.

6-methoxy-2,4-diphenylquinazoline⁶ (3fa)

Yellow solid; yield 87%; m.p.140-142 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.58 (d, J = 6.6Hz, 2H), 8.01 (d, J = 9.3 Hz, 1H), 7.84-7.83 (m, 2H), 7.54-7.43 (m, 7H), 7.33-7.32 (m, 1H), 3.79 (s, 3H); ^{13}C NMR (75MHz, CDCl_3) δ 166.7, 158.6, 158.1, 148.0, 138.2, 138.0, 130.6, 130.2, 129.9, 129.8, 128.6, 128.5, 128.3, 126.3, 122.4, 104.4, 55.6.

4-phenyl-2-(p-tolyl)quinazoline⁶(3ga)

White solid; yield 98%; m.p.162-163 °C; Lit.m.p:166-168°C, ^1H NMR (300 MHz, CDCl_3) δ 8.52 (d, J = 7.8 Hz, 2H), 8.06-8.02 (m, 2H), 7.81-7.77(m, 3H), 7.52-7.42(m, 4H), 7.26 (d, J = 7.8 Hz, 2H), 2.37 (s, 3H); ^{13}C NMR (75MHz, CDCl_3) δ 167.2, 159.2, 150.9, 139.8, 136.7, 134.3, 132.5, 129.2, 128.9, 128.3, 128.0, 127.6, 127.5, 126.0, 125.8, 120.6, 20.5.

4-phenyl-2-(m-tolyl)quinazoline⁶(3ha)

White solid; yield 95%; m.p.106 °C; Lit.m.p:115-117°C, ^1H NMR (300 MHz, CDCl_3) δ 8.42 (d, J = 6.9 Hz, 2H), 8.11-8.04 (m, 2H), 7.84-7.80 (m, 3H), 7.54-7.45 (m, 4H), 7.38-7.33 (m, 1H), 7.25 (d, J = 7.5 Hz, 1H) 2.42(s, 3H); ^{13}C NMR (75 MHz, CDCl_3) δ 168.3, 160.4, 152.0, 138.2, 138.1, 137.7, 133.6, 131.4, 130.2, 129.9, 129.2, 129.1, 128.6, 128.5, 127.0, 126.9, 125.9, 121.7, 21.6.

4-phenyl-2-(o-tolyl)quinazoline⁶(3ia)

White solid; yield 92%; m.p.67-69 °C; Lit.m.p:72-74°C, ^1H NMR (300MHz, CDCl_3) δ 8.22-8.16 (m, 2H), 7.99-7.85 (m, 4H), 7.63-7.58 (m, 4H), 7.36-7.31 (m, 3H); ^{13}C NMR (75 MHz, CDCl_3) δ 168.1, 163.4, 151.6, 138.7, 137.4, 133.7, 131.3, 130.8, 130.2, 130.0, 129.3, 129.0, 128.6, 127.4, 127.0, 126.0, 121.0, 21.3.

2-(4-chlorophenyl)-4-phenylquinazoline⁶(3ja)

Yellow solid; yield 93%; m.p.187 °C; Lit.m.p:190-192°C, ^1H NMR (300 MHz, CDCl_3) δ 8.58 (d, J = 8.5 Hz, 2H), 8.08 (t, J = 9.3 Hz, 2H), 7.85-7.79 (m, 3H), 7.54-7.40 (m, 6H); ^{13}C NMR (75 MHz, CDCl_3) δ 167.4, 158.1, 150.8, 136.5, 135.7, 135.6, 132.7, 129.1, 129.0, 128.9, 128.0, 127.7, 127.5, 126.2, 126.0, 120.7.

2-(2-chlorophenyl)-4-phenylquinazoline (3ka)

Yellow solid; yield 89%; m.p.53-54°C; ^1H NMR (300 MHz, CDCl_3) δ 8.11 (d, J = 9.1

Hz, 2H), 7.87-7.78 (m, 4H), 7.57-7.44 (m, 5H), 7.35-7.27 (m, 2H). ^{13}C NMR (75 MHz, CDCl_3) δ 168.3, 161.3, 151.6, 138.5, 137.2, 133.9, 133.1, 131.8, 130.6, 130.2, 130.2, 130.1, 129.1, 128.7, 127.8, 127.1, 126.9, 121.4. HRMS(ESI) calcd for $\text{C}_{20}\text{H}_{14}\text{N}_2\text{Cl}$ ($[\text{M}+\text{H}]^+$) 317.0845, found 317.0840.

2-(4-chlorophenyl)-6-methyl-4-phenylquinazoline (3la)

Yellow solid; yield 92%; m.p. 179-180 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.62 (d, J = 8.6 Hz, 2H), 8.04 (d, J = 8.6 Hz, 1H), 7.87-7.85 (m, 3H), 7.75-7.71 (m, 1H), 7.6-7.60 (m, 3H), 7.48 (d, J = 8.6 Hz, 2H), 2.52 (s, 3H); ^{13}C NMR (75 MHz, CDCl_3) δ 167.7, 158.5, 150.4, 137.7, 137.4, 136.5, 136.0, 130.1, 129.9, 128.8, 128.7, 128.6, 125.7, 121.7, 21.9. HRMS (ESI) calcd for $\text{C}_{21}\text{H}_{16}\text{N}_2\text{Cl}$ ($[\text{M}+\text{H}]^+$) 331.1002, found 331.0997.

6-chloro-4-phenyl-2-(p-tolyl)quinazoline² (3ma)

White solid; yield 90%; m.p. 197-199 °C; Lit. m.p. 216-218 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.50 (d, J = 8.2 Hz, 2H), 8.04-8.00 (m, 2H), 7.81-7.72 (m, 3H), 7.56-7.54 (m, 3H), 7.26 (d, J = 8.1 Hz, 2H), 2.37 (s, 3H); ^{13}C NMR (75 MHz, CDCl_3) δ 167.5, 160.5, 150.4, 141.1, 137.1, 135.0, 134.4, 132.3, 130.7, 130.2, 130.1, 129.4, 128.7, 128.6, 125.8, 122.1, 21.6.

2-(4-chlorophenyl)-7-methyl-4-phenylquinazoline (3na)

White solid; yield 91%; m.p. 202-203 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.66 (d, J = 8.4 Hz, 2H), 8.03 (d, J = 8.4 Hz, 2H), 7.89-7.86 (m, 2H), 7.61-7.39 (m, 6H), 2.62 (s, 3H); ^{13}C NMR (75 MHz, CDCl_3) δ 167.9, 159.3, 152.2, 144.7, 137.7, 136.8, 136.6, 130.1, 129.9, 129.5, 128.7, 128.5, 128.0, 126.7, 119.9, 22.1. HRMS (ESI) calcd for $\text{C}_{21}\text{H}_{16}\text{N}_2\text{Cl}$ ($[\text{M}+\text{H}]^+$) 331.1002, found 331.1002.

4-(4-chlorophenyl)-2-phenylquinazoline (3cb)

White solid; yield 95%; m.p. 143-145 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.66-8.63 (m, 2H), 8.25 (d, J = 7.8 Hz, 2H), 8.05-8.02 (m, 1H), 7.89-7.78 (m, 4H), 7.56-7.47 (m, 6H); ^{13}C NMR (75 MHz, CDCl_3) δ 167.1, 160.2, 152.0, 137.9, 136.3, 136.1, 133.7, 131.5, 130.7, 129.3, 128.9, 128.7, 128.6, 127.2, 126.6, 121.5. HRMS (ESI) calcd for $\text{C}_{20}\text{H}_{14}\text{N}_2\text{Cl}$ ($[\text{M}+\text{H}]^+$) 317.0845, found 317.0840.

4-(2-phenylquinazolin-4-yl)benzonitrile (3cc)

White solid; yield 91%; m.p.129-130 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.67(br, 2H), 8.24 (d, J = 8.4 Hz, 1H), 8.02-7.89 (m, 6H), 7.63-7.53 (m, 4H); ^{13}C NMR (75 MHz, CDCl_3) δ 166.3, 160.2, 152.0, 141.9, 137.6, 134.1, 132.3, 130.9, 130.8, 129.5, 128.7, 127.6, 126.1, 121.2, 118.4, 113.6. HRMS (ESI) calcd for $\text{C}_{21}\text{H}_{14}\text{N}_3$ ($[\text{M}+\text{H}]^+$) 308.1188, found 308.1182.

4-(4-nitrophenyl)-2-phenylquinazoline (3cd)

Yellow solid; yield 83%; m.p.179-180 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.61-8.59 (m, 2H), 8.40 (d, J = 8.1 Hz, 2H), 8.15 (d, J = 8.1 Hz, 1H), 8.00-7.85 (m, 4H), 7.56-7.46 (m, 4H); ^{13}C NMR (75MHz, CDCl_3) δ 165.9, 160.3, 152.1, 148.7, 143.7, 137.6, 134.1, 131.1, 130.9, 129.5, 128.6, 127.7, 126.0, 123.8, 121.3. HRMS (ESI) calcd for $\text{C}_{20}\text{H}_{14}\text{N}_3\text{O}_2$ ($[\text{M}+\text{H}]^+$) 328.1086, found 328.1081.

4-(furan-2-yl)-2-phenylquinazoline (3ce)

Yellow solid; yield 78%; m.p.72-74 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.83 (d, J = 8.4, 1H), 8.60-8.58 (m, 1H), 8.01(d, J = 8.4 Hz, 1H), 7.78-7.71 (m, 2H), 7.63-7.44 (m, 5H), 6.61(s, 1H); ^{13}C NMR (75 MHz, CDCl_3) δ 160.0, 155.2, 154.1, 152.6, 145.8, 138.1, 133.5, 130.5, 129.1, 128.5, 127.3, 126.7, 119.6, 118.5, 116.0, 112.4. HRMS (ESI) calcd for $\text{C}_{18}\text{H}_{13}\text{N}_2\text{O}$ ($[\text{M}+\text{H}]^+$) 273.1028, found 273.1022.

4-(4- methylphenyl)-2-phenylquinazoline⁶ (3cf)

Yellow solid; yield 96%; m.p.112-113 °C; Lit.m.p:128-130°C, ^1H NMR (300 MHz, CDCl_3) δ 8.71-8.68 (m, 2H), 8.16-8.13 (m, 2H), 7.91-7.79 (m, 3H), 7.53-7.40 (m, 6H), 2.50 (s, 3H); ^{13}C NMR (75 MHz, CDCl_3) δ 168.3, 160.2, 151.9, 140.2, 138.3, 134.9, 133.5, 130.5, 130.2, 129.3, 129.1, 128.7, 128.5, 127.1, 126.9, 121.7, 21.5.

4-(4-methoxyphenyl)-2-phenylquinazoline (3cg)

White solid; yield 85%; m.p.129-130 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.71 (d, J = 6.3 Hz, 2H), 8.18 (d, J = 8.1 Hz, 2H), 7.92-7.86 (m, 3H), 7.58-7.52 (m, 4H), 7.13 (d, J = 8.4 Hz, 2H), 3.94(s, 3H); ^{13}C NMR (75 MHz, CDCl_3) δ 167.8, 161.3, 160.1, 151.9, 138.2, 133.5, 131.9,130.5, 130.1, 129.0, 128.7,128.5, 127.1, 126.9, 121.6, 114.0, 55.5. HRMS (ESI) calcd for $\text{C}_{21}\text{H}_{17}\text{N}_2\text{O}$ ($[\text{M}+\text{H}]^+$)313.1341, found 313.1335.

4-(2-ethoxyphenyl)-2-phenylquinazoline (3ch)

White solid; yield 73%; m.p.125-126 °C; ^1H NMR(300MHz, CDCl_3) δ 8.61-8.57(m, 2H), 8.06-8.03 (m, 1H), 7.78-7.65 (m, 2H), 7.48-7.35 (m, 6H), 7.10-6.97 (m, 2H), 3.93 (q, $J = 6.9$ Hz, 2H), 0.98 (t, $J = 6.9$ Hz, 3H). ^{13}C NMR(75MHz, CDCl_3) δ 166.8, 159.5, 155.4, 149.9, 137.4, 132.3, 130.3, 129.9, 129.3, 127.7, 127.6, 127.4, 126.8, 126.2, 125.3, 121.8, 119.9, 111.3, 63.0, 13.5. HRMS(ESI) calcd for $\text{C}_{22}\text{H}_{19}\text{N}_2\text{O}$ ($[\text{M}+\text{H}]^+$) 327.1497, found 327.1492.

6-chloro-4-(4-chlorophenyl)-2-phenylquinazoline (3ab)

White solid; yield 92%; m.p.186-188 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.63-8.61 (m, 2H), 8.15 (d, $J = 9.0$ Hz, 1H), 7.99-7.98 (m, 1H), 7.81-7.75(m, 3H), 7.56-7.47(m, 5H); ^{13}C NMR (75 MHz, CDCl_3) δ 165.3, 159.4, 149.5, 136.5, 135.7, 134.5, 133.7, 131.9, 130.4, 129.9, 129.9, 128.1, 127.7, 127.6, 124.3, 121.0. HRMS (ESI) calcd for $\text{C}_{20}\text{H}_{13}\text{Cl}_2\text{N}_2$ ($[\text{M}+\text{H}]^+$) 351.0456, found 351.0450.

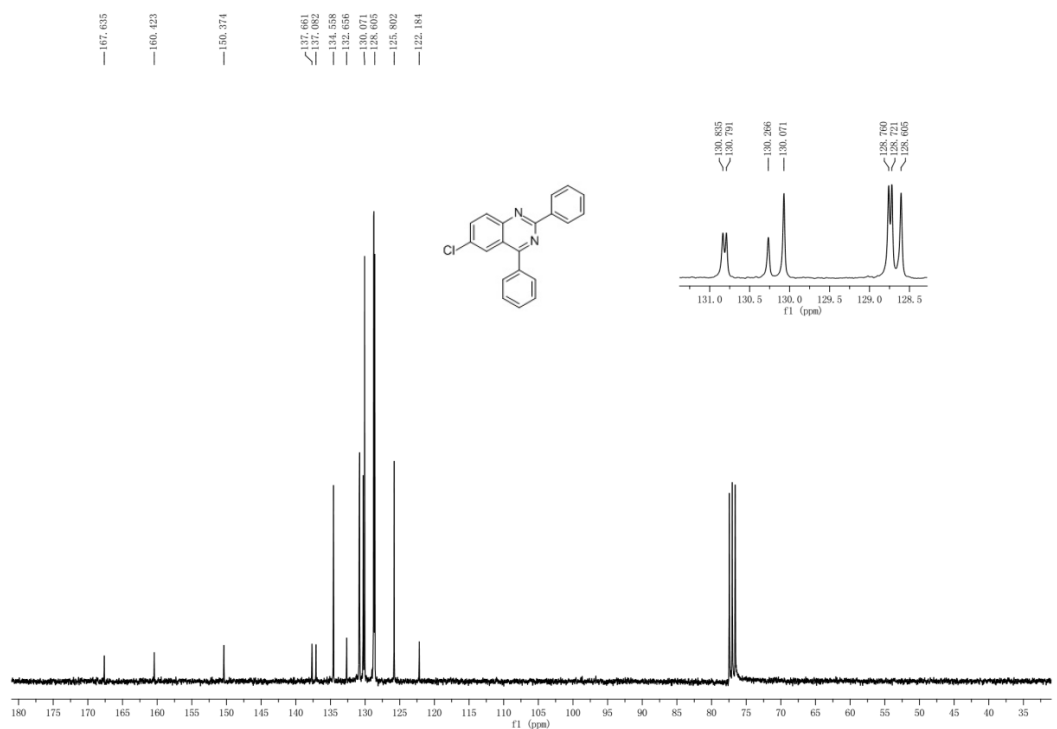
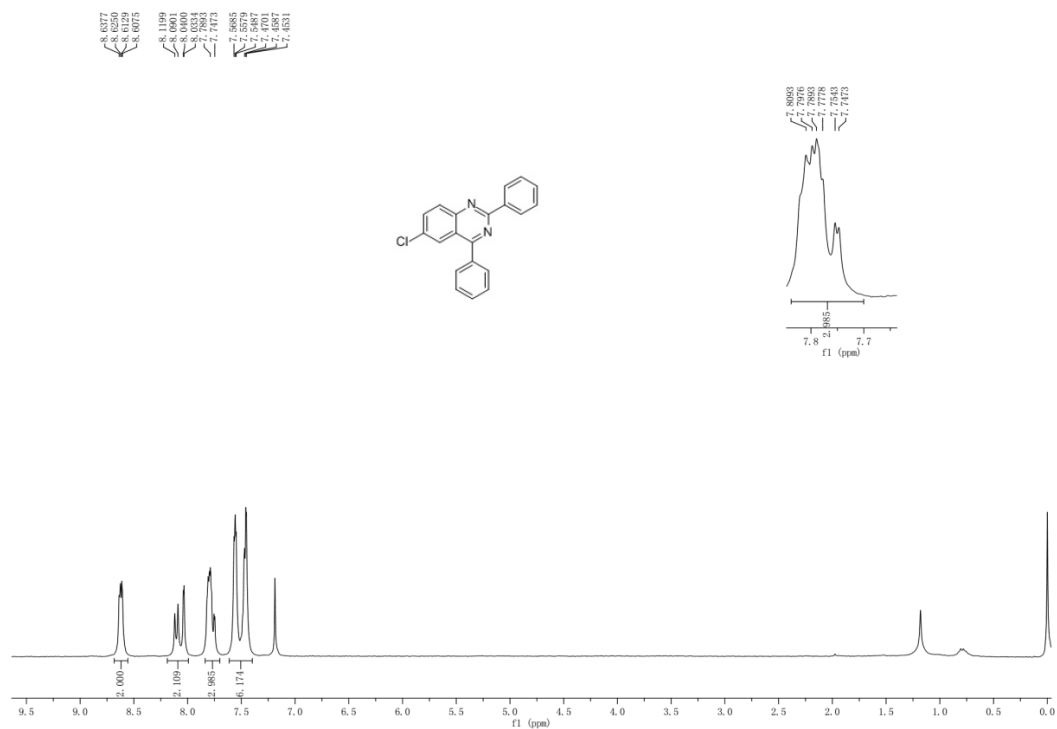
4-(3-nitrophenyl)-2-phenylquinazoline (3cj)

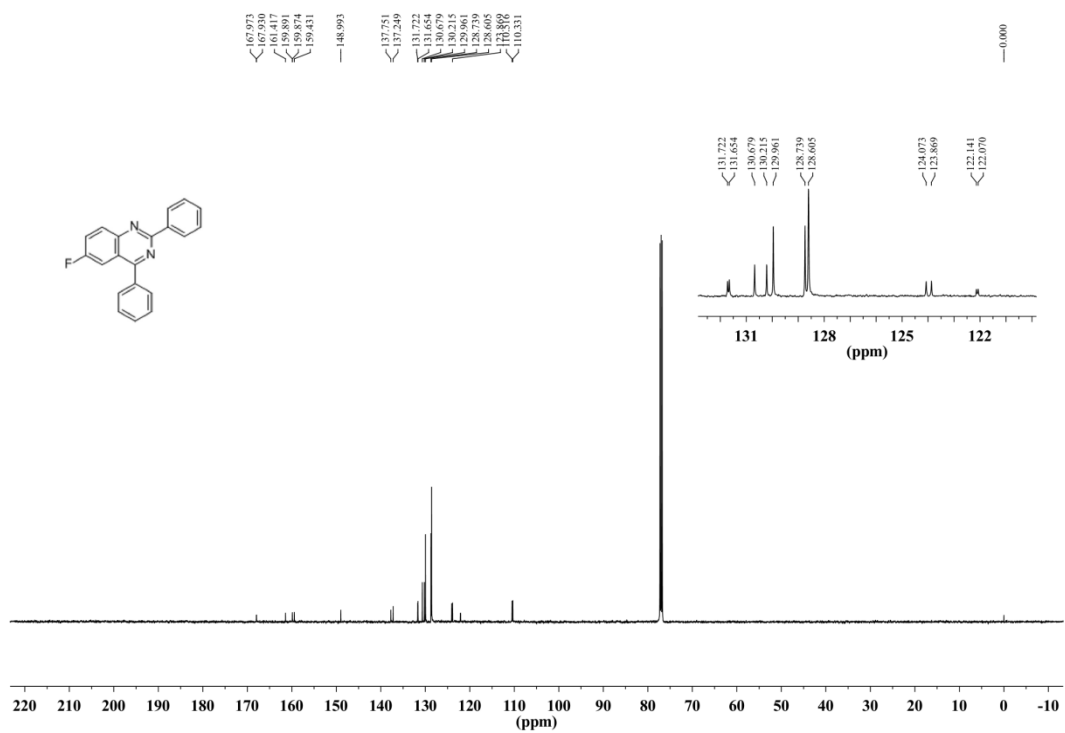
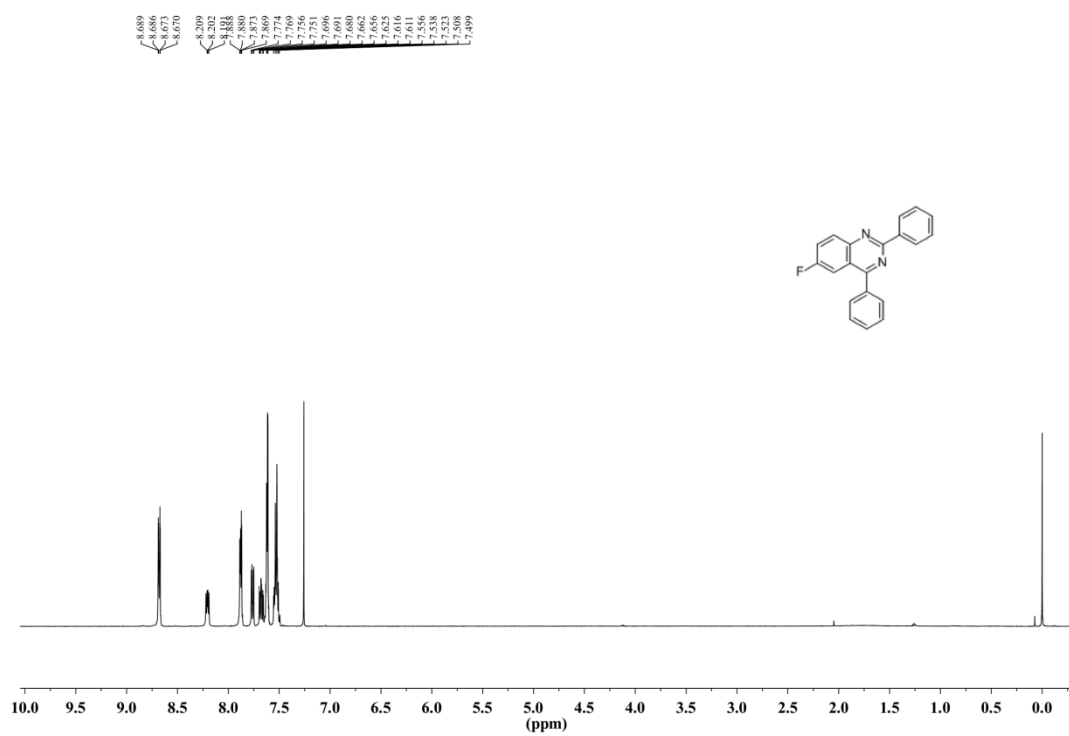
White solid; yield 76%; m.p.188-189 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.69-8.59 (m, 3H), 8.39-8.36 (m, 1H), 8.16-8.12 (m, 2H), 7.96-7.93 (m, 1H), 7.89-7.84 (m, 1H), 7.74-7.69 (m, 1H), 7.56-7.45 (m, 4H); ^{13}C NMR (75 MHz, CDCl_3) δ 168.6, 162.3, 161.5, 151.3, 136.2, 136.0, 129.5, 129.3, 129.3, 128.8, 128.3, 127.6, 124.6, 124.1, 114.3. HRMS(ESI) calcd for $\text{C}_{20}\text{H}_{14}\text{N}_3\text{O}_2$ ($[\text{M}+\text{H}]^+$) 328.1086, found 328.1081.

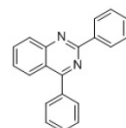
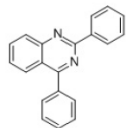
4-(2-nitrophenyl)-2-phenylquinazoline (3ck)

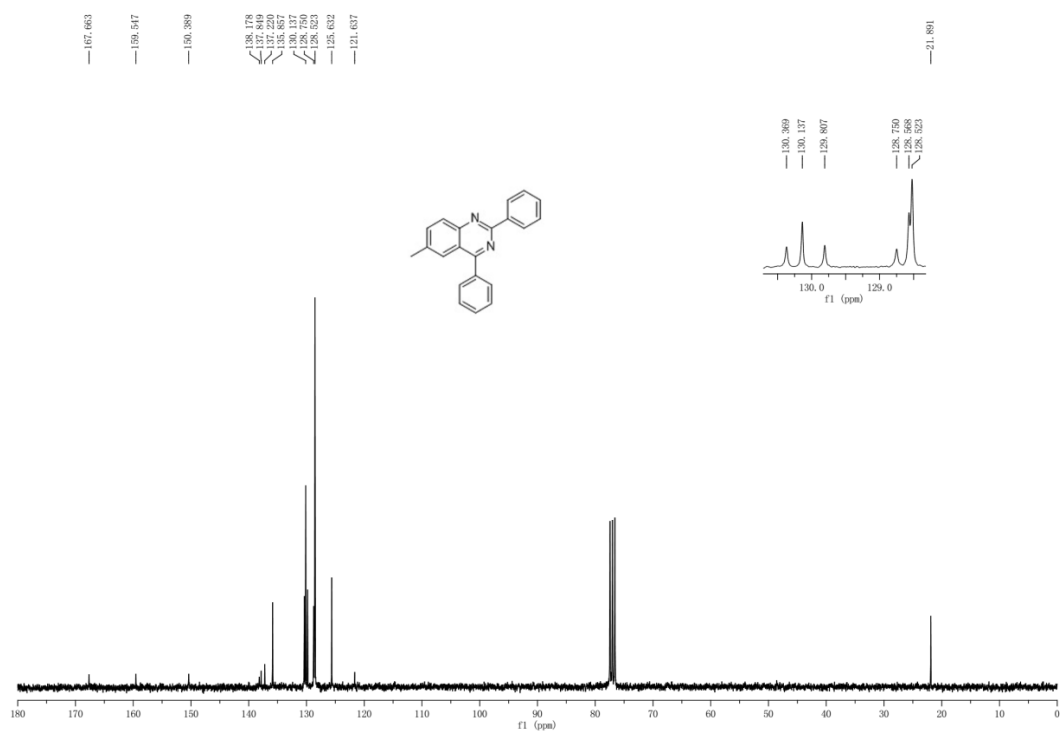
White solid; yield 73%; m.p.162-163 °C; ^1H NMR (300 MHz, CDCl_3) δ 8.60-8.57 (m, 2H), 8.28-8.12 (m, 2H), 7.93-7.63 (m, 5H), 7.54-7.49 (m, 4H); ^{13}C NMR (75 MHz, CDCl_3) δ 166.7, 160.7, 151.7, 138.1, 134.4, 133.6, 133.4, 132.3, 131.0, 130.7, 129.7, 129.1, 129.0, 127.9, 125.8, 125.3, 122.2. HRMS (ESI) calcd for $\text{C}_{20}\text{H}_{14}\text{N}_3\text{O}_2$ ($[\text{M}+\text{H}]^+$) 328.1086, found 328.1081.

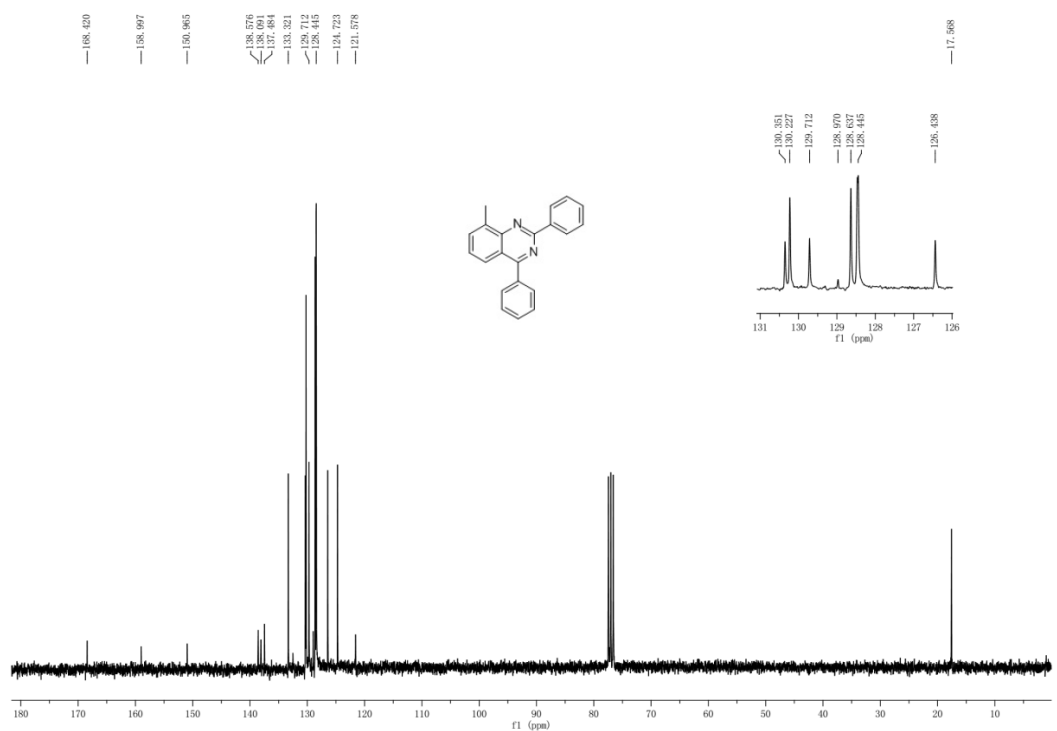
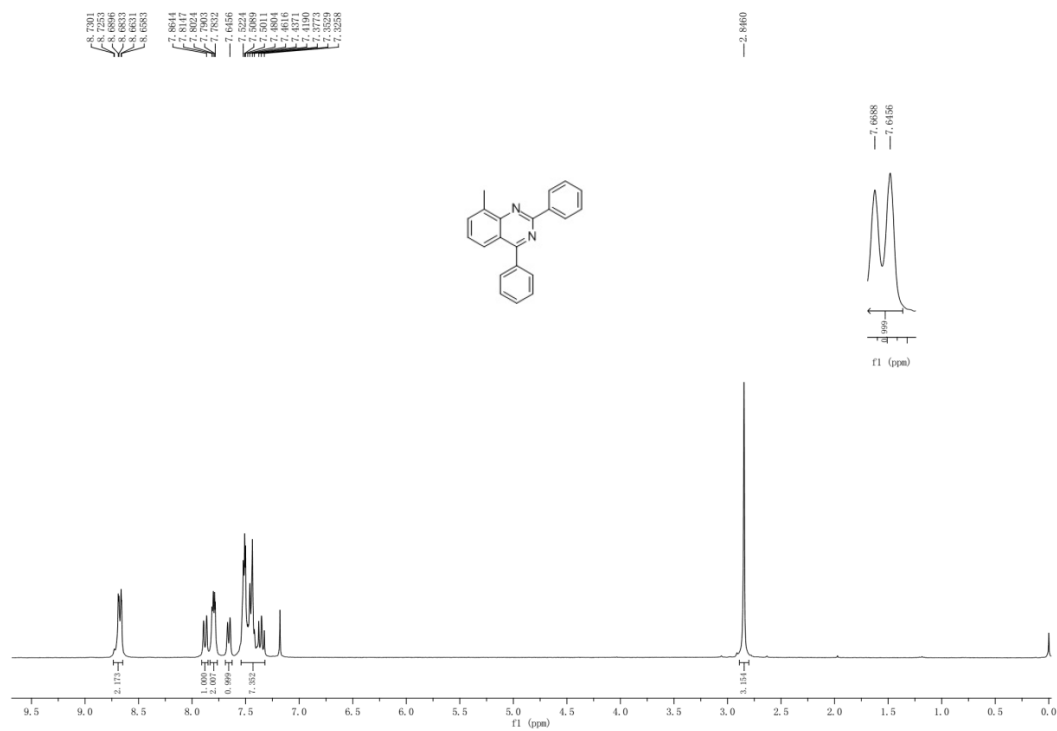
^1H and ^{13}C NMR spectra of the products.

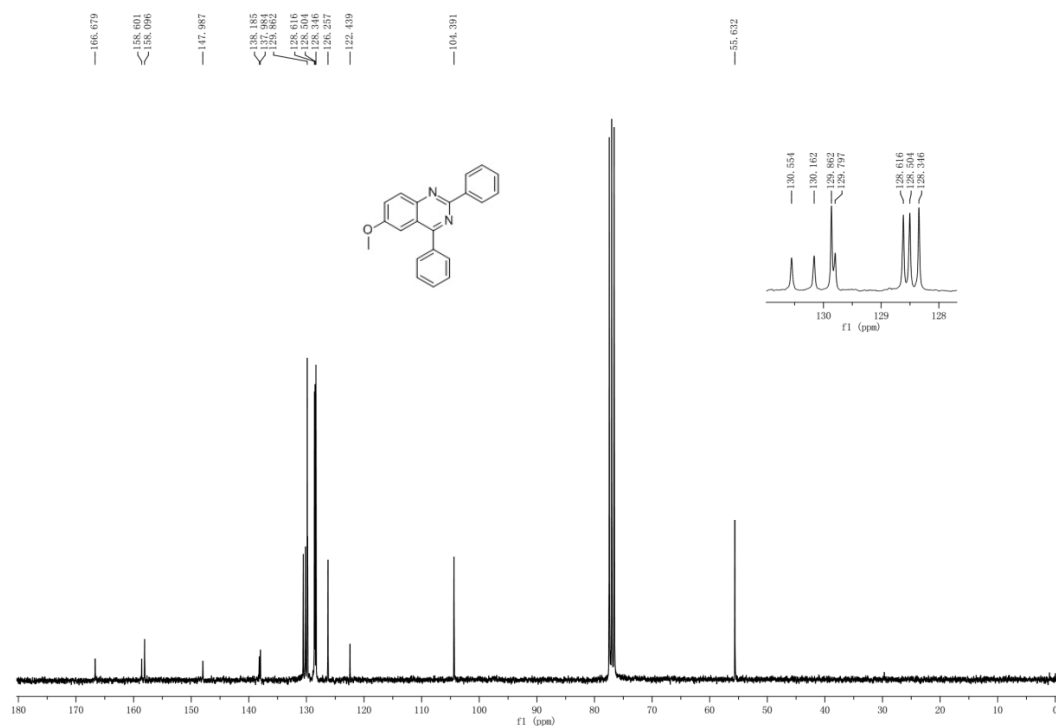
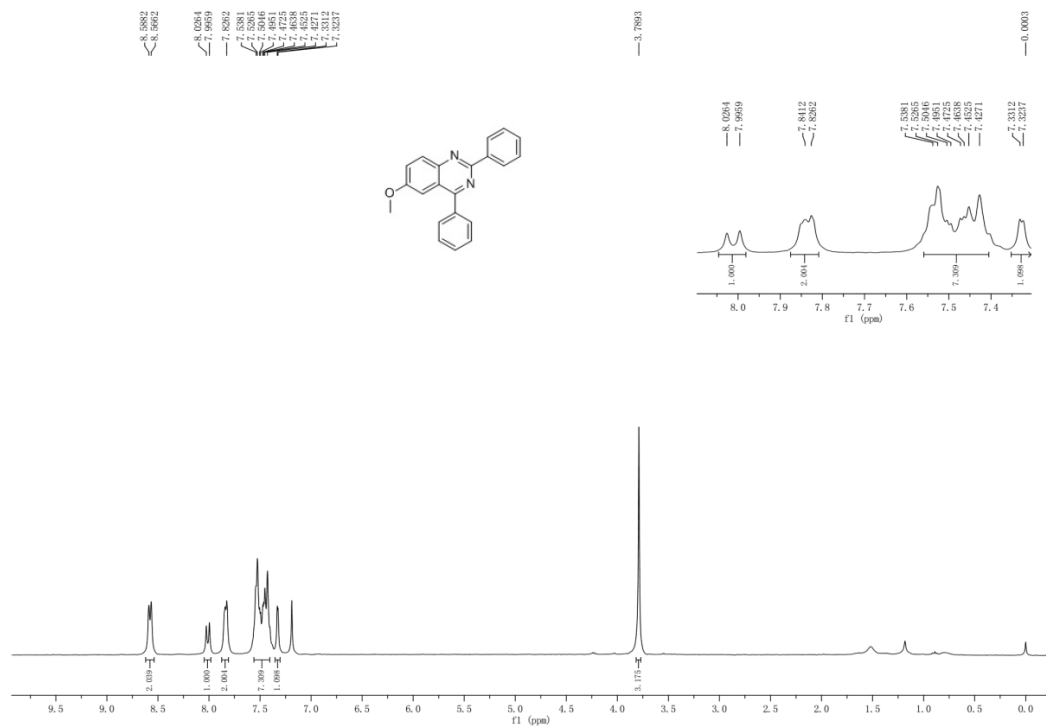


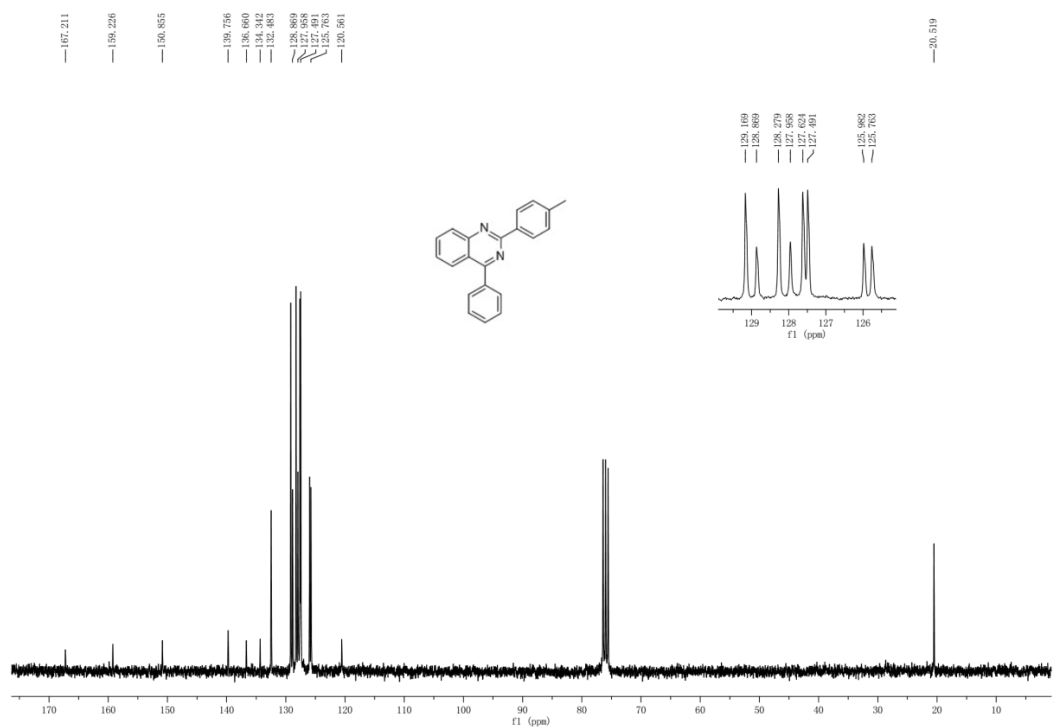
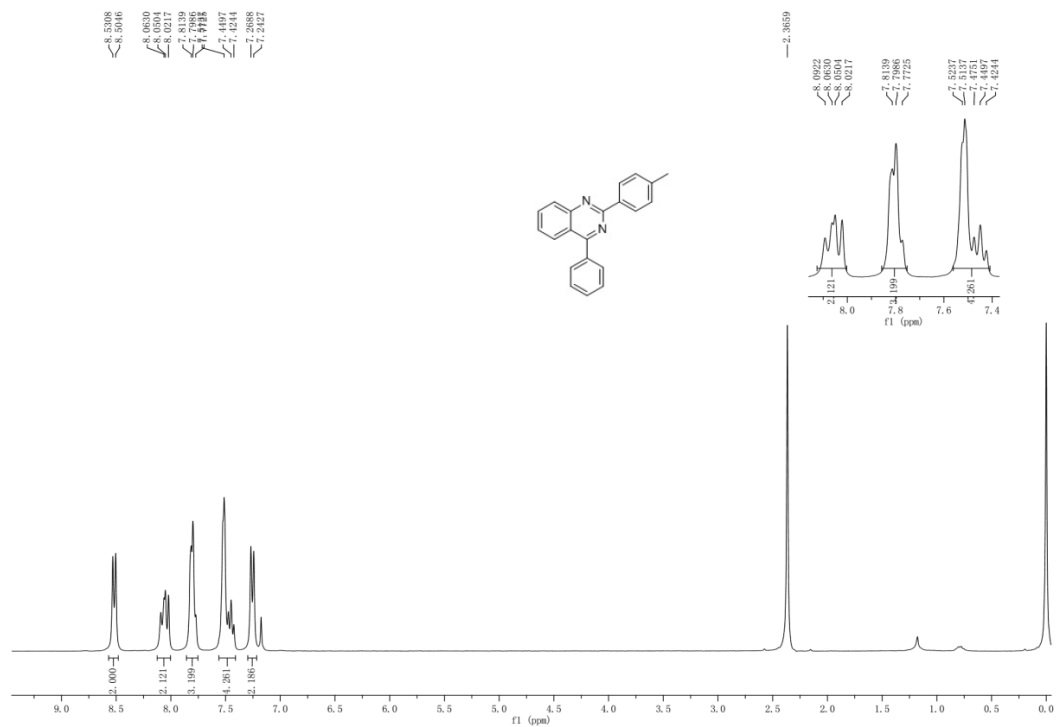


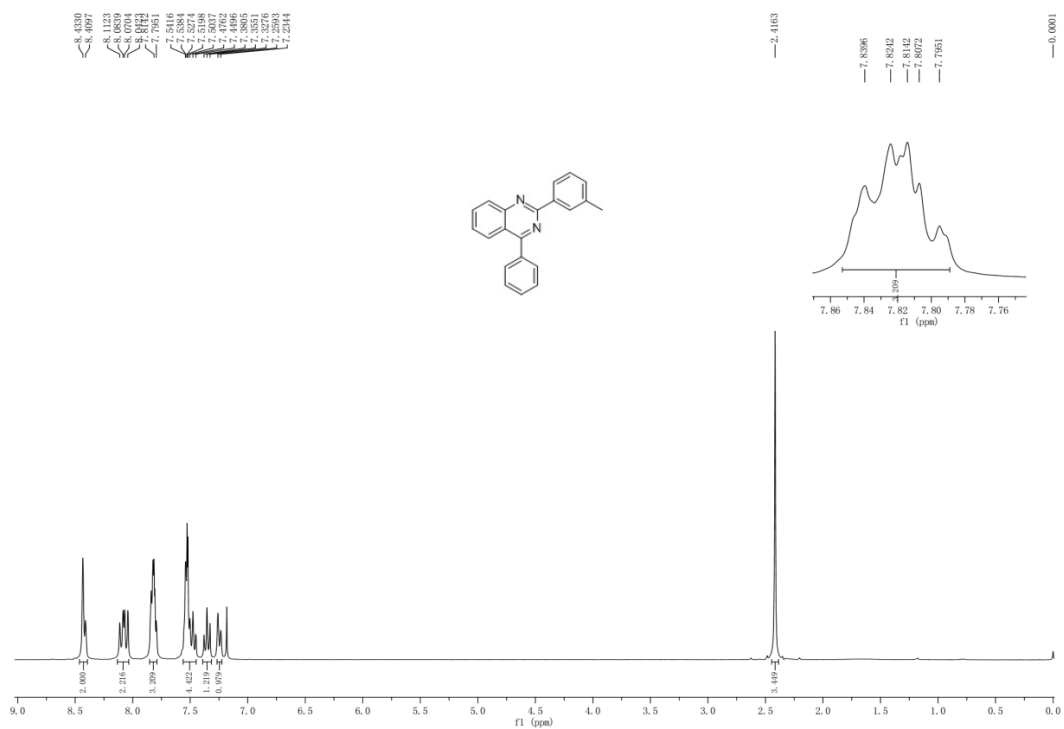


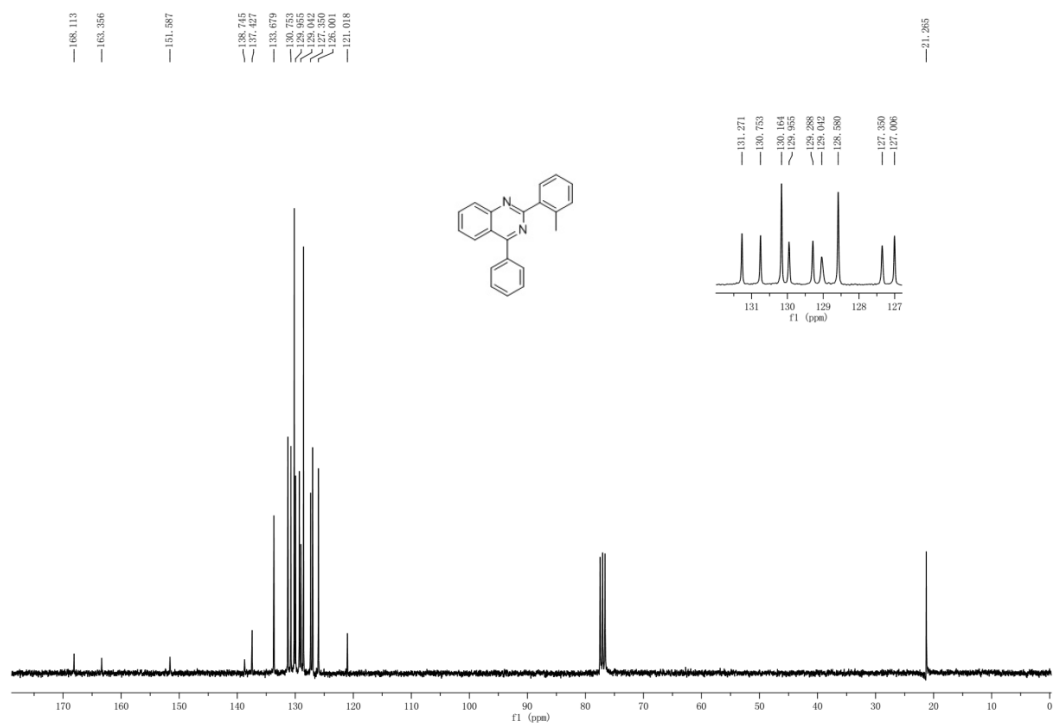
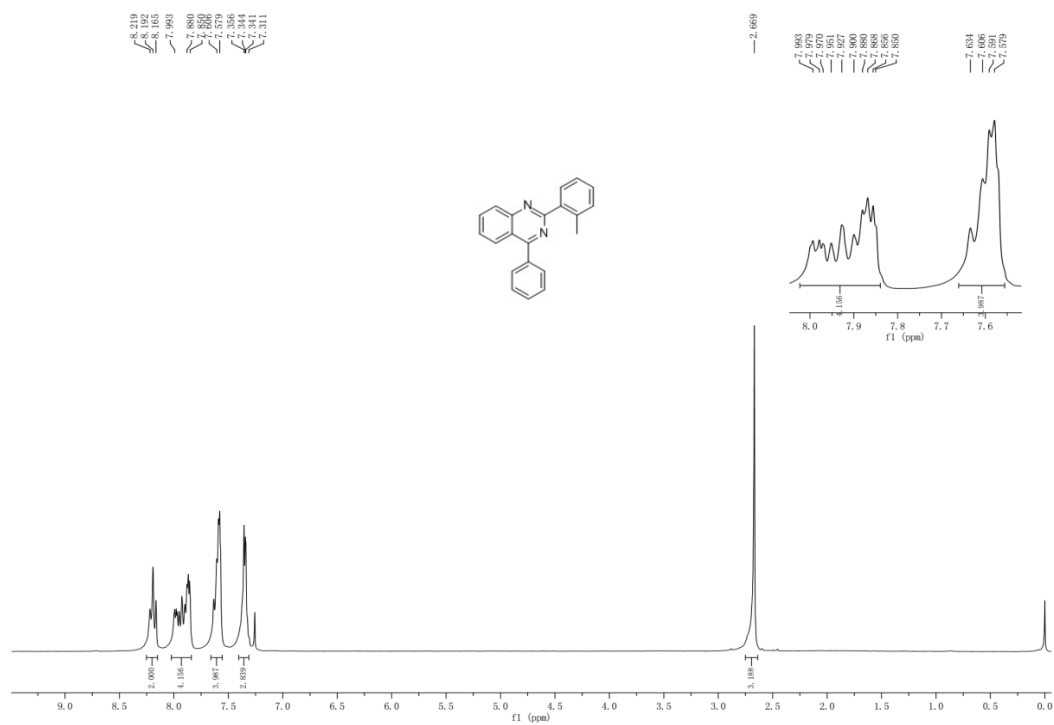


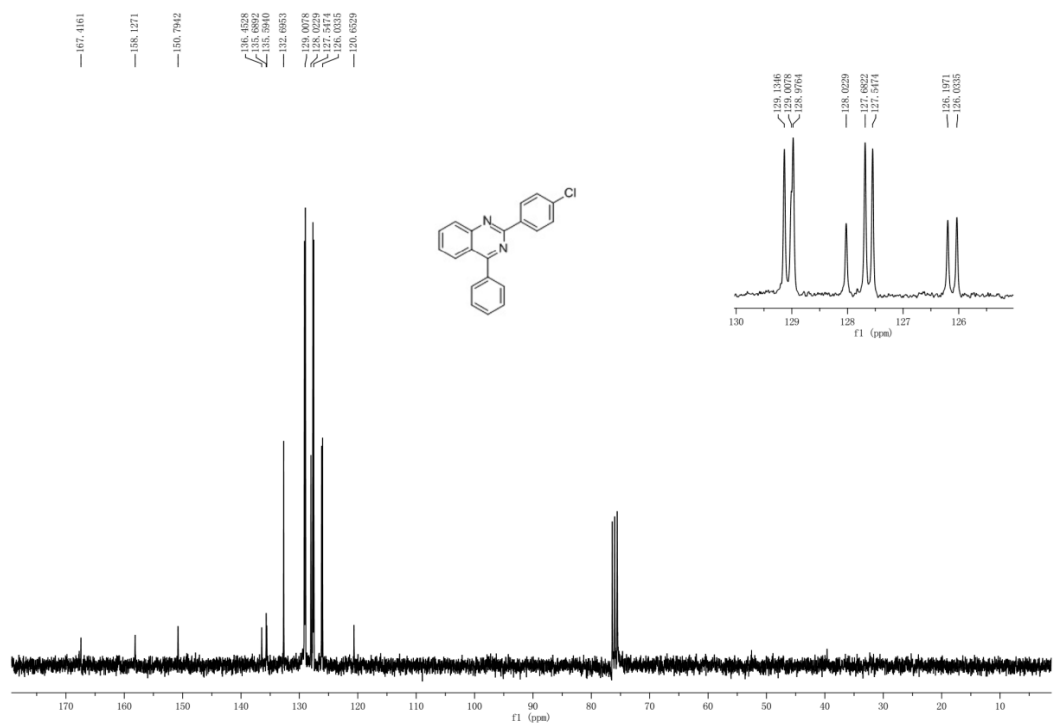
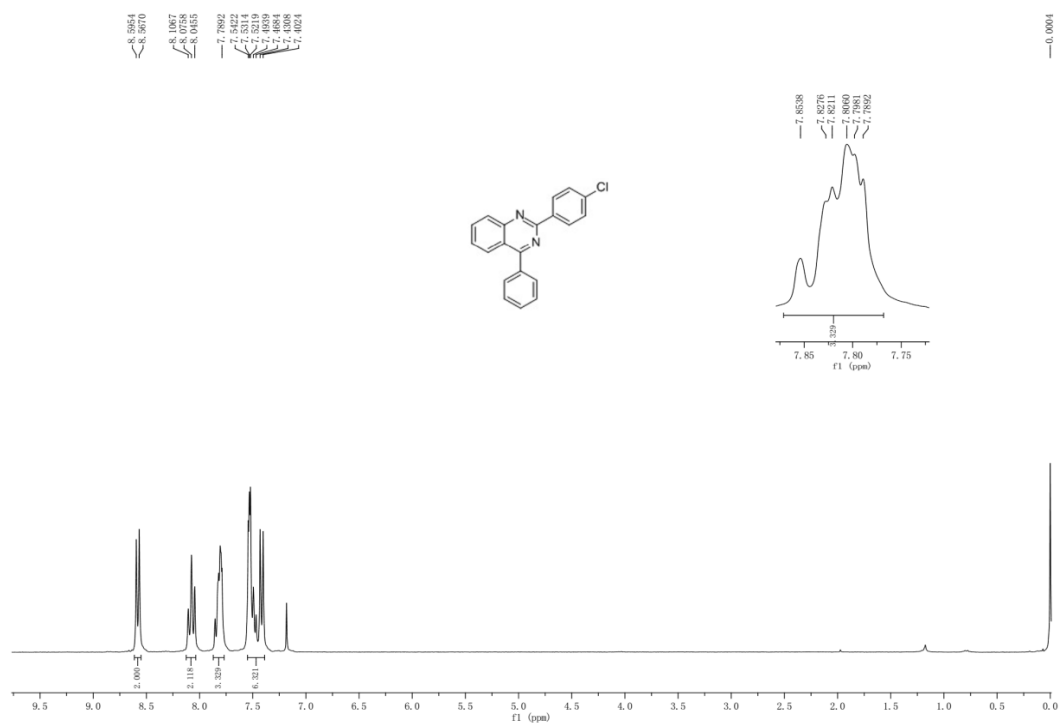


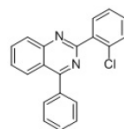


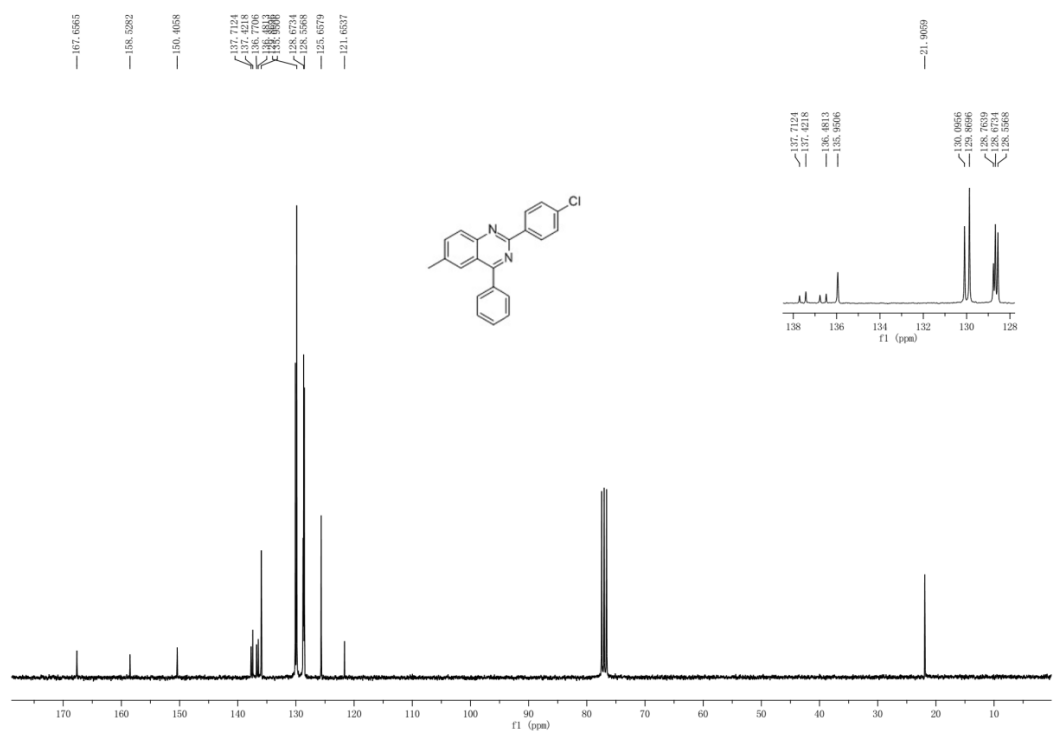
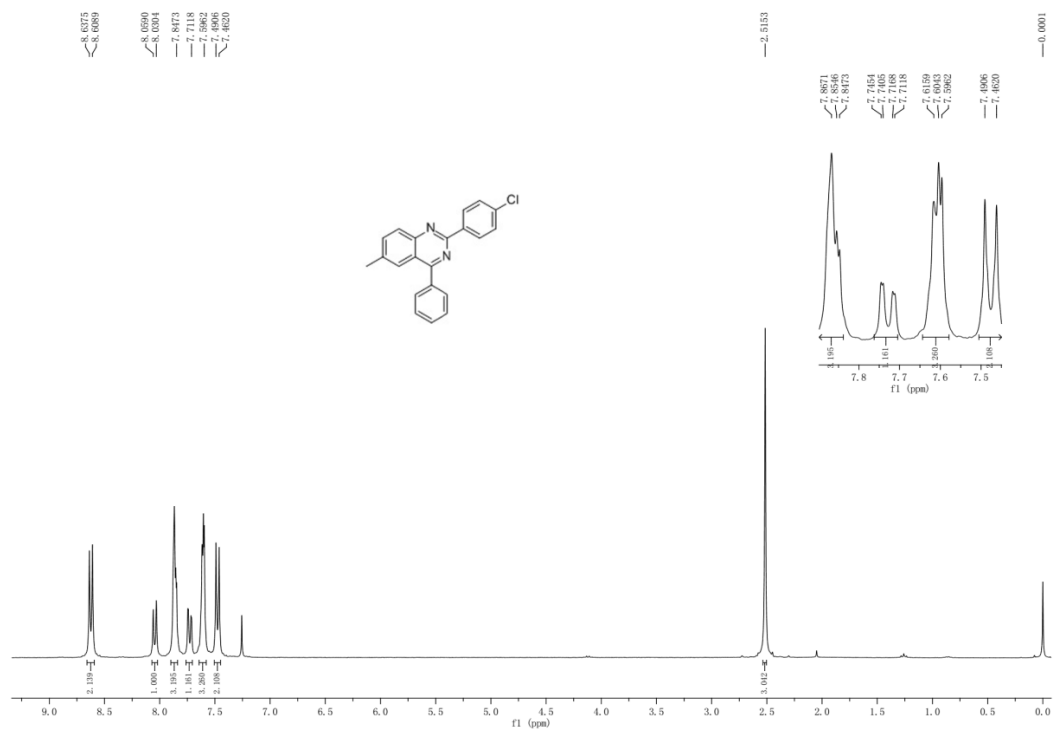


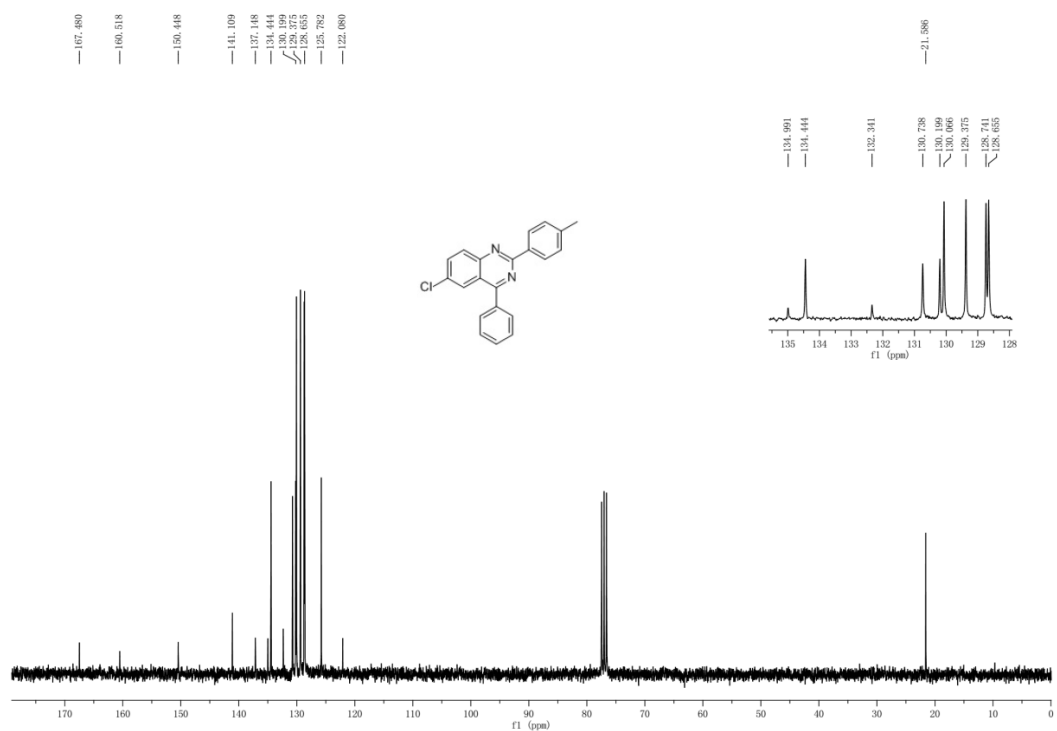
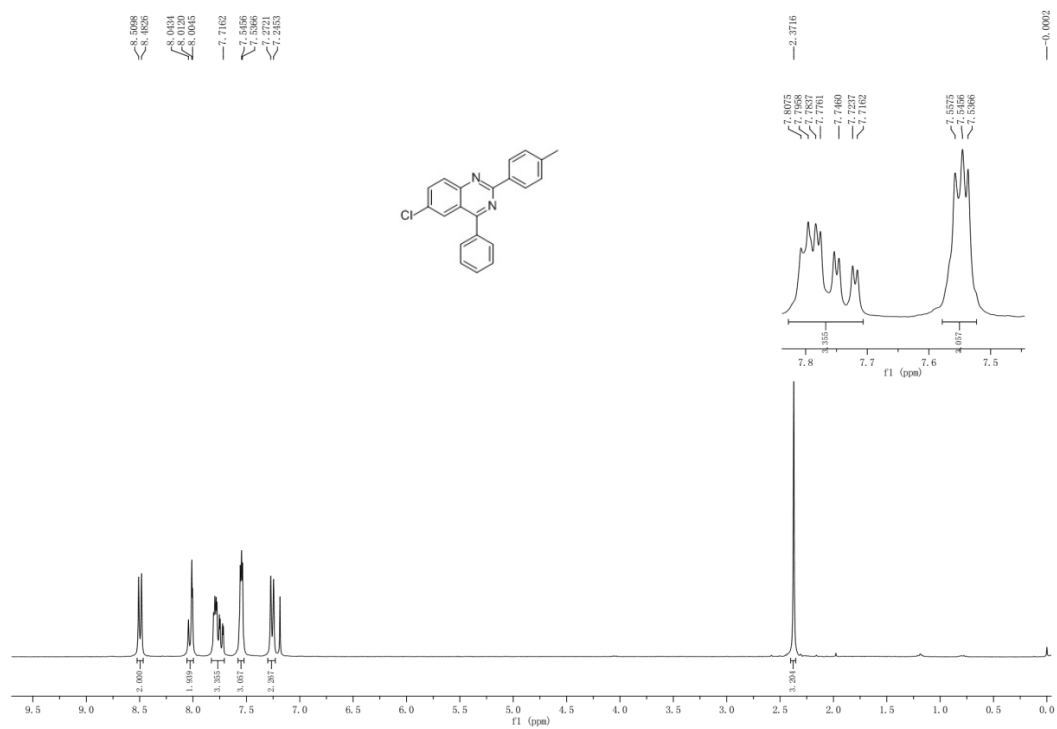


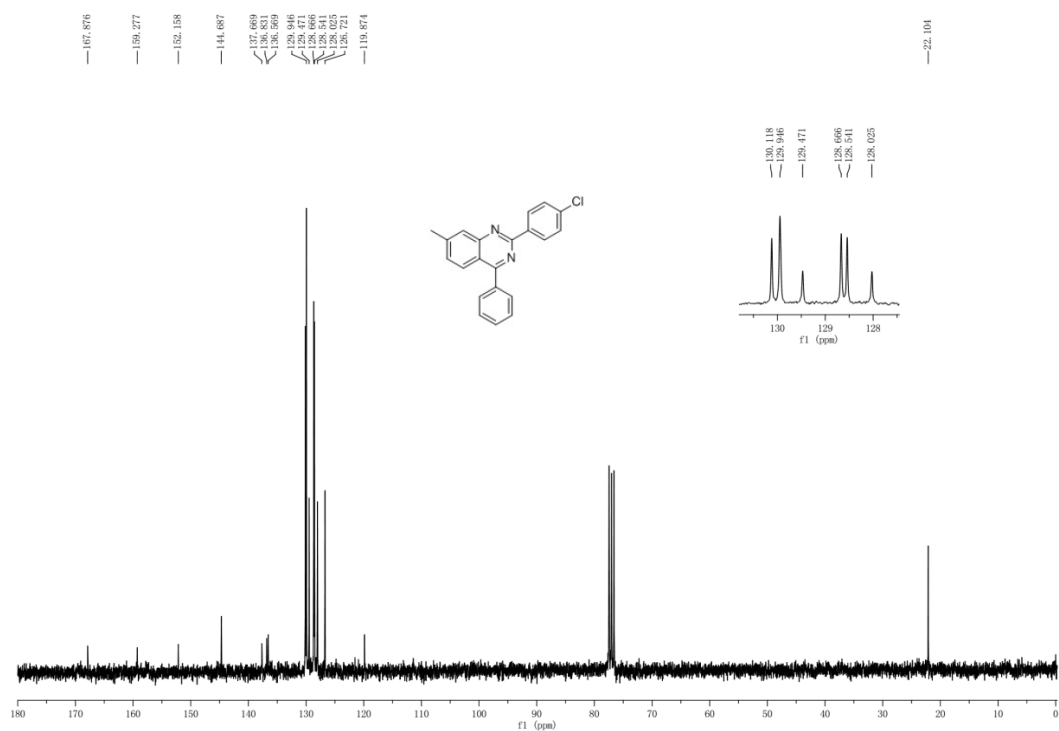
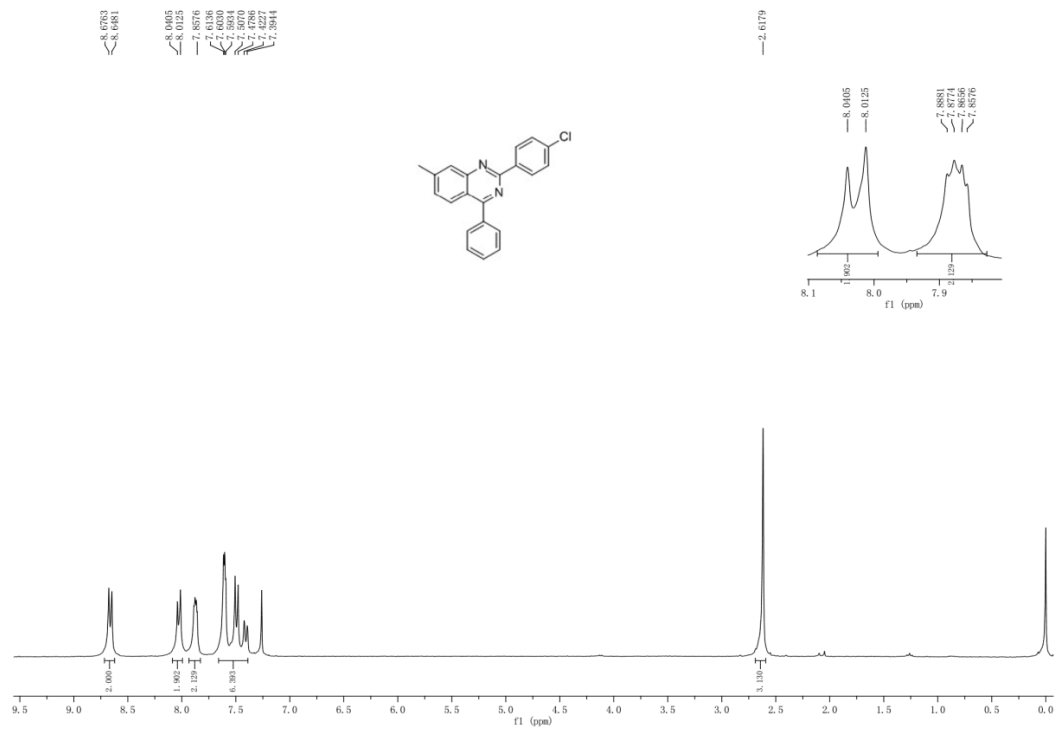


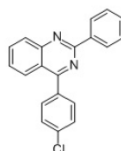
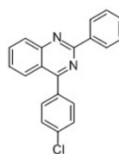


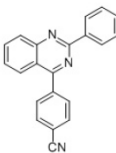
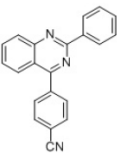


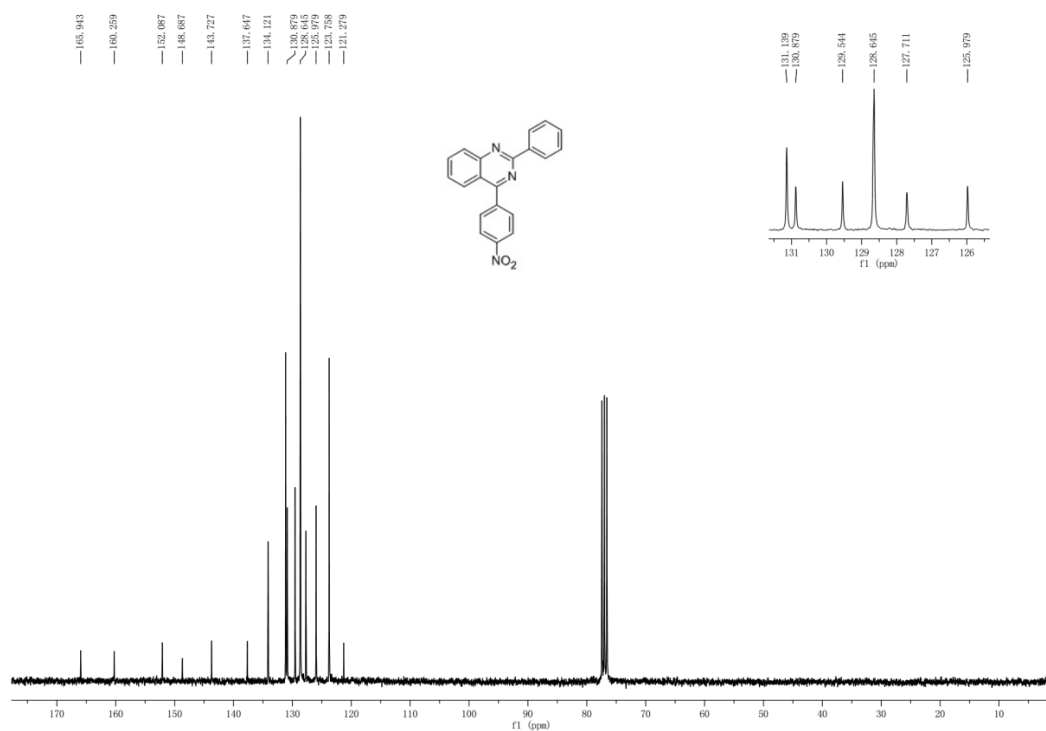
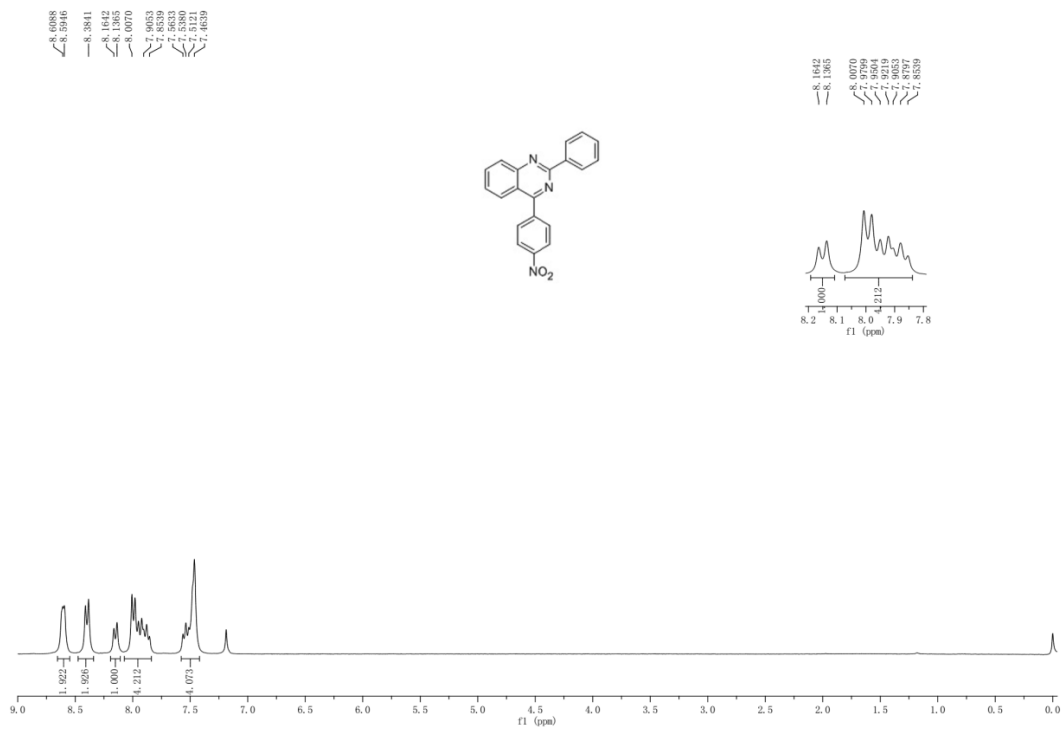


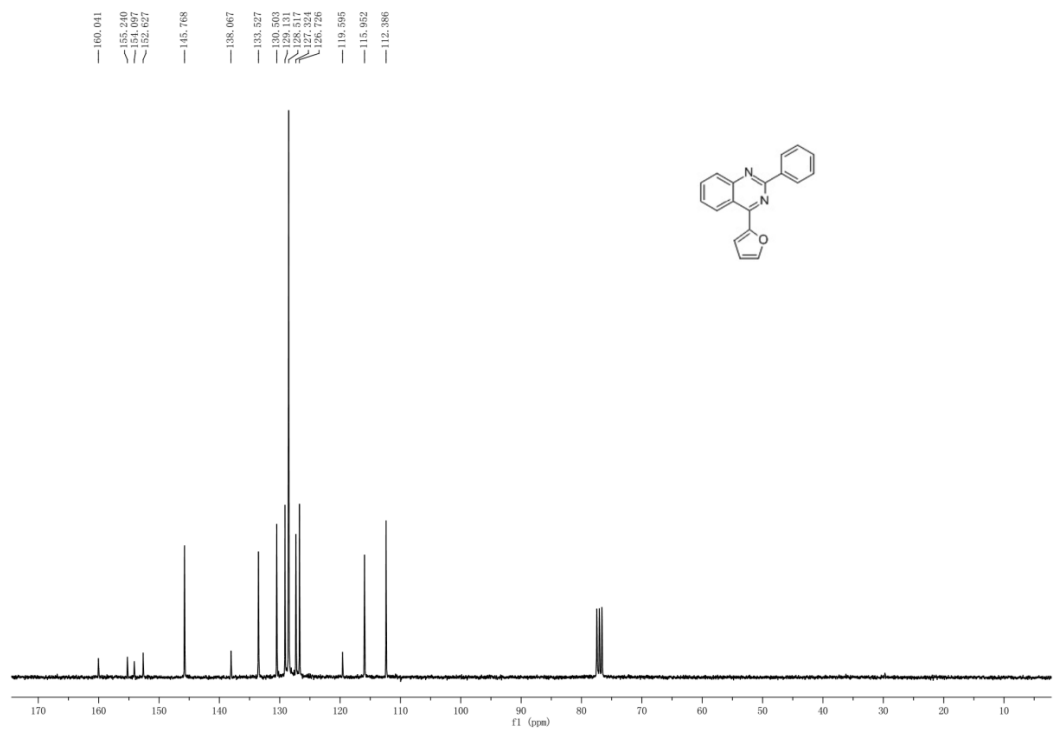
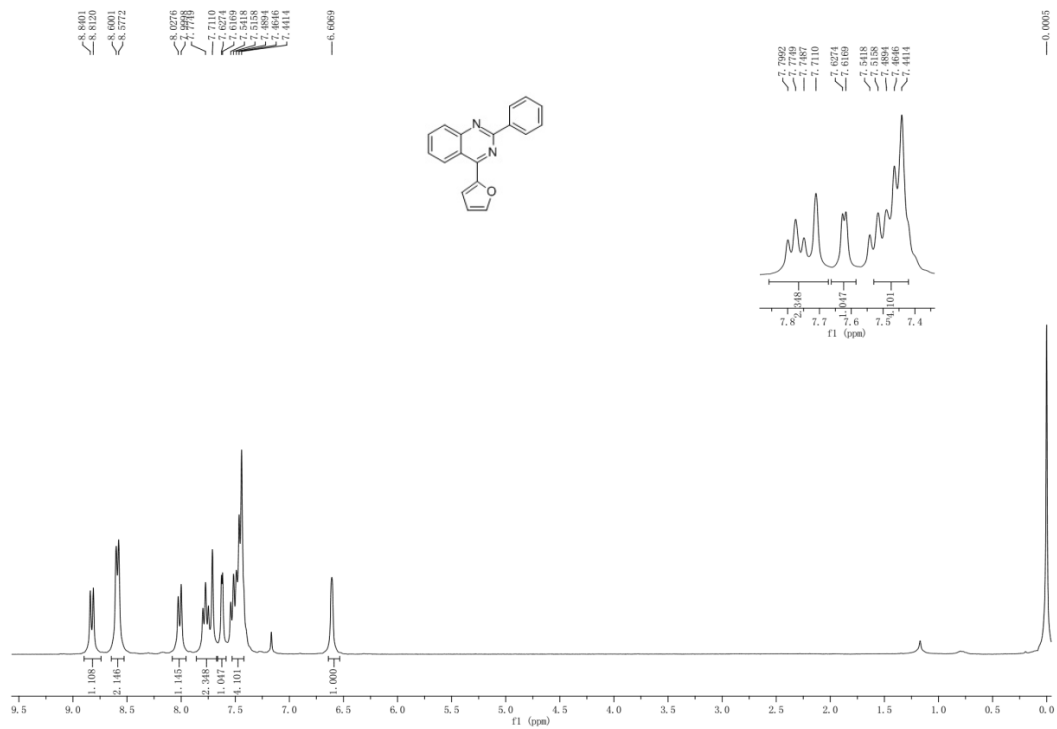


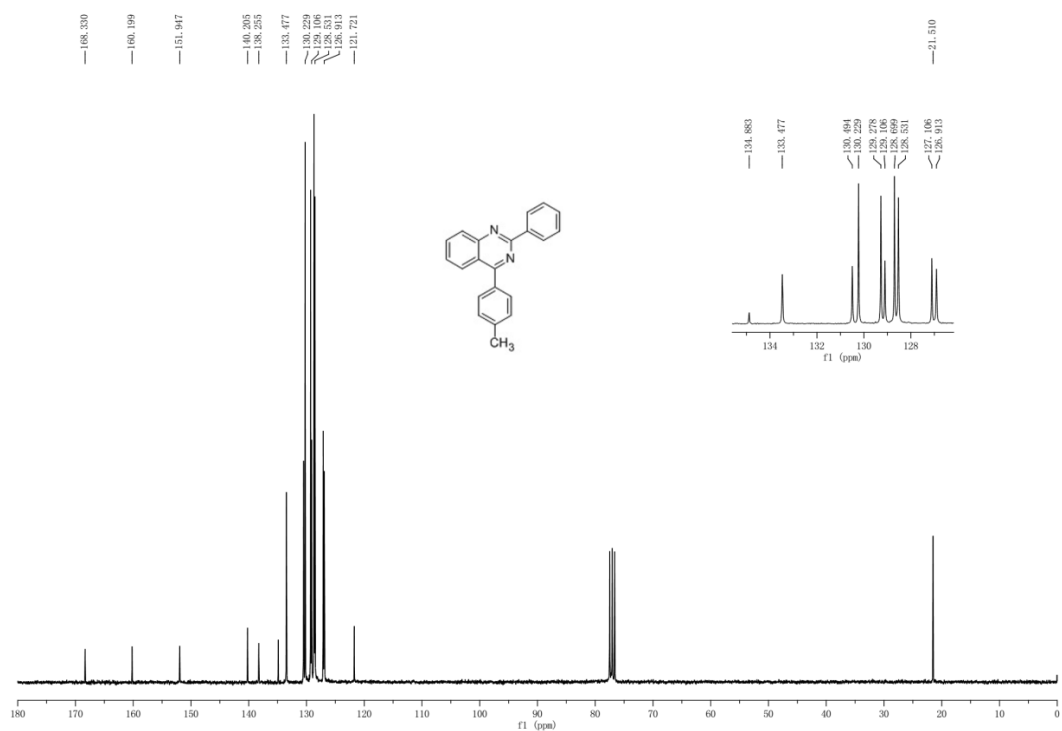
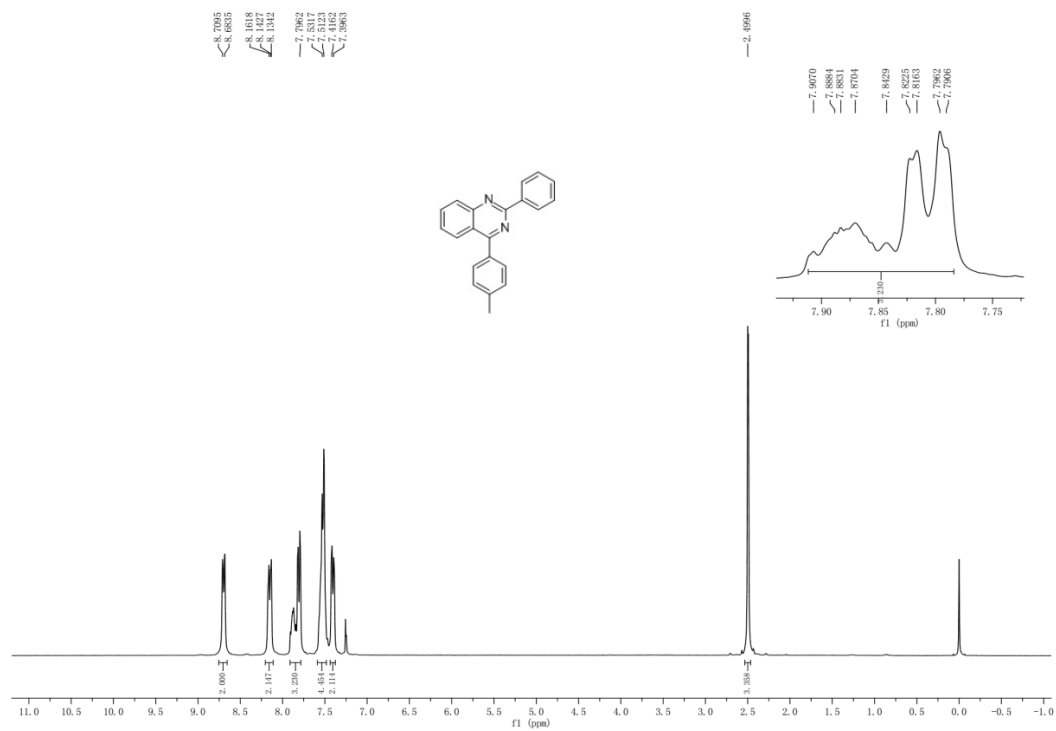


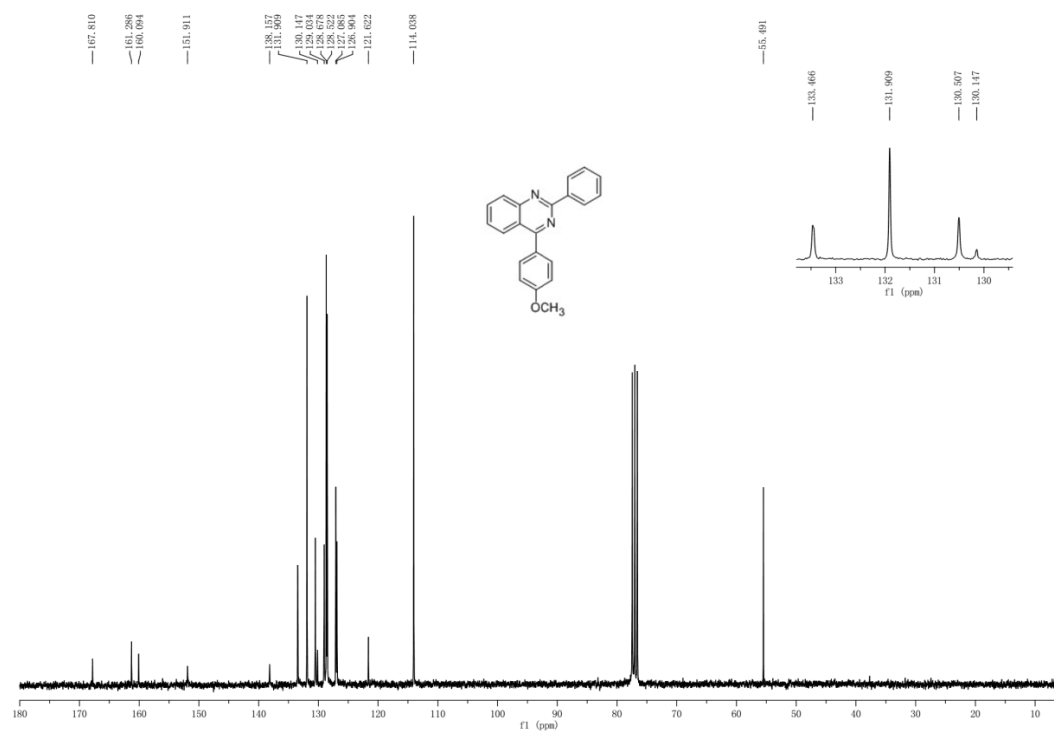
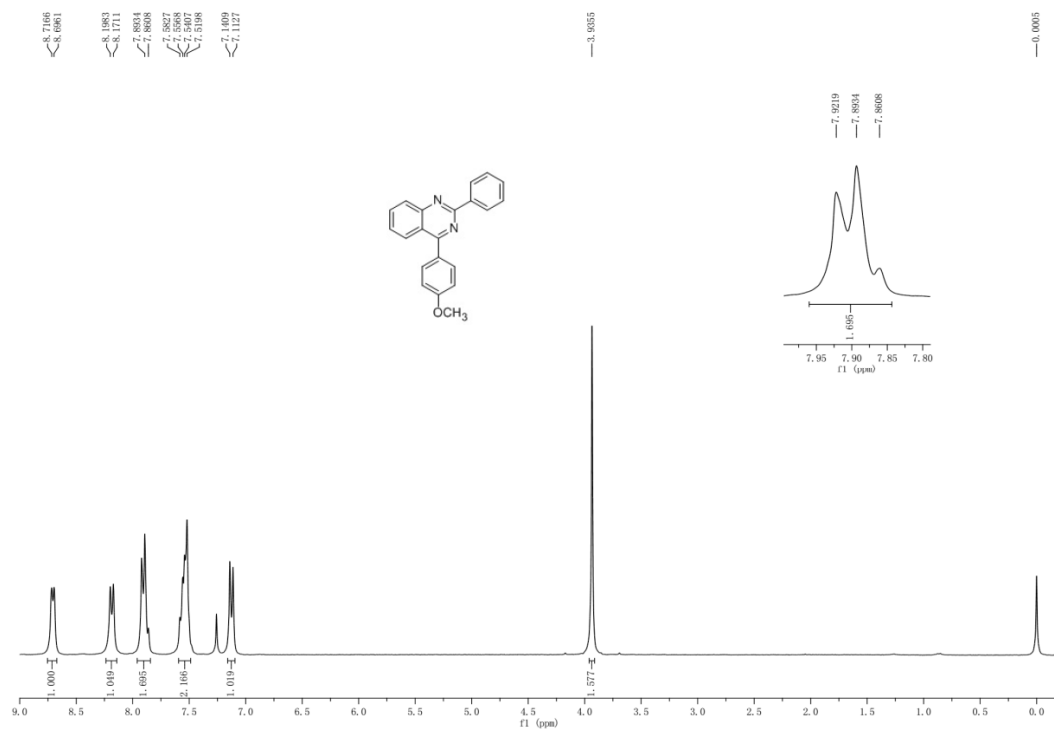


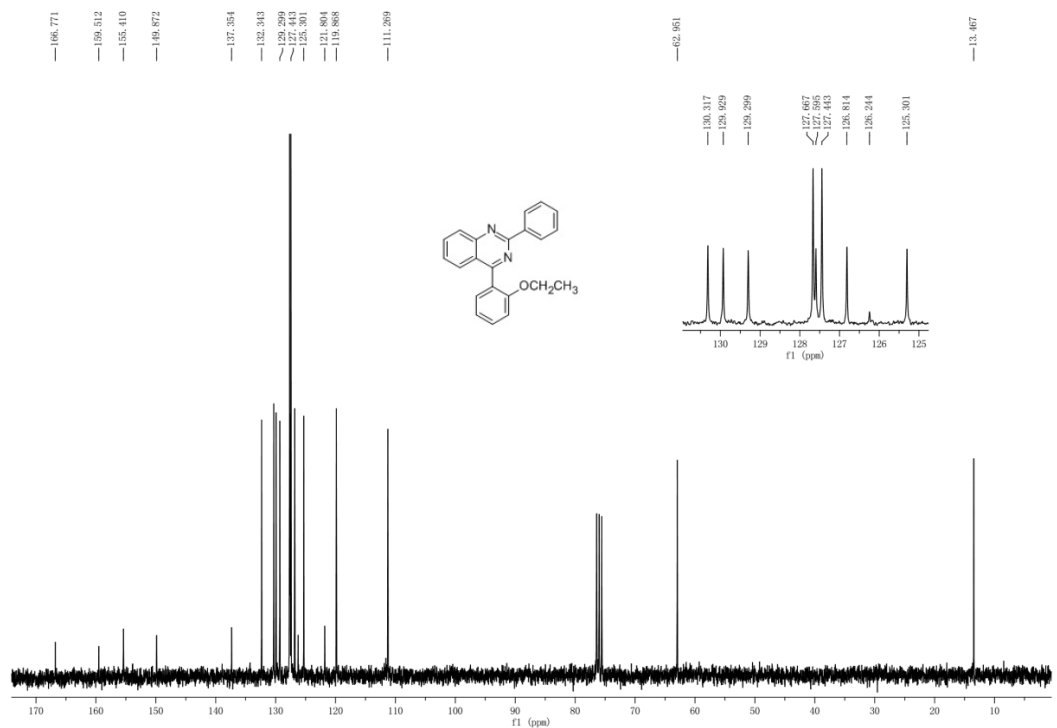
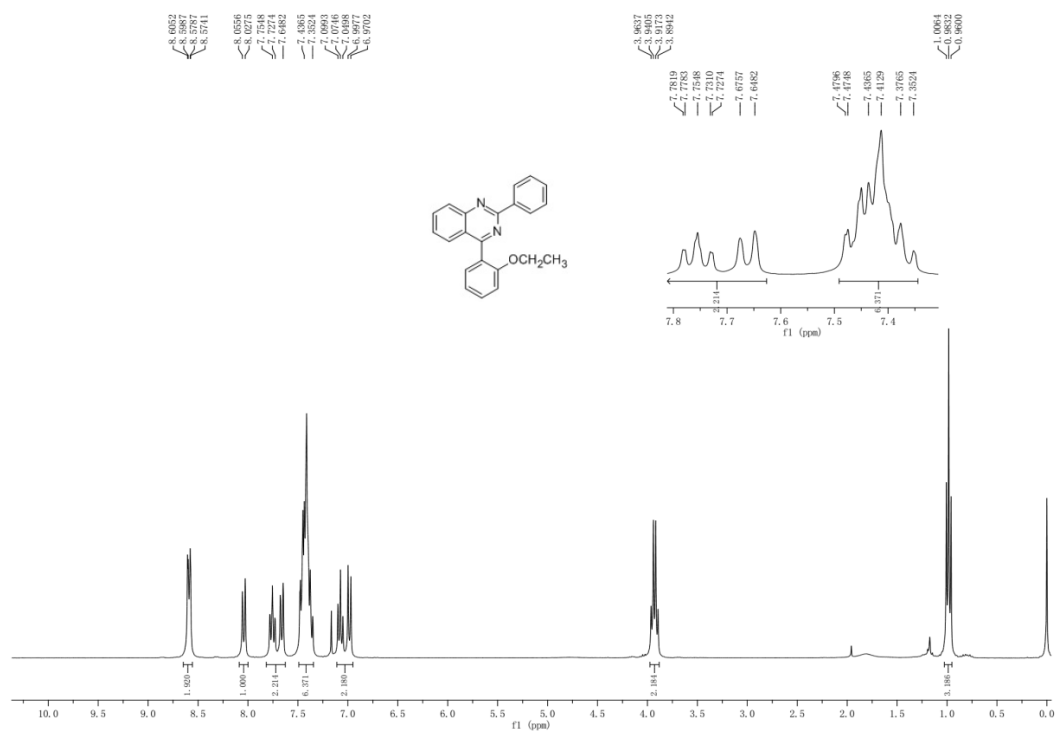


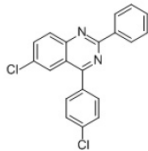
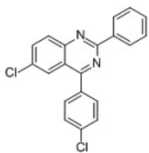


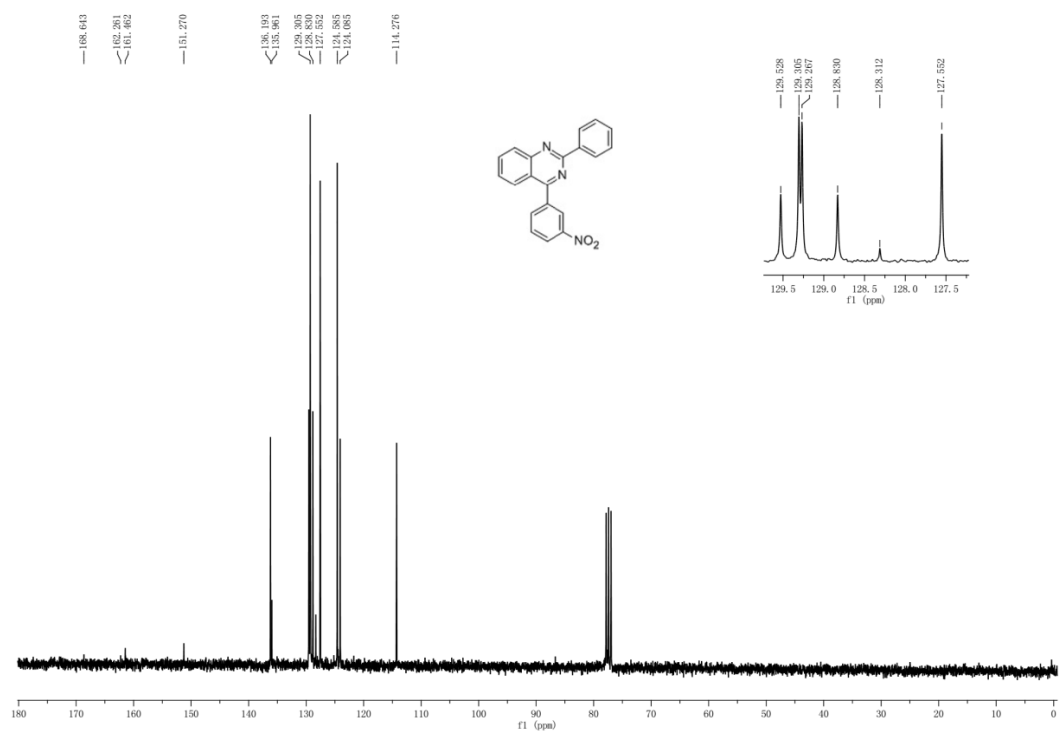
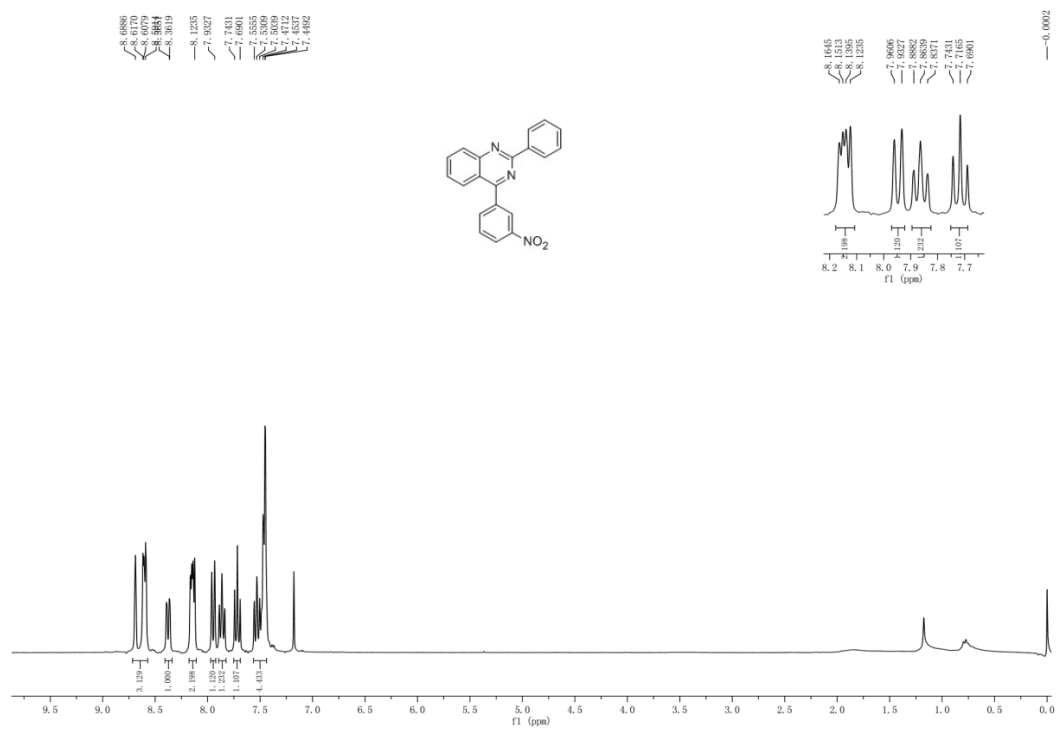


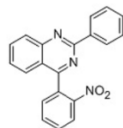
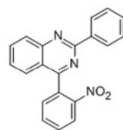












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