Supporting Information

Zn/H_2O-Mediated Tandem Reductive Cyclization of 2-(2-Nitrophenyl)-1*H*-benzimidazoles with Aldehydes to 5,6-Dihydrobenzo[4,5]imidazo[1,2-*c*]quinazolines

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1. General Information

Solvents and reagents: All solvents and reagents were purchased from commercial suppliers and used without further purification.

Reactions: All sample preparation reactions were performed in oven-dried glassware under reflux.

Chromatography: Reactions were monitored by thin-layer chromatography (TLC) using silica gel plates (silica gel 60 F254) and components were visualized by observation under UV light. Flash column chromatography was performed with 200-300 mesh silica gel, eluting with a mixture of petroleum ether (b.p. 60 – 90 ℃) and ethyl acetate.

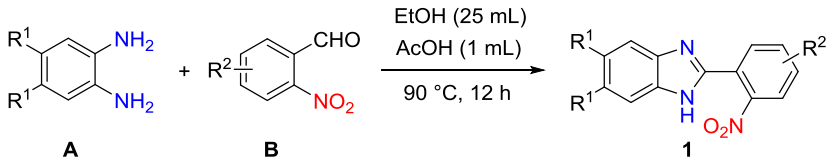
Melting point: Melting points were determined using a Yuhua X-5 apparatus (Gongyi, China) and were not corrected.

NMR spectroscopy: ^{1}H NMR and ^{13}C NMR spectra were recorded with Bruker Avance NEO 400 and Bruker Avance III HD 600 spectrometers with tetramethylsilane (TMS) as an internal standard. All chemical shifts (δ) are given in ppm, and are referenced to residual solvent peaks (2.50 ppm for ^{1}H NMR and 39.56 $\pm$0.06 ppm for ^{13}C NMR spectra in $(CD_3)_2SO$, 7.26 ppm for ^{1}H NMR and 77.16 ppm for ^{13}C NMR spectra in $CDCl_3$) or TMS peak (0.00 ppm). Coupling constants (J) were reported in Hertz (Hz). The following abbreviations are used to indicate the multiplicity of the signals: s = singlet, d = doublet, t = triplet, m = multiplet, and associated combinations, e.g. dd = doublet of doublets.

Mass spectrometry: High resolution mass spectrometry (HRMS) was performed on a Bruker Micro ToF II mass spectrometer.

2. Experimental Procedures

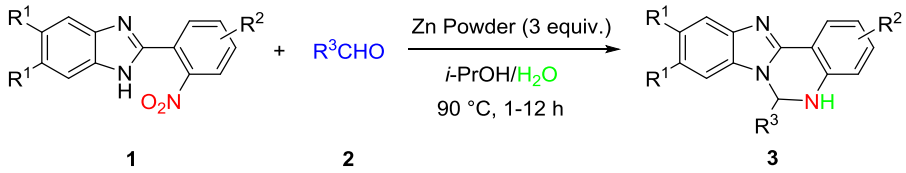
2.1 Preparation of 2-(2-nitrophenyl)-1*H*-benzimidazoles substrates



A B 1

To a mixture of **A** (10 mmol, 1 equiv.) and **B** (10 mmol, 1 equiv.) in EtOH (25 mL) was added AcOH (1 mL) and the mixture was stirred under reflux for 12 hours[1]. The mixture was cooled to room temperature, a yellowish-white precipitate that was collected by filtration and washed with H_2O and then dried to afford the corresponding product **1**.

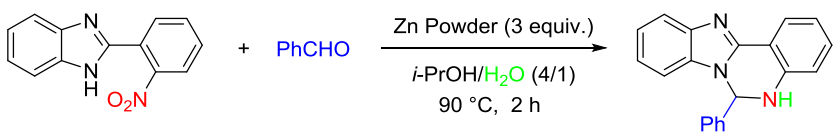
2.2 General procedures for Zn/H_2O-mediated, one-pot reductive cyclization of 2-(2-nitrophenyl)-1*H*-benzimidazoles with aldehydes



1 2 3

In a 10 mL tube with a stir bar, 2 mL *i*-PrOH and 0.5 mL H_2O were added to a mixture of **1** (0.5 mmol, 1.0 equiv.), Zn powder (1.5 mmol, 3 equiv.), aldehyde **2** (0.75 mmol, 1.5 equiv.) under air. The reaction mixture was refluxed in an oil bath for 1-12 h. After cooling to room temperature, the mixture was filtered over celite and anhydrous Na_2SO_4, and concentrated under vacuum. The crude reaction mixture was purified by silica gel column chromatography to give the corresponding product with petroleum ether/ethyl acetate (PE/EA = 3/1) as the eluent.

2.3 General procedure for gram-scale reaction

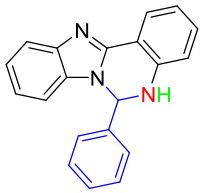


1a, 5 mmol (1.19 g) **2a**, 1.5 equiv. **3a**, 78%, (1.16 g)

In a 50 mL round-bottom flask with a stir bar, 20 mL *i*-PrOH and 5 mL H_2O were added to a mixture of **1a** (1.19 g, 5 mmol, 1.0 equiv.), Zn powder (980.70 mg, 15 mmol, 3 equiv.), **2a** (795.90 mg, 7.5 mmol, 1.5 equiv.) under air. The reaction

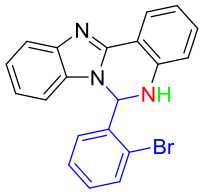
mixture was allowed to reflux in an oil bath for 2 h. After cooling to room temperature, the mixture was filtered over celite and anhydrous Na_2SO_4, and concentrated under vacuum. The crude reaction mixture was purified by silica gel column chromatography to give the desired product **3a** with petroleum ether/ethyl acetate (PE/EA = 3/1) as the eluent.

3. Characterization Data



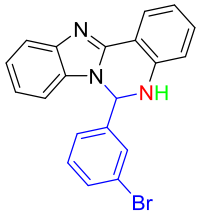
6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3a)

White solid (132.32 mg, 89%), R_f = 0.23 (PE/EA = 3/1), melting point: 202 – 203 ℃ (Lit.[2] 220 ℃), CAS registry number: 305851-84-7, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.97 (d, J = 7.6 Hz, 1H), 7.66 (d, J = 8.7 Hz, 2H), 7.31 – 7.23 (m, 6H), 7.19 – 7.09 (m, 4H), 6.88 – 6.81 (m, 2H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.4, 144.3, 143.7, 140.9, 133.3, 132.2, 129.4, 129.3, 126.5, 125.1, 122.7, 122.5, 119.2, 118.7, 115.3, 112.4, 111.0, 68.3.



6-(2-bromophenyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3b)

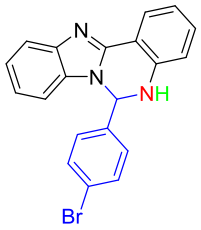
White solid (158.03 mg, 84%), R_f = 0.32 (PE/EA = 3/1), melting point: 192 – 194 ℃, (Lit.[3] 195 – 197 ℃), CAS registry number: 38027-33-9, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.99 (d, J = 7.7 Hz, 1H), 7.75 (d, J = 7.6 Hz, 1H), 7.67 (d, J = 8.0 Hz, 1H), 7.53 (s, 1H), 7.33 – 7.24 (m, 4H), 7.18 (t, J = 7.7 Hz, 1H), 7.09 – 7.02 (m, 4H), 6.89 – 6.80 (m, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.5, 144.4, 143.0, 138.5, 133.9, 133.0, 132.3, 131.8, 129.2, 128.6, 125.2, 122.9, 122.8, 121.8, 119.3, 118.8, 115.4, 111.7, 110.4, 68.6.



6-(3-bromophenyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3c)

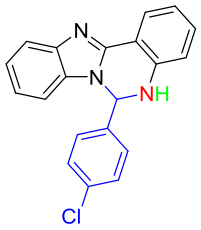
White solid (127.92 mg, 68%), R_f = 0.23 (PE/EA = 3/1), melting point: 225 – 227 ℃, (Lit.[3] 234 – 236 ℃), CAS registry number: 325763-56-2, **^{1}H NMR** (400 MHz,

DMSO-d_6) δ 7.97 (dd, J = 7.6, 1.1 Hz, 1H), 7.68 (d, J = 7.6 Hz, 2H), 7.56 (t, J = 1.6 Hz, 1H), 7.51 – 7.49 (m, 1H), 7.29 – 7.12 (m, 6H), 7.07 (d, J = 7.8 Hz, 1H), 6.88 – 6.83 (m, 2H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.2, 144.3, 143.5, 143.2, 133.2, 132.3, 132.8, 131.6, 129.3, 125.2, 125.0, 122.9, 122.8, 122.4, 119.3, 119.0, 115.4, 112.4, 110.9, 67.2.



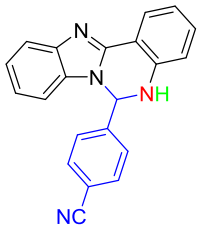
6-(4-bromophenyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3d)

White solid (152.39 mg, 81%), R_f = 0.18 (PE/EA = 3/1), melting point: 102 – 103 ℃, (Lit.[3] 209 – 210 ℃), CAS registry number: 378199-61-2, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.95 (d, J = 7.6 Hz, 1H), 7.67 – 7.65 (m, 2H), 7.54 (d, J = 8.4 Hz, 2H), 7.28 – 7.11 (m, 7H), 6.86 – 6.82 (m, 2H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.3, 144.3, 143.3, 140.3, 133.2, 132.3, 128.5, 125.2, 122.8, 122.7, 122.6, 119.3, 118.9, 115.4, 112.4, 110.9, 67.5.



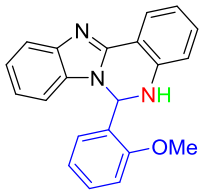
6-(4-chlorophenyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3e)

White solid (119.45 mg, 72%), R_f = 0.18 (PE/EA = 3/1), melting point: 176 – 177 ℃, (Lit.[2] 207 ℃), CAS registry number: 387371-80-4, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.96 (d, J = 7.4 Hz, 1H), 7.66 (d, J = 8.3 Hz, 2H), 7.40 (d, J = 8.4 Hz, 2H), 7.27 – 7.11 (m, 7H), 6.87 – 6.82 (m, 2H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.2, 144.3, 143.3, 139.9, 134.0, 133.2, 132.2, 129.3, 128.2, 125.2, 122.8, 122.7, 119.2, 118.9, 115.4, 112.3, 110.9, 67.4.



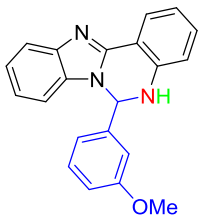
4-(5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazolin-6-yl)benzonitrile (3f)

Yellow solid (32.23 mg, 20%), R_f = 0.36 (PE/EA = 1/1), melting point: 232 – 233 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.96 (dd, J = 8.1, 1.6 Hz, 1H), 7.83 – 7.74 (m, 3H), 7.68 (d, J = 8.0 Hz, 1H), 7.37 – 7.32 (m, 3H), 7.28 – 7.15 (m, 4H), 6.86 – 6.83 (m, 2H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.1, 146.0, 144.2, 143.0, 133.4, 133.2, 132.5, 132.4, 127.4, 127.1, 125.2, 123.0, 122.9, 119.3, 119.1, 118.8, 115.5, 112.4, 112.1, 110.8, 67.1; **HRMS (ESI)** calcd for $C_{21}H_{15}N_4^+$ $[M+H]^+$: 323.1291; found: 323.1292.



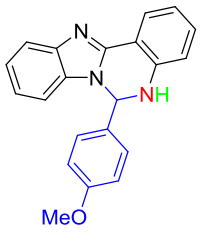
6-(2-methoxyphenyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3h)

White solid (127.68 mg, 78%), R_f = 0.20 (PE/EA = 3/1), melting point: 167 – 168 ℃, CAS registry number: 1947372-92-0, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 8.03 (dd, J = 7.7, 1.2 Hz, 1H), 7.69 (d, J = 8.0 Hz, 1H), 7.32 (d, J = 2.5 Hz, 2H), 7.29 – 7.17 (m, 3H), 7.12 – 7.07 (m, 3H), 6.91 (d, J = 8.0 Hz, 1H), 6.82 (td, J = 7.8, 0.6 Hz 1H), 6.74 (t, J = 7.4 Hz, 1H), 6.66 (dd, J = 7.6, 1.6 Hz, 1H), 3.85 (s, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 156.6, 147.9, 144.3, 143.7, 133.3, 132.0, 130.7, 128.1, 126.6, 125.0, 122.7, 121.0, 119.1, 118.4, 115.3, 112.1, 110.6, 63.5, 56.18; **HRMS (ESI)** calcd for $C_{21}H_{18}N_3O^+$ $[M+H]^+$: 328.1444; found: 328.1444.



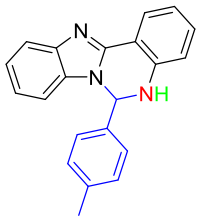
6-(3-methoxyphenyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3i)

White solid (130.96 mg, 80%), R_f = 0.17 (PE/EA = 3/1), melting point: 183 – 184 ℃, CAS registry number: 387372-04-5, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.96 (t, J = 7.9 Hz, 1H), 7.69 – 7.62 (m, 2H), 7.29 – 7.05 (m, 6H), 6.91 – 6.81 (m, 4H), 6.74 (t, J = 7.7 Hz, 1H), 3.68 (d, J = 9.6 Hz, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 159.9, 147.5, 144.3, 143.6, 142.5, 133.4, 132.2, 130.5, 125.2, 122.7, 122.6, 119.2, 118.8, 118.3, 115.4, 114.1, 112.8, 112.5, 111.0, 68.1, 55.5; **HRMS (ESI)** calcd for $C_{21}H_{18}N_3O^+$ $[M+H]^+$: 328.1444; found: 328.1438.



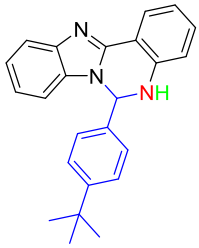
6-(4-methoxyphenyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3j)

White solid (150.59 mg, 92%), R_f = 0.16 (PE/EA = 3/1), melting point: 103 – 104 ℃, (Lit.[2] 187 ℃), CAS registry number: 325765-30-8, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 8.02 (dd, J = 7.7, 1.1 Hz, 1H), 7.68 (d, J = 8.0 Hz, 1H), 7.57 (d, J = 1.0 Hz, 1H), 7.30 – 7.25 (m, 3H), 7.18 (ddd, J = 8.2, 4.9, 3.4 Hz, 1H), 7.08 (d, J = 3.9 Hz, 2H), 7.02 (d, J = 1.3 Hz, 1H), 6.91 (dd, J = 8.5, 3.3 Hz, 3H), 6.85 (t, J = 7.5 Hz, 1H), 3.69 (s, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 160.2, 147.6, 144.4, 143.9, 133.4, 132.9, 132.1, 128.0, 125.2, 122.6, 122.5, 119.1, 118.6, 115.3, 114.6, 112.4, 111.1, 68.3, 55.6**.**



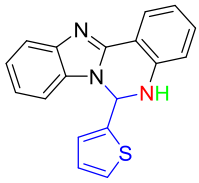
6-(*p*-tolyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3k)

White solid (116.77 mg, 75%), R_f = 0.26 (PE/EA = 3/1), melting point: 104 – 105 ℃, CAS registry number: 387372-24-9, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.96 (d, J = 7.1 Hz, 1H), 7.65 (d, J = 7.9 Hz, 1H), 7.56 (s, 1H), 7.24 (t, J = 7.2 Hz, 1H), 7.17 – 7.06 (m, 7H), 7.02 (s, 1H), 6.87 – 6.80 (m, 2H), 2.23 (s, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.5, 144.3, 143.7, 138.9, 138.0, 133.4, 132.1, 129.7, 126.4, 125.1, 122.6, 122.5, 119.1, 118.6, 115.3, 112.4, 111.0, 68.2; **HRMS (ESI)** calcd for $C_{21}H_{18}N_3^+$ $[M+H]^+$: 312.1495; found: 312.1495.



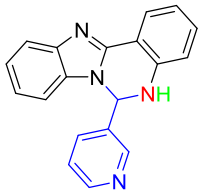
6-(4-(*tert*-butyl)phenyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3l)

White solid (139.62 mg, 79%), R_f = 0.27 (PE/EA = 3/1), melting point: 94 – 95 ℃, CAS registry number: 387372-44-3, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.97 (dd, J = 7.7, 1.1 Hz, 1H), 7.66 (d, J = 8.0 Hz, 1H), 7.60 (d, J = 1.5 Hz, 1H), 7.32 (d, J = 8.4 Hz, 2H), 7.27 – 7.16 (m, 5H), 7.10 (td, J = 7.9, 0.7 Hz, 1H), 7.05 (d, J = 1.6 Hz, 1H), 6.88 – 6.81 (m, 2H), 1.19 (s, 9H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 151.9, 147.4, 144.3, 143.7, 138.2, 133.3, 132.1, 126.0, 126.0, 125.1, 122.7, 122.5, 119.1, 118.6, 115.3, 112.4, 111.0, 67.9, 34.8, 31.4; **HRMS (ESI)** calcd for $C_{24}H_{24}N_3^+$ $[M+H]^+$: 354.1965; found: 354.1965.



6-(thiophen-2-yl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3m)

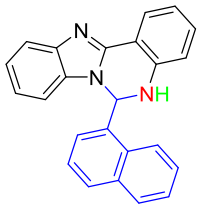
White solid (125.90 mg, 83%), R_f = 0.23 (PE/EA = 3/1), melting point: 174 – 175 ℃, (Lit.[4] 210 – 212 ℃), CAS registry number: 387371-68-8, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.98 (dd, J = 7.7, 1.2 Hz, 1H), 7.77 (d, J = 1.9 Hz, 1H), 7.66 (dd, J = 6.9, 1.5 Hz, 1H), 7.47 – 7.44 (m, 2H), 7.37 (dd, J = 5.0, 1.1 Hz, 1H), 7.31 (td, J = 8.5, 1.5 Hz 1H), 7.22 – 7.15 (m, 3H), 6.97 – 6.87 (m, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 146.9, 144.3, 144.2, 143.3, 133.0, 132.2, 127.0, 126.9, 126.5, 125.1, 122.8, 122.7, 119.3, 199.2, 115.9, 112.8, 111.0, 64.2.



6-(pyridin-3-yl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3n)

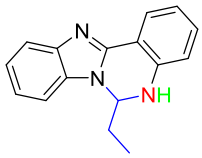
White solid (107.40 mg, 72%), R_f = 0.30 (EA), melting point: 221 – 222 ℃, (Lit.[4] 216 – 217 ℃), CAS registry number: 326618-68-2, **^{1}H NMR** (400 MHz, DMSO-d_6)

δ 8.55 (d, J = 2.1 Hz, 1H), 8.51 (dd, J = 4.8, 1.6 Hz, 1H), 7.99 (dd, J = 7.7, 1.2 Hz, 1H), 7.73 (d, J = 1.8 Hz, 1H), 7.69 (d, J = 7.9 Hz, 1H), 7.52 (dt, J = 8.0, 1.9 Hz, 1H), 7.34 – 7.12 (m, 6H), 6.90 – 6.85 (m, 2H). **^{13}C NMR** (100 MHz, DMSO-d_6) δ 150.6, 147.7, 147.3, 144.3, 143.2, 136.4, 133.8, 133.1, 132.3, 125.2, 124.5, 122.9, 122.8, 119.3, 119.1, 115.5, 112.5, 110.8, 66.1.



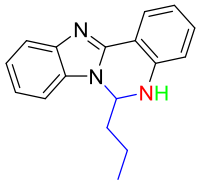
6-(naphthalen-1-yl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3o)

White solid (145.92 mg, 84%), R_f = 0.24 (PE/EA = 3/1), melting point: 180 – 181 ℃, CAS registry number: 387371-76-8, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 8.54 – 8.51 (m, 1H)., 8.09 – 7.97 (m, 3H), 7.75 (s, 1H), 7.59 – 7.55 (m, 3H), 7.44 (t, J = 7.7 Hz, 1H), 7.24 (td, J = 7.7, 1.6 Hz, 1H), 7.20 – 7.18 (m, 1H), 7.12 – 7.08 (m, 1H), 6.88 – 6.83 (m, 3H), 6.47 (d, J = 8.1 Hz, 1H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 148.3, 144.5, 143.8, 134.5, 134.3, 133.4, 132.1, 130.6, 130.5, 129.3, 127.2, 126.7, 125.8, 125.3, 124.2, 122.6, 122.4, 119.2, 118.7, 115.4, 112.1, 110.9; **HRMS (ESI)** calcd for $C_{24}H_{18}N_3^+$ $[M+H]^+$: 348.1495; found: 348.1495.



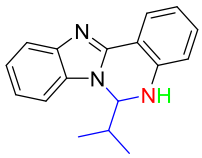
6-ethyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3p)

White solid (74.79 mg, 60%), R_f = 0.14 (PE/EA = 3/1), melting point: 161 – 162 ℃, CAS registry number: 3018902-72-9, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.88 (dd, J = 7.7, 1.3 Hz, 1H), 7.65 – 7.59 (m, 2H), 7.26 – 7.17 (m, 4H), 6.90 (d, J = 7.9 Hz, 1H), 6.78 (td, J = 7.7, 0.84 Hz,1H), 6.05-6.02 (m, 1H), 1.88 – 1.71 (m, 2H), 0.82 (t, J = 7.4 Hz, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.1, 144.2, 143.9, 133.2, 132.0, 125.0, 122.4, 122.4, 119.0, 118.2, 115.3, 112.5, 110.6, 67.0, 29.4, 9.1; **HRMS (ESI)** calcd for $C_{16}H_{16}N_3^+$ $[M+H]^+$: 250.1339; found: 250.1339.



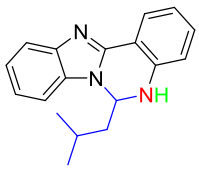
6-propyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3q)

White solid (93.49 mg, 71%), R_f = 0.20 (PE/EA = 3/1), melting point: 70 – 71 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.88 (d, J = 7.7 Hz, 1H), 7.65 – 7.58 (m, 2H), 7.26 – 7.16 (m, 4H), 6.89 (d, J = 8.1 Hz, 1H), 6.78 (t, J = 7.5 Hz, 1H), 6.07 (s, 1H), 1.84 – 1.75 (m, 1H), 1.71 – 1.62 (m, 1H), 1.33 – 1.24 (m, 2H), 0.79 (t, J = 7.3 Hz, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.0, 144.2, 143.8, 133.1, 132.0, 125.0, 122.4, 119.0, 118.3, 115.4, 112.5, 110.5, 65.8, 38.5, 17.7, 14.0; **HRMS (ESI)** calcd for $C_{17}H_{18}N_3^+$ $[M+H]^+$: 264.1495; found: 264.1495.



6-isopropyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3r)

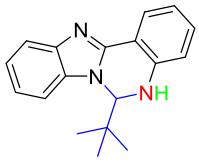
White solid (23.70 mg, 18%), R_f = 0.18 (PE/EA = 3/1), melting point: 195 – 196 ℃, CAS registry number: 2674076-31-2, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.85 (dd, J = 7.7, 1.6 Hz, 1H), 7.64 – 7.59 (m, 2H), 7.24 – 7.17 (m, 4H), 6.91 (dd, J = 8.1, 1.0 Hz, 1H), 6.74 (td, J = 7.5, 1.1 Hz, 1H), 5.89 (dd, J = 5.1, 2.9 Hz, 1H), 2.24 – 2.16 (m, 1H), 0.88 (d, J = 6.9 Hz, 3H), 0.71 (d, J = 6.7 Hz, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.3, 144.5, 144.1, 133.6, 132.0, 124.9, 122.4, 122.3, 119.0, 117.9, 114.7, 112.6, 111.0, 70.3, 35.3, 18.7, 17.04; **HRMS (ESI)** calcd for $C_{17}H_{18}N_3^+$ $[M+H]^+$: 264.1495; found: 264.1495.



6-isobutyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3s)

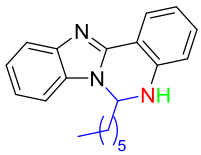
White solid (70.73 mg, 51%), R_f = 0.27 (PE/EA = 3/1), melting point: 169 – 170 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.89 (d, J = 7.6 Hz, 1H), 7.64 (dd, J = 7.1, 1.5 Hz 1H), 7.52 (d, J = 7.6 Hz, 1H), 7.27 – 7.19 (m, 4H), 6.92 (d, J = 8.1 Hz, 1H), 6.80 (t, J = 7.5 Hz, 1H), 6.12 – 6.10 (m, 1H), 1.78 – 1.69 (m, 2H), 1.45 – 1.39 (m, 1H), 0.96 (d,

J = 6.3 Hz, 3H), 0.82 (d, *J* = 6.3 Hz, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 147.0, 144.2, 143.4, 132.9, 132.1, 125.0, 122.4, 119.1, 118.4, 115.8, 112.7, 110.2, 64.5, 44.4, 23.7, 22.3; **HRMS (ESI)** calcd for $C_{18}H_{20}N_3^+$ $[M+H]^+$: 278.1652; found: 278.1651.



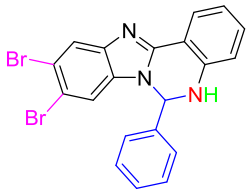
6-(*tert*-butyl)-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3t)

White solid (15.26 mg, 11%), R_f = 0.23 (PE/EA = 3/1), melting point: 156 – 157 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 7.83 (dd, *J* = 7.6, 1.2 Hz, 1H), 7.66 (dd, *J* = 6.3, 2.5 Hz, 1H), 7.62 – 7.61 (m, 1H), 7.23 – 7.16 (m, 4H), 6.91 (d, *J* = 7.9 Hz, 1H), 6.73 (td, *J* = 7.6, 0.8 Hz,1H), 5.81 (d, *J* = 3.1 Hz, 1H), 0.85 (s, 9H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 151.9, 147.4, 144.3, 143.7, 138.2, 133.3, 132.1, 126.0, 126.0, 125.1, 122.7, 122.5, 119.1, 118.6, 115.3, 112.4, 111.0, 67.9, 34.8, 31.4; **HRMS (ESI)** calcd for $C_{18}H_{20}N_3^+$ $[M+H]^+$: 278.1652; found: 278.1652.



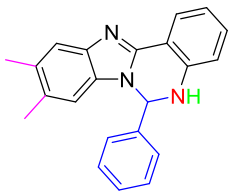
6-hexyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3u)

White solid (88.57 mg, 58%), R_f = 0.25 (PE/EA = 3/1), melting point: 162 – 163 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 7.86 (dd, *J* = 7.7, 1.1 Hz, 1H), 7.62 (dd, *J* = 6.6, 2.0 Hz, 1H), 7.59 (dd, *J* = 6.7, 1.8 Hz, 1H), 7.25 – 7.19 (m, 3H), 7.16 (d, *J* = 2.0 Hz, 1H), 6.88 (d, *J* = 8.0 Hz, 1H), 6.78 (t, *J* = 7.4 Hz, 1H), 6.08 – 6.05 (m, 1H), 1.99 (s, 1H), 1.82 – 1.77 (m, 1H), 1.70 – 1.65 (m, 1H), 1.29 – 1.25 (m, 2H), 1.18 – 1.15 (m, 5H), 0.78 (t, *J* = 7.1 Hz, 3H); **^{13}C NMR** (150 MHz, DMSO-d_6) δ 147.0, 144.2, 143.8, 133.1, 132.1, 125.0, 122.4, 122.4, 119.0, 118.3, 115.4, 112.5, 110.5, 66.0, 36.2, 31.6, 28.7, 24.2, 22.4, 14.3; **HRMS (ESI)** calcd for $C_{20}H_{24}N_3^+$ $[M+H]^+$: 306.1965; found: 306.1965.



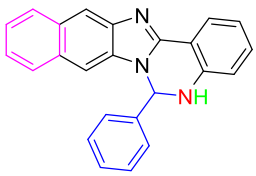
9,10-dibromo-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3v)

Yellow solid (186.61 mg, 82%), R_f = 0.30 (PE/EA = 3/1), melting point: 218 – 220 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 8.06 (s, 1H), 7.95 (dd, J = 7.7, 1.2 Hz, 1H), 7.78 (d, J = 1.8 Hz, 1H), 7.59 (s, 1H), 7.36 – 7.34 (m, 3H), 7.31 – 7.27 (m, 3H), 7.13 (d, J = 1.9 Hz, 1H), 6.88 – 6.81 (m, 2H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 149.7, 144.9, 144.0, 140.1, 133.8, 133.0, 129.7, 129.5, 129.4, 127.3, 126.5, 125.6, 123.3, 118.9, 117.1, 116.5, 115.6, 115.5, 111.6, 68.2; **HRMS (ESI)** calcd for $C_{20}H_{14}Br_2N_3^+$ $[M+H]^+$: 455.9540; found: 455.9538.



9,10-dimethyl-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3w)

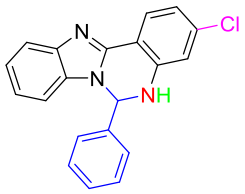
White solid (125.28 mg, 77%), R_f = 0.21 (PE/EA = 3/1), melting point: 238 – 240 ℃, (Lit.[2] 230 ℃), CAS registry number, 932022-75-8; **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.17 (dd, J = 7.7, 0.9 Hz 1H), 7.52 (s, 1H), 7.42 – 7.33 (m, 5H), 7.21 (td, J = 8.0, 1.4 Hz 1H), 6.93 (t, J = 7.5 Hz, 1H), 6.67 (d, J = 8.0 Hz, 1H), 6.58 (s, 1H), 6.30 (s, 1H), 4.78 (s, 1H), 2.29 (s, 3H), 2.16 (s, 3H); **^{13}C NMR** (100 MHz, $CDCl_3$) δ 146.9, 142.9, 141.8, 138.9, 131.7, 131.5, 131.5, 131.2, 129.9, 129.2, 126.9, 125.4, 120.1, 119.5, 114.8, 113.7, 110.6, 70.3, 20.6, 20.3.



6-phenyl-5,6-dihydronaphtho[2',3':4,5]imidazo[1,2-*c*]quinazoline (3x)

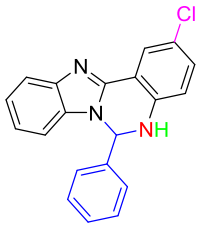
White solid (112.82 mg, 65%), R_f = 0.22 (PE/EA = 3/1), melting point: 148 – 145 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 8.20 (s, 1H), 8.07 – 7.99 (m, 3H), 7.81 (s, 2H), 7.68 (s, 1H), 7.39 – 7.36 (m, 2H), 7.33 – 7.31 (m, 5H), 7.21 (s, 1H), 6.91 (d, J = 8.1 Hz, 1H), 6.87 (t, J = 7.4 Hz, 1H); **^{13}C NMR** (150 MHz, DMSO-d_6) δ 151.2, 144.5,

144.3, 140.9, 134.2, 133.2, 130.5, 130.2, 129.4, 129.3 128.5, 127.7, 126.3, 125.9, 124.5, 123.9, 118.7, 115.5, 115.4, 111.7, 106.5, 68.0; **HRMS (ESI)** calcd for $C_{24}H_{18}N_3^+$ $[M+H]^+$: 348.1495; found: 348.1495.



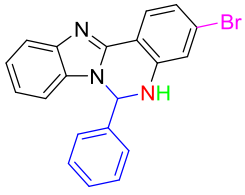
2-chloro-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3y)

White solid (109.26 mg, 66%), R_f = 0.31 (PE/EA = 4/1), melting point: 236 – 238 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 7.97 (d, J = 8.3 Hz, 1H), 7.88 (s, 1H), 7.67 (d, J = 8.0 Hz, 1H), 7.36 – 7.39 (m, 5H), 7.20 (t, J = 7.4 Hz, 1H), 7.14-7.10 (m, 3H), 6.91 (s, 1H), 6.86 (d, J = 8.3 Hz, 1H); **^{13}C NMR** (150 MHz, DMSO-d_6) δ 146.5, 144.7, 144.3 140.6, 136.4, 133.3, 129.6, 129.4, 126.8, 126.5, 122.9, 122.8, 119.3, 118.6, 114.4 111.1, 68.2; **HRMS (ESI)** calcd for $C_{20}H_{15}ClN_3^+$ $[M+H]^+$: 332.0949; found: 332.0951.



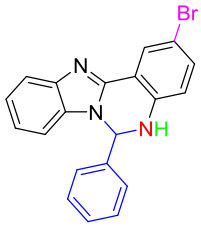
2-chloro-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3z)

White solid (84.43 mg, 51%), R_f = 0.20 (PE/EA = 3/1), melting point: 188 – 190 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 7.92 (s, 1H), 7.83 (s, 1H), 7.69 (d, J = 8.0 Hz, 1H), 7.35 (s, 3H), 7.30 (d, J = 7.3 Hz, 3H), 7.21 (t, J = 7.4 Hz, 1H), 7.16 – 7.12 (m, 3H), 6.90 (d, J = 8.6 Hz, 1H); **^{13}C NMR** (150 MHz, DMSO-d_6) δ 146.1, 144.2, 142.4, 140.6, 133.3, 131.8, 129.6, 129.4, 126.5, 124.1, 123.0, 122.2, 119.4, 117.1, 113.6, 111.2, 68.3; **HRMS (ESI)** calcd for $C_{20}H_{15}ClN_3^+$ $[M+H]^+$: 332.0949; found: 332.0949.



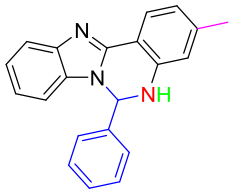
3-bromo-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3aa)

White solid (116.26 mg, 62%), R_f = 0.30 (PE/EA = 4/1), melting point: 236 – 238 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 7.90 (d, J = 8.2 Hz, 1H), 7.87 (s, 1H), 7.68 (d, J = 8.0 Hz, 1H), 7.36 – 7.30 (m, 5H), 7.20 (t, J = 7.3 Hz, 1H), 7.15 – 7.10 (m, 3H), 7.07 (s, 1H), 7.00 (d, J = 8.2 Hz, 1H); **^{13}C NMR** (150 MHz, DMSO-d_6) δ 146.5, 144.8, 144.3, 140.5, 133.3, 129.6, 129.4, 126.9, 126.4, 125.2, 122.9, 122.8, 121.4, 119.3, 117.3, 111.4, 111.1, 68.2; **HRMS (ESI)** calcd for $C_{20}H_{15}BrN_3^+$ $[M+H]^+$: 376.0444; found: 376.0442.



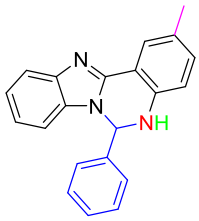
2-bromo-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3ab)

White solid (112.88 mg, 60%), R_f = 0.32 (PE/EA = 3/1), melting point: 207 – 209 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 8.04 (d, J = 2.4 Hz, 1H), 7.85 (d, J = 1.5 Hz, 1H), 7.68 (d, J = 8.0 Hz, 1H), 7.40 (dd, J = 8.7, 2.4 Hz, 1H), 7.35 – 7.28 (m, 5H), 7.20 (td, J = 8.2, 1.7 Hz, 1H), 7.16 – 7.09 (m, 3H), 6.84 (d, J = 8.7 Hz, 1H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 146.0, 144.2, 142.7, 140.5, 134.5, 133.3, 129.6, 129.4, 127.0, 126.5, 123.0, 119.4, 117.5, 114.0, 111.2, 109.5, 68.3; **HRMS (ESI)** calcd for $C_{20}H_{15}BrN_3^+$ $[M+H]^+$: 376.0444; found: 376.0444.



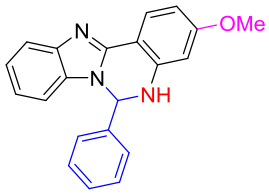
3-methyl-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3ac)

White solid (91.79 mg, 59%), R_f = 0.33 (PE/EA = 3/1), melting point: 102 – 104 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 7.86 (d, J = 7.7 Hz, 1H), 7.64 (d, J = 7.9 Hz, 1H), 7.56 (s, 1H), 7.32 (d, J = 5.9 Hz, 3H), 7.27 (d, J = 7.1 Hz, 2H), 7.17 (t, J = 7.2 Hz, 2H), 7.09 (t, J = 7.6 Hz, 1H), 7.06 (s, 1H), 6.68 – 6.66 (m, 2H), 2.26 (s, 3H); **^{13}C NMR** (150 MHz, DMSO-d_6) δ 147.6, 144.4, 143.6, 142.1, 141.0, 133.3, 129.3, 129.2, 126.4, 125.1, 122.6, 122.3, 119.9, 119.0, 115.4, 110.9, 110.0, 68.2, 21.9; **HRMS (ESI)** calcd for $C_{21}H_{18}N_3^+$ $[M+H]^+$: 312.1495; found: 312.1495.



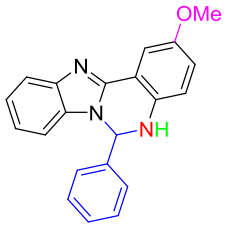
2-methyl-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3ad)

White solid (85.56 mg, 55%), R_f = 0.33 (PE/EA = 3/1), melting point: 196 – 198 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 7.80 (s, 1H), 7.66 (d, J = 7.9 Hz, 1H), 7.45 (s, 1H), 7.32 (s, 3H), 7.28 (s, 2H), 7.20 – 7.16 (m, 2H), 7.11 – 7.08 (m, 2H), 7.05 (s, 1H), 6.80 (d, J = 8.1 Hz, 1H), 2.28 (s, 3H); **^{13}C NMR** (150 MHz, DMSO-d_6) δ 147.6, 144.3, 141.4, 141.0, 133.4, 133.0, 129.3, 129.2, 126.4, 125.1, 122.6, 122.6, 122.5, 119.1, 115.5, 112.5, 111.0, 68.3, 20.7; **HRMS (ESI)** calcd for $C_{21}H_{18}N_3^+$ $[M+H]^+$: 312.1495; found: 312.1495.



3-methoxy-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3ae)

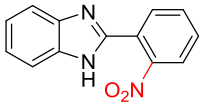
White solid (91.60 mg, 56%), R_f = 0.25 (PE/EA = 2/1), melting point: 198 – 200 ℃, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 7.95 (d, J = 8.4 Hz, 1H), 7.69 (s, 1H), 7.65 (d, J = 7.9 Hz, 1H), 7.33 (s, 5H), 7.16 – 7.15 (m, 2H), 7.08 – 7.06 (m, 2H), 6.49 – 6.47 (m, 2H), 3.76 (s, 3H); **^{13}C NMR** (150 MHz, DMSO-d_6) δ 162.9, 147.8, 145.3, 144.5, 141.0, 133.3, 129.4, 129.3, 126.9, 126.5, 122.5, 122.1, 118.8, 110.7, 106.1, 105.8, 99.4, 68.3, 55.5; **HRMS (ESI)** calcd for $C_{21}H_{18}N_3O^+$ $[M+H]^+$: 328.1444; found: 328.1444.



2-methoxy-6-phenyl-5,6-dihydrobenzo[4,5]imidazo[1,2-*c*]quinazoline (3af)

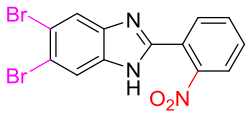
White solid (85.12 mg, 52%), R_f = 0.16 (PE/EA = 3/1), melting point: 89 – 90 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 7.67 (d, J = 8.0 Hz, 1H), 7.51 (d, J = 2.9 Hz, 1H), 7.33

– 7.25 (m, 6H), 7.21 – 7.08 (m, 3H), 7.02 (d, *J* = 1.9 Hz, 1H), 6.94 – 6.91 (m, 1H), 6.85 (d, *J* = 8.8 Hz, 1H), 3.78 (s, 3 H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 152.7, 147.5, 144.2, 140.8, 137.8, 133.5, 129.3, 129.2, 126.5, 122.7, 120.0, 119.2, 117.2, 113.3, 111.0, 108.1, 68.5, 55.8; **HRMS (ESI)** calcd for $C_{21}H_{18}N_3O^+$ $[M+H]^+$: 328.1444; found: 328.1444.



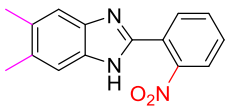
2-(2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1a)

Yellow solid (1.87 g, 78%), R_f = 0.29 (PE/EA = 1/1), melting point: 225 – 227 ℃ (Lit.[5] 259 – 262 ℃), CAS Registry Number: 2208-58-4, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 13.10 (s, 1H), 8.04 – 7.99 (m, 2H), 7.85 (t, *J* = 7.5 Hz, 1H), 7.74 (t, *J* = 7.5 Hz, 1H), 7.68 (s, 1H), 7.61 (s, 1H), 7.26 (s, 2H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 149.5, 147.9, 144.1, 135.1, 133.1, 131.4, 131.3, 124.8, 123.6, 122.4, 119.7, 112.2.



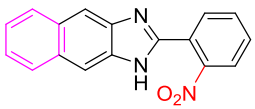
5,6-dibromo-2-(2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1v)

Yellow solid (2.57 g, 65%), R_f = 0.16 (PE/EA = 3/1), melting point: 120 – 122 ℃, (Lit.[6] 213 – 215 ℃), CAS Registry Number: 2829334-38-3, **^{1}H NMR** (400 MHz, DMSO-*d*6) δ 13.39 (s, 1H), 8.10 – 8.01 (m, 2H), 8.00 – 7.97 (m, 2H), 7.91 – 7.87 (m, 1H), 7.83 – 7.78 (m, 1H).



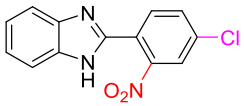
5,6-dimethyl-2-(2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1w)

White solid (1.66 g, 62%), R_f = 0.23 (PE/EA = 3/1), melting point: 120 – 122 ℃ (Lit.[6] 203 – 204 ℃), CAS registry number: 10173-74-7, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 12.78 (s, 1H), 7.99 – 7.94 (m, 2H), 7.83 (t, *J* = 7.5 Hz, 1H), 7.72 (t, *J* = 7.6 Hz, 1H), 7.42 (s, 1H), 7.33 (s, 1H), 2.33 (s, 6H).



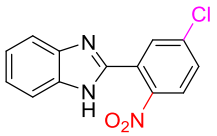
2-(2-nitrophenyl)-1*H*-naphtho[2,3-*d*]imidazole (1x)

White solid (0.95 g, 33%), R_f = 0.25 (PE/EA = 2/1), melting point: 249 – 251 ℃, (Lit.[7] 270 ℃), CAS Registry Number: 1449387-77-2, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 13.02 (s, 1H), 8.16 (s, 1H), 8.04 – 7.94 (m, 5H), 7.85 (td, J = 7.6, 1.1 Hz, 1H), 7.75 (td, J = 7.8, 1.3 Hz, 1H), 7.37 – 7.30 (m, 2H).



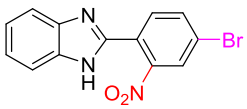
2-(4-chloro-2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1y)

Yellow solid (1.67 g, 61%), R_f = 0.33 (PE/EA = 2/1), melting point: 210 – 212 ℃, (Lit.[8] 256 – 258 ℃), CAS Registry Number: 2409006-77-3, **^{1}H NMR** (600 MHz, DMSO-d_6) δ 13.14 (s, 1H), 8.25 (d, J = 1.8 Hz, 1H), 8.04 (d, J = 8.4 Hz, 1H), 7.99 (dd, J = 8.3, 1.9 Hz, 1H), 7.67 (d, J = 6.6 Hz, 1H), 7.59 (d, J = 6.7 Hz, 1H), 7.29 (d, J = 6.0 Hz, 1H), 7.25 (d, J = 6.2 Hz, 1H).



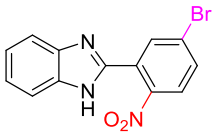
2-(5-chloro-2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1z)

Yellow solid (1.34 g, 49%), R_f = 0.3 (PE/EA = 3/1), melting point: 215 – 217 ℃, (Lit. [9] not reported), CAS Registry Number: 10173-75-8, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 13.16 (s, 1H), 8.12 (s, 2H), 7.85 (s, 1H), 7.64 (s, 2H), 7.28 (s, 2H).



2-(4-bromo-2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1aa)

Yellow solid (1.65 g, 52%), R_f = 0.33 (PE/EA = 2/1), melting point: 218 – 220 ℃, (Lit.[9] not reported), CAS Registry Number: 1392427-60-9, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 13.13 (s, 1H), 8.33 (d, J = 2.0 Hz, 1H), 8.10 (dd, J = 8.3, 2.1 Hz, 1H), 7.95 (d, J = 8.3 Hz, 1H), 7.70 – 7.57 (m, 2H), 7.31 – 7.20 (m, 2H).

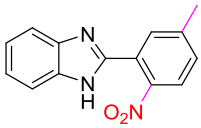


5-bromo-2-(2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1ab)

Yellow solid (1.91 g, 60%), R_f = 0.25 (PE/EA = 3/1), melting point: 204 – 206 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 13.15 (s, 1H), 8.25 (d, J = 1.7 Hz, 1H), 7.99 – 7.94 (m, 2H), 7.64 (s, 2H), 7.26 (d, J = 2.1 Hz, 2H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 148.3, 146.3, 144.0, 135.1, 134.0, 133.8, 126.7, 126.3, 126.1, 123.9, 122.6, 119.9, 112.3, **HRMS (ESI)** calcd for $C_{13}H_9BrN_3O_2^+$ $[M+H]^+$: 317.9873; found: 317.9871.

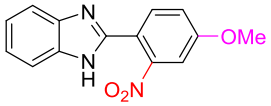
2-(4-methyl-2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1ac)

Yellow solid (1.42 g, 56%), R_f = 0.25 (PE/EA = 2/1), melting point: 242 – 245 ℃, (Lit.[8] 260 – 262 ℃), CAS Registry Number: 2409006-80-8, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 13.02 (s, 1H), 7.91 – 7.88 (m, 1H), 7.70 – 7.58 (m, 3H), 7.31 – 7.22 (m, 2H), 2.50 (s, 3H).



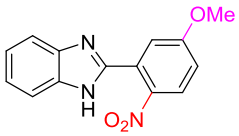
2-(5-methyl-2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1ad)

Yellow solid (1.27 g, 50%), R_f = 0.25 (PE/EA = 1/1), melting point: 226 – 228 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 12.99 (s, 1H), 7.96 (d, J = 8.3 Hz, 1H), 7.78 (s, 1H), 7.66 (s, 1H), 7.55 (d, J = 8.0 Hz, 2H), 7.25 (s, 2H), 2.49 (s, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 148.2, 147.2, 143.9, 135.1, 132.1, 131.5, 125.1, 124.9, 123.4, 122.3, 119.7, 112.1, 21.3; **HRMS (ESI)** calcd for $C_{14}H_{12}N_3O_2^+$ $[M+H]^+$: 254.0924; found: 254.0924.



2-(4-methoxy-2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1ae)

White solid (1.02 g, 38%), R_f = 0.25 (PE/EA = 1/1), melting point: 190 – 192 ℃, (Lit.[8] 242 – 244 ℃), CAS Registry Number: 942614-32-6, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 12.92 (s, 1H), 7.91 (dd, J = 9.9, 5.0 Hz, 1H), 7.63 – 7.59 (m, 2H), 7.54 (d, J = 5.8 Hz, 1H), 7.44 – 7.40 (m, 1H), 7.24 – 7.20 (m, 2H), 3.93 (s, 3H).



2-(5-methoxy-2-nitrophenyl)-1*H*-benzo[*d*]imidazole (1af)

Brown solid (1.67 g, 62%), R_f = 0.29 (PE/EA = 1/1), melting point: 207 – 209 ℃, **^{1}H NMR** (400 MHz, DMSO-d_6) δ 12.97 (s, 1H), 8.09 (d, J = 9.0 Hz, 1H), 7.67 (d, J = 4.3 Hz, 1H), 7.57 (d, J = 5.1 Hz), 7.41 (d, J = 2.2 Hz, 1H), 7.26 (dd, J = 8.5, 1.9 Hz, 3H), 3.94 (s, 3H); **^{13}C NMR** (100 MHz, DMSO-d_6) δ 162.6, 148.3, 144.0, 142.3, 135.0, 128.1, 127.5, 123.4, 122.2, 119.7, 117.1, 116.0, 112.1, 56.9; **HRMS (ESI)** calcd for $C_{14}H_{12}N_3O_3^+$ $[M+H]^+$: 270.0873; found: 270.0870.

4. References

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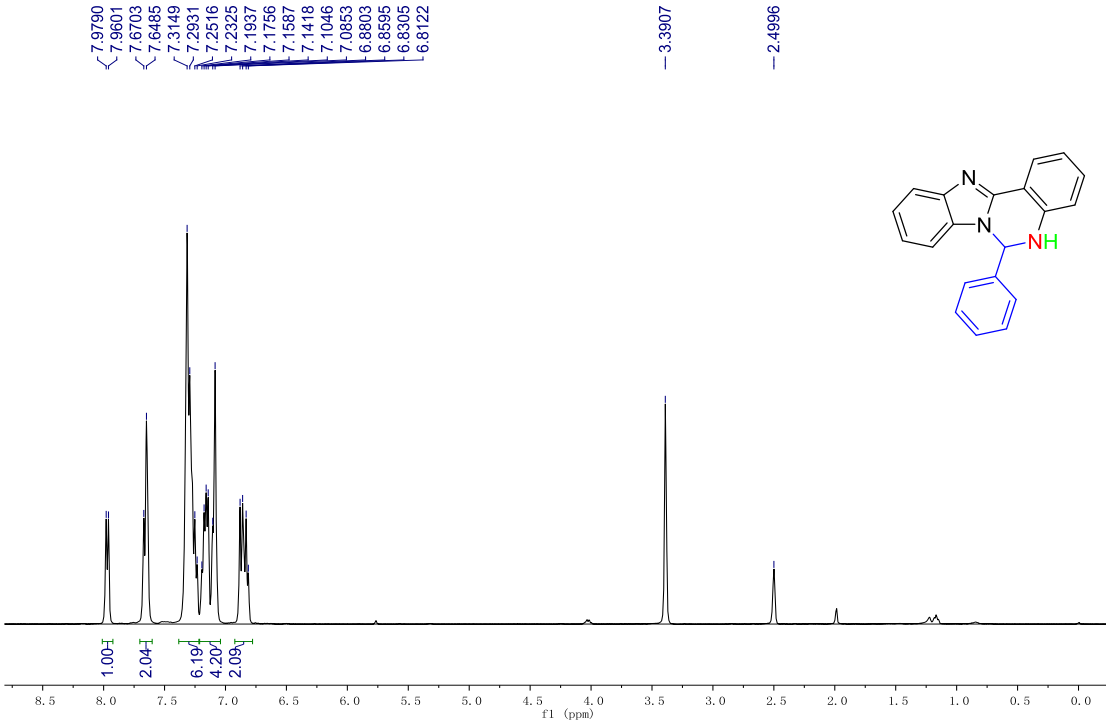
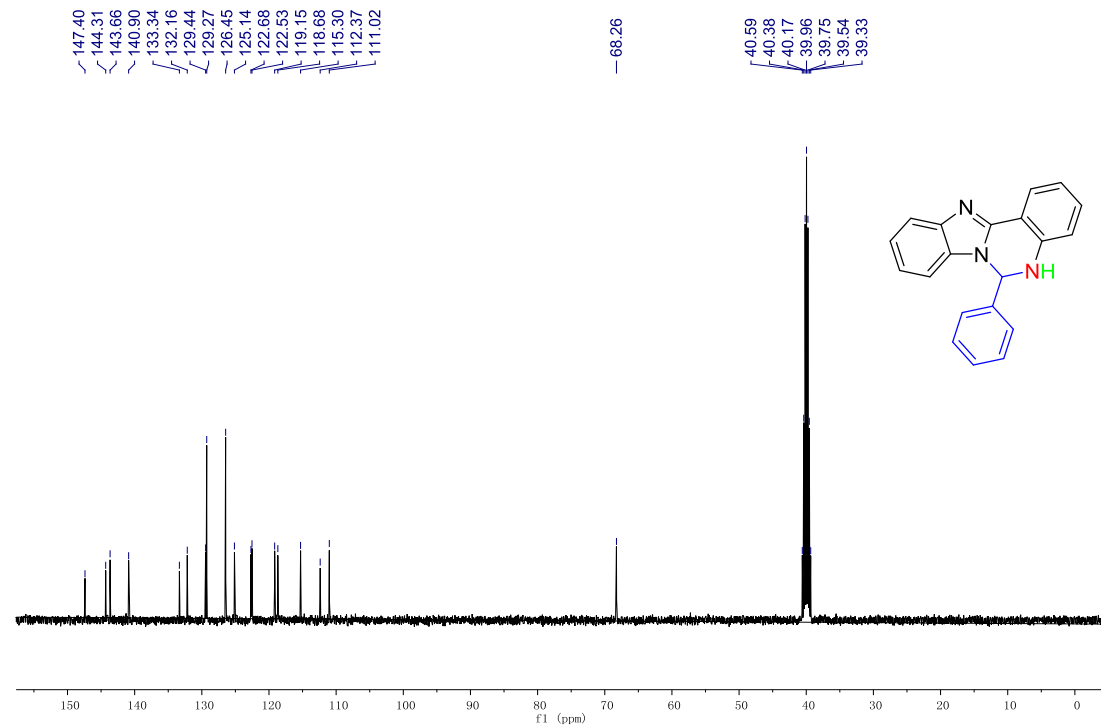
5. Copies of NMR Spectra for the Products



3a, ^{1}H NMR (400 MHz, DMSO-d_6)



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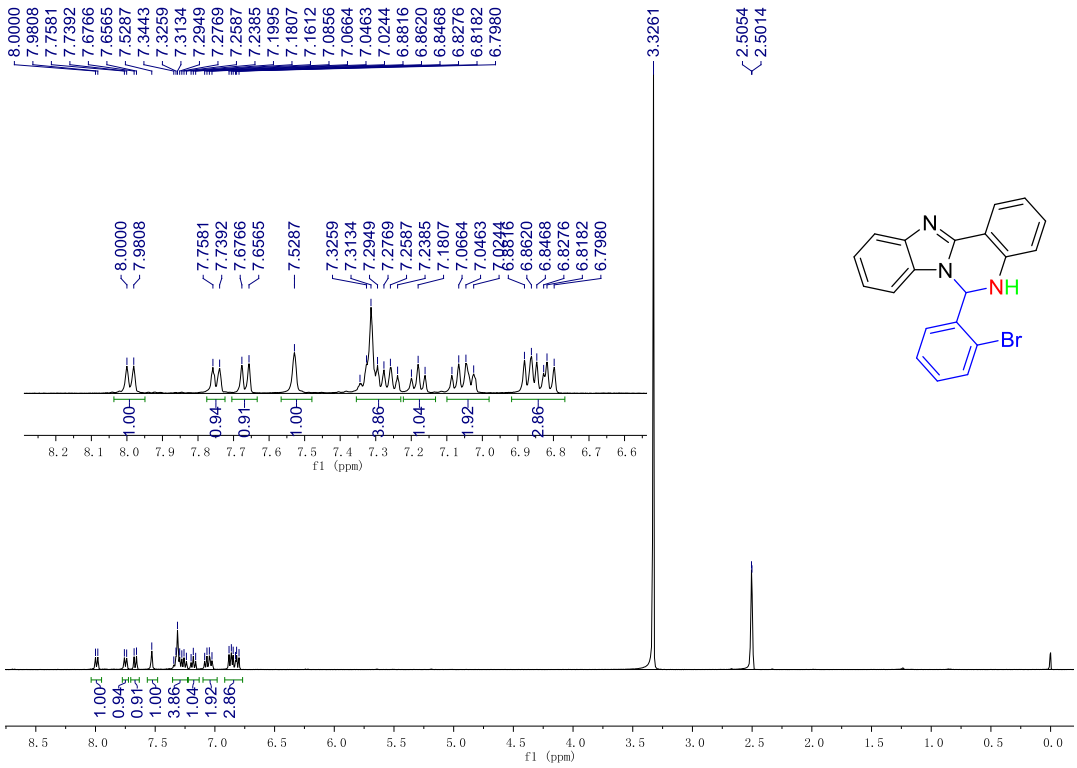
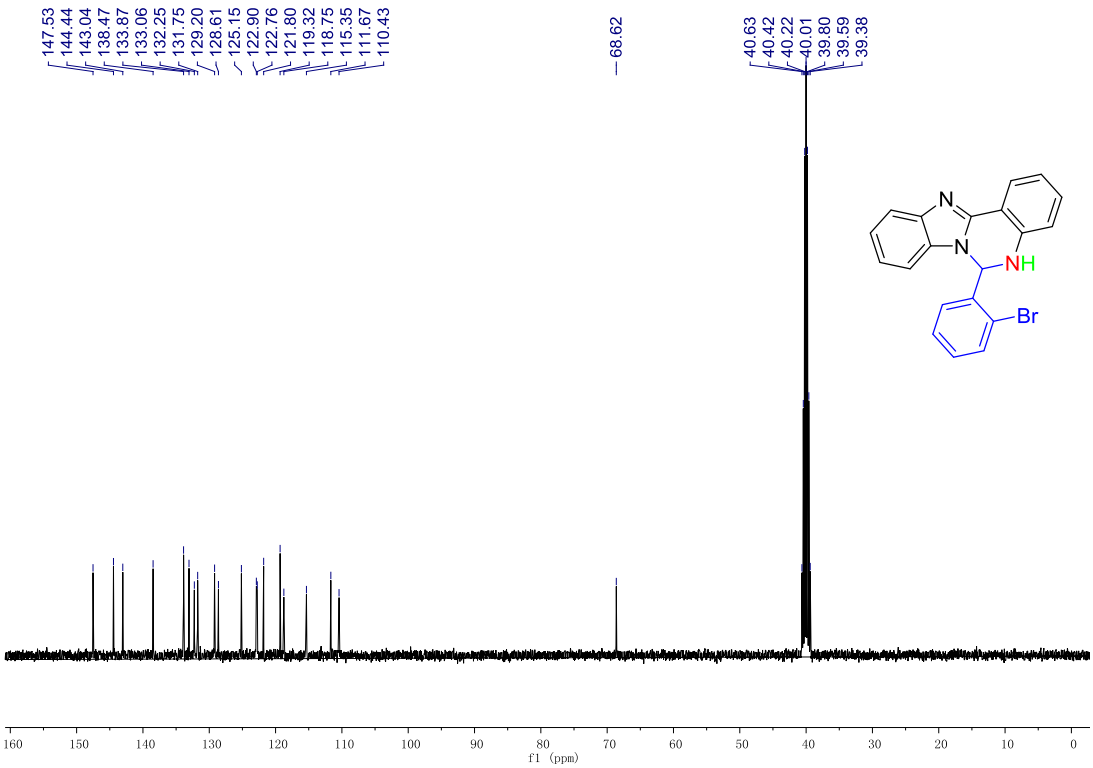
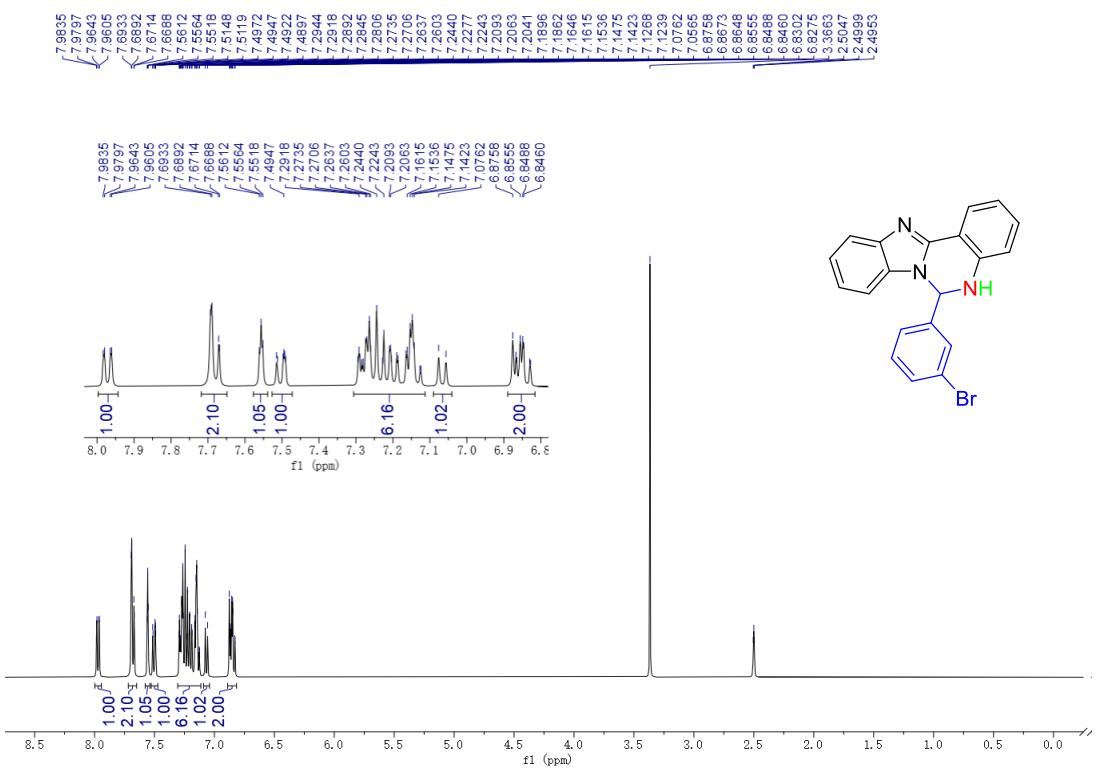


3b, ^{1}H NMR (400 MHz, DMSO-d_6)



3b, ^{13}C NMR (100 MHz, DMSO-d_6)



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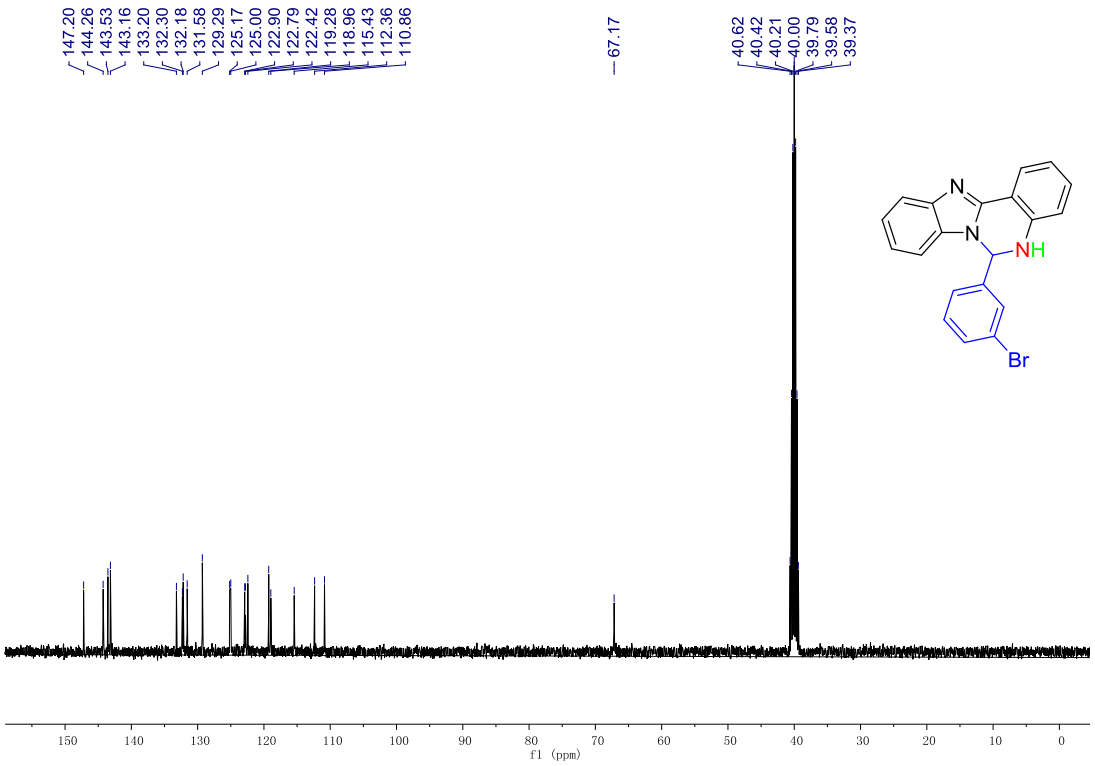

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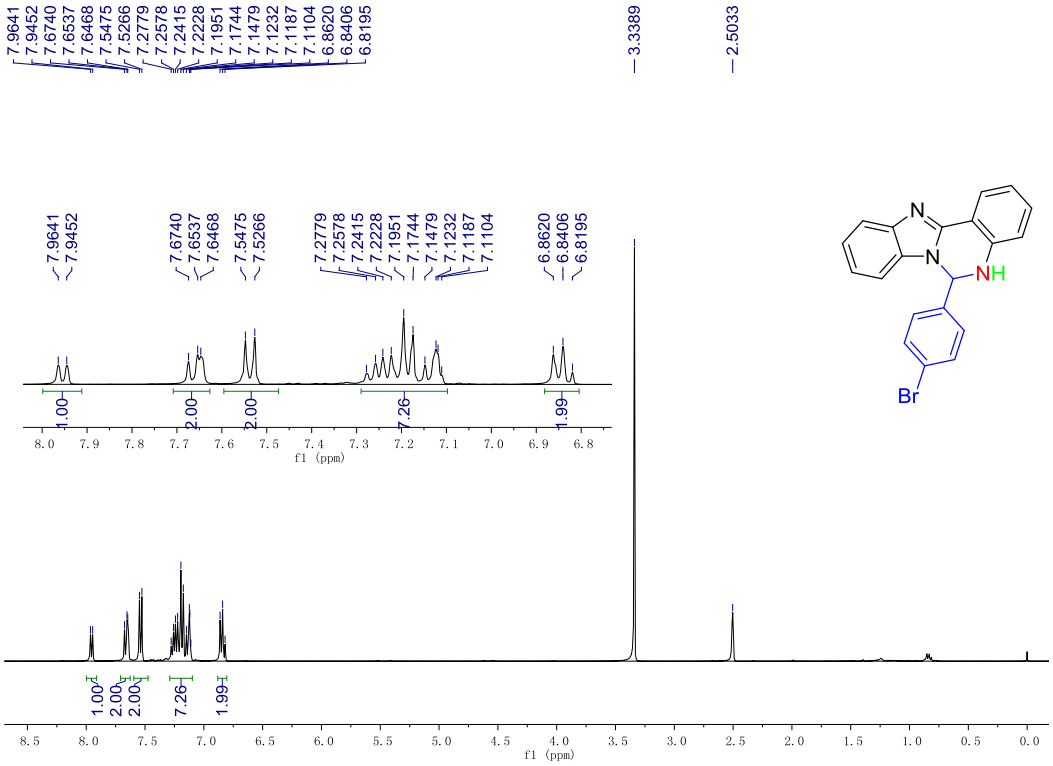

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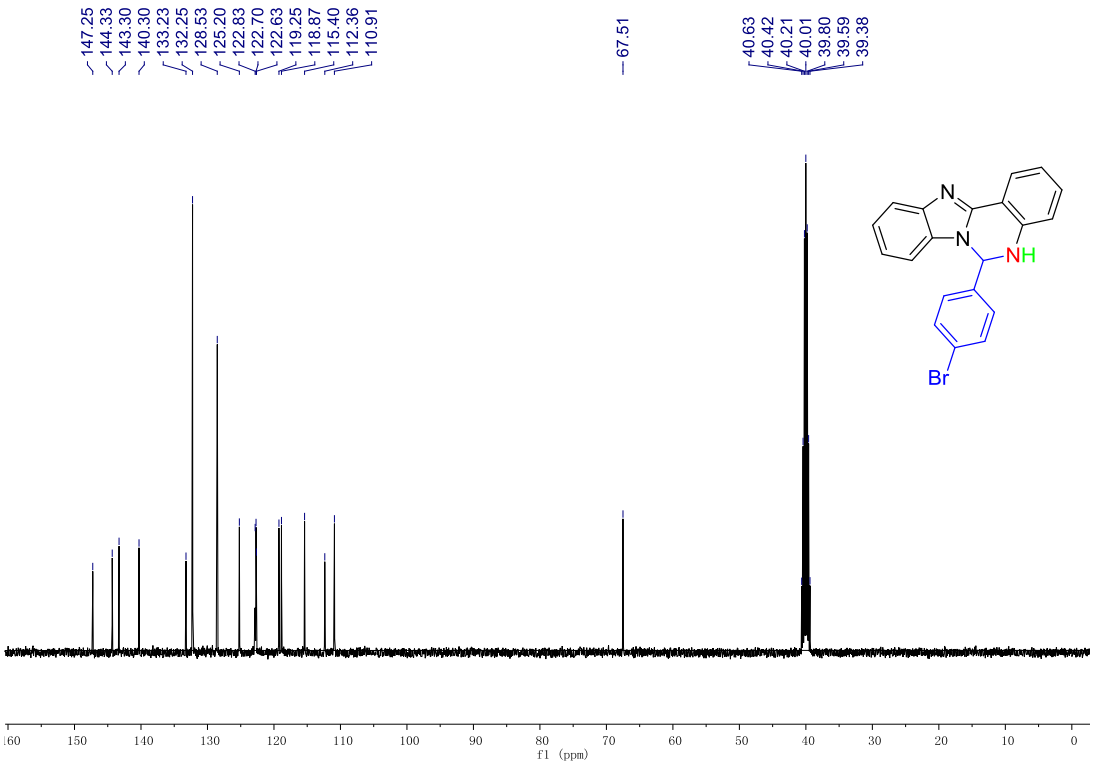
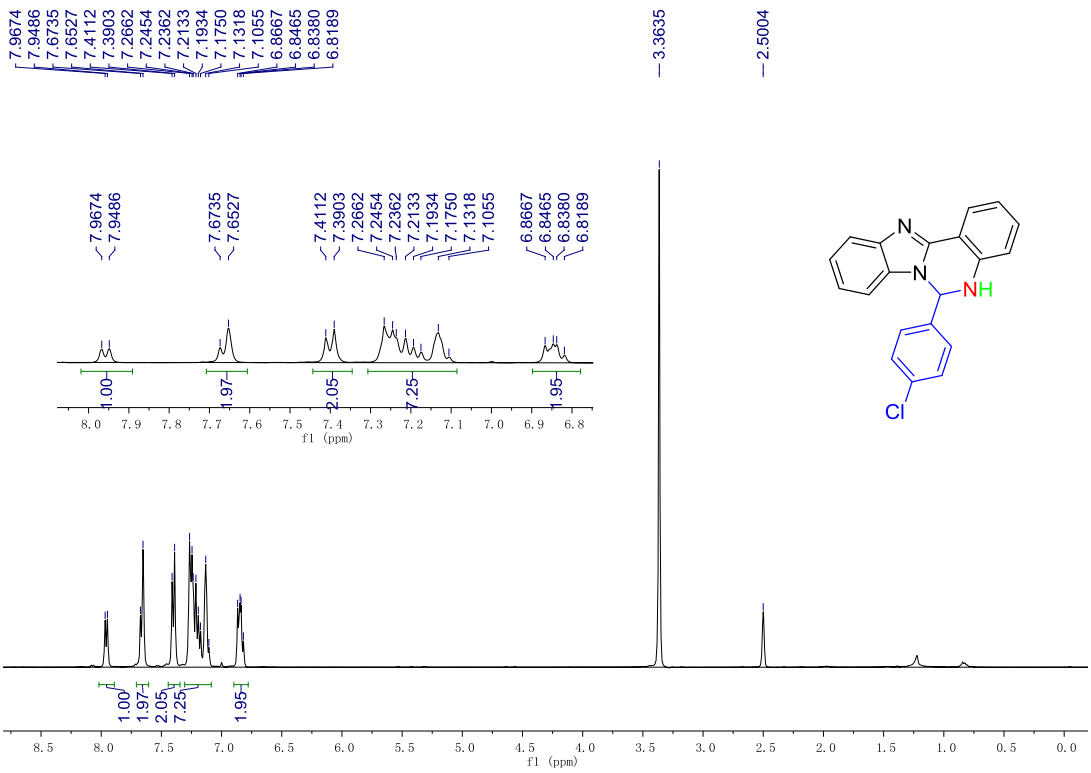
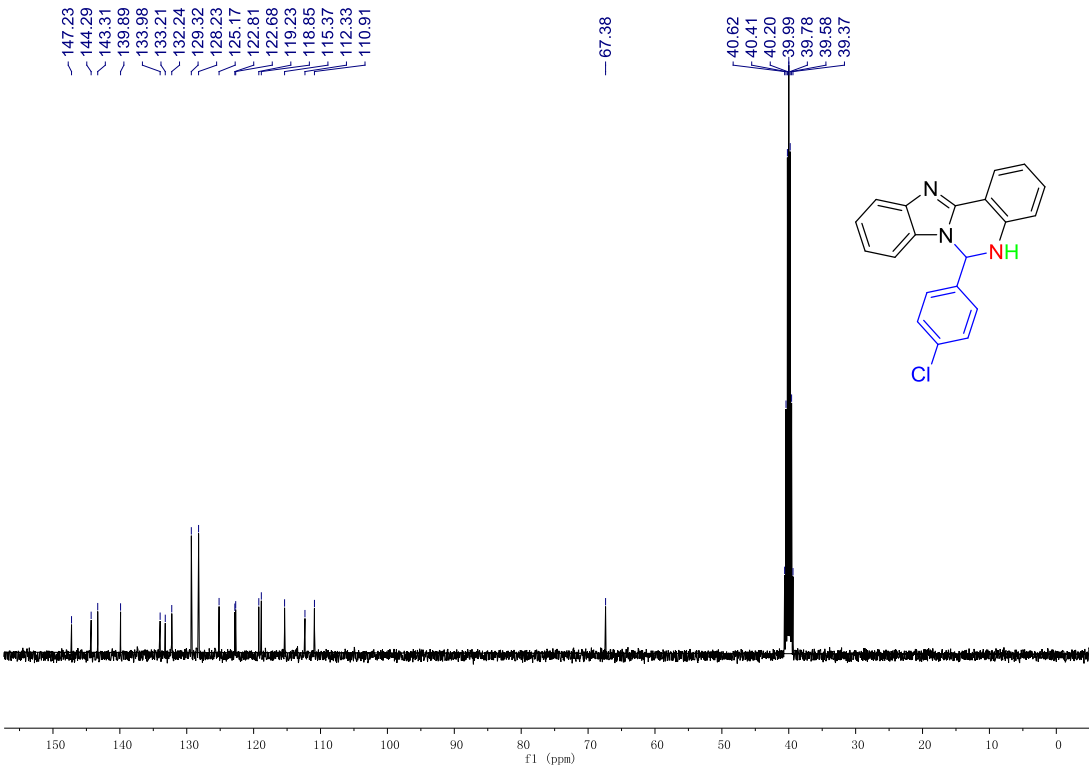


3d, ^{13}C NMR (100 MHz, DMSO-d_6)



3e, ^{1}H NMR (400 MHz, DMSO-d_6)



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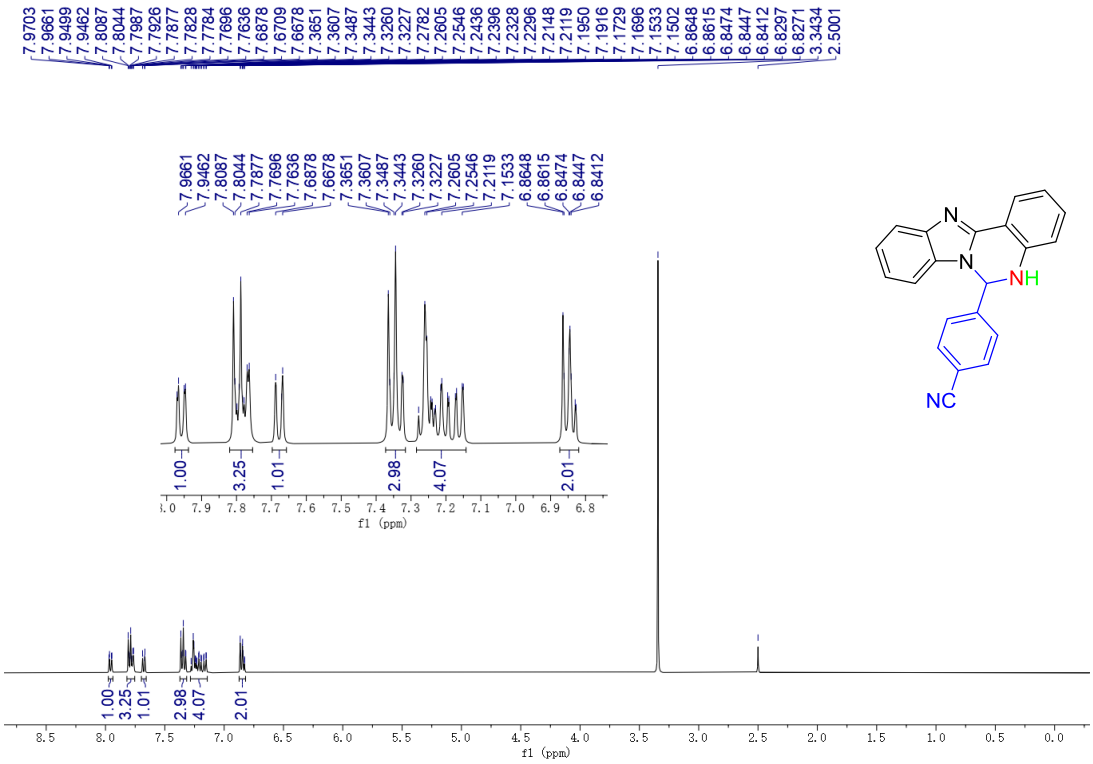

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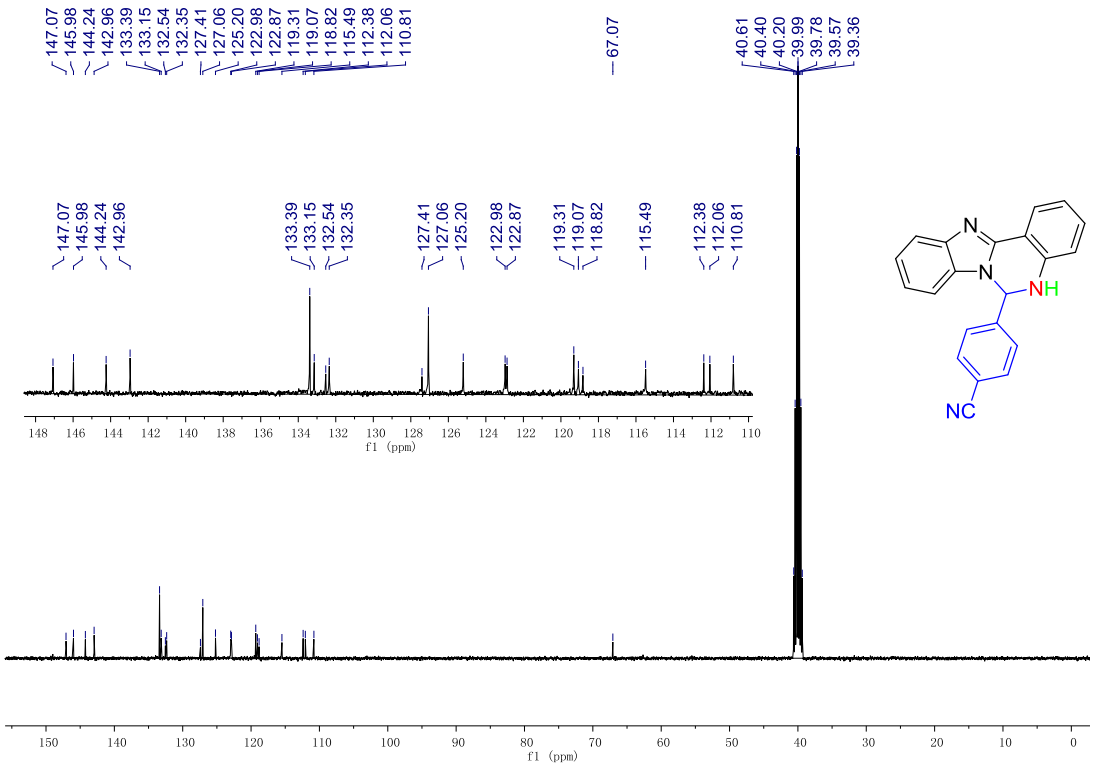

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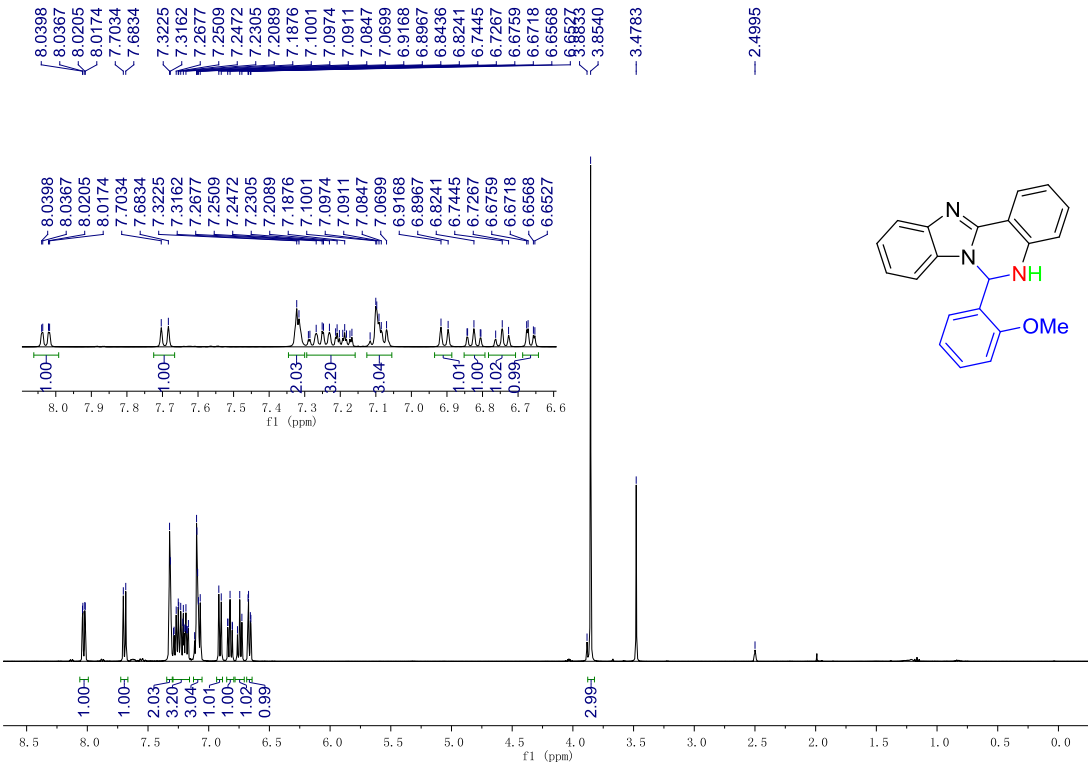

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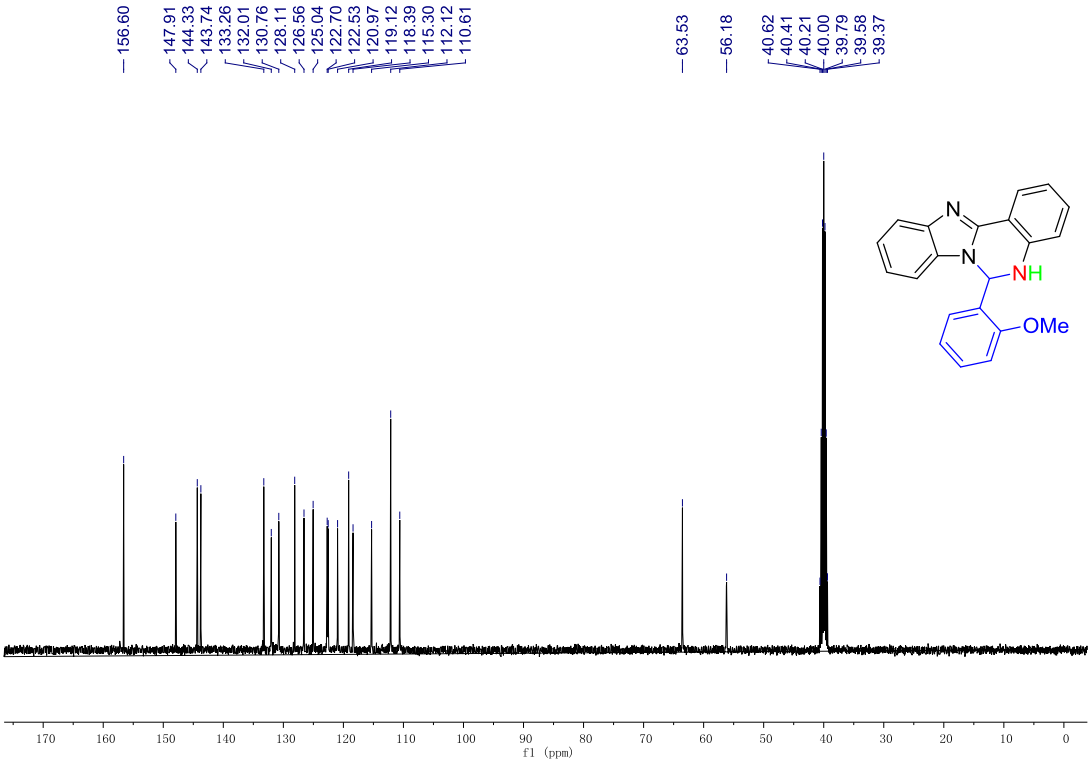

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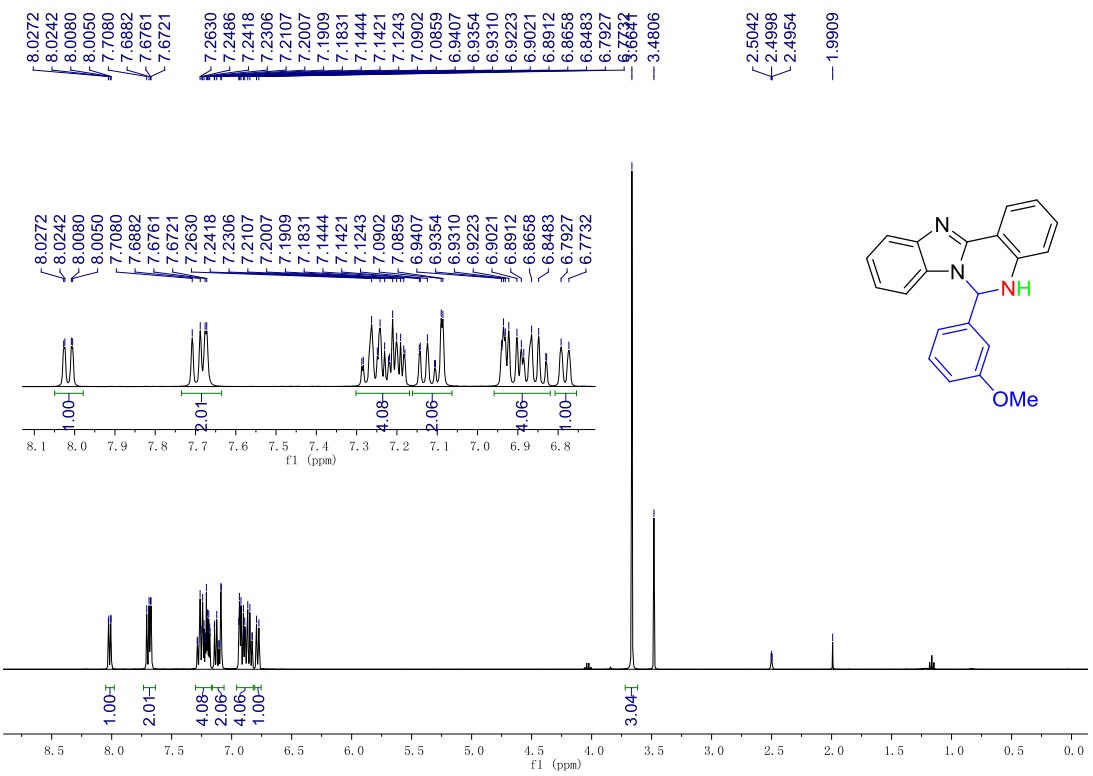
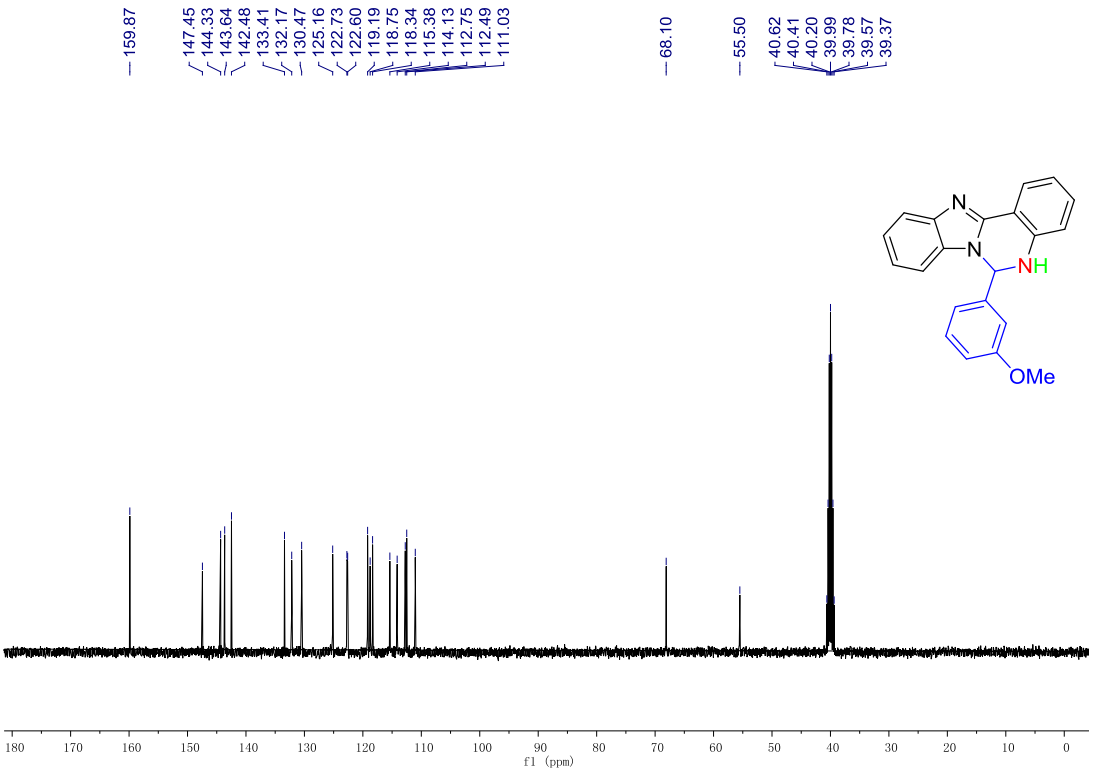
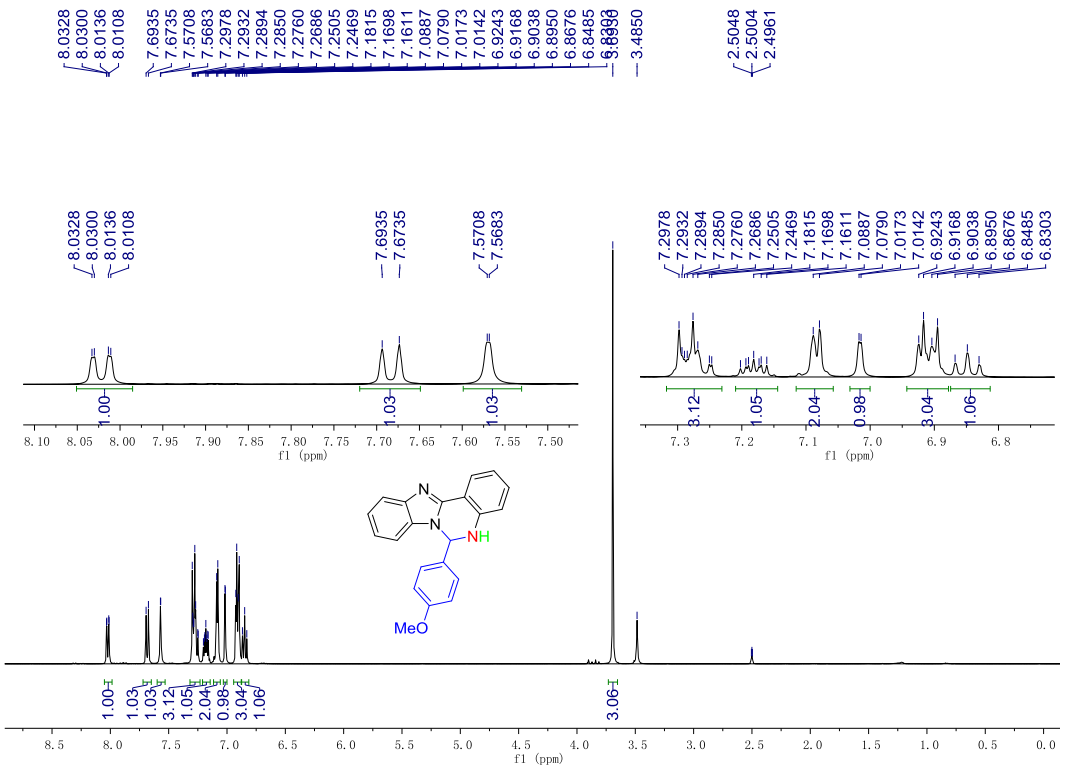


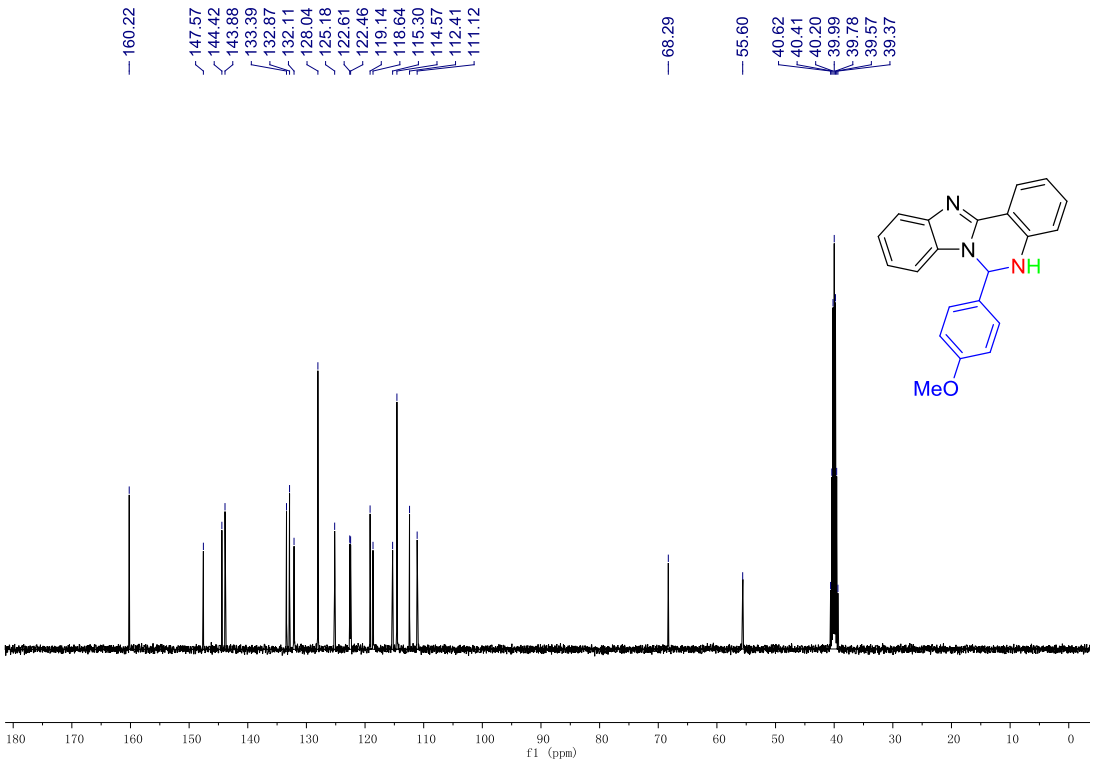
3i, ^{1}H NMR (400 MHz, DMSO-d_6)



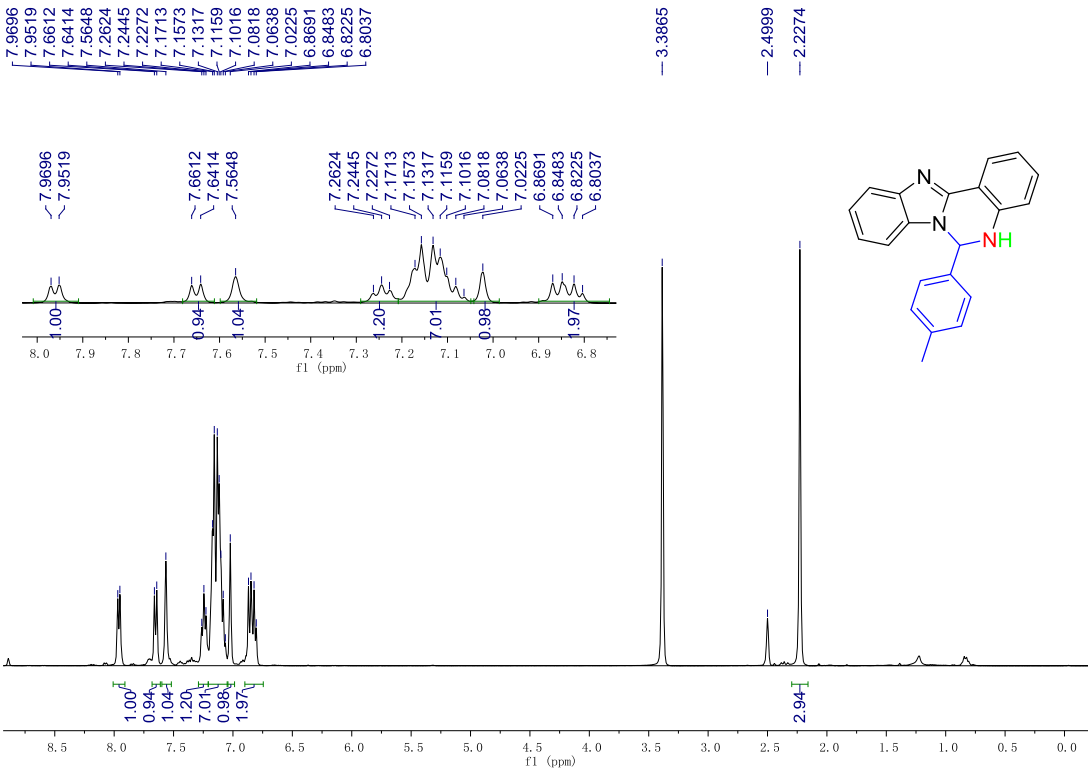
3i, ^{13}C NMR (100 MHz, DMSO-d_6)



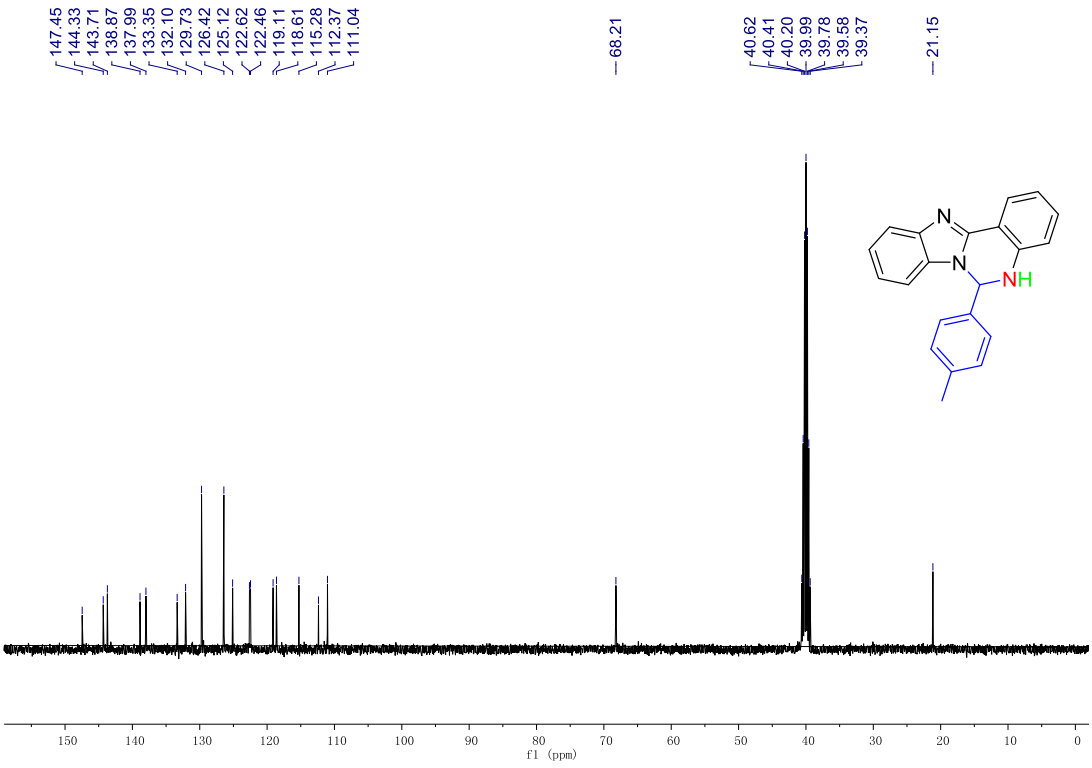
3j, ^{1}H NMR (400 MHz, DMSO-d_6)



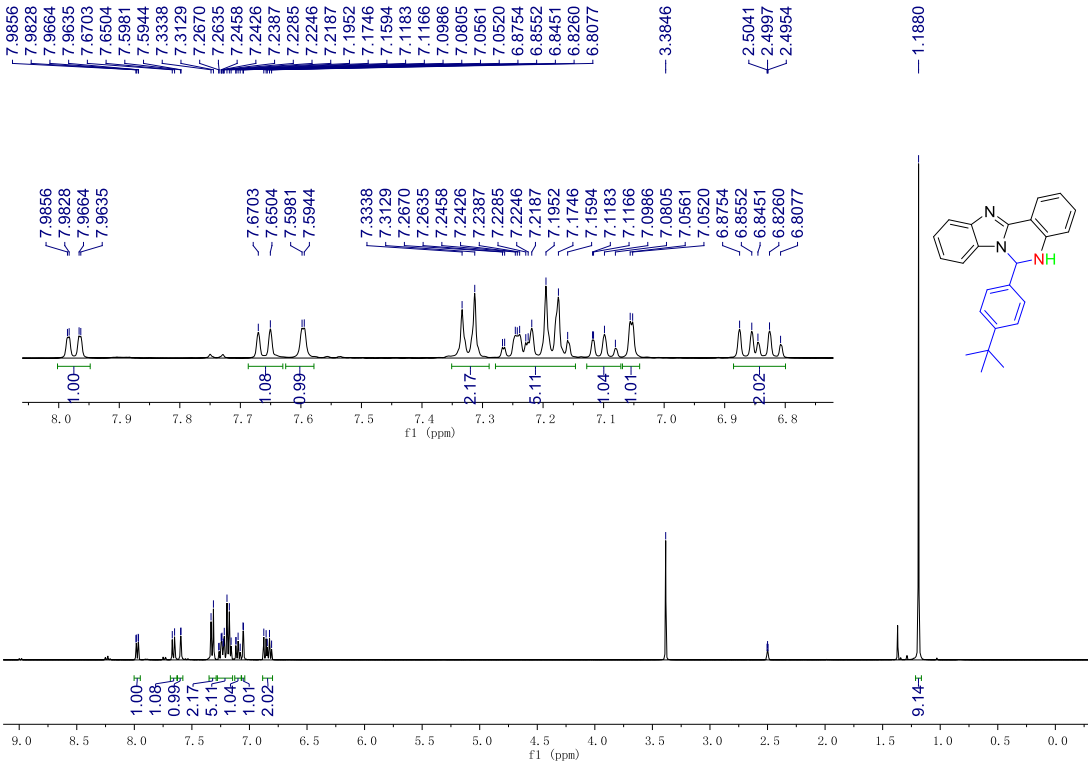
3j, ^{13}C NMR (100 MHz, DMSO-d_6)



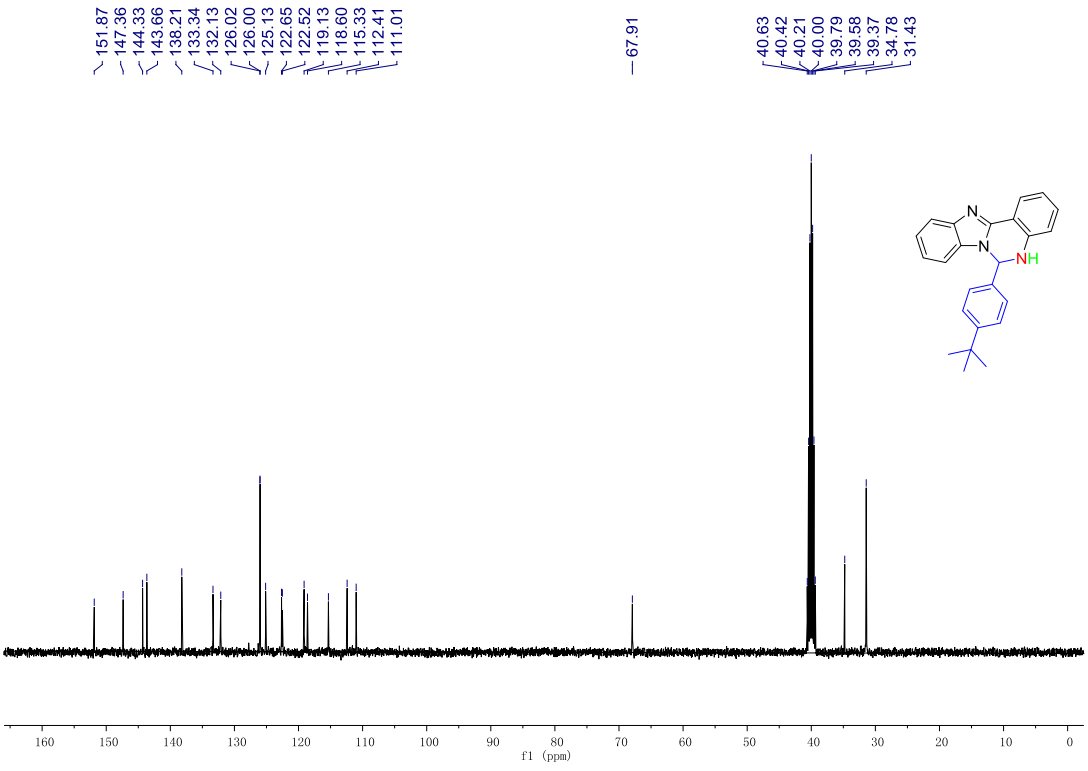
3k, ^{1}H NMR (DMSO-d_6)



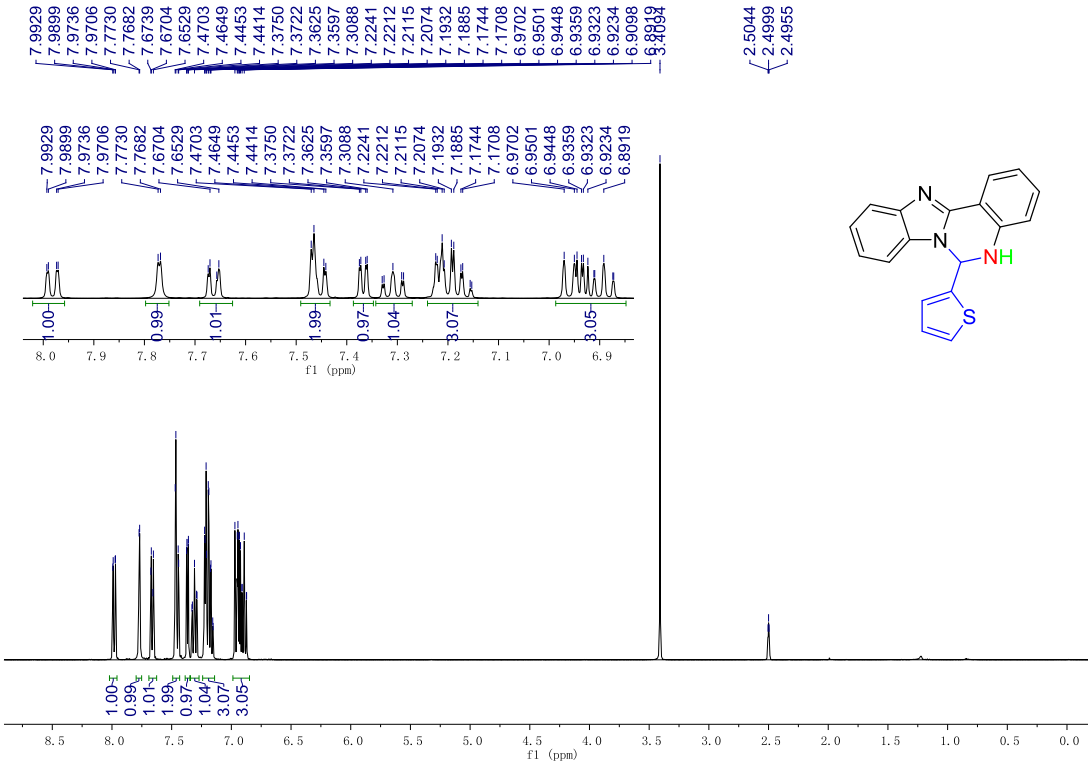
3k, ^{13}C NMR (100 MHz, DMSO-d_6)



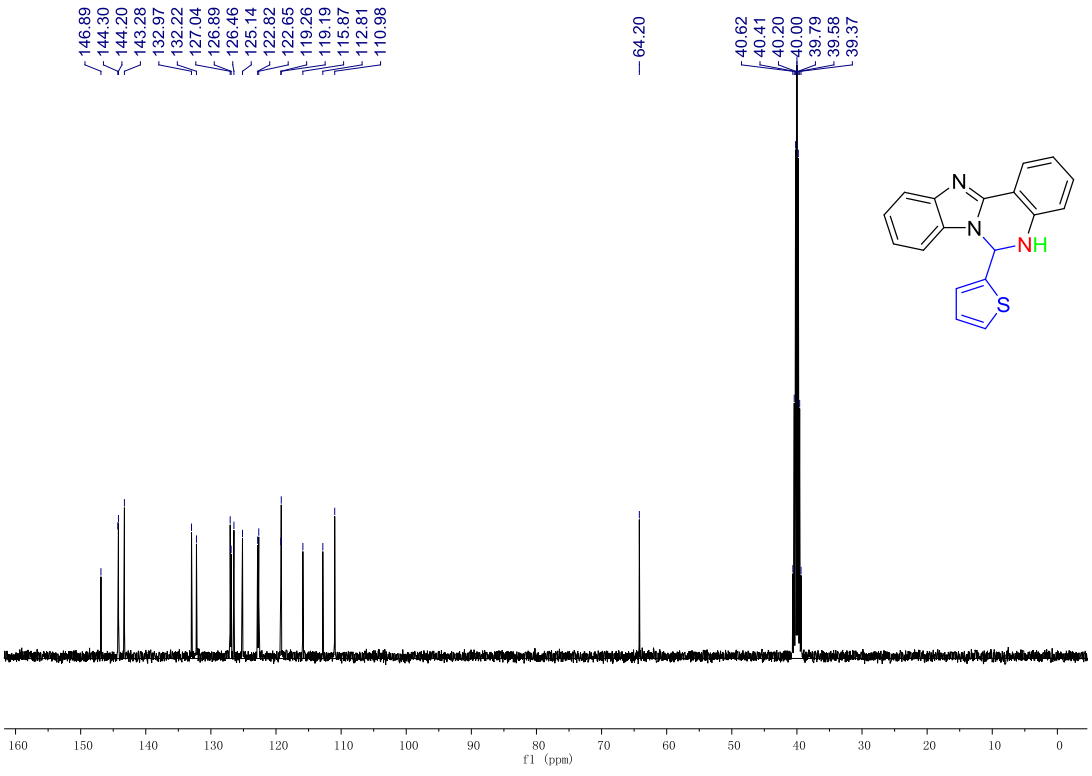
3l, ^{1}H NMR (400 MHz, DMSO-d_6)



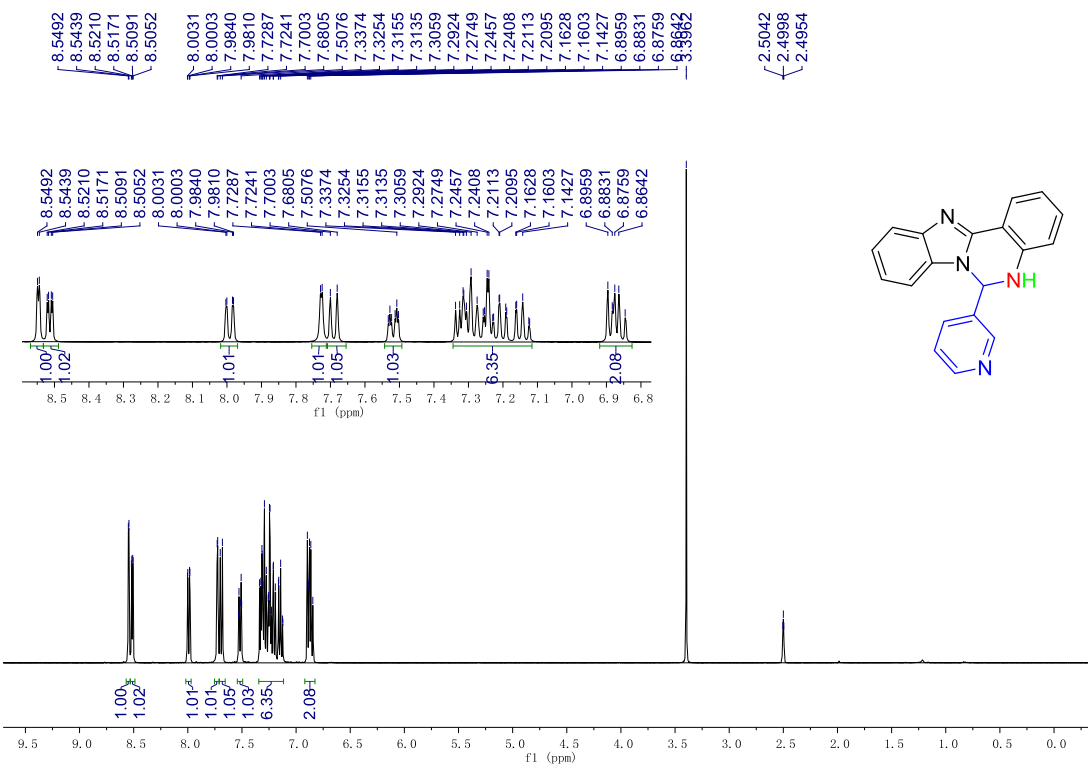
3l, ^{13}C NMR (100 MHz, DMSO-d_6)



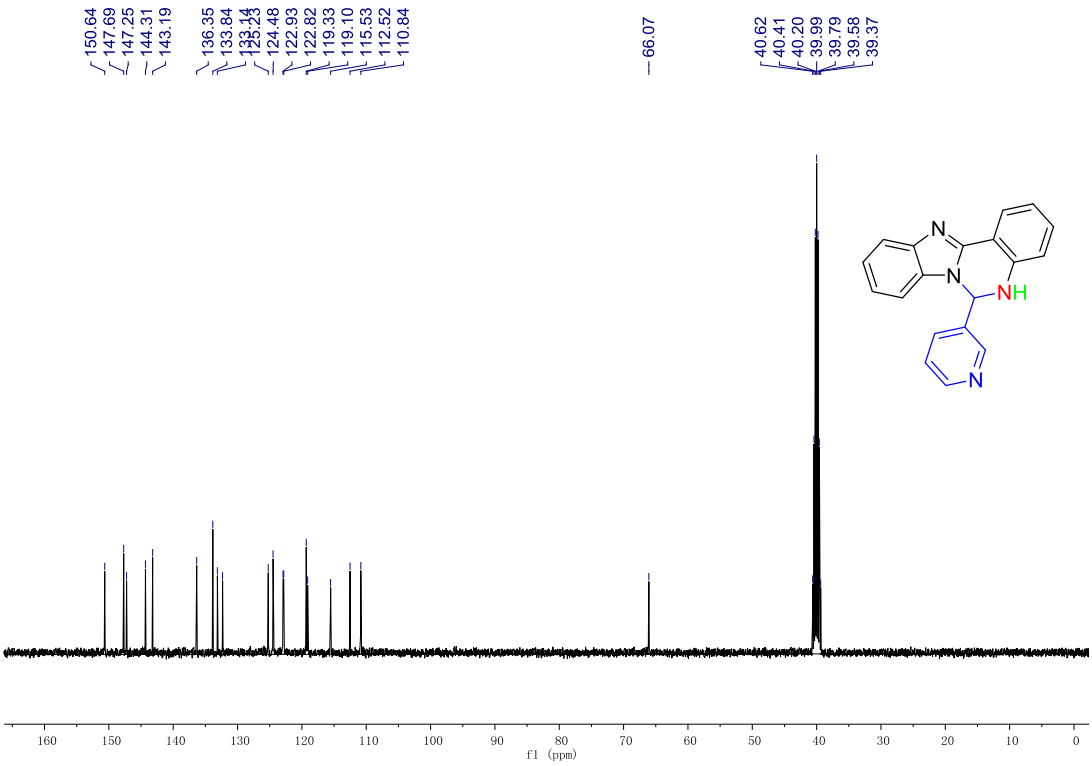
3m, ^{1}H NMR (400 MHz, DMSO-d_6)



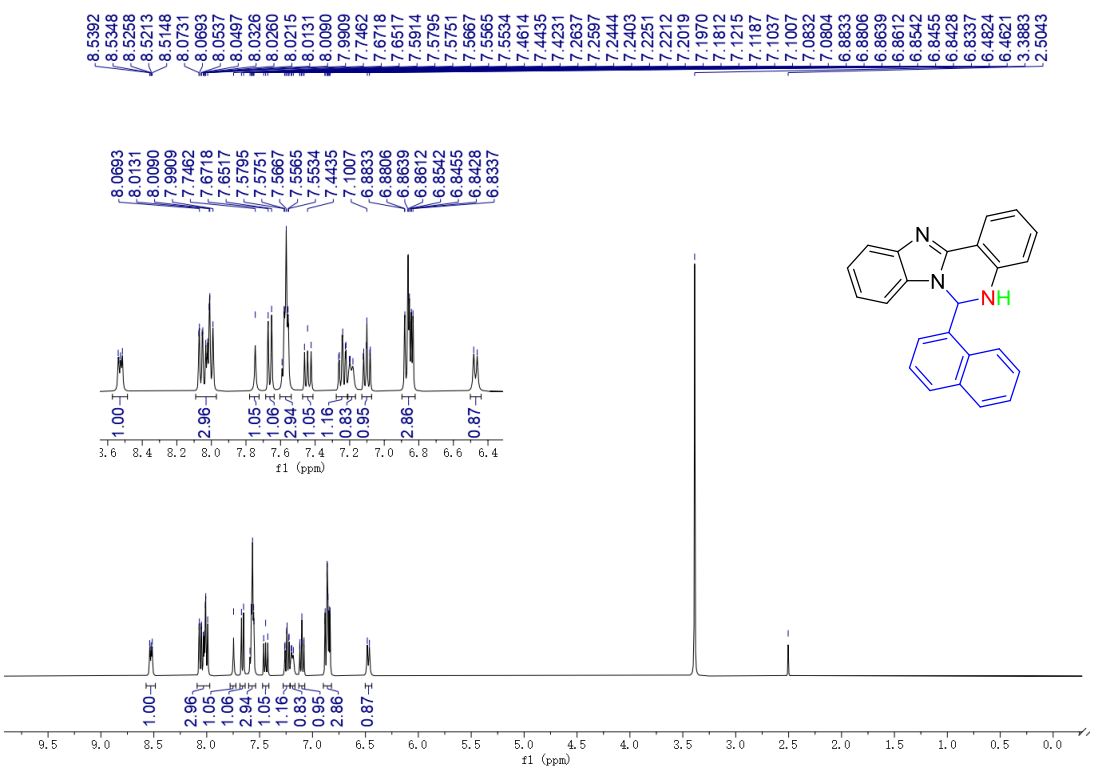
3m, ^{13}C NMR (100 MHz, DMSO-d_6)



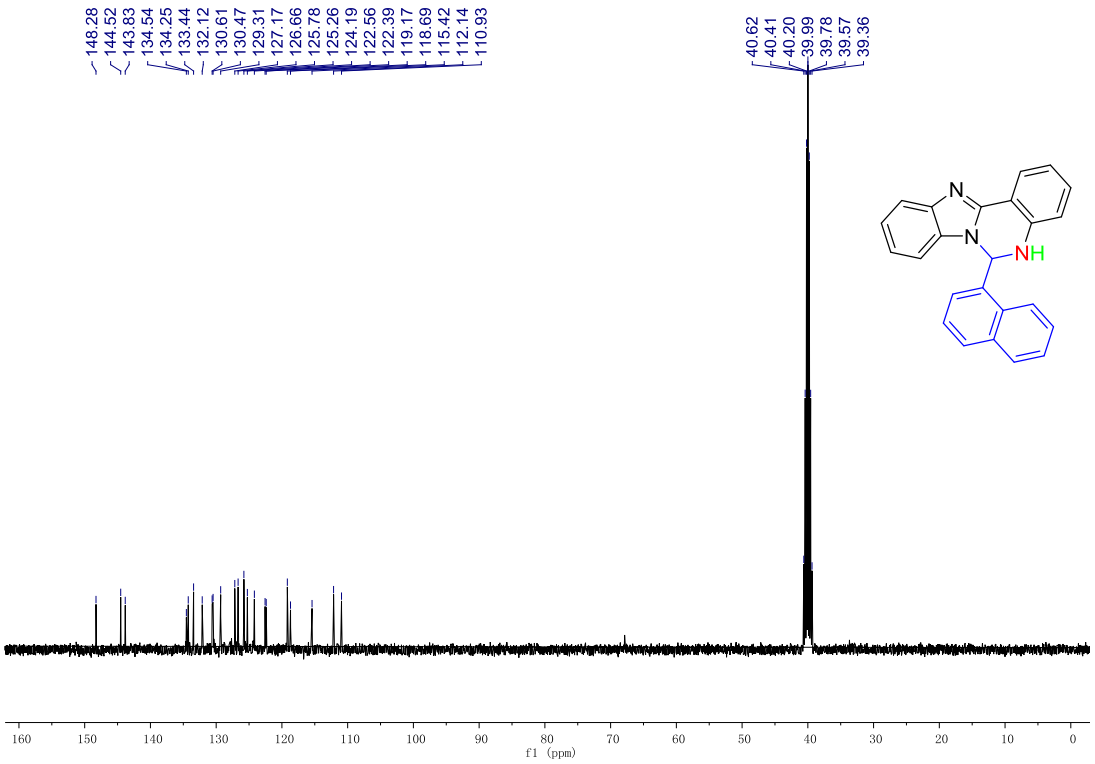
3n, ^{1}H NMR (400 MHz, DMSO-d_6)



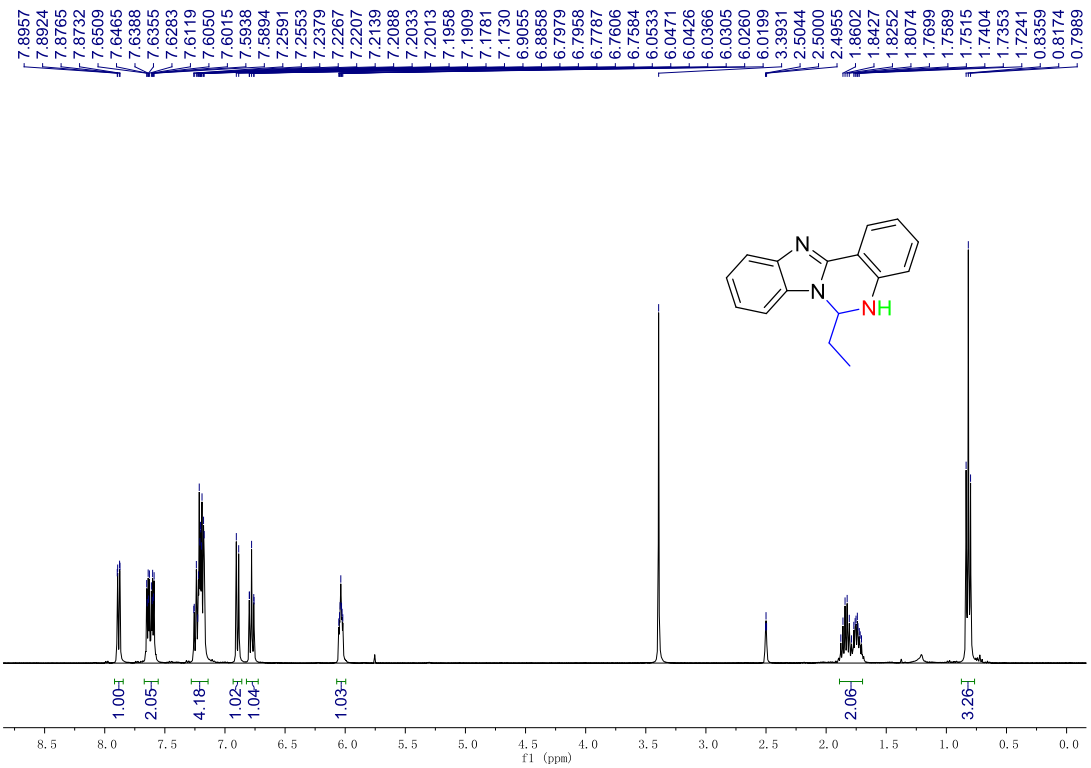
3n, ^{13}C NMR (100 MHz, DMSO-d_6)



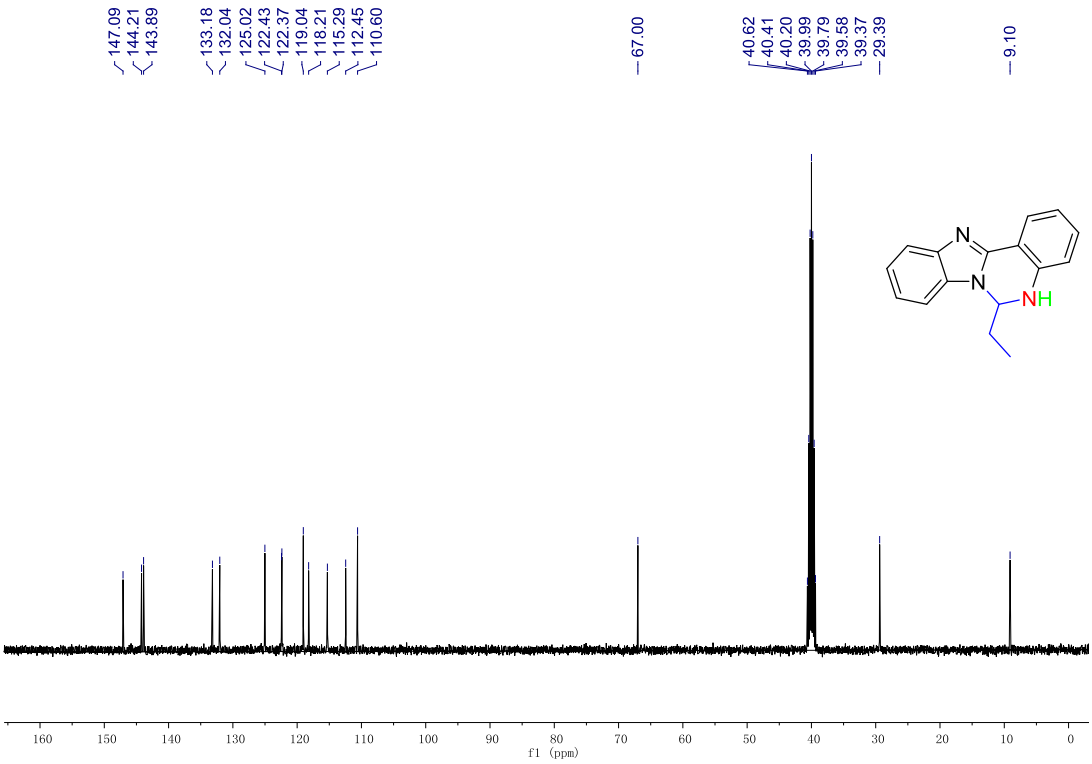
3o, ^{1}H NMR (400 MHz, DMSO-d_6)



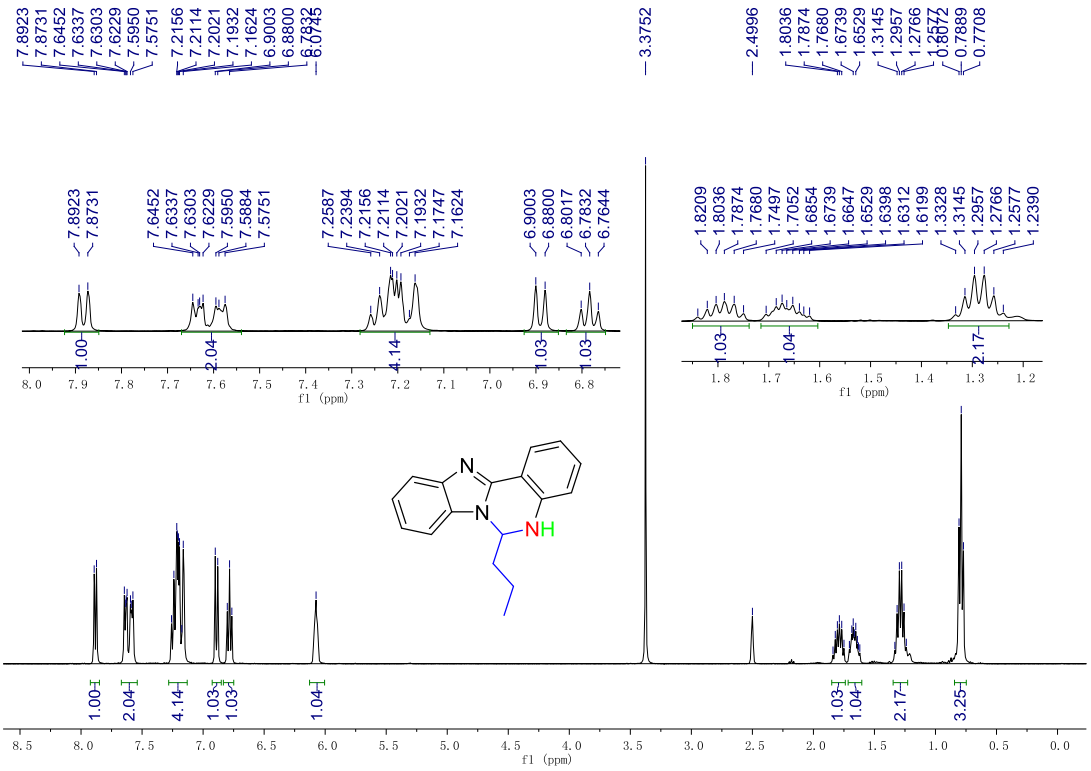
3o, ^{13}C NMR (100 MHz, DMSO-d_6)



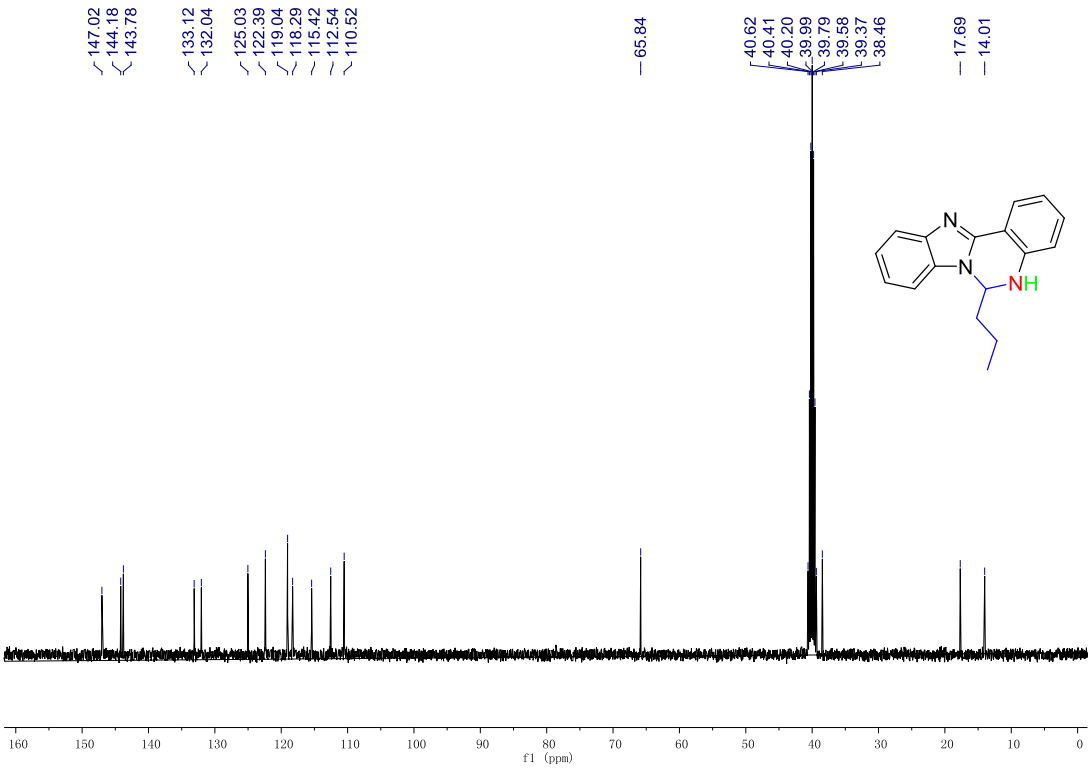
3p, ^{1}H NMR (400 MHz, DMSO-d_6)



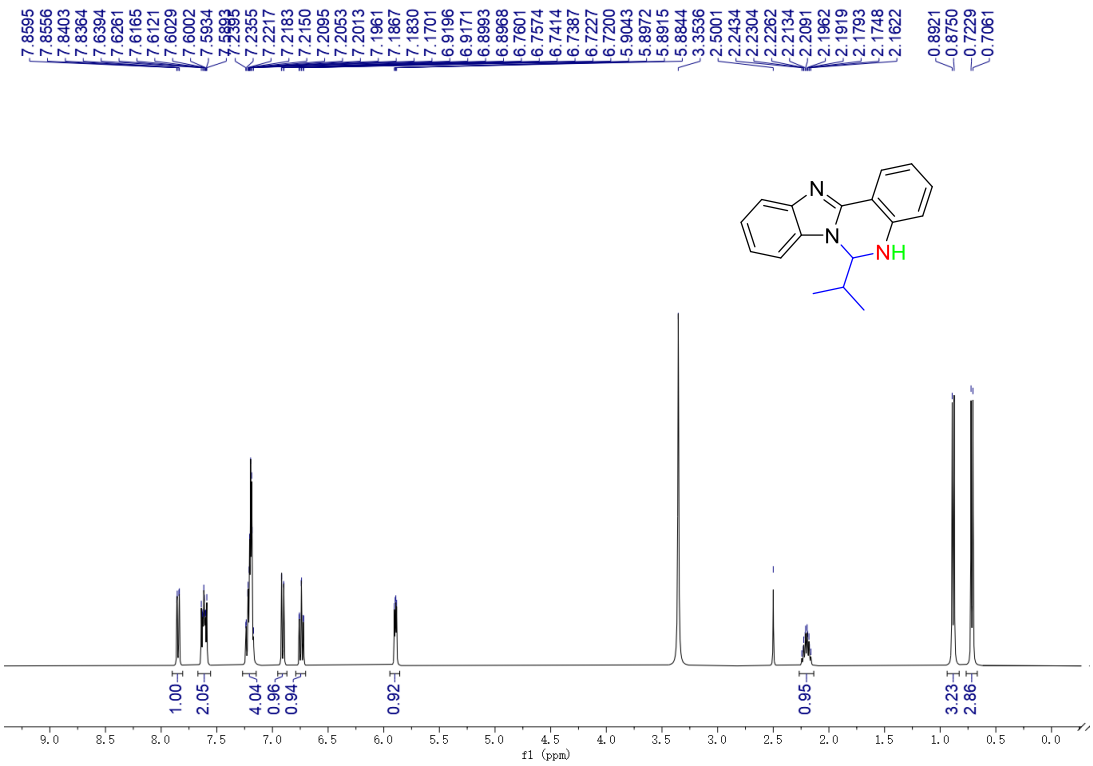
3p, ^{13}C NMR (100 MHz, DMSO-d_6)



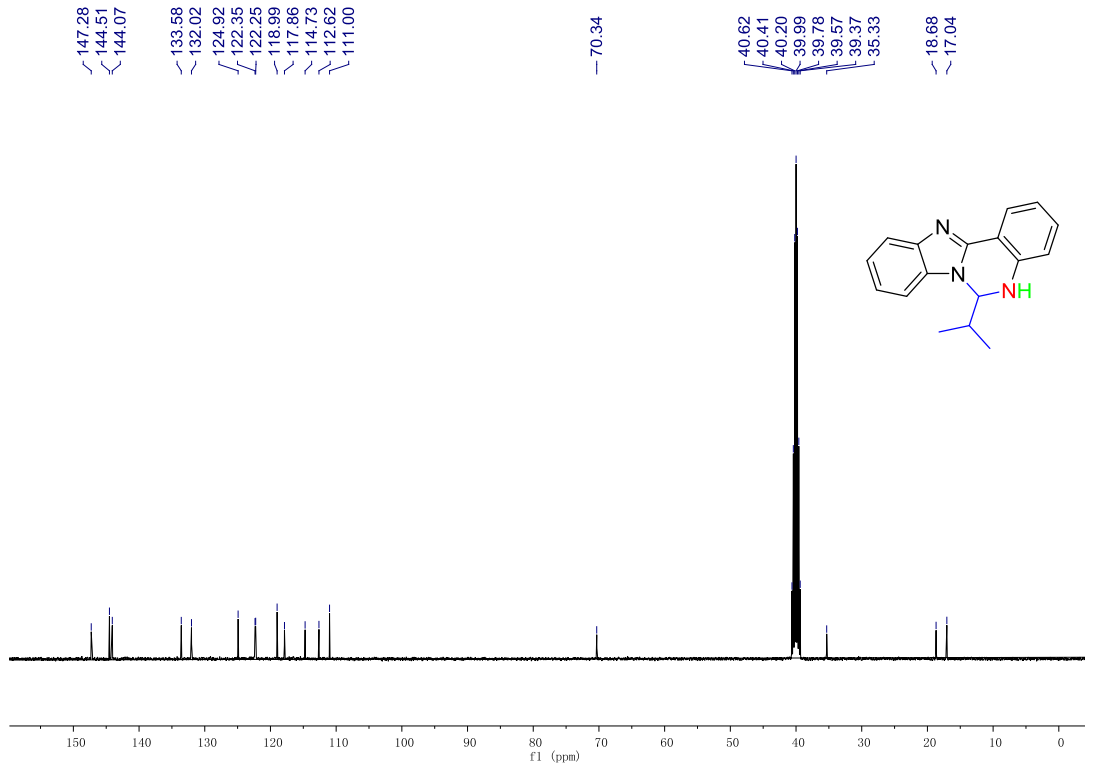
3q, ^{1}H NMR (400 MHz, DMSO-d_6)



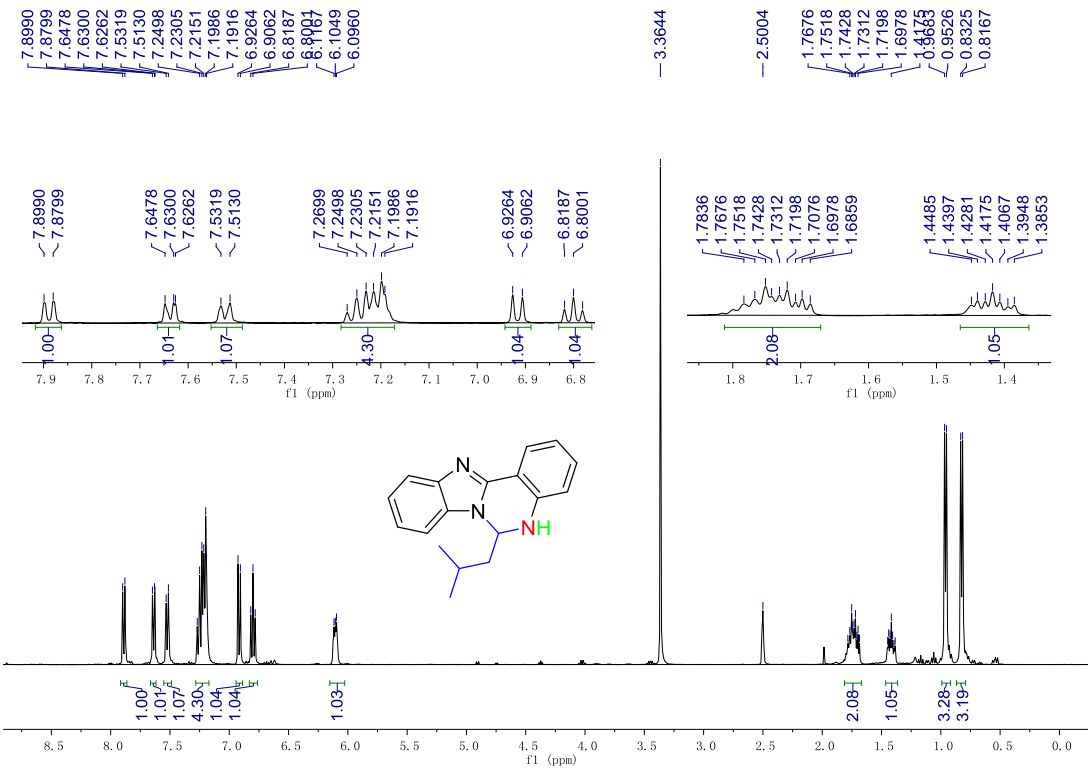
3q, ^{13}C NMR (100 MHz, DMSO-d_6)



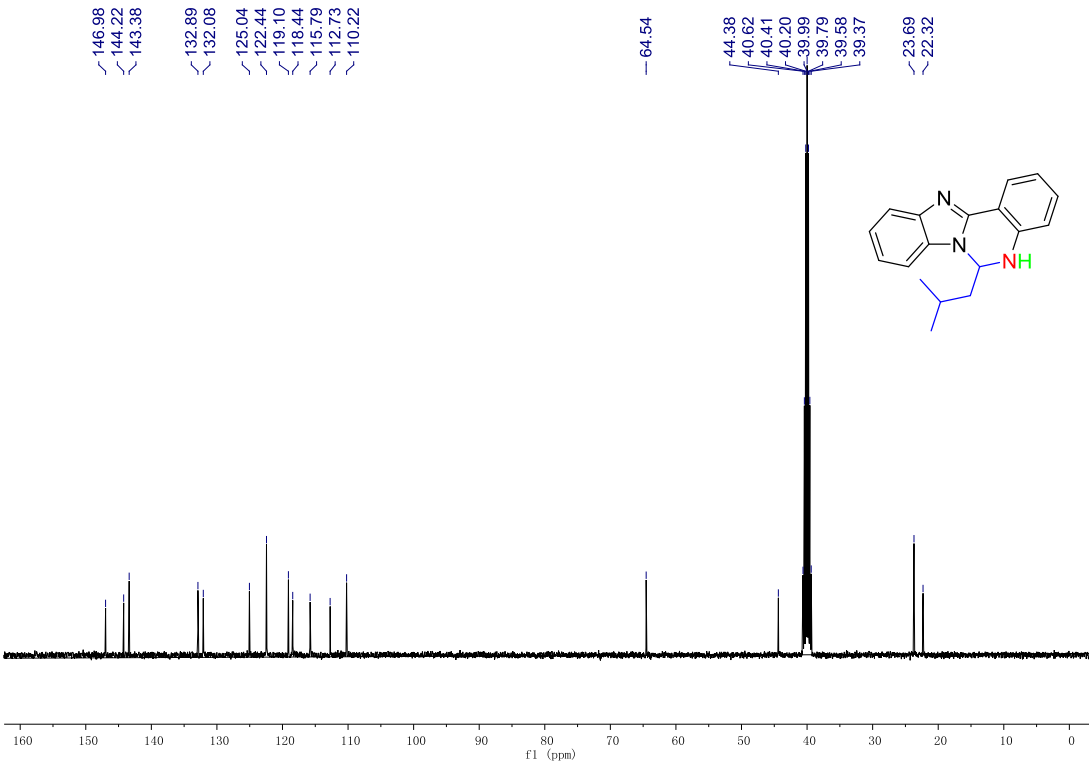
3r, ^{1}H NMR (400 MHz, DMSO-d_6)



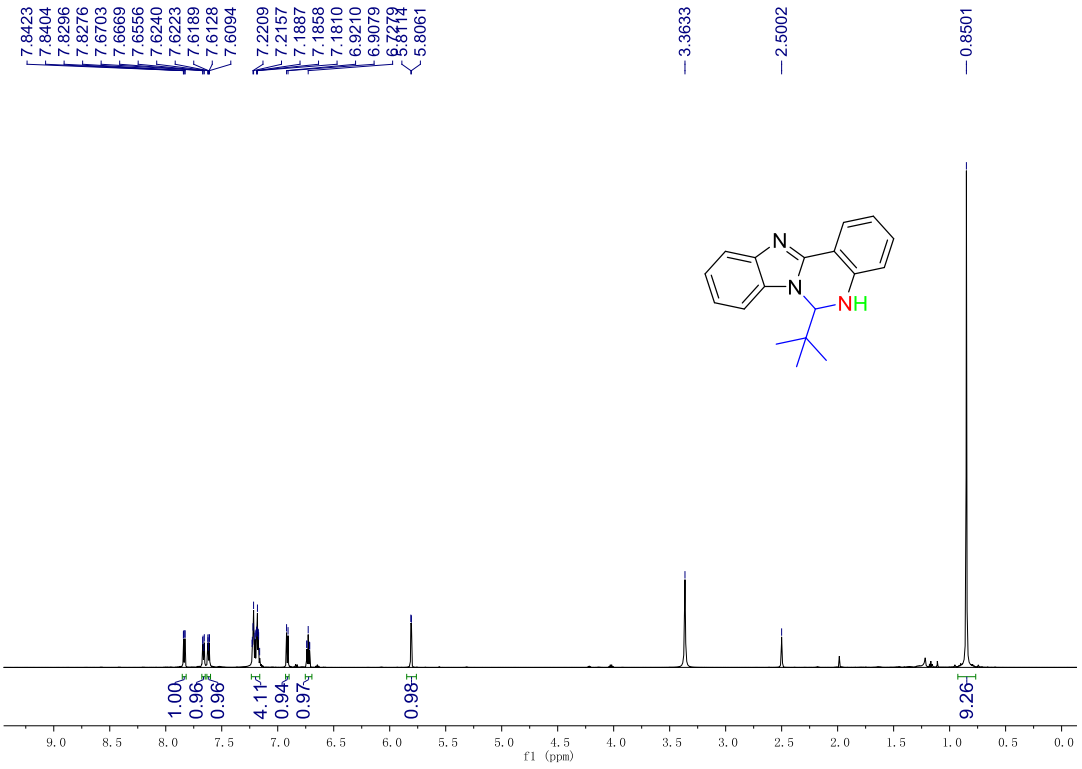
3r, ^{13}C NMR (100 MHz, DMSO-d_6)



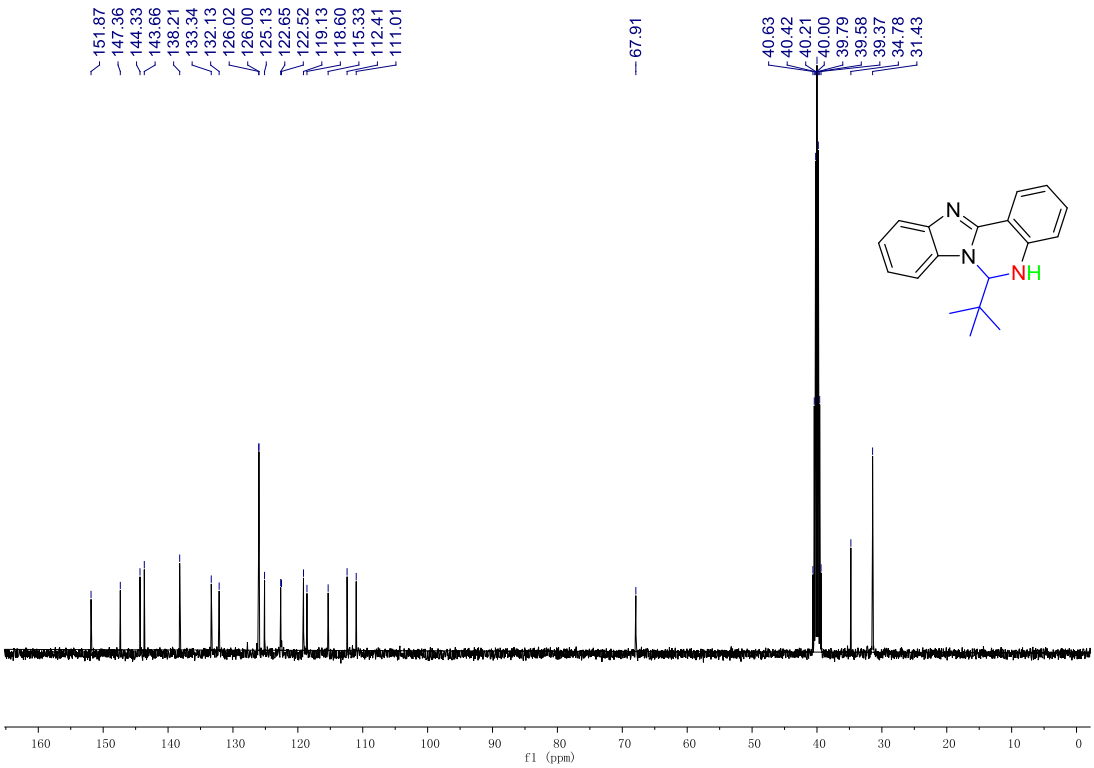
3s, ^{1}H NMR (400 MHz, DMSO-d_6)



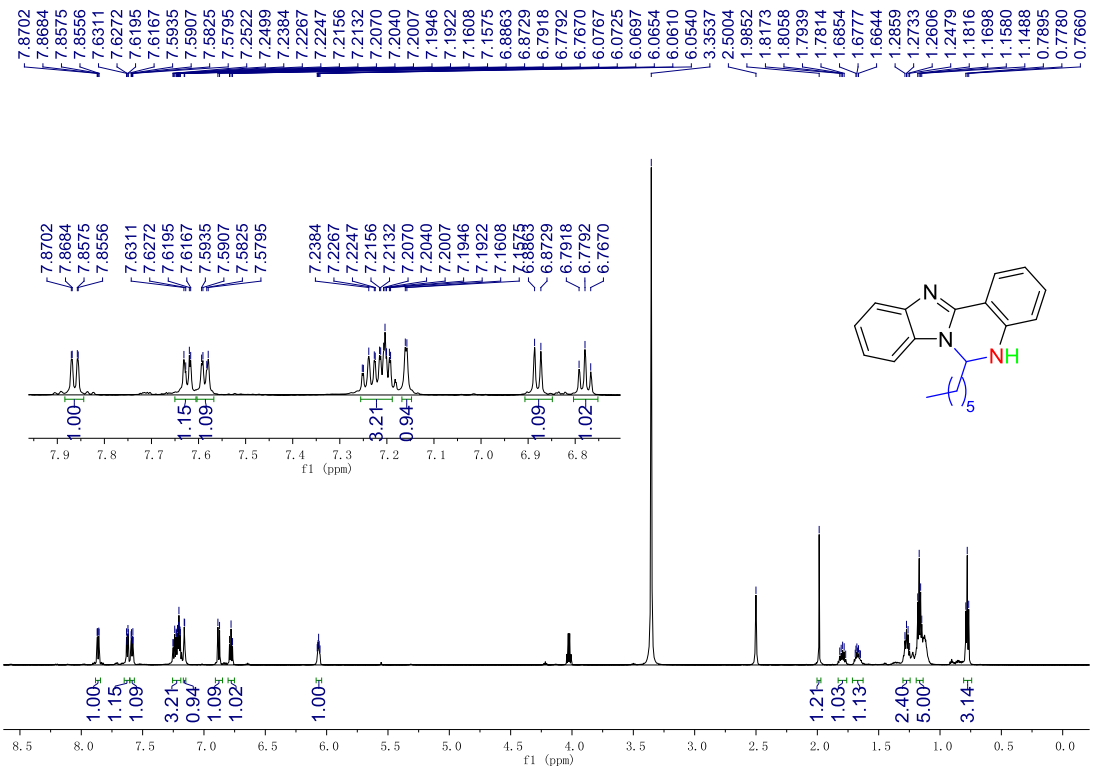
3s, ^{13}C NMR (100 MHz, DMSO-d_6)



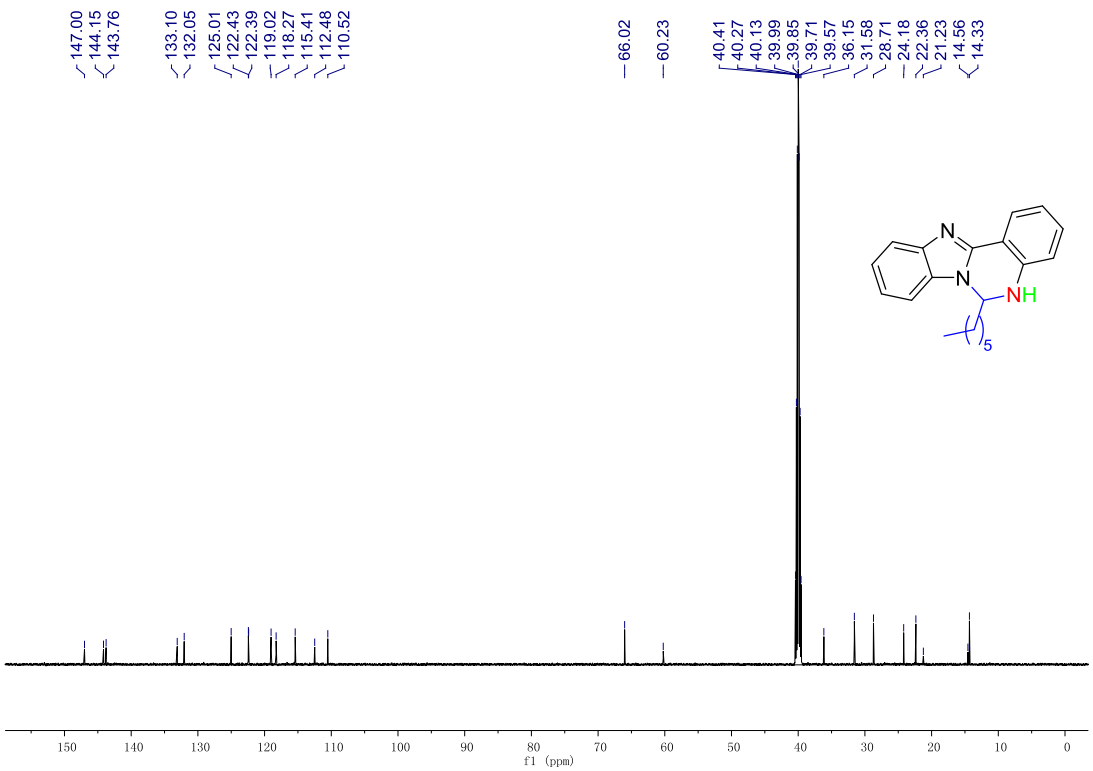
3t, ^{1}H NMR (600 MHz, DMSO-d_6)



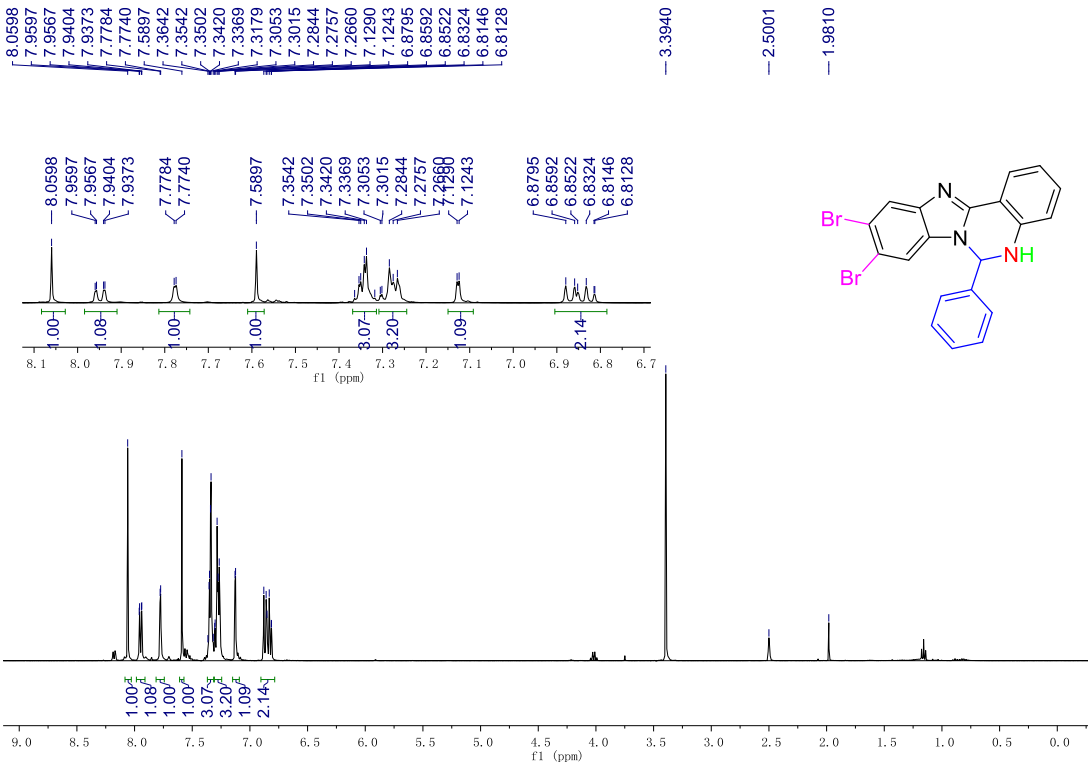
3t, ^{13}C NMR (100 MHz, DMSO-d_6)



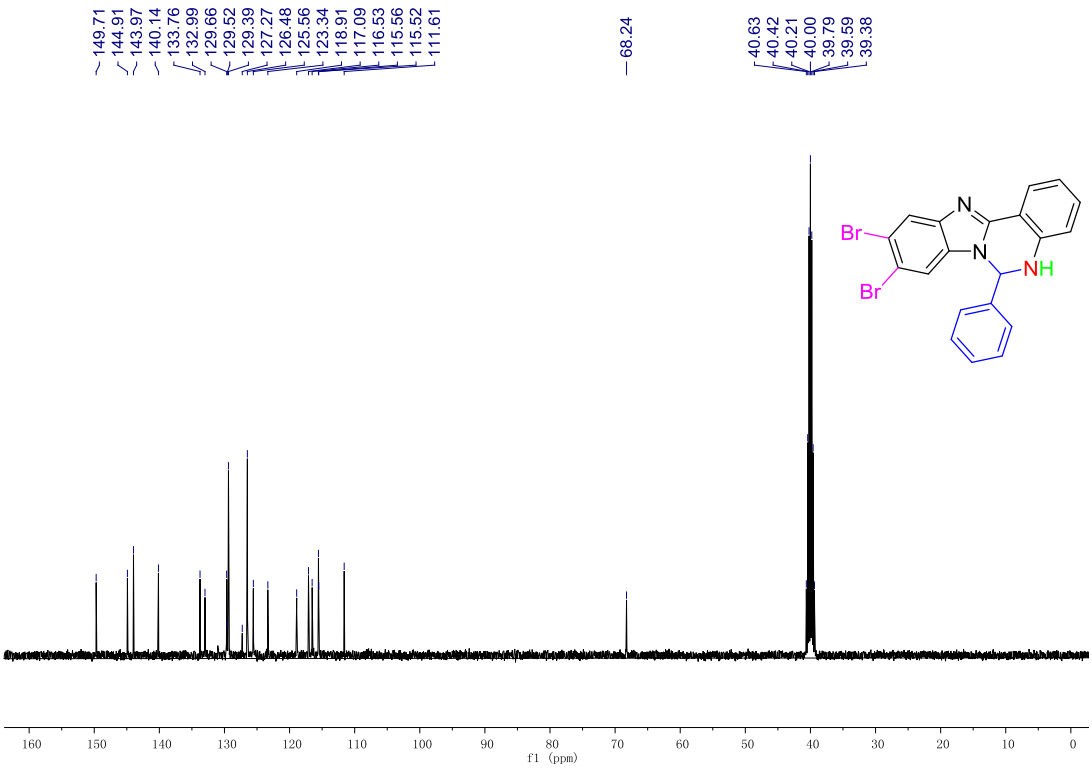
3u, ^{1}H NMR (600 MHz, DMSO-d_6)



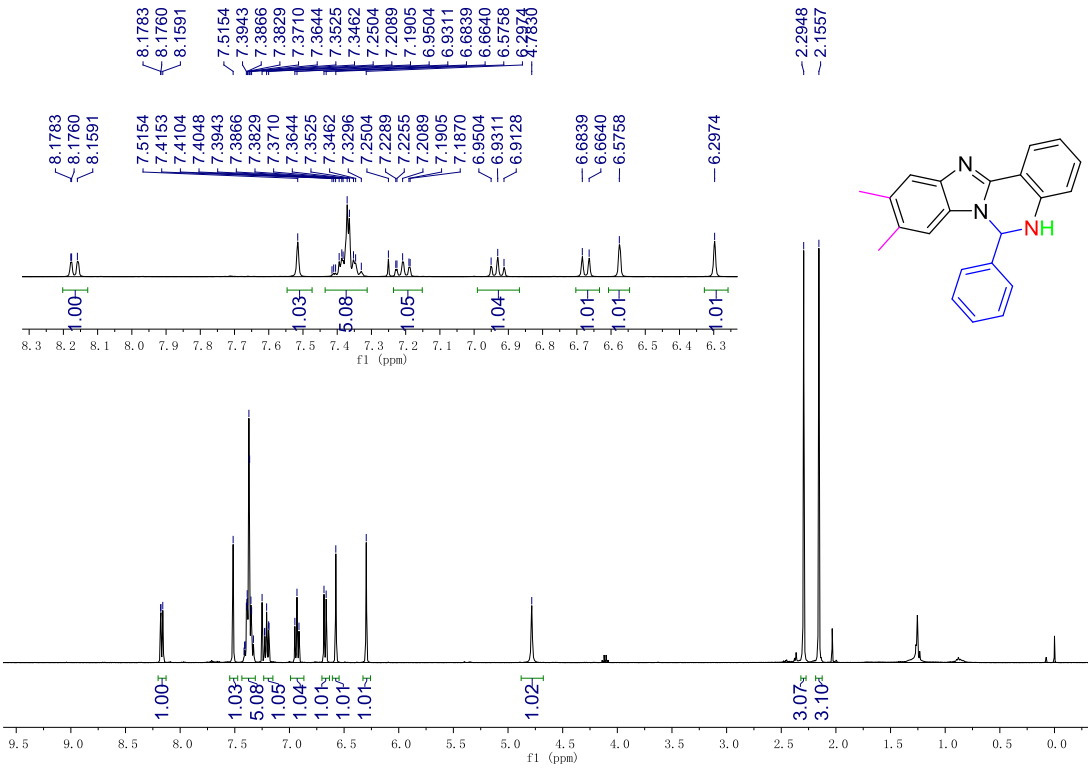
3u, ^{13}C NMR (150 MHz, DMSO-d_6)



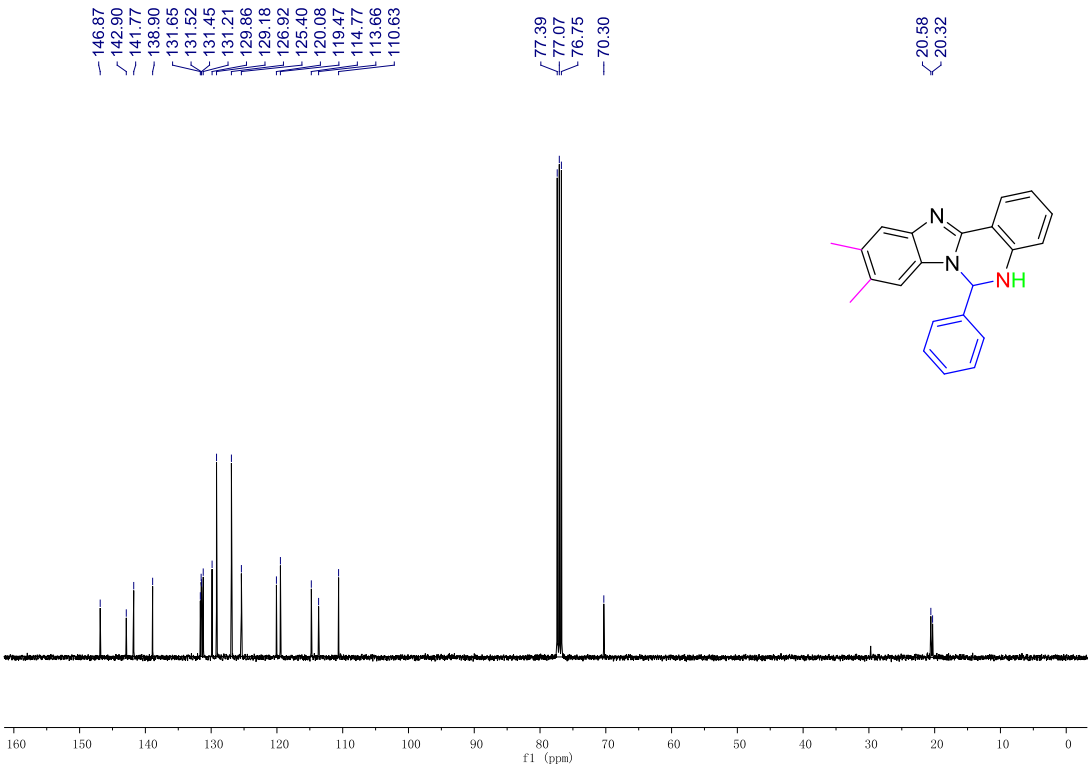
3v, ^{1}H NMR (400 MHz, DMSO-d_6)



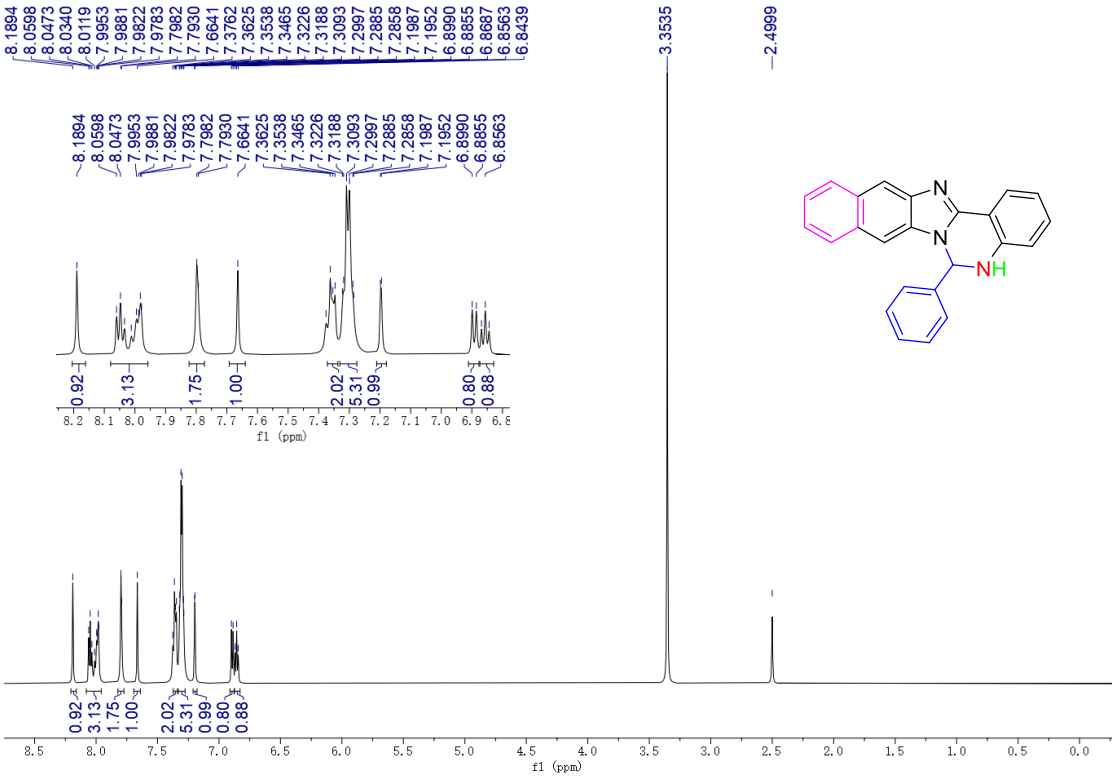
3v, ^{13}C NMR (100 MHz, DMSO-d_6)



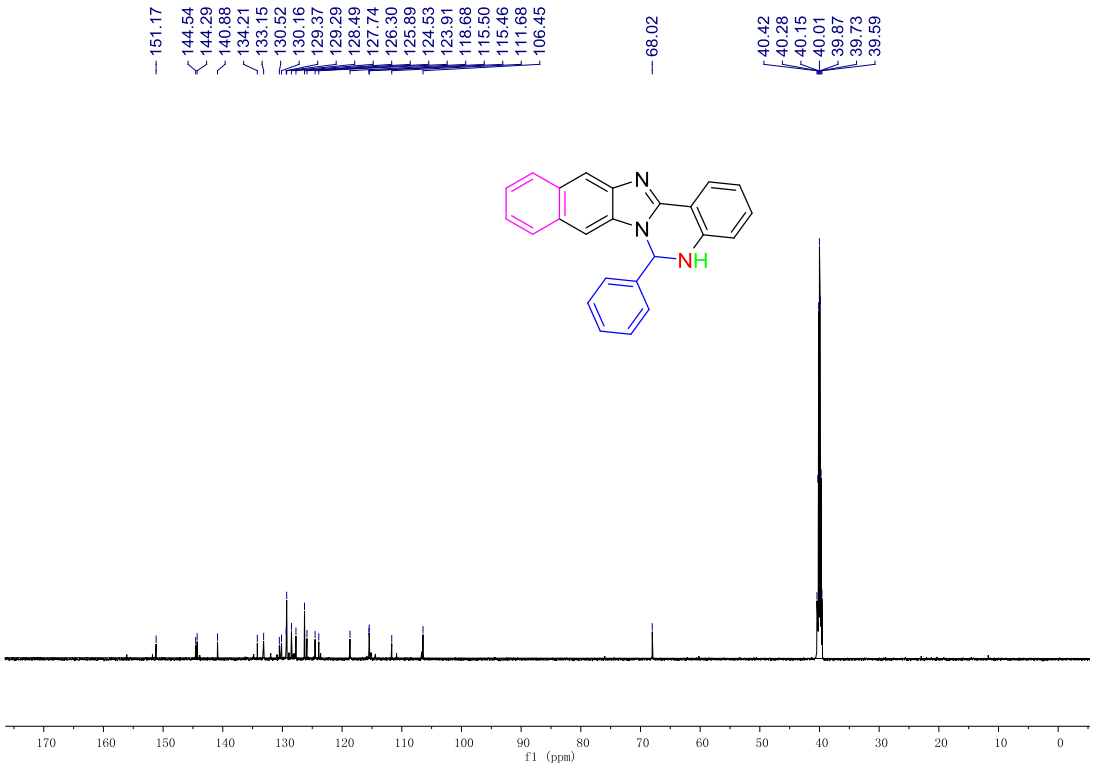
3w, ^{1}H NMR (400 MHz, $CDCl_3$)



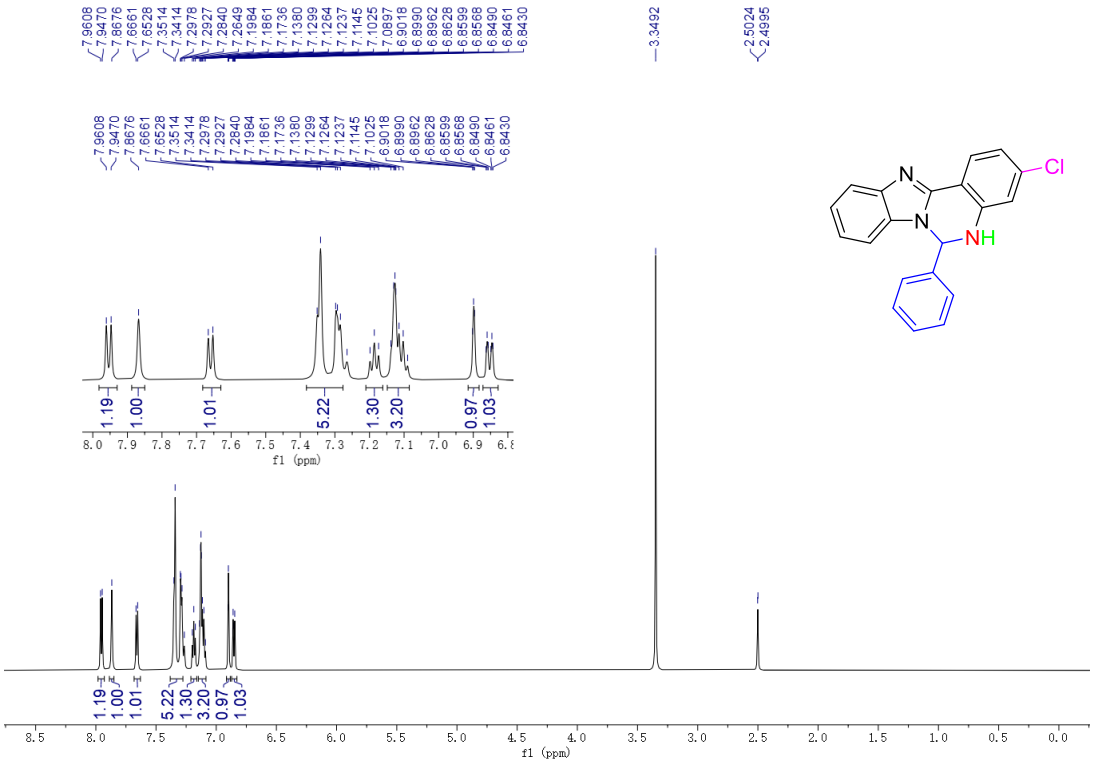
3w, ^{13}C NMR (100 MHz, $CDCl_3$)



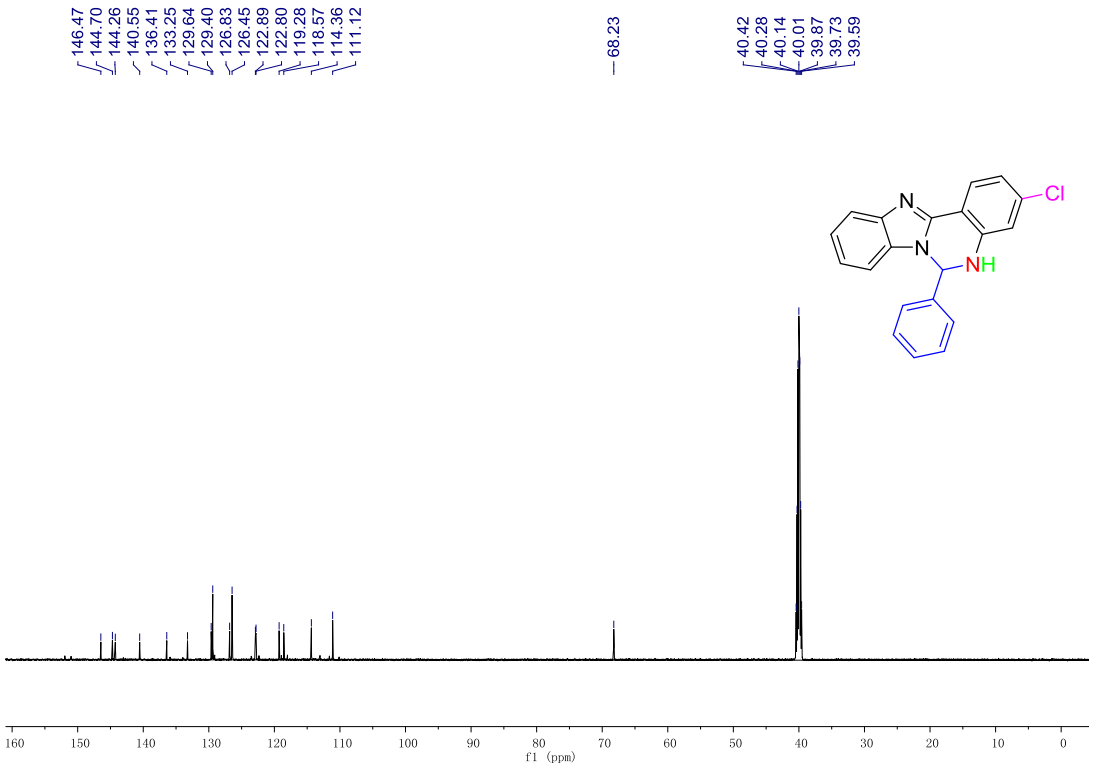
3x, ^{1}H NMR (600 MHz, DMSO-d_6)



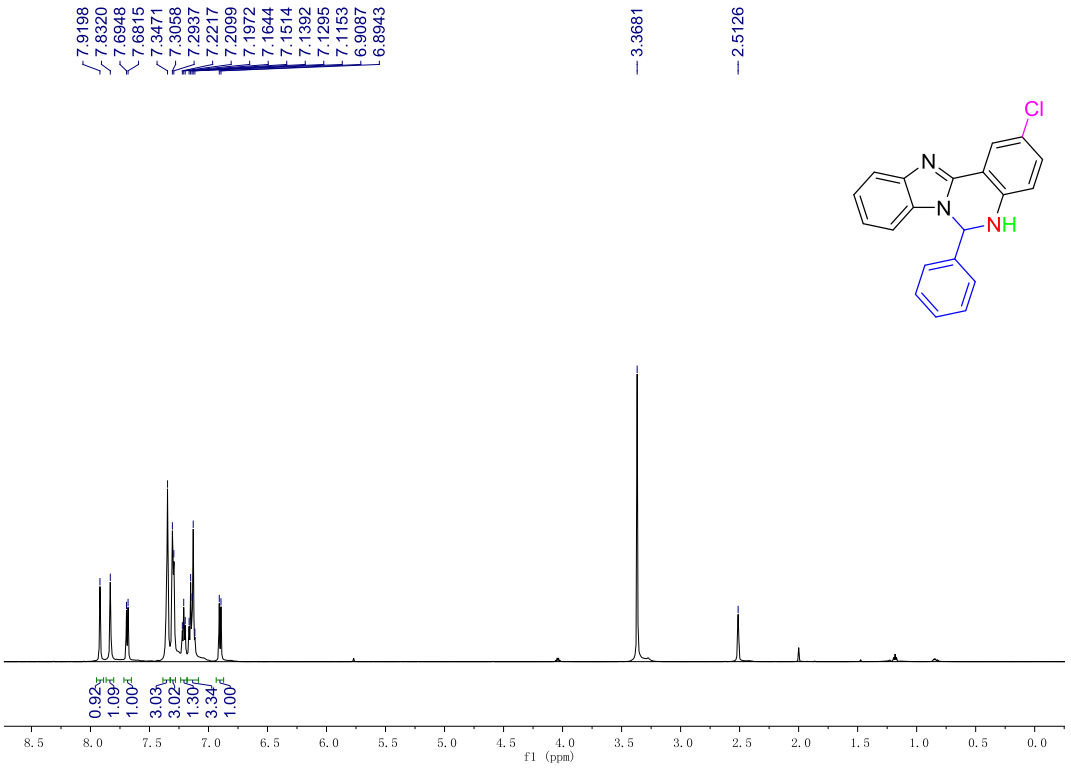
3x, ^{13}C NMR (150 MHz, DMSO-d_6)



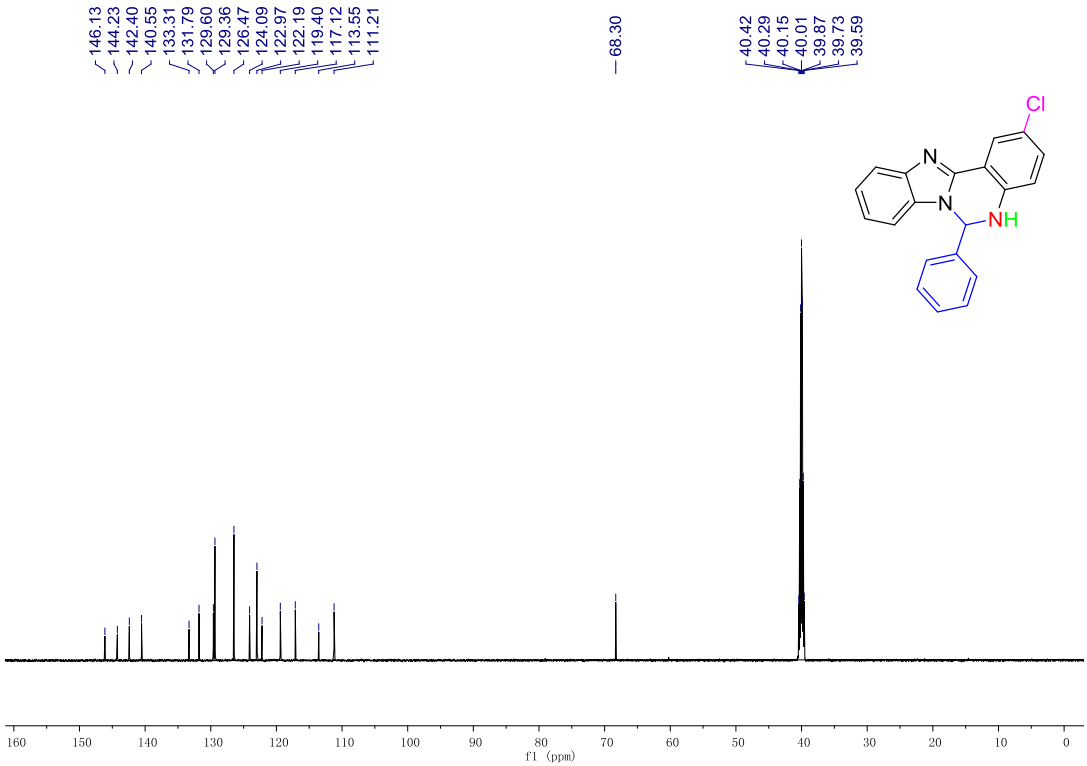
3y, ^{1}H NMR (600 MHz, DMSO-d_6)



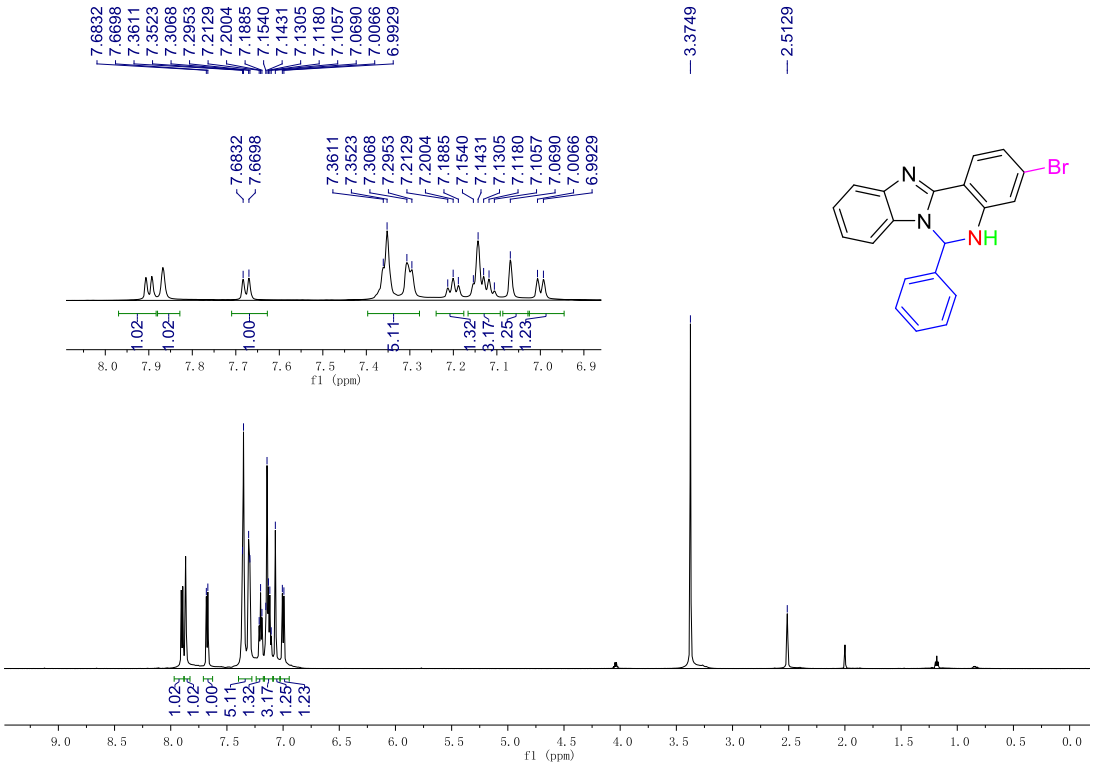
3y, ^{13}C NMR (150 MHz, DMSO-d_6)



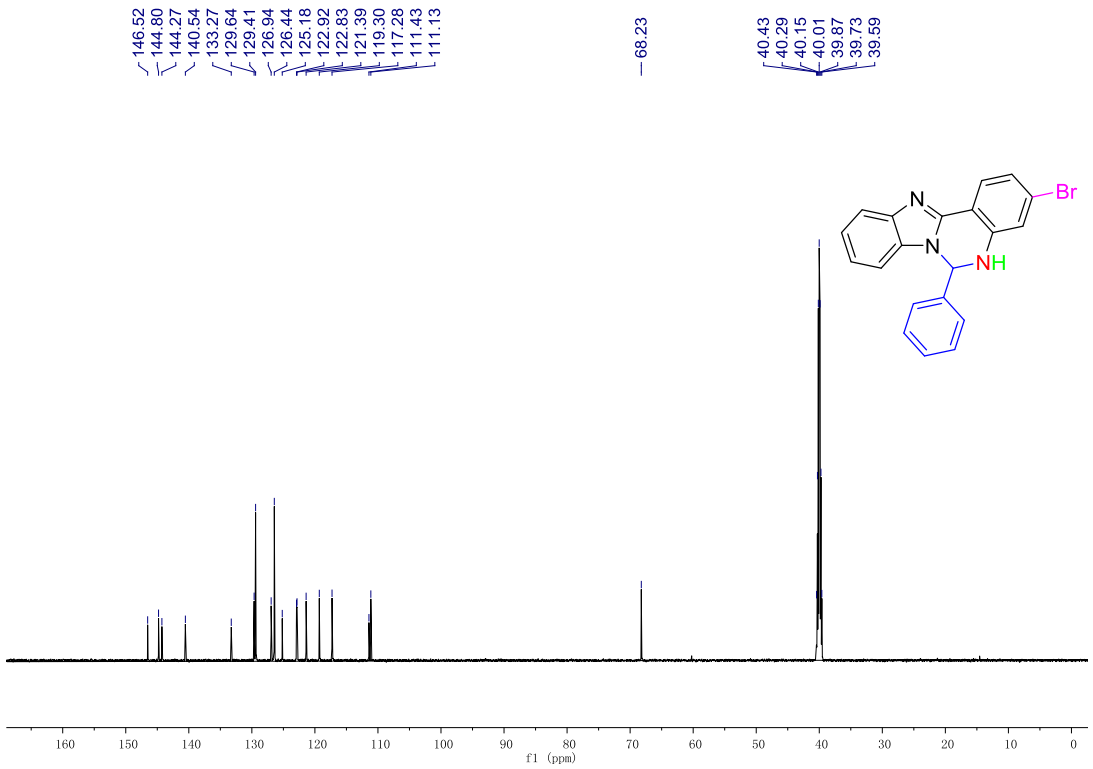
3z, ^{1}H NMR (600 MHz, DMSO-d_6)



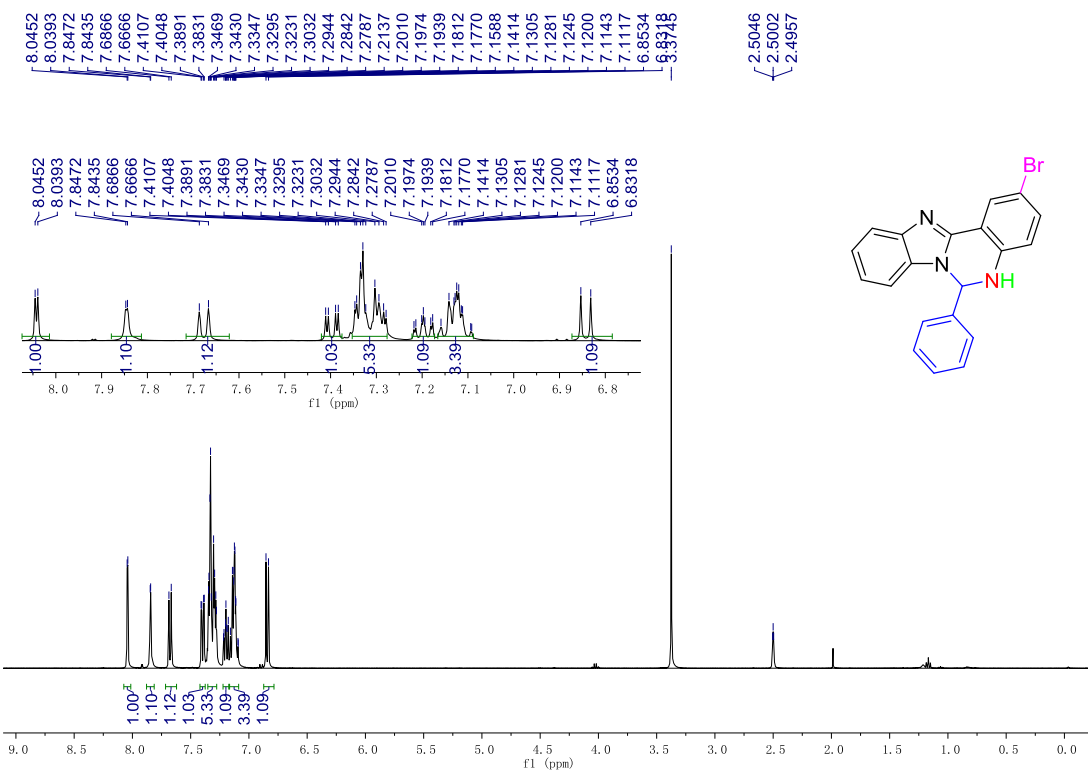
3z, ^{13}C NMR (150 MHz, DMSO-d_6)



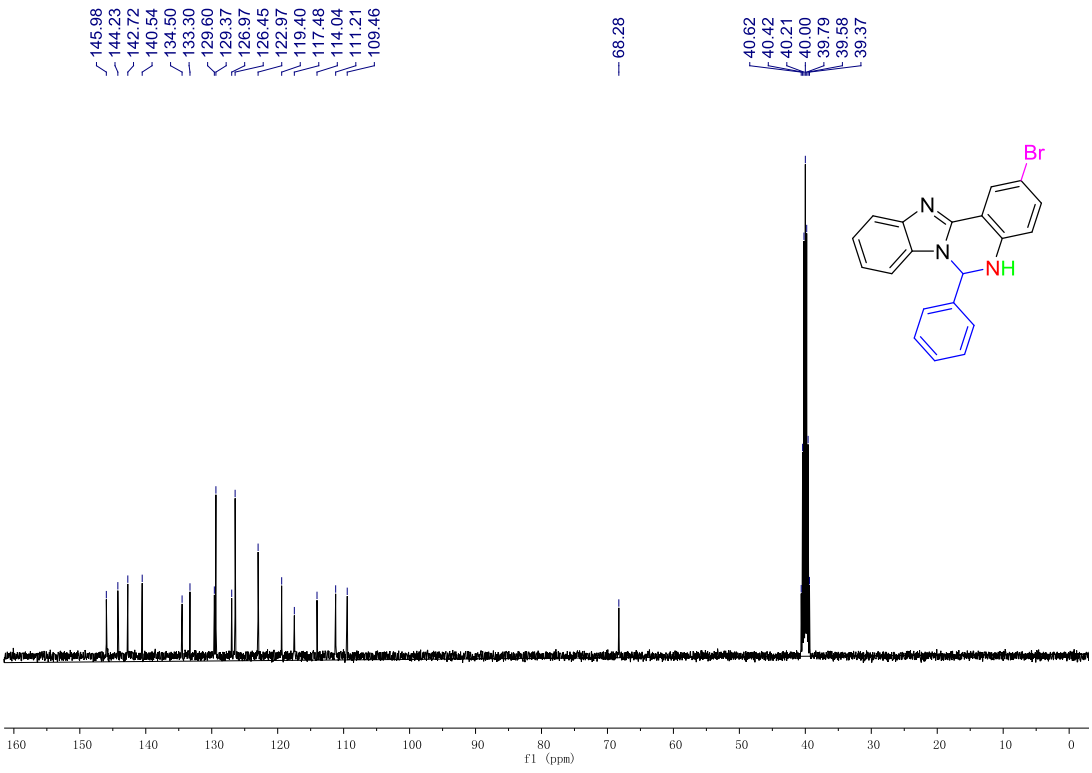
3aa, ^{1}H NMR (600 MHz, DMSO-d_6)



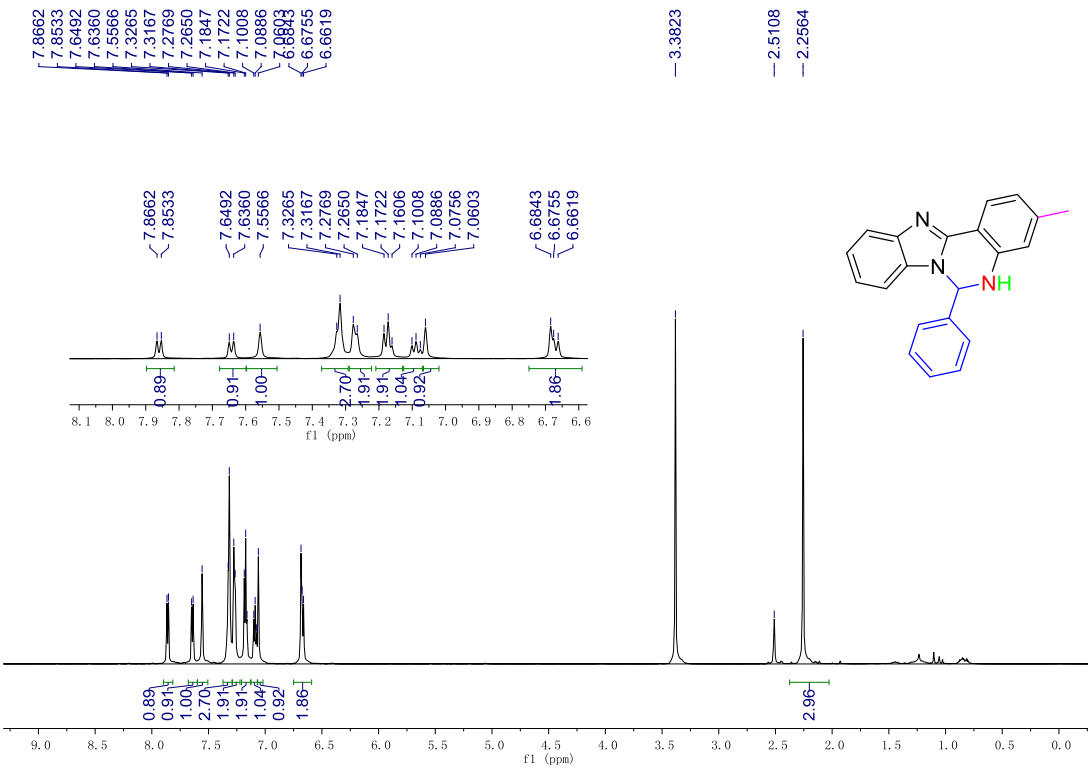
3aa, ^{13}C NMR (150 MHz, DMSO-d_6)



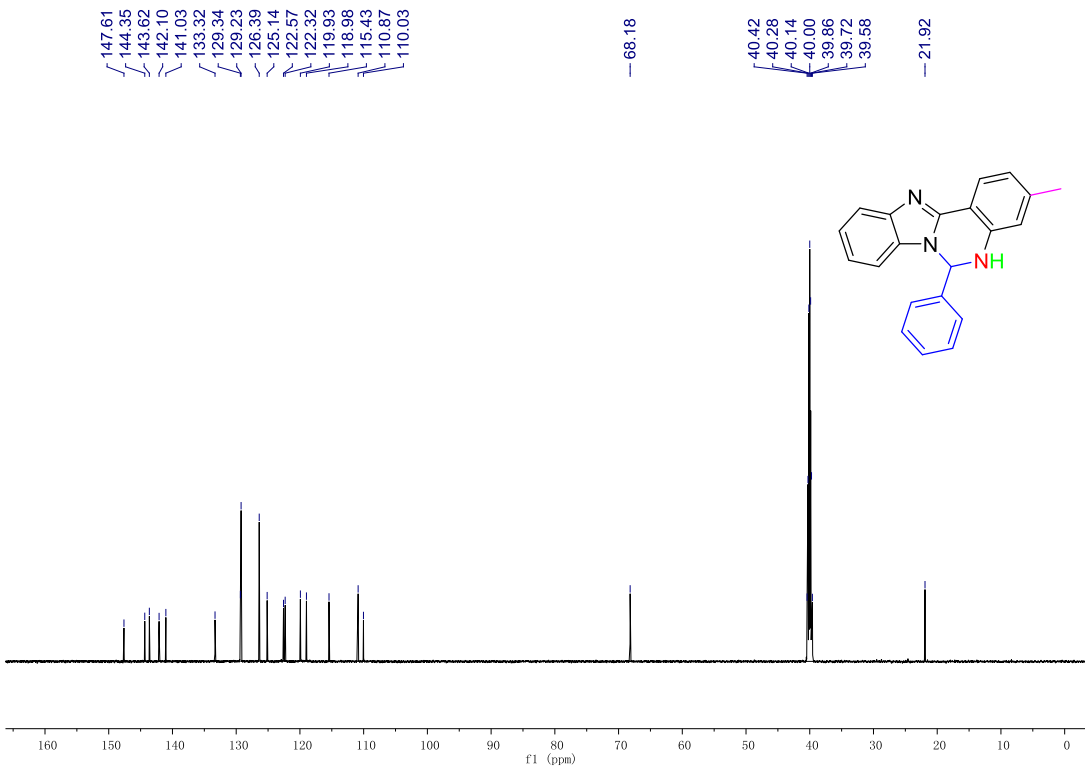
3ab, ^{1}H NMR (400 MHz, DMSO-d_6)



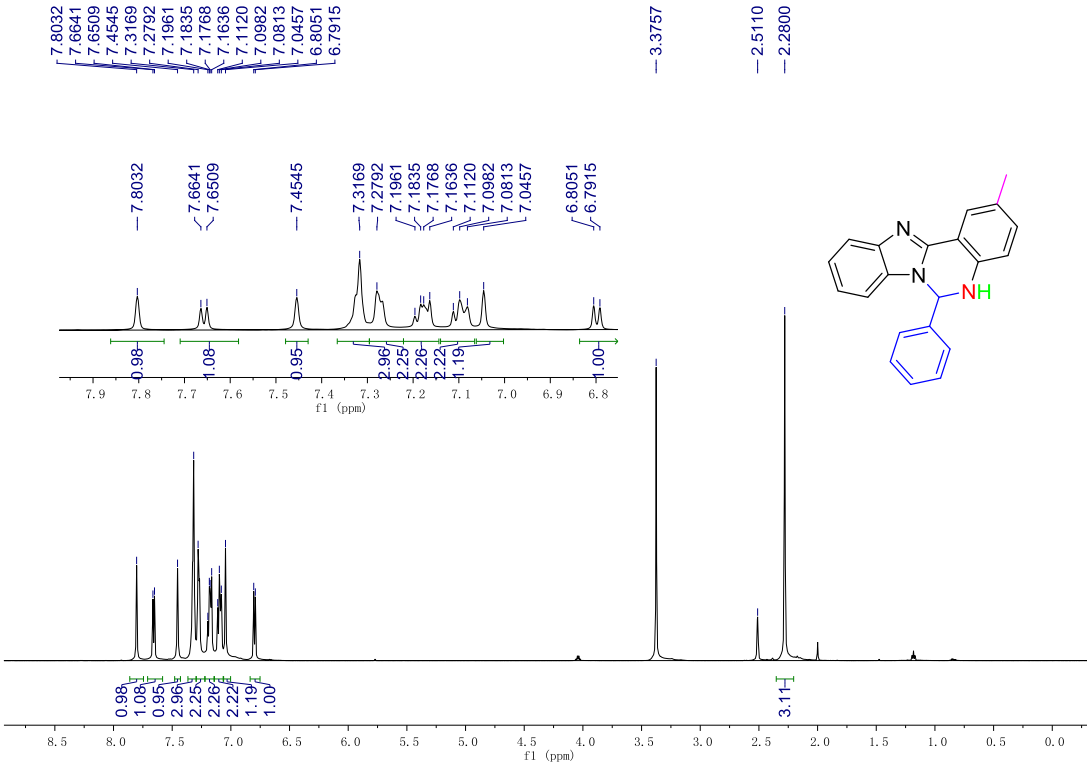
3ab, ^{13}C NMR (100 MHz, DMSO-d_6)



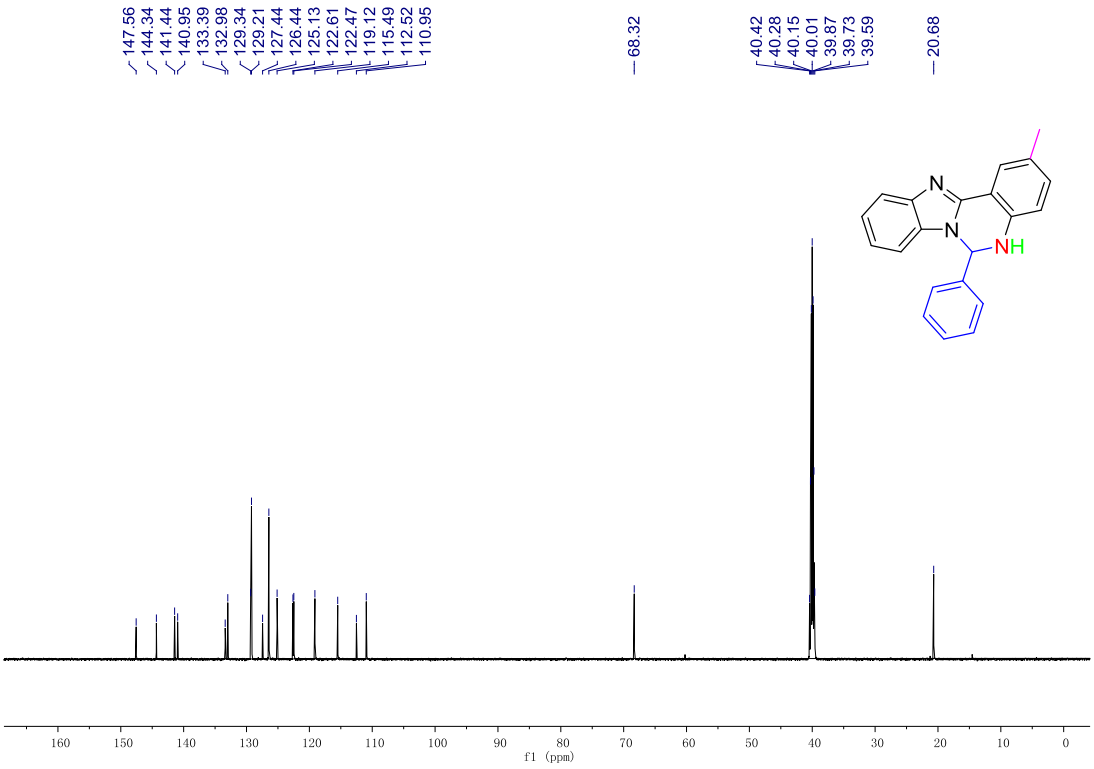
3ac, ^{1}H NMR (600 MHz, DMSO-d_6)



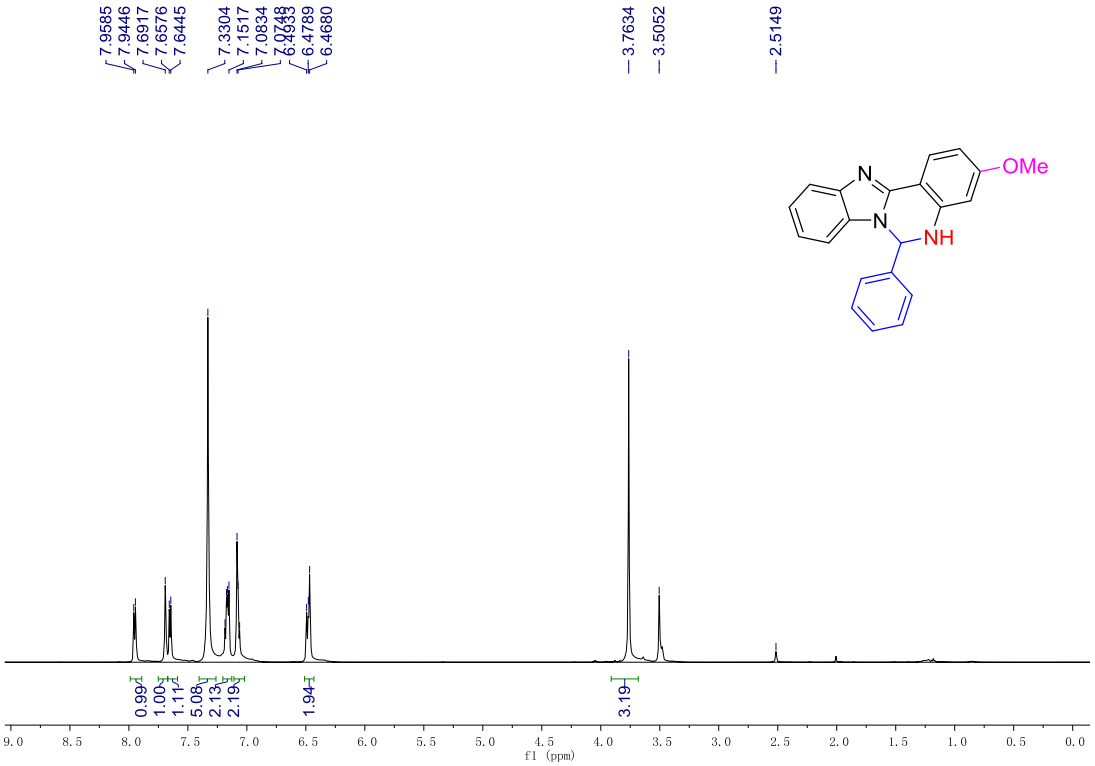
3ac, ^{13}C NMR (150 MHz, DMSO-d_6)



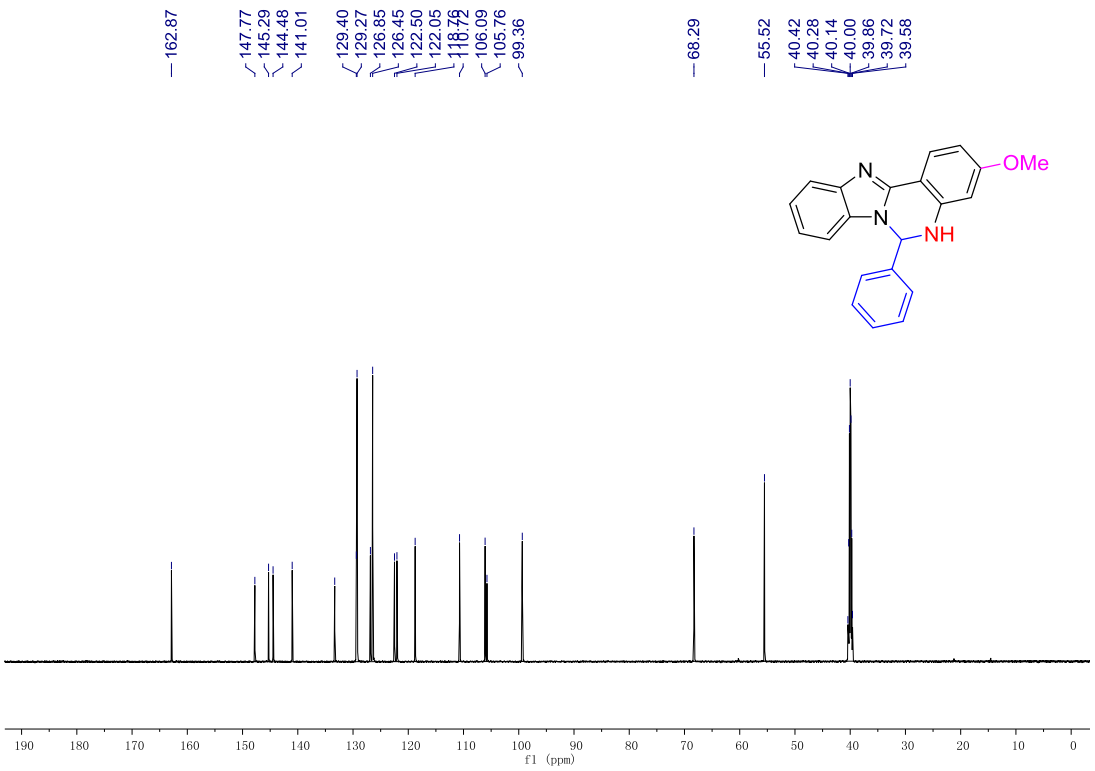
3ad, ^{1}H NMR (600 MHz, DMSO-d_6)



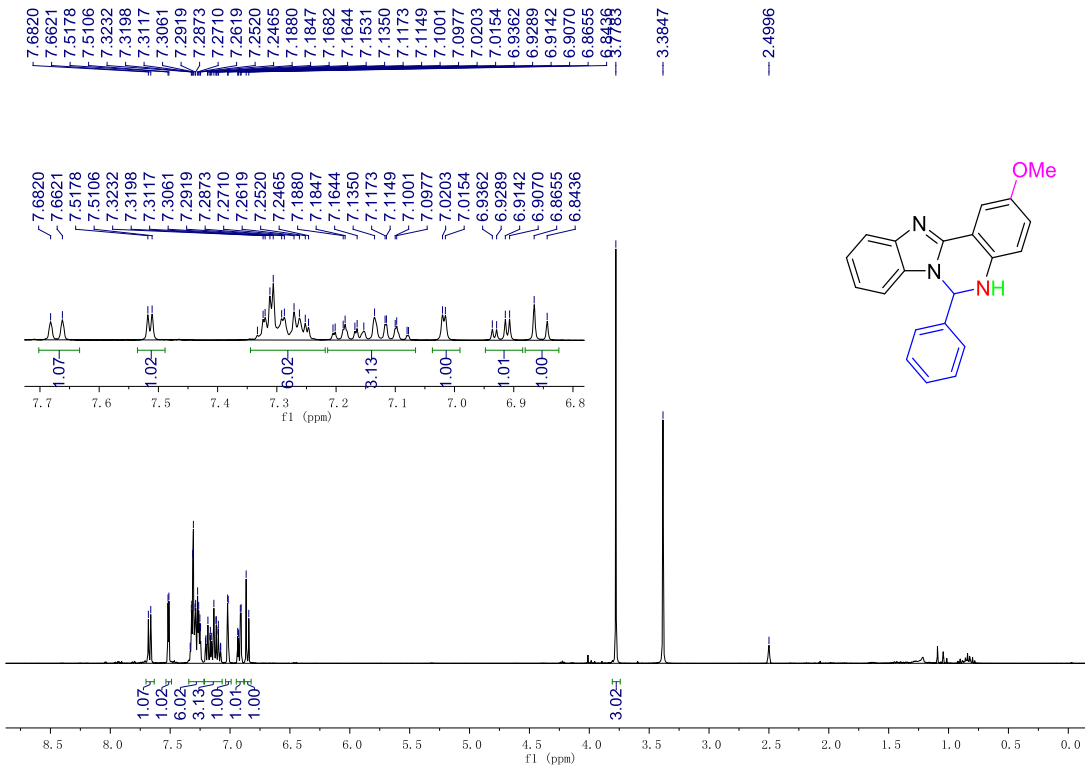
3ad, ^{13}C NMR (150 MHz, DMSO-d_6)



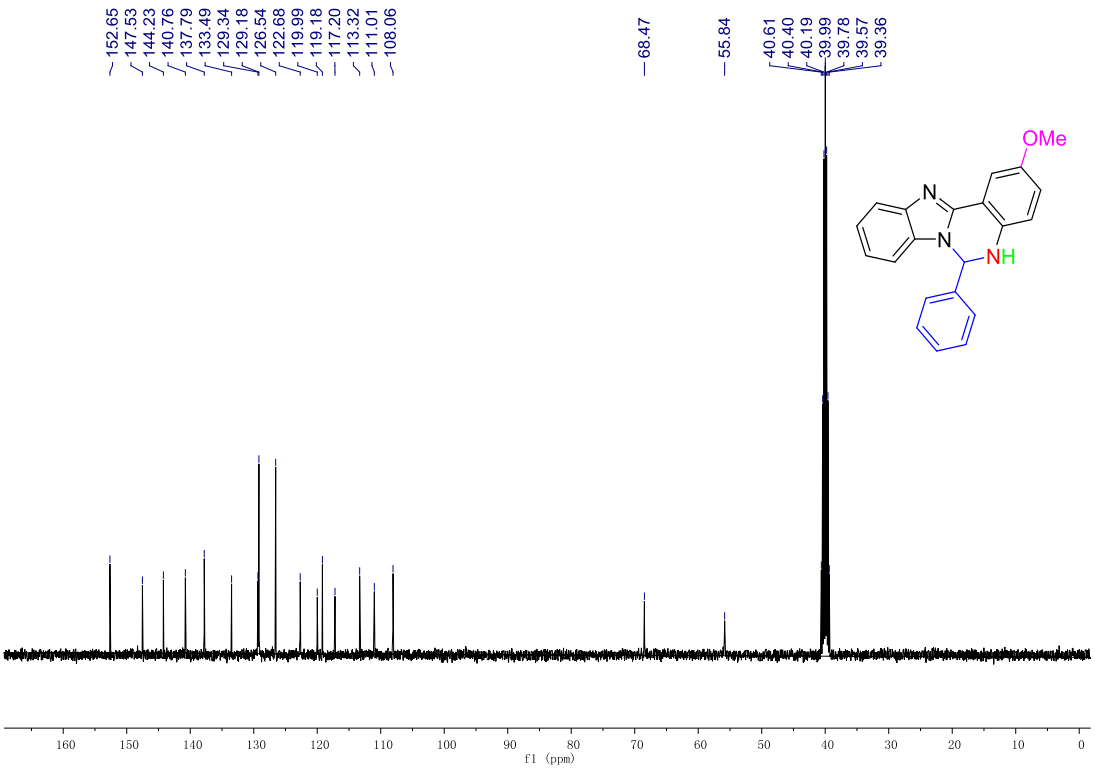
3ae, ^{1}H NMR (600 MHz, DMSO-d_6)



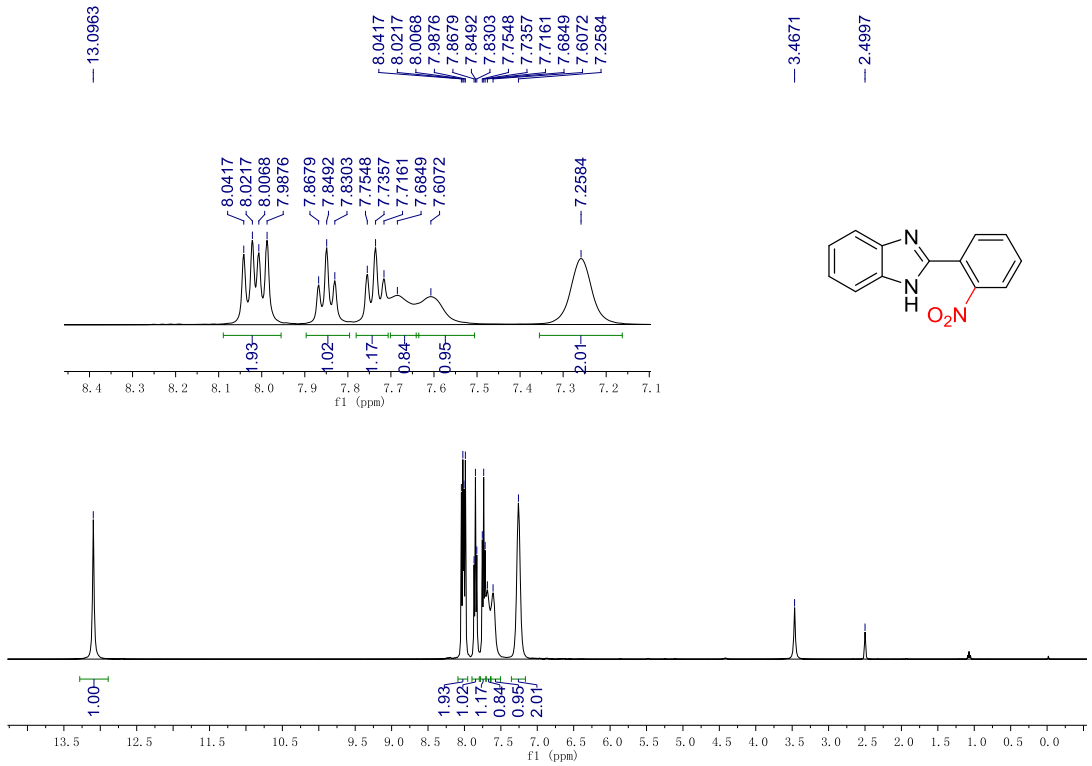
3ae, ^{13}C NMR (150 MHz, DMSO-d_6)



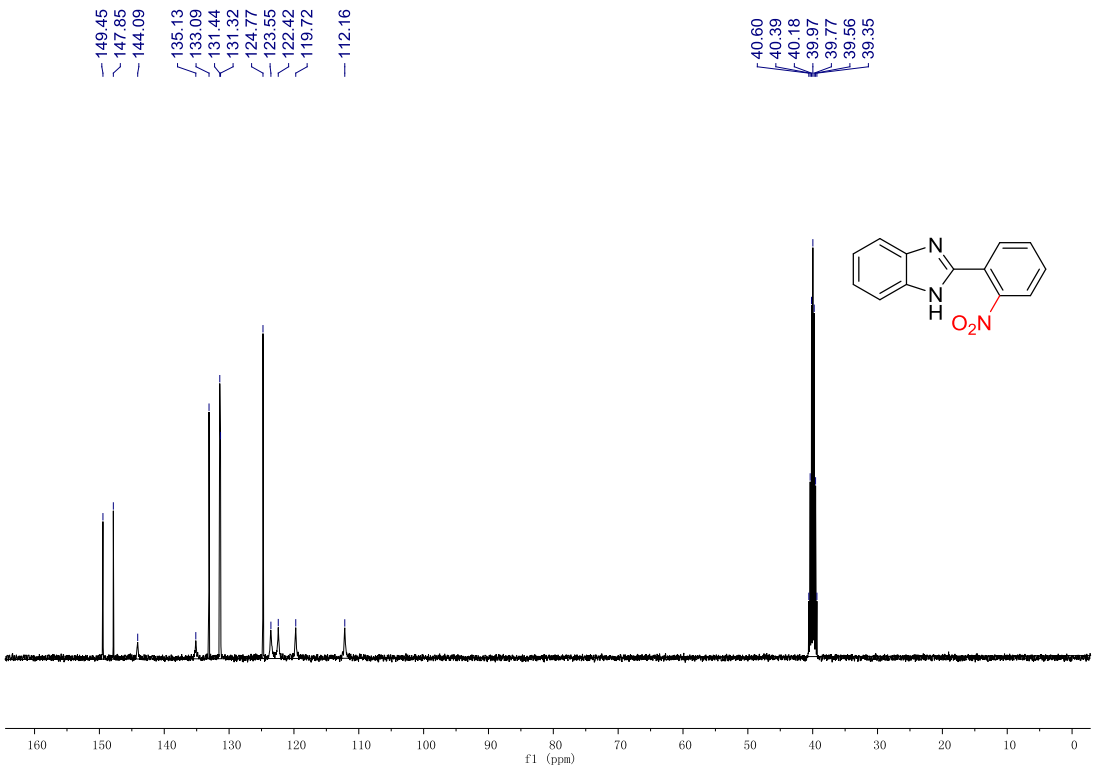
3af, ^{1}H NMR (400 MHz, DMSO-d_6)



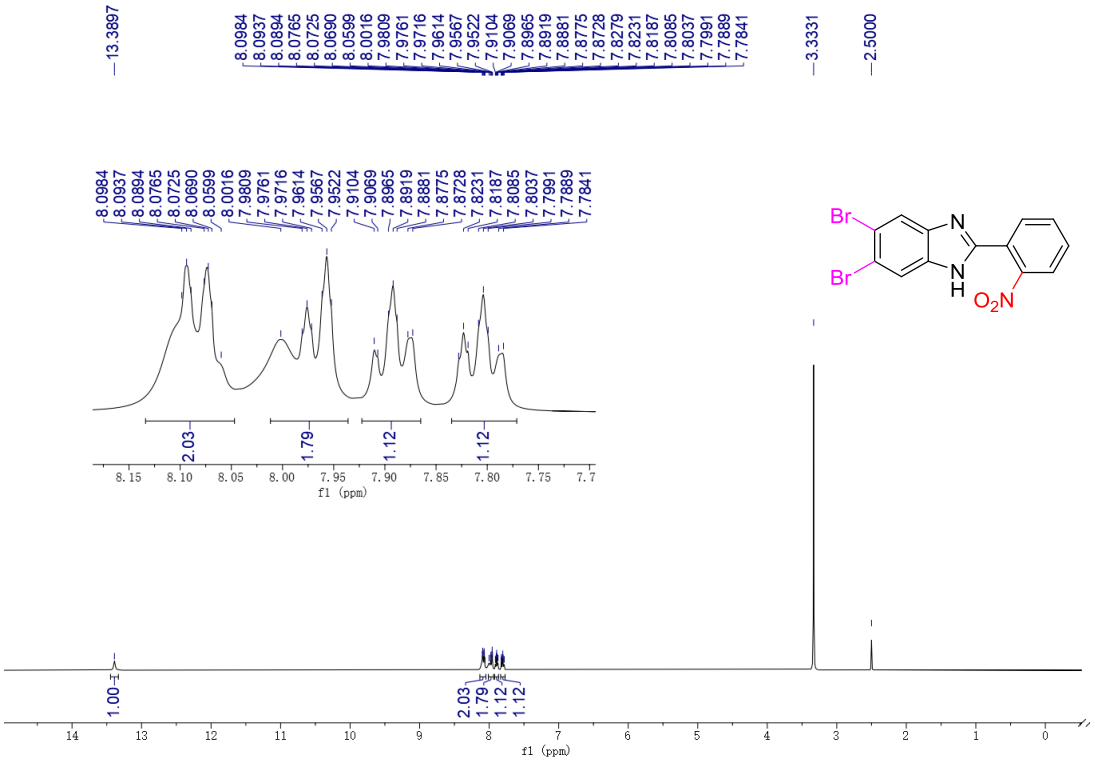
3af, ^{13}C NMR (100 MHz, DMSO-d_6)



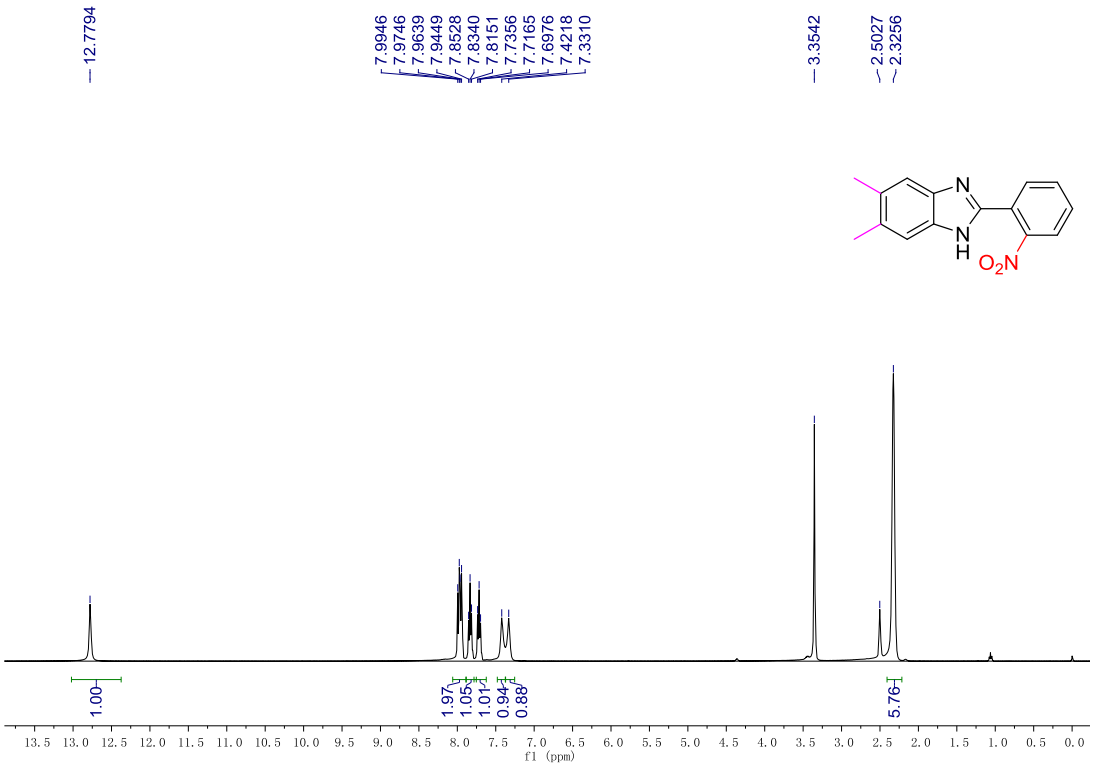
1a, ^{1}H NMR (400 MHz, DMSO-d_6)



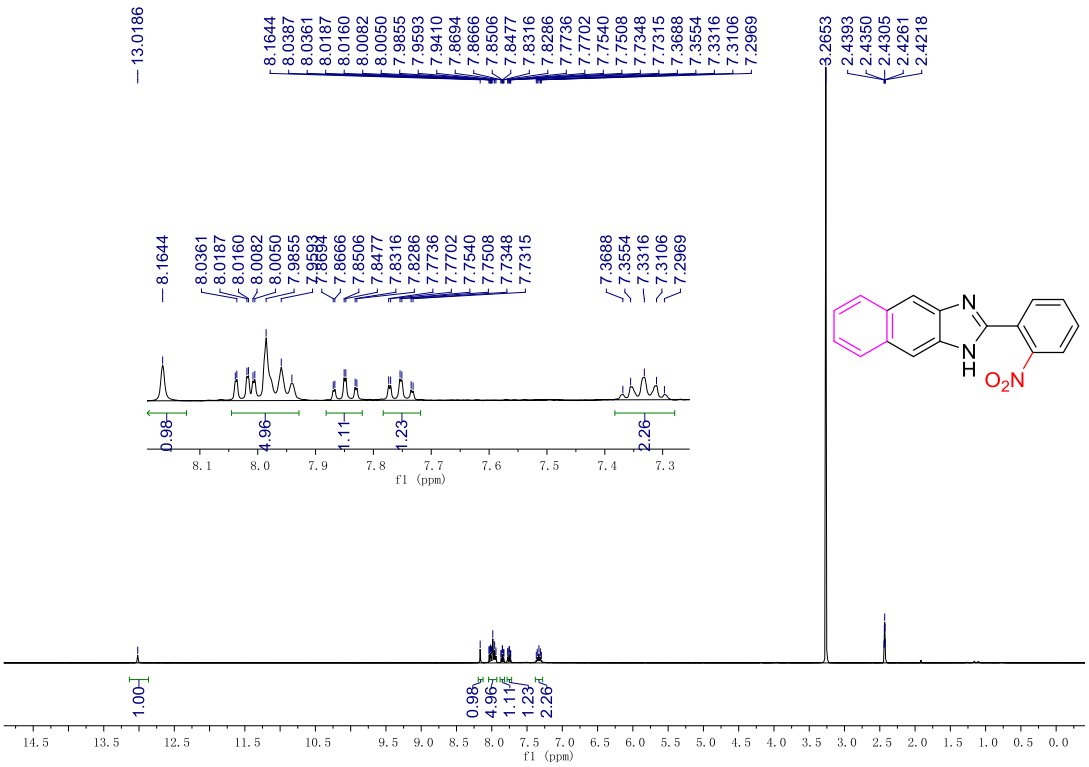
1a, ^{13}C NMR (400 MHz, DMSO-d_6)



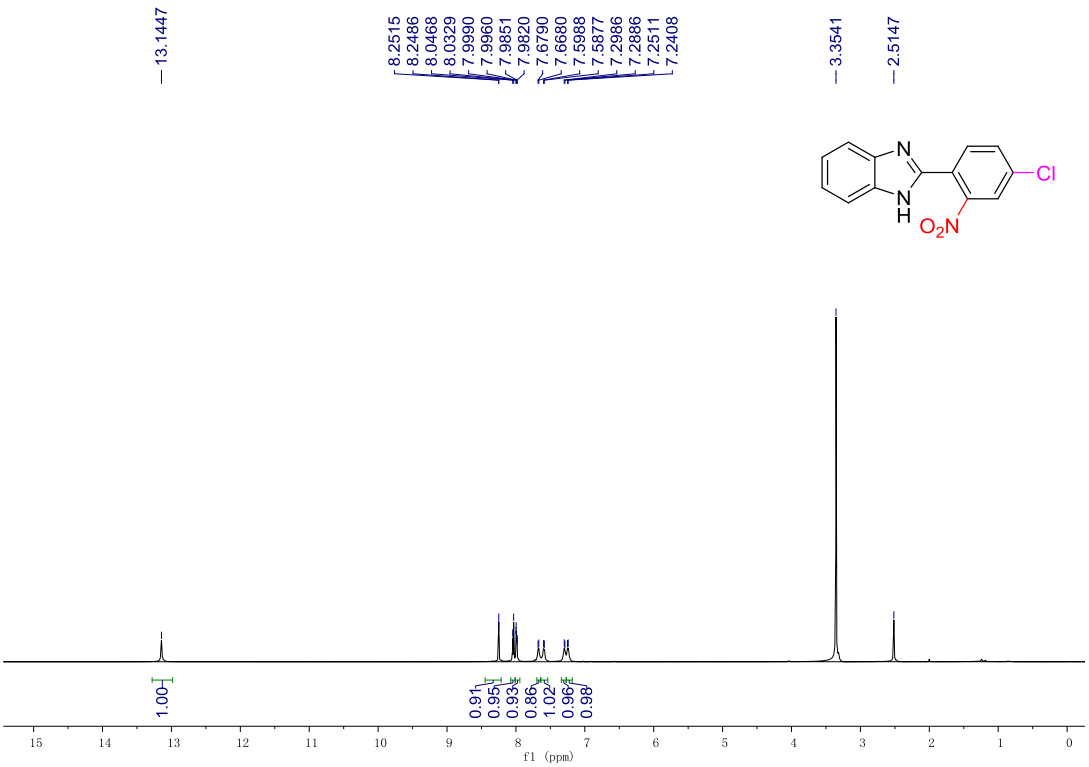
1v, ^{1}H NMR (400 MHz, DMSO-d_6)



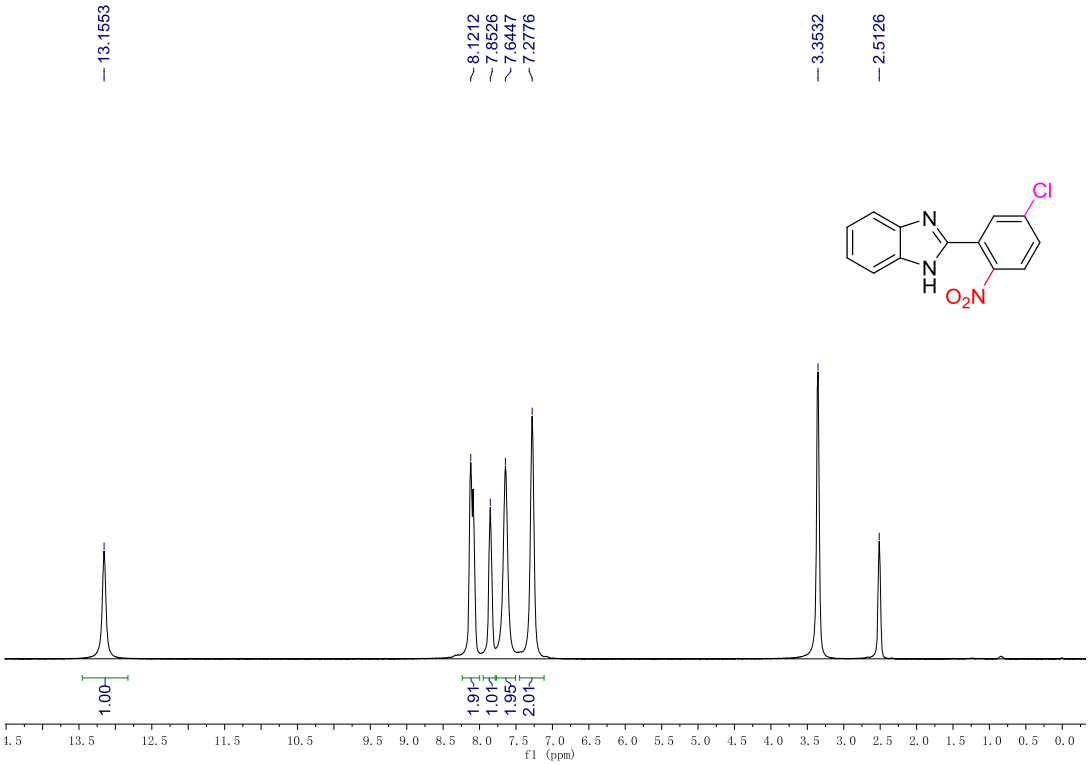
1w, ^{1}H NMR (400 MHz, DMSO-d_6)



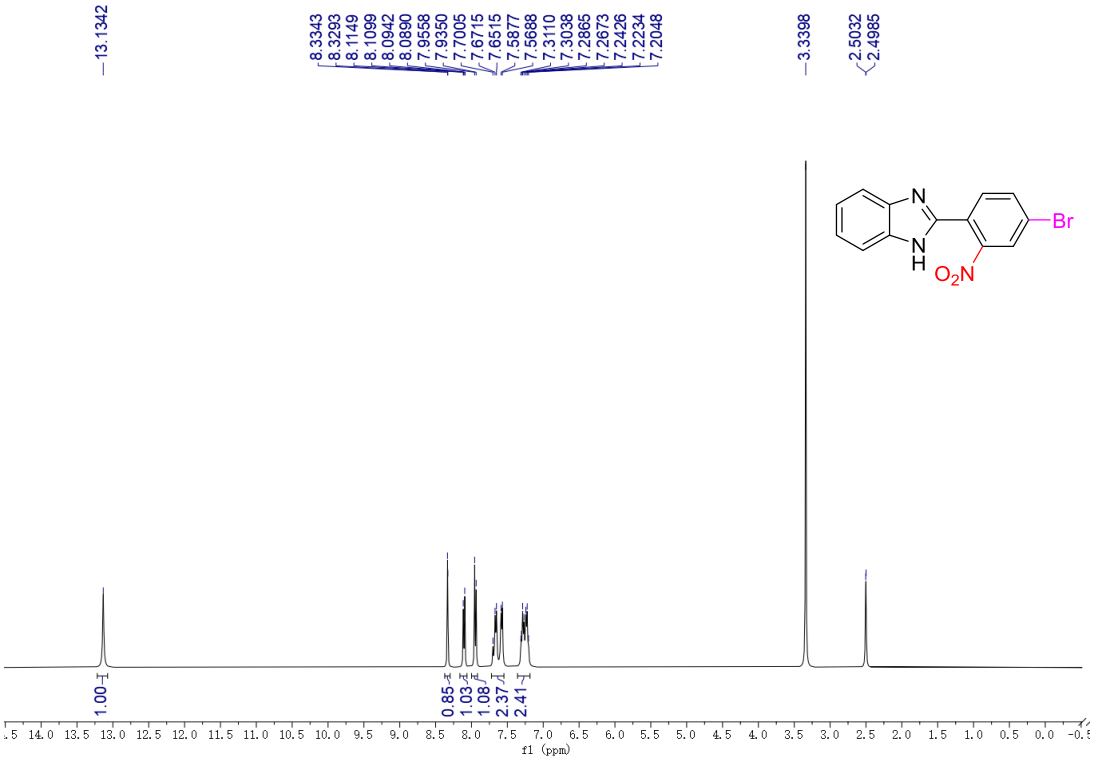
1x, ^{1}H NMR (400 MHz, DMSO-d_6)



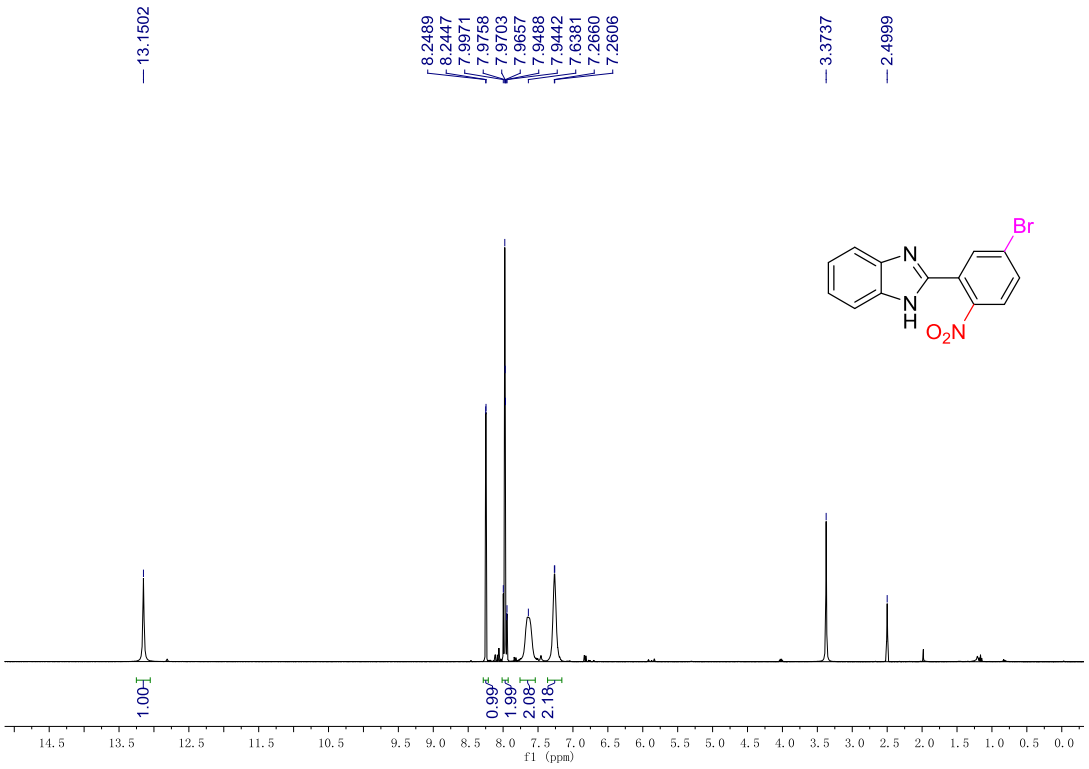
1y, ^{1}H NMR (600 MHz, DMSO-d_6)



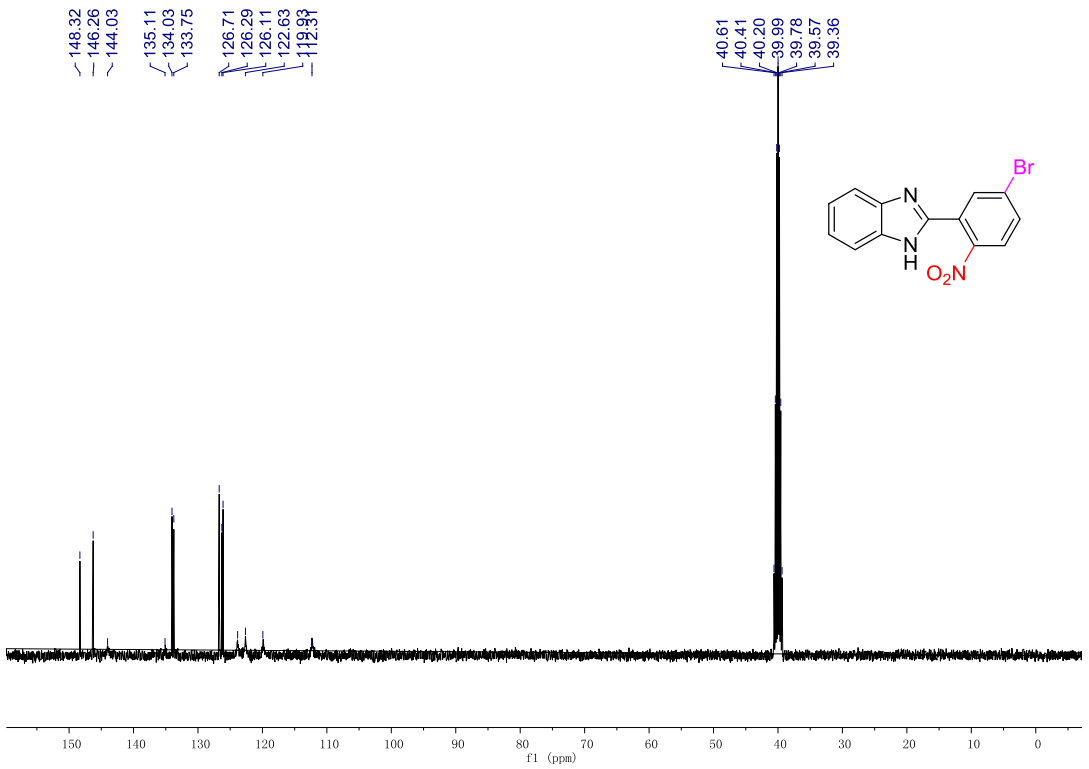
1z, ^{1}H NMR (400 MHz, DMSO-d_6)



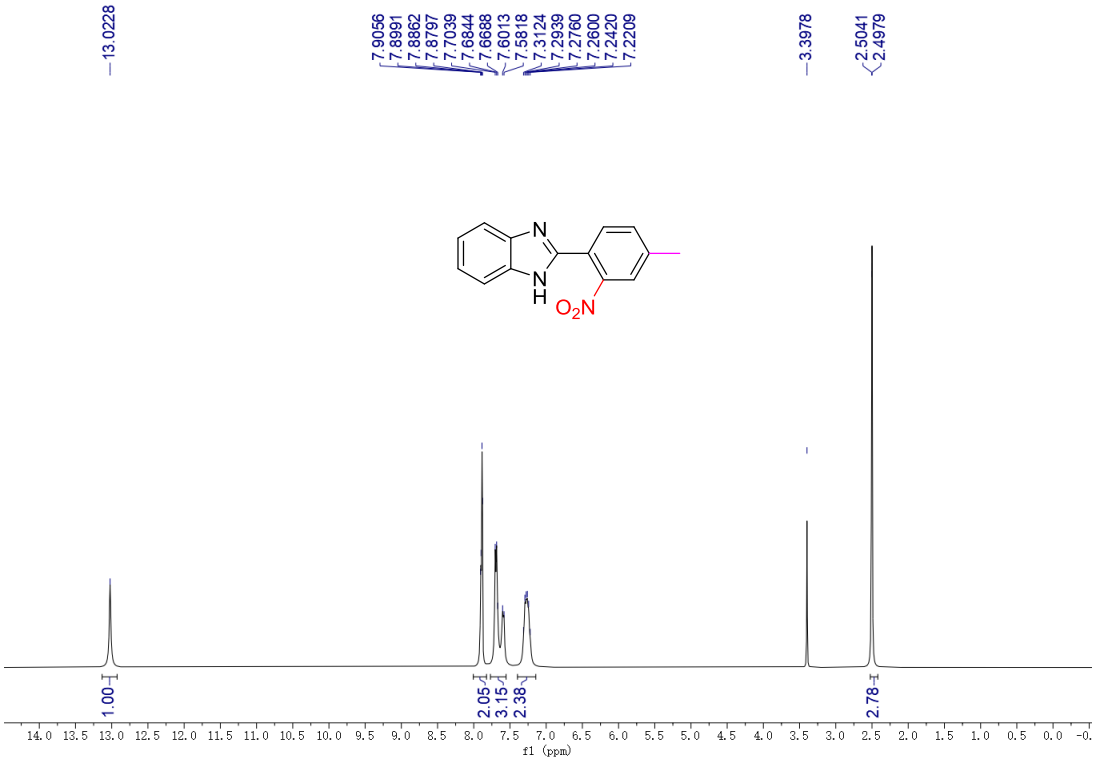
1aa, ^{1}H NMR (400 MHz, DMSO-d_6)



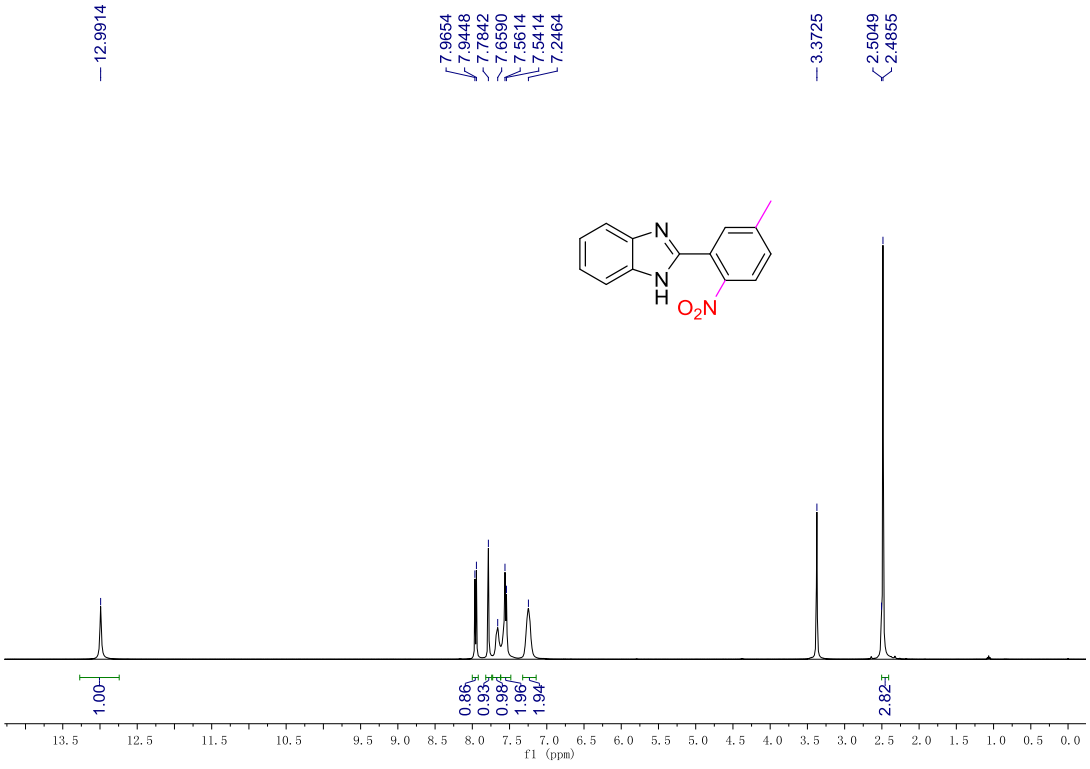
1ab, ^{1}H NMR (400 MHz, DMSO-d_6)



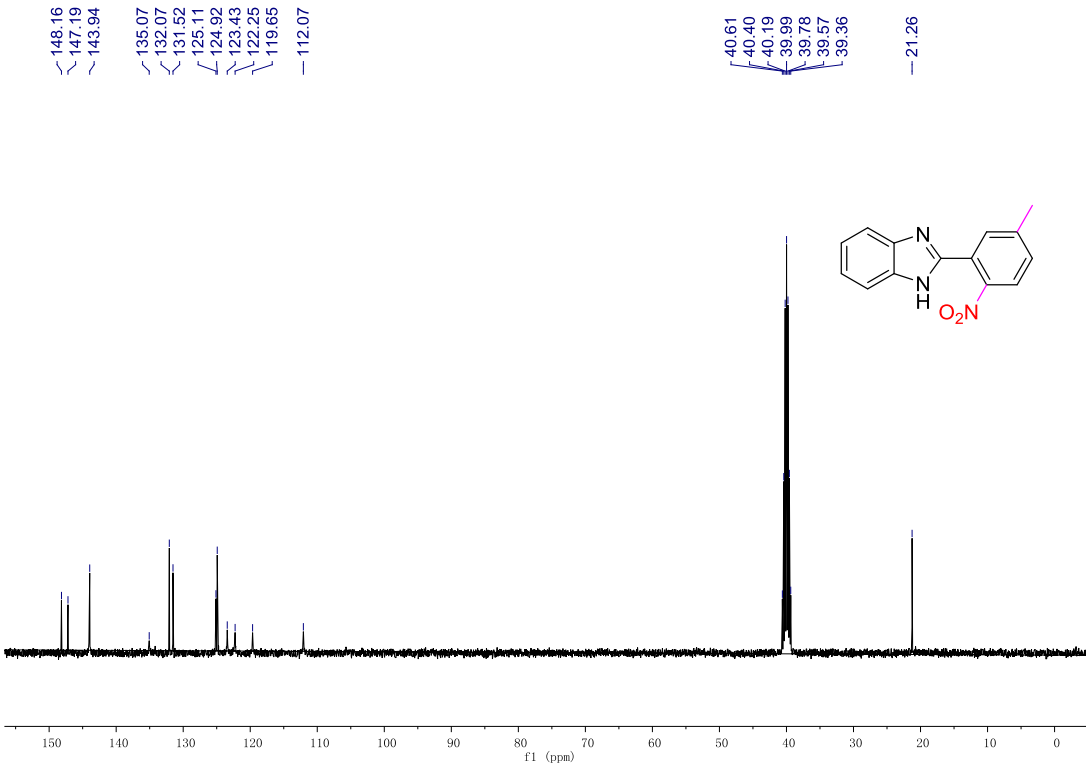
1ab, ^{13}C NMR (100 MHz, DMSO-d_6)



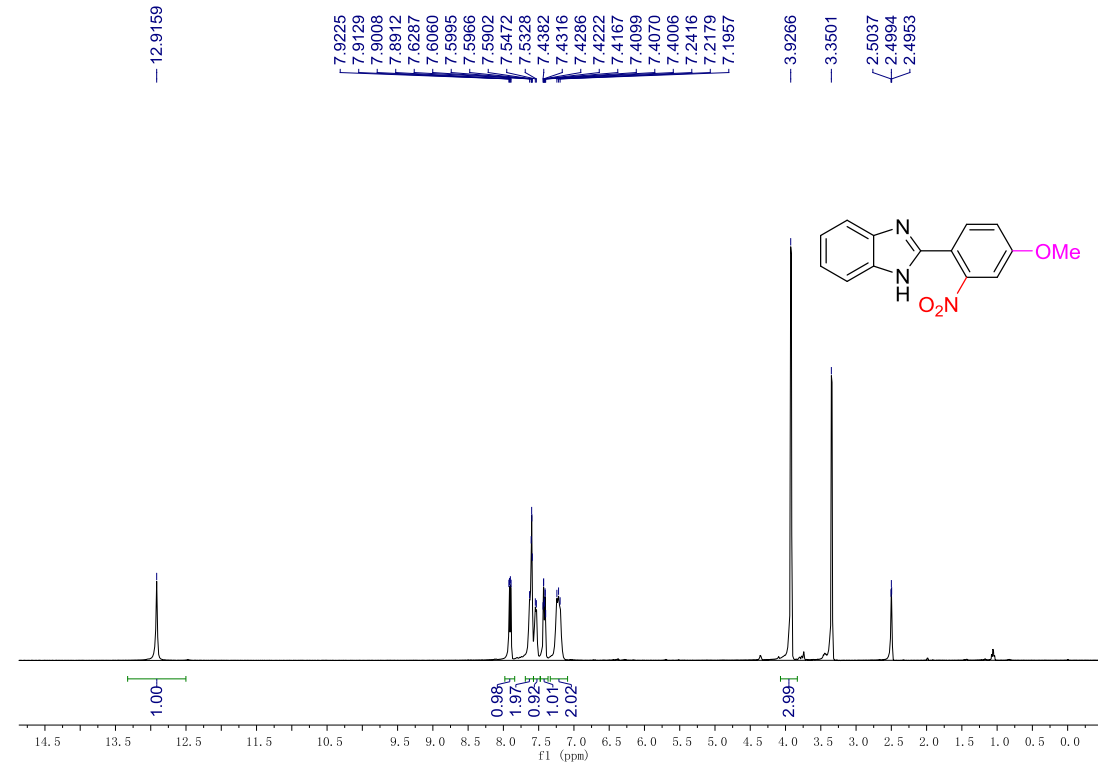
1ac, ^{1}H NMR (400 MHz, DMSO-d_6)



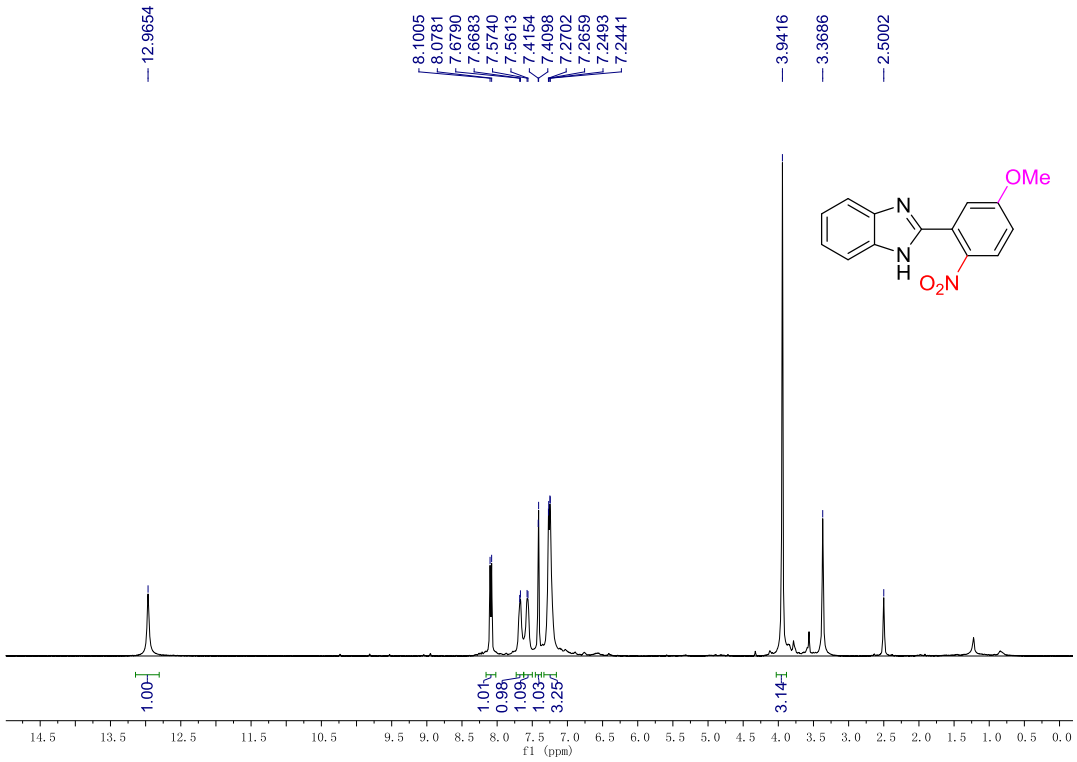
1ad, ^{1}H NMR (400 MHz, DMSO-d_6)



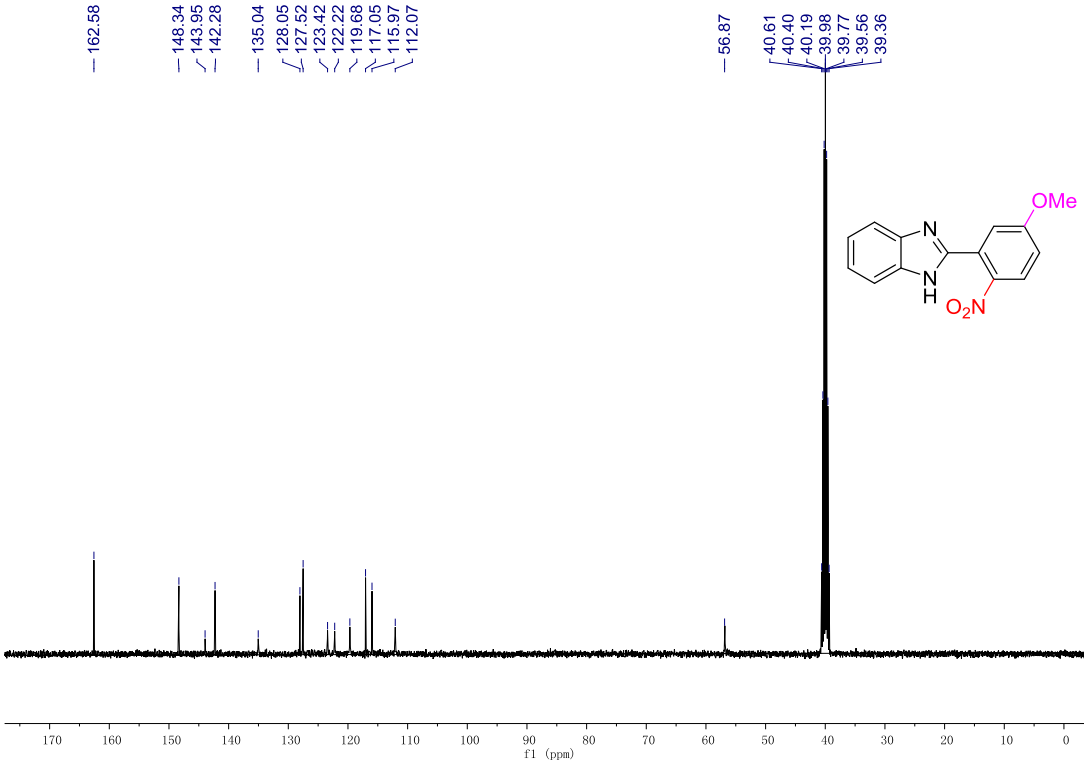
1ad, ^{13}C NMR (400 MHz, DMSO-d_6)



1ae, ^{1}H NMR (400 MHz, DMSO-d_6)



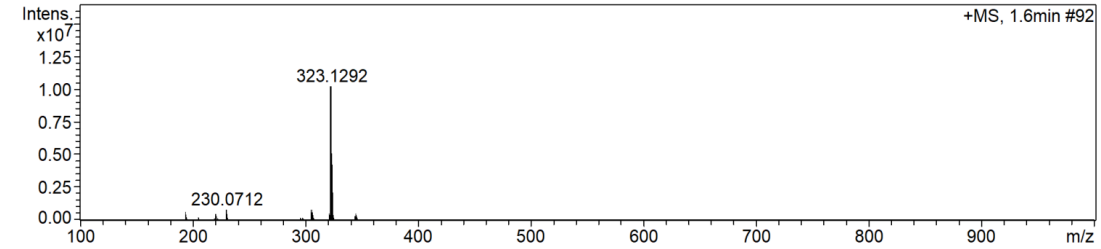
1af, ^{1}H NMR (400 MHz, DMSO-d_6)



1af, ^{13}C NMR (400 MHz, DMSO-d_6)

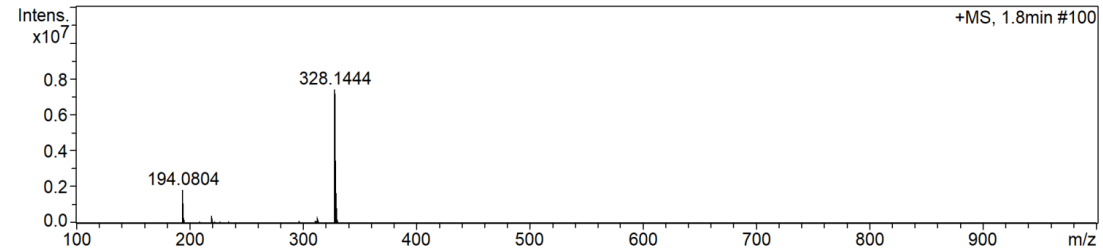
6. Copies of HRMS Spectra for the Products

+MS, 1.6min #92



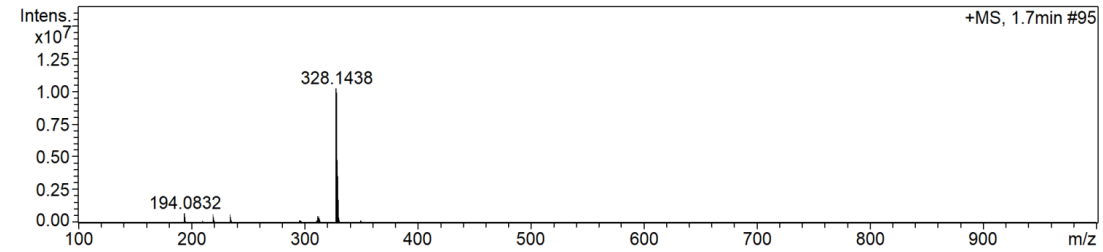
3f, HRMS calcd for $C_{21}H_{15}N_4^+$ $[M+H]^+$: 323.1291; found: 323.1292

+MS, 1.8min #100



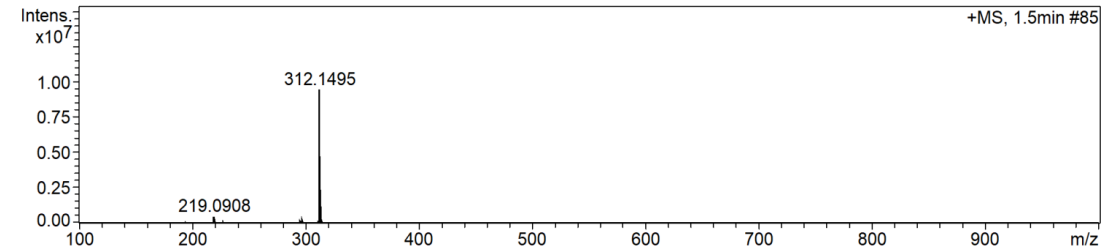
3h, HRMS calcd for $C_{21}H_{18}N_3O^+$ $[M+H]^+$: 328.1444; found: 328.1444

+MS, 1.7min #95

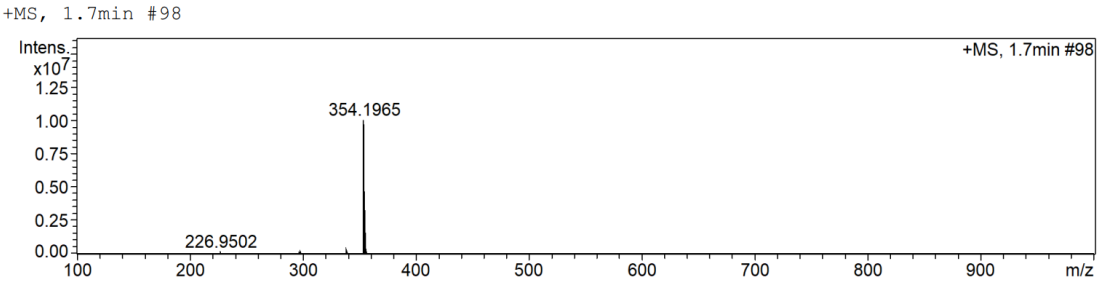


3i, HRMS calcd for $C_{21}H_{18}N_3O^+$ $[M+H]^+$: 328.1444; found: 328.1438

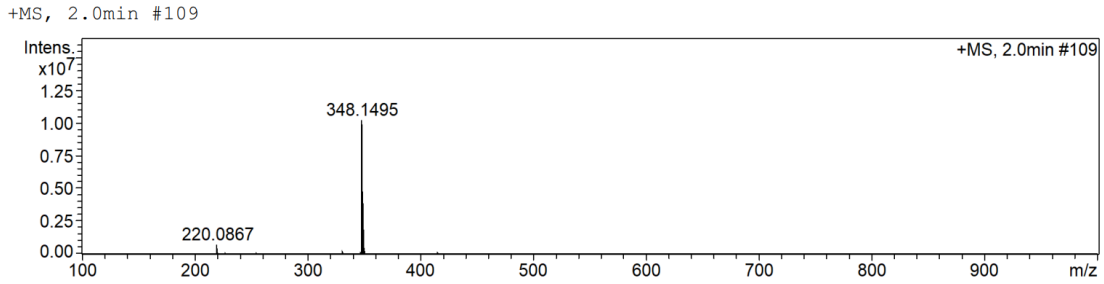
+MS, 1.5min #85



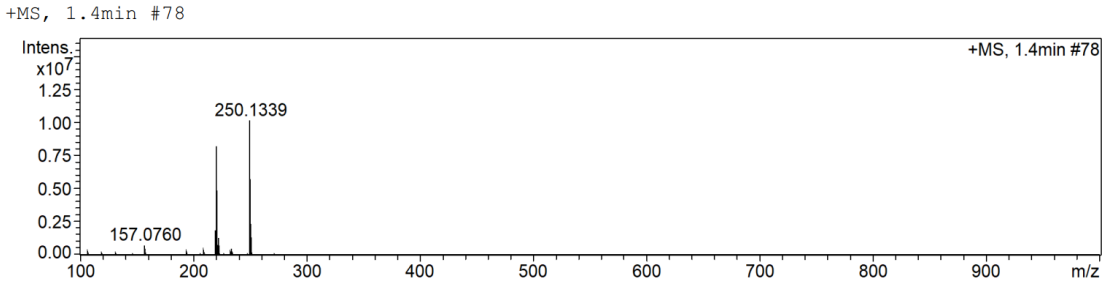
3k, HRMS calcd for $C_{21}H_{18}N_3^+$ $[M+H]^+$: 312.1495; found: 312.1495



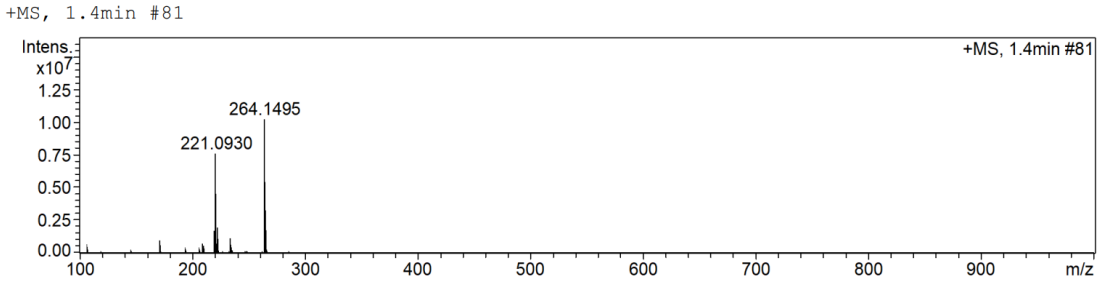
3l, HRMS calcd for $C_{24}H_{24}N_3^+$ $[M+H]^+$: 354.1965; found: 354.1965



3o, HRMS calcd for $C_{24}H_{18}N_3^+$ $[M+H]^+$: 348.1495; found: 348.1495

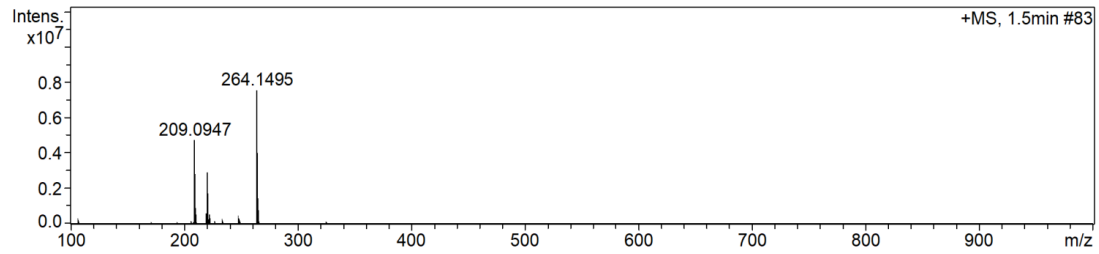


3p, HRMS calcd for $C_{16}H_{16}N_3^+$ $[M+H]^+$: 250.1339; found: 250.1339



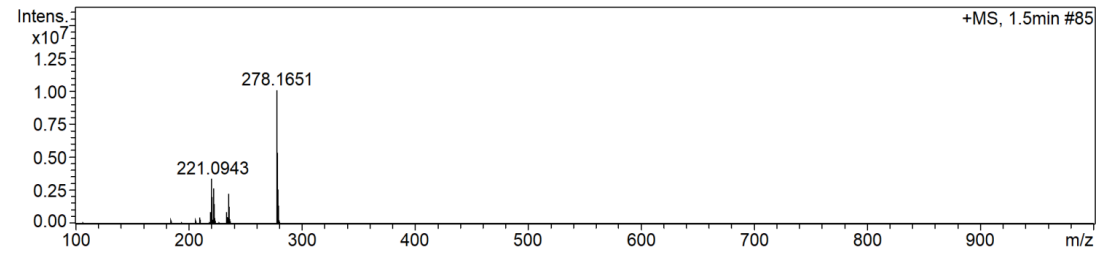
3q, HRMS calcd for $C_{17}H_{18}N_3^+$ $[M+H]^+$: 264.1495; found: 264.1495

+MS, 1.5min #83



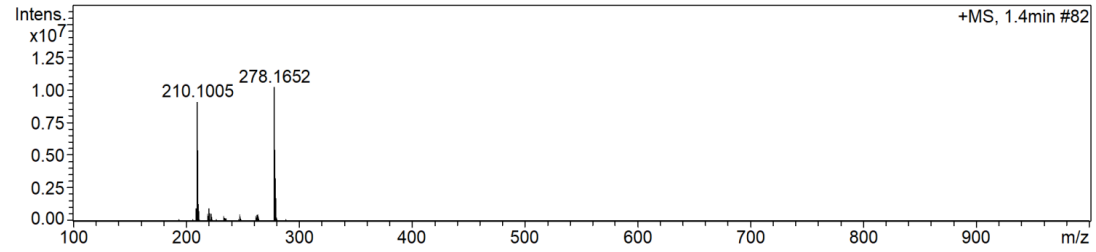
3r, HRMS calcd for $C_{17}H_{18}N_3^+$ $[M+H]^+$: 264.1495; found: 264.1495

+MS, 1.5min #85



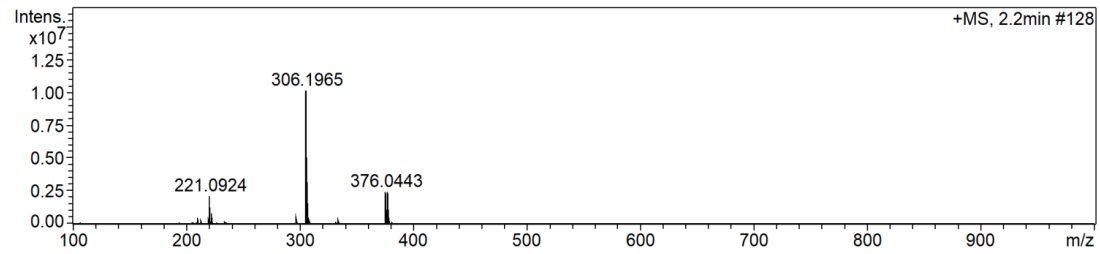
3s, HRMS calcd for $C_{18}H_{20}N_3^+$ $[M+H]^+$: 278.1652; found: 278.1651

+MS, 1.4min #82

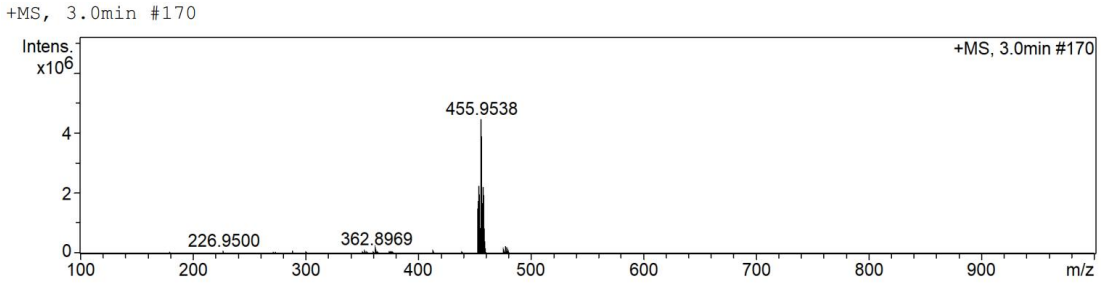


3t, HRMS calcd for $C_{18}H_{20}N_3^+$ $[M+H]^+$: 278.1652; found: 278.1652

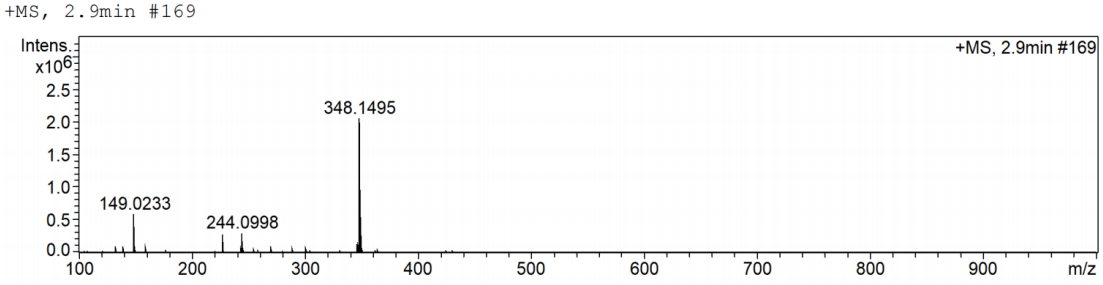
+MS, 2.2min #128



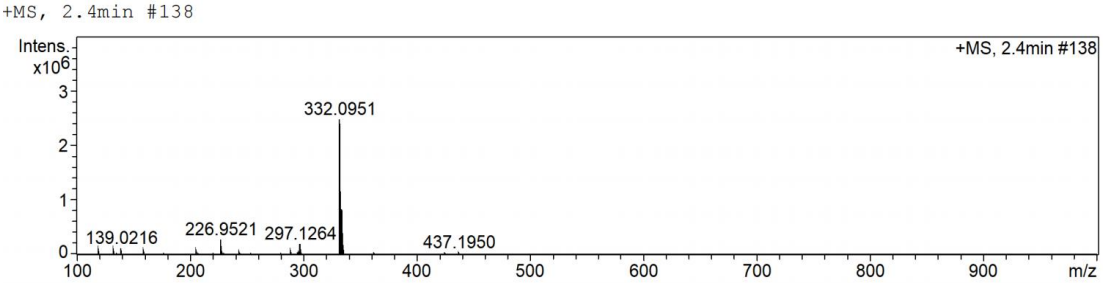
3u, HRMS calcd for $C_{20}H_{24}N_3^+$ $[M+H]^+$: 306.1965; found: 306.1965



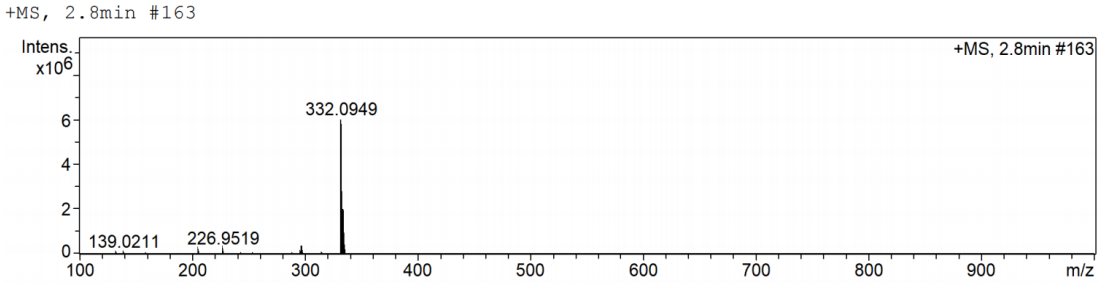
3v, HRMS calcd for $C_{20}H_{14}Br_2N_3^+$ $[M+H]^+$: 455.9540; found: 455.9538



3x, HRMS calcd for $C_{24}H_{18}N_3^+$ $[M+H]^+$: 348.1495; found: 348.1495

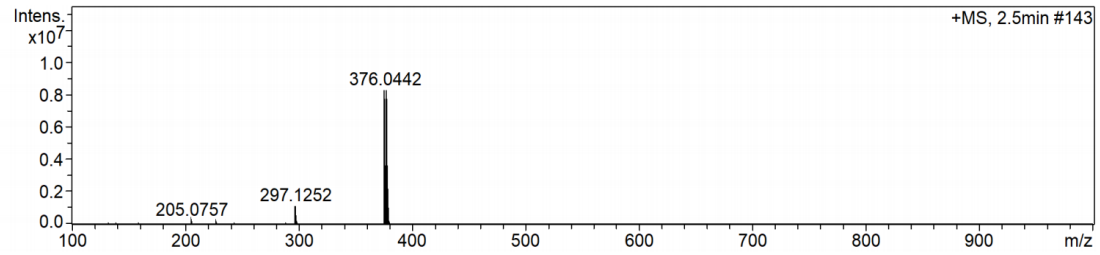


3y, HRMS calcd for $C_{20}H_{15}ClN_3^+$ $[M+H]^+$: 332.0949; found: 332.0951



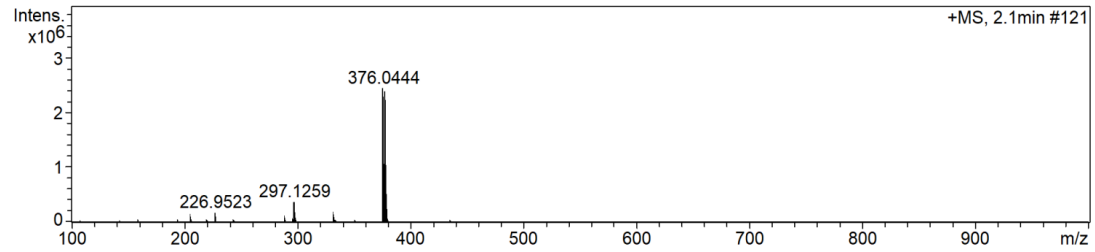
3z, HRMS calcd for $C_{20}H_{15}ClN_3^+$ $[M+H]^+$: 332.0949; found: 332.0949

+MS, 2.5min #143



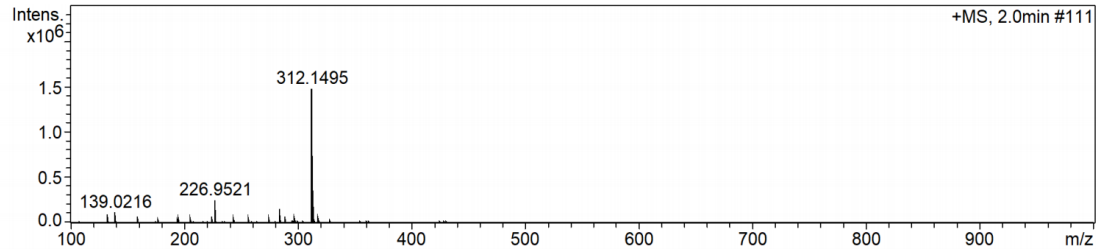
3aa, HRMS calcd for $C_{20}H_{15}BrN_3^+$ $[M+H]^+$: 376.0444; found: 376.0442

+MS, 2.1min #121



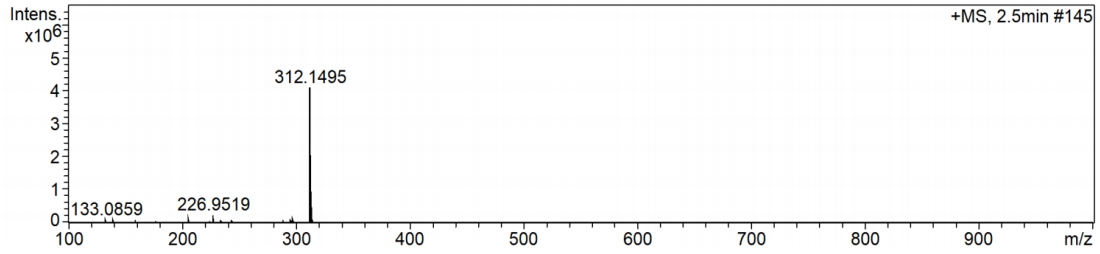
3ab, HRMS calcd for $C_{20}H_{15}BrN_3^+$ $[M+H]^+$: 376.0444; found: 376.0444

+MS, 2.0min #111

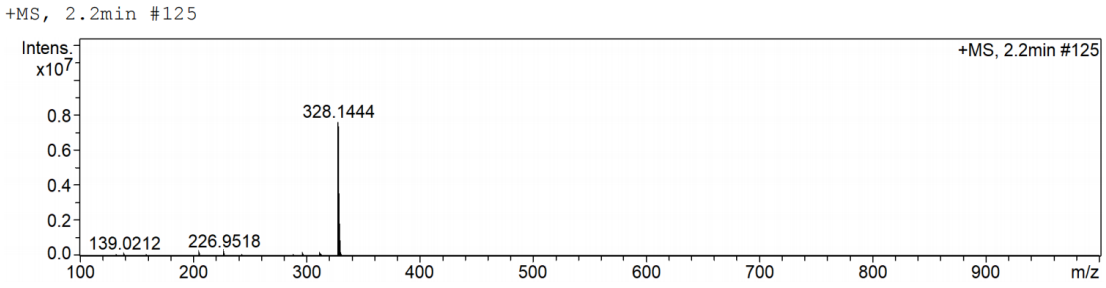


3ac, HRMS calcd for $C_{21}H_{18}N_3^+$ $[M+H]^+$: 312.1495; found: 312.1495

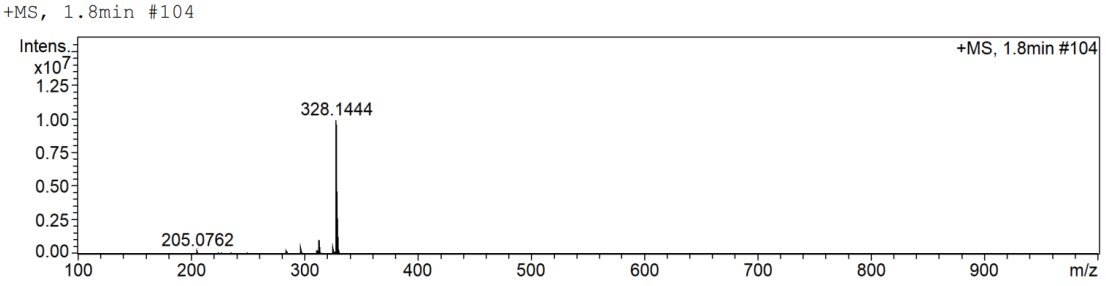
+MS, 2.5min #145



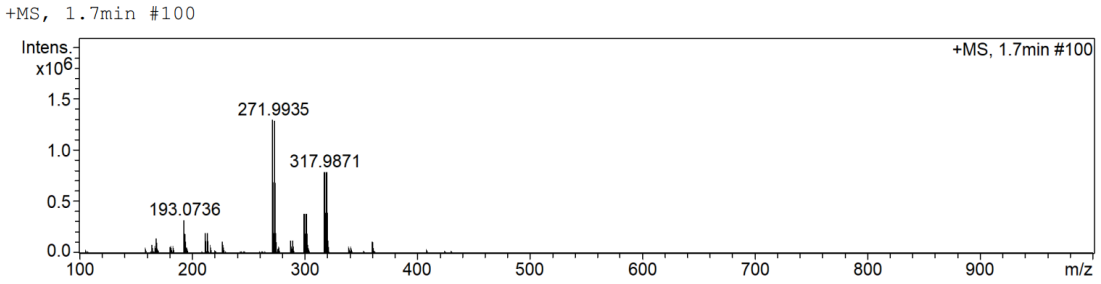
3ad HRMS calcd for $C_{21}H_{18}N_3^+$ $[M+H]^+$: 312.1495; found: 312.1495



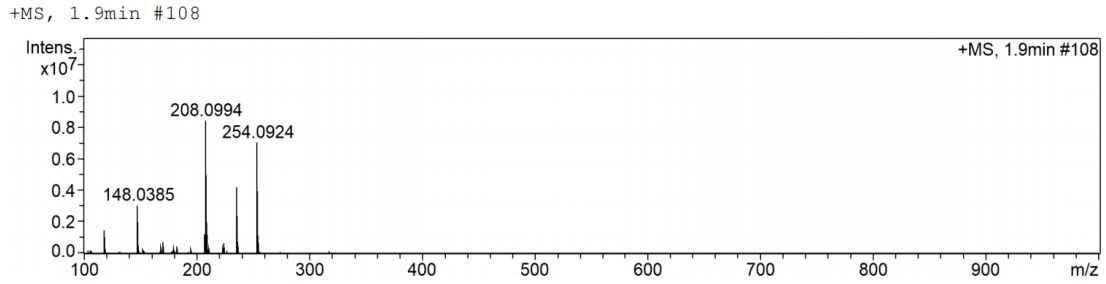
3ae HRMS calcd for $C_{21}H_{18}N_3O^+$ $[M+H]^+$: 328.1444; found: 328.1444



3af, HRMS calcd for $C_{21}H_{18}N_3O^+$ $[M+H]^+$: 328.1444; found: 328.1444

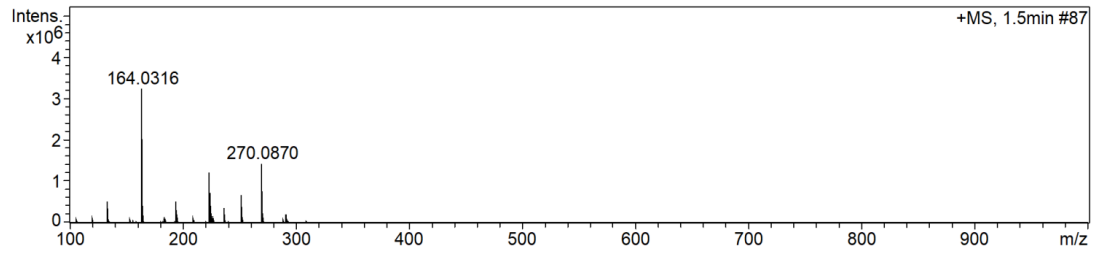


1ab, HRMS calcd for $C_{13}H_9BrN_3O_2^+$ $[M+H]^+$: 317.9873; found: 317.9871



1ad HRMS calcd for $C_{14}H_{12}N_3O_2^+$ $[M+H]^+$: 254.0924; found: 254.0924

+MS, 1.5min #87



1af, HRMS calcd for $C_{14}H_{12}N_3O_3^+$ $[M+H]^+$: 270.0873; found: 270.0870