Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supplementary Information

Electrocatalytic Oxidative C-H Cycloamination Towards Tricyclic [1,2,4]Triazolo-[3,4-*i*]purine Nucleosides Mediated by Bromide Ions

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Table of Contents

1. General Information

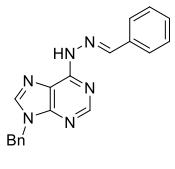
All the electrochemical oxidations were performed in an undivided cell equipped with two platinum electrodes (1.5 × 1.5 × 0.2 cm^2) unless otherwise noted. All commercial reagents were purchased from TCI, Sigma-Aldrich, Accela, Bidepharm and Adamas-beta of the highest purity grade and used without further purification. The conversion of starting materials was monitored by thin layer chromatography using silica gel plates, and components were visualized by observation under UV light (254 and 365 nm).

^{1}H NMR spectra was recorded at 400 MHz or 600 MHz. The ^{13}C NMR spectra were recorded at 100 MHz or 150 MHz. The ^{19}F NMR spectra were recorded at 376 MHz or 565 MHz. The ^{11}B NMR spectra were recorded at 128 MHz or 193 MHz. Chemical shifts were expressed in parts per million (δ) downfield from the internal standard tetramethylsilane (TMS), and were reported as s (singlet), d (doublet), t (triplet), dd (doublets of doublet), dt (doublets of triplet), td (triplets of doublet), ddd (doublets of doublets of doublet), dddd (doublets of doublets of doublets of doublet), dddt (doublets of doublets of doublets of triplet) and m (multiplet). The residual solvent signals were used as references and the chemical shifts were converted to the TMS scale ($CDCl_3$: δ H = 7.26 ppm, δ C = 77.16 ppm). CD_2Cl_2 (δ H = 5.32 ppm, δ C = 53.84 ppm), CD_3OD (δ H = 3.31 ppm, δ C = 49.00 ppm), Acetone-d_6 (δ H = 2.05 ppm, δ C = 206.26, 29.84 ppm), CD_3CN (δ H = 1.94 ppm, δ C = 1.32 ppm), DMSO-d_6 (δ H = 2.50 ppm, δ C = 39.52 ppm). The coupling constants J were given in Hz. High resolution mass spectra (HRMS) were obtained via ESI mode by using an Agilent

Q-TOF 6540 mass spectrometer. Unless otherwise noted, all other compounds have been reported in the literature or are commercially available.

2 Preparation of Substrate

2.1 Structures of Starting Materials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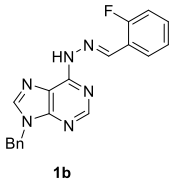
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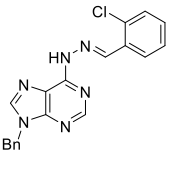
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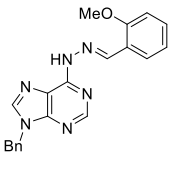
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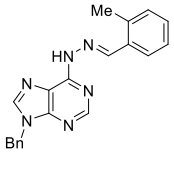
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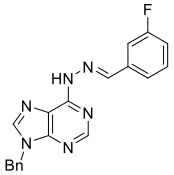
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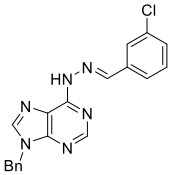
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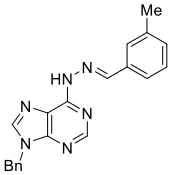
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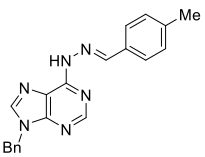
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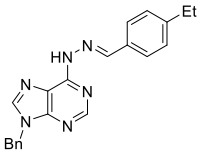
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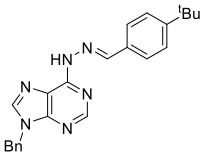
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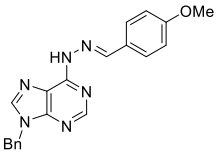
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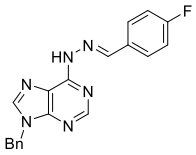
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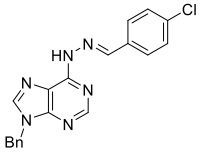
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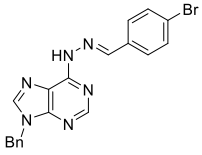
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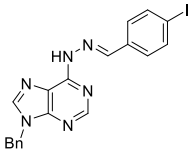
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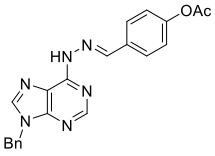
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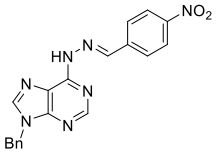
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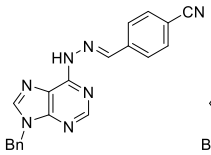
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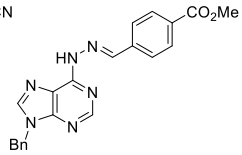
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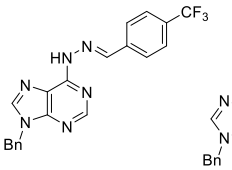
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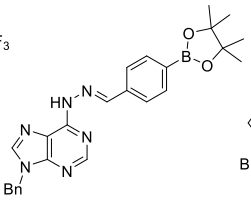
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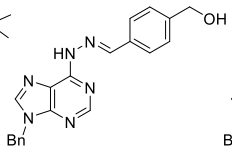
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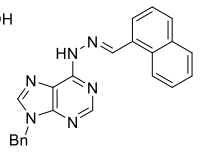
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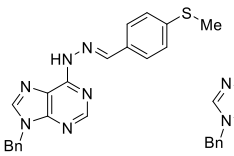
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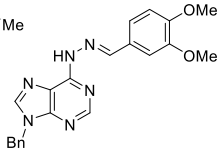
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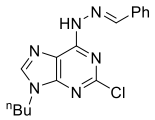
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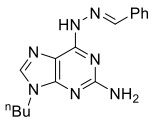
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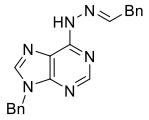
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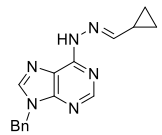
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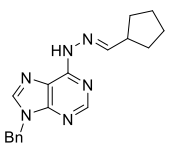
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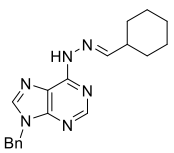
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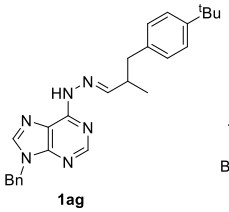
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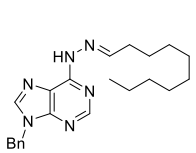
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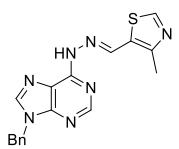
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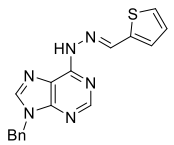
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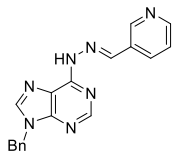
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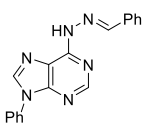
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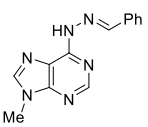
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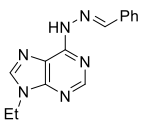
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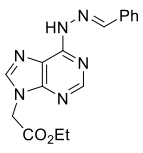

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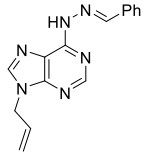

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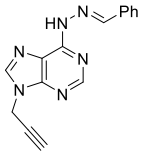

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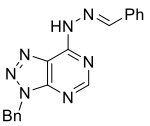

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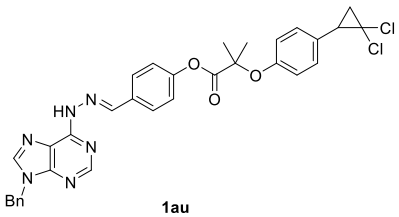

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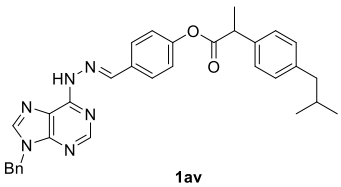

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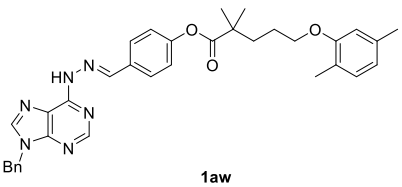

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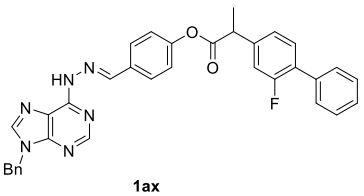

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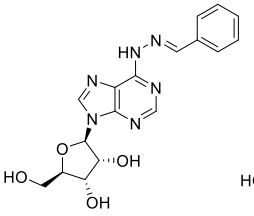

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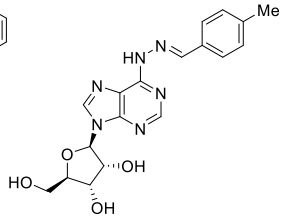

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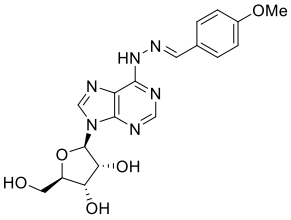

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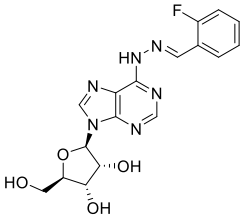

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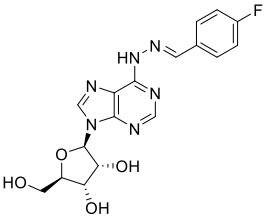

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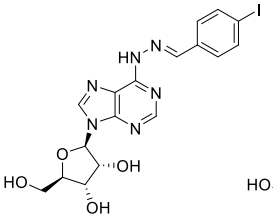

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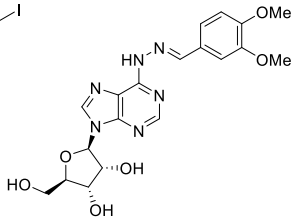
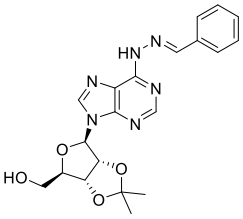
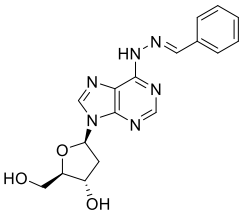


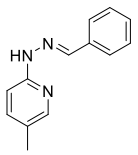
1aae



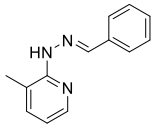
1aaf



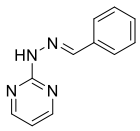
1aag



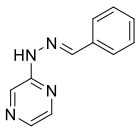
1aah



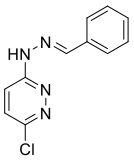
1aai



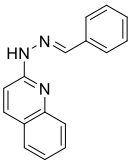
1aaj



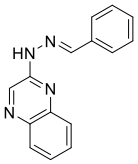
1aak



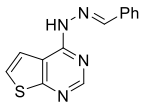
1aal



1aam

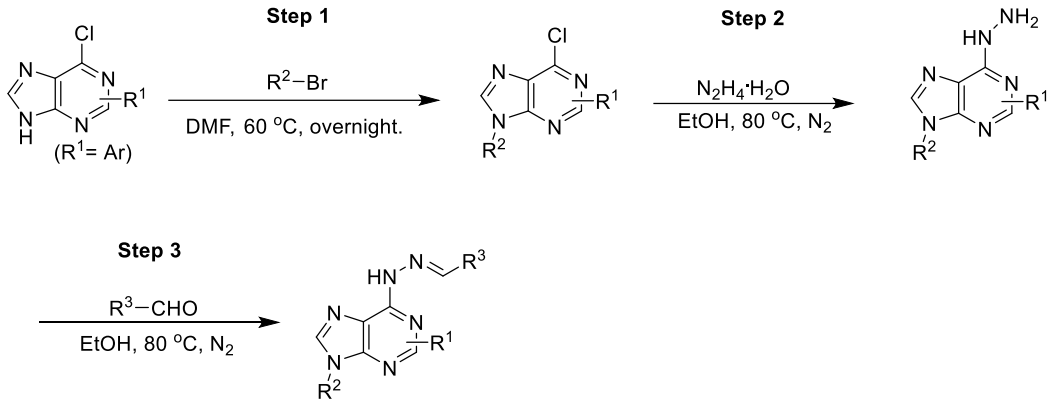


1aan



1aao

2.2 General Substrate Synthesis Steps:



Step 1.

The procedures were according to the reported method.[1] 6-Chloro-9*H*-purine (7.7280 g, 50.0 mmol), K_2CO_3 (10.3655 g, 75.0 mmol), and the appropriate alkyl bromide or benzyl bromide derivative (60.0 mmol) were combined in DMF (100 mL) in a 250 mL round-bottomed flask, and the resulting mixture was stirred for overnight at 60 °C (oil bath). After the reaction was completed (TLC), the mixture was poured into water (50.0 mL), and then extracted with CH_2Cl_2. The combined organic extracts were washed with water, dried (Na_2SO_4), and concentrated to dryness in vacuo. The residue was purified by chromatography (PE/EA = 10/1 - 4/1) to afford substrate.

Step 2.

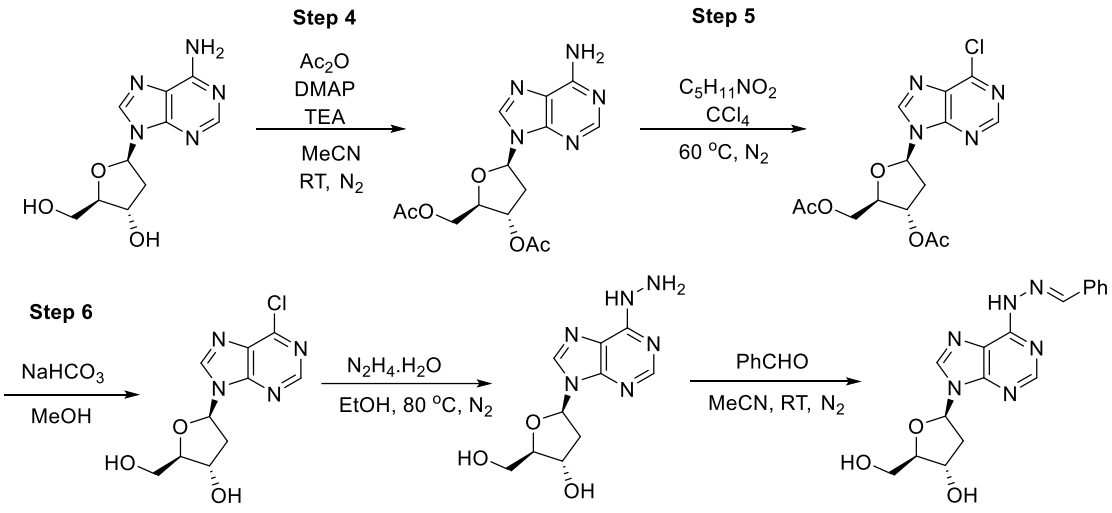
The procedures were according to the reported method.[2] To a flask equipped with a stir bar, 2-Chlorosubstituted heterocycle (5.0 mmol, 1.0 equiv), $N_2H_4 \cdot H_2O$ (80%, 3.0 mmol, 3.0 equiv) and absolute ethanol (10.0 mL, 0.1 M) were added. The solution was stirred at 80 °C (oil bath) for 12 h (TLC tracking detection). After the reaction was completed, the mixture was cooled to room temperature and solid was

precipitated. The solid was washed with petroleum ether to afford the corresponding hydrazine.

Step 3.

Method A: The procedures were according to the reported method.[3] A mixture of the hydrazine (2.0 mmol) and aldehyde (3.0 mmol) in EtOH (20.0 mL) was heated at 80 °C (oil bath) for 2 hours (TLC tracking detection) at nitrogen atmosphere. Then, the solvent was evaporated under reduced pressure. Purify the mixture residue by flash column chromatography (DCM/MeOH = 80:1) to afford substrate.

Method B: The procedures were according to the reported method.[4] A mixture of the hydrazine (2.0 mmol) and aldehyde (3.0 mmol) in 1,4-dioxane or acetonitrile was stirred at room temperature for 3 - 10 hours. Then, the solvent was evaporated under reduced pressure. Purify the mixture residue by flash column chromatography (DCM/MeOH = 80:1) to afford substrate.



Step 4.

In an oven dried round bottom flask (250 mL), free hydroxyl starting material (5.0250 g, 20.0 mmol) was taken and then dry acetonitrile (100.0 mL) was added

followed by acetic anhydride (7.5 mL, 80.0 mmol), triethyl amine (11.2 mL, 80.0 mmol) and 4-dimethylaminopyridine (977.0 mg, 8.0 mmol) were added respectively and stirred for 16 h at room temperature. The mixture was poured into water (50.0 mL), and then extracted with CH_2Cl_2. The combined organic extracts were washed with water, dried (Na_2SO_4), and concentrated to dryness in vacuo. The residue was purified by chromatography (DCM/MeOH = 100/1 - 50/1) to afford substrate.

Step 5

Isoamylnitrite (2.5 mL, 19 mmol) was added to a mixture of ((2R,3*S*,5*R*)-3-acetoxy-5-(6-amino-9*H*-purin-9-yl)tetrahydrofuran-2-yl)methyl acetate (1.4335 g, 4.3 mmol) in CCl_4 (50.0 mL) and reaction mixture was heated under nitrogen atmosphere at 60 ^oC (oil bath) overnight, the mixture was poured into water (50.0 mL), and then extracted with CH_2Cl_2. The combined organic extracts were washed with water, dried (Na_2SO_4), and concentrated to dryness in vacuo. The residue was purified by chromatography (PE/EA = 10/1 - 1/1) to afford substrate.

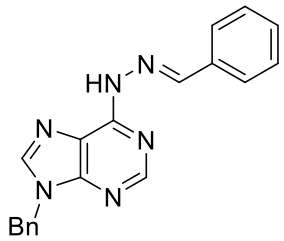
Step 6

(2*R*,3*S*,5*R*)-5-(6-chloro-9*H*-purin-9-yl)-2-(hydroxymethyl)tetrahydrofuran-3-ol (1.9014 g, 5.4 mmol), $NaHCO_3$ (0.3024 g, 3.0 mmol) were combined in MeOH (15.0 mL) in a 50 mL round-bottomed flask, and the resulting mixture was stirred for 24 h at room temperature. After the reaction was completed (TLC), dissolvent were concentrated to dryness in vacuo. The residue was purified by chromatography (DCM/MeOH = 50/1 - 20/1) to afford substrate.

1az, **1aae**, **1aan** were prepared according to a protocol reported in the literature.[5,6]

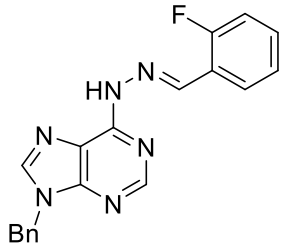
2.3 Characterization Data for New Starting Materials

(E)-9-Benzyl-6-(2-benzylidenehydrazinyl)-9*H*-purine (1a)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1a** (558.2 mg, 85%) as a white solid. **M.P.:** 201.2 - 203.6 ℃. **^{1}H NMR (600 MHz, DMSO-*d*$_6$):** δ 11.76 (s, 1 H), 8.45 (s, 1 H), 8.41 (s, 1 H), 8.37 (s, 1 H), 7.76 (d, *J* = 6.6 Hz, 2 H), 7.45 -7.42 (m, 2 H), 7.39 -7.35 (m, 1 H), 7.34 -7.31 (m, 4 H), 7.29 -7.26 (m, 1 H), 5.45 (s, 2 H). **^{13}C NMR (150 MHz, DMSO-*d*$_6$):** δ 152.2, 144.3, 142.3, 137.0, 135.0, 129.3, 128.73, 128.70, 127.8, 127.5, 126.7, 118.5, 46.3. **ESI-HRMS**: m/z calcd for $C_{19}H_{17}N_6$ $[M + H]^+$: 329.1509; found: 329.1509.

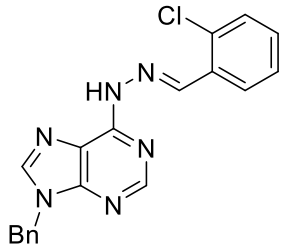
(*E*)-9-Benzyl-6-(2-(2-fluorobenzylidene)hydrazinyl)-9*H*-purine (1b)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1b** (561.1 mg, 81%) as a white solid. **M.P.:** 193.4 - 194.4 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 10.21 (s, 1 H), 8.63 (s, 1 H), 8.34 (s, 1 H), 8.20 (t, *J* = 7.2 Hz, 1 H), 7.85 (s, 1 H), 7.35 – 7.25 (m, 6 H), 7.15 (t, *J* = 7.8 Hz, 1 H), 7.03 (t, *J* = 9.6 Hz, 1 H), 5.40 (s, 2 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ

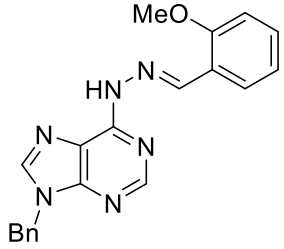
161.2 (d, $J_{C\text{-}F}$ = 249.0 Hz), 153.6, 151.6, 151.1, 141.4, 138.5, 135.4, 131.4 (d, $J_{C\text{-}F}$ = 9.0 Hz), 129.2, 128.6, 128.0, 127.4, 124.5 (d, $J_{C\text{-}F}$ = 1.2 Hz), 121.9 (d, $J_{C\text{-}F}$ = 10.5 Hz), 118.9, 115.6 (d, $J_{C\text{-}F}$ = 19.5 Hz), 47.4. **^{19}F NMR (565 MHz, $CDCl_3$):** δ -120.52. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}FN_6$ $[M + H]^+$: 347.1415; found: 347.1415.

(*E*)-9-Benzyl-6-(2-(2-chlorobenzylidene)hydrazinyl)-9*H*-purine (1c)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1c** (544.2 mg, 75%) as a light yellow solid. **M.P.:** 205.7 - 207.2 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 10.26 (s, 1 H), 8.65 (s, 1 H), 8.50 (s, 1 H), 8.29 – 8.26 (m, 1 H), 7.87 (s, 1 H), 7.37 – 7.27 (m, 8 H), 5.41 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.6, 151.6, 141.8, 141.5, 135.4, 133.8, 131.5, 130.9, 129.7, 129.3, 128.7, 128.02, 127.98, 127.2, 119.1, 47.5. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}ClN_6$ $[M + H]^+$: 363.1119; found: 363.1119.

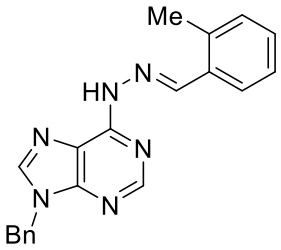
(*E*)-9-Benzyl-6-(2-(2-methoxybenzylidene)hydrazinyl)-9*H*-purine (1d)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1d** (508.9 mg, 71%) as a light yellow

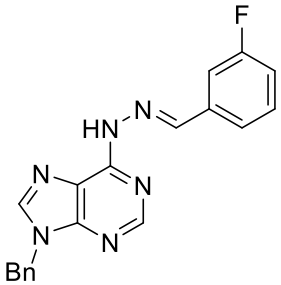
solid. **M.P.:** 184.4 - 187.5 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 9.97 (s, 1 H), 8.62 (s, 1 H), 8.50 (s, 1 H), 8.17 (d, *J* = 7.8 Hz, 1 H), 7.84 (s, 1 H), 7.36 -7.26 (m, 6 H), 6.96 (t, *J* = 7.2 Hz, 1 H), 6.84 (d, *J* = 8.4 Hz, 1 H), 5.38 (s, 2 H), 3.77 (s, 3 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 157.8, 153.6, 151.7, 150.9, 141.7, 141.1, 135.5, 131.2, 129.1, 128.5, 127.9, 127.1, 122.5, 121.0, 118.8, 110.8, 55.6, 47.3. **ESI-HRMS**: m/z calcd for $C_{20}H_{19}N_6O$ $[M + H]^+$: 359.1615; found: 359.1612.

(*E*)-9-Benzyl-6-(2-(2-methylbenzylidene)hydrazinyl)-9*H*-purine (1e)



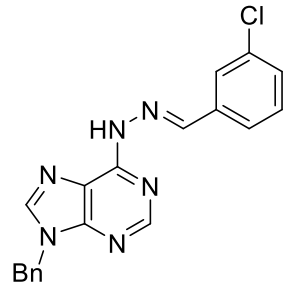
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1e** (472.5 mg, 69%) as a yellow solid. **M.P.:** 178.8 - 179.8 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.67 (s, 1 H), 8.67 (s, 1 H), 8.44 (s, 1 H), 8.39 (s, 1 H), 7.95 (t, *J* = 5.2 Hz, 1 H), 7.35 – 7.21 (m, 8 H), 5.44 (s, 2 H), 2.46 (s, 3 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 152.3, 143.1, 142.3, 136.9, 136.2, 132.8, 130.7, 129.0, 128.7, 127.7, 127.6, 126.0, 125.7, 118.4, 46.3, 19.1. **ESI-HRMS**: m/z calcd for $C_{20}H_{19}N_6$ $[M + H]^+$: 343.1666; found: 343.1667.

(*E*)-9-Benzyl-6-(2-(3-fluorobenzylidene)hydrazinyl)-9*H*-purine (1f)



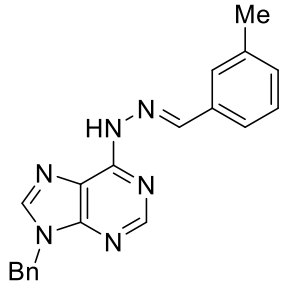
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1f** (519.5 mg, 75%) as a white solid. **M.P.:** 208.9 - 210.1 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 10.04 (s, 1 H), 8.62 (s, 1 H), 8.03 (s, 1 H), 7.86 (s, 1 H), 7.53 (d, *J* = 9.6 Hz, 1 H), 7.46 (d, *J* = 7.8 Hz, 1 H), 7.36 – 7.28 (m, 6 H), 7.05 – 7.01 (m, 1 H), 5.40 (s, 2 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 163.1(d, $J_{C\text{-}F}$ = 244.5 Hz), 153.5, 151.6, 143.9, 141.4, 136.3 (d, $J_{C\text{-}F}$ = 9.0 Hz), 135.4, 130.3 (d, $J_{C\text{-}F}$ = 7.5 Hz), 129.2, 128.7, 128.0, 123.5, 123.4, 119.1, 116.9 (d, $J_{C\text{-}F}$ = 21.0 Hz), 113.7 (d, $J_{C\text{-}F}$ = 23.0 Hz), 47.4. **^{19}F NMR (565 MHz, $CDCl_3$):** δ -112.76. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}FN_6$ $[M + H]^+$: 347.1415; found: 347.1410.

(*E*)-9-Benzyl-6-(2-(3-chlorobenzylidene)hydrazinyl)-9*H*-purine (1g)



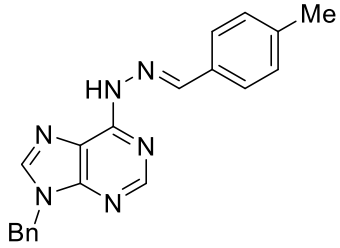
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1g** (537.0 mg, 74%) as a white solid. **M.P.:** 195.6 - 196.5 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 10.22 (s, 1 H), 8.62 (s, 1 H), 8.00 (s, 1 H), 7.84 (s, 1 H), 7.78 – 7.72 (m, 1 H), 7.60 – 7.54 (m, 1 H), 7.36 – 7.26 (m, 7 H), 5.38 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.5, 151.4, 143.8, 141.3, 135.8, 135.3, 134.8, 129.9, 129.8, 129.2, 128.6, 128.0, 127.0, 125.7, 118.6, 48.0. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}ClN_6$ $[M + H]^+$: 363.1119; found: 363.1120.

(*E*)-9-Benzyl-6-(2-(3-methylbenzylidene)hydrazinyl)-9*H*-purine (1h)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1h** (554.7 mg, 81%) as a light yellow solid. **M.P.:** 204.9 - 208.4 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.71 (s, 1 H), 8.45 (s, 1 H), 8.40 (s, 1 H), 8.32 (s, 1 H), 7.54 (d, *J* = 9.2 Hz, 2 H), 7.35 – 7.27 (m, 6 H), 7.20 (d, *J* = 7.6 Hz, 1 H), 5.44 (s, 2 H), 2.35 (s, 3 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 152.4, 151.8, 150.7, 144.8, 142.8, 138.4, 137.4, 135.4, 130.5, 129.2, 129.1, 128.2, 128.0, 127.6, 124.5, 118.8, 46.7, 21.4. **ESI-HRMS**: m/z calcd for $C_{20}H_{19}N_6$ $[M + H]^+$: 343.1666; found: 343.1670.

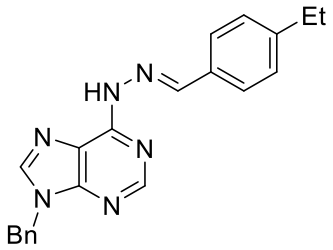
(*E*)-9-Benzyl-6-(2-(4-methylbenzylidene)hydrazinyl)-9*H*-purine (1i)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1i** (513.6 mg, 75%) as a light yellow solid. **M.P.:** 216.3 - 219.6 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.67 (s, 1 H), 8.44 (s, 1 H), 8.39 (s, 1 H), 8.31 (s, 1 H), 7.64 (d, *J* = 8.0 Hz, 2 H), 7.36 – 7.28 (m, 5 H), 7.25 (d, *J* = 8.0 Hz, 2 H), 5.44 (s, 2 H), 2.32 (s, 3 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 152.3, 144.3, 142.2, 139.0, 137.0, 132.2, 129.4, 128.7, 127.8, 127.5,

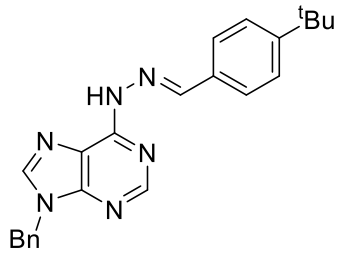
126.7, 118.3, 46.3, 21.0. **ESI-HRMS**: m/z calcd for $C_{20}H_{19}N_6$ $[M + H]^+$: 343.1666; found: 343.1668.

(*E*)-9-Benzyl-6-(2-(4-ethylbenzylidene)hydrazinyl)-9*H*-purine (1j)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1j** (520.4 mg, 73%) as a light yellow solid. **M.P.:** 216.7 - 220.8 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11. 67 (s, 1 H), 8.43 (s, 1 H), 8.38 (s, 1 H), 8.32 (s, 1 H), 7.67 (d, *J* = 8.0 Hz, 2 H), 7.35 – 7.32 (m, 4 H), 7.31 – 7.27 (m, 3 H), 5.44 (s, 2 H), 2.63 (q, *J* = 7.6 Hz, 2 H), 1.19 (t, *J* = 7.6 Hz, 3 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 145.3, 137.0, 132.5, 128.7, 128.2, 127.8, 127.5, 126.8, 46.3, 28.1, 15.4. **ESI-HRMS**: m/z calcd for $C_{21}H_{21}N_6$ $[M + H]^+$: 357.1822; found: 357.1814.

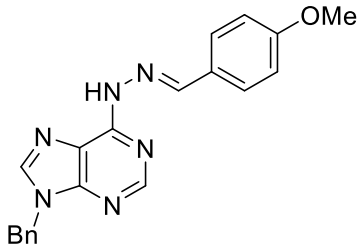
(*E*)-9-Benzyl-6-(2-(4-(*tert*-butyl)benzylidene)hydrazinyl)-9*H*-purine (1k)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1k** (630.6 mg, 82%) as a white solid. **M.P.:** >300 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.67 (s, 1 H), 8.44 (s, 1 H), 8.39

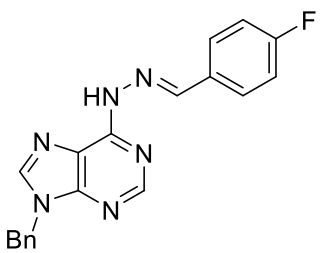
(s, 1 H), 8.33 (s, 1 H), 7.67 (d, *J* = 8.0 Hz, 2 H), 7.45 (d, *J* = 8.4 Hz, 2 H), 7.35 -7.32 (m, 4 H), 7.30 -7.26 (m, 1 H), 5.44 (s, 2 H), 1.29 (s, 9 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 152.4, 152.1, 141.2, 137.0, 132.2, 128.7, 127.8, 127.5, 126.5, 125.5, 118.3, 46.2, 40.2, 34.5, 31.0. **ESI-HRMS**: m/z calcd for $C_{23}H_{25}N_6$ $[M + H]^+$: 385.2135; found: 385.2126.

(*E*)-9-Benzyl-6-(2-(4-methoxybenzylidene)hydrazinyl)-9*H*-purine (1l)



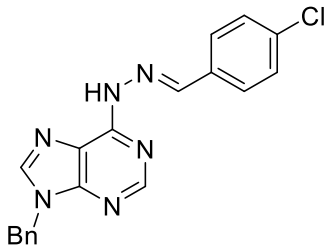
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 80/1-40/1) yielded **1l** (566.3 mg, 79%) as a white solid. **M.P.:** 203.0 - 204.5 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 9.78 (s, 1 H), 8.60 (s, 1 H), 8.01 (s, 1 H), 7.82 (s, 1 H), 7.70 (d, *J* = 8.4 Hz, 2 H), 7.35 – 7.26 (m, 5 H), 6.88 (d, *J* = 8.4 Hz, 2 H), 5.38 (s, 2 H), 3.81 (s, 3 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 161.3, 153.6, 151.5, 145.6, 141.0, 135.5, 129.2, 129.1, 128.6, 127.9, 126.7, 118.8, 114.2, 55.4, 47.3. **ESI-HRMS**: m/z calcd for $C_{20}H_{19}N_6O$ $[M + H]^+$: 359.1615; found: 359.1618.

(*E*)-9-Benzyl-6-(2-(4-fluorobenzylidene)hydrazinyl)-9*H*-purine (1m)



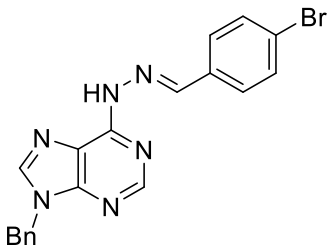
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1m** (554.2 mg, 80%) as a white solid. **M.P.:** 191.8 - 193.0 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 10.21 (s, 1 H), 8.60 (s, 1 H), 8.01 (s, 1 H), δ 7.83 (s, 1 H), 7.71 – 7.65 (m, 2 H), 7.34 – 7.25 (m, 5 H), 7.01 (t, *J* = 8.4 Hz, 2 H), 5.37 (s, 2 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 163.8 (d, $J_{C\text{-}F}$ = 249.0 Hz), 153.5, 151.7, 144.2, 141.3, 135.4, 130.2, 129.2 (d, $J_{C\text{-}F}$ = 7.5 Hz), 129.1, 128.6, 127.9, 118.9, 115.9, 115.7, 47.3. **^{19}F NMR (565 MHz, $CDCl_3$):** δ -110.12. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}FN_6$ $[M + H]^+$: 347.1415; found: 347.1413.

(*E*)-9-Benzyl-6-(2-(4-chlorobenzylidene)hydrazinyl)-9*H*-purine (1n)



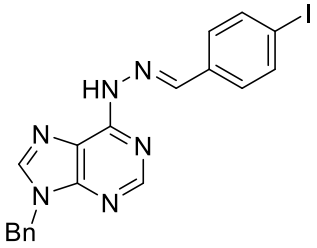
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1n** (587.8 mg, 81%) as a yellow solid. **M.P.:** 197.1 - 198.6 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.70 (s, 1 H), 8.63 (s, 1 H), 8.02 (s, 1 H), 7.85 (s, 1 H), 7.72 (dt, *J* = 9.2, 2.4 Hz, 2 H), 7.38 – 7.28 (m, 7 H), 5.41 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.6, 151.4, 144.0, 141.4, 135.9, 135.4, 132.5, 131.5, 129.3, 129.1, 128.8, 128.7, 128.0, 119.0, 47.5. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}ClN_6$ $[M + H]^+$: 363.1119; found: 363.1121.

(*E*)-9-Benzyl-6-(2-(4-bromobenzylidene)hydrazinyl)-9*H*-purine (1o)



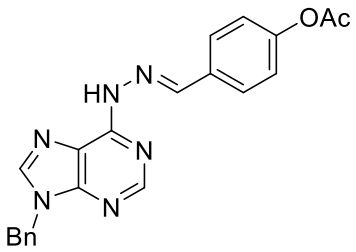
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1o** (586.4 mg, 72%) as a light yellow solid. **M.P.:** 203.8 - 206.0 ℃. **^{1}H NMR (600 MHz, DMSO-d_6):** δ 10.82 (s, 1 H), 8.46 (s, 1 H), 8.40 (s, 1 H), 8.30 (s, 1 H), 7.70 (d, *J* = 7.8 Hz, 2 H), 7.64 (d, *J* = 7.8 Hz, 2 H), 7.36 – 7.27 (m, 5 H), 5.44 (s, 2 H). **^{13}C NMR (150 MHz, DMSO-d_6):** δ 152.3, 142.7, 142.5, 136.9, 134.3, 131.7, 128.7, 128.5, 127.8, 127.5, 122.4, 118.5, 46.3. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}BrN_6$ $[M + H]^+$: 407.0614; found: 407.0612.

(*E*)-9-Benzyl-6-(2-(4-iodobenzylidene)hydrazinyl)-9*H*-purine (1p)



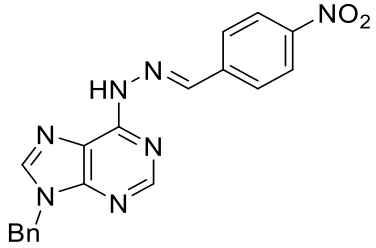
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1p** (626.9 mg, 69%) as a white solid. **M.P.:** 211.9 - 213.8 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.80 (s, 1 H), 8.46 (s, 1 H), 8.40 (s, 1 H), 8.28 (s, 1 H), 7.83 – 7.78 (m, 2 H), 7.56 – 7.51 (m, 2 H), 7.36 – 7.27 (m, 5 H), 5.44 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 152.3, 151.0, 143.0, 142.5, 137.6, 136.9, 134.5, 128.7, 128.4, 127.8, 127.5, 118.4, 95.7, 46.3. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}IN_6$ $[M + H]^+$: 455.0476; found: 455.0472.

(*E*)-4-((2-(9-Benzyl-9*H*-purin-6-yl)hydrazono)methyl)phenyl acetate (1q)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1q** (633.7 mg, 82%) as a white solid. **M.P.:** 253.3 - 256.1 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.58 (s, 1 H), 8.63 (s, 1 H), 8.03 (s, 1 H), 7.83 (s, 1 H), 7.79 (d, *J* = 8.4 Hz, 2 H), 7.37 -7.28 (m, 5 H), 7.13 (d, *J* = 8.4 Hz, 2 H), 5.40 (s, 2 H), 2.30 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 169.3, 152.0, 144.3, 141.3, 135.5, 131.7, 129.3, 128.7, 128.6, 128.0, 122.0, 47.4, 21.3. **ESI-HRMS**: m/z calcd for $C_{21}H_{19}N_6O_2$ $[M + H]^+$: 387.1564; found: 387.1565.

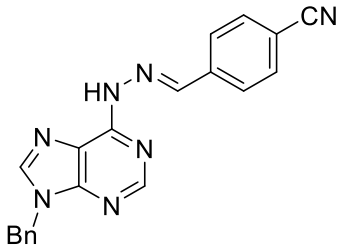
(*E*)-9-Benzyl-6-(2-(4-nitrobenzylidene)hydrazinyl)-9*H*-purine (1r)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1r** (485.4 mg, 65%) as a yellow solid. **M.P.:** 241.1 - 242.6 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 12.11 (s, 1 H), 8.51 (s, 1 H), 8.45 (s, 1 H), 8.41 (s, 1 H), 8.29 (d, *J* = 8.8 Hz, 2 H), 7.99 (d, *J* = 8.8 Hz, 2 H), 7.35 – 7.26 (m, 5 H), 5.46 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 152.3, 151.3,

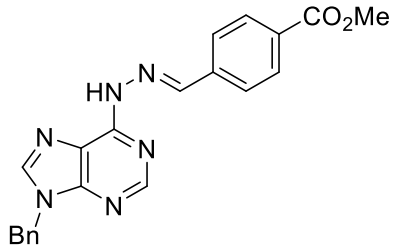
147.2, 143.0, 141.4, 136.9, 128.7, 127.8, 127.6, 127.3, 124.1, 118.7, 46.3. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}N_7O_2$ $[M + H]^+$: 374.1360; found: 374.1357.

(*E*)-4-((2-(9-Benzyl-9*H*-purin-6-yl)hydrazono)methyl)benzonitrile (1s)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1s** (494.7 mg, 70%) as a yellow solid. **M.P.:** 228.0 - 232.1 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 12.03 (s, 1 H), 8.50 (s, 1 H), 8.43 (s, 1 H), 8.34 (s, 1 H), 7.94 – 7.87 (m, 4 H), 7.36 – 7.28 (m, 5 H), 5.45 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 152.2, 142.9, 141.7, 139.5, 136.9, 132.7, 128.7, 127.8, 127.5, 127.1, 118.9, 118.7, 110.9, 46.3. **ESI-HRMS**: m/z calcd for $C_{20}H_{16}N_7$ $[M + H]^+$: 354.1462; found: 354.1464.

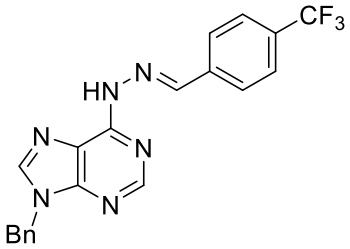
Methyl (*E*)-4-((2-(9-benzyl-9*H*-purin-6-yl)hydrazono)methyl)benzoate (1t)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1t** (494.6mg, 64%) as a white solid. **M.P.:** 216.3 - 217.8 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.95 (s, 1 H), 8.48 (s, 1 H), 8.43 (s, 1 H), 8.31 (s, 1 H), 8.02 (d, *J* = 8.4 Hz, 2 H), 7.88 (d, *J* = 8.0 Hz, 2 H),

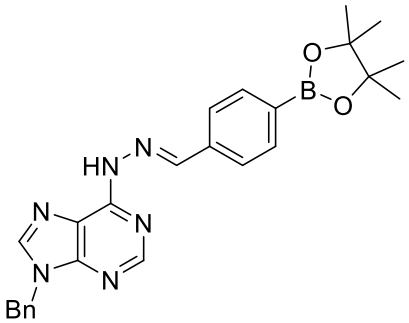
7.35 – 7.28 (m, 5 H), 5.45 (s, 2 H), 3.87 (s, 3 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 165.9, 152.4, 142.7, 139.5, 136.9, 129.6, 128.7, 127.8, 127.6, 126.7, 118.6, 52.2, 46.3. **ESI-HRMS**: m/z calcd for $C_{21}H_{19}N_6O_2$ $[M + H]^+$: 387.1564; found: 387.1563.

(*E*)-9-Benzyl-6-(2-(4-(trifluoromethyl)benzylidene)hydrazinyl)-9*H*-purine (1u)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1u** (562.8 mg, 71%) as a yellow solid. **M.P.:** 210.2 - 211.9 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 10.01 (s, 1 H), 8.64 (s, 1 H), 8.08 (s, 1 H), 7.87 – 7.84 (m, 3 H), 7.61 (d, J = 8.4 Hz, 2 H), 7.36 – 7.28 (m, 5 H), 5.41 (s, 2 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 153.5, 151.5, 143.3, 141.6, 137.4, 135.4, 131.5, 131.3, 129.3, 128.7, 128.0, 127.6, 125.7(d, $J_{C\text{-}F}$ = 3.0 Hz), 125.0, 123.2, 119.2, 47.5. **^{19}F NMR (565 MHz, $CDCl_3$):** δ - 62.68. **ESI-HRMS**: m/z calcd for $C_{20}H_{16}F_3N_6$ $[M + H]^+$: 397.1383; found: 397.1384.

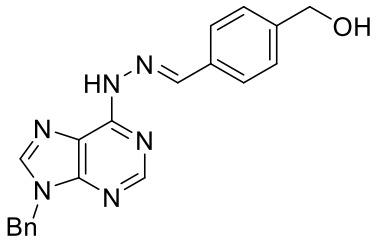
(*E*)-9-Benzyl-6-(2-(4-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)benzylidene)hydrazinyl)-9*H*-purine (1v)



Follow the general procedure, product purification by column chromatography on

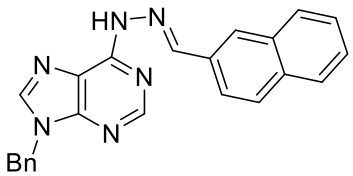
silica gel (DCM/ MeOH = 100/1-50/1) yielded **1v** (581.6 mg, 64%) as a white solid. **M.P.:** 287.2 - 291.3 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.29 (s, 1 H), 8.66 (s, 1 H), 8.05 (s, 1 H), 7.85 – 7.80 (m, 5 H), 7.38 – 7.30 (m, 5 H), 5.41 (s, 2 H), 1.36 (s, 12 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.8, 151.3, 151.0, 145.5, 141.2, 136.3, 135.4, 135.1, 129.3, 128.7, 128.0, 126.8, 118.8, 84.1, 47.5, 25.0. **^{11}B NMR (193 MHz, $CDCl_3$):** δ 31.51. **ESI-HRMS**: m/z calcd for $C_{25}H_{28}BN_6O_2$ $[M + H]^+$: 455.2361; found: 455.2351.

(*E*)-(4-((2-(9-Benzyl-9*H*-purin-6-yl)hydrazono)methyl)phenyl)methanol (1w)



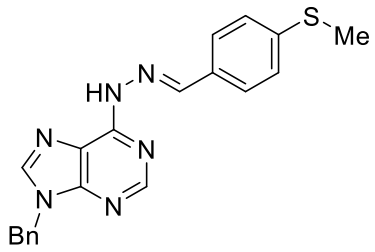
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 60/1-20/1) yielded **1w** (595.0 mg, 83%) as a yellow solid. **M.P.:** 275.2 - 278.7 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.72 (s, 1 H), 8.45 (s, 1 H), 8.42 (s, 1 H), 8.37 (s, 1 H), 7.72 (d, *J* = 8.0 Hz, 2 H), 7.40 (d, *J* = 4.4 Hz, 2 H), 7.35 – 7.26 (m, 5 H), 5.45 (s, 2 H), 4.54 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 152.4, 150.9, 144.3, 143.99, 143.95, 142.3, 137.0, 133.4, 128.7, 127.8, 127.6, 126.7, 126.5, 118.3, 62.6, 46.3. **ESI-HRMS**: m/z calcd for $C_{20}H_{19}N_6O$ $[M + H]^+$: 359.1615; found: 359.1616.

(*E*)-9-Benzyl-6-(2-(naphthalen-2-ylmethylene)hydrazinyl)-9*H*-purine (1x)



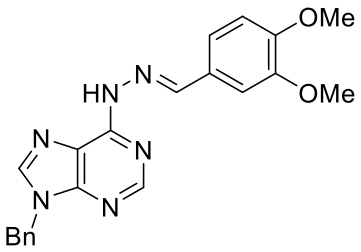
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1x** (590.4 mg, 78%) as a yellow solid. **M.P.:** 93.6 - 97.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 10.25 (s, 1 H), 8.77 (d, *J* = 10.4 Hz, 2 H), 8.65 (s, 1 H), 8.02 (d, *J* = 7.2 Hz, 1 H), 7.88 (s, 1 H), 7.85 (dd, *J* = 8.4, 2.8 Hz, 2 H), 7.58 – 7.53 (m, 1 H), 7.52 – 7.44 (m, 2 H), 7.34 – 7.26 (m, 5 H), 5.38 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.4, 151.9, 144.6, 141.3, 135.5, 133.9, 130.9, 130.6, 129.6, 129.2, 128.8, 128.6, 127.9, 127.6, 127.3, 126.1, 125.4, 124.2, 119.1, 47.4. **ESI-HRMS**: m/z calcd for $C_{23}H_{19}N_6$ $[M + H]^+$: 379.1666; found: 379.1664.

(*E*)-9-Benzyl-6-(2-(4-(methylthio)benzylidene)hydrazinyl)-9*H*-purine (1y)



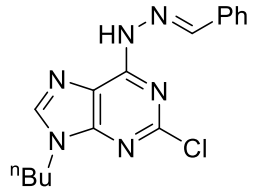
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1y** (539.2 mg, 72%) as a yellow solid. **M.P.:** 206.2 - 208.9 ℃. **^{1}H NMR (600 MHz, DMSO-d_6):** δ 11.69 (s, 1 H), 8.44 (s, 1 H), 8.38 (s, 1 H), 7.68 (d, *J* = 8.4 Hz, 2 H), 7.37 – 7.27 (m, 8 H), 5.44 (s, 2 H), 2.51 (s, 3 H). **^{13}C NMR (150 MHz, DMSO-d_6):** δ 142.3, 139.9, 137.0, 131.5, 128.7, 127.8, 127.5, 127.1, 125.7, 118.3, 46.2, 30.7, 14.4. **ESI-HRMS**: m/z calcd for $C_{20}H_{18}N_6SNa$ $[M + Na]^+$: 397.1206; found: 397.1200.

(*E*)-9-Benzyl-6-(2-(3,4-dimethoxybenzylidene)hydrazinyl)-9*H*-purine (1z)



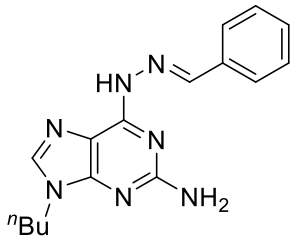
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1z** (637.0 mg, 82%) as a yellow solid. **M.P.:** 213.5 - 214.9 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.62 (s, 1 H), 8.60 (s, 1 H), 7.99 (s, 1 H), 7.80 (s, 1 H), 7.50 (d, *J* = 2.0 Hz, 1 H), 7.37 – 7.27 (m, 5 H), 7.12 (dd, *J* = 4.4, 2.0 Hz, 1 H), 6.83 (d, *J* = 4.4 Hz, 1 H), 5.38 (s, 2 H), 3.56 (s, 3 H) 3.89 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.7, 151.5, 151.1, 149.4, 145.8, 141.0, 135.4, 129.2, 128.6, 127.9, 127.0, 122.3, 118.8, 110.7, 108.7, 56.2, 56.0, 47.4. **ESI-HRMS**: m/z calcd for $C_{21}H_{21}N_6O_2$ $[M + H]^+$: 389.1721; found: 389.1719.

(E)-6-(2-Benzylidenehydrazinyl)-9-butyl-2-chloro-9H-purine (1aa)



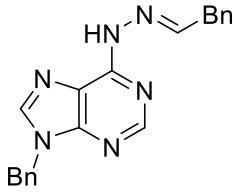
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1aa** (427.4 mg, 65%) as a white solid. **M.P.:** 190.1 - 193.6 ℃. **^{1}H NMR (600 MHz, DMSO-*d_6*):** δ 8.39 (s, 1 H), 7.89 (s, 2 H), 7.47 -7.41 (m, 4 H), 4.26 (s, 2 H), 1.85 - 1.80 (m, 2 H), 1.33 - 1.26 (m, 2 H), 0.90 (t, *J* = 7.2 Hz, 3 H). **^{13}C NMR (150 MHz, DMSO-*d_6*):** δ 153.9, 151.3, 142.8, 133.6, 130.3, 128.8, 127.7, 30.9, 19.1, 13.4. **ESI-HRMS**: m/z calcd for $C_{16}H_{18}ClN_6$ $[M + H]^+$: 329.1276; found: 329.1275.

(*E*)-9-Butyl-6-(2-(2-phenylethylidene)hydrazinyl)-9*H*-purin-2-amine (1ab)



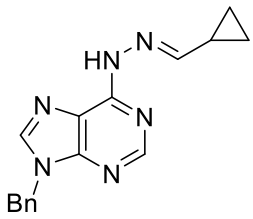
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ab** (451.7 mg, 73%) as a yellow solid. **M.P.:** 178.2 - 181.4 ℃. **^{1}H NMR (400 MHz, CD_3OD):** δ 8.17 (s, 1 H), 7.87 -7.83 (m, 2 H), 7.83 (s, 1 H), 7.44 – 7.35 (m, 3 H), 4.09 (t, *J* = 7.2 Hz, 2 H), 1.87 – 1.78 (m, 2 H), 1.41 – 1.31 (m, 2 H), 0.97 (t, *J* = 7.2 Hz, 3 H). **^{13}C NMR (100 MHz, CD_3OD):** δ 161.4, 153.5, 152.7, 147.0, 140.6, 136.1, 130.8, 129.7, 128.4, 113.5, 33.0, 20.8, 13.9. **ESI-HRMS**: m/z calcd for $C_{16}H_{20}N_7$ $[M + H]^+$: 310.1775; found: 310.1775.

(*E*)-9-Benzyl-6-(2-(2-phenylethylidene)hydrazinyl)-9*H*-purine (1ac)



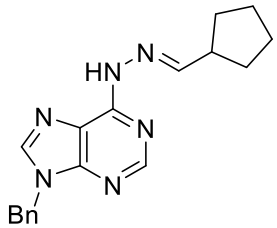
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ac** (390.3 mg, 57%) as a white solid. **M.P.:** 201.1 - 203.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.60 (s, 1 H), 7.71 (s, 1 H), 7.45 (t, *J* = 6.0 Hz, 1 H), 7.36 – 7.28 (m, 8 H), 7.25 – 7.21 (m, 2 H), 5.34 (s, 2 H), 3.79 (d, *J* = 5.6 Hz, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.7, 151.5, 147.7, 141.0, 136.6, 135.5, 129.22, 129.15, 128.9, 128.6, 128.0, 127.0, 118.4, 47.4, 39.0. **ESI-HRMS**: m/z calcd for $C_{20}H_{19}N_6$ $[M + H]^+$: 343.1666; found: 343.1666.

(*E*)-9-Benzyl-6-(2-(cyclopropylmethylene)hydrazinyl)-9*H*-purine (1ad)



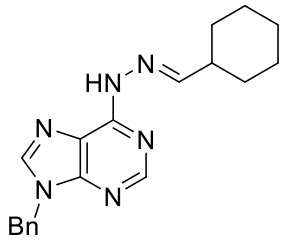
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ad** (309.9 mg, 53%) as a white solid. **M.P.:** 168.5 - 171.9 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 8.83 (s, 1 H), 8.52 (s, 1 H), 7.63 (s, 1 H), 7.40 - 7.27 (m, 6 H), 5.54 (s, 2 H), 1.84 (s, 1 H), 1.04 – 0.98 (m, 2 H), 0.90 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 160.3, 148.9, 144.6, 135.9, 128.8, 128.1, 127.8, 47.1, 14.1, 7.3. **ESI-HRMS**: m/z calcd for $C_{16}H_{17}N_6$ $[M + H]^+$: 293.1509; found: 293.1508.

(*E*)-9-Benzyl-6-(2-(cyclopentylmethylene)hydrazinyl)-9*H*-purine (1ae)



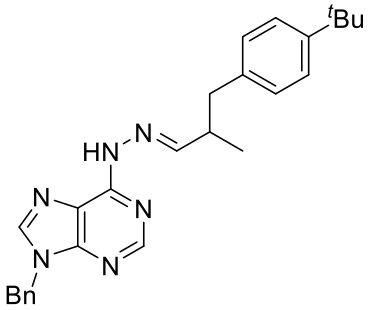
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ae** (339.6 mg, 53%) as a yellow solid. **M.P.:** 143.2 - 147.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.32 (s, 1 H), 8.55 (s, 1 H), 7.75 (s, 1 H), 7.36 – 7.26 (m, 6 H), 5.36 (s, 2 H), 3.00 – 2.90 (m, 1 H), 1.96 – 1.87 (m, 2 H), 1.68 – 1.60 (m, 4 H), 1.51 – 1.43 (m, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.7, 153.6, 151.5, 150.6, 140.8, 135.5, 129.2, 128.6, 128.0, 118.3, 47.3, 42.7, 31.2, 25.7. **ESI-HRMS**: m/z calcd for $C_{18}H_{21}N_6$ $[M + H]^+$: 321.1822; found: 321.1819.

(*E*)-9-Benzyl-6-(2-(cyclohexylmethylene)hydrazinyl)-9*H*-purine (1af)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1af** (327.7 mg, 49%) as a white solid. **M.P.:** 208.3 - 210.9 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.82 (s, 1 H), 8.57 (s, 1 H), 7.78 (s, 1 H), 7.38 – 7.27 (m, 5 H), 7.23 (d, *J* = 6.8 Hz, 1 H), 5.38 (s, 2 H), 2.58 – 2.50 (m, 1 H), 1.89 – 1.76 (m, 5 H), 1.32 – 1.26 (m, 5 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.8, 153.5, 140.9, 135.6, 129.3, 128.6, 128.0, 47.4, 41.0, 30.6, 25.9, 25.5. **ESI-HRMS**: m/z calcd for $C_{19}H_{23}N_6$ $[M + H]^+$: 335.1979; found: 335.1980.

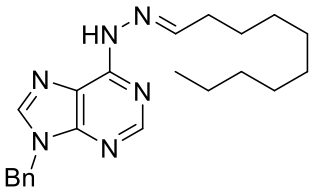
(*E*)-9-Benzyl-6-(2-(3-(4-(*tert*-butyl)phenyl)-2-methylpropylidene)hydrazinyl)-9*H*-purine (1ag)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ag** (486.3 mg, 57%) as a yellow solid. **M.P.:** 127.2 - 132.3 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.92 (s, 1 H), 8.60 (s, 1 H), 7.77 (s, 1 H), 7.36 – 7.27 (m, 9 H), 7.12 (dd, *J* = 6.8, 2.0 Hz, 2 H), 5.38 (s, 2 H), 2.91 (q, *J* = 6.4 Hz, 1 H), 2.65 (dd, *J* = 13.6, 8.4 Hz, 1 H), 1.30 (s, 9 H), 1.14 (d, *J* = 6.8 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.9, 153.4, 151.4, 150.7, 149.2, 140.8, 136.0,

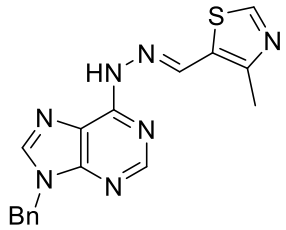
135.5, 129.3, 129.1, 128.7, 128.0, 125.4, 118.4, 47.4, 40.6, 38.1, 34.5, 31.5, 17.8. **ESI-HRMS**: m/z calcd for $C_{26}H_{31}N_6$ $[M + H]^+$: 427.2605; found: 427.2608.

6-(*E*)-9-Benzyl-6-(2-decylidenehydrazinyl)-9*H*-purine (1ah)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ah** (476.9 mg, 63%) as a yellow solid. **M.P.:** 121.3 - 124.8 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.03 (s, 1 H), 8.57 (s, 1 H), 7.77 (s, 1 H), 7.39 – 7.27 (m, 6 H), 5.37 (s, 2 H), 2.47 – 2.41 (m, 2 H), 1.52 (q, *J* = 7.2 Hz, 2 H), 1.35 – 1.24 (m, 12 H), 0.86 (t, *J* = 6.4 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.8, 151.5, 150.7, 149.8, 140.9, 135.5, 129.2, 128.6, 128.0, 127.8, 118.4, 47.4, 32.5, 32.0, 29.6, 29.5, 29.42, 29.36, 27.1, 22.8, 14.2. **ESI-HRMS**: m/z calcd for $C_{22}H_{31}N_6$ $[M + H]^+$: 379.2605; found: 379.2606.

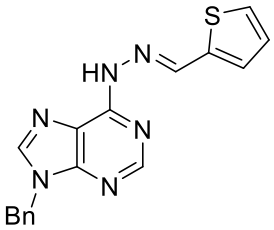
(*E*)-5-((2-(9-Benzyl-9*H*-purin-6-yl)hydrazono)methyl)-4-methylthiazole (1ai)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 60/1-30/1) yielded **1ai** (496.2 mg, 71%) as a yellow solid. **M.P.:** 230.1 - 232.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.93 (s, 1 H), 8.69 (s, 1 H), 8.62 (s, 1 H), 8.37(s, 1 H), 7.83 (s, 1 H), 7.38 -7.31 (m, 3 H), 7.30 -7.28 (m, 2 H), 5.39 (s, 2 H), 2.51 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.5, 141.4, 137.8, 135.4,

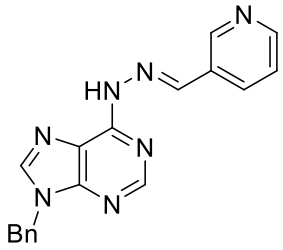
129.3, 128.7, 128.0, 47.5, 15.7. **ESI-HRMS**: m/z calcd for $C_{17}H_{16}N_7S$ $[M + H]^+$: 350.1182; found: 350.1181.

(*E*)-9-Benzyl-6-(2-(thiophen-2-ylmethylene)hydrazinyl)-9*H*-purine (1aj)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1aj** (548.4 mg, 82%) as a yellow solid. **M.P.:** 201.7 - 202.8 ℃. **^{1}H NMR (600 MHz, DMSO-*d*$_6$):** δ 11.73 (s, 1 H), 8.59 (s, 1 H), 8.43 (s, 1 H), 8.41 (s, 1 H), 7.60 -7.59 (m, 1 H), 7.36 (dd, *J* = 3.6, 1.2 Hz, 1 H), 7.35 -7.32 (m, 4 H), 7.29 -7.26 (m, 1 H), 7.10 (dd, *J* = 5.4, 3.6 Hz, 1 H), 5.44 (s, 2 H). **^{13}C NMR (150 MHz, DMSO-*d*$_6$):** δ 152.5, 151.1, 150.6, 142.2, 139.8, 136.9, 129.4, 128.7, 128.0, 127.8, 127.7, 118.2, 46.3. **ESI-HRMS**: m/z calcd for $C_{17}H_{15}N_6S$ $[M + H]^+$: 335.1073; found: 335.1074.

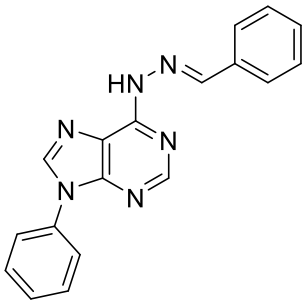
(*E*)-9-Benzyl-6-(2-(pyridin-3-ylmethylene)hydrazinyl)-9*H*-purine (1ak)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 60/1-30/1) yielded **1ak** (513.8 mg, 78%) as a yellow solid. **M.P.:** 233.4 - 236.7 ℃. **^{1}H NMR (600 MHz, CDCl$_3$):** δ 10.90 (s, 1 H), 8.64 -8.59 (m, 3 H), 8.04 (s, 1 H), 7.91 (s, 1 H), 7.57 (s, 2 H), 7.34 -7.30 (m, 5 H), 5.41 (s, 2 H). **^{13}C**

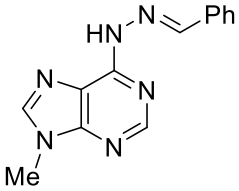
NMR (150 MHz, $CDCl_3$): δ 153.0, 151.5, 150.1, 142.1, 141.7, 141.4, 135.2, 129.1, 128.6, 127.8, 121.0, 119.2, 47.3. **ESI-HRMS**: m/z calcd for $C_{18}H_{16}N_7$ $[M + H]^+$: 330.1462; found: 330.1459.

(*E*)-6-(2-Benzylidenehydrazinyl)-9-phenyl-9*H*-purine (1al)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1al** (503.0 mg, 80%) as a white solid. **M.P.:** 277.5 - 280.3 ℃. **^{1}H NMR (600 MHz, DMSO-*d*$_6$):** δ 11.85 (s, 1 H), 8.46 (s, 1 H), 8.39 (s, 1 H), 7.92 (d, *J* = 7.8 Hz, 2 H), 7.78 (d, *J* = 7.8 Hz, 2 H), 7.61 (t, *J* = 7.8 Hz, 2 H), 7.48 -7.44 (m, 3 H), 7.40 (t, *J* = 7.2 Hz, 1 H). **^{13}C NMR (150 MHz, DMSO-*d*$_6$):** δ 152.9, 144.4, 141.2, 134.9, 129.5, 128.8, 127.7, 126.7, 123.3, 119.0. **ESI-HRMS**: m/z calcd for $C_{18}H_{15}N_6$ $[M + H]^+$: 315.1353; found: 315.1353.

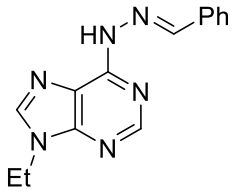
(*E*)-6-(2-Benzylidenehydrazinyl)-9-methyl-9*H*-purine (1am)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1am** (418.8 mg, 83%) as a white solid. **M.P.:** 205.1 - 209.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.42 (s, 1 H), 7.90 (s, 1 H), 7.75 – 7.71 (m, 2 H), 7.46 – 7.41 (m, 2 H), 7.41 – 7.37 (m, 1 H), 7.11 (d, *J* = 3.2 Hz, 1

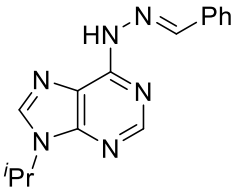
H), 7.04 (d, *J* = 3.6 Hz, 1 H), 3.86 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 154.8, 151.0, 142.4, 139.9, 134.6, 129.7, 129.0, 127.0, 126.6, 103.2, 102.5, 31.4, 30.0. **ESI-HRMS**: m/z calcd for $C_{13}H_{13}N_6$ $[M + H]^+$: 253.1196; found: 253.1196.

(*E*)-6-(2-Benzylidenehydrazinyl)-9-ethyl-9*H*-purine (1an)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1an** (410.1 mg, 77%) as a white solid. **M.P.:** 167.8 - 170.3 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 9.81 (s, 1 H), 8.59 (s, 1 H), 8.06 (s, 1 H), 7.88 (s, 1 H), 7.77 (d, *J* = 7.2 Hz, 2 H), 7.39 – 7.33 (m, 3 H), 4.27 (q, *J* = 7.2 Hz, 2 H), 1.53 (t, *J* = 7.2 Hz, 3 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 153.3, 151.5, 150.7, 145.4, 140.8, 133.9, 130.1, 128.7, 127.5, 119.1, 36.0, 15.6. **ESI-HRMS**: m/z calcd for $C_{14}H_{15}N_6$ $[M + H]^+$: 267.1353; found: 267.1353.

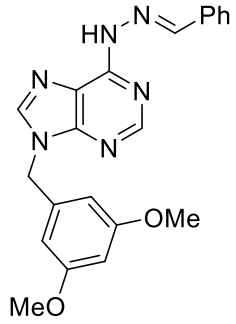
(*E*)-6-(2-Benzylidenehydrazinyl)-9-isopropyl-9*H*-purine (1ao)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ao** (431.7 mg, 77%) as a yellow solid. **M.P.:** 163.3 - 164.2 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.62 (s, 1 H), 8.59 (s, 1 H), 8.06 (s, 1 H), 7.94 (s, 1 H), 7.81 – 7.77 (m, 2 H), 7.41 – 7.35 (m, 3 H), 4.93 – 4.84 (m, 1 H), 1.62 (d, *J* = 6.8 Hz, 6 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.1, 151.4, 145.3,

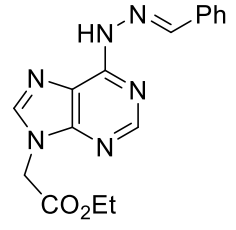
138.9, 133.9, 130.1, 128.7, 127.6, 119.4, 47.3, 22.8. **ESI-HRMS**: m/z calcd for $C_{15}H_{17}N_6$ $[M + H]^+$: 281.1509; found: 281.1502.

(*E*)-6-(2-Benzylidenehydrazinyl)-9-(3,5-dimethoxybenzyl)-9*H*-purine (1ap)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ap** (621.5 mg, 80%) as a white solid. **M.P.:** 182.1 - 183.4 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.74 (s, 1 H), 8.44 (s, 1 H), 8.41 (s, 1 H), 8.34 (s, 1 H), 7.77 – 7.73 (m, 2 H), 7.46 – 7.38 (m, 3 H), 6.49 (d, *J* = 2.0 Hz, 2 H), 6.43 (t, *J* = 2.4 Hz, 1 H), 5.36 (s, 2 H), 3.70 (s, 6 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 160.7, 152.4, 151.8, 150.9, 144.1, 142.4, 139.1, 134.9, 129.3, 128.7, 126.6, 118.3, 105.7, 99.2, 55.2, 46.3. **ESI-HRMS**: m/z calcd for $C_{21}H_{21}N_6O_2$ $[M + H]^+$: 389.1721; found: 389.1721.

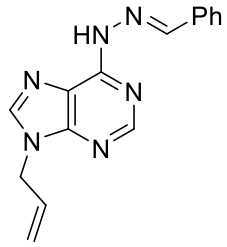
Ethyl (*E*)-2-(6-(2-benzylidenehydrazinyl)-9*H*-purin-9-yl)acetate (1aq)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1aq** (499.5 mg, 77%) as a white solid. **M.P.:** 253.4 - 256.1 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.76 (s, 1 H), 8.38 (s, 1

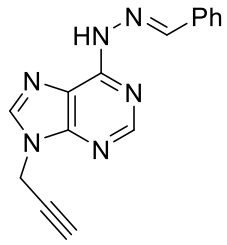
H), 8.35 (s, 1 H), 8.30 (s, 1 H), 7.78 -7.75 (m, 2 H), 7.48 -7.43 (m, 2 H), 7.41 -7.37 (m, 1 H), 5.15 (s, 2 H), 4.18 (q, *J* = 6.8 Hz, 2 H), 1.22 (t, *J* = 7.2 Hz, 3 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 167.9, 152.4, 144.2, 142.8, 134.9, 129.3, 128.8, 126.7, 118.0, 61.4, 44.0, 14.0. **ESI-HRMS**: m/z calcd for $C_{16}H_{17}N_6O_2$ $[M + H]^+$: 325.1408; found: 325.1408.

(*E*)-9-Allyl-6-(2-benzylidenehydrazinyl)-9*H*-purine (1ar)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ar** (439.7 mg, 79%) as a white solid. **M.P.:** 109.9 - 111.8 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 10.14 (s, 1 H), 8.58 (s, 1 H), 8.06 (s, 1 H), 7.86 (s, 1 H), 7.74 -7.72 (m, 2 H), 7.35 -7.29 (m, 3 H), 6.04 -5.97 (m, 1 H), 5.29 -5.26 (m, 1 H), 5.19 -5.15 (m, 1 H), 4.81 -4.80 (m, 2 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 153.4, 151.6, 145.6, 141.1, 133.9, 131.7, 130.0, 128.6, 127.5, 119.2, 45.8. **ESI-HRMS**: m/z calcd for $C_{15}H_{15}N_6$ $[M + H]^+$: 279.1353; found: 279.1353.

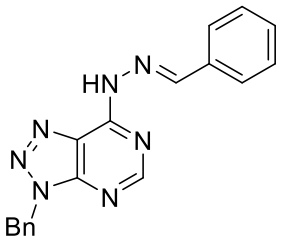
(*E*)-6-(2-Benzylidenehydrazinyl)-9-(prop-2-yn-1-yl)-9*H*-purine (1as)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1as** (414.5 mg, 75%) as a light red

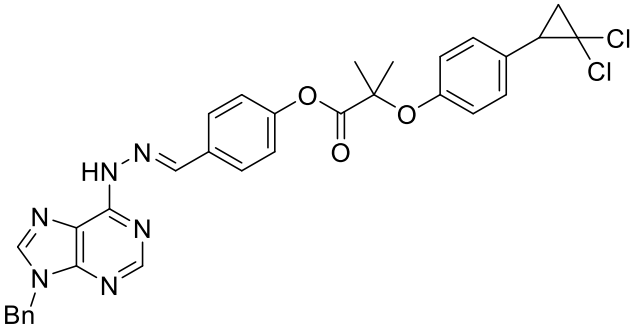
solid. **M.P.:** 215.3 - 218.6 ℃. **^{1}H NMR (600 MHz, DMSO-*d*$_6$):** δ 11.77 (s, 1 H), 8.44 (s, 1 H), 8.40 (s, 1 H), 8.36 (s, 1 H), 7.76 (d, *J* = 7.2 Hz, 2 H), 7.43 (t, *J* = 7.8 Hz, 2 H), 7.39 -7.35 (m, 1 H), 5.12 (d, *J* = 3.0 Hz, 2 H), 3.51 (t, *J* = 2.4 Hz, 1 H). **^{13}C NMR (150 MHz, DMSO-*d*$_6$):** δ 152.4, 151.8, 150.5, 144.3, 141.6, 134.9, 129.3, 128.7, 126.7, 118.3, 78.2, 76.1, 32.5. **ESI-HRMS**: m/z calcd for $C_{15}H_{13}N_6$ $[M + H]^+$: 277.1196; found: 277.1196.

(*E*)-3-Benzyl-7-(2-(2-phenylethylidene)hydrazinyl)-3*H*-[1,2,3]triazolo[4,5-d]pyrimidine (1at)



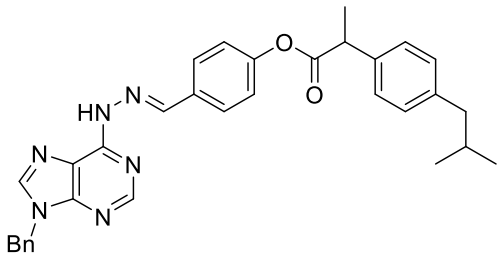
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1at** (329.4 mg, 63%) as a white solid. **M.P.:** 198.6 - 201.2 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 12.55 (s, 1 H), 8.52 (s, 1 H), 8.33 (s, 1 H), 7.99 – 7.79 (m, 2 H), 7.48 – 7.33 (m, 8 H), 5.83 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 135.8, 134.4, 130.0, 128.84, 128.77, 128.1, 127.8, 127.1, 49.5. **ESI-HRMS**: m/z calcd for $C_{18}H_{16}N_7$ $[M + H]^+$: 330.1462; found: 330.1455.

(*E*)-4-((2-(9-Benzyl-9*H*-purin-6-yl)hydrazono)methyl)phenyl 2-(4-(2,2-Dichlorocyclopropyl)phenoxy)-2-methylpropanoate (1au)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1au** (800.2 mg, 65%) as a yellow solid. **M.P.:** 102.3 - 105.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.61 (s, 1 H), 8.00 (s, 1 H), 7.82 (s, 1 H), 7.74 (dd, *J* = 6.4, 2.0 Hz, 2 H), 7.36 – 7.26 (m, 6 H), 7.16 (dd, *J* = 6.4, 2.0 Hz, 2 H), 6.99 (dd, *J* = 6.8, 2.0 Hz, 2 H), 6.93 (dt, *J* = 10.0, 3.2 Hz, 2 H), 5.38 (s, 2 H), 2.85 (dd, *J* = 10.4, 8.0 Hz, 1 H), 1.94 (dd, *J* = 10.4, 7.2 Hz, 1 H), 1.80 (d, *J* = 8.0 Hz, 1 H), 1.75 (s, 6 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 172.7, 155.0, 153.5, 151.8, 151.5, 144.2, 141.3, 135.4, 132.0, 129.9, 129.2, 128.64, 128.61, 127.9, 127.8, 121.7, 119.0, 118.7, 79.4, 61.0, 47.4, 34.9, 29.8, 25.9, 25.6. **ESI-HRMS**: m/z calcd for $C_{32}H_{29}Cl_2N_6O_3$ $[M + H]^+$: 615.1673; found: 615.1663.

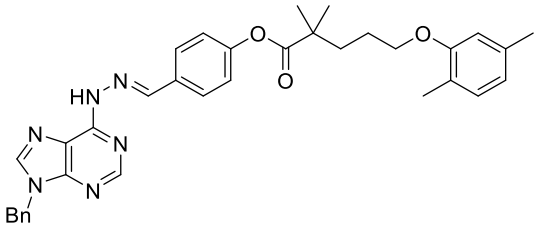
(*E*)-4-((2-(9-Benzyl-9*H*-purin-6-yl)hydrazono)methyl)phenyl 2-(4-isobutylphenyl) propanoate (1av)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1av** (767.0 mg, 72%) as a yellow solid. **M.P.:** 118.3 - 121.1 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 10.82 (s, 1 H), 8.59 (s, 1 H),

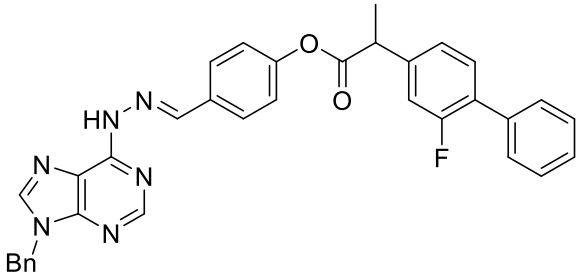
7.93 (s, 1 H), 7.77 (s, 1 H), 7.41 (s, 1 H), 7.34 (d, *J* = 7.6 Hz, 1 H), 7.28 (s, 1 H), 7.24 – 7.06 (m, 9 H), 6.88 (dd, *J* = 8.4, 2.4 Hz, 1 H), 5.24 (s, 2 H), 3.87 (q, *J* = 7.2 Hz, 1 H), 2.41 (d, *J* = 7.2 Hz, 2 H) 1.87 – 1.57 (m, 1 H), 1.54 (d, *J* = 7.2 Hz, 3 H), 0.86 (d, *J* = 6.8 Hz, 6 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 172.9, 153.1, 151.3, 150.9, 150.6, 144.1, 141.2, 140.6, 136.9, 135.4, 135.3, 129.3, 129.1, 128.8, 128.1, 127.6, 127.1, 124.8, 122.6, 119.5, 118.6, 46.9, 45.0, 44.8, 29.9, 22.2, 18.4. **ESI-HRMS**: m/z calcd for $C_{32}H_{33}N_6O_2$ $[M + H]^+$: 533.2660; found: 533.2662.

(*E*)-4-((2-(9-Benzyl-9*H*-purin-6-yl)hydrazono)methyl)phenyl 5-(2,5-dimethyl phenoxy)-2,2-dimethylpentanoate (1aw)



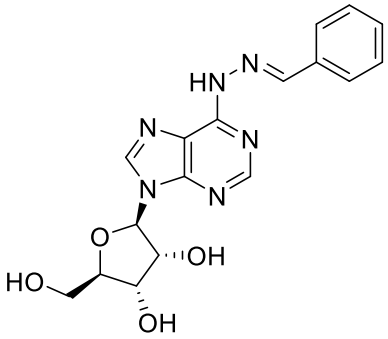
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1aw** (807.4 mg, 70%) as a yellow solid. **M.P.:** 134.5 - 138.7 °C. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 10.07 (s, 1 H), 8.61 (s, 1 H), 7.53 (s, 1 H), 7.78 (s, 1 H), 7.51 – 7.49 (m, 2 H), 7.32 – 7.27 (m, 4 H), 7.23 (d, *J* = 6.0 Hz, 2 H), 7.00 (dd, *J* = 7.2, 1.8 Hz, 1 H), 6.97 (d, *J* = 7.8 Hz, 1 H), 6.63 (d, *J* = 7.8 Hz, 1 H), 6.60 (s, 1 H), 5.32 (s, 2 H), 3.96 (s, 2 H), 2.63 (s, 3 H), 2.15 (s, 3 H), 1.87 (s, 4 H), 1.35 (s, 6 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 176.3, 156.9, 153.5, 151.4, 144.4, 141.3, 136.5, 135.5, 135.4, 130.4, 129.6, 129.1, 128.5, 127.9, 125.1, 123.6, 123.2, 120.8, 120.0, 112.0, 67.8, 47.3, 42.5, 37.1, 25.3, 25.2, 21.4, 15.9. **ESI-HRMS**: m/z calcd for $C_{34}H_{37}N_6O_3$ $[M + H]^+$: 577.2922; found: 577.2918.

(*E*)-4-((2-(9-Benzyl-9*H*-purin-6-yl)hydrazono)methyl)phenyl 2-(2-fluoro-[1,1'-biphenyl]-4-yl)propanoate (1ax)



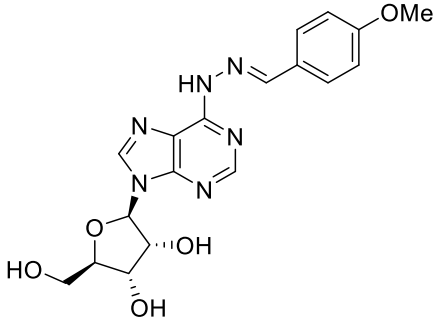
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1ax** (821.7 mg, 72%) as a yellow solid. **M.P.:** 107.9 - 111.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.36 (s, 1 H), 8.64 (s, 1 H), 8.02 (s, 1 H), 7.81 (t, *J* = 6.8 Hz, 3 H), 7.58 – 7.54 (m, 2 H), 7.48 – 7.43 (m, 3 H), 7.40 – 7.27 (m, 6 H), 7.25 – 7.20 (m, 2 H), 7.10 (d, *J* = 8.8 Hz ,2 H), 5.41 (s, 2 H), 4.01 (q, *J* = 7.2 Hz ,1 H), 1.66 (d, *J* = 7.2 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 172.3, 161.5, 153.7, 152.1, 144.2, 141.3, 141.2, 135.5 (d, $J_{C\text{-}F}$ = 5.0 Hz), 131.7, 131.2 (d, $J_{C\text{-}F}$ = 4.0 Hz), 129.3, 129.1 (d, $J_{C\text{-}F}$ = 3.0 Hz), 128.7, 128.6, 128.0, 127.9, 123.7 (d, $J_{C\text{-}F}$ = 3.0 Hz), 121.8, 115.6, 115.4, 47.5, 45.3, 18.6. **^{19}F NMR (376 MHz, $CDCl_3$):** δ -117.12. **ESI-HRMS**: m/z calcd for $C_{34}H_{28}FN_6O_2$ $[M + H]^+$: 571.2252; found: 571.2243.

(2*R*,3*R*,4*S*,5*R*)-2-(6-(2-((*E*)-Benzylidene)hydrazinyl)-9*H*-purin-9-yl)-5-(hydroxymethyl)tetrahydrofuran-3,4-diol (1ay)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 50/1-10/1) yielded **1ay** (592.6 mg, 80%) as a white solid. **M.P.:** 210.1 - 213.3 ℃. **^{1}H NMR (600 MHz, DMSO-*d*$_6$):** δ 8.87 (s, 1 H), 8.64 (s, 1 H), 8.59 (s, 1 H), 8.00 – 7.98 (m, 2 H), 7.48 – 7.45 (m, 3 H), 6.03 (d, *J* = 5.4 Hz, 1 H), 4.57 (t, *J* = 4.8 Hz, 1 H), 4.22 (t, *J* = 4.2 Hz, 1 H), 4.03 (q, *J* = 4.2 Hz, 1 H), 3.71 (dd, *J* = 12.0, 3.6 Hz, 1 H), 3.61 (dd, *J* = 12.0, 4.2 Hz, 1 H). **^{13}C NMR (150 MHz, DMSO-*d*$_6$):** δ 149.4, 142.9, 133.3, 129.0, 128.8, 128.4, 128.2, 117.6, 88.0, 86.0, 74.5, 70.2, 61.1. **ESI-HRMS**: m/z calcd for $C_{17}H_{19}N_6O_4$ $[M + H]^+$: 371.1462; found: 371.1468.

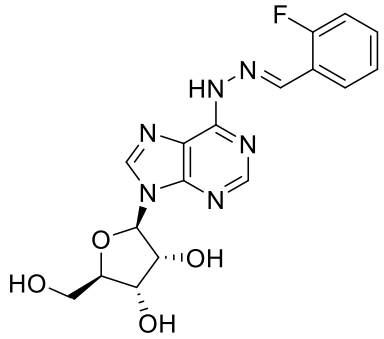
(2*R*,3*S*,4*R*,5*R*)-2-(Hydroxymethyl)-5-(6-(2-((*E*)-4-methoxybenzylidene)hydrazinyl)-9*H*-purin-9-yl)tetrahydrofuran-3,4-diol (1aaa)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 50/1-10/1) yielded **1aaa** (624.6 mg, 78%) as a white solid. **M.P.:** 162.6 - 164.0 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 8.57 (s, 1 H), 8.40 (s, 1 H),

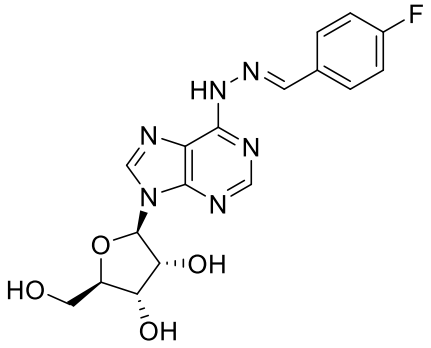
8.34 (s, 1 H), 7.72 (d, *J* = 9.2 Hz, 2 H), 7.02 (d, *J* = 8.8 Hz, 2 H), 5.97 (d, *J* = 6.0 Hz, 1 H), 5.53 (s, 1 H), 5.26 (s, 1 H), 4.62 (t, *J* = 5.6 Hz, 1 H), 4.18 (t, *J* = 4.0 Hz, 1 H), 3.99 (q, *J* = 4.0 Hz, 1 H), 3.80 (s, 3 H), 3.70 (dd, *J* = 12.4, 4.0 Hz, 1 H), 3.58 (dd, *J* = 12.0, 3.6 Hz, 1 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 160.6, 151.3, 141.2, 130.0, 128.6, 127.3, 118.7, 114.4, 114.3, 87.8, 85.9, 73.8, 70.5, 61.5, 55.4, 55.3. **ESI-HRMS**: m/z calcd for $C_{18}H_{21}N_6O_5$ $[M + H]^+$: 401.1568; found: 401.1568.

(2R,3R,4S,5R)-2-(6-(2-((*E*)-2-Fluorobenzylidene)hydrazinyl)-9*H*-purin-9-yl)-5-(hydroxymethyl)tetrahydrofuran-3,4-diol (1aab)



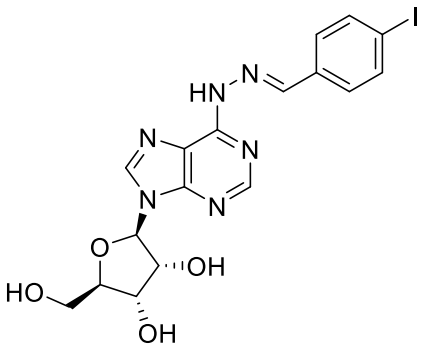
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 50/1-10/1) yielded **1aab** (629.1 mg, 81%) as a white solid. **M.P.:** 215.7 - 217.4 °C. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 8.60 (s, 1 H), 8.59 (s, 1 H), 8.43 (s, 1 H), 8.09 (t, *J* = 7.6 Hz, 1 H), 7.48 – 7.41 (m, 1 H), 7.32 – 7.25 (m, 2 H), 5.99 (d, *J* = 6.0 Hz, 1 H), 5.23 (s, 1 H), 5.25 (s, 1 H), 4.63 (t, *J* = 5.6 Hz, 1 H), 4.20 – 4.17 (m, 1 H), 4.00 (q, *J* = 3.6 Hz, 1 H), 3.71 (dd, *J* = 12.0, 3.6 Hz, 1 H), 3.59 (dd, *J* = 12.0, 3.6 Hz, 1 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 160.5 (d, $J_{C\text{-}F}$ = 247.0 Hz), 152.1, 151.9, 141.4, 131.3, 126.3, 124.9, 122.5 (d, $J_{C\text{-}F}$ = 10.0 Hz), 119.1, 115.9 (d, $J_{C\text{-}F}$ = 22.0 Hz), 87.8, 85.8, 73.7, 70.5, 61.5. **^{19}F NMR (376 MHz, DMSO-*d*$_6$):** δ -121.65. **ESI-HRMS**: m/z calcd for $C_{17}H_{17}FN_6O_4Na$ $[M + Na]^+$: 411.1188; found: 411.1181.

(2*R*,3*R*,4*S*,5*R*)-2-(6-(2-((*E*)-4-Fluorobenzylidene)hydrazinyl)-9*H*-purin-9-yl)-5-(hydroxymethyl)tetrahydrofuran-3,4-diol (1aac)



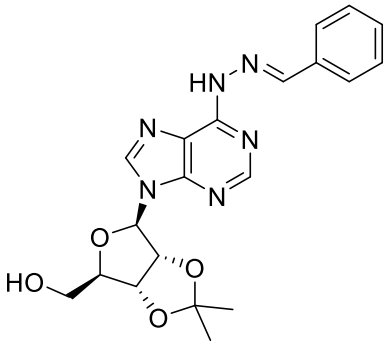
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 50/1-10/1) yielded **1aac** (621.4 mg, 80%) as a white solid. **M.P.:** 213.4 - 216.3 ℃. **^{1}H NMR (600 MHz, DMSO-*d*$_6$):** δ 8.88 (s, 1 H), 8.65 (s, 1 H), 8.58 (s, 1 H), 8.11 (dd, *J* = 8.4, 6.0 Hz, 2 H), 7.92 (dd, *J* = 9.0, 6.0 Hz, 1 H), 7.33 (t, *J* = 8.4 Hz, 2 H), 6.03 (d, *J* = 5.4 Hz, 1 H), 4.56 (t, *J* = 4.8 Hz, 1 H), 4.21 (t, *J* = 4.8 Hz, 1 H), 4.02 (q, *J* = 3.6 Hz, 1 H), 3.71 (dd, *J* = 12.0, 3.6 Hz, 1 H), 3.61 (dd, *J* = 12.6, 4.2 Hz, 1 H). **^{13}C NMR (150 MHz, DMSO-*d*$_6$):** δ 164.7 (d, $J_{C\text{-}F}$ = 20.0 Hz), 163.0 (d, $J_{C\text{-}F}$ = 24.0 Hz), 160.5, 142.9, 130.7 (d, $J_{C\text{-}F}$ = 7.5 Hz), 130.4, 129.9, 117.5, 116.1 (d, $J_{C\text{-}F}$ = 16.5 Hz), 116.0 (d, $J_{C\text{-}F}$ = 18.0 Hz), 88.0, 86.0, 74.5, 70.2, 61.0. **^{19}F NMR (565 MHz, DMSO-*d*$_6$):** δ -108.49. **ESI-HRMS**: m/z calcd for $C_{17}H_{18}FN_6O_4$ $[M + H]^+$: 389.1368; found: 389.1368.

(2*R*,3*S*,4*R*,5*R*)-2-(Hydroxymethyl)-5-(6-(2-((*E*)-4-iodobenzylidene)hydrazinyl)-9*H*-purin-9-yl)tetrahydrofuran-3,4-diol (1aad)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 50/1-10/1) yielded **1aad** (843.7 mg, 85%) as a yellow solid. **M.P.:** 172.5 - 176.8 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 8.58 (s, 1 H), 8.42 (s, 1 H), 8.32 (s, 1 H), 7.82 (d, *J* = 8.4 Hz, 2 H), 7.57 (d, *J* = 8.4 Hz, 2 H), 5.98 (d, *J* = 6.0 Hz, 1 H), 4.61 (t, *J* = 5.2 Hz, 1 H), 4.18 (dd, *J* = 4.8, 4.0 Hz, 1 H), 3.99 (q, *J* = 3.6 Hz, 1 H), 3.70 (dd, *J* = 12.4, 4.0 Hz, 1 H), 3.58 (dd, *J* = 12.4, 4.0 Hz, 1 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 161.0, 151.5, 141.4, 137.9, 137.6, 134.4, 130.1, 128.7, 118.9, 96.1, 87.8, 85.8, 73.7, 70.5, 61.5. **ESI-HRMS**: m/z calcd for $C_{17}H_{18}IN_6O_4$ $[M + H]^+$: 497.0429; found: 497.0422.

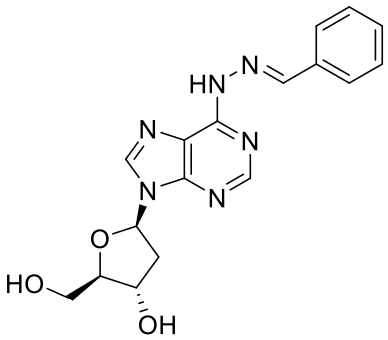
((3a*R*,4*R*,6*R*,6a*R*)-6-(6-(2-((*E*)-Benzylidene)hydrazinyl)-9*H*-purin-9-yl)-2,2-dimethyltetrahydrofuro[3,4-d][1,3]dioxol-4-yl)methanol (1aaf)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 50/1-10/1) yielded **1aaf** (656.7 mg, 80%) as a white solid.

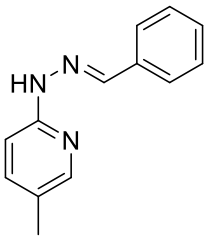
M.P.: 190.2 - 193.8 ℃. **^{1}H NMR (400 MHz, CD_3OD):** δ 8.73 (s, 1 H), 8.60 (s, 1 H), 8.51 (s, 1 H), 8.00 – 7.96 (m, 2 H), 7.56 – 7.49 (m, 3 H), 6.35 – 6.33 (m, 1 H), 5.37 – 5.32 (m, 1 H), 5.19 – 5.05 (m, 1 H), 4.48 – 4.43 (m, 1 H), 3.84 – 3.74 (m, 2 H), 1.64 (s, 3 H), 1.42 (s, 3 H). **^{13}C NMR (100 MHz, CD_3OD):** δ 153.9, 150.6, 145.0, 134.3, 132.8, 130.1, 129.5, 115.3, 93.0, 89.2, 86.3, 83.0, 63.2, 27.5, 25.5. **ESI-HRMS**: m/z calcd for $C_{20}H_{22}N_6O_4Na$ $[M + Na]^+$: 433.1595; found: 433.1589.

(2*R*,3*S*,5*R*)-5-(6-((*Z*)-Benzyldiazenyl)-9*H*-purin-9-yl)-2-(hydroxymethyl)tetrahydrofuran-3-ol (1aag)



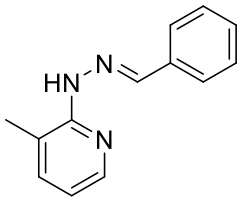
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 50/1-10/1) yielded **1aag** (581.2 mg, 82%) as a yellow solid. **M.P.:** 187.4 - 189.5 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 11.78 (s, 1 H), 8.53 (s, 1 H), 8.38 (s, 1 H), 8.36 (s, 1 H), 7.76 (d, *J* = 7.2 Hz, 2 H), 7.48 -7.36 (m, 3 H), 6.42 (q, *J* = 6.4 Hz, 1 H), 5.35 (d, *J* = 4.0 Hz, 1 H), 5.15 (s, 1 H), 4.46 - 4.41 (m, 1 H), 3.90 (dd, *J* = 6.8, 4.0 Hz, 1 H), 3.64 (dd, *J* = 11.6, 4.4 Hz, 1 H), 3.54 (dd, *J* = 12.0, 4.8 Hz, 1 H), 2.78 - 2.71 (m, 1 H), 2.35 - 2.29 (m, 1 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 140.8, 134.9, 129.4, 128.8, 126.7, 118.9, 88.0, 83.8, 70.9, 70.8, 61.8, 54.9. **ESI-HRMS**: m/z calcd for $C_{17}H_{18}N_6NaO_3$ $[M + Na]^+$: 377.1333; found: 377.1333.

(*E*)-2-(2-Benzylidenehydrazinyl)-5-methylpyridine (1aah)



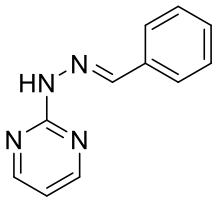
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-70/1) yielded **1aah** (295.8 mg, 70%) as a light brown solid. **M.P.:** 163.1 - 166.2 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 10.74 (s, 1 H), 7.99 (s, 1 H), 7.94 (s, 1 H), 7.65 -7.63 (m, 2 H), 7.47 (dd, *J* = 8.4, 2.0 Hz, 1 H), 7.39 (t, *J* = 7.2 Hz, 2 H), 7.32 -7.28 (m, 1 H), 7.18 (d, *J* = 8.4 Hz, 1 H), 2.18 (s, 3 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 155.3, 147.3, 138.6, 138.0, 135.6, 128.7, 128.3, 125.8, 123.4, 106.0, 17.1. **ESI-HRMS**: m/z calcd for $C_{13}H_{14}N_3$ $[M + H]^+$: 212.1182; found: 212.1180.

(*E*)-2-(2-Benzylidenehydrazinyl)-3-methylpyridine (1aai)



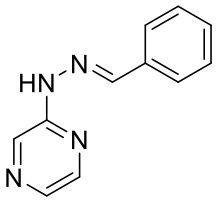
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-70/1) yielded **1aai** (316.9 mg, 75%) as a yellow solid. **M.P.:** 115.6 - 118.3 ℃. **^{1}H NMR (600 MHz, CD$_3$OD):** δ 8.13 (s, 1 H), 7.99 (d, *J* = 3.6 Hz, 1 H), 7.80 -7.78 (m, 2 H), 7.40 -7.34 (m, 3 H), 7.31 -7.27 (m, 1 H), 6.71 (dd, ***J*** = 7.2, 5.4 Hz, 1 H), 2.23 (s, 3 H). **^{13}C NMR (150 MHz, CD$_3$OD):** δ 154.8, 145.4, 143.7, 139.9, 137.0, 130.0, 129.5, 127.9, 118.8, 116.1, 17.2. **ESI-HRMS**: m/z calcd for $C_{13}H_{14}N_3$ $[M + H]^+$: 212.1182; found: 212.1183.

(*E*)-2-(2-Benzylidenehydrazinyl)pyrimidine (1aaj)



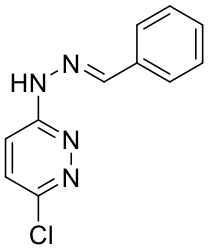
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1aaj** (293.4 mg, 74%) as a white solid. **M.P.:** 177.8 - 180.3 ℃. **H NMR (600 MHz, CD_3OD):** δ 8.42 (d, *J* = 4.8 Hz, 2 H), 8.05 (s, 1 H), 7.79 -7.77 (m, 2 H), 7.38 -7.35 (m, 2 H), 7.34 -7.30 (m, 1 H), 6.81 (t, *J* = 4.8 Hz, 1 H). **^{13}C NMR (150 MHz, CD_3OD):** δ 161.2, 159.6, 144.7, 136.4, 130.4, 129.6, 128.1, 114.0. **ESI-HRMS**: m/z calcd for $C_{11}H_{10}N_4Na$ $[M + Na]^+$: 221.0798; found: 221.0796.

(*E*)-2-(2-Benzylidenehydrazinyl)pyrazine (1aak)



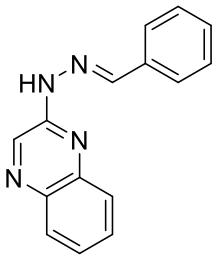
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-80/1) yielded **1aak** (285.4 mg, 72%) as a white solid. **M.P.:** 271.3 - 275.4 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 8.80 (s, 1 H), 8.34 (s, 1 H), 8.08 – 8.04 (m, 2 H), 7.79 (s, 1 H), 7.70 (d, *J* = 7.2 Hz, 2 H), 7.42 (t, *J* = 7.8 Hz, 2 H), 7.39 – 7.35 (m, 1 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 152.5, 141.7, 141.2, 136.4, 134.4, 132.2, 129.7, 128.9, 126.9. **ESI-HRMS**: m/z calcd for $C_{11}H_{11}N_4$ $[M + H]^+$: 199.0978; found: 199.0981.

(*E*)-3-(2-Benzylidenehydrazinyl)-6-chloropyridazine (1aal)



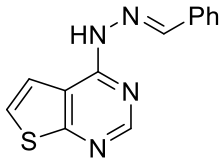
Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1aal** (329.5 mg, 71%) as a yellow solid. **M.P.:** 257.2 - 260.2 ℃. **^{1}H NMR (600 MHz, DMSO-*d*$_6$):** δ 11.71 (s, 1 H), 8.13 (s, 1 H), 7.70 (d, *J* = 10.8 Hz, 2 H), 7.69 -7.64 (m, 2 H), 7.42 (t, *J* = 7.2 Hz, 2 H), 7.39 -7.35 (m, 1 H). **^{13}C NMR (150 MHz, DMSO-*d*$_6$):** δ 158.9, 147.4, 141.9, 134.7, 130.0, 129.2, 128.8, 126.4, 115.9. **ESI-HRMS**: m/z calcd for $C_{11}H_{10}ClN_4$ $[M + H]^+$: 233.0589; found: 233.0582.

(*E*)-2-(2-Benzylidenehydrazinyl)quinoxaline (1aan)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1aan** (392.3 mg, 79%) as a yellow solid. **M.P.:** 194.6 - 197.1 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 11.71 (s, 1 H), 9.11 (s, 1 H), 8.14 (s, 1 H), 7.91 (d, *J* = 7.6 Hz, 1 H), 7.75 (d, *J* = 7.2 Hz, 2 H), 7.70 -7.62 (m, 2 H), 7.49 -7.34 (m, 4 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 150.3, 141.8, 141.0, 137.9, 136.4, 134.7, 130.3, 129.2, 128.8, 126.5, 126.1, 125.1. **ESI-HRMS**: m/z calcd for $C_{15}H_{13}N_4$ $[M + H]^+$: 249.1135; found: 249.1128.

(*E*)-4-(2-Benzylidenehydrazinyl)thieno[2,3-d]pyrimidine (1aao)



Follow the general procedure, product purification by column chromatography on silica gel (DCM/ MeOH = 100/1-50/1) yielded **1aao** (432.3 mg, 85%) as a light yellow solid. **M.P.:** 215.2 - 217.0 ℃. **^{1}H NMR (600 MHz, DMSO-d_6):** δ 11.88 (s, 1 H), 8.49 (s, 1 H), 8.27 (s, 1 H), 8.10 (s, 1 H), 7.75 – 7.70 (m, 3 H), 7.48 (t, *J* = 7.2 Hz, 2 H), 7.43 (t, *J* = 7.2 Hz, 1 H). **^{13}C NMR (150 MHz, DMSO-d_6):** δ 168.8, 155.5, 153.0, 144.5, 134.4, 129.7, 129.0, 126.7, 123.5, 122.6, 114.6. **ESI-HRMS**: m/z calcd for $C_{13}H_{11}N_4S$ $[M + H]^+$: 255.0699; found: 255.0697.

3. Graphical Supporting Information for Electrochemical Cycloamination

Figure S1: General equipment for electrolysis.

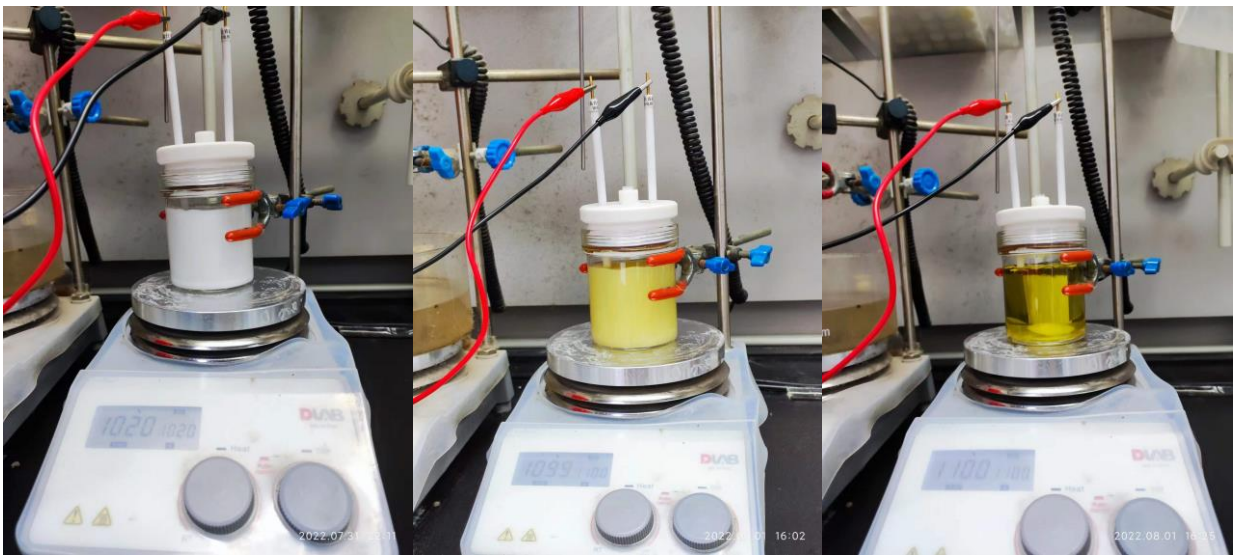


Figure S2: (*Left and Center*): The reaction mixture was subjected to constant current electrolysis (I = 20.0 mA). **(*Right*):** The reaction mixture after 18 h of electrolysis.

4. Optimization of the Reaction Conditions[a]

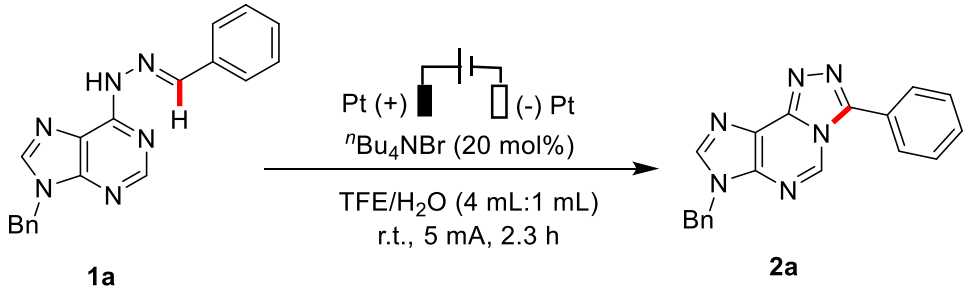


Table S1 Optimization Study

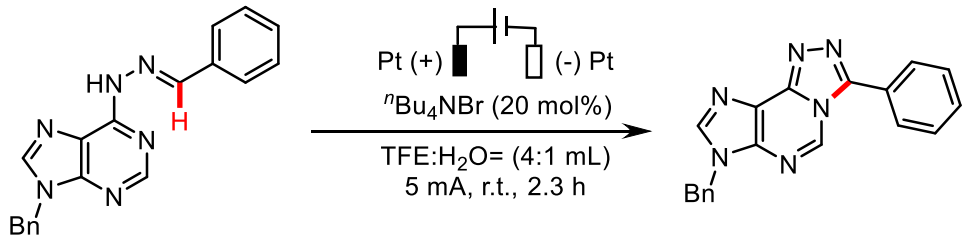
Entry	Variation from standard conditions above[a]	Yield (%)[b]
1	None	98(99)[c]
2	NaBr as electrolyte	86
3	KBr as electrolyte	94
4	HBr as electrolyte	95
5	NH_4Br as electrolyte	97
6	$^{n}Bu_4NI$ as electrolyte	48
7	$^{n}Bu_4NPF_4$ as electrolyte	10
8	$^{n}Bu_4NCl$ as electrolyte	18
9	LiCl as electrolyte	14
10	$LiClO_4$ as electrolyte	8
11	KI as electrolyte	15
12	$^{n}Bu_4NBr$ (5 mol%)(E_{cell}=19.5V→23.3V)	94
13	$^{n}Bu_4NBr$ (10 mol%)(E_{cell} =12.6V→14.0V)	96
14	CH_3CN	74
15	TFE	86
16	H_2O	nr[d]
17	MeOH	12
18	CH_3CN/ H_2O =4/1	70
19	EtOH/ MeOH =4/1	38
20	EtOH/ TFE =4/1	93

21	MeOH/ TFE =4/1	85
22	MeOH/ AcOH =4.8/0.2	82
23	TFE/ H_2O =3/2	95
24	TFE/ H_2O =2.5/2.5	95
25	TFE/ H_2O =2/3	93
26	$^tBuOH/H_2O$=1/1	40
27	$EtOH/H_2O$=4/1	34
28	CH_3CN/MeOH =4/1	51
29	$CH_3CN/AcOH/H_2O$=3.5/0.5/2	90
30	$EtOH/AcOH/H_2O$=4/0.5/0.5	76
31	Graphite felt as the anode	66
32	Graphite rod (6 mm diam.) as the anode	90
33	Reticulated vitreous carbon as the anode	70
34	Pt net as the anode	94
35	Ni foam (1×1 cm^2) as the cathode	97
36	Fe sheet (1×1 cm^2) as the cathode	97
37	Graphite rod (6 mm diam.) as the cathode	96
38	3 mA, 3.9 h	90
39	7 mA, 1.6 h	90
40	9 mA, 1.3 h	90
41	11 mA, 1.1 h	87
42	Under N_2 atmosphere	98[c]

43	Under O_2 atmosphere	94[c]
44	No electrolyte	23
45	In the dark	98
46	No electric current	nr[d]

[a]Standard conditions: substrate **1a** (0.2 mmol), [n]Bu_4NBr (20 mol%) in TFE/H_2O (4/1 mL), two platinum electrodes (1.5 × 1.5 × 0.2 cm^2), undivided cell, r.t., 5 mA(j_{anode} = 0.9 mA cm^{-2}), 2.3 h. [b]Yield determined by ^1H NMR analysis with CH_2Br_2 as an internal standard. [c]Isolated yield. [d]nr: no reaction.

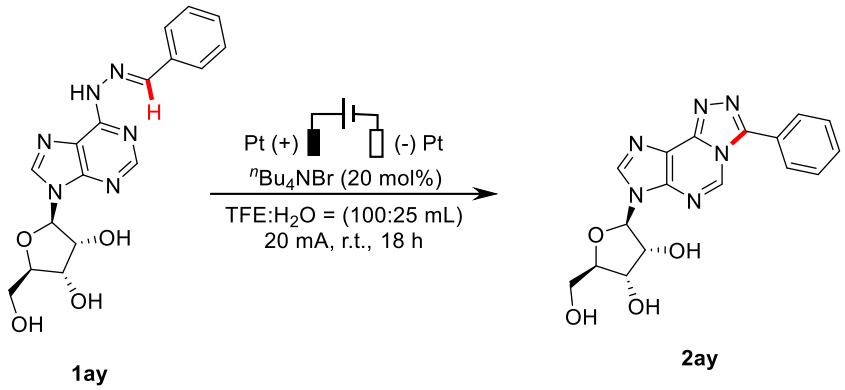
5. General Procedure for Intramolecular C-H Cycloamination



1a **2a**

The electrocatalysis was carried out in an undivided cell equipped with two platinum electrode (1.5 × 1.5 × 0.2 cm^2). The substrates **1a** (0.2 mmol) and [n]Bu_4NBr (12.9 mg, 0.04 mmol, 20 mol%) were dissolved in the mixture solvent TFE/H_2O (4/1 mL). The electrolysis was carried out at room temperature using a constant current of 5.0 mA until complete consumption of the substrate (monitored by TLC or ^1H NMR analysis). After the reaction, the solvent was removed under reduced pressure. The resulting residue was chromatographed through silica gel eluting with DCM/MeOH to afford the corresponding product.

Electrochemistry General Procedure for a Gram-Scale Experiment

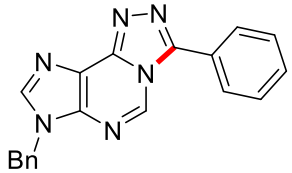


1ay **2ay**

The gram scale reaction was conducted in a 150 mL straight undivided five port electrolytic cell. The substrates **1ay** (1.8520 g, 5.0 mmol) and ^{n}Bu$_4$NBr (322.4 mg, 1.0 mmol, 20 mol%) were dissolved in the mixture solvent TFE/H$_2$O (100/25 mL). The electrolysis was carried out at room temperature using a constant current of 20 mA for 18 hours. After the reaction, the solvent was removed under reduced pressure. The resulting residue was chromatographed through silica gel eluting with DCM/ MeOH (20/1 - 10/1) to afford the corresponding product **2ay** as white solid with 88% yield (1.6748 g).

6. Characterization Data for Electrolysis Products

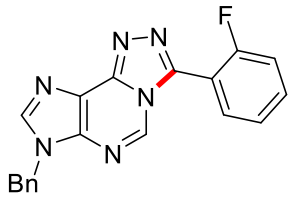
7-Benzyl-3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2a)



Following the general procedure, the electrochemical reaction was carried out with **1a** (65.5 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2a** (64.6 mg, 99%) as a white solid. **M.P.:** 239.2 - 241.4 °C. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 9.22 (s, 1 H),

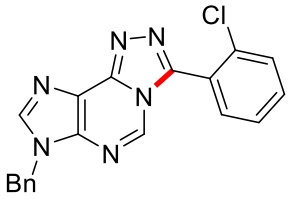
8.59 (s, 1 H), 7.98 – 7.94 (m, 2 H), 7.65 – 7.60 (m, 3 H), 7.38 – 7.26 (m, 5 H), 5.59 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 146.3, 145.7, 142.3, 139.2, 136.6, 135.3, 130.4, 129.2, 128.7, 127.9, 127.5, 126.1, 120.1, 47.2. **ESI-HRMS**: m/z calcd for $C_{19}H_{15}N_6$ $[M + H]^+$: 327.1353; found: 327.1351. Spectral data matched those previously reported[7].

7-Benzyl-3-(2-fluorophenyl)-7*H*-[1,2,4]triazolo[3,4-i]purine (2b)



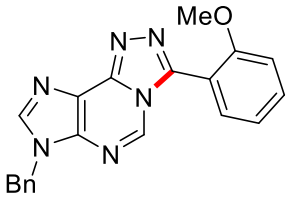
Following the general procedure, the electrochemical reaction was carried out with **1b** (69.3 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2b** (62.0 mg, 90%) as a white solid. **M.P.:** 185.3 - 188.5 ℃. **^{1}H NMR (400 MHz, CDCl$_3$):** δ 8.66 (d, *J* = 3.6 Hz, 1 H), 8.04 (s, 1 H), 7.88 (ddd, *J* = 14.4, 7.2, 1.6 Hz, 1 H), 7.64 – 7.58 (m, 1 H), 7.42 – 7.29 (m, 7 H), 5.51 (s, 2 H). **^{13}C NMR (100 MHz, CDCl$_3$):** δ 159.7 (d, $J_{C\text{-}F}$ = 248.0 Hz), 146.4, 142.3, 141.5, 139.7, 135.2, 133.9 (d, $J_{C\text{-}F}$ = 10.0 Hz), 133.2 (d, $J_{C\text{-}F}$ = 9.0 Hz), 132.6 (d, $J_{C\text{-}F}$ = 2.0 Hz), 129.2, 128.7, 127.9, 125.5 (d, $J_{C\text{-}F}$ = 4.0 Hz), 121.2, 116.5 (d, $J_{C\text{-}F}$ = 20.0 Hz), 114.2 (d, $J_{C\text{-}F}$ = 14.0 Hz), 48.3. **^{19}F NMR (376 MHz, CDCl$_3$):** δ -111.16. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}FN_6$ $[M + H]^+$: 345.1258; found: 345.1256.

7-Benzyl-3-(2-chlorophenyl)-7*H*-[1,2,4]triazolo[3,4-i]purine (2c)



Following the general procedure, the electrochemical reaction was carried out with **1c** (72.6 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2c** (68.6 mg, 95%) as a white solid. **M.P.:**122.7 - 124.8 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 8.91 (s, 1 H), 8.62 (s, 1 H), 7.79 (dd, *J* = 7.6, 1.6 Hz, 1 H), 7.76 (dd, *J* = 8.0, 0.8 Hz, 1 H), 7.70 (td, *J* = 7.6, 1.6 Hz, 1 H), 7.60 (td, *J* = 7.2, 1.2 Hz, 1 H), 7.34 (d, *J* = 4.8 Hz, 4 H), 7.32 – 7.28 (m, 1 H), 5.59 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 145.4, 143.8, 142.5, 139.3, 136.5, 135.3, 133.30, 133.26, 132.7, 130.1, 128.7, 127.9, 127.8, 127.5, 125.1, 120.0, 47.2. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}ClN_6$ $[M + H]^+$: 361.0963; found: 361.0958.

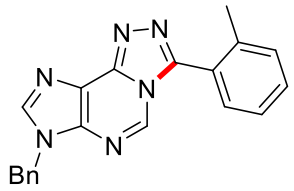
7-Benzyl-3-(2-methoxyphenyl)-7*H*-[1,2,4]triazolo[3,4-i]purine (2d)



Following the general procedure, the electrochemical reaction was carried out with **1d** (71.7 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2d** (56.3 mg, 79%) as a white solid, **M.P.:** 174.9 - 177.3 ℃. **^{1}H NMR (400 MHz, CDCl$_3$):** δ 8.53 (s, 1 H), 8.00 (s, 1 H), 7.76 (dd, *J* = 7.6, 1.6 Hz, 1 H), 7.61 – 7.55 (m, 1 H), 7.40 – 7.28 (m, 5 H), 7.17 (t, *J* = 7.6 Hz, 1 H), 7.11 (d, *J* = 8.4 Hz, 1 H), 5.50 (s, 2 H), 3.84 (s, 3 H). **^{13}C**

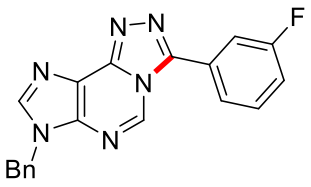
NMR (100 MHz, $CDCl_3$): δ 157.2, 146.2, 145.0, 141.0, 139.6, 135.3, 135.2, 133.0, 132.8, 129.3, 128.8, 127.9, 121.8, 121.4, 115.1, 111.5, 55.9, 48.3. **ESI-HRMS**: m/z calcd for $C_{20}H_{17}N_6O$ $[M + H]^+$: 357.1458; found: 357.1448.

7-Benzyl-3-(o-tolyl)-7*H*-[1,2,4]triazolo[3,4-i]purine (2e)



Following the general procedure, the electrochemical reaction was carried out with **1e** (68.4 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2e** (62.0 mg, 91%) as a white solid. **M.P.:** 165.7 - 168.9 ℃. **^{1}H NMR (400 MHz, CD_3OD):** δ 8.72 (s, 1 H), 8.41 (s, 1 H), 7.60 – 7.48 (m, 3 H), 7.46 – 7.37 (m, 3 H), 7.35 – 7.25 (m, 3 H), 5.59 (s, 2 H), 2.28 (s, 3 H). **^{13}C NMR (100 MHz, CD_3OD):** δ 147.6, 146.8, 143.5, 141.4, 140.1, 137.3, 135.9, 132.4, 132.3, 131.7, 130.0, 129.4, 129.0, 127.6, 125.8, 121.2, 19.8. **ESI-HRMS**: m/z calcd for $C_{20}H_{17}N_6$ $[M + H]^+$: 341.1509; found: 341.1508.

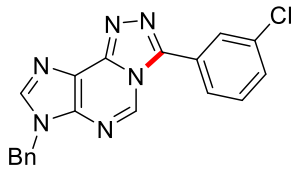
7-Benzyl-3-(3-fluorophenyl)-7*H*-[1,2,4]triazolo[3,4-i]purine (2f)



Following the general procedure, the electrochemical reaction was carried out with **1f** (69.3 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2f** (37.2 mg, 54%) as a white solid. **M.P.:** 244.7 - 244.9 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 9.27 (s, 1 H),

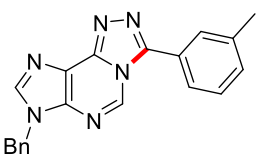
8.60 (s, 1 H), 7.86 – 7.80 (m, 2 H), 7.70 – 7.64 (m, 1 H), 7.46 (td, *J* = 8.0, 2.0 Hz, 1 H), 7.37 – 7.34 (m, 4 H), 7.33 – 7.27 (m, 1 H), 5.59 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 162.2 (d, $J_{C\text{-}F}$ = 243.0 Hz), 145.9, 145.3 (d, $J_{C\text{-}F}$ = 3.0 Hz), 142.4, 139.2, 136.6, 135.4, 131.4 (d, $J_{C\text{-}F}$ = 8.0 Hz), 128.7, 128.1 (d, $J_{C\text{-}F}$ = 9.0 Hz), 128.0, 127.5, 125.0 (d, $J_{C\text{-}F}$ = 3.0 Hz), 120.0, 117.3 (d, $J_{C\text{-}F}$ = 21.0 Hz), 115.6 (d, $J_{C\text{-}F}$ = 24.0 Hz), 47.2. **^{19}F NMR (376 MHz, DMSO-*d*$_6$):** δ -111.58. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}FN_6$ $[M + H]^+$: 345.1258; found: 345.1252.

7-Benzyl-3-(3-chlorophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2g)



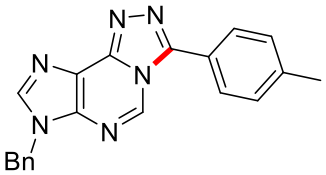
Following the general procedure, the electrochemical reaction was carried out with **1g** (72.6 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2g** (65.7 mg, 91%) as a white solid. **M.P.:** 185.4 - 188.6 ℃. **^{1}H NMR (400 MHz, CDCl$_3$):** δ 8.95 (s, 1 H), 8.02 (s, 1 H), 7.84 (s, 1 H), 7.76 – 7.71 (m, 1 H), 7.55 – 7.50 (m, 2 H), 7.37 – 7.28 (m, 5 H), 5.50 (s, 2 H). **^{13}C NMR (100 MHz, CDCl$_3$):** δ 146.4, 145.2, 141.6, 139.7, 135.7, 135.1, 133.1, 131.0, 130.9, 129.3, 128.8, 128.7, 127.9, 127.7, 126.7, 121.4, 48.4. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}ClN_6$ $[M + H]^+$: 361.0963; found: 361.0954.

7-Benzyl-3-(m-tolyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2h)



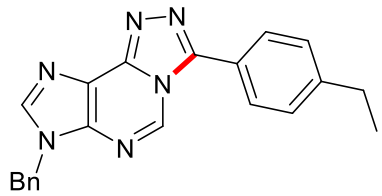
Following the general procedure, the electrochemical reaction was carried out with **1h** (68.4 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2h** (62.6 mg, 92%) as a white solid. **M.P.:** 163.2 - 166.5 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.94 (s, 1 H), 8.00 (s, 1 H), 7.66 (s, 1 H), 7.61 (d, *J* = 7.6 Hz, 1 H), 7.46 (t, *J* = 7.6 Hz, 1 H), 7.40 – 7.28 (m, 6 H), 5.50 (s, 2 H), 2.45 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.7, 146.1, 141.4, 139.6, 139.5, 135.2, 133.5, 131.7, 129.44, 129.41, 129.2, 128.7, 127.8, 125.9, 125.6, 121.4, 48.3, 21.5. **ESI-HRMS**: m/z calcd for $C_{20}H_{16}N_6Na$ $[M + Na]^+$: 363.1329; found: 363.1329.

7-Benzyl-3-(p-tolyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2i)



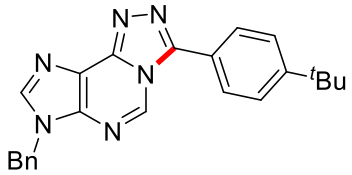
Following the general procedure, the electrochemical reaction was carried out with **1i** (68.4 mg, 0.2 mmol) at room temperature for 2.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2i** (62.6 mg, 92%) as a white solid. **M.P.:** 192.7 - 195.8 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.91 (s, 1 H), 7.99 (s, 1 H), 7.71 (d, *J* = 8.0 Hz, 2 H), 7.37 (d, *J* = 8.0 Hz, 2 H), 7.36 – 7.27 (m, 5 H), 5.49 (s, 2 H), 2.44 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.6, 146.1, 141.3, 141.2, 139.5, 135.2, 133.5, 130.2, 129.2, 128.7, 128.6, 127.8, 123.1, 121.4, 48.3, 21.6. **ESI-HRMS**: m/z calcd for $C_{20}H_{17}N_6$ $[M + H]^+$: 341.1509; found: 341.1508.

7-Benzyl-3-(4-ethylphenyl)-7*H*-[1,2,4]triazolo[3,4-i]purine (2j)



Following the general procedure, the electrochemical reaction was carried out with **1j** (71.3 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2j** (60.2 mg, 85%) as a white solid. **M.P.:** 221.9 - 223.5 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.94 (s, 1 H), 8.00 (s, 1 H), 7.76 (dt, *J* = 8.4, 2.4 Hz, 2 H), 7.42 (dd, *J* = 6.4, 1.6 Hz, 2 H), 7.40 – 7.28 (m, 5 H), 5.49 (s, 2 H), 2.76 (q, *J* = 7.6 Hz, 2 H), 1.30 (t, *J* = 7.6 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 147.6, 146.7, 146.1, 141.3, 139.6, 135.6, 133.6, 129.3, 129.1, 128.8, 127.9, 123.3, 121.5, 47.9, 28.6, 16.3. **ESI-HRMS**: m/z calcd for $C_{21}H_{19}N_6$ $[M + H]^+$: 355.1666; found: 355.1666.

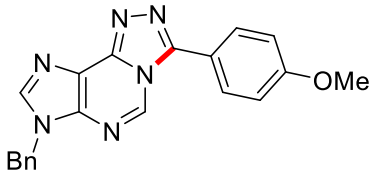
7-Benzyl-3-(4-(*tert*-butyl)phenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2k)



Following the general procedure, the electrochemical reaction was carried out with **1k** (76.9 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2k** (72.7 mg, 95%) as a white solid. **M.P.:** 247.6 - 250.3 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.94 (s, 1 H), 7.99 (s, 1 H), 7.78 (dt, *J* = 8.8, 2.4 Hz, 2 H), 7.60 (dt, *J* = 8.8, 2.4 Hz, 2 H), 7.35 – 7.27 (m, 5 H), 5.49 (s, 2 H), 1.37 (s, 9 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 154.8, 146.6, 146.1, 141.3, 139.5, 135.9, 133.9, 129.2, 128.7, 128.5, 127.9, 126.6, 123.1, 121.4, 48.3,

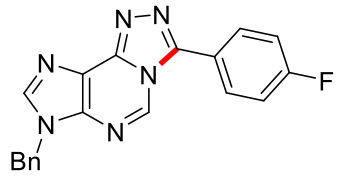
35.1, 31.3. **ESI-HRMS**: m/z calcd for $C_{23}H_{23}N_6$ $[M + H]^+$: 383.1979; found: 383.1980.

7-Benzyl-3-(4-methoxyphenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2l)



Following the general procedure, the electrochemical reaction was carried out with **1l** (71.7 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2l** (63.4 mg, 89%) as a white solid. **M.P.:** 218.2 - 220.3 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 9.16 (s, 1 H), 8.57 (s, 1 H), 7.89 (dt, *J* = 9.6, 2.8 Hz, 2 H), 7.35 (d, *J* = 4.8 Hz, 4 H), 7.33 – 7.26 (m, 1 H), 7.15 (dt, *J* = 10.0, 3.2 Hz, 2 H), 5.58 (s, 2 H), 3.85 (s, 3 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 160.8, 146.2, 145.5, 142.2, 139.1, 136.7, 135.3, 130.3, 128.8, 128.0, 127.5, 120.1, 118.2, 114.7, 55.4, 47.2. **ESI-HRMS**: m/z calcd for $C_{20}H_{17}N_6O$ $[M + H]^+$: 357.1458; found: 357.1449.

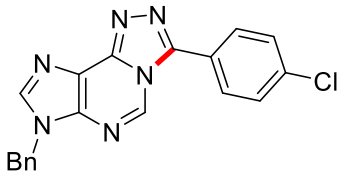
7-Benzyl-3-(4-fluorophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2m)



Following the general procedure, the electrochemical reaction was carried out with **1m** (69.3 mg, 0.2 mmol) at room temperature for 2.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2m** (59.9 mg, 87%) as a white solid. **M.P.:** 160.3 - 164.8 ℃. **^{1}H NMR (400 MHz, CDCl$_3$):** δ 8.90 (s, 1 H),

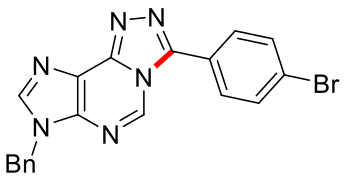
8.02 (s, 1 H), 7.85 (dd, *J* = 8.8, 5.2 Hz, 2 H), 7.38 – 7.25 (m, 7 H), 5.50 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 164.2 (d, $J_{C\text{-}F}$ = 251.0 Hz), 146.3, 145.7, 141.5, 139.6, 135.1, 133.1, 130.9 (d, $J_{C\text{-}F}$ = 9.0 Hz), 129.3, 128.8, 127.9, 122.3, 122.2, 121.4, 117.1, 116.8, 48.4. **^{19}F NMR (376 MHz, $CDCl_3$):** δ -108.29. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}FN_6$ $[M + H]^+$: 345.1258; found: 345.1250.

7-Benzyl-3-(4-chlorophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2n)



Following the general procedure, the electrochemical reaction was carried out with **1n** (72.6 mg, 0.2 mmol) at room temperature for 3.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2n** (56.3 mg, 78%) as a white solid. **M.P.:** 211.3 - 211.8 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 9.23 (s, 1 H), 8.59 (s, 1 H), 7.99 (dt, *J* = 9.2, 2.8 Hz, 2 H), 7.71 – 7.65 (m, 2 H), 7.38 – 7.26 (m, 5 H), 5.59 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 145.8, 145.4, 142.3, 139.2, 136.6, 135.4, 135.2, 130.6, 129.3, 128.7, 128.0, 127.5, 125.0, 120.0, 47.2. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}ClN_6$ $[M + H]^+$: 361.0963; found: 361.0965.

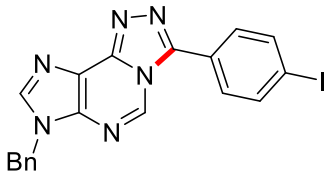
7-Benzyl-3-(4-bromophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2o)



Following the general procedure, the electrochemical reaction was carried out with **1o** (81.5 mg, 0.2 mmol) at room temperature for 4.5 h. Purification by column

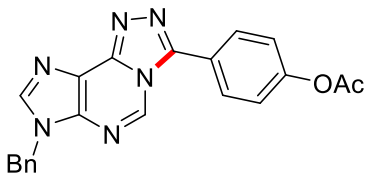
chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2o** (71.3 mg, 88%) as a white solid. **M.P.:** 228.7 - 231.1 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 9.23 (s, 1 H), 8.59 (s, 1 H), 7.94 – 7.90 (m, 2 H), 7.83 – 7.79 (m, 2 H), 7.37 – 7.27 (m, 5 H), 5.59 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 145.9, 145.5, 142.3, 139.2, 136.6, 135.3, 132.2, 130.7, 128.7, 127.9, 127.5, 125.3, 123.9, 120.0, 47.2. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}BrN_6$ $[M + H]^+$: 405.0458; found: 405.0453.

7-Benzyl-3-(4-iodophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2p)



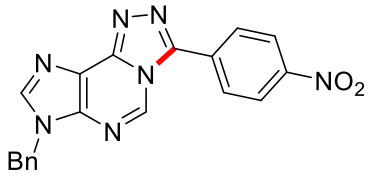
Following the general procedure, the electrochemical reaction was carried out with **1p** (90.9 mg, 0.2 mmol) at room temperature for 4.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2p** (78.7 mg, 87%) as a white solid. **M.P.:** 193.7 - 195.2 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 9.22 (s, 1 H), 8.59 (s, 1 H), 7.98 (d, *J* = 8.4 Hz, 2 H), 7.76 (d, *J* = 8.4 Hz, 2 H), 7.36 – 7.27 (m, 5 H), 5.58 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 145.9, 145.7, 142.4, 139.2, 138.0, 136.6, 135.3, 130.5, 128.8, 128.0, 127.5, 125.6, 120.0, 97.5, 47.2. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}IN_6$ $[M + H]^+$: 453.0319; found: 453.0310.

4-(7-Benzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-3-yl)phenyl acetate (2q)



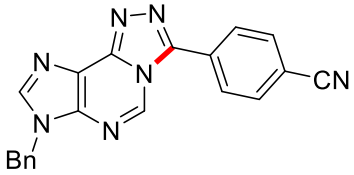
Following the general procedure, the electrochemical reaction was carried out with **1q** (77.3 mg, 0.2 mmol) at room temperature for 3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2q** (53.0 mg, 69%) as a white solid. **M.P.:** 186.0 - 188.3 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 8.93 (s, 1 H), 7.99 (s, 1 H), 7.85 (dt, *J* = 9.6, 3.0 Hz, 2 H), 7.34 – 7.27 (m, 7 H), 5.48 (s, 2 H), 2.32 (s, 3 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 169.6, 152.5, 146.2, 145.8, 141.5, 139.5, 135.2, 133.3, 129.9, 129.2, 128.7, 127.9, 123.6, 122.9, 121.3, 48.3, 20.7. **ESI-HRMS**: m/z calcd for $C_{21}H_{17}N_6O_2$ $[M + H]^+$: 385.1408; found: 385.1405.

7-Benzyl-3-(4-nitrophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2r)



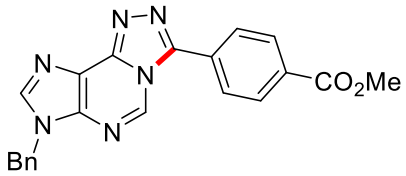
Following the general procedure, the electrochemical reaction was carried out with **1r** (74.6 mg, 0.2 mmol) at room temperature for 3.0 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2r** (27.6 mg, 37%) as a yellow solid. **M.P.:** 136.2 - 140.9 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 9.35 (s, 1 H), 8.63 (s, 1 H), 8.44 (dt, *J* = 9.6, 2.8 Hz, 2 H), 8.29 (dt, *J* = 9.6, 2.4 Hz, 2 H), 7.37 – 7.29 (m, 5 H), 5.61 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 148.2, 146.3, 144.9, 142.6, 139.4, 136.6, 135.5, 132.3, 129.9, 128.8, 128.0, 127.6, 124.2, 120.0, 47.3. **ESI-HRMS**: m/z calcd for $C_{19}H_{14}N_7O_2$ $[M + H]^+$: 372.1203; found: 372.1203.

4-(7-Benzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-3-yl)benzonitrile (2s)



Following the general procedure, the electrochemical reaction was carried out with **1s** (70.7 mg, 0.2 mmol) at room temperature for 2.8 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2s** (64.0 mg, 91%) as a white solid, **M.P.:** 236.6 - 237.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.99 (s, 1 H), 8.04 (dd, *J* = 8.0, 1.2 Hz, 3 H), 7.90 (dd, *J* = 6.8, 2.0 Hz, 2 H), 7.40 – 7.29 (m, 5 H), 5.51 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.8, 144.9, 141.9, 139.8, 135.0, 133.4, 132.7, 130.5, 129.4, 129.1, 129.0, 127.9, 121.6, 118.0, 114.6, 48.5. **ESI-HRMS**: m/z calcd for $C_{20}H_{14}N_7$ $[M + H]^+$: 352.1305; found: 352.1303.

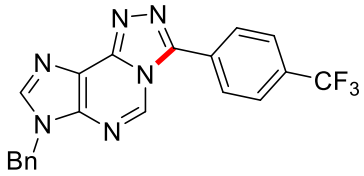
Methyl 4-(7-benzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-3-yl)benzoate (2t)



Following the general procedure, the electrochemical reaction was carried out with **1t** (77.2 mg, 0.2 mmol) at room temperature for 2.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2t** (69.1 mg, 90%) as a white solid. **M.P.:** 219.2 - 221.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.99 (s, 1 H), 8.24 (dd, *J* = 6.8, 2.0 Hz, 2 H), 8.03 (s, 1 H), 7.96 (dd, *J* = 6.8, 1.6 Hz, 2 H), 7.38 – 7.28 (m, 5 H), 5.51 (s, 2 H), 3.97 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 166.2, 146.5, 145.7, 141.7, 139.7, 135.1, 133.1, 132.2, 130.7, 130.2, 129.3, 128.8, 128.6,

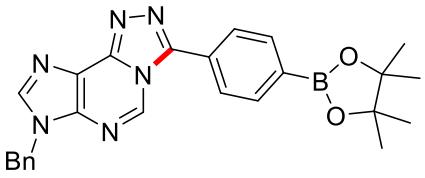
127.9, 121.5, 52.6, 48.5. **ESI-HRMS**: m/z calcd for $C_{21}H_{17}N_6O_2$ $[M + H]^+$: 385.1408; found: 385.1408.

7-Benzyl-3-(4-(trifluoromethyl)phenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2u)



Following the general procedure, the electrochemical reaction was carried out with **1u** (79.3 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2u** (71.8 mg, 91%) as a white solid, **M.P.:** 193.7 - 195.2 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.99 (s, 1 H), 8.00 (d, *J* = 7.6 Hz, 3 H), 7.83 (d, *J* = 8.0 Hz, 2 H), 7.36 – 7.28 (m, 5 H), 5.50 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.5, 145.2, 141.7, 139.7, 135.1, 133.0, 132.8, 132.5, 129.6, 129.2, 129.0, 128.8, 127.9, 126.5 (q, $J_{C\text{-}F}$ = 4.0 Hz), 125.0, 122.3, 121.3, 48.4. **^{19}F NMR (376 MHz, $CDCl_3$):** δ -62.93. **ESI-HRMS**: m/z calcd for $C_{20}H_{14}F_3N_6$ $[M + H]^+$: 395.1227; found: 395.1229.

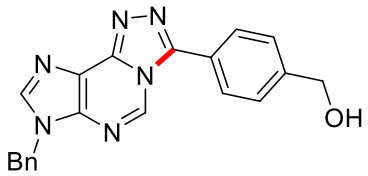
7-Benzyl-3-(4-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)phenyl)-7*H*-[1,2,4]tria zolo[3,4-*i*]purine (2v)



Following the general procedure, the electrochemical reaction was carried out with **1v** (90.9 mg, 0.2 mmol) at room temperature for 2.9 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2v** (57.0 mg, 63%) as a

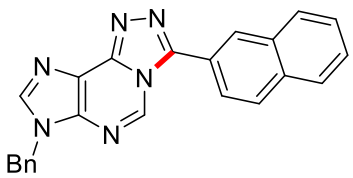
white solid, **M.P.:** 157.9 - 160.1 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.97 (s, 1 H), 8.05 (s, 1 H), 8.03 (d, *J* = 3.6 Hz, 2 H), 7.87 (d, *J* = 8.4 Hz, 2 H), 7.39 – 7.28 (m, 5 H), 5.50 (s, 2 H), 1.39 (s, 12 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.5, 146.2, 141.5, 139.6, 135.7, 135.2, 133.4, 129.2, 128.7, 128.3, 127.80, 127.75, 121.3, 84.4, 75.0, 48.3, 25.0. **^{11}B NMR (193 MHz, $CDCl_3$):** δ 31.01. **ESI-HRMS**: m/z calcd for $C_{25}H_{25}BN_6O_2Na$ [M + Na]$^+$: 475.2024; found: 475.2017.

(4-(7-Benzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-3-yl)phenyl)methanol (2w)



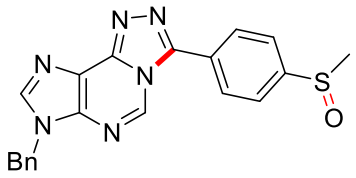
Following the general procedure, the electrochemical reaction was carried out with **1w** (71.7 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2w** (69.1 mg, 97%) as a white solid. **M.P.:** 113.8 - 115.6 ℃. **^{1}H NMR (400 MHz, CD_3OD):** δ 9.12 (s, 1 H), 8.36 (s, 1 H), 7.86 (dd, *J* = 6.4, 2.0 Hz, 2 H), 7.60 (d, *J* = 8.0 Hz, 2 H), 7.40 – 7.35 (m, 2 H), 7.33 – 7.23 (m, 3 H), 5.56 (s, 2 H), 4.71 (s, 2 H). **^{13}C NMR (100 MHz, CD_3OD):** δ 148.4, 147.2, 146.4, 143.5, 141.3, 137.3, 136.1, 129.97, 129.95, 129.4, 129.0, 128.7, 125.4, 121.2, 64.5. **ESI-HRMS**: m/z calcd for $C_{20}H_{16}N_6ONa$ [M + Na]$^+$: 379.1278; found: 379.1276.

7-Benzyl-3-(naphthalen-2-yl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2x)



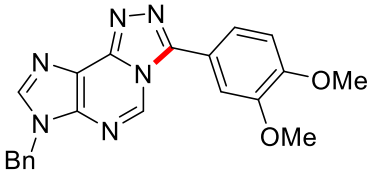
Following the general procedure, the electrochemical reaction was carried out with **1x** (75.7 mg, 0.2 mmol) at room temperature for 2.9 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2x** (67.8 mg, 90%) as a white solid. **M.P.:** 129.7 - 133.9 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.48 (s, 1 H), 8.11 (d, *J* = 8.4 Hz, 1 H), 8.06 (s, 1 H), 7.99 (dd, *J* = 8.0, 1.6 Hz, 1 H), 7.82 (dd, *J* = 6.8, 1.2 Hz, 1 H), 7.75 (dd, *J* = 8.4, 0.8 Hz, 1 H), 7.69 – 7.64 (m, 1 H) , 7.62 – 7.57 (m, 1 H), 7.54 – 7.49 (m, 1 H), 7.38 – 7.29 (m, 5 H), 5.49 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.1, 145.3, 141.4, 139.8, 135.2, 134.1, 133.7, 131.8, 129.5, 129.3, 128.94, 128.85, 128.0, 127.1, 125.4, 124.9, 123.0, 121.5, 48.4. **ESI-HRMS**: m/z calcd for $C_{23}H_{17}N_6$ $[M + H]^+$: 377.1509; found: 377.1500.

7-Benzyl-3-(4-(methylsulfinyl)phenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2y)



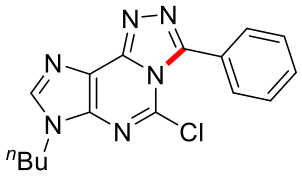
Following the general procedure, the electrochemical reaction was carried out with **1y** (74.9 mg, 0.2 mmol) at room temperature for 3.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2y** (57.4 mg, 74%) as a white solid. **M.P.:** >300 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.99 (s, 1 H), 8.06 (s, 1 H), 8.04 (d, *J* = 2.4 Hz, 2 H), 7.87 (dt, *J* = 8.4, 1.6 Hz, 2 H), 7.38 – 7.28 (m, 5 H), 5.51 (s, 2 H), 2.81 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 148.9, 146.6, 145.4, 141.7, 139.7, 135.1, 133.0, 129.5, 129.3, 128.9, 128.8, 127.9, 124.8, 121.5, 48.5, 44.1. **ESI-HRMS**: m/z calcd for $C_{20}H_{16}N_6OSNa$ $[M + Na]^+$: 411.0999; found: 411.0997.

7-Benzyl-3-(3,4-dimethoxyphenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2z)



Following the general procedure, the electrochemical reaction was carried out with **1z** (77.7 mg, 0.2 mmol) at room temperature for 2.8 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2z** (64.9 mg, 84%) as a white solid, **M.P.:** 170.0 - 173.8 ℃. **^{1}H NMR (400 MHz, CDCl$_3$):** δ 8.95 (s, 1 H), 7.99 (s, 1 H), 7.40 (s, 1 H), 7.34 – 7.26 (m, 6 H), 7.03 (d, *J* = 8.0 Hz, 1 H), 5.48 (s, 2 H), 3.95 (s, 6 H). **^{13}C NMR (100 MHz, CDCl$_3$):** δ 151.2, 150.0, 146.5, 146.1, 141.3, 139.5, 135.2, 133.6, 129.2, 128.7, 127.8, 121.4, 120.9, 118.4, 112.1, 111.5, 56.3, 56.2, 48.3. **ESI-HRMS**: m/z calcd for $C_{21}H_{19}N_6O_2$ $[M + H]^+$: 387.1564; found: 387.1564.

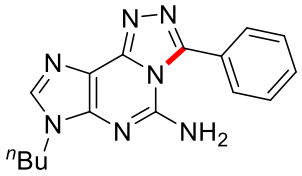
7-Butyl-5-chloro-3-phenyl-7*H*-[1,2,4]triazolo[3,4-i]purine (2aa)



Following the general procedure, the electrochemical reaction was carried out with **1aa** (65.8 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aa** (55.6 mg, 85%) as a white solid. **M.P.:** 102.8 - 107.3 ℃. **^{1}H NMR (400 MHz, CDCl$_3$):** δ 7.98 (s, 1 H), 7.61 – 7.53 (m, 3 H), 7.52 – 7.46 (m, 2 H), 4.29 (t, *J* = 6.8 Hz, 2 H), 1.95 – 1.86 (m, 2 H), 1.42 – 1.32 (m, 2 H), 0.96 (t, *J* = 7.2 Hz, 3 H). **^{13}C NMR (100 MHz, CDCl$_3$):** δ 147.2, 147.0, 141.5, 139.4, 132.0, 131.4, 130.5, 128.0, 127.8, 120.9, 44.7, 32.4, 19.9,

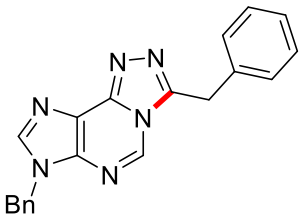
13.6. **ESI-HRMS**: m/z calcd for $C_{16}H_{15}ClN_6Na$ $[M + Na]^+$: 349.0939; found: 349.0937. Spectral data matched those previously reported[7].

7-Butyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-5-amine (2ab)



Following the general procedure, the electrochemical reaction was carried out with **1ab** (61.9 mg, 0.2 mmol) at room temperature for 3.4 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ab** (20.3 mg, 33%) as a yellow solid. **M.P.:** 212.4 - 216.3 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 7.73 – 7.53 (m, 6 H), 5.28 (s, 2 H), 4.11 (t, *J* = 7.2 Hz, 2 H), 1.87 – 1.79 (m, 2 H), 1.38 – 1.29 (m, 2 H), 0.94 (t, *J* = 7.2 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 147.8, 145.2, 142.8, 141.4, 138.2, 131.2, 130.9, 129.1, 127.6, 115.2, 44.0, 32.3, 19.9, 13.6. **ESI-HRMS**: m/z calcd for $C_{16}H_{18}N_7$ $[M + H]^+$: 308.1618; found: 308.1617.

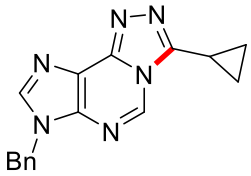
3,7-Dibenzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ac)



Following the general procedure, the electrochemical reaction was carried out with **1ac** (68.5 mg, 0.2 mmol) at room temperature for 2.8 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ac** (51.1 mg, 75%) as a white solid. **M.P.:** 163.5 - 166.6 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.46 (s, 1 H), 7.97 (s, 1 H), 7.36 – 7.23 (m, 10 H), 5.43 (s, 2 H), 4.64 (s, 2 H). **^{13}C NMR (100 MHz,**

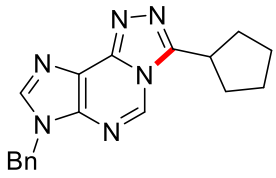
$CDCl_3$): δ 146.1, 145.1, 141.2, 139.3, 135.1, 134.3, 132.9, 129.4, 129.2, 128.8, 128.5, 127.9, 127.8, 121.4, 48.3, 31.8. **ESI-HRMS**: m/z calcd for $C_{20}H_{17}N_6$ $[M + H]^+$: 341.1509; found: 341.1510.

7-Benzyl-3-cyclopropyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ad)



Following the general procedure, the electrochemical reaction was carried out with **1ad** (58.5 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ad** (47.0 mg, 81%) as a white solid, **M.P.:** 201.8 - 206.4 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 8.86 (s, 1 H), 7.96 (s, 1 H), 7.35 – 7.27 (m, 5 H), 5.48 (s, 2 H), 1.30 – 1.15 (m, 5 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 148.0, 145.6, 141.0, 139.5, 135.3, 133.4, 129.2, 128.7, 127.8, 121.4, 47.7, 6.8, 5.1. **ESI-HRMS**: m/z calcd for $C_{16}H_{15}N_6$ $[M + H]^+$: 291.1353; found: 291.1354.

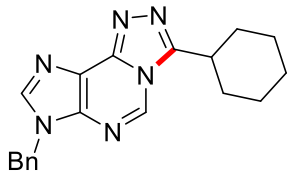
7-Benzyl-3-cyclopentyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ae)



Following the general procedure, the electrochemical reaction was carried out with **1ae** (49.1 mg, 0.2 mmol) at room temperature for 1.7 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ae** (31.4 mg, 50%) as a white solid. **M.P.:** 151.0 - 155.1 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.69 (s, 1 H),

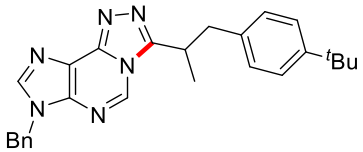
7.97 (s, 1 H), 7.37 – 7.30 (m, 3 H), 7.29 – 7.25 (m, 2 H), 5.48 (s, 2 H), 3.59 – 3.49 (m, 1 H), 2.28 – 2.11 (m, 4 H), 1.96 – 1.85 (m, 2 H), 1.82 – 1.70 (m, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 150.1, 145.8, 141.0, 139.3, 135.3, 133.0, 129.2, 128.7, 127.8, 121.4, 48.3, 35.6, 31.0, 25.6. **ESI-HRMS**: m/z calcd for $C_{18}H_{19}N_6$ $[M + H]^+$: 319.1666; found: 319.1665.

7-Benzyl-3-cyclopentyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2af)



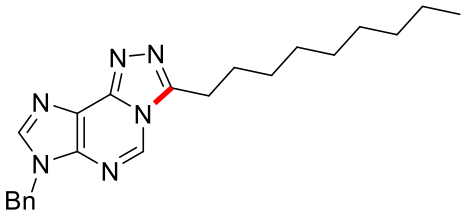
Following the general procedure, the electrochemical reaction was carried out with **1af** (66.9 mg, 0.2 mmol) at room temperature for 1.6 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2af** (49.9 mg, 75%) as a white solid. **M.P.:** 103.9 - 105.9 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.69 (s, 1 H), 8.07 (s, 1 H), 7.36 – 7.28 (m, 5 H), 5.49 (s, 2 H), 3.16 – 3.10 (m, 1 H), 2.14 – 2.10 (m, 2 H), 1.96 – 1.77 (m, 5 H), 1.52 – 1.35 (m, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 150.4, 145.3, 141.3, 139.3, 135.2, 132.9, 129.2, 128.7, 127.9, 121.3, 48.9, 35.1, 30.8, 26.0, 25.8. **ESI-HRMS**: m/z calcd for $C_{19}H_{21}N_6$ $[M + H]^+$: 333.1822; found: 333.1823.

7-Benzyl-3-(1-(4-(*tert*-butyl)phenyl)propan-2-yl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ag)



Following the general procedure, the electrochemical reaction was carried out with **1ag** (85.3 mg, 0.2 mmol) at room temperature for 2.0 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ag** (63.6 mg, 75%) as a white solid, **M.P.:** 183.2 - 187.5 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 8.01 (d, *J* = 1.8 Hz, 1 H), 7.92 (s, 1 H), 7.34 – 7.27 (m, 3 H), 7.22 (dd, *J* = 6.0, 1.8 Hz, 2 H), 7.14 (dd, *J* = 7.8, 1.8 Hz, 2 H), 6.94 (d, *J* = 7.8 Hz, 2 H), 5.39 (s, 2 H), 3.51 (q, *J* = 7.2 Hz, 1 H), 3.14 (dd, *J* = 8.4, 4.2 Hz, 2 H), 1.65 (dd, *J* = 7.2, 2.4 Hz, 3 H), 1.19 (s, 9 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 150.1, 149.9, 145.3, 140.9, 139.0, 136.0, 135.2, 132.6, 129.2, 128.64, 128.62, 127.7, 125.6, 121.1, 48.0, 42.4, 34.4, 34.1, 31.3, 19.2. **ESI-HRMS**: m/z calcd for $C_{26}H_{28}N_6Na$ $[M + Na]^+$: 447.2268; found: 447.2266.

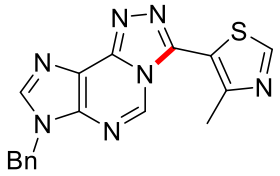
7-Benzyl-3-decyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ah)



Following the general procedure, the electrochemical reaction was carried out with **1ah** (75.7 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ah** (58.7 mg, 78%) as a white solid, **M.P.:** 119.2 - 121.9 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.64 (s, 1 H), 7.98 (s, 1 H), 7.38 – 7.27 (m, 5 H), 5.48 (s, 2 H), 3.16 (t, *J* = 7.6 Hz, 2 H), 1.98 – 1.89 (m, 2 H), 1.50 – 1.42 (m, 2 H), 1.37 – 1.33 (m, 2 H), 1.29 – 1.24 (m, 8 H), 0.86 (t, *J* = 6.4 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.7, 145.6, 141.1, 139.3, 135.3,132.7, 129.3, 128.8, 127.8, 121.5, 48.3, 31.9, 29.5, 29.4, 29.3, 27.0, 25.1, 22.7, 14.2.

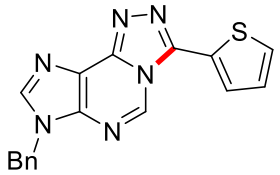
ESI-HRMS: m/z calcd for $C_{22}H_{29}N_6$ $[M + H]^+$: 377.2448; found: 377.2446.

5-(7-Benzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-3-yl)-4-methylthiazole (2ai)



Following the general procedure, the electrochemical reaction was carried out with **1ai** (69.9 mg, 0.2 mmol) at room temperature for 2.4 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ai** (59.1 mg, 85%) as a white solid, **M.P.:** 235.5 - 237.8 ℃. **^{1}H NMR (400 MHz, $CDCl_3$)**: δ 9.01 (s, 1 H), 8.73 (s, 1 H), 8.07 (s, 1 H), 7.39 – 7.29 (m, 5 H), 5.51 (s, 2 H), 2.61 (s, 3 H).**^{13}C NMR (100 MHz, $CDCl_3$):** δ 156.6, 154.6, 146.4, 141.8, 139.9, 138.9, 135.0, 133.0, 129.3, 128.9, 127.9, 121.4, 114.4, 48.5, 16.9. **ESI-HRMS**: m/z calcd for $C_{17}H_{13}N_7SNa$ $[M + Na]^+$: 370.0845; found: 370.0841.

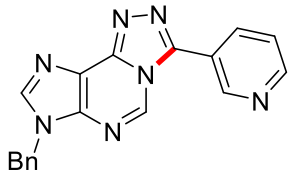
7-Benzyl-3-(thiophen-2-yl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2aj)



Following the general procedure, the electrochemical reaction was carried out with **1aj** (66.9 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aj** (43.9 mg, 66%) as a white solid, **M.P.:** 221.4 - 224.7 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.10 (s, 1 H), 8.03 (s, 1 H), 7.71 (dd, *J* = 3.6, 0.8 Hz, 1 H), 7.61 (dd, *J* = 5.2, 0.8 Hz, 1 H), 7.39 – 7.33 (m, 3 H), 7.32 – 7.28 (m, 2 H), 7.28 – 7.26 (m, 1 H), 5.51 (s, 2 H). **^{13}C NMR**

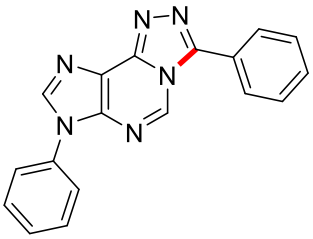
(100 MHz, $CDCl_3$): δ 146.2, 141.7, 141.6, 139.6, 135.1, 133.4, 129.3, 129.1, 128.9, 128.4, 128.3, 127.9, 126.8, 121.5, 48.4. **ESI-HRMS**: m/z calcd for $C_{17}H_{12}N_6SNa$ [M + Na]$^+$: 355.0736; found: 355.0731.

7-Benzyl-3-(pyridin-3-yl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ak)



Following the general procedure, the electrochemical reaction was carried out with **1ak** (65.9 mg, 0.2 mmol) at room temperature for 2.0 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ak** (30.0 mg, 46%) as a white solid, **M.P.:** 101.2 - 105.3 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 9.29 (s, 1 H), 9.13 (dd, *J* = 2.4, 0.8 Hz, 1 H), 8.80 (dd, *J* = 4.8, 1.6 Hz, 1 H), 8.61 (s, 1 H), 8.41 (dt, *J* = 8.0, 1.6 Hz, 1 H), 7.66 (ddd, *J* = 8.0, 4.8, 0.8 Hz, 1 H), 7.36 (d, *J* = 4.4 Hz, 4 H), 7.33 – 7.27 (m, 1 H), 5.60 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 151.1, 149.1, 146.0, 144.2, 142.4, 139.3, 136.6, 136.3, 135.5, 128.8, 128.0, 127.5, 124.1, 122.7, 120.0, 47.2. **ESI-HRMS**: m/z calcd for $C_{18}H_{14}N_7$ [M + H]$^+$: 328.1305; found: 328.1300.

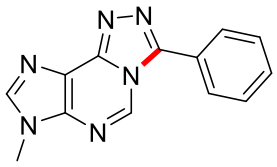
3,7-Diphenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2al)



Following the general procedure, the electrochemical reaction was carried out with **1al** (62.9 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column

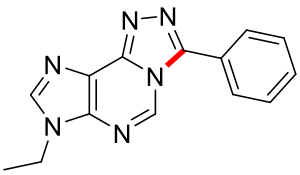
chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2al** (54.8 mg, 88%) as a white solid. **M.P.:** 109.2 - 114.3 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.98 (s, 1 H), 8.26 (s, 1 H), 7.89 – 7.85 (m, 2 H), 7.69 (dd, *J* = 7.2, 1.2 Hz, 2 H), 7.64 – 7.56 (m, 5 H), 7.55 – 7.49 (m, 1 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.8, 146.1, 140.8, 139.2, 134.4, 133.7, 131.0, 130.1, 129.7, 129.1, 128.7, 126.0, 124.2, 122.3. **ESI-HRMS**: m/z calcd for $C_{18}H_{13}N_6$ $[M + H]^+$: 313.1196; found: 313.1195. Spectral data matched those previously reported[7].

7-Methyl-3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2am)



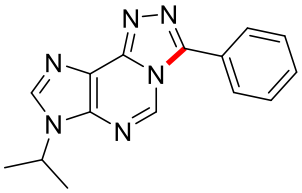
Following the general procedure, the electrochemical reaction was carried out with **1am** (50.5 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2am** (46.1 mg, 92%) as a white solid. **M.P.:** 233.5 - 236.7 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.94 (s, 1 H), 7.98 (s, 1 H), 7.86 (dd, *J* = 7.6, 5.6 Hz, 2 H), 7.65 – 7.59 (m, 3 H), 3.99 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.5, 142.0, 140.0, 133.2, 131.0, 129.7, 128.8, 126.1, 121.4, 30.8. **ESI-HRMS**: m/z calcd for $C_{13}H_{11}N_6$ $[M + H]^+$: 251.1040; found: 251.1039.

7-Ethyl-3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2an)



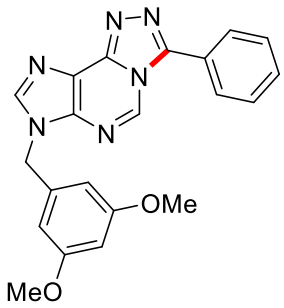
Following the general procedure, the electrochemical reaction was carried out with **1an** (53.3 mg, 0.2 mmol) at room temperature for 2.4 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2an** (50.2 mg, 95%) as a white solid, **M.P.:** 229.7 - 232.5 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.94 (s, 1 H), 8.02 (s, 1 H), 7.86 (dd, *J* = 8.0, 5.6 Hz, 2 H), 7.64 – 7.58 (m, 3 H), 4.39 (q, *J* = 7.6 Hz, 2 H), 1.60 (t, *J* = 7.2 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.5, 146.3, 140.9, 139.4, 133.0, 130.9, 129.6, 128.8, 126.2, 121.6, 40.0, 16.0. **ESI-HRMS**: m/z calcd for $C_{14}H_{13}N_6$ $[M + H]^+$: 265.1196; found: 265.1199.

7-Isopropyl-3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ao)



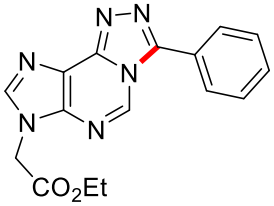
Following the general procedure, the electrochemical reaction was carried out with **1ao** (56.1 mg, 0.2 mmol) at room temperature for 2.4 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ao** (54.0 mg, 97%) as a white solid. **M.P.:** 144.9 - 146.8 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.92 (s, 1 H), 8.03 (s, 1 H), 7.83 (dd, *J* = 7.6, 5.6 Hz, 2 H), 7.61 – 7.52 (m, 3 H), 4.91 (dt, *J* = 6.8 Hz, 1 H), 1.65 (d, *J* = 6.8 Hz, 6 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.4, 146.2, 139.1, 132.7, 130.8, 129.5, 128.7, 126.1, 121.7, 48.5, 22.9. **ESI-HRMS**: m/z calcd for $C_{15}H_{15}N_6$ $[M + H]^+$: 279.1353; found: 279.1351.

7-(3,5-Dimethoxybenzyl)-3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ap)



Following the general procedure, the electrochemical reaction was carried out with **1ap** (77.7 mg, 0.2 mmol) at room temperature for 3.8 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ap** (62.6 mg, 81%) as a yellow solid. **M.P.:** 223.5 – 225.7 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 9.23 (s, 1 H), 8.57 (s, 1 H), 8.00 – 7.94 (m, 2 H), 7.66 – 7.60 (m, 3 H), 6.52 (d, *J* = 2.4 Hz, 2 H), 6.42 (t, *J* = 2.4 Hz, 1 H), 5.49 (s, 2 H), 3.70 (s, 6 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 160.7, 146.3, 145.8, 142.3, 139.2, 138.8, 135.3, 130.4, 129.2, 128.7, 126.1, 120.0, 105.7, 99.3, 55.2, 47.2. **ESI-HRMS**: m/z calcd for $C_{21}H_{18}N_6O_2Na$ $[M + Na]^+$: 409.1383; found: 409.1380.

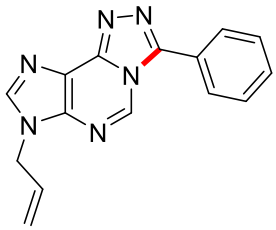
Ethyl 2-(3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)acetate (2aq)



Following the general procedure, the electrochemical reaction was carried out with **1aq** (64.9 mg, 0.2 mmol) at room temperature for 2.7 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aq** (23.9 mg, 37%) as a white solid, **M.P.:** 238.6 - 242.3 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 9.23 (s, 1 H), 8.42 (s, 1 H), 8.00 – 7.95 (m, 2 H), 7.67 – 7.59 (m, 3 H), 5.32 (s, 2 H), 4.20 (q, *J* = 7.2

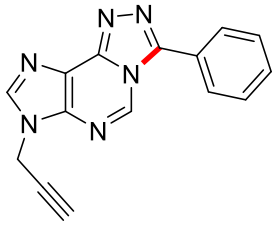
Hz, 2 H), 1.23 (t, *J* = 7.2 Hz, 3 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 167.6, 146.4, 145.6, 142.9, 139.4, 135.5, 130.4, 129.2, 128.7, 126.1, 119.6, 61.6, 44.9, 14.0. **ESI-HRMS**: m/z calcd for $C_{16}H_{15}N_6O_2$ $[M + H]^+$: 323.1251; found: 323.1247. Spectral data matched those previously reported[7].

7-Allyl-3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2ar)



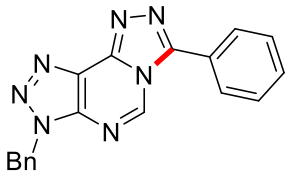
Following the general procedure, the electrochemical reaction was carried out with **1ar** (55.7 mg, 0.2 mmol) at room temperature for 2.3 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ar** (44.2 mg, 80%) as a white solid. **M.P.:** 206.5 - 209.3 °C. **^{1}H NMR (600 MHz, CDCl$_3$):** δ 8.92 (s, 1 H), 7.98 (s, 1 H), 7.83 (dd, *J* = 7.8, 1.8 Hz, 2 H), 7.60 – 7.54 (m, 3 H), 6.10 – 6.02 (m, 1 H), 5.32 (d, *J* = 10.2 Hz, 1 H), 5.22 (dt, *J* = 16.8, 1.8 Hz, 1 H), 4.94 (dt, *J* = 6.0, 1.8 Hz, 2 H). **^{13}C NMR (150 MHz, CDCl$_3$):** δ 146.5, 146.2, 141.3, 139.4, 133.3, 131.6, 130.9, 129.6, 128.7, 126.0, 121.3, 119.5, 46.8. **ESI-HRMS**: m/z calcd for $C_{15}H_{13}N_6$ $[M + H]^+$: 277.1196; found: 277.1198. Spectral data matched those previously reported[7].

3-Phenyl-7-(prop-2-yn-1-yl)-7*H*-[1,2,4]triazolo[3,4-*i*]purine (2as)



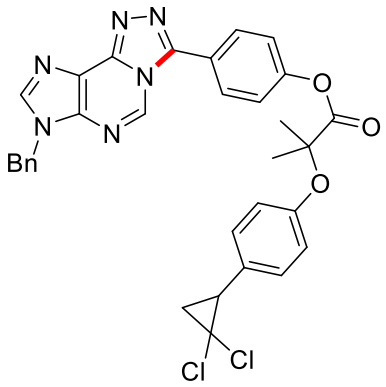
Following the general procedure, the electrochemical reaction was carried out with **1as** (55.3 mg, 0.2 mmol) at room temperature for 2.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2as** (45.0 mg, 82%) as a white solid. **M.P.:** 244.6 - 247.9 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 9.27 (s, 1 H), 8.50 (s, 1 H), 8.02 – 7.97 (m, 2 H), 7.67 – 7.63 (m, 3 H), 5.27 (d, *J* = 2.8 Hz, 2 H), 3.57 (t, *J* = 2.8 Hz, 1 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 146.4, 145.6, 141.7, 138.8, 135.5, 130.5, 129.3, 128.7, 126.1, 120.0, 77.9, 76.5. **ESI-HRMS**: m/z calcd for $C_{15}H_{11}N_6$ $[M + H]^+$: 275.1040; found: 275.1036. Spectral data matched those previously reported[7].

3-Benzyl-7-phenyl-3*H*-[1,2,3]triazolo[4,5-e][1,2,4]triazolo[4,3-c]pyrimidine (2at)



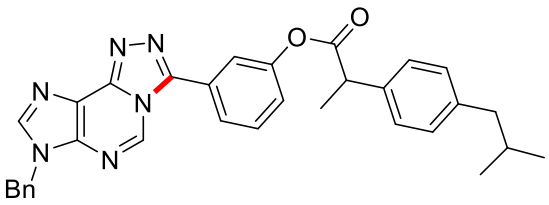
Following the general procedure, the electrochemical reaction was carried out with **1at** (65.9 mg, 0.2 mmol) at room temperature for 3.0 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2at** (56.8 mg, 87%) as a white solid. **M.P.:** 185.3 - 189.8 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 9.38 (s, 1 H), 7.98 – 7.33 (m, 10 H), 5.98 (s, 2 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 147.8, 143.4, 140.9, 140.6, 135.2, 130.9, 129.3, 129.0, 128.8, 128.3, 127.9, 125.4, 125.1, 50.6. **ESI-HRMS**: m/z calcd for $C_{18}H_{13}N_7Na$ $[M + Na]^+$: 350.1125; found: 350.1122. Spectral data matched those previously reported[8].

4-(7-Benzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-3-yl)phenyl 2-(4-(2,2-dichlorocy clopropyl)phenoxy)-2-methylpropanoate (2au)



Following the general procedure, the electrochemical reaction was carried out with **1au** (125.9 mg, 0.2 mmol) at room temperature for 5.8 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2au** (103.1 mg, 84%) as a white solid, **M.P.:** 85.8 - 89.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.92 (s, 1 H), 8.01 (s, 1 H), 7.85 (dt, *J* = 8.8, 2.8 Hz, 2 H), 7.36 – 7.28 (m, 5 H), 7.23 – 7.16 (m, 4 H), 6.97 – 6.93 (m, 2 H), 5.49 (s, 2 H), 2.86 (dd, *J* = 10.8, 8.4 Hz, 1 H), 1.96 (dd, *J* = 10.4, 7.2 Hz, 1 H), 1.81 (d, *J* = 8.4 Hz, 1 H), 1.79 (s, 6 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 172.7, 155.0, 152.4, 146.3, 145.7, 141.5, 139.6, 135.1, 133.2, 130.1, 130.0, 129.3, 128.8, 127.9, 124.0, 122.8, 121.4, 118.7, 79.5, 61.0, 48.4, 34.9, 25.9, 25.62, 25.58. **ESI-HRMS**: m/z calcd for $C_{32}H_{27}Cl_2N_6O_3$ $[M + H]^+$: 613.1516; found: 613.1516.

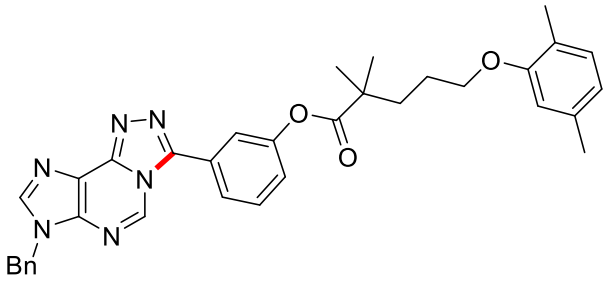
3-(7-Benzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-3-yl)phenyl 2-(4-isobutylphenyl) propanoate (2av)



Following the general procedure, the electrochemical reaction was carried out with **1av** (106.5 mg, 0.2 mmol) at room temperature for 5.8 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2av** (49.8 mg, 47%) as a

yellow solid, **M.P.:** 104.4 - 108.7 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.94 (s, 1 H), 8.03 (s, 1 H), 7.70 (dt, *J* = 7.6, 1.2 Hz, 1 H), 7.60 – 7.54 (m, 2 H), 7.39 – 7.30 (m, 7 H), 7.25 – 7.21 (m, 1 H), 7.15 (dd, *J* = 6.4, 2.0 Hz, 2 H), 5.50 (s, 2 H), 3.98 (q, *J* = 7.2 Hz, 1 H), 2.46 (d, *J* = 7.2 Hz, 2 H), 1.90 – 1.82 (m, 1 H), 1.63 (d, *J* = 7.2 Hz, 3 H), 0.90 (d, *J* = 6.4 Hz, 6 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 173.1, 151.6, 146.4, 145.7, 141.5, 141.2, 139.7, 136.9, 135.1, 133.3, 130.7, 129.8, 129.3, 128.9, 127.9, 127.3, 127.2, 126.0, 124.0, 122.1, 121.4, 48.4, 45.4, 45.2, 30.3, 22.5, 18.6. **ESI-HRMS**: m/z calcd for $C_{32}H_{31}N_6O_2$ $[M + H]^+$: 531.2503; found: 531.2502.

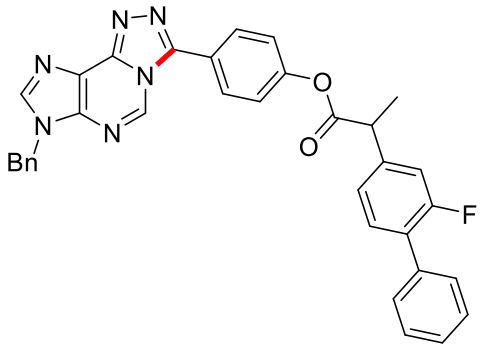
3-(7-Benzyl-7H-[1,2,4]triazolo[3,4-*i*]purin-3-yl)phenyl 5-(2,5-dimethylphenoxy) -2,2-dimethylpentanoate (2aw)



Following the general procedure, the electrochemical reaction was carried out with **1aw** (115.3 mg, 0.2 mmol) at room temperature for 6.0 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aw** (36.8 mg, 32%) as a white solid, **M.P.:** 198.8 - 201.7 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.98 (s, 1 H), 8.04 (s, 1 H), 7.73 (dt, *J* = 7.2, 1.2 Hz, 1 H), 7.64 – 7.59 (m, 2 H), 7.38 – 7.29 (m, 5 H), 7.26 – 7.21 (m, 3 H), 6.65 (s, 1 H), 5.50 (s, 2 H), 3.97 (t, *J* = 5.6 Hz, 2 H), 2.31 (s, 3 H), 2.13 (s, 3 H), 1.92 – 1.87 (m, 4 H), 1.40 (s, 6 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 176.2, 156.3, 151.8, 146.4, 145.7, 141.6, 139.7, 135.8, 135.1, 133.8, 133.3, 130.8, 129.4, 128.9, 127.9, 127.3, 126.4, 125.8, 124.1, 122.3, 121.6, 114.8, 113.6, 68.2, 48.4,

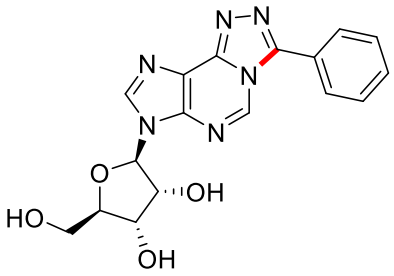
42.7, 37.2, 25.3, 23.0, 15.6. **ESI-HRMS**: m/z calcd for $C_{34}H_{35}N_6O_3$ $[M + H]^+$: 575.2765; found: 575.2764.

4-(7-Benzyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-3-yl)phenyl 2-(2-fluoro-[1,1'-biphenyl]-4-yl)propanoate (2ax)



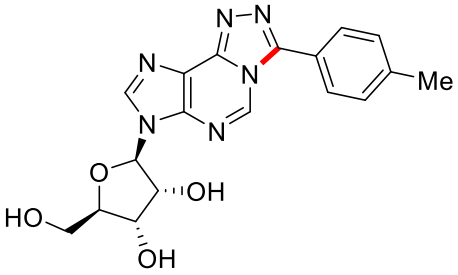
Following the general procedure, the electrochemical reaction was carried out with **1ax** (114.1 mg, 0.2 mmol) at room temperature for 6.4 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2ax** (56.9 mg, 50%) as a white solid, **M.P.:** 153.5 - 165.2 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.92 (s, 1 H), 8.02 (s, 1 H), 7.87 (d, *J* = 8.8 Hz, 2 H), 7.58 – 7.55 (m, 2 H), 7.50 – 7.43 (m, 3 H), 7.40 – 7.26 (m, 9 H), 7.23 (d, *J* = 1.6 Hz, 1 H), 5.49 (s, 2 H), 4.06 (q, *J* = 7.2 Hz, 1 H), 1.70 (d, *J* = 6.8 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 172.3, 161.2, 158.7, 152.7, 146.3, 145.8, 141.5, 141.0, 140.9, 139.6, 135.4, 135.1, 133.2, 131.3 (d, $J_{C\text{-}F}$ = 4.0 Hz), 130.0, 129.3, 129.1 (d, $J_{C\text{-}F}$ = 2.0 Hz), 128.9, 128.6, 128.0, 127.9, 123.8(t, $J_{C\text{-}F}$ = 4.0 Hz), 122.9, 121.5, 115.6, 115.4, 48.4, 45.3, 29.8, 18.5. **^{19}F NMR (376 MHz, $CDCl_3$):** δ -117.00. **ESI-HRMS**: m/z calcd for $C_{34}H_{25}FN_6O_2Na$ $[M + Na]^+$: 591.1915; found: 591.1905.

(2*R*,3*S*,4*R*,5*R*)-2-(Hydroxymethyl)-5-(3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl) tetrahydrofuran-3,4-diol (2ay)



Following the general procedure, the electrochemical reaction was carried out with **1ay** (74.1 mg, 0.2 mmol) at room temperature for 3.0 h. Purification by column chromatography on silica gel (DCM/ MeOH = 20/1) yielded **2ay** (63.4 mg, 86%) as a white solid. **M.P.:** 198.4 - 201.6 ℃. **^{1}H NMR (400 MHz, CD$_3$OD):** δ 9.14 (s, 1 H), 8.57 (s, 1 H), 7.94 – 7.85 (m, 2 H), 7.70 – 7.58 (m, 3 H), 6.15 (d, *J* = 4.8 Hz, 1 H), 4.69 (t, *J* = 4.8 Hz, 1 H), 4.39 (t, *J* = 4.8 Hz, 1 H), 4.12 (q, *J* = 3.6 Hz, 1 H), 3.88 (dd, *J* = 12.0, 3.2 Hz, 1 H), 3.78 (dd, *J* = 12.4, 3.6 Hz, 1 H). **^{13}C NMR (100 MHz, CD$_3$OD):** δ 148.5, 147.0, 142.4, 140.6, 136.2, 132.2, 130.6, 130.0, 126.6, 121.6, 90.7, 87.2, 76.6, 71.8, 62.7. **ESI-HRMS**: m/z calcd for $C_{17}H_{17}N_6O_4$ $[M + H]^+$: 369.1306; found: 369.1307. Spectral data matched those previously reported[5].

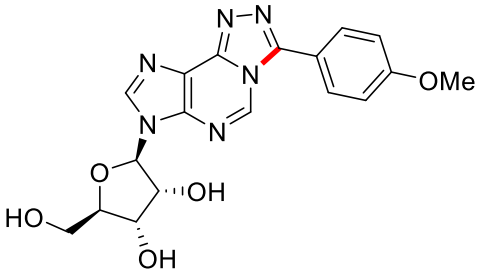
(2*R*,3*S*,4*R*,5*R*)-2-(Hydroxymethyl)-5-(3-(p-tolyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)tetrahydrofuran-3,4-diol (2az)



Following the general procedure, the electrochemical reaction was carried out with **1az** (71.2 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 20/1) yielded **2az** (55.7 mg, 73%) as a

white solid. **M.P.:** 256.9 - 259.8 ℃. **^{1}H NMR (400 MHz, CD_3OD):** δ 9.16 (s, 1 H), 8.65 (s, 1 H), 7.83 (d, *J* = 8.0 Hz, 2 H), 7.71 (d, *J* = 8.0 Hz, 2 H), 6.21 (d, *J* = 5.2 Hz, 1 H), 4.69 (t, *J* = 5.2 Hz, 1 H), 4.38 (t, *J* = 4.8 Hz, 1 H), 4.16 (q, *J* = 3.6 Hz, 1 H), 3.90 (dd, *J* = 12.0, 2.8 Hz, 1 H), 3.80 (dd, *J* = 12.4, 3.6 Hz, 1 H) , 2.48 (s, 3 H). **^{13}C NMR (100 MHz, CD_3OD):** δ 148.8, 147.1, 143.0, 142.4, 140.8, 136.3, 131.3, 130.0, 123.8, 121.9, 90.8, 87.3, 76.6, 71.9, 62.7, 21.5. **ESI-HRMS**: m/z calcd for $C_{18}H_{18}N_6O_4Na$ [M + Na]$^+$: 405.1282; found: 405.1282. Spectral data matched those previously reported[5].

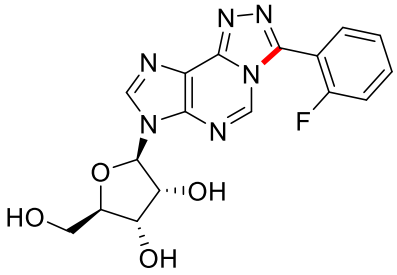
(2*R*,3*S*,4*R*,5*R*)-2-(Hydroxymethyl)-5-(3-(4-methoxyphenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)tetrahydrofuran-3,4-diol (2aaa)



Following the general procedure, the electrochemical reaction was carried out with **1aaa** (80.1 mg, 0.2 mmol) at room temperature for 2.4 h. Purification by column chromatography on silica gel (DCM/ MeOH = 20/1) yielded **2aaa** (62.0 mg, 78%) as a white solid, **M.P.:** 196.3 - 200.6 ℃. **^{1}H NMR (400 MHz, DMSO-d_6):** δ 9.20 (s, 1 H), 8.70 (s, 1 H), 7.92 (dt, *J* = 10.0, 2.8 Hz, 2 H), 7.19 (dt, *J* = 10.0, 3.2 Hz, 2 H), 6.08 (d, *J* = 5.6 Hz, 1 H), 5.58 (d, *J* = 6.0 Hz, 1 H), 5.29 (d, *J* = 5.2 Hz, 1 H), 5.08 (t, *J* = 5.2 Hz, 1 H), 4.58 (q, *J* = 5.2 Hz, 1 H), 4.20 (q, *J* = 4.4 Hz, 1 H), 4.00 (q, *J* = 4.0 Hz, 1 H), 3.87 (s, 3 H), 3.75 – 3.68 (m, 1 H), 3.64 – 3.56 (m, 1 H). **^{13}C NMR (100 MHz, DMSO-d_6):** δ 160.9, 146.3, 145.3, 140.6, 138.8, 135.5, 130.3, 120.4, 118.2, 114.7,

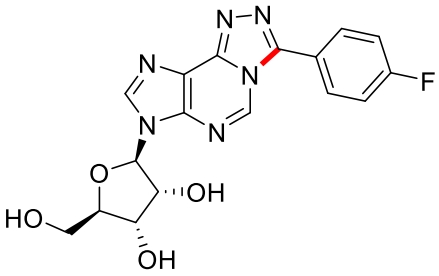
87.9, 85.7, 74.4, 70.2, 61.2, 55.4. **ESI-HRMS**: m/z calcd for $C_{18}H_{18}N_6O_5Na$ $[M + Na]^+$: 421.1231; found: 421.1228.

(2*R*,3*R*,4*S*,5*R*)-2-(3-(2-Fluorophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)-5-(hydroxymethyl)tetrahydrofuran-3,4-diol (2aab)



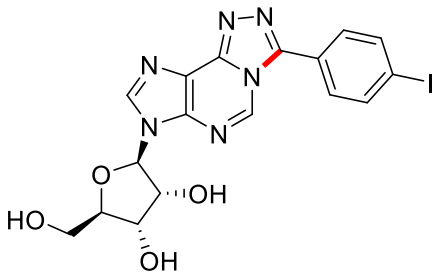
Following the general procedure, the electrochemical reaction was carried out with **1aab** (77.7 mg, 0.2 mmol) at room temperature for 2.9 h. Purification by column chromatography on silica gel (DCM/ MeOH = 20/1) yielded **2aab** (69.4 mg, 90%) as a white solid, **M.P.:** 191.4 - 196.2 ℃. **^{1}H NMR (400 MHz, CD_3OD):** δ 8.96 (d, *J* = 2.8 Hz, 1 H), 8.64 (s, 1 H), 7.85 (td, *J* = 7.2, 1.6 Hz, 1 H), 7.73 – 7.66 (m, 1 H), 7.48 – 7.39 (m, 2 H), 6.19 (d, *J* = 5.2 Hz, 1 H), 4.70 (t, *J* = 4.8 Hz, 1 H), 4.39 (t, *J* = 4.8 Hz, 1 H), 4.14 (q, *J* = 3.6 Hz, 1 H), 3.89 (dd, *J* = 12.4, 3.6 Hz, 1 H), 3.79 (dd, *J* = 12.0, 3.6 Hz, 1 H). **^{13}C NMR (100 MHz, CD_3OD):** δ 161.4 (d, $J_{C\text{-}F}$ = 249.0 Hz), 147.2, 144.0, 142.6, 140.8, 136.4 (d, $J_{C\text{-}F}$ = 7.0 Hz), 134.9 (d, $J_{C\text{-}F}$ = 8.0 Hz), 133.3 (d, $J_{C\text{-}F}$ = 2.0 Hz), 126.6 (d, $J_{C\text{-}F}$ = 4.0 Hz), 121.7, 117.6 (d, $J_{C\text{-}F}$ = 21.0 Hz), 114.6 (d, $J_{C\text{-}F}$ = 14.0 Hz), 90.8, 86.8, 76.6, 71.8, 62.6. **^{19}F NMR (376 MHz, CD_3OD):** δ -113.53. **ESI-HRMS**: m/z calcd for $C_{17}H_{16}FN_6O_4$ $[M + H]^+$: 387.1212; found: 387.1207.

(2*R*,3*R*,4*S*,5*R*)-2-(3-(4-Fluorophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)-5-(hydroxymethyl)tetrahydrofuran-3,4-diol (2aac)



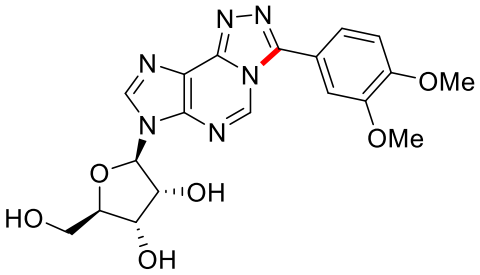
Following the general procedure, the electrochemical reaction was carried out with **1aac** (77.7 mg, 0.2 mmol) at room temperature for 5.0 h. Purification by column chromatography on silica gel (DCM/ MeOH = 20/1) yielded **2aac** (32.0 mg, 41%) as a light yellow solid, **M.P.:** 105.4 - 109.4 ℃. **^{1}H NMR (400 MHz, DMSO-*d*$_6$):** δ 9.24 (s, 1 H), 8.71 (s, 1 H), 8.08 – 8.01 (m, 2 H), 7.52 – 7.45 (m, 2 H), 6.08 (d, *J* = 5.6 Hz, 1 H), 7.59 (d, *J* = 6.0 Hz, 1 H), 5.30 (d, *J* = 5.2 Hz, 1 H), 5.09 (t, *J* = 5.6 Hz, 1 H), 4.61 – 4.56 (m, 1 H), 4.20 (q, *J* = 4.8 Hz, 1 H), 4.00 (q, *J* = 4.0 Hz, 1 H), 3.74 – 3.68 (m, 1 H), 3.63 – 3.57 (m, 1 H). **^{13}C NMR (100 MHz, DMSO-*d*$_6$):** δ 163.3 (d, $J_{C\text{-}F}$ = 247.0 Hz), 145.6 (d, $J_{C\text{-}F}$ = 6.0 Hz), 140.7, 139.0, 135.5, 131.3 (d, $J_{C\text{-}F}$ = 8.0 Hz), 122.6 (d, $J_{C\text{-}F}$ = 4.0 Hz), 120.3, 116.5, 116.3, 87.9, 85.8, 74.4 (d, $J_{C\text{-}F}$ = 10.0 Hz), 70.2 (d, $J_{C\text{-}F}$ = 11.0 Hz), 61.2 (d, $J_{C\text{-}F}$ = 11.0 Hz). **^{19}F NMR (376 MHz, DMSO-*d*$_6$):** δ -110.07. **ESI-HRMS**: m/z calcd for $C_{17}H_{16}FN_6O_4$ $[M + H]^+$: 387.1212; found: 387.1205.

(2*R*,3*S*,4*R*,5*R*)-2-(Hydroxymethyl)-5-(3-(4-iodophenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)tetrahydrofuran-3,4-diol (2aad)



Following the general procedure, the electrochemical reaction was carried out with **2aad** (99.3 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 20/1) yielded **2aad** (62.3 mg, 63%) as a white solid. **M.P.:** 201.2 - 205.2 ℃. **^{1}H NMR (400 MHz, CD_3OD)**: δ 9.19 (s, 1 H), 8.62 (s, 1 H), 8.02 (dt, *J* = 8.8, 2.4 Hz, 2 H), 7.71 (dt, *J* = 8.8, 2.4 Hz, 2 H), 6.19 (d, *J* = 5.2 Hz, 1 H), 4.69 (t, *J* = 4.8 Hz, 1 H), 4.39 (t, *J* = 4.4 Hz, 1 H), 4.15 (q, *J* = 3.6 Hz, 1 H), 3.89 (dd, *J* = 12.0, 3.2 Hz, 1 H), 3.79 (dd, *J* = 12.4, 3.6 Hz, 1 H). **^{13}C NMR (100 MHz, CD_3OD):** δ 147.9, 147.3, 142.5, 140.8, 139.9, 136.2, 131.5, 126.3, 121.8, 98.3, 90.8, 87.3, 76.6, 71.9, 62.7. **ESI-HRMS**: m/z calcd for $C_{17}H_{16}IN_6O_4$ $[M + H]^+$: 495.0272; found: 495.0269.

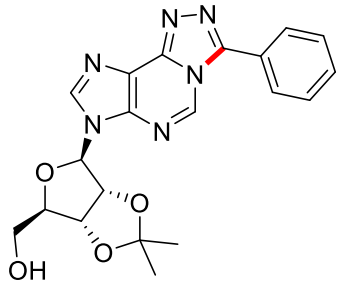
(*2R,3R,4S,5R*)-2-(3-(3,4-Dimethoxyphenyl)-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)-5-(hydroxymethyl)tetrahydrofuran-3,4-diol (2aae)



Following the general procedure, the electrochemical reaction was carried out with **1aae** (86.1 mg, 0.2 mmol) at room temperature for 3.1 h. Purification by column chromatography on silica gel (DCM/ MeOH = 20/1) yielded **2aae** (70.0 mg, 82%) as a white solid. **M.P.:** 264.7 - 266.1 ℃. **^{1}H NMR (400 MHz, DMSO-d_6)**: δ 9.25 (s, 1 H), 8.70 (s, 1 H), 7.50 (d, *J* = 8.0 Hz, 2 H), 7.21 (d, *J* = 8.4 Hz, 1 H), 6.08 (d, *J* = 5.6 Hz, 1 H), 5.57 (s, 1 H), 5.28 (s, 1 H), 5.08 (s, 1 H), 4.57 (s, 1 H), 4.19 (s, 1 H), 3.99 (d, *J* = 4.0 Hz, 1 H), 3.87 (s, 6 H), 3.71 (d, *J* = 12.0 Hz, 1 H), 3.60 (d, *J* = 12.0 Hz, 1 H).**^{13}C**

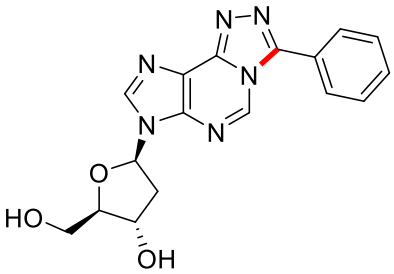
NMR (100 MHz, DMSO-d_6): δ 150.6, 149.1, 146.4, 145.3, 140.5, 138.9, 135.7, 121.7, 120.3, 118.2, 112.1, 112.0, 87.8, 85.7, 74.4, 70.2, 61.1, 55.7. **ESI-HRMS**: m/z calcd for $C_{19}H_{21}N_6O_6$ $[M + H]^+$: 429.1517; found: 429.1520.

((3*aR*,4*R*,6*R*,6*aR*)-2,2-Dimethyl-6-(3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)tetrahydrofuro[3,4-*d*][1,3]dioxol-4-yl)methanol (2aaf)



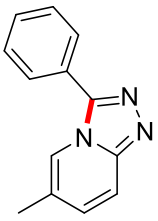
Following the general procedure, the electrochemical reaction was carried out with **1aaf** (82.0 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 20/1) yielded **2aaf** (59.6 mg, 73%) as a white solid, **M.P.:** 210.1 - 213.3 °C. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.98 (s, 1 H), 8.43 (s, 1 H), 7.84 – 7.77 (m, 2 H), 7.60 – 7.55 (m, 3 H), 6.20 (d, *J* = 3.2 Hz, 1 H), 5.31 – 5.23 (m, 2 H), 5.13 (dd, *J* = 6.0, 1.2 Hz, 1 H), 4.55 (d, *J* = 1.6 Hz, 1 H), 4.05 (dd, *J* = 12.0, 2.4 Hz, 1 H), 3.93 (dd, *J* = 12.0, 2.4 Hz, 1 H), 1.63 (s, 3 H), 1.37 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.5, 145.5, 141.1, 138.3, 133.5, 130.9, 129.4, 128.6, 125.4, 121.7, 114.0, 92.9, 87.2, 85.0, 81.7, 62.6, 27.3, 25.2. **ESI-HRMS**: m/z calcd for $C_{20}H_{21}N_6O_4$ $[M + H]^+$: 409.1619; found: 409.1613.

(2*R*,3*S*,5*R*)-2-(hydroxymethyl)-5-(3-phenyl-7*H*-[1,2,4]triazolo[3,4-*i*]purin-7-yl)tetrahydrofuran-3-ol (2aag)



Following the general procedure, the electrochemical reaction was carried out with **1aag** (70.9 mg, 0.2 mmol) at room temperature for 2.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 15/1) yielded **2aag** (67.8 mg, 96%) as a white solid, **M.P.:** 118.5 - 120.7 ℃. **^{1}H NMR (400 MHz, CD_3OD):** δ 9.17 (s, 1 H), 8.60 (s, 1 H), 7.96 – 7.92 (m, 2 H), 7.68 – 7.63 (m, 3 H), 6.58 (t, *J* = 6.8 Hz, 1 H), 4.64 – 4.60 (m, 1 H), 4.05 (q, *J* = 3.6 Hz, 1 H), 3.83 (dd, *J* = 12.0, 4.0 Hz, 1 H), 3.77 (dd, *J* = 12.0 , 4.0 Hz, 1 H), 2.86 – 2.79 (m, 1 H), 2.59 – 2.53 (m, 1 H). **^{13}C NMR (100 MHz, CD_3OD):** δ 148.5, 147.1, 142.1, 140.4, 136.1, 132.2, 130.6, 130.0, 126.7, 121.6, 89.6, 86.6, 72.4, 63.1, 42.0. **ESI-HRMS**: m/z calcd for $C_{17}H_{17}N_6O_3$ $[M + H]^+$: 353.1357; found: 353.1350.

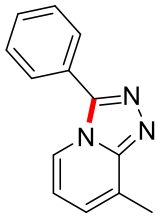
6-Methyl-3-phenyl-[1,2,4]triazolo[4,3-*a*]pyridine (2aah)



Following the general procedure, the electrochemical reaction was carried out with **1aah** (42.3 mg, 0.2 mmol) at room temperature for 2.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aah** (38.1 mg, 91%) as a white solid. **M.P.:** 144.5 - 147.8 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.02 (s, 1 H), 7.80 (dd, *J* = 8.4, 6.8 Hz, 2 H), 7.71 (d, *J* = 9.2 Hz, 1 H), 7.60 – 7.50 (m, 3 H), 7.12

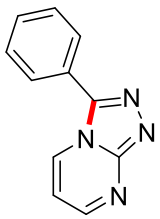
(dd, *J* = 9.2, 0.8 Hz, 1 H), 2.32 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 130.6, 130.2, 129.4, 128.4, 127.0, 124.2, 119.7, 116.1, 18.4. **ESI-HRMS**: m/z calcd for $C_{13}H_{12}N_3$ $[M + H]^+$: 210.1026; found: 210.1025. Spectral data matched those previously reported[3].

8-Methyl-3-phenyl-[1,2,4]triazolo[4,3-*a*]pyridine (2aai)



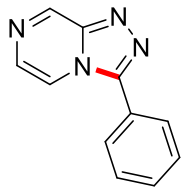
Following the general procedure, the electrochemical reaction was carried out with **1aai** (42.3 mg, 0.2 mmol) at room temperature for 2.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aai** (34.7 mg, 83%) as a white solid. **M.P.:** 96.3 - 99.5 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.11 (d, *J* = 6.8 Hz, 1 H), 7.82 – 7.77 (m, 2 H), 7.59 – 7.48 (m, 3 H), 7.02 (dt, *J* = 6.8, 1.6 Hz, 1 H), 6.76 (t, *J* = 6.8 Hz, 1 H), 2.70 (s, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 151.4, 147.4, 130.2, 129.4, 128.5, 127.5, 127.1, 125.3, 120.4, 114.5, 17.0. **ESI-HRMS**: m/z calcd for $C_{13}H_{12}N_3$ $[M + H]^+$: 210.1026; found: 210.1024. Spectral data matched those previously reported[9].

3-Phenyl-[1,2,4]triazolo[4,3-*a*]pyrimidine (2aaj)



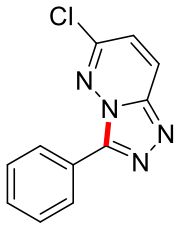
Following the general procedure, the electrochemical reaction was carried out with **1aaj** (39.6 mg, 0.2 mmol) at room temperature for 1.5 h. Purification by column chromatography on silica gel (DCM/ MeOH = 60/1) yielded **2aaj** (36.5 mg, 93%) as a white solid. **M.P.:** 210.7 - 213.5 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.86 (dd, *J* = 6.8, 2.0 Hz, 1 H), 8.81 (q, *J* = 2.0 Hz, 1 H), 8.37 – 8.84 (m, 2 H), 7.54 – 7.49 (m, 3 H), 7.10 (dd, *J* = 6.8, 4.0 Hz, 1 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 166.6, 154.5, 135.6, 131.0, 128.9, 127.7, 110.1. **ESI-HRMS**: m/z calcd for $C_{11}H_9N_4$ $[M + H]^+$: 197.0822; found: 197.0824. Spectral data matched those previously reported[10].

3-Phenyl-[1,2,4]triazolo[4,3-a]pyrazine (2aak)



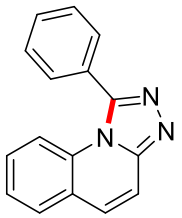
Following the general procedure, the electrochemical reaction was carried out with **1aak** (39.6 mg, 0.2 mmol) at room temperature for 2.4 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aak** (34.5 mg, 88%) as a white solid. **M.P.:** 168.5 - 171.8 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.39 (s, 1 H), 8.21 (d, *J* = 4.4 Hz, 1 H), 7.91 (d, *J* = 4.8 Hz, 1 H), 7.85 (d, *J* = 6.0 Hz, 2 H), 7.59 (d, *J* = 6.0 Hz, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 147.2, 145.9, 145.0, 130.9, 130.4, 129.5, 128.1, 125.5, 115.2. **ESI-HRMS**: m/z calcd for $C_{11}H_8N_4Na$ $[M + Na]^+$: 219.0641; found: 219.0640. Spectral data matched those previously reported[10].

6-Chloro-3-phenyl-[1,2,4]triazolo[4,3-*b*]pyridazine (2aal)



Following the general procedure, the electrochemical reaction was carried out with **1aal** (46.5 mg, 0.2 mmol) at room temperature for 3.2 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aal** (44.3 mg, 96%) as a white solid, **M.P.:** 174.7 - 177.9 ℃. **^{1}H NMR (600 MHz, $CDCl_3$):** δ 8.43 (d, *J* = 7.2 Hz, 2 H), 8.13 (d, *J* = 9.6 Hz, 1 H), 7.60 – 7.50 (m, 3 H), 7.14 (d, *J* = 9.6 Hz, 1 H). **^{13}C NMR (150 MHz, $CDCl_3$):** δ 150.1, 148.1, 143.7, 130.8, 128.9, 127.8, 126.8, 125.6, 121.9. **ESI-HRMS**: m/z calcd for $C_{11}H_8ClN_4$ $[M + H]^+$: 231.0432; found: 231.0431. Spectral data matched those previously reported[10].

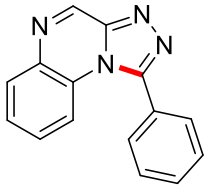
1-Phenyl-[1,2,4]triazolo[4,3-*a*]quinoline (2aam)



Following the general procedure, the electrochemical reaction was carried out with **1aam** (49.4 mg, 0.2 mmol) at room temperature for 2.4 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aam** (45.1 mg, 92%) as a white solid, **M.P.:** 107.6 - 110.5 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 7.78 (dd, *J* = 8.0, 1.2 Hz, 1 H), 7.70 – 7.66 (m, 3 H), 7.65 – 7.52 (m, 5 H), 7.44 (td, *J* = 7.2, 0.8 Hz, 1 H), 7.35 – 7.30 (m, 1 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 150.0, 149.2, 132.0, 130.6, 130.1, 129.8, 129.7, 129.4, 129.2, 129.0, 126.2, 124.7, 116.8, 115.2.

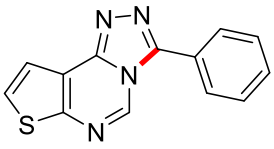
ESI-HRMS: m/z calcd for $C_{16}H_{12}N_3$ $[M + H]^+$: 246.1026; found: 246.1028. Spectral data matched those previously reported[3].

1-Phenyl-[1,2,4]triazolo[4,3-*a*]quinoxaline (2aan)



Following the general procedure, the electrochemical reaction was carried out with **1aan** (49.7 mg, 0.2 mmol) at room temperature for 2.1 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aan** (41.9 mg, 85%) as a white solid, **M.P.:** 153.0 - 154.6 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.28 (s, 1 H), 8.11 (dd, *J* = 8.4, 1.2 Hz, 1 H), 7.72 – 7.53 (m, 7 H), 7.45 – 7.36 (m, 1 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 149.6, 144.8, 143.8, 136.7, 131.2, 131.1, 130.1, 129.5, 129.3, 128.3, 128.1, 127.8, 126.1, 116.1. **ESI-HRMS**: m/z calcd for $C_{15}H_{10}N_4Na$ $[M + Na]^+$: 269.0798; found: 269.0798.

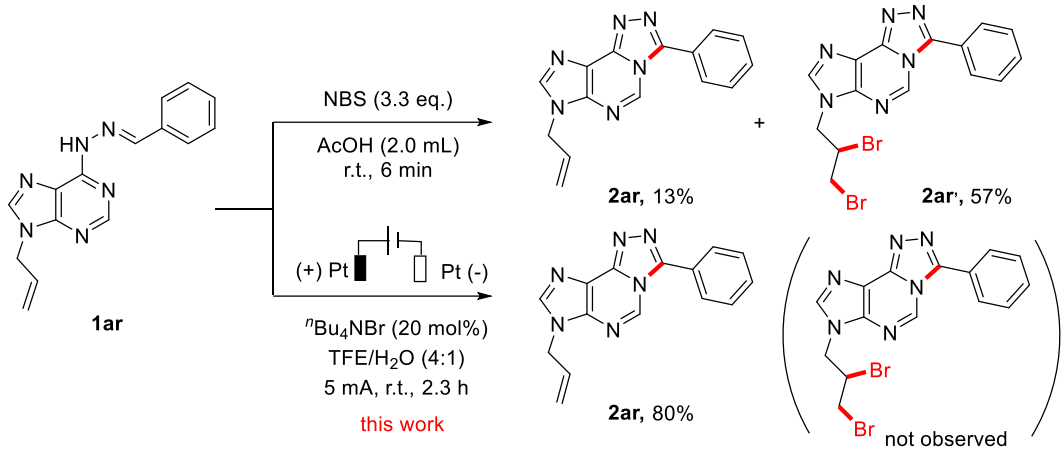
3-Phenylthieno[3,2-*e*][1,2,4]triazolo[4,3-*c*]pyrimidine (2aao)



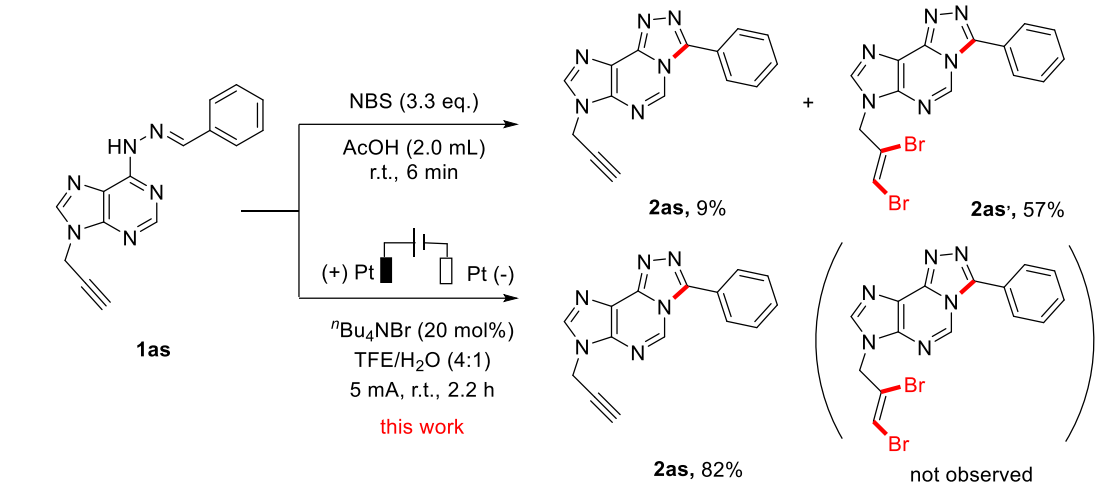
Following the general procedure, the electrochemical reaction was carried out with **1aao** (50.9 mg, 0.2 mmol) at room temperature for 2.6 h. Purification by column chromatography on silica gel (DCM/ MeOH = 50/1) yielded **2aao** (44.9 mg, 89%) as a white solid, **M.P.:** 252.3 - 255.4 ℃. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 8.98 (s, 1 H), 7.91 (d, *J* = 5.6 Hz, 1 H), 7.88 – 7.84 (m, 2 H), 7.68 (d, *J* = 6.0 Hz, 1 H), 7.65 – 7.58 (m, 3 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 152.5, 147.0, 146.8, 133.1, 131.1, 129.7,

128.7, 128.4, 125.8, 120.6, 119.3. **ESI-HRMS**: m/z calcd for $C_{13}H_{19}N_4S$ $[M + H]^+$: 253.0542; found: 253.0543.

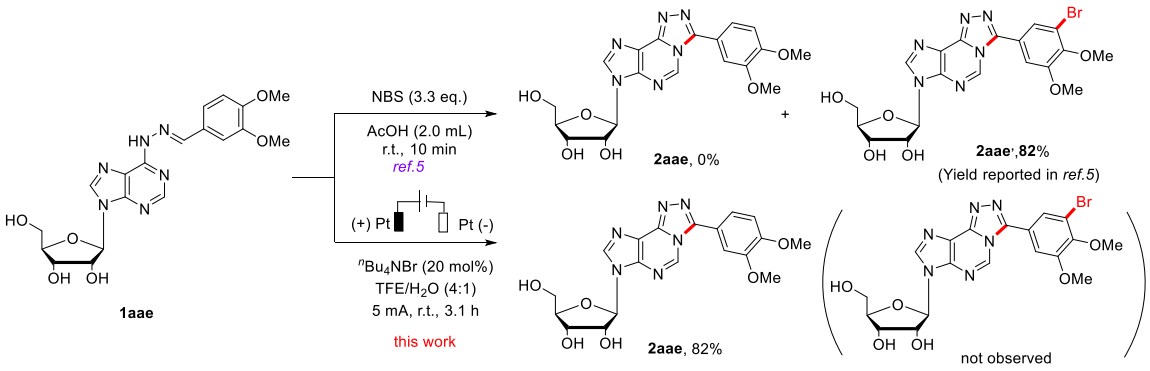
7 Substrate scope comparison between our electrocatalytic method and a previous report using *N*-Bromosuccinimide



The procedures were according to the reported method.[5] *N*-Bromosuccinimide (0.66 mmol) was added portionwise to a mixture of hydrazone (0.2 mmol) in acetic acid (2.0 ml). The contents were stirred at room temperature, and the reaction was monitored by using TLC. Purification by column chromatography on silica gel (DCM/ MeOH = 60/1) yielded **2ar** (7.2 mg, 13%) and **2ar'**(49.7 mg, 57%) as a white solid, **M.P.:** 90.5 - 94.8 °C. **^{1}H NMR (400 MHz, $CDCl_3$)**: δ 8.97 (s, 1 H), 8.15 (s, 1 H), 7.87 – 7.83 (m, 2 H), 7.64 – 7.59 (m, 3 H), 5.12 – 5.04 (m, 1 H), 4.74 – 4.63 (m, 2 H), 3.90 (dd, *J* = 10.4, 3.2 Hz, 1 H), 3.74 (dd, *J* = 10.8, 8.8 Hz, 1 H).**^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.7, 146.0, 142.0, 139.5, 133.7, 131.1, 129.7, 128.8, 125.9, 121.6, 49.3, 48.1, 33.1. **ESI-HRMS**: m/z calcd for $C_{15}H_{13}Br_2N_6$ $[M + H]^+$: 436.9542; found: 436.9547.

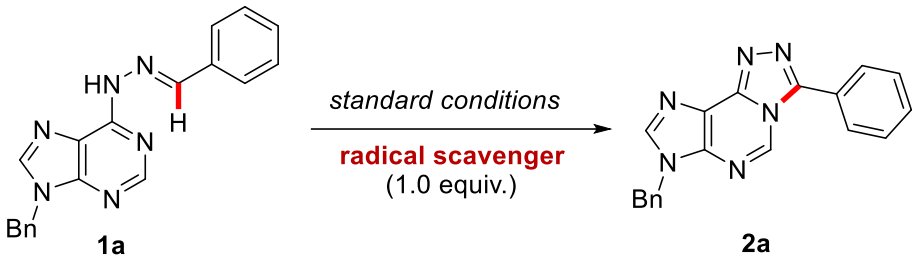


The procedures were according to the reported method.[5] *N*-Bromosuccinimide (0.66 mmol) was added portionwise to a mixture of hydrazone (0.2 mmol) in acetic acid (2.0 ml). The contents were stirred at room temperature, and the reaction was monitored by using TLC. Purification by column chromatography on silica gel (DCM/ MeOH = 60/1) yielded **2as** (4.9 mg, 9%) and **2as'**(49.5 mg, 57%) as a yellow solid, **M.P.:** 195.0 - 198.8 °C. **^{1}H NMR (400 MHz, $CDCl_3$)**: δ 8.97 (s, 1 H), 8.09 (s, 1 H), 7.86 – 7.83 (m, 2 H), 7.61 – 7.58 (m, 3 H), 6.82 (s, 1 H), 5.04 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 146.7, 146.1, 141.4, 139.6, 133.7, 131.0, 129.7, 128.8, 125.9, 121.3, 118.7, 108.5, 48.3. **ESI-HRMS**: m/z calcd for $C_{15}H_{11}Br_2N_6$ $[M + H]^+$: 434.9386; found: 434.9387.



8 Preliminary Mechanistic Studies

8.1 Radical Scavenger Addition Experiments



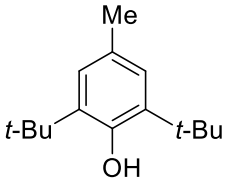
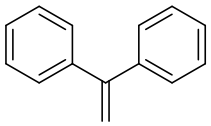
Radical scavenger	none		
		BHT	DPE
Yield[%]	98	27	55

Figure S3: Radical scavenger addition experiments

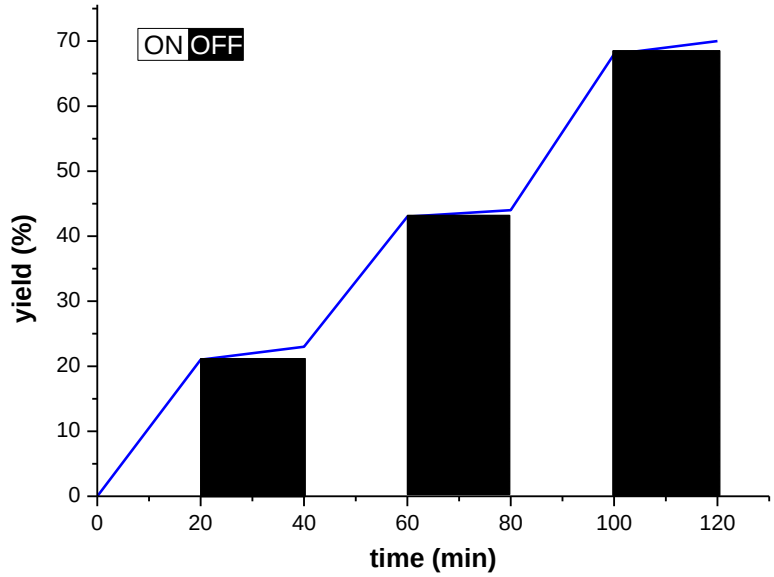
Three parallel radical scavenger addition experiments were conducted under standard conditions using **1a** (65.5 mg, 0.2 mmol) as substrates with BHT (44.1 mg, 0.2 mmol), and 1,1-diphenylethylene (36.1 mg, 0.2 mmol) respectively. The electrochemistry enabled intramolecular dehydrogenative aminooxygenation was not completely inhibited but led to a significant decrease in the yield of **2a**, it indicates that the reaction may be a free radical pathway.

8.2 Power on/off experiment

The electrocatalysis was carried out in an undivided cell equipped with two platinum electrodes (1.5 × 1.5 × 0.2 cm^2). The substrates **1a** (0.2 mmol) and $^nBu_4NBr$ (12.9 mg, 0.04 mmol, 20 mol%) were dissolved in the mixture solvent TFE/H_2O (4/1 mL). The electrolysis was carried out at room temperature using a constant current of 5.0 mA. Aliquots of 0.2 mL were taken at each of the times listed in the table below. The mixture was evaporated under reduced pressure. The residues were diluted with

$CDCl_3$ and analyzed by ^{1}H-NMR spectroscopy based on CH_2Br_2 as an internal standard.

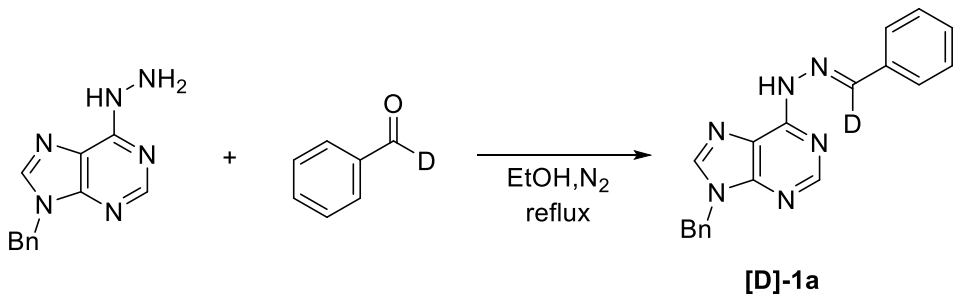
Time [min]	0	20	40	60	80	100	120
Yield of 2a [%]	0	21	23	43	44	68	70



Figures S4: Power on/off experiment

8.3 Kinetic Isotope Effect Studies

General Procedure for the Preparation of (*E*)-9-benzyl-6-(2-(phenylmethylene-d) hydrazinyl)-9*H*-purine [D]-1a

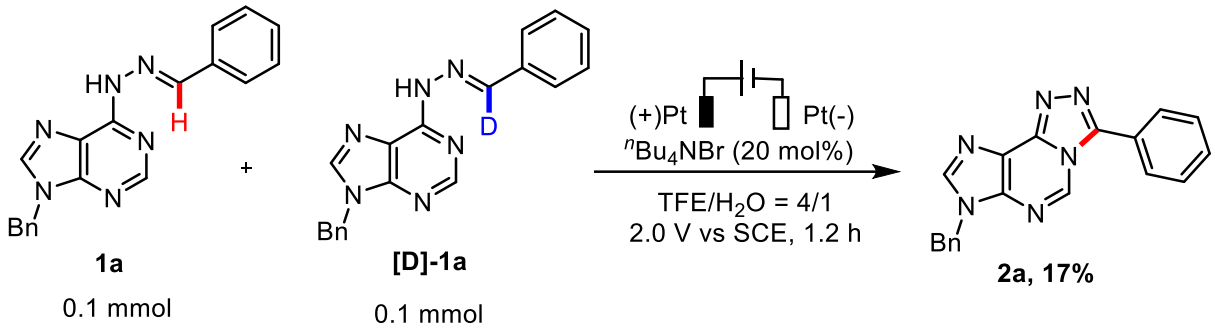


[D]-1a

Following a known literature report, [11] a mixture of the hydrazine (0.5 mmol) and aldehyde (0.5 mmol) in EtOH (6.0 mL) was heated at reflux for 2 hours (TLC tracking detection) at nitrogen atmosphere. Then, the solvent was evaporated under

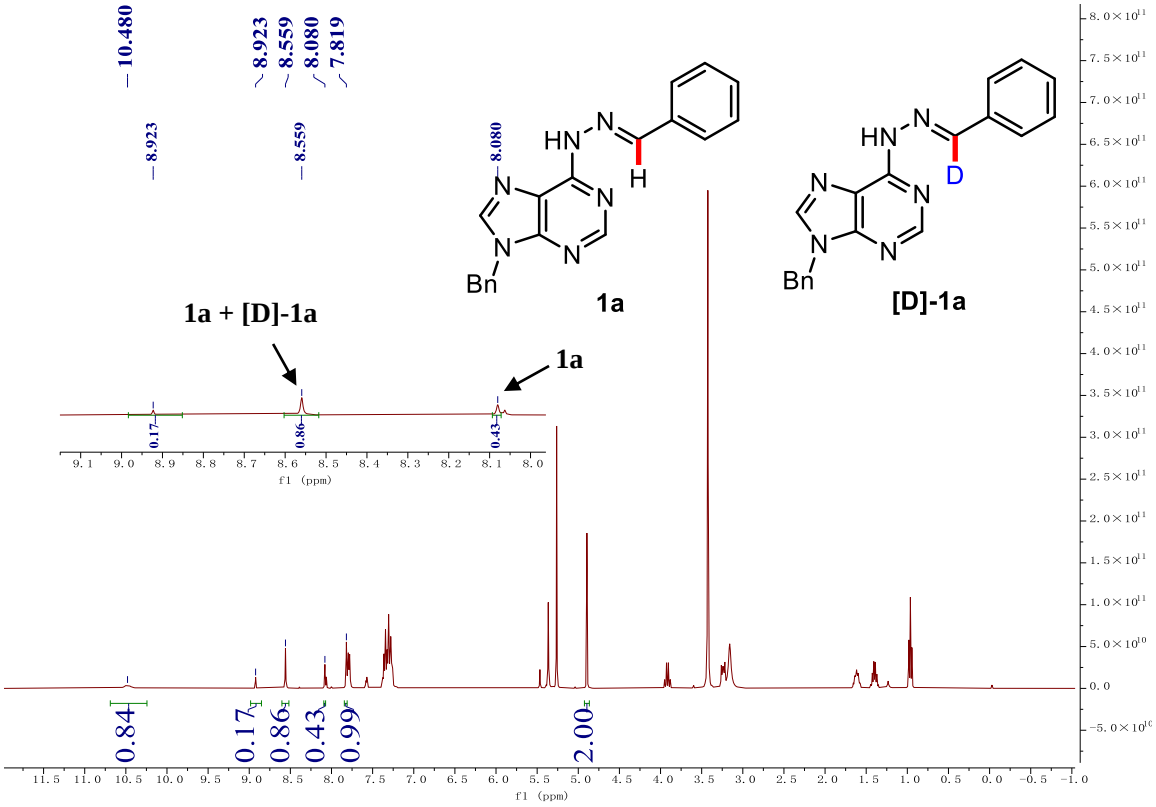
reduced pressure, evaporate the solvent under vacuum. Purify the mixture residue by flash column chromatography (DCM/MeOH = 80:1) to afford **[D]-1a** (131.8 mg, 80%) as a white solid. **M. p.:** 194.4 – 196.7 °C. **^{1}H NMR (400 MHz, $CDCl_3$):** δ 9.79 (s, 1 H), 8.63 (s, 1 H), 7.83 (s, 1 H), 7.77 (dd, *J* = 7.6, 2.4 Hz, 2 H), 7.40 – 7.27 (m, 8 H), 5.39 (s, 2 H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 153.6, 151.6, 141.2, 135.5, 133.9, 130.1, 129.2, 128.7, 128.6, 128.0, 127.6, 119.0, 47.4. **ESI-HRMS**: m/z calcd for $C_{19}H_{16}DN_6$ $[M + H]^+$: 330.1572; found: 330.1574. The deuterization rate was determined to be 97% by NMR.

a) Intermolecular competition experiment:

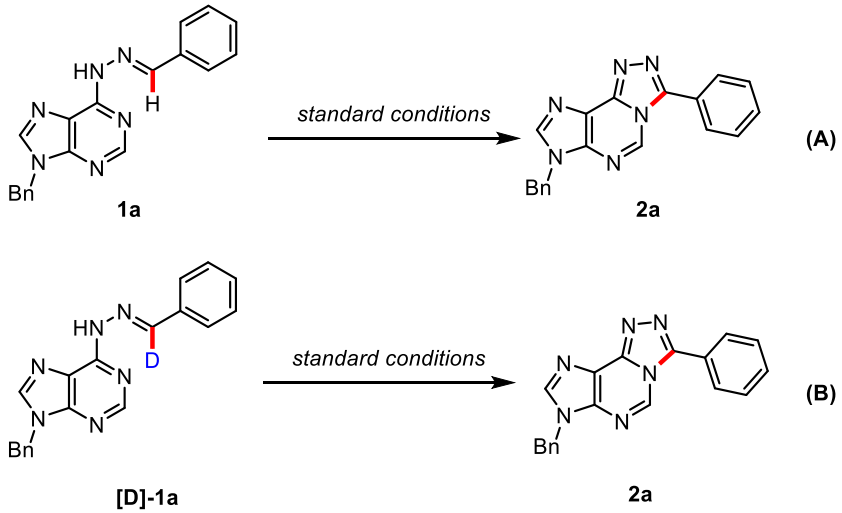


According to general procedure, the electrocatalysis was carried out in an undivided cell equipped with platinum electrode (1.5 × 1.5 × 0.2 cm^2) as the working electrode and platinum electrode (1.0 × 1.0 × 0.2 cm^2) as the auxiliary electrode, and saturated calomel electrode as the reference electrode. The (*E*)-9-Benzyl-6-(2-benzylidenehydrazinyl)-9*H*-purine **1a** (0. 1 mmol) and **[D]-1a** (0. 1 mmol), and ^{n}Bu_4NBr (12.9 mg, 0.04 mmol, 20 mol%) were dissolved in the solvent TFE/H_2O (4/1 mL). Electrolysis was conducted at a constant voltage of 2.0 V at room temperature for 1.2 h. The reaction mixture was concentrated in vacuo. Both reaction mixtures were determined by ^{1}H NMR analysis of the crude product using CH_2Br_2 as

the internal standard. The ratio of unreacted **[D]-1a** vs **1a** in the reaction mixture was 0.43:0.43 as determined by ^{1}H NMR, thus giving a calculated K_H/K_D = 0.43/0.43 = 1.00.



b) Parallel experiments:



Reaction (A): according to general procedure, the electrocatalysis was carried out in an undivided cell equipped with two platinum electrode (1.5 × 1.5 × 0.2 cm^2). The

(*E*)-9-Benzyl-6-(2-benzylidenehydrazinyl)-9*H*-purine **1a** (0.2 mmol), and $^{n}Bu_4NBr$ (12.9 mg, 0.04 mmol, 20 mol%) were dissolved in the solvent TFE/H_2O (4/1 mL). The reaction mixture was electrolyzed at room temperature and stopped respectively at 10 min, 20 min, 30 min, 40 min, and 50 min. In similar, substrate **[D]-1a** (65.9 mg, 0.2 mmol) was used instead of **1a** for the reaction (B). The yield of products was determined by ^1H NMR with CH_2Br_2 as internal standard and the reaction rate was obtained by plotting the percentage yield of the product versus time. The kinetic isotope effect (k_H/k_D) was determined to be 1.00.

Table S2 Kinetic Isotope Effect Studies for **1a**

Time [min]	10	20	30	40	50
Yield [%]	9	18	25	34	43

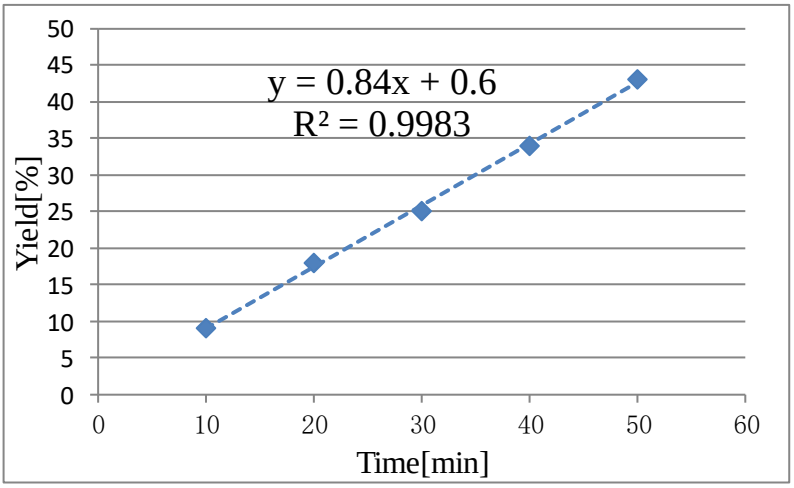


Figure S5: Plots to Determine the KIE were taken for **1a**.

Table S3 Kinetic Isotope Effect Studies for **[D]-1a**

Time [min]	10	20	30	40	50
Yield [%]	8	16	24	31	41

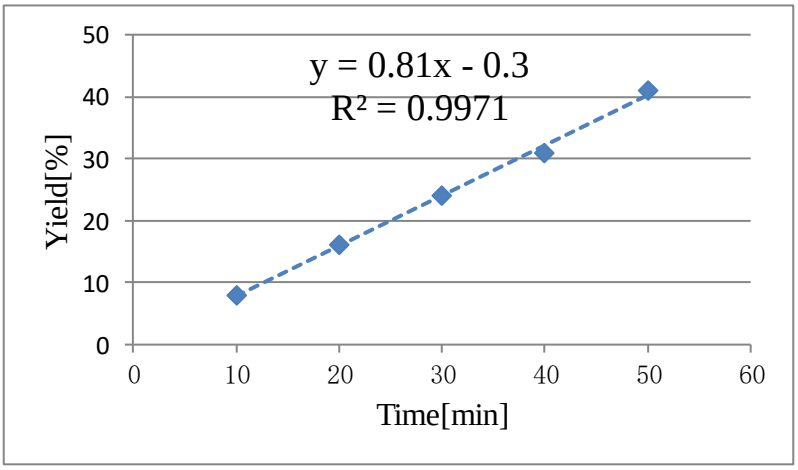
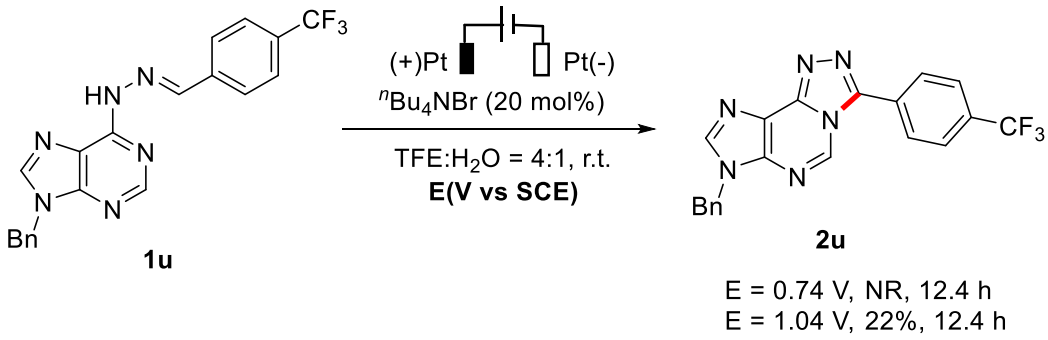


Figure S6: Plots to Determine the KIE were taken for **[D]-1a**.

8.4 Controlled potential electrolysis



The electrocatalysis was carried out in an undivided cell equipped with platinum electrode (1.5 × 1.5 × 0.2 cm^2) as the working electrode and platinum electrode (1.0 × 1.0 × 0.2 cm^2) as the auxiliary electrode, and saturated calomel electrode as the reference electrode. The (*E*)-9-Benzyl-6-(2-(4-(trifluoromethyl) benzylidene) hydrazinyl)-9*H*-purine **1u** (0.2 mmol) and $^{n}Bu_4NBr$ (12.9 mg, 0.04 mmol, 20 mol%) were dissolved in the solvent TFE/H_2O (4/1 mL). The electrolysis was carried out at

room temperature using a constant voltage. After the reaction, the solvent was removed under reduced pressure. The yield of products was determined by ^{1}H NMR with CH_2Br_2 as internal standard. The controlled potential electrolysis (E = 0.74 V) does not afford the desired product, whereas the product **2u** is obtained in 22% yield at higher potential (E = 1.04 V).

9 Proposed Mechanism with ESI-Mass (m/z) Analysis

When the reaction mixture was subjected to detailed ESI-MS study at the reaction time of 50 min, we identified various masses at 329.1506, 329.1428, 327.1354, and those are representing for substrate **1a** ($[M+H]^+$, calcd mass: 329.1509), intermediate **B**, **C**, or **D** ($[M+H]^+$, calcd mass: 328.1431), and product **2a** ($[M+H]^+$, calcd mass: 327.1353), respectively.

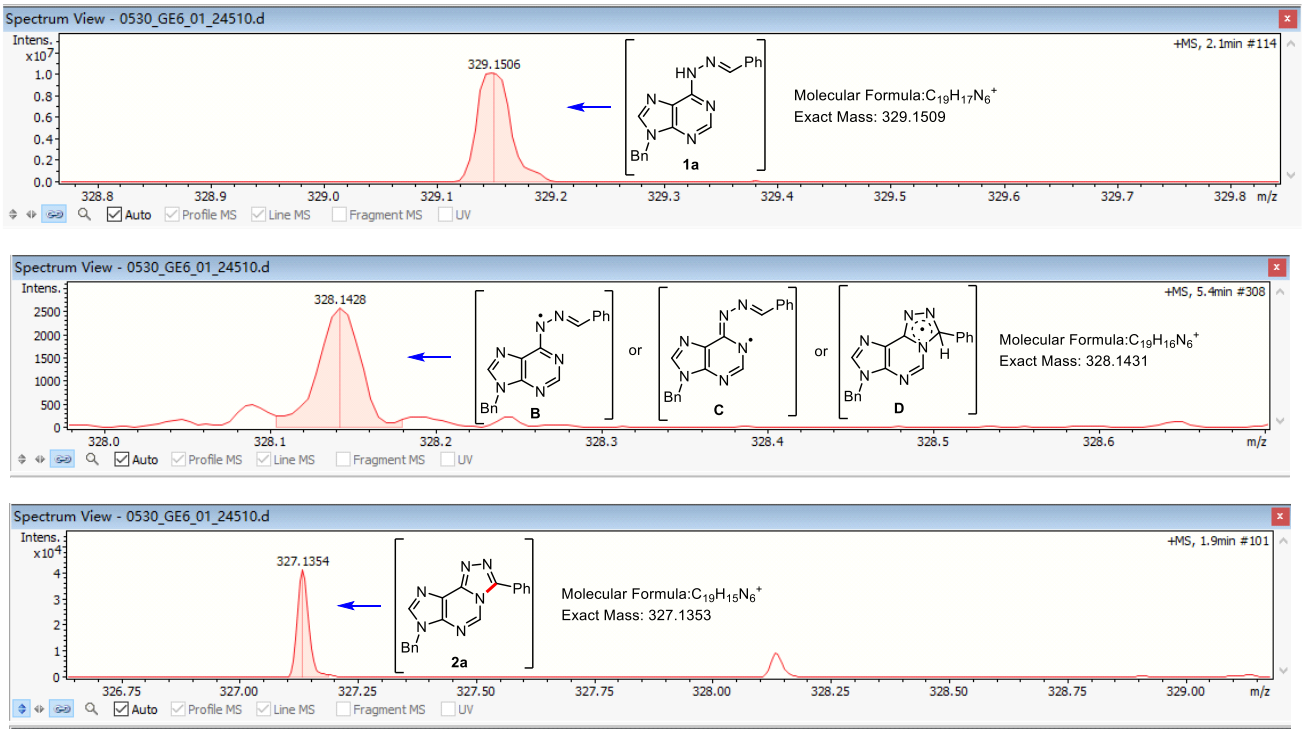


Figure S7: Detection of reaction intermediates by ESI-Mass Spectroscopy

10 Cyclic Voltammetry Studies

Cyclic voltammograms were recorded with a CHI660E potentiostat at room temperature in MeCN. $^{n}Bu_4NPF_6$ (0.1 M) was used as the supporting electrolyte, and a Pt electrode (area = 0.03 cm^2) was used as the working electrode. The auxiliary electrode was a Pt sheet. All potentials are referenced against the SCE redox couple.

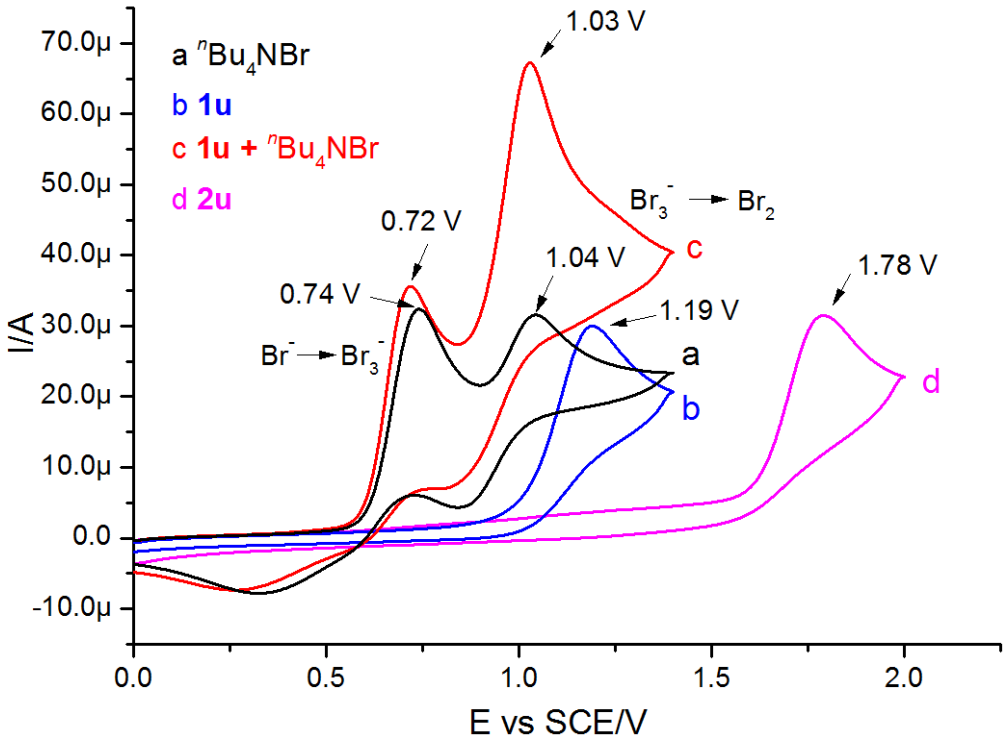


Figure S8: Cyclic voltammograms obtained in 0.1 M $^{n}Bu_4NPF_6$/MeCN using glass carbon (diameter, 3 mm) as the working electrode, Pt wire, and saturated calomel electrode (SCE) as the auxiliary and reference electrode, respectively, at a scan rate of 0.1 $V \cdot s^{-1}$**:** (a) background, (b) 3 mM **1u**, (c) 3 mM **1u+** 2 mM $^{n}Bu_4NBr$, and (d) 3 mM **2u**.

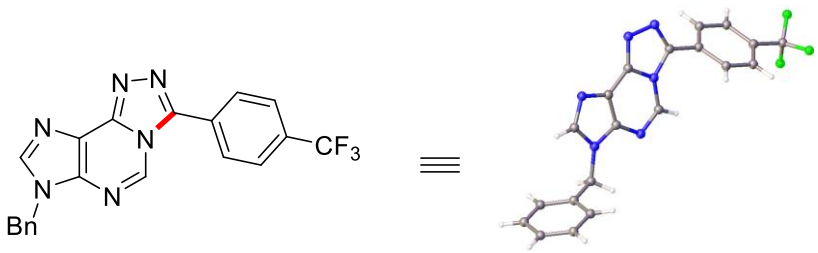
11 X-ray Crystallographic Data

General Procedure for Compound 2u Crystal Preparation:

Compound **2u** (around 20 mg) was dissolved in DCM-PE (1:1, 10 mL). The single crystal was grown by slow evaporation of solvents at room temperature.

Compound **2u** was collected at 100 K on a Rigaku Oxford Diffraction Supernova Dual Source, Cu at Zero equipped with an AtlasS2 CCD. The data were collected and processed using CrysAlisPro. The structures were solved by direct methods using Olex2 software, and the non-hydrogen atoms were located from the trial structure and S26 then refined anisotropically with SHELXL-2018 using a full-matrix least squares procedure based on F^2. The weighted R factor, wR and goodness-of-fit S values were obtained based on F^2. The hydrogen atom positions were fixed geometrically at the calculated distances and allowed to ride on their parent atoms. Crystallographic data for the structure reported in this paper have been deposited at the Cambridge Crystallographic Data Center and allocated with the deposition number: CCDC 2222383 for compound **2u**, basic information pertaining to crystal parameters and structure refinement are summarized in Figure S9.

Compound 2u (CCDC 2222383)



2u **2u**

Figure S9: X-ray structure of **2u** with 50% ellipsoid probability

Empirical formula	$C_{20}H_{13}F_3N_6$
Formula weight	394.36
Temperature/K	293(2)
Crystal system	monoclinic

Space group	$P2_1/c$
a/Å	13.9286(5)
b/Å	9.9839(4)
c/Å	13.3315(5)
α/°	90
β/°	94.139(3)
γ/°	90
Volume/$Å^3$	1849.07(12)
Z	4
$\rho_{calc}g/cm^3$	1.417
μ/mm^{-1}	0.939
F(000)	808.0
Crystal size/mm^3	0.3 × 0.3 × 0.2
Radiation	Cu Kα (λ = 1.54184)
2Θ range for data collection/°	10.912 to 142.514
Index ranges	$-17 \leq h \leq 17$, $-11 \leq k \leq 8$, $-16 \leq l \leq 12$
Reflections collected	8232
Independent reflections	3433 [R_{int} = 0.0385, R_{sigma} = 0.0384]
Data/restraints/parameters	3433/0/262
Goodness-of-fit on F^2	1.071
Final R indexes [I>=2σ (I)]	R_1 = 0.0832, wR_2 = 0.2351
Final R indexes [all data]	R_1 = 0.0985, wR_2 = 0.2638
Largest diff. peak/hole / e $Å^{-3}$	0.30/-0.46

General Procedure for Compound 2y Crystal Preparation:

Compound **2y** (around 20 mg) was dissolved in DCM-PE (1:1, 10 mL). The single crystal was grown by slow evaporation of solvents at room temperature.

Compound **2y** was collected at 100 K on a Rigaku Oxford Diffraction Supernova Dual Source, Cu at Zero equipped with an AtlasS2 CCD. The data were collected and processed using CrysAlisPro. The structures were solved by direct methods using Olex2 software, and the non-hydrogen atoms were located from the trial structure and S26 then refined anisotropically with SHELXL-2018 using a full-matrix least squares procedure based on F^2. The weighted R factor, wR and goodness-of-fit S values were obtained based on F^2. The hydrogen atom positions were fixed geometrically at the

calculated distances and allowed to ride on their parent atoms. Crystallographic data for the structure reported in this paper have been deposited at the Cambridge Crystallographic Data Center and allocated with the deposition number: CCDC 2222382 for compound **2y**, basic information pertaining to crystal parameters and structure refinement are summarized in Figure S10.

Compound 2y (CCDC 2222382)

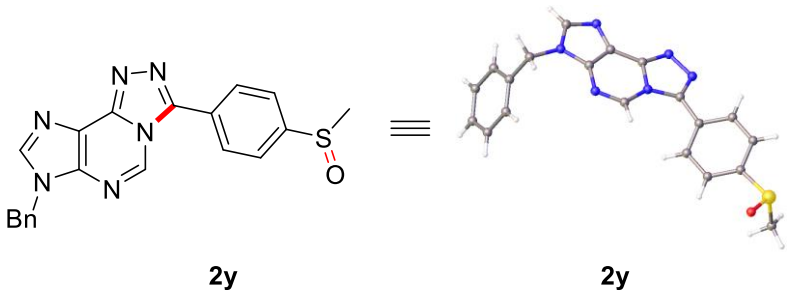


Figure S10: X-ray structure of **2y** with 50% ellipsoid probability

Empirical formula	$C_{20}H_{16}N_6OS$
Formula weight	388.45
Temperature/K	169.99(10)
Crystal system	orthorhombic
Space group	$Pna2_1$
a/Å	11.3223(3)
b/Å	15.5202(6)
c/Å	10.3674(4)
α/°	90
β/°	90
γ/°	90
Volume/$Å^3$	1821.80(11)
Z	4
$\rho_{calc}g/cm^3$	1.416
μ/mm^{-1}	1.783
F(000)	808.0
Crystal size/mm^3	0.14 × 0.12 × 0.1
Radiation	Cu Kα (λ = 1.54184)
2Θ range for data collection/°	9.67 to 147.518
Index ranges	$-7 \leq h \leq 13$, $-13 \leq k \leq 19$, $-12 \leq l \leq 12$
Reflections collected	6353
Independent reflections	3278 [R_{int} = 0.0328, R_{sigma} = 0.0400]

Data/restraints/parameters	3278/1/265
Goodness-of-fit on F^2	1.053
Final R indexes [I>=2σ (I)]	R_1 = 0.0466, wR_2 = 0.1119
Final R indexes [all data]	R_1 = 0.0457, wR_2 = 0.1132
Largest diff. peak/hole / e $Å^{-3}$	0.46/-0.39

General Procedure for Compound 2ay Crystal Preparation:

Compound **2ay** (around 20 mg) was dissolved in DCM-PE (1:1, 10 mL). The single crystal was grown by slow evaporation of solvents at room temperature.

Compound **2ay** was collected at 100 K on a Rigaku Oxford Diffraction Supernova Dual Source, Cu at Zero equipped with an AtlasS2 CCD. The data were collected and processed using CrysAlisPro. The structures were solved by direct methods using Olex2 software, and the non-hydrogen atoms were located from the trial structure and S26 then refined anisotropically with SHELXL-2018 using a full-matrix least squares procedure based on F^2. The weighted R factor, wR and goodness-of-fit S values were obtained based on F^2. The hydrogen atom positions were fixed geometrically at the calculated distances and allowed to ride on their parent atoms. Crystallographic data for the structure reported in this paper have been deposited at the Cambridge Crystallographic Data Center and allocated with the deposition number: CCDC 2222385 for compound **2ay**, basic information pertaining to crystal parameters and structure refinement are summarized in Figure S11.

Compound 2ay (CCDC 2222385)

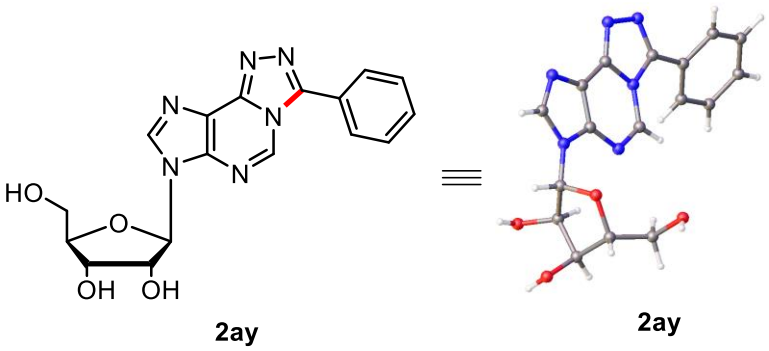


Figure S11: X-ray structure of **2ay** with 50% ellipsoid probability

Empirical formula	$C_{17}H_{16}N_6O_4$
Formula weight	368.36
Temperature/K	170.00(10)
Crystal system	orthorhombic
Space group	$P2_12_12_1$
a/Å	7.16540(10)
b/Å	11.4212(2)
c/Å	19.8938(4)
α/°	90
β/°	90
γ/°	90
Volume/Å^3	1628.06(5)
Z	4
ρ_{calc}g/cm^3	1.503
μ/mm^{-1}	0.932
F(000)	768.0
Crystal size/mm^3	0.15 × 0.11 × 0.09
Radiation	Cu Kα (λ = 1.54184)
2Θ range for data collection/°	8.89 to 147.722
Index ranges	$-8 \le h \le 5$, $-12 \le k \le 13$, $-24 \le l \le 20$
Reflections collected	5776
Independent reflections	3195 [R_{int} = 0.0231, R_{sigma} = 0.0316]
Data/restraints/parameters	3195/0/255
Goodness-of-fit on F^2	1.086
Final R indexes [I>=2σ (I)]	R_1 = 0.0298, wR_2 = 0.0752
Final R indexes [all data]	R_1 = 0.0306, wR_2 = 0.0761
Largest diff. peak/hole / e Å^{-3}	0.14/-0.22

General Procedure for Compound 2aaf Crystal Preparation:

Compound **2aaf** (around 20 mg) was dissolved in DCM-PE (1:1, 10 mL). The single crystal was grown by slow evaporation of solvents at room temperature.

Compound **2aaf** was collected at 100 K on a Rigaku Oxford Diffraction Supernova Dual Source, Cu at Zero equipped with an AtlasS2 CCD. The data were collected and processed using CrysAlisPro. The structures were solved by direct methods using Olex2 software, and the non-hydrogen atoms were located from the trial structure and S26 then refined anisotropically with SHELXL-2018 using a full-matrix least squares procedure based on F^2. The weighted R factor, wR and goodness-of-fit S values were obtained based on F^2. The hydrogen atom positions were fixed geometrically at the calculated distances and allowed to ride on their parent atoms. Crystallographic data for the structure reported in this paper have been deposited at the Cambridge Crystallographic Data Center and allocated with the deposition number: CCDC 2222384 for compound **2aaf**, basic information pertaining to crystal parameters and structure refinement are summarized in Figure S12.

Compound 2aaf (CCDC 2222384)

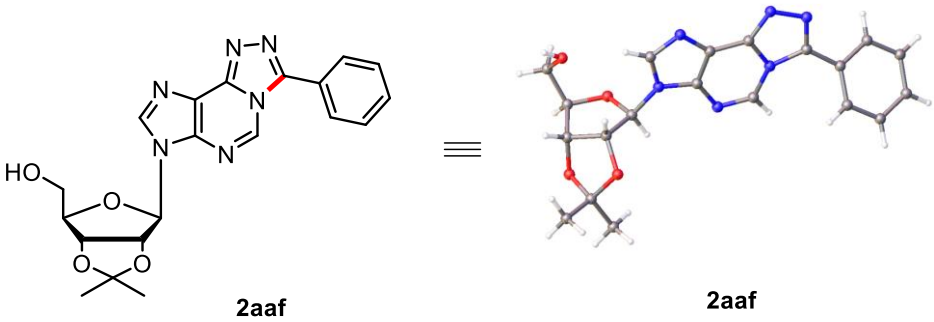


Figure S12: X-ray structure of **2aaf** with 50% ellipsoid probability

Empirical formula	$C_{20}H_{20}N_6O_4$
Formula weight	408.42
Temperature/K	293(2)
Crystal system	orthorhombic
Space group	$P2_12_12_1$
a/Å	8.58890(10)
b/Å	13.3226(2)
c/Å	17.0548(3)

α/°	90
β/°	90
γ/°	90
Volume/Å^3	1951.52(5)
Z	4
ρ_{calc}g/cm^3	1.390
μ/mm^{-1}	0.833
F(000)	856.0
Crystal size/mm^3	0.3 × 0.2 × 0.2
Radiation	Cu Kα (λ = 1.54184)
2Θ range for data collection/°	8.422 to 143.028
Index ranges	-9 ≤ h ≤ 10, -10 ≤ k ≤ 16, -20 ≤ l ≤ 20
Reflections collected	5868
Independent reflections	3534 [R_{int} = 0.0179, R_{sigma} = 0.0265]
Data/restraints/parameters	3534/0/275
Goodness-of-fit on F^2	1.071
Final R indexes [I>=2σ (I)]	R_1 = 0.0332, wR_2 = 0.0855
Final R indexes [all data]	R_1 = 0.0363, wR_2 = 0.0875
Largest diff. peak/hole / e Å^{-3}	0.15/-0.12

General Procedure for Compound 2aal Crystal Preparation:

Compound **2aal** (around 20 mg) was dissolved in DCM-PE (1:1, 10 mL). The single crystal was grown by slow evaporation of solvents at room temperature.

Compound **2aal** was collected at 100 K on a Rigaku Oxford Diffraction Supernova Dual Source, Cu at Zero equipped with an AtlasS2 CCD. The data were collected and processed using CrysAlisPro. The structures were solved by direct methods using Olex2 software, and the non-hydrogen atoms were located from the trial structure and S26 then refined anisotropically with SHELXL-2018 using a full-matrix least squares procedure based on F^2. The weighted R factor, wR and goodness-of-fit S values were obtained based on F^2. The hydrogen atom positions were fixed geometrically at the calculated distances and allowed to ride on their parent atoms. Crystallographic data for the structure reported in this paper have been

deposited at the Cambridge Crystallographic Data Center and allocated with the deposition number: CCDC 2222386 for compound **2aal**, basic information pertaining to crystal parameters and structure refinement are summarized in Figure S13 [12].

Compound 2aal (CCDC 2222386)

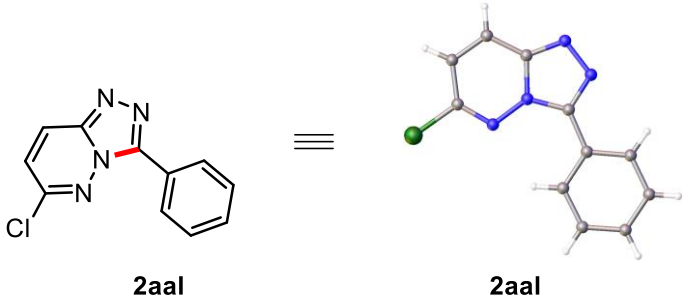


Figure S13: X-ray structure of **2aal** with 50% ellipsoid probability

Empirical formula	$C_{11}H_7ClN_4$
Formula weight	230.66
Temperature/K	293(2)
Crystal system	monoclinic
Space group	$P2_1/c$
a/Å	7.1946(5)
b/Å	11.1720(8)
c/Å	12.8438(9)
α/°	90
β/°	92.899(6)
γ/°	90
Volume/Å^3	1031.04(13)
Z	4
ρ_{calc}g/cm^3	1.486
μ/mm^{-1}	3.075
F(000)	472.0
Crystal size/mm^3	0.3 × 0.3 × 0.3
Radiation	Cu Kα (λ = 1.54184)
2Θ range for data collection/°	10.5 to 142.76
Index ranges	$-7 \le h \le 8$, $-12 \le k \le 13$, $-15 \le l \le 15$
Reflections collected	4548
Independent reflections	1953 [R_{int} = 0.0453, R_{sigma} = 0.0454]
Data/restraints/parameters	1953/0/146
Goodness-of-fit on F^2	1.144
Final R indexes [I>=2σ (I)]	R_1 = 0.0806, wR_2 = 0.2273

Final R indexes [all data]	R_1 = 0.0870, wR_2 = 0.2336
Largest diff. peak/hole / e $Å^{-3}$	0.62/-0.47

12. Reference

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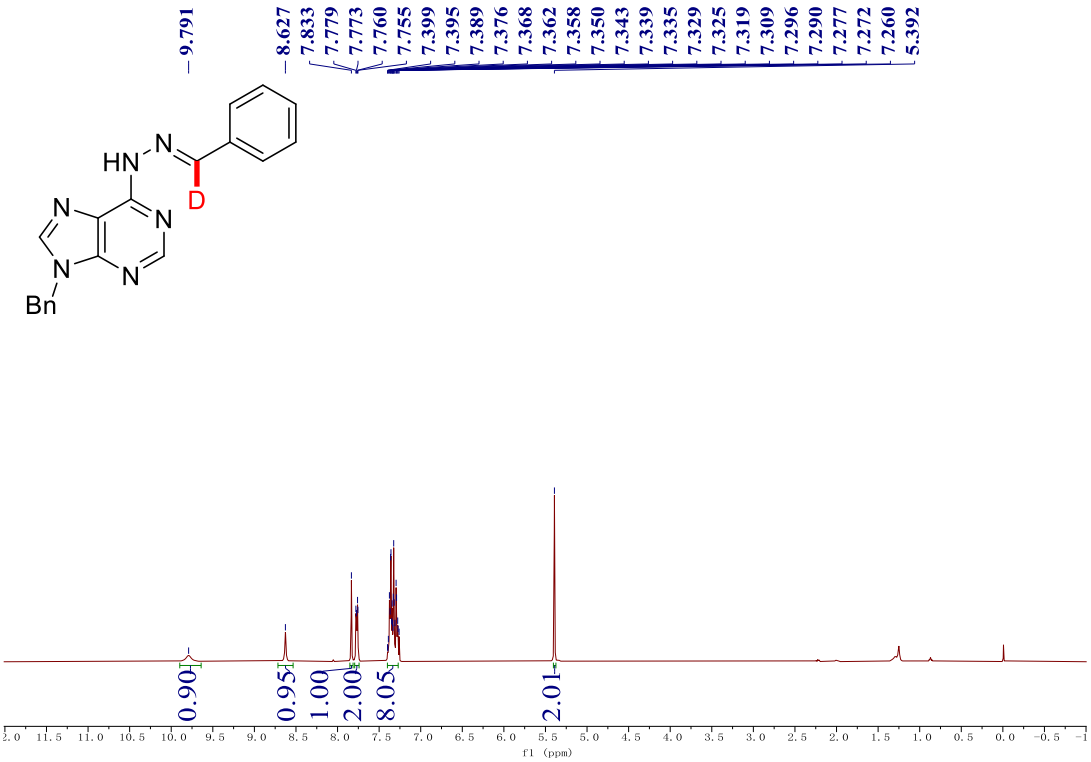
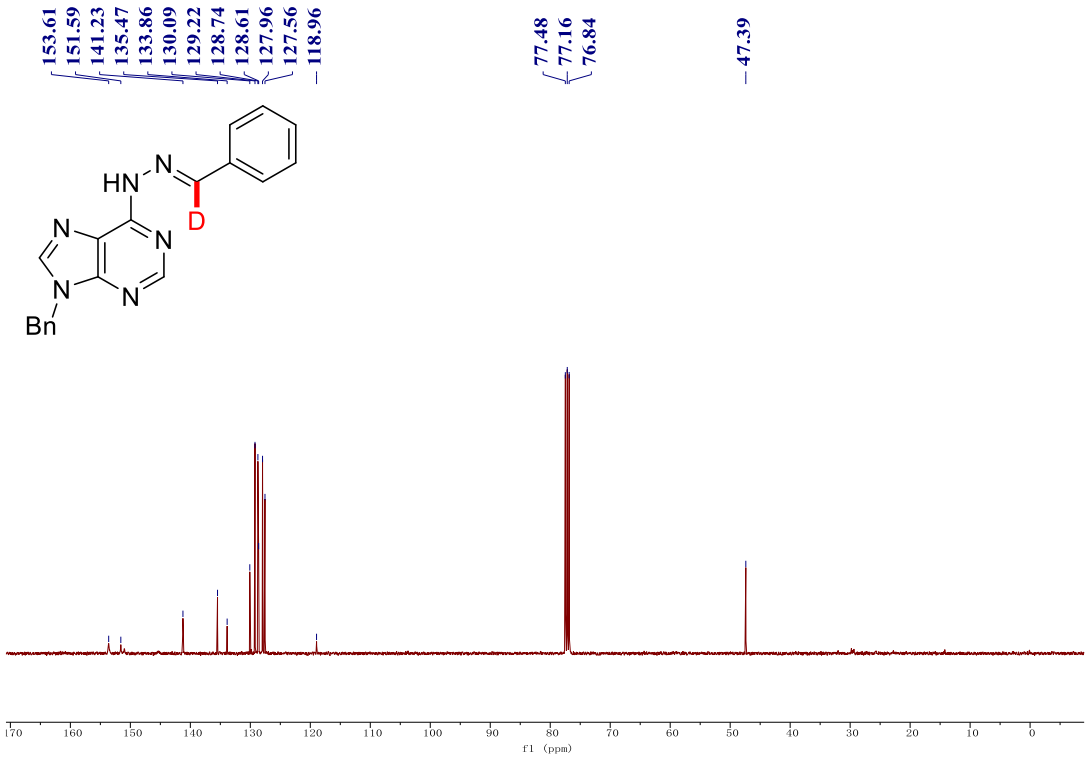
13 NMR Spectra

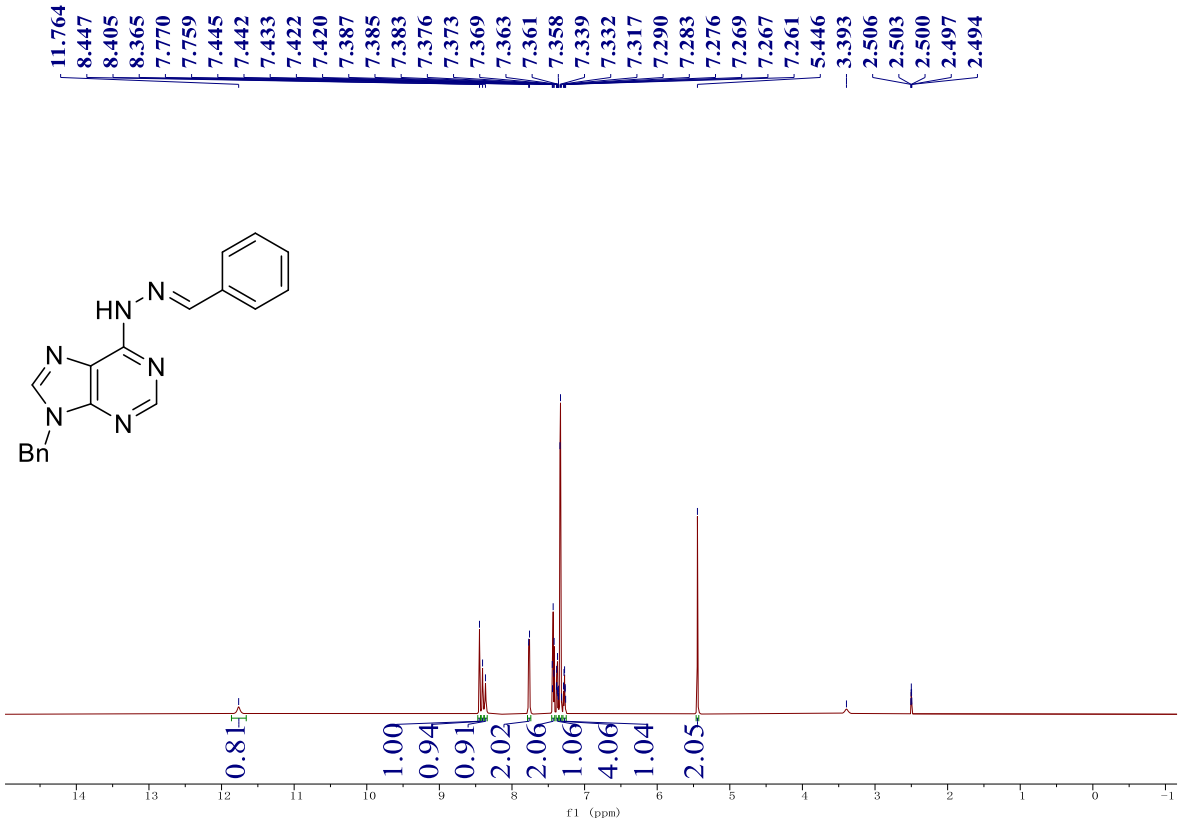
^{1}H NMR Spectrum of [D]-1a at 25 °C ($CDCl_3$, 400 MHz)



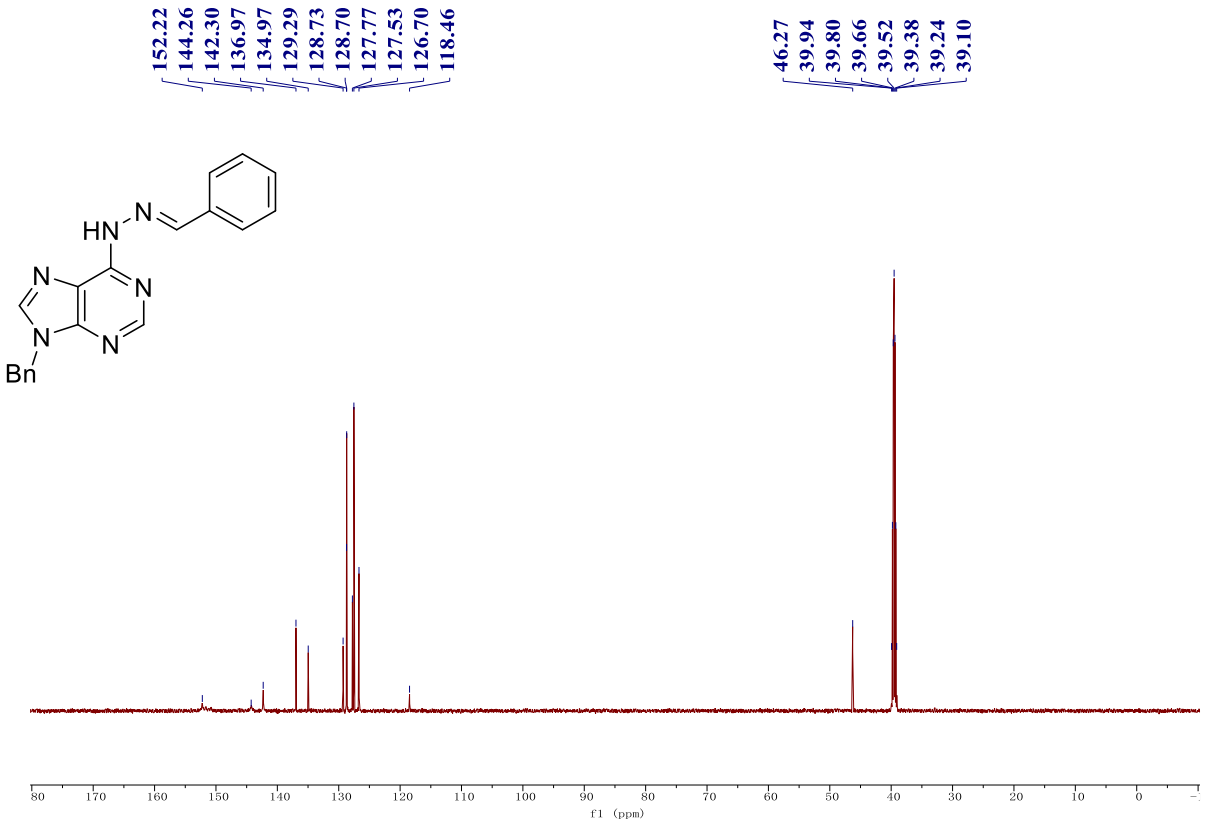
^{13}C NMR Spectrum of [D]-1a at 25 °C ($CDCl_3$, 100 MHz)



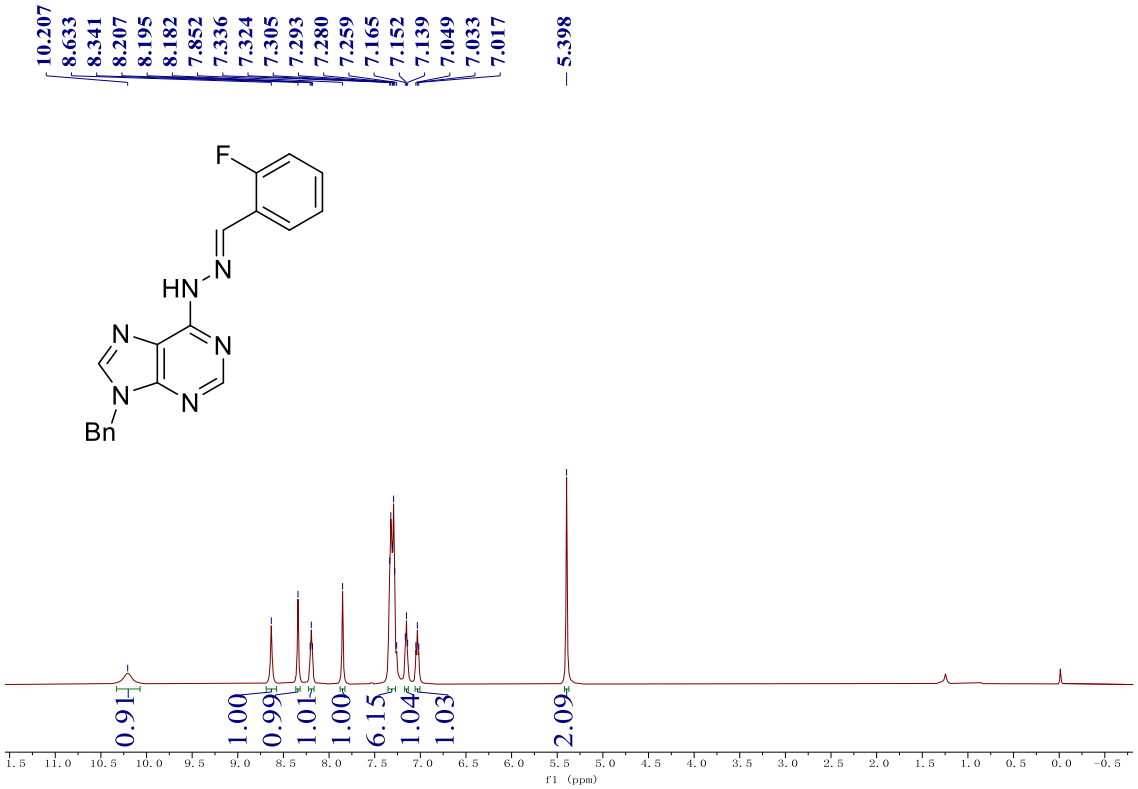
^{1}H NMR Spectrum of 1a at 25 °C (DMSO-d_6, 600 MHz)



^{13}C NMR Spectrum of 1a at 25 °C (DMSO-d_6, 150 MHz)



^{1}H NMR Spectrum of 1b at 25 ^{o}C ($CDCl_3$, 600 MHz)



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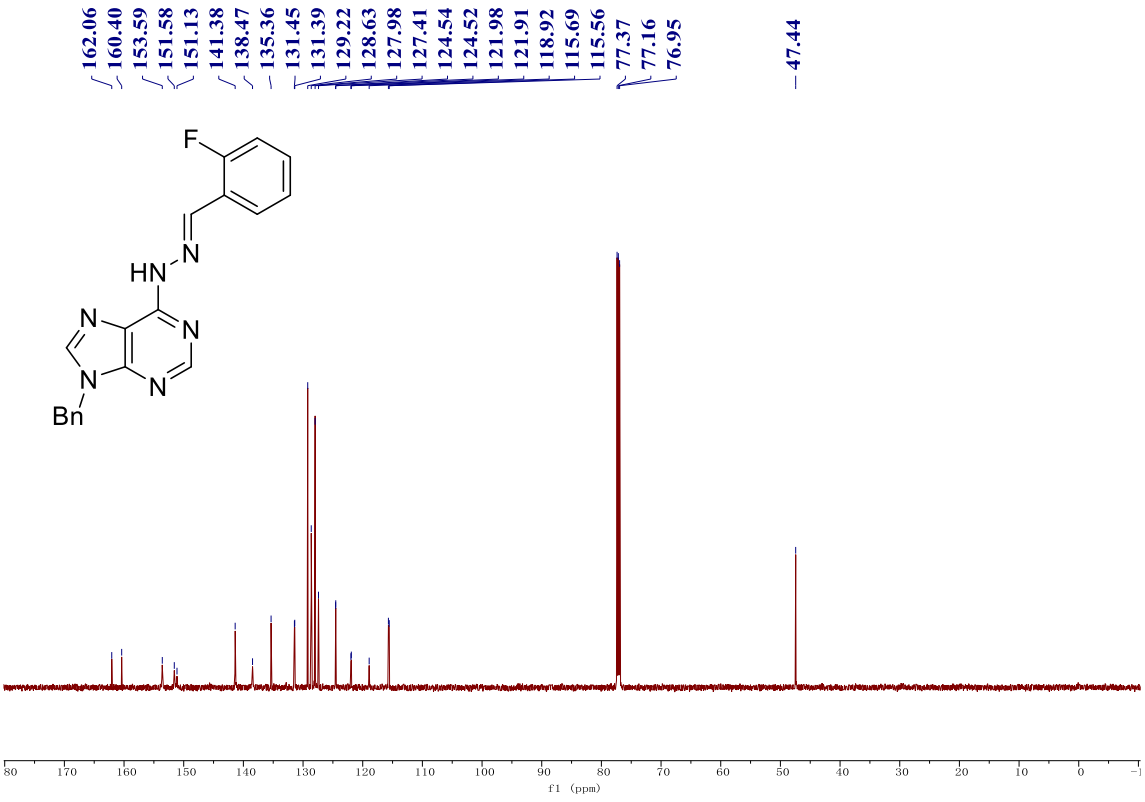
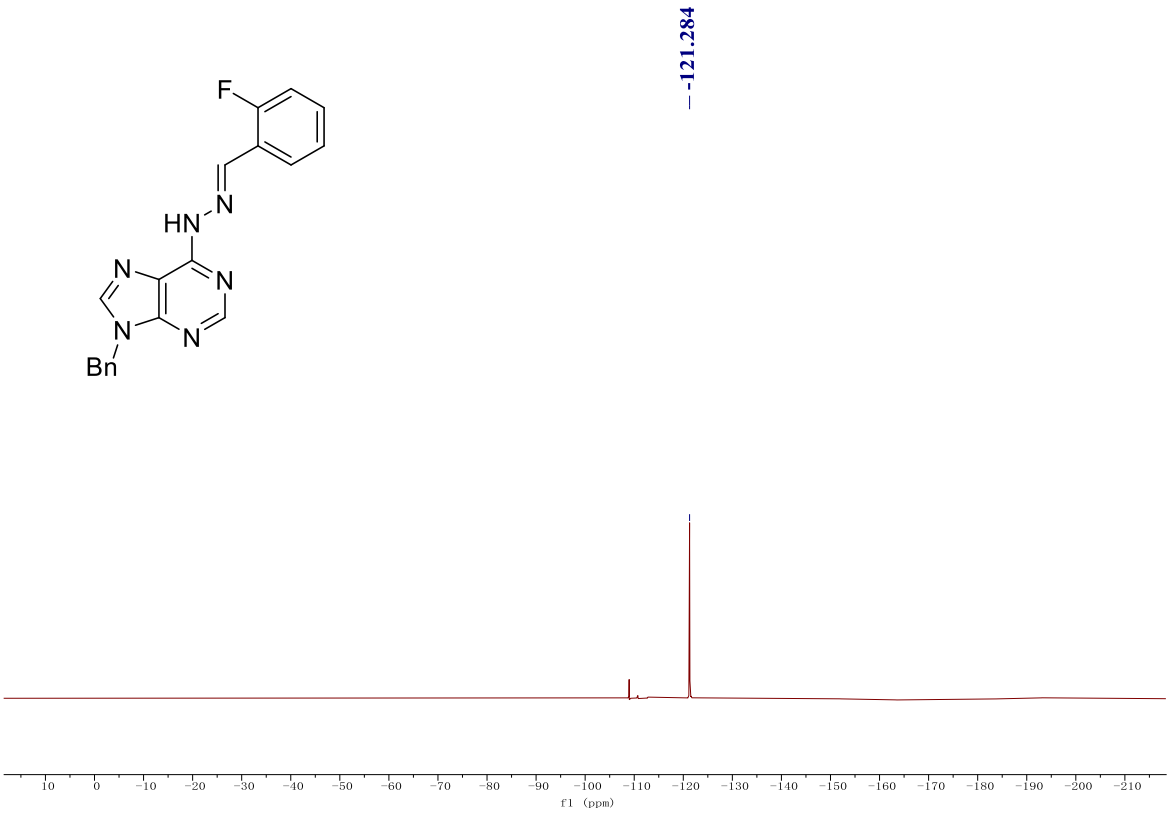
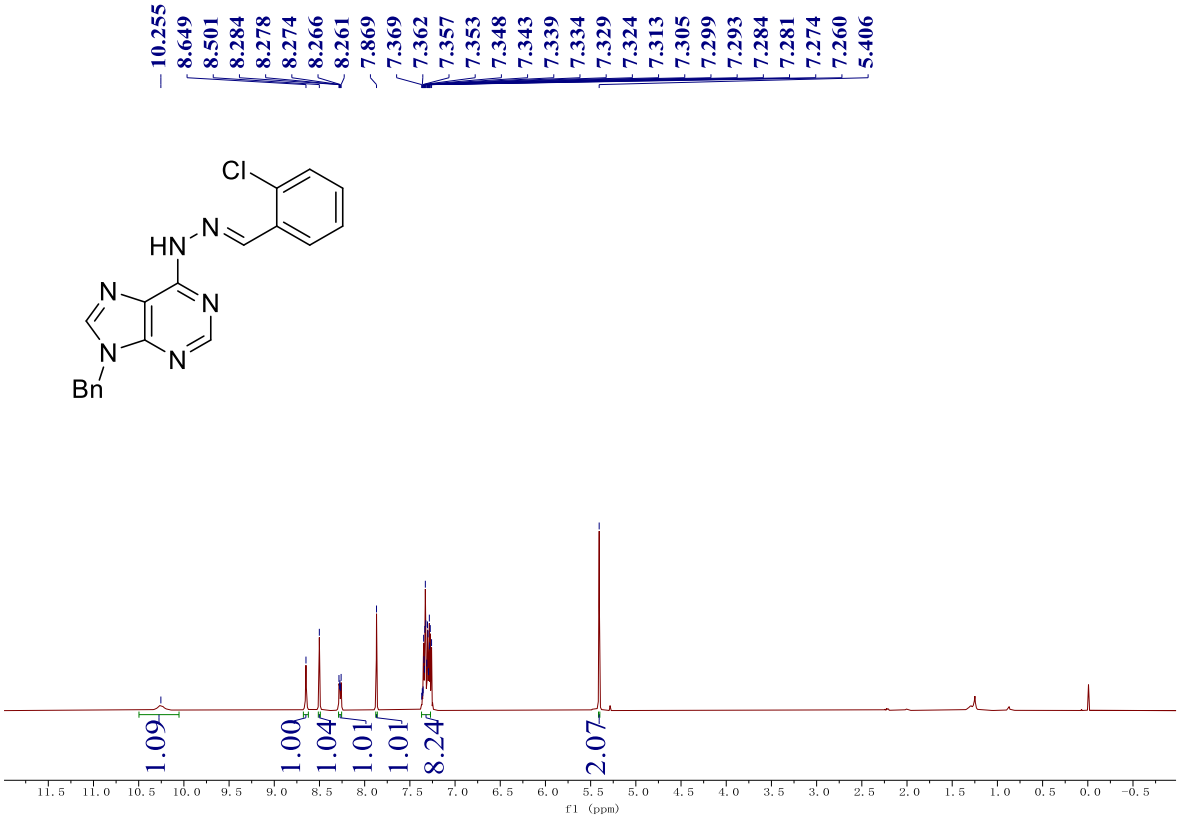


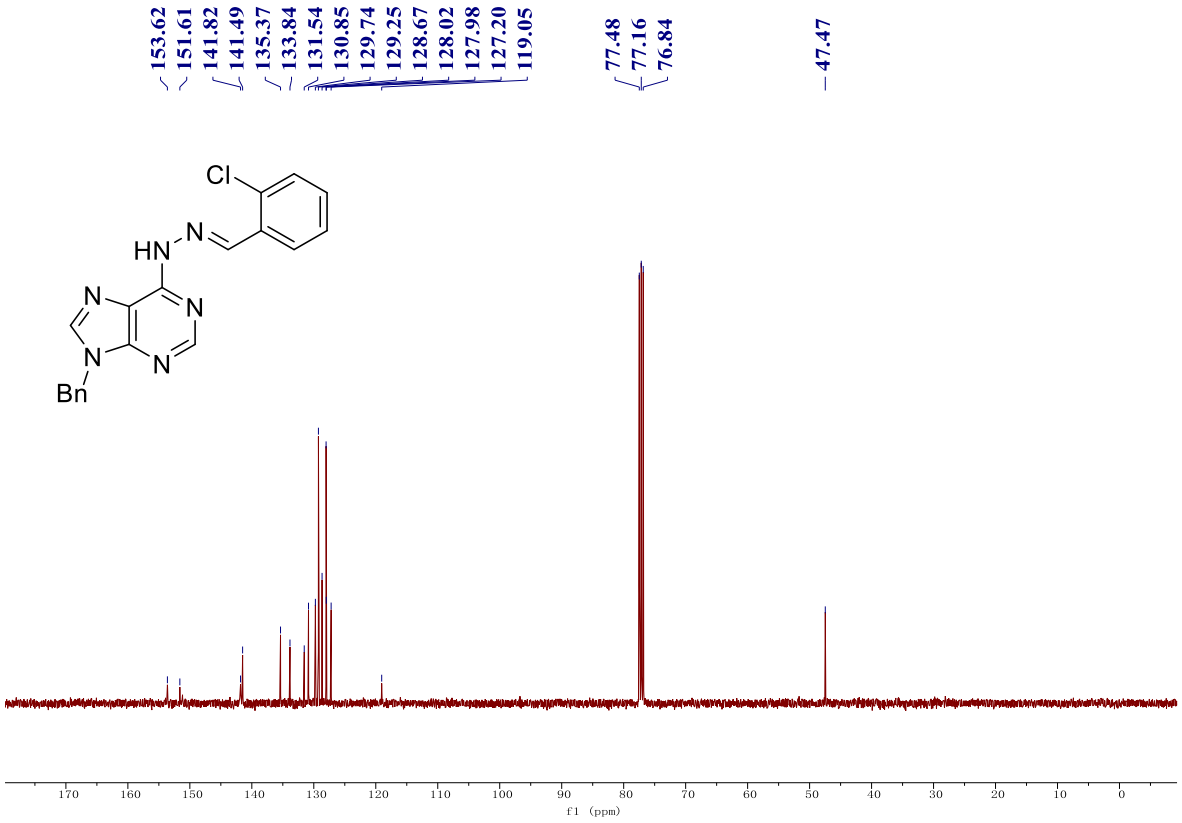
^{19}F NMR Spectrum of 1b at 25 °C ($CDCl_3$, 565 MHz)



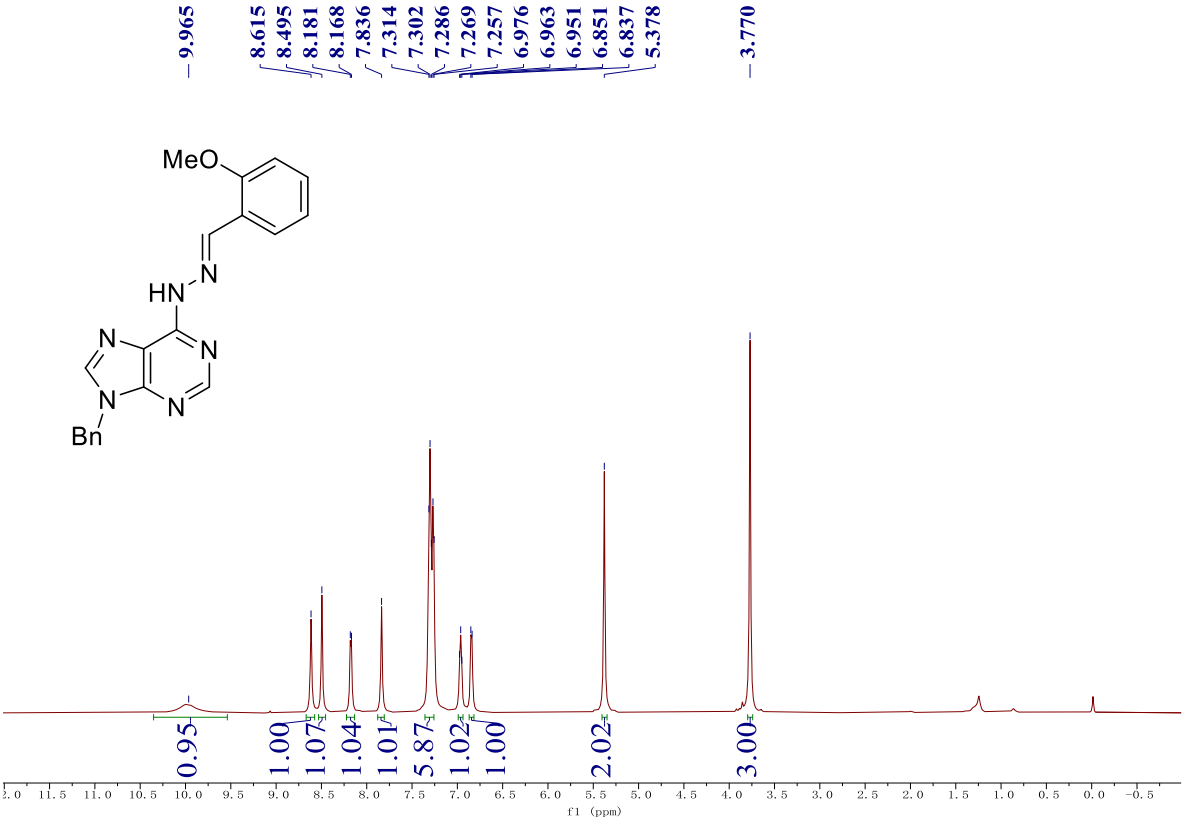
^{1}H NMR Spectrum of 1c at 25 °C ($CDCl_3$, 400 MHz)



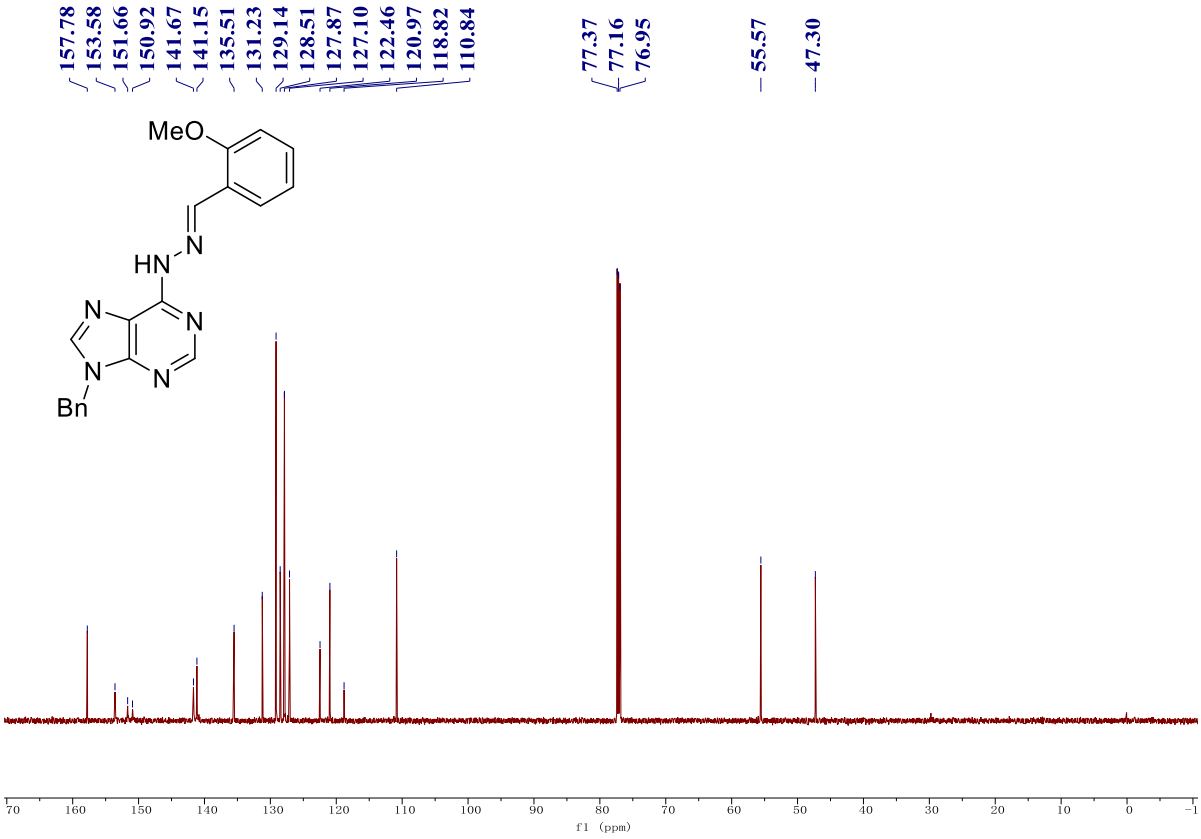
^{13}C NMR Spectrum of 1c at 25 °C ($CDCl_3$, 100 MHz)



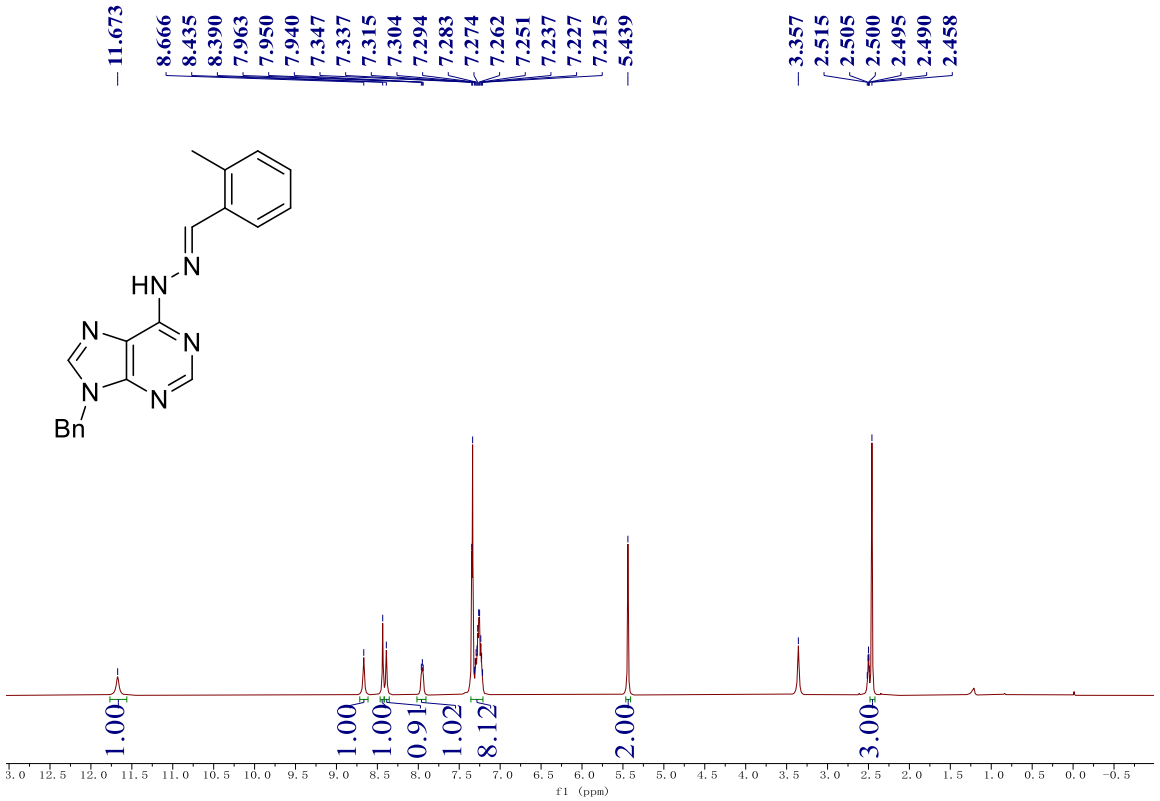
^{1}H NMR Spectrum of 1d at 25 °C ($CDCl_3$, 600 MHz)



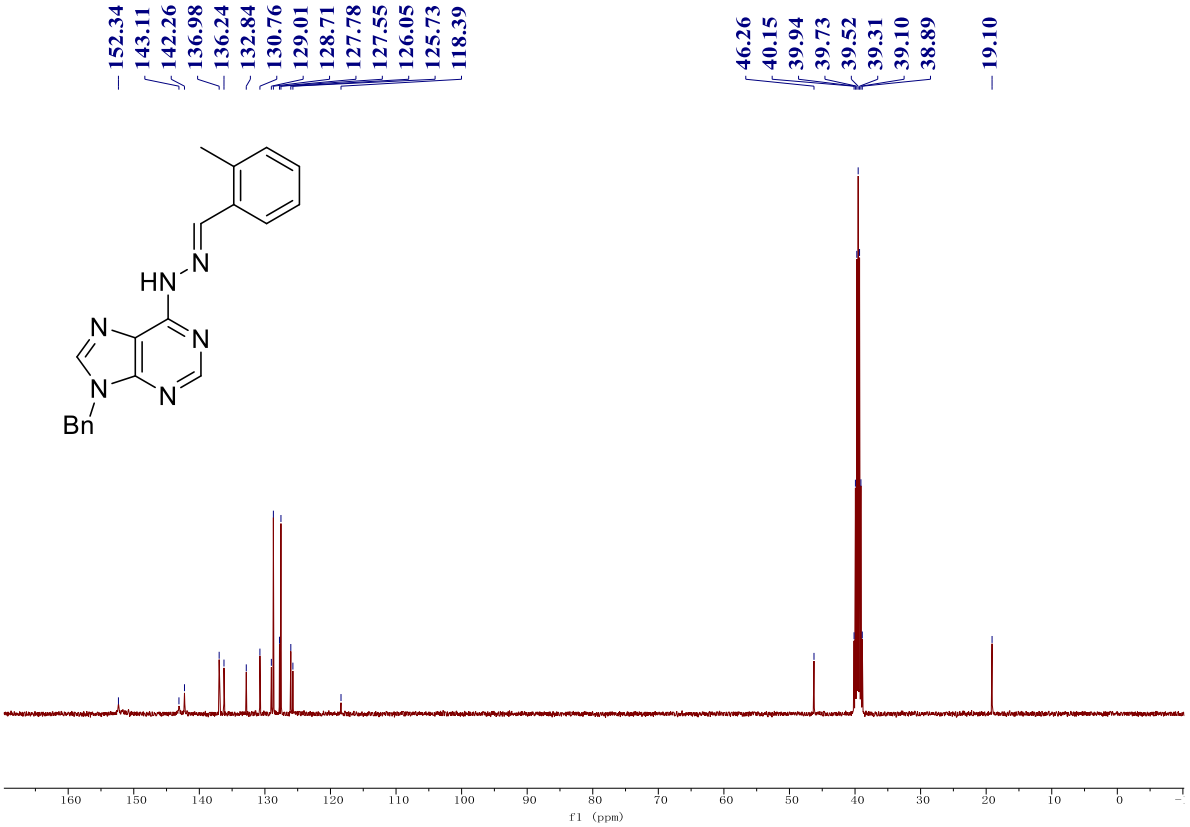
^{13}C NMR Spectrum of 1d at 25 °C ($CDCl_3$, 150 MHz)



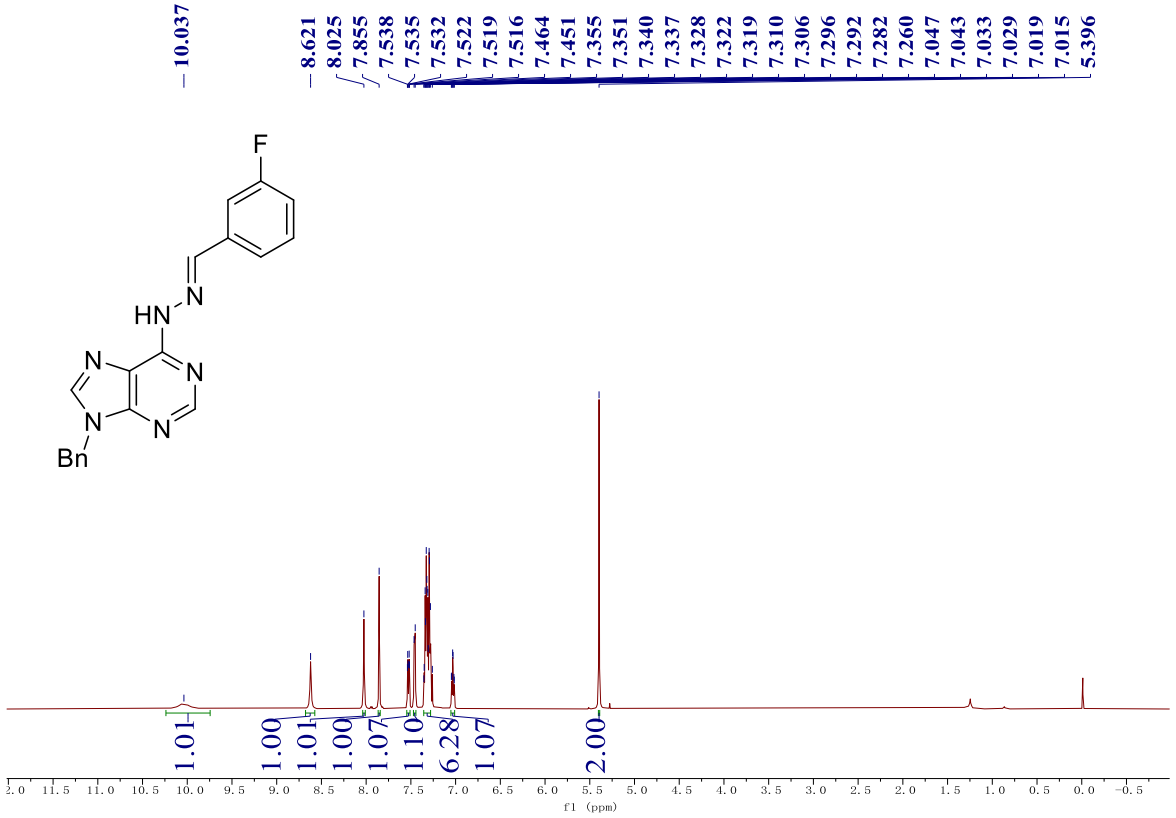
^{1}H NMR Spectrum of 1e at 25 °C (DMSO-d_6, 400 MHz)



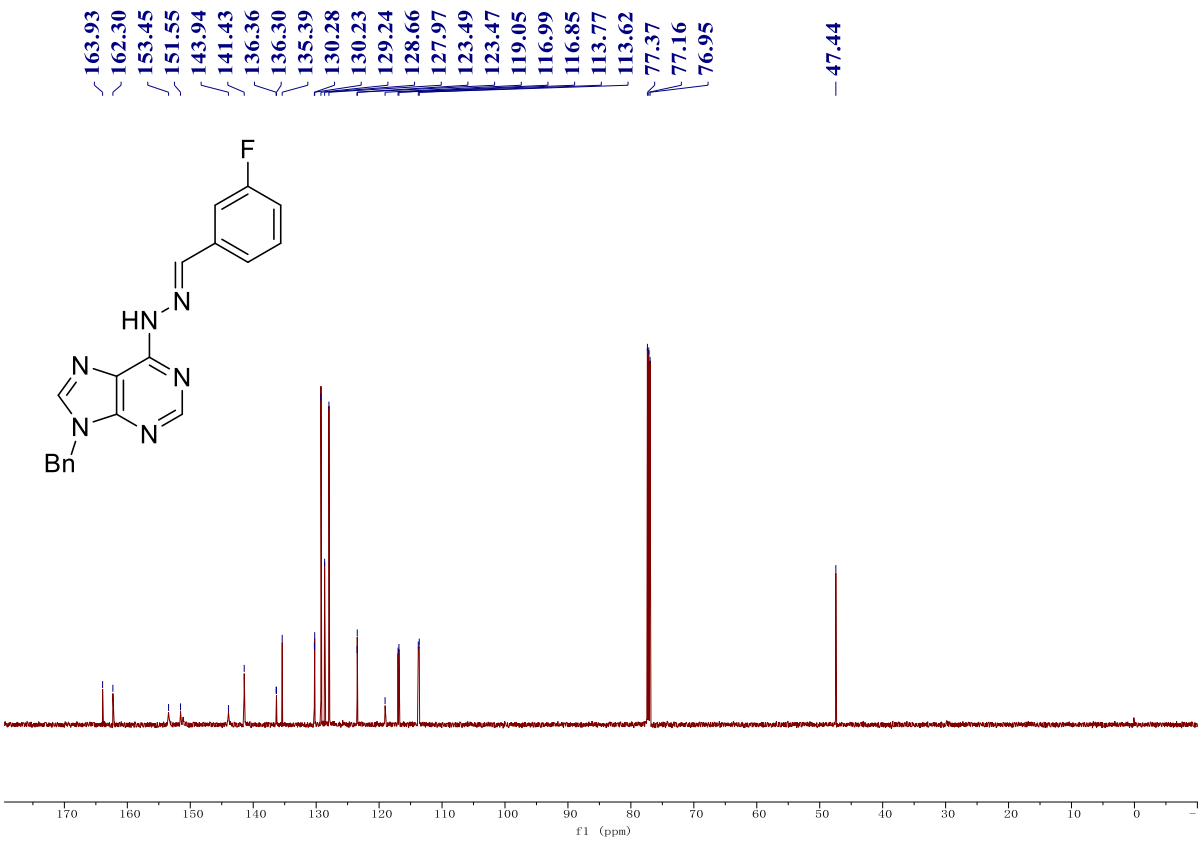
^{13}C NMR Spectrum of 1e at 25 °C (DMSO-d_6, 100 MHz)



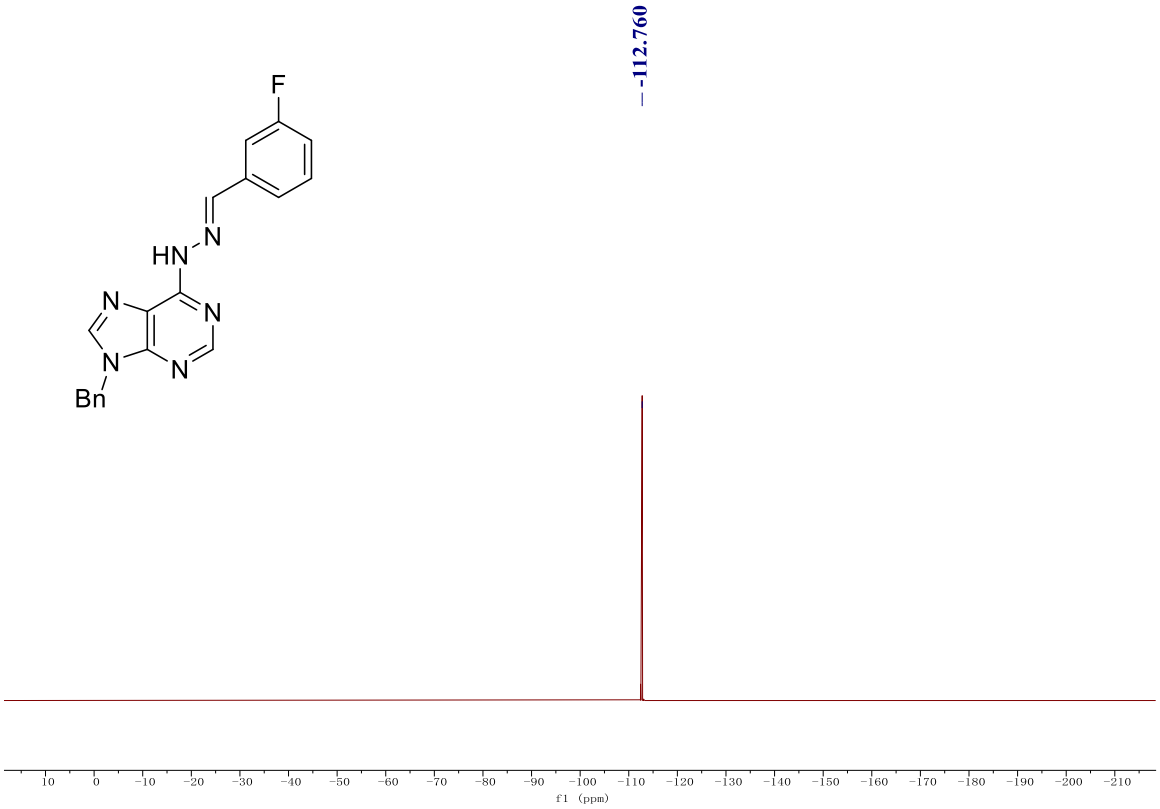
^{1}H NMR Spectrum of 1f at 25 °C ($CDCl_3$, 600 MHz)



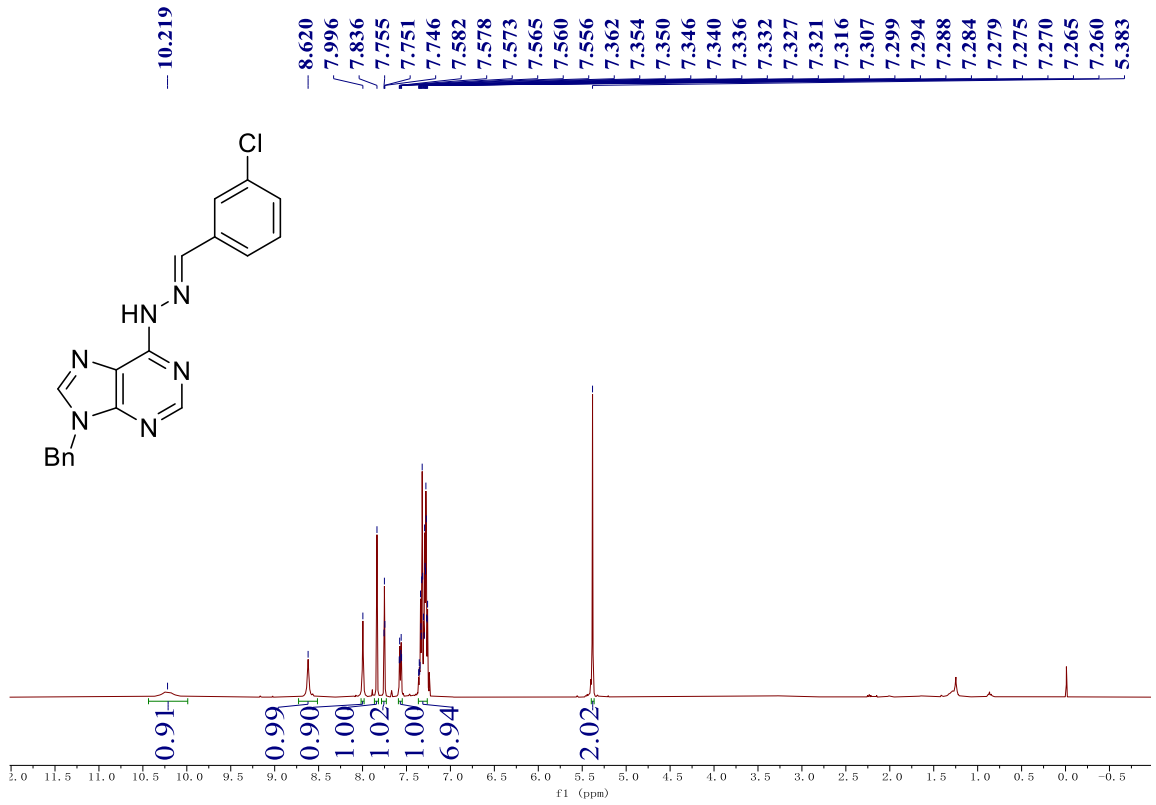
^{13}C NMR Spectrum of 1f at 25 °C ($CDCl_3$, 150 MHz)



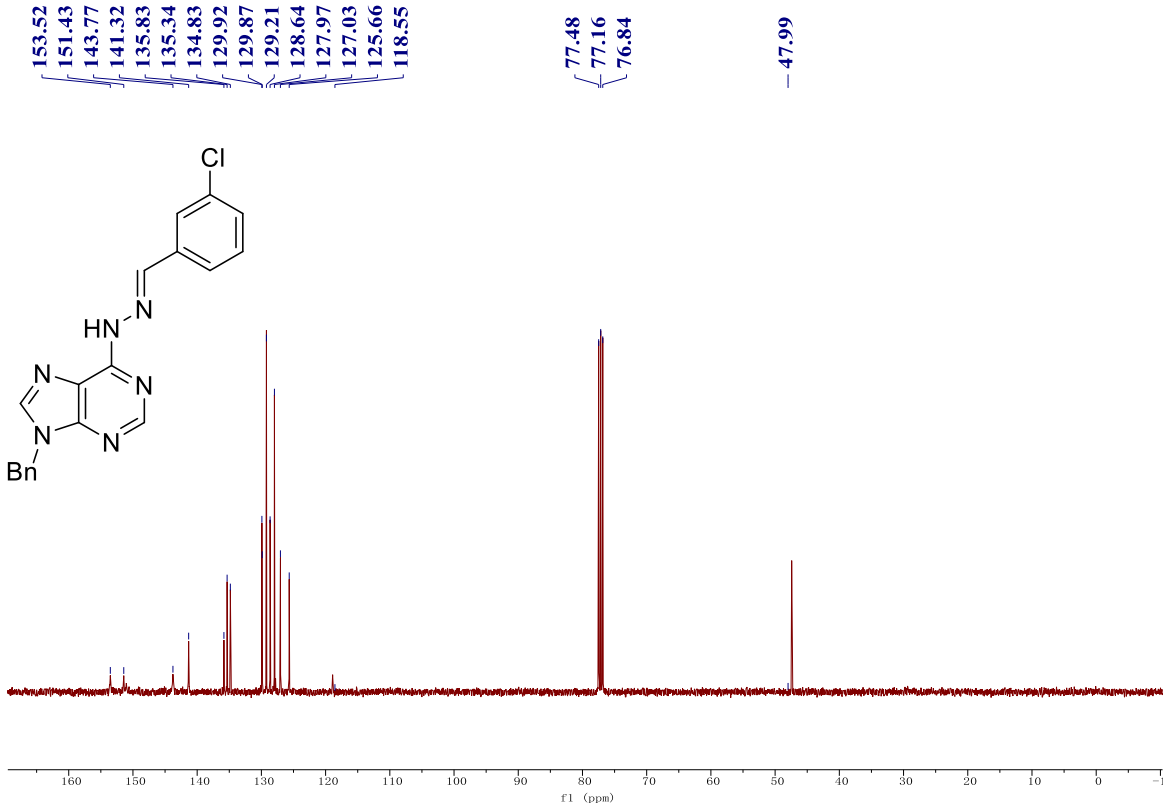
^{19}F NMR Spectrum of 1f at 25 °C ($CDCl_3$, 565 MHz)



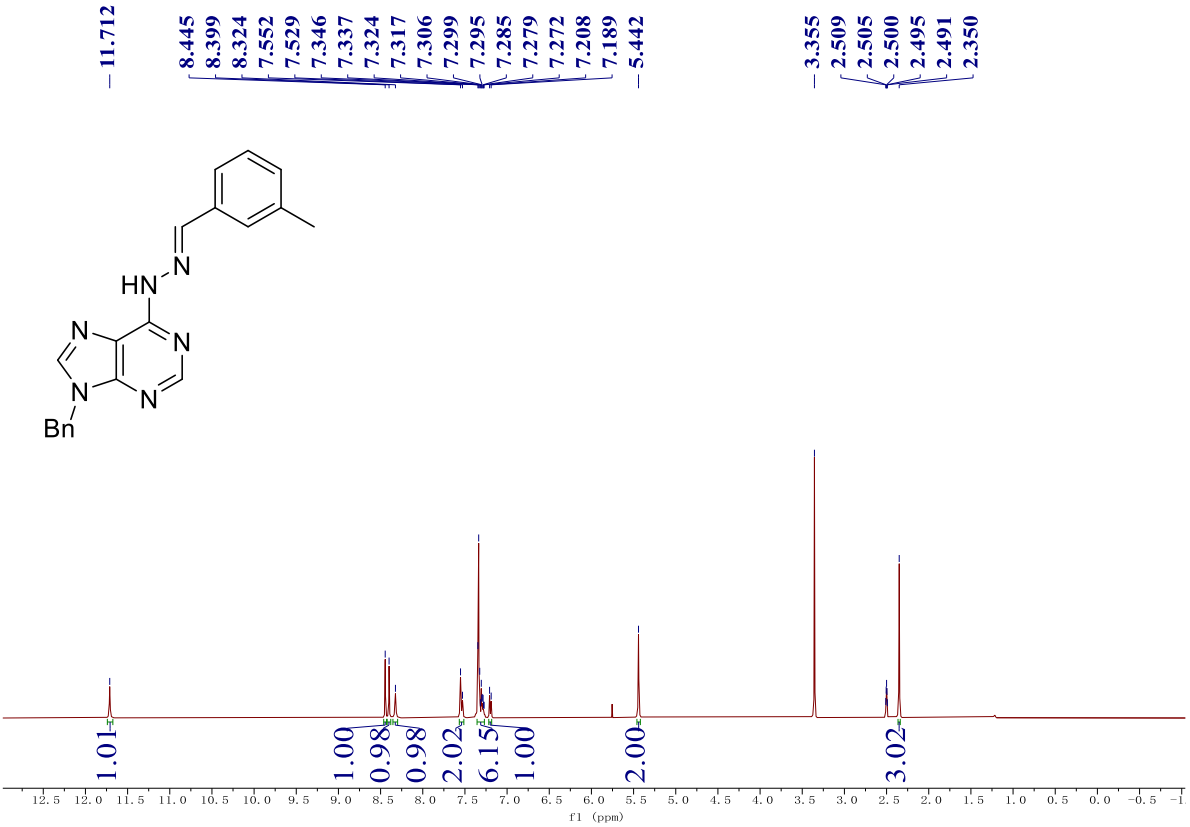
^{1}H NMR Spectrum of 1g at 25 °C ($CDCl_3$, 400 MHz)



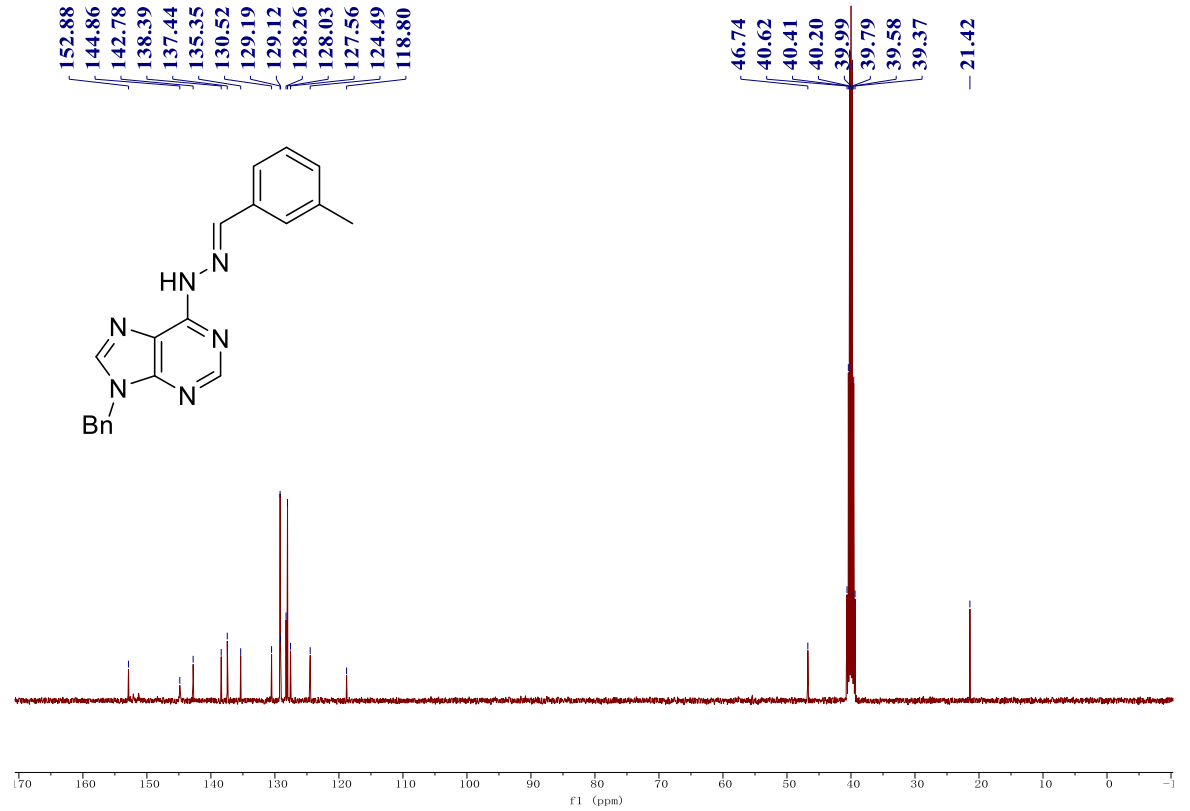
^{13}C NMR Spectrum of 1g at 25 °C ($CDCl_3$, 100 MHz)



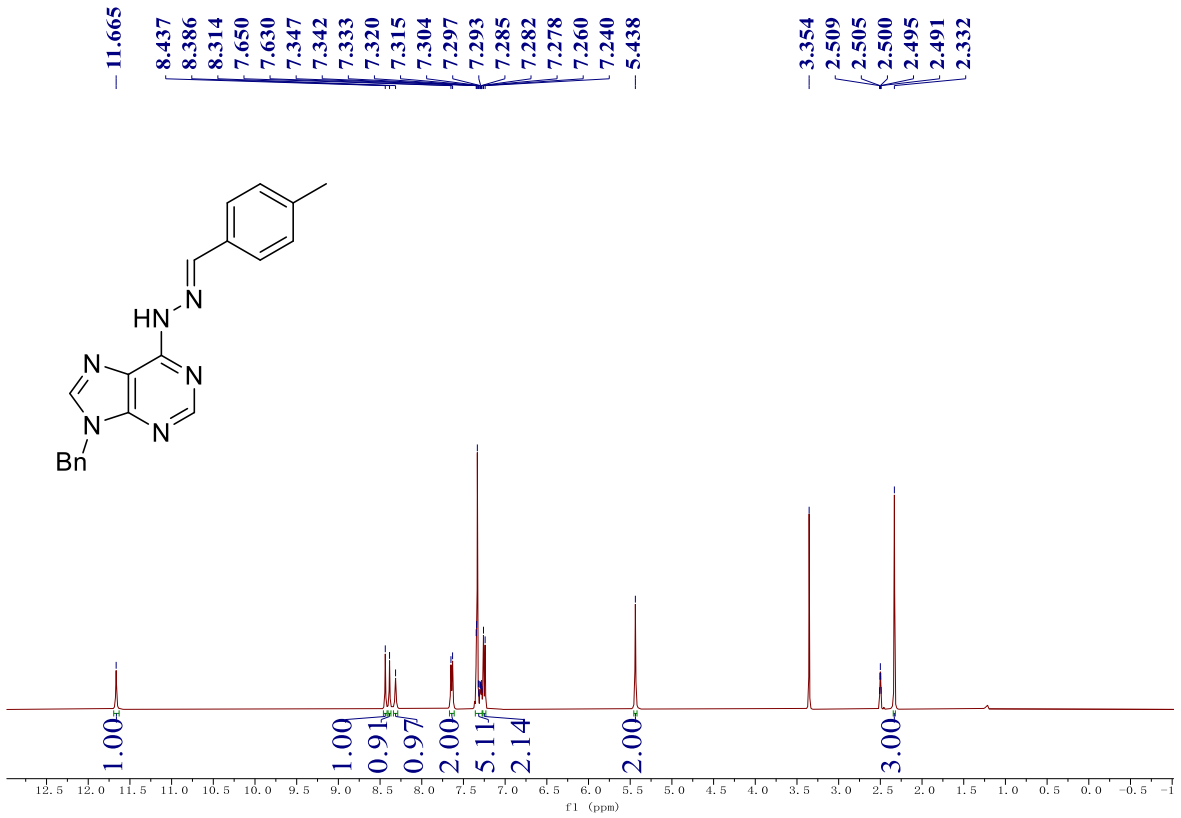
^{1}H NMR Spectrum of 1h at 25 °C (DMSO-d_6, 400 MHz)



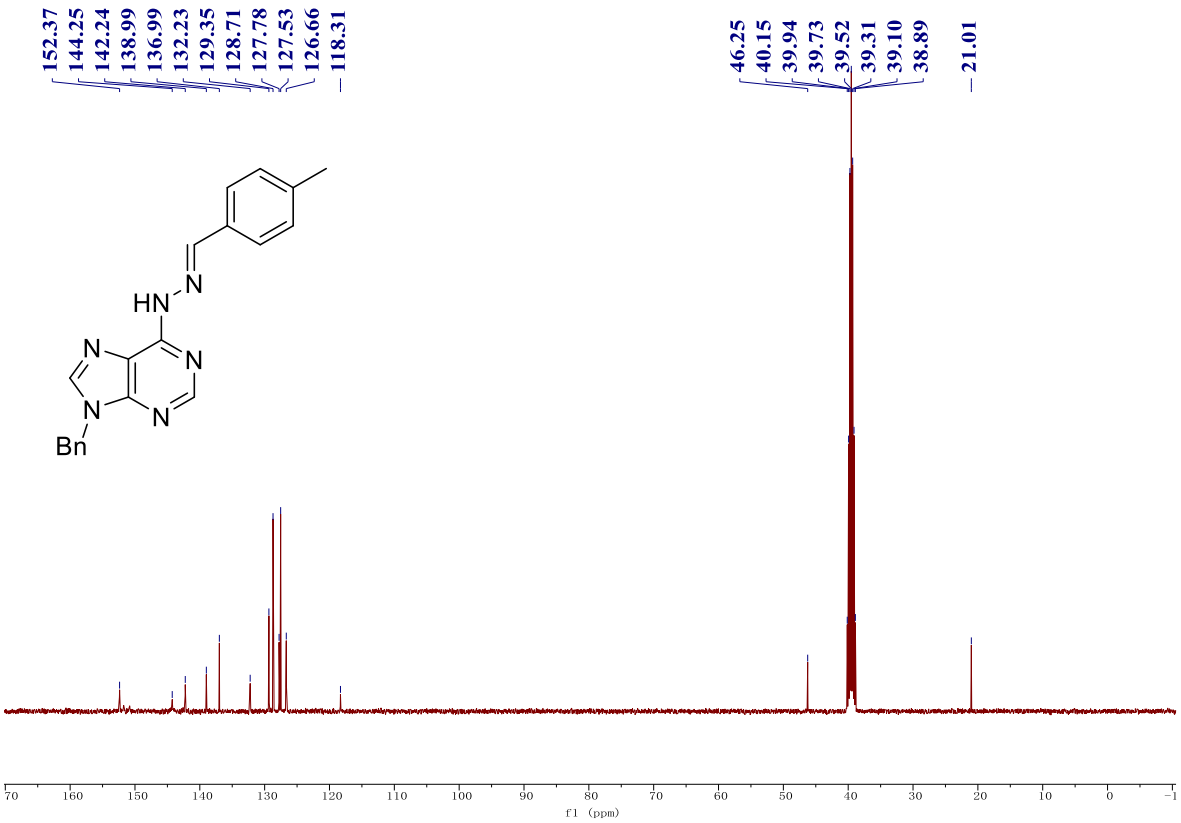
^{13}C NMR Spectrum of 1h at 25 °C (DMSO-d_6, 100 MHz)



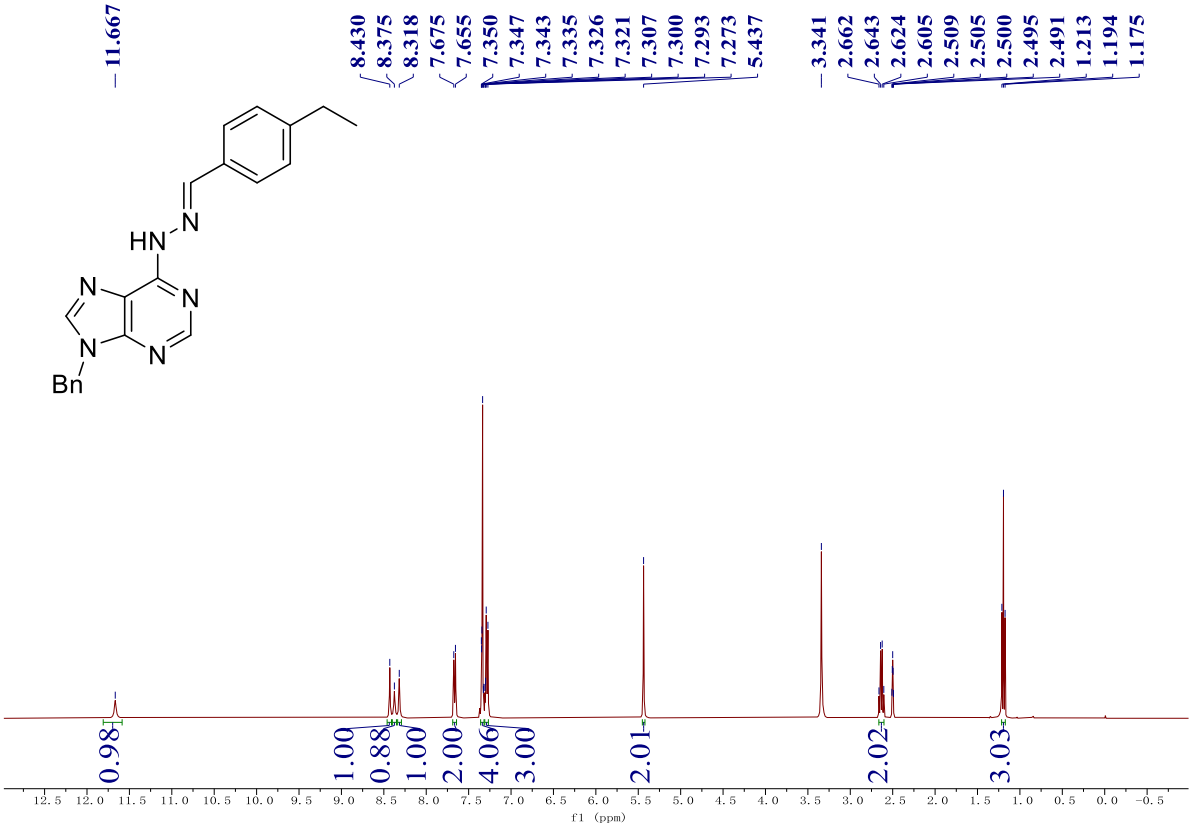
^{1}H NMR Spectrum of 1i at 25 °C (DMSO-d_6, 400 MHz)



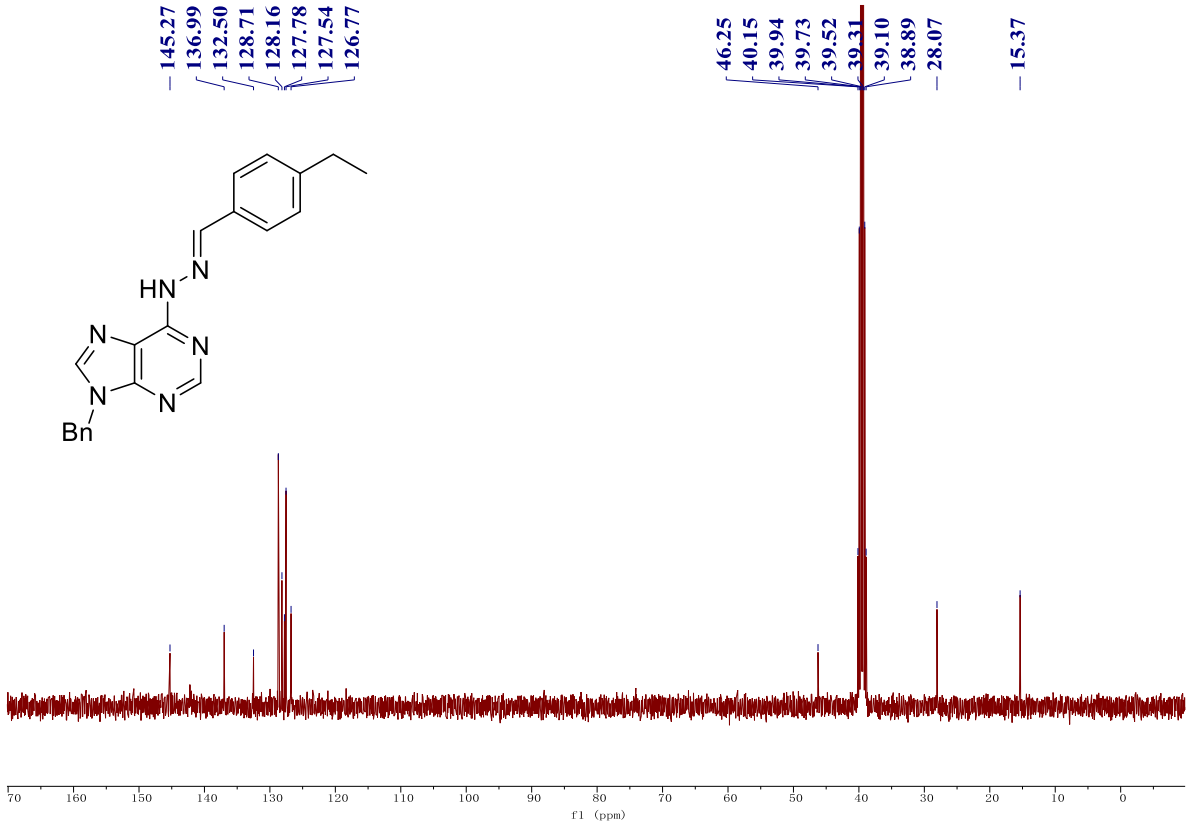
^{13}C NMR Spectrum of 1i at 25 °C (DMSO-d_6, 100 MHz)



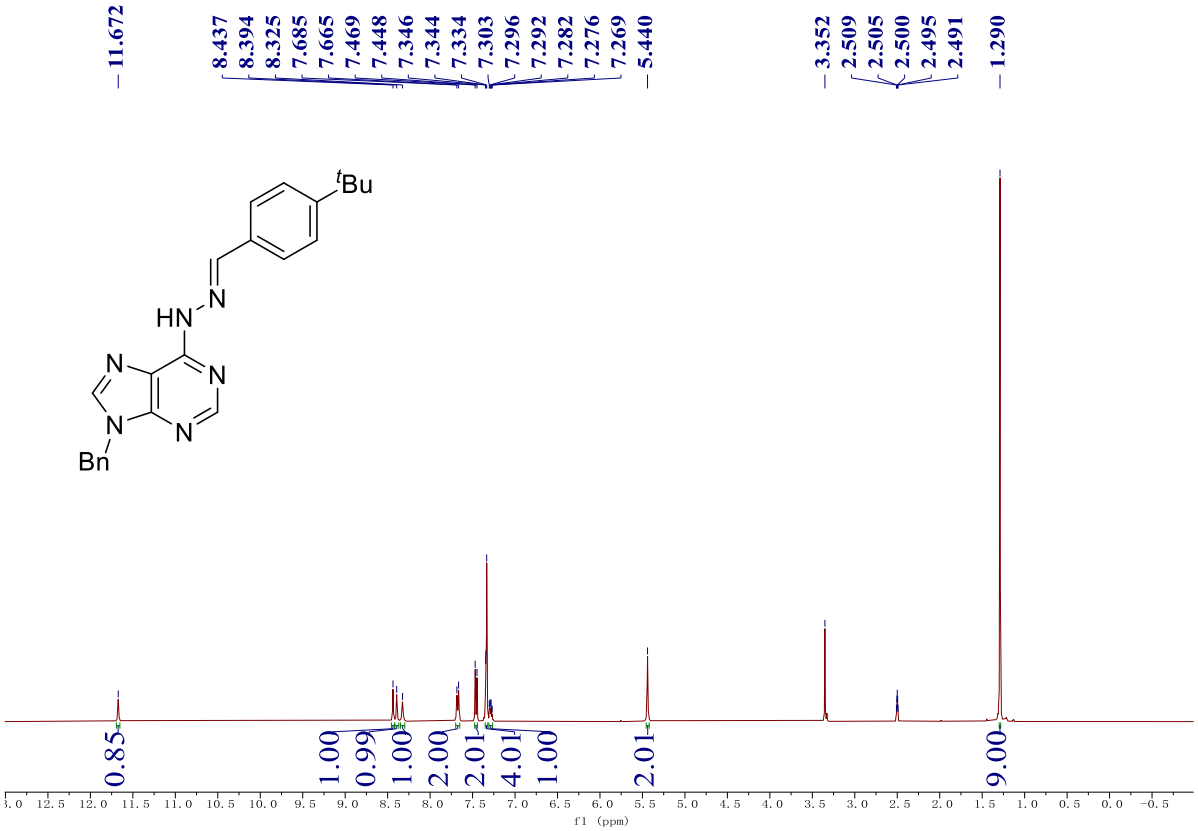
^{1}H NMR Spectrum of 1j at 25 °C (DMSO-d_6, 400 MHz)



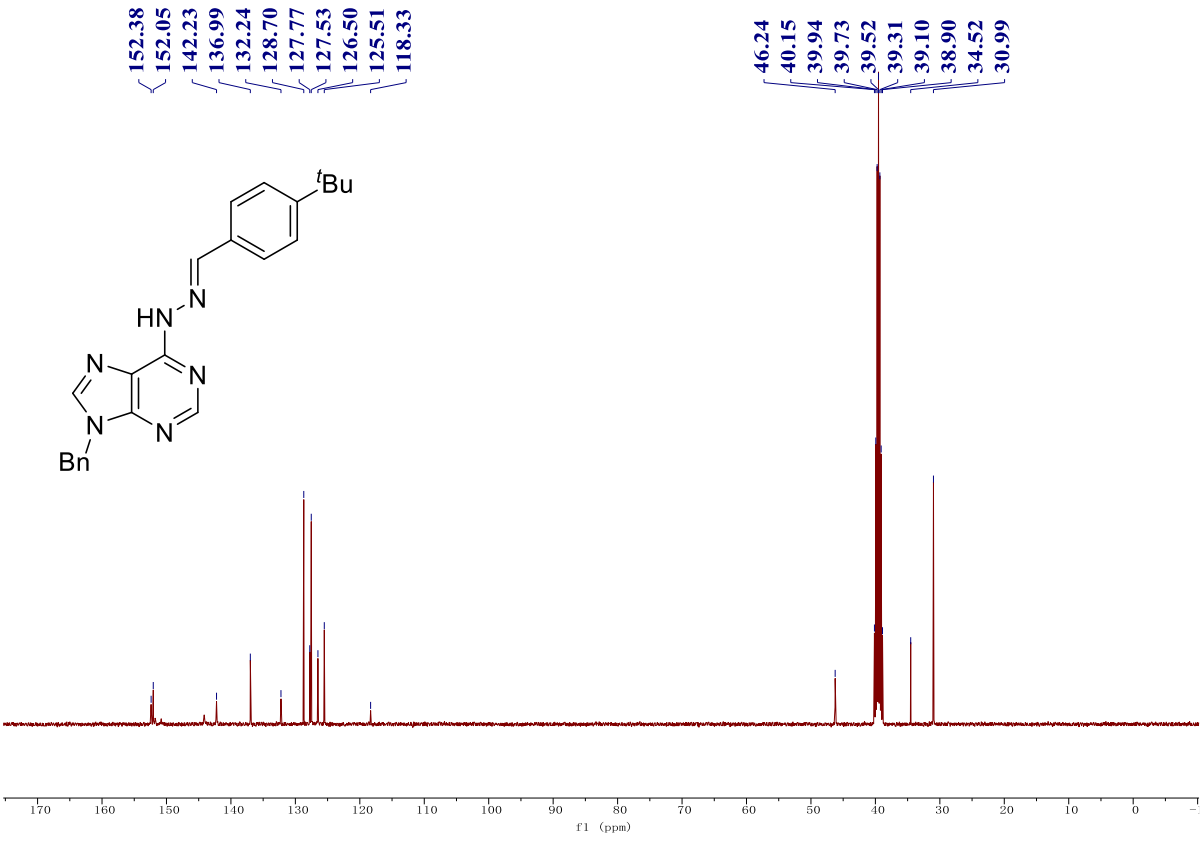
^{13}C NMR Spectrum of 1j at 25 °C (DMSO-d_6, 100 MHz)



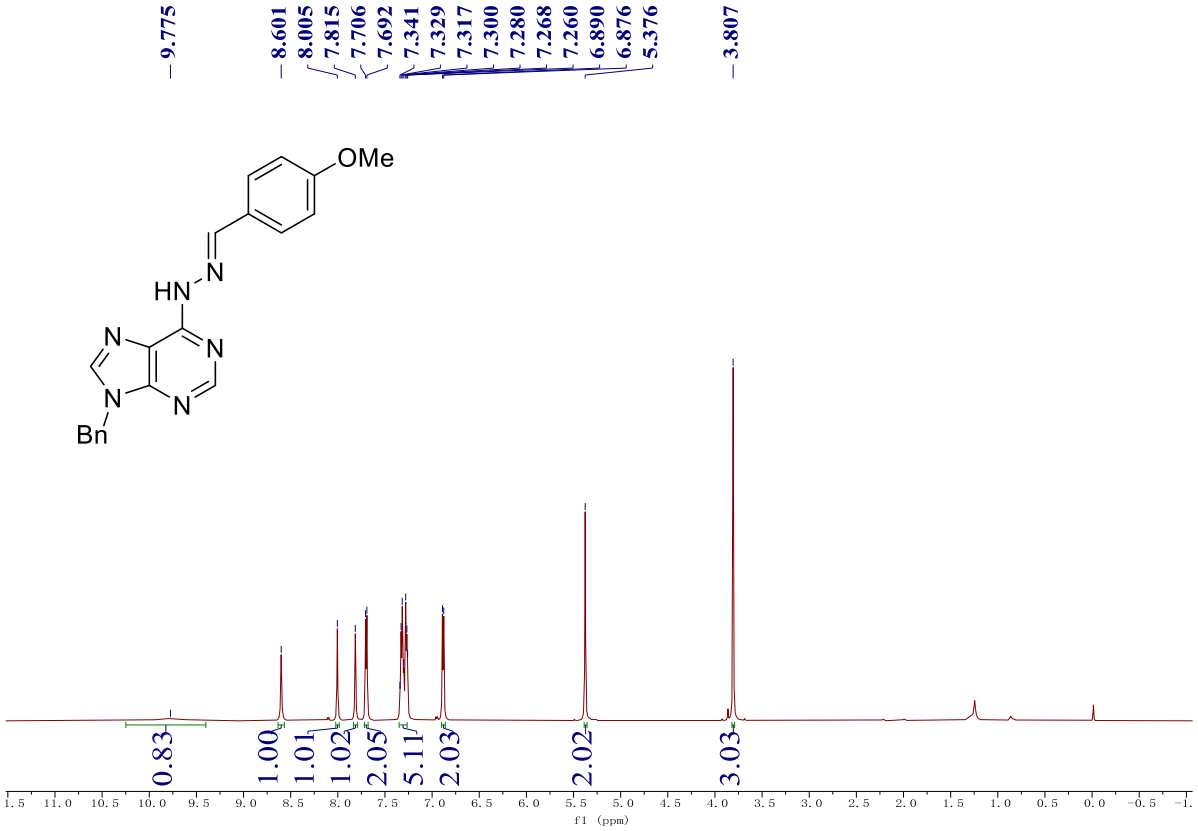
^{1}H NMR Spectrum of 1k at 25 °C (DMSO-d_6, 400 MHz)



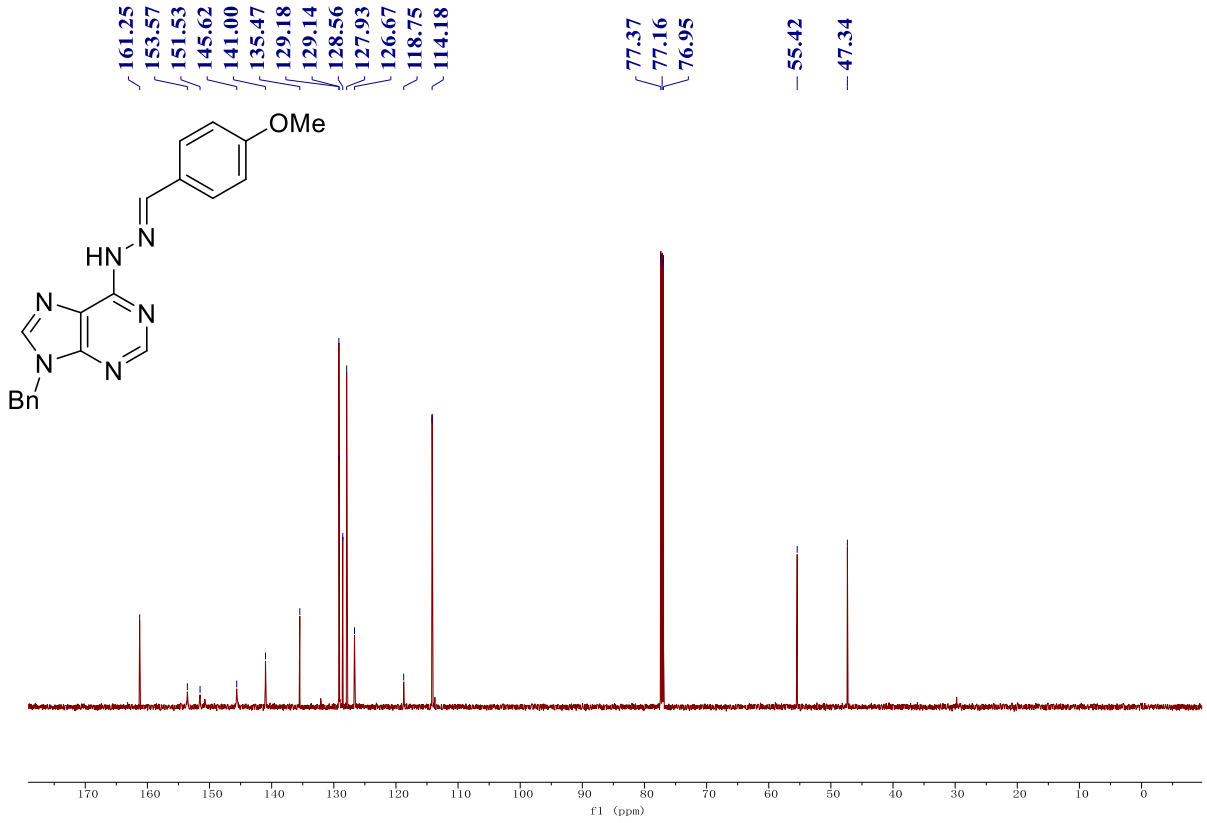
^{13}C NMR Spectrum of 1k at 25 °C (DMSO-d_6, 100 MHz)



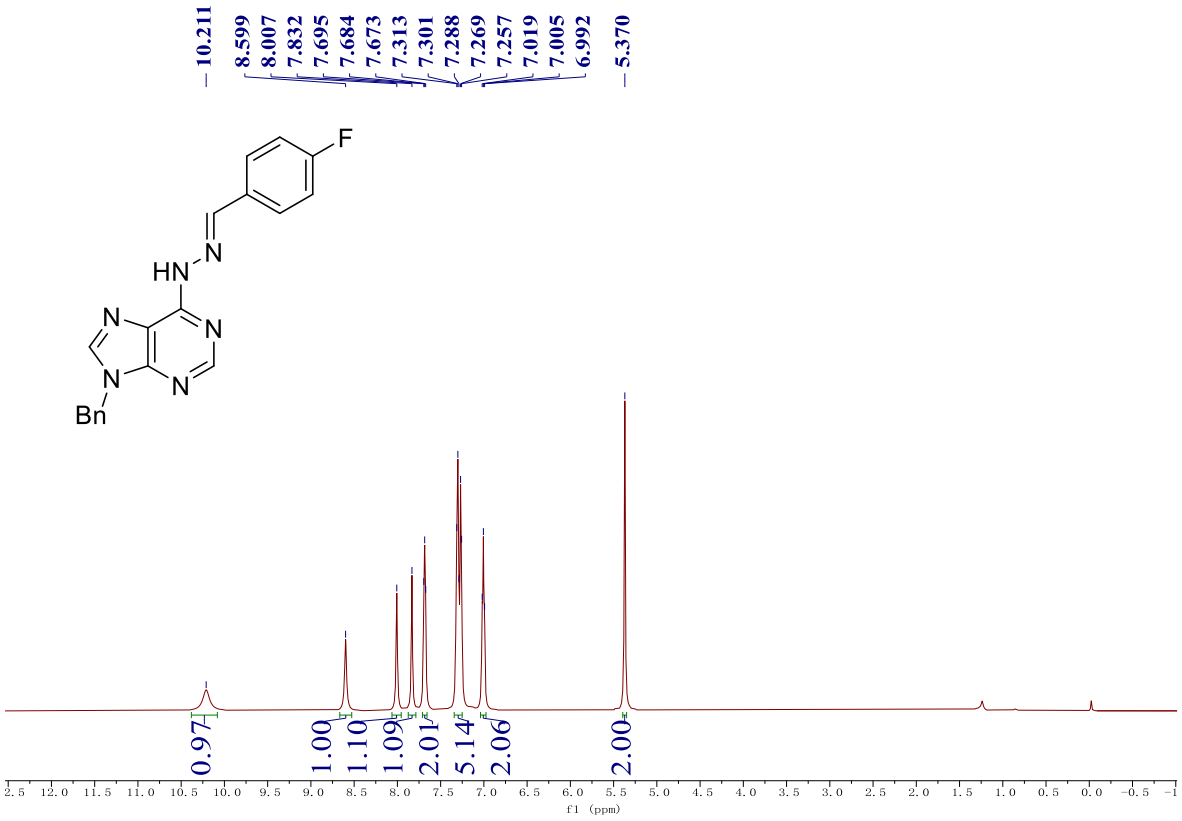
^{1}H NMR Spectrum of 1l at 25 °C ($CDCl_3$, 600 MHz)



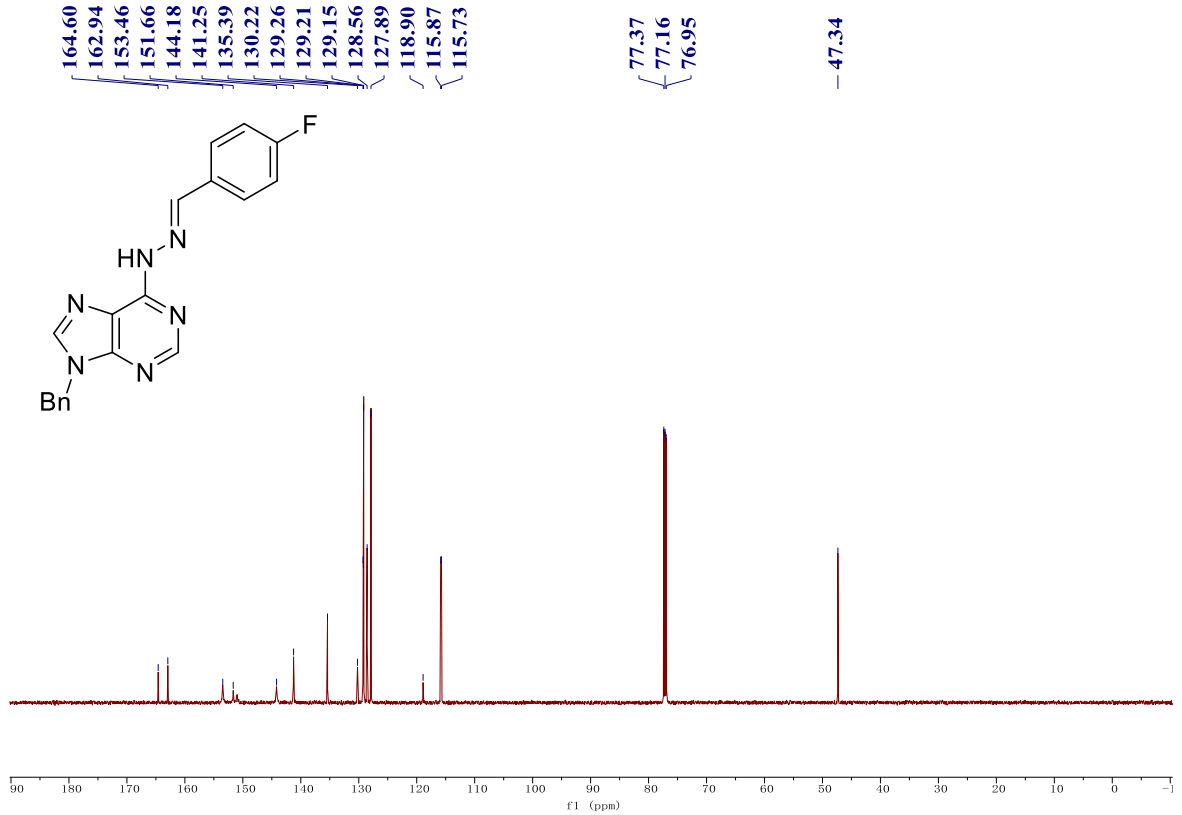
^{13}C NMR Spectrum of 1l at 25 °C ($CDCl_3$, 150 MHz)



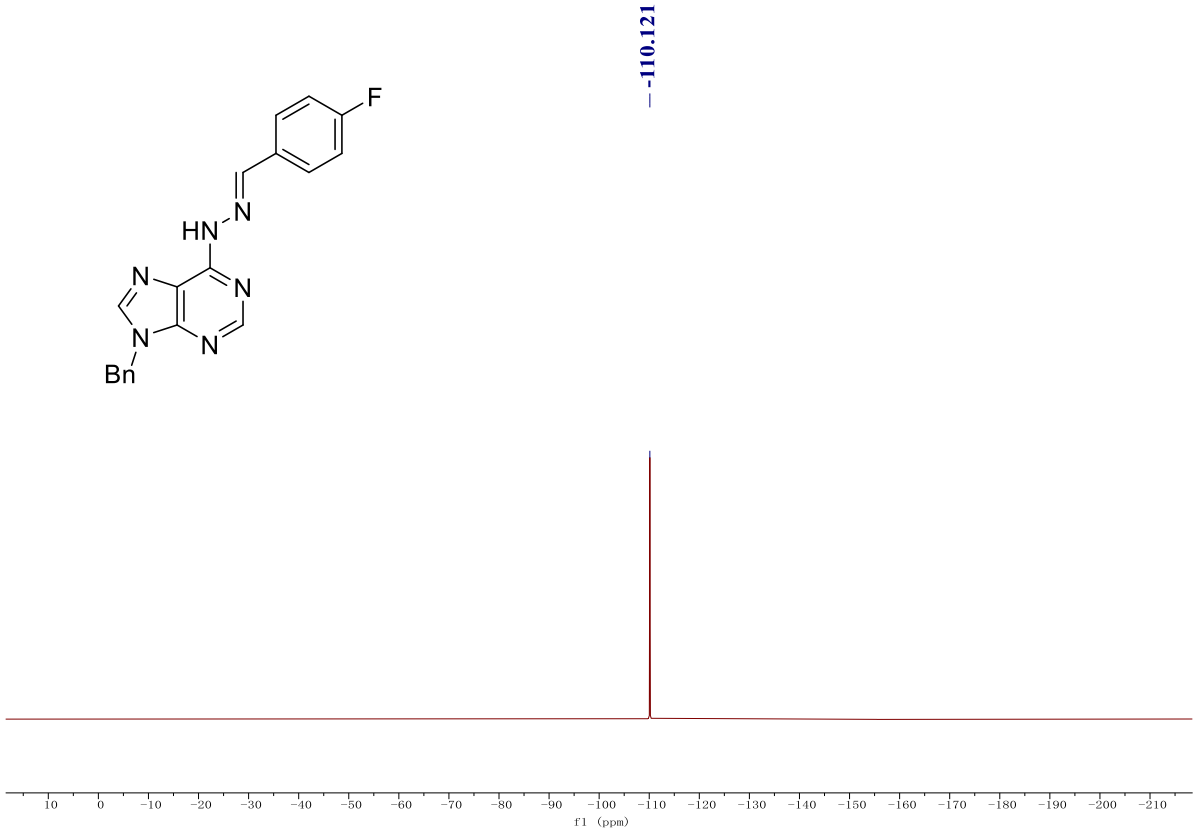
^{1}H NMR Spectrum of 1m at 25 ^{o}C ($CDCl_3$, 600 MHz)



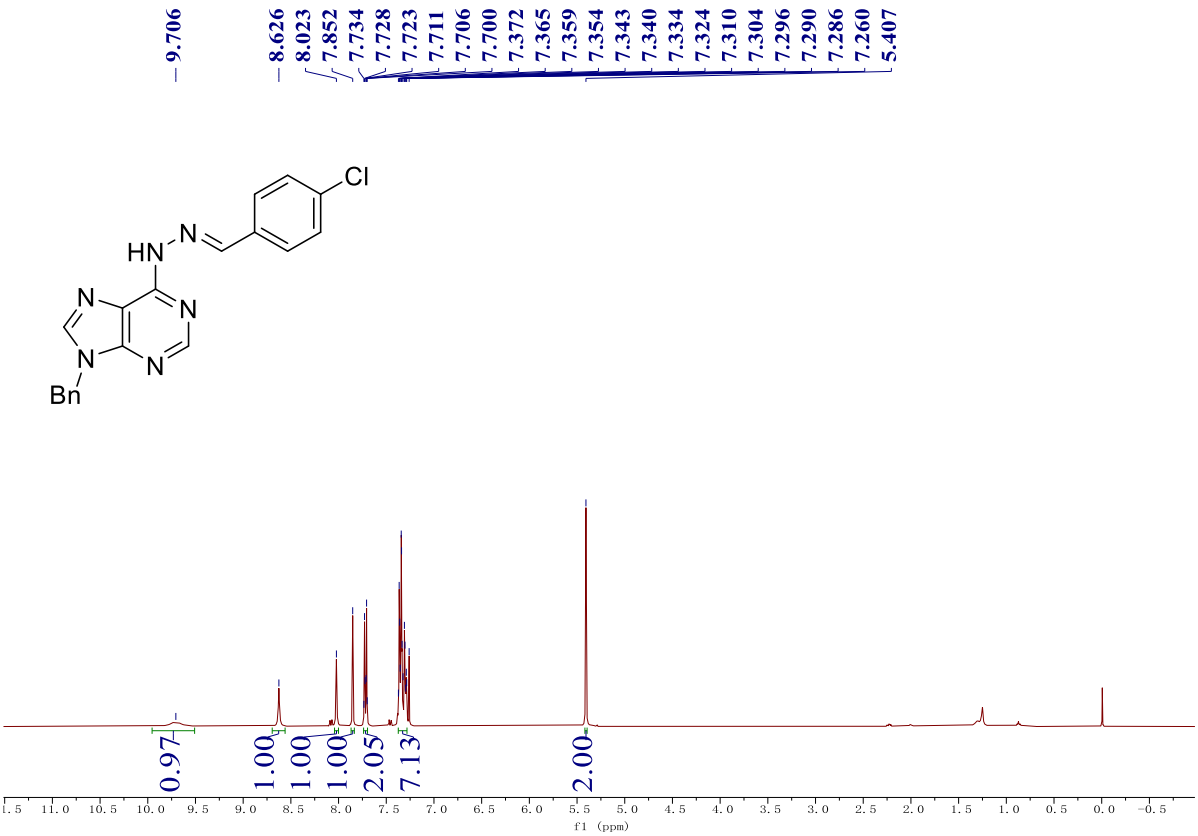
^{13}C NMR Spectrum of 1m at 25 ^{o}C ($CDCl_3$, 150 MHz)



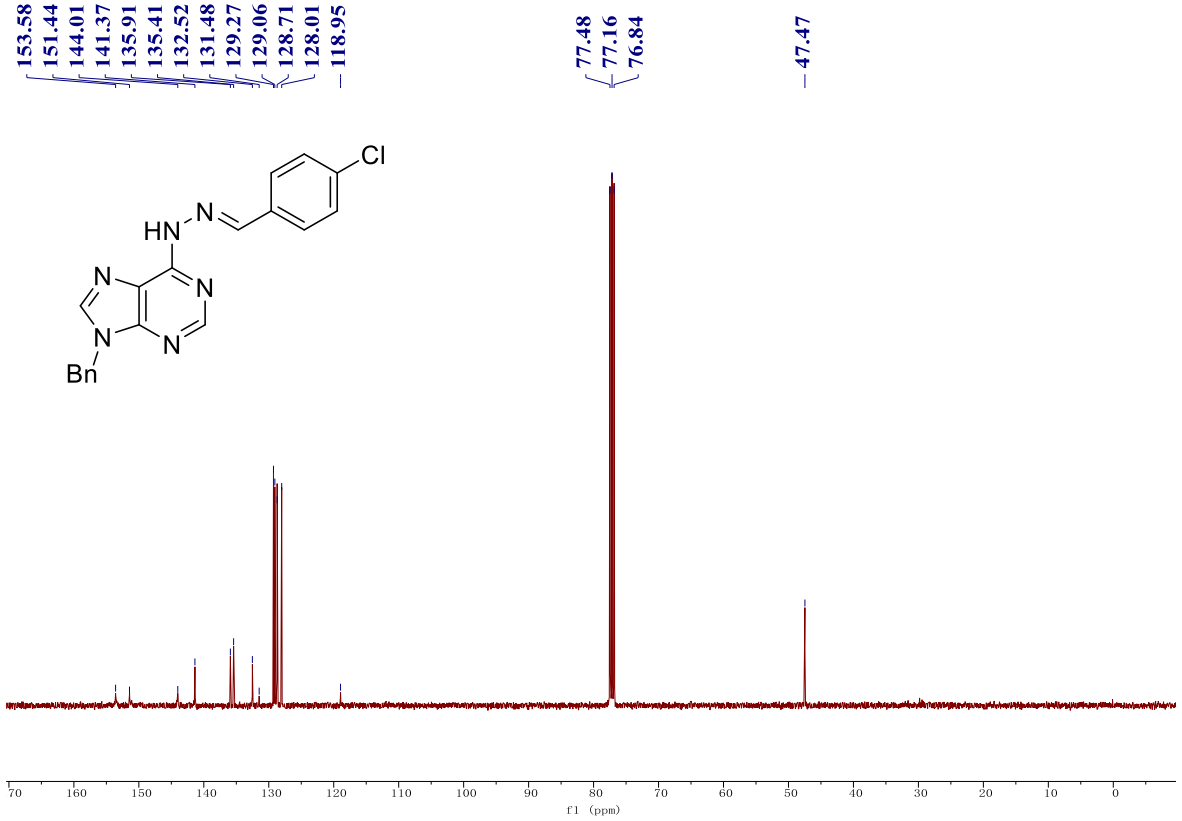
^{19}F NMR Spectrum of 1m at 25 °C ($CDCl_3$, 565 MHz)



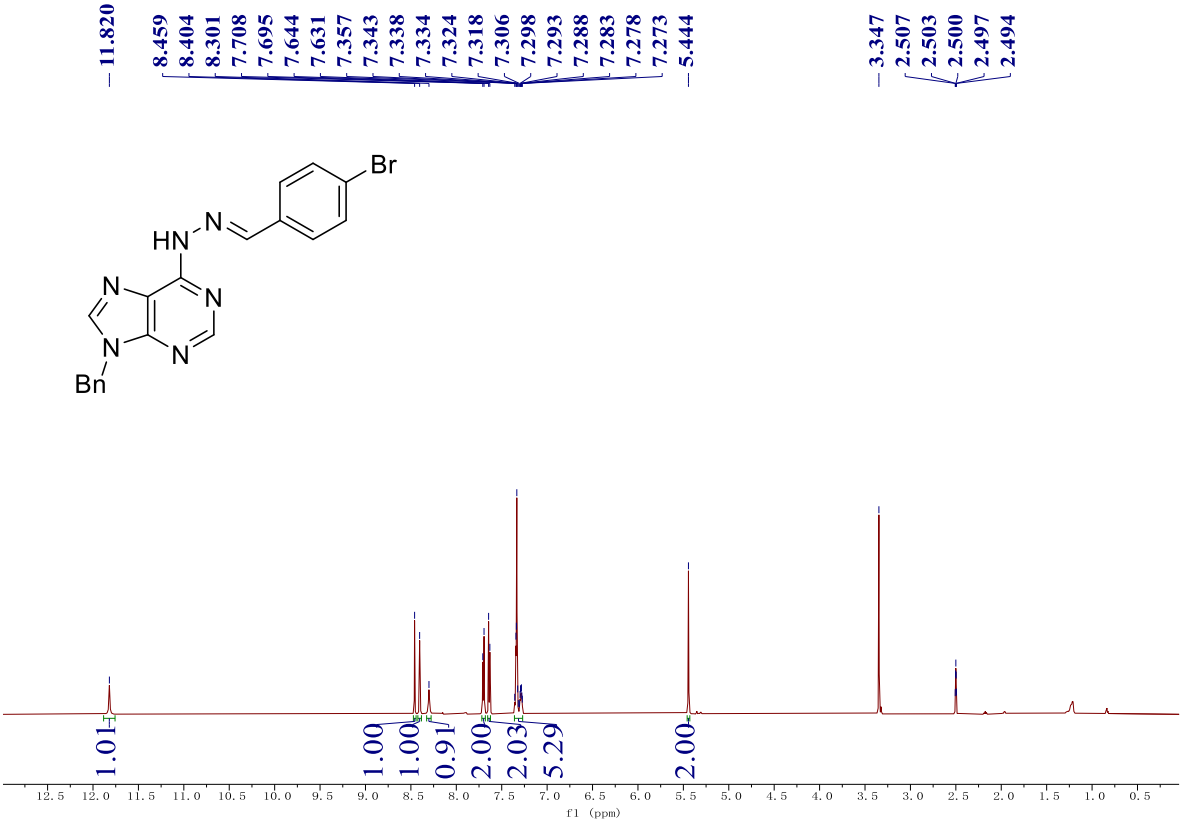
^{1}H NMR Spectrum of 1n at 25 °C ($CDCl_3$, 400 MHz)



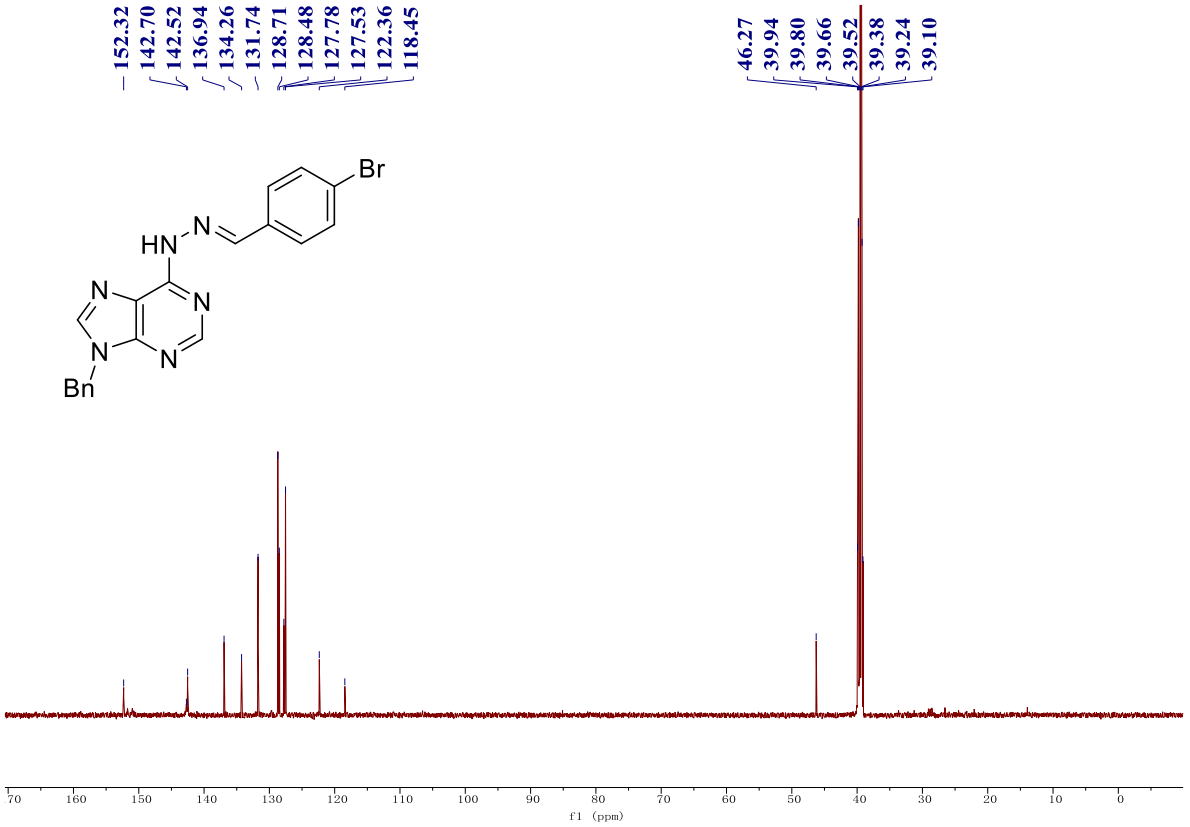
^{13}C NMR Spectrum of 1n at 25 °C ($CDCl_3$, 100 MHz)



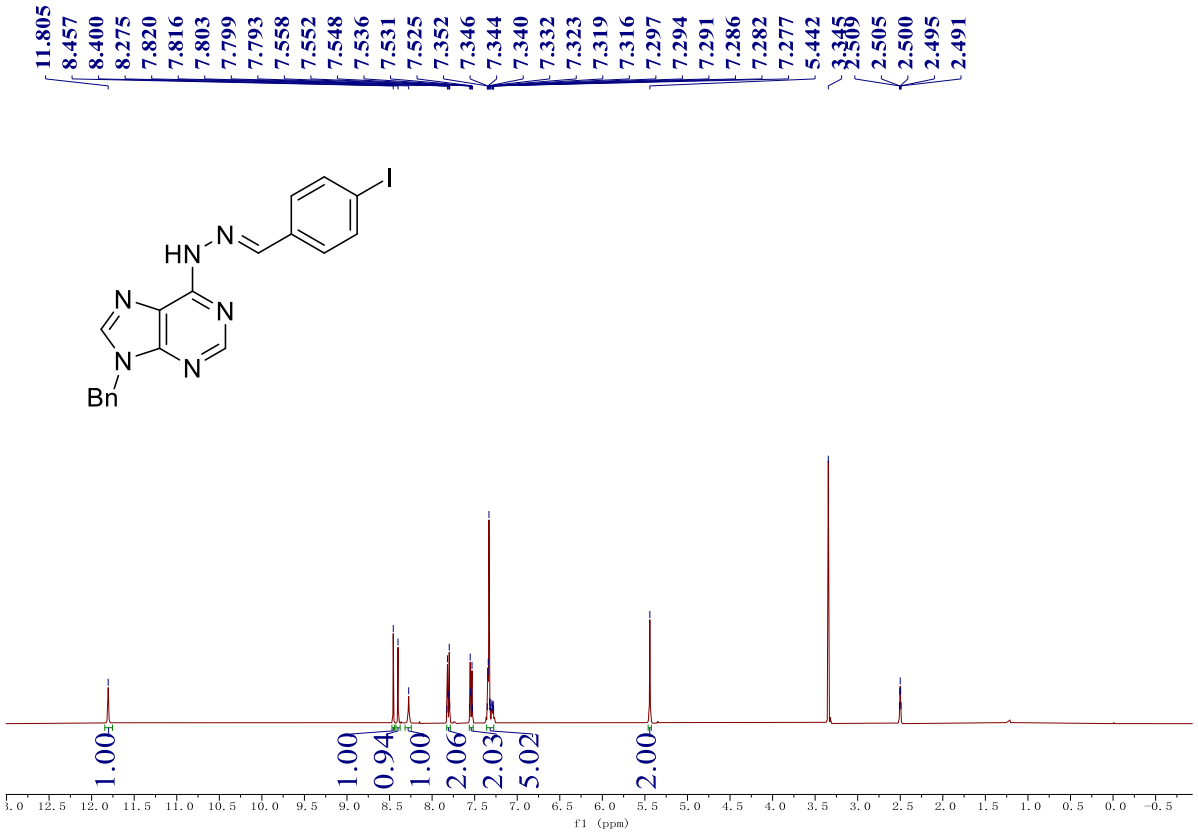
^{1}H NMR Spectrum of 1o at 25 °C (DMSO-d_6, 600 MHz)



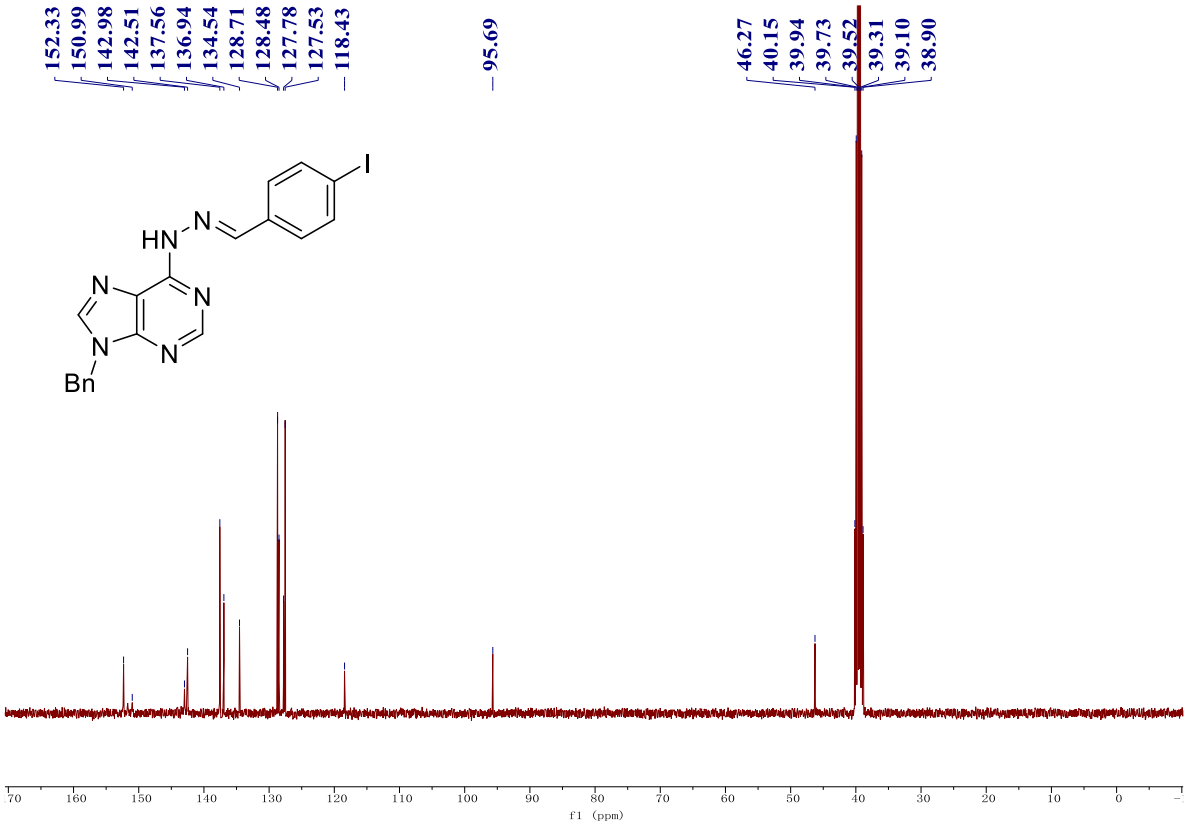
^{13}C NMR Spectrum of 1o at 25 °C (DMSO-d_6, 150 MHz)



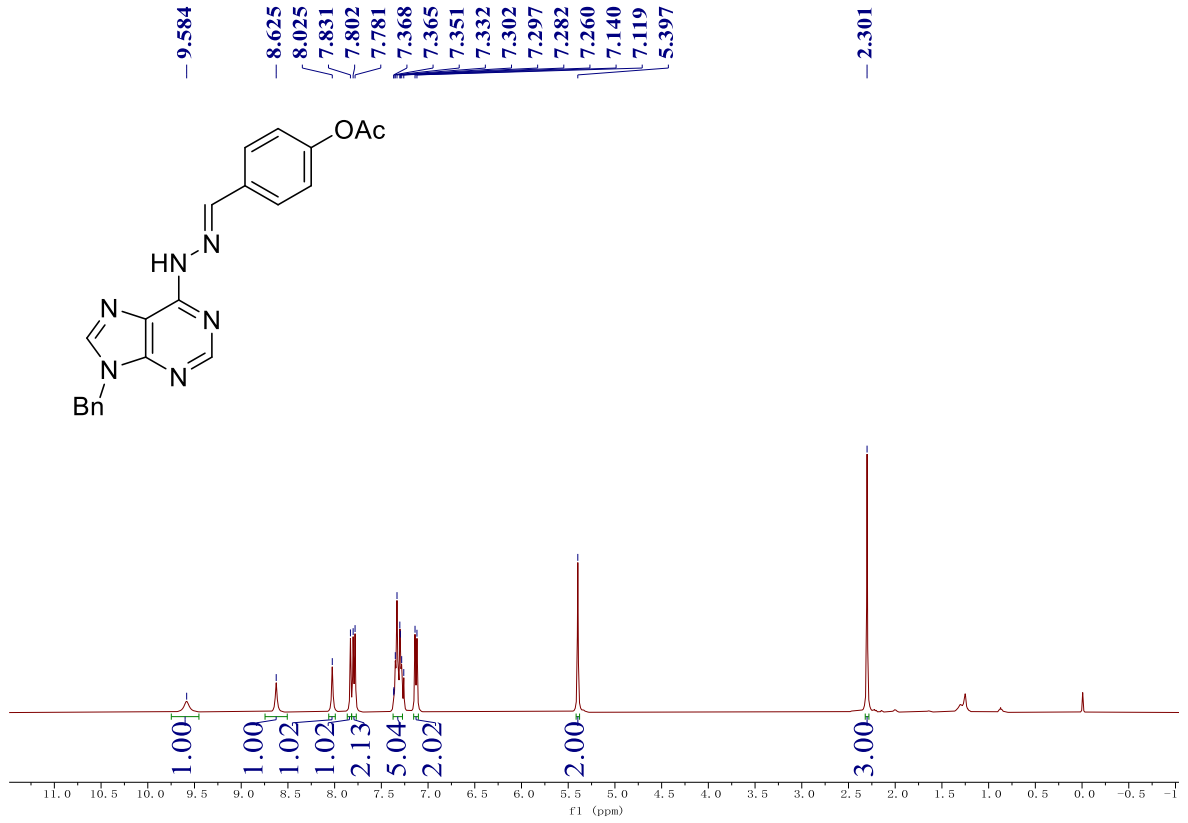
^{1}H NMR Spectrum of 1p at 25 °C (DMSO-d_6, 400 MHz)



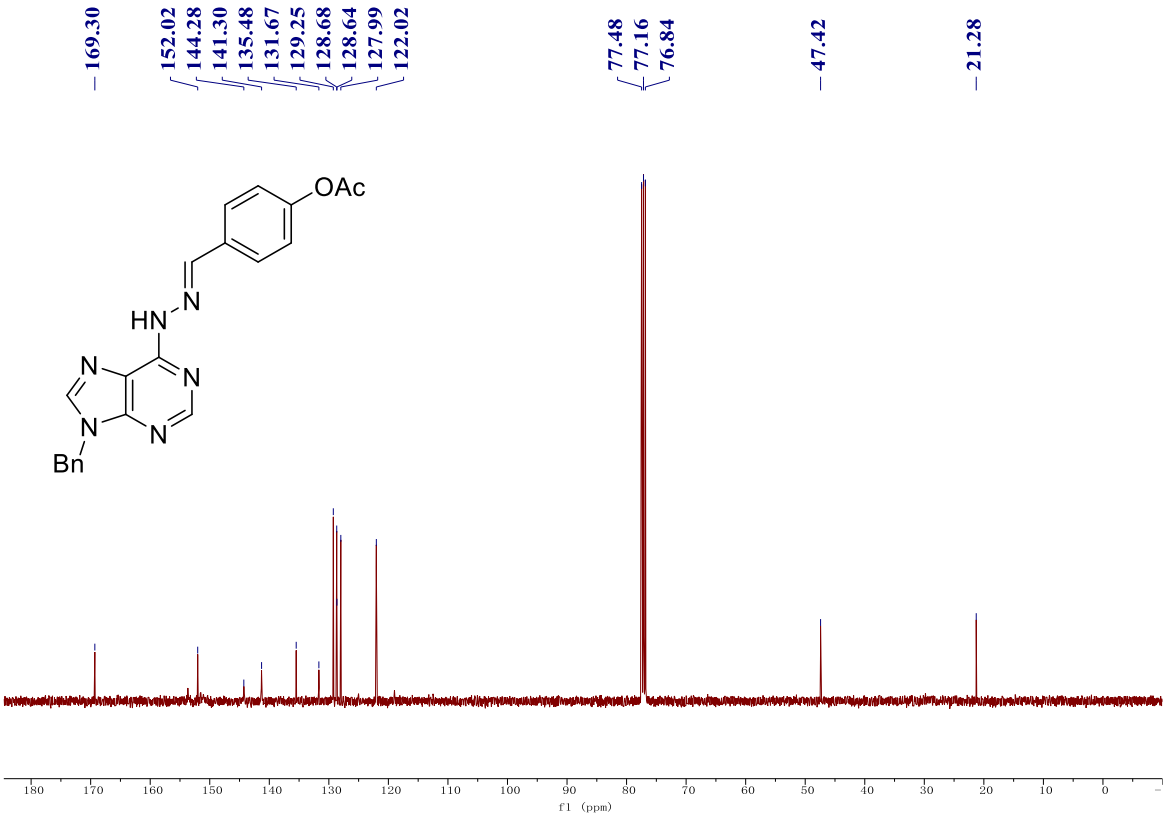
^{13}C NMR Spectrum of 1p at 25 °C (DMSO-d_6, 100 MHz)



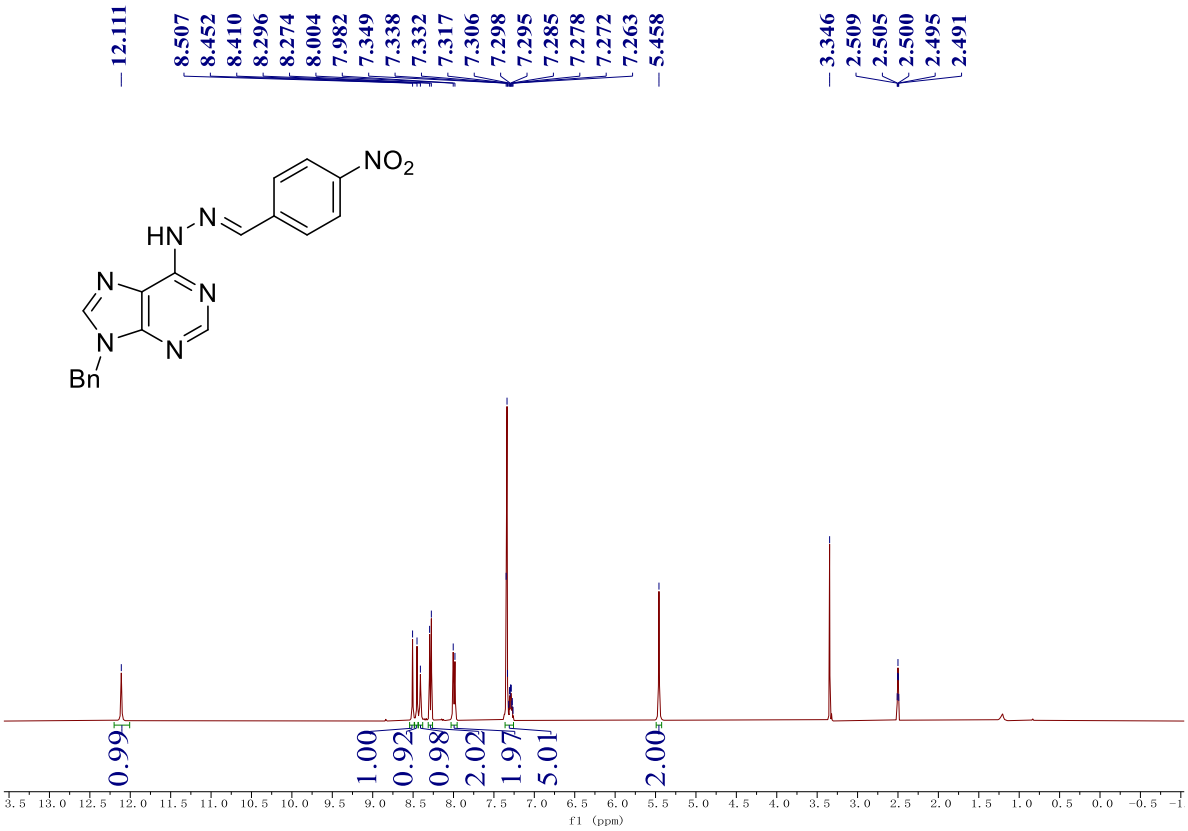
^{1}H NMR Spectrum of 1q at 25 °C ($CDCl_3$, 400 MHz)



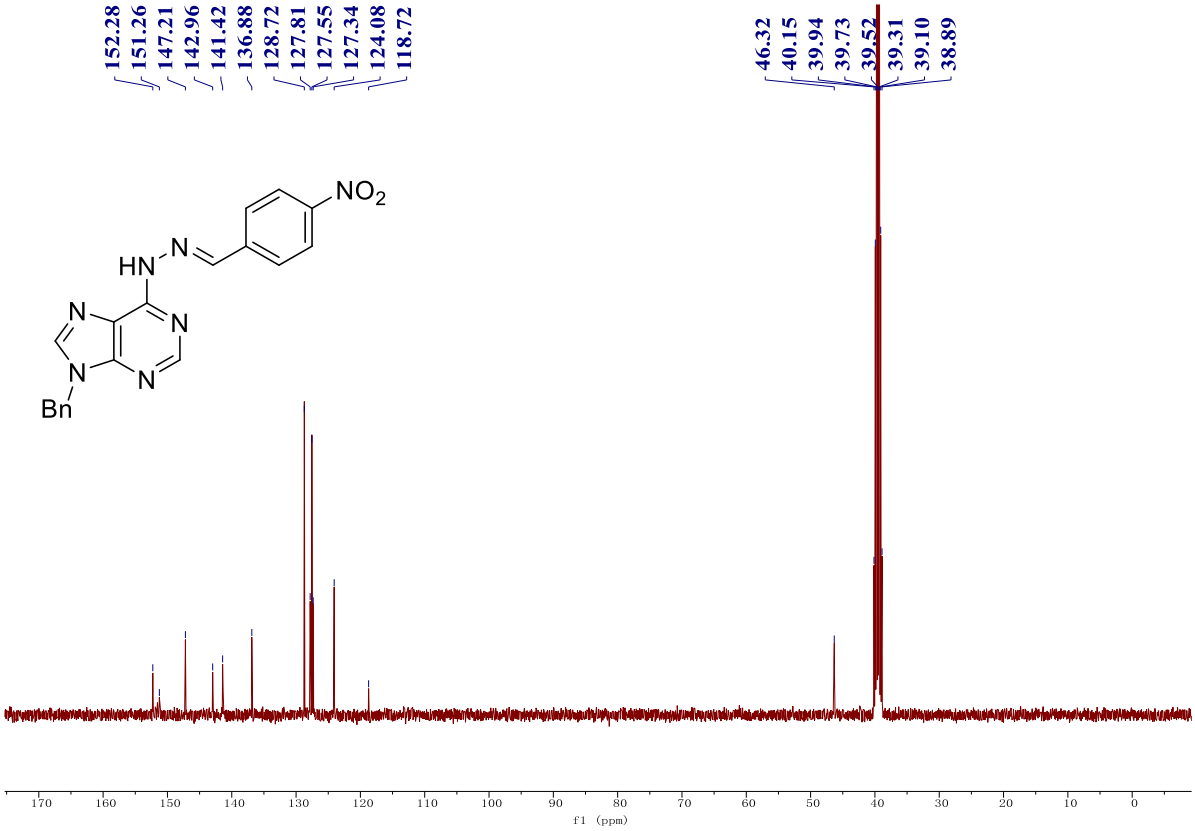
^{13}C NMR Spectrum of 1q at 25 °C ($CDCl_3$, 100 MHz)



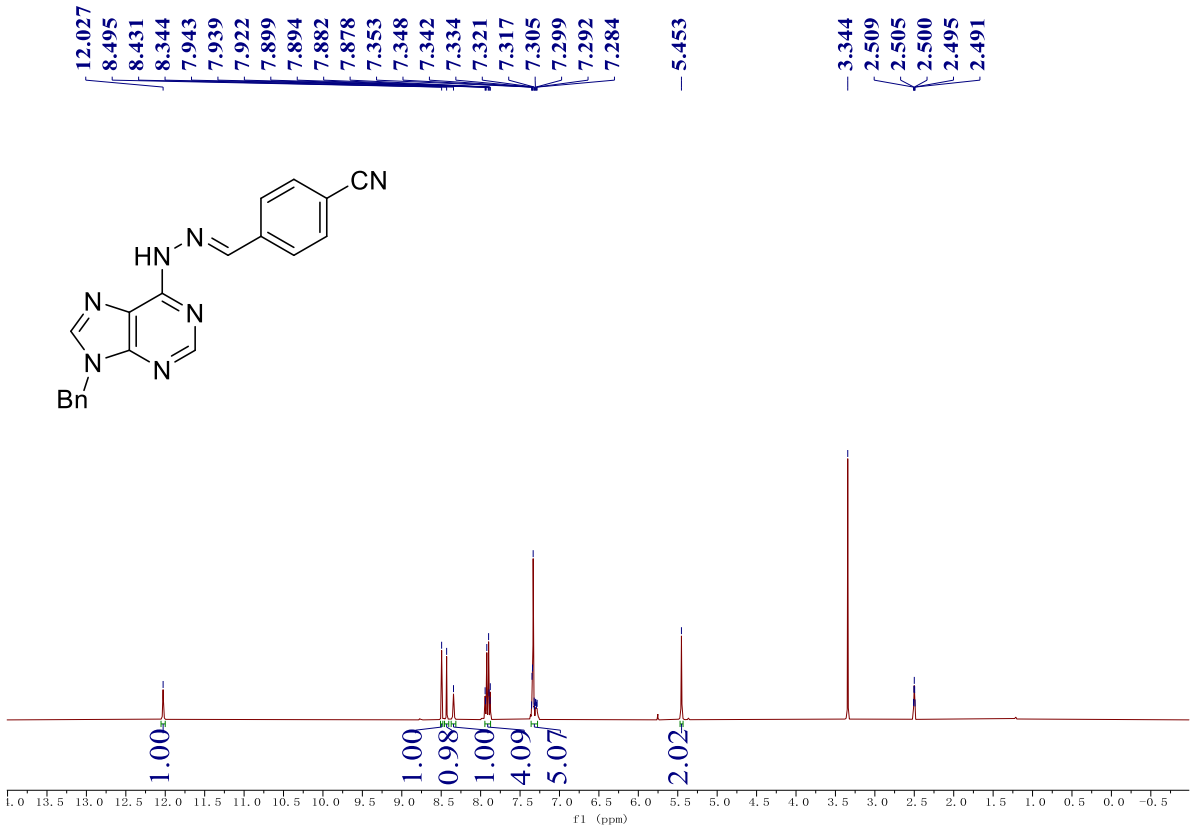
^{1}H NMR Spectrum of 1r at 25 °C (DMSO-d_6, 400 MHz)



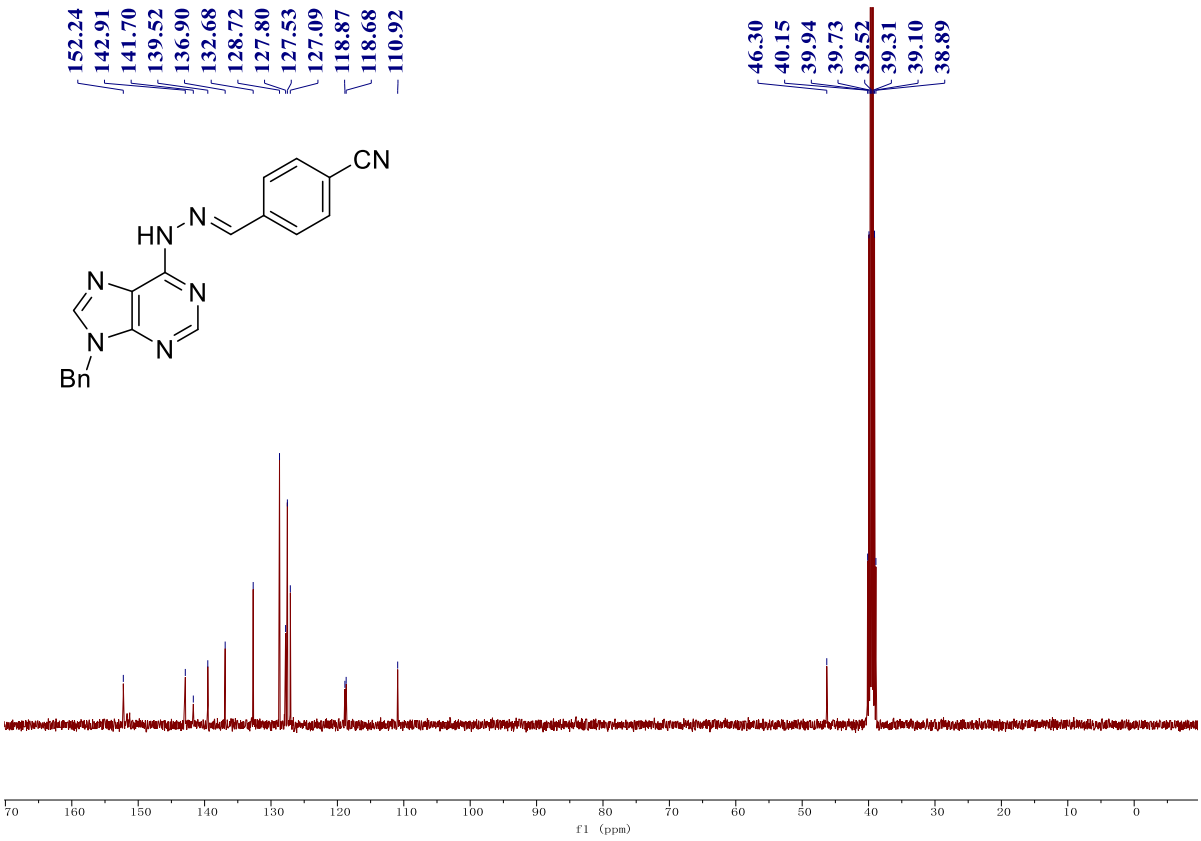
^{13}C NMR Spectrum of 1r at 25 °C (DMSO-d_6, 100 MHz)



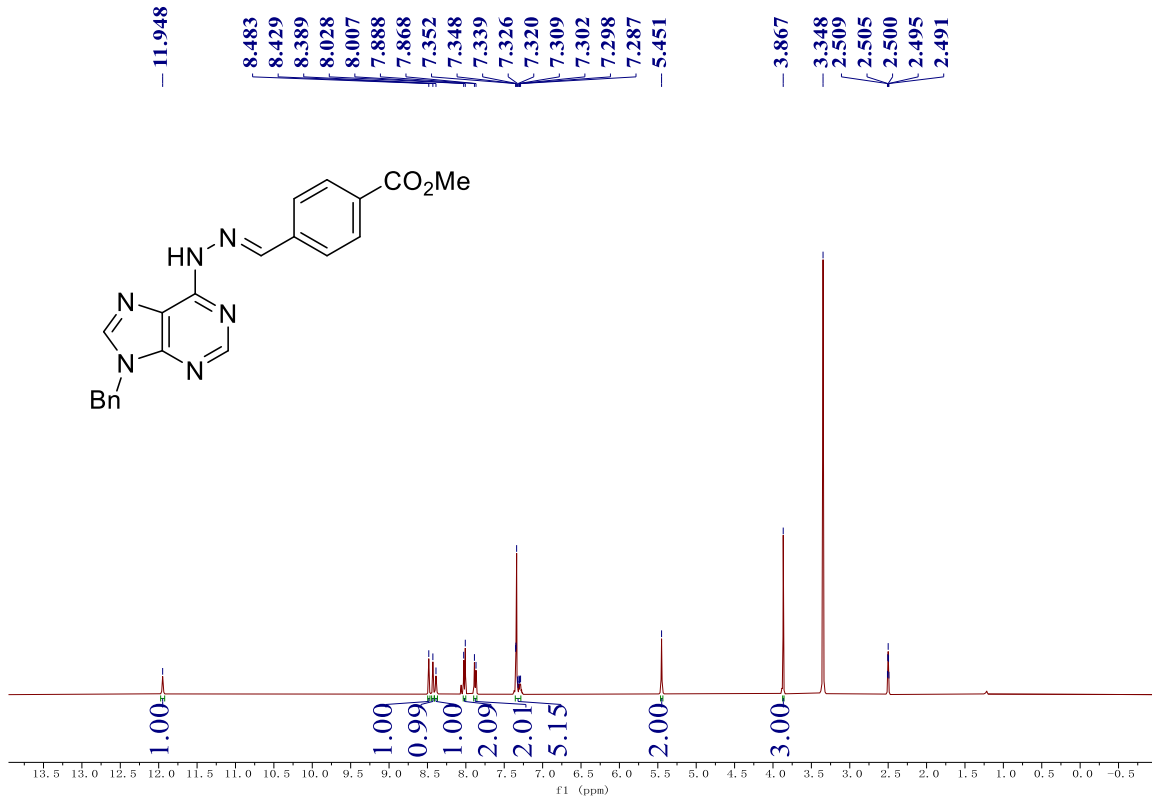
^{1}H NMR Spectrum of 1s at 25 °C (DMSO-d_6, 400 MHz)



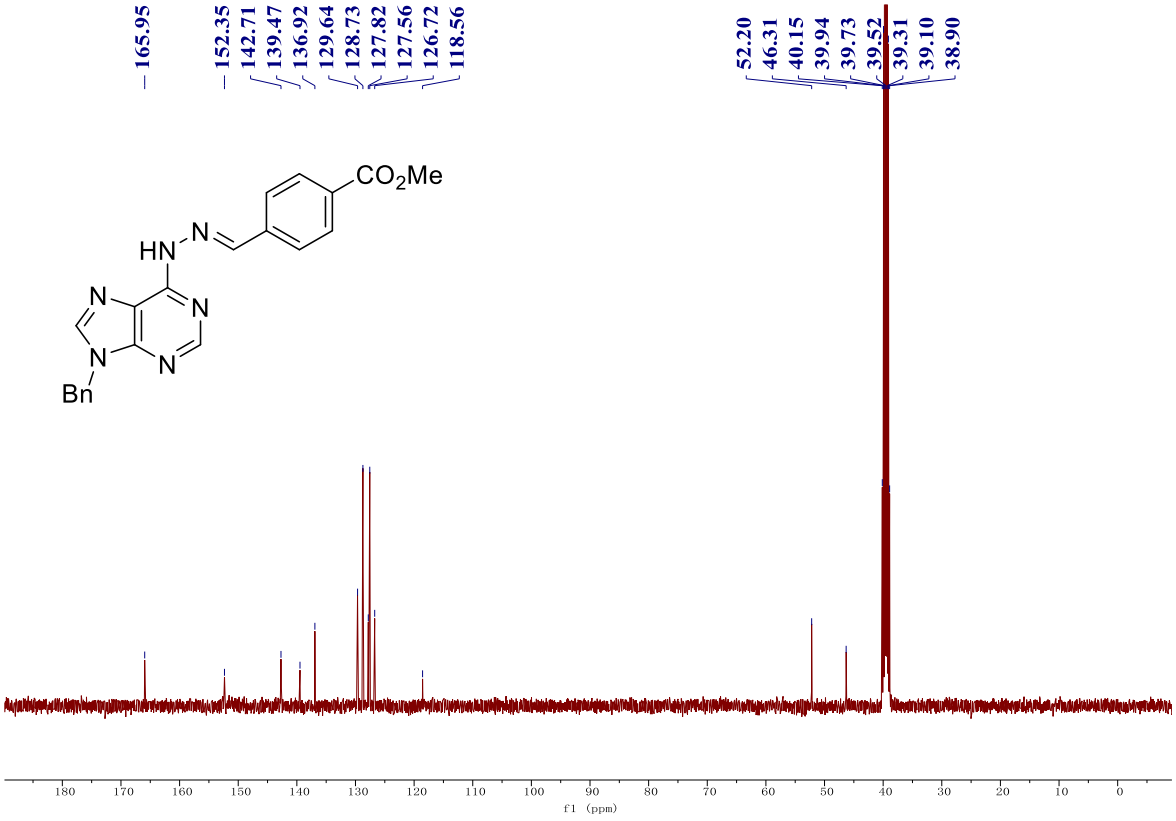
^{13}C NMR Spectrum of 1s at 25 °C (DMSO-d_6, 100 MHz)



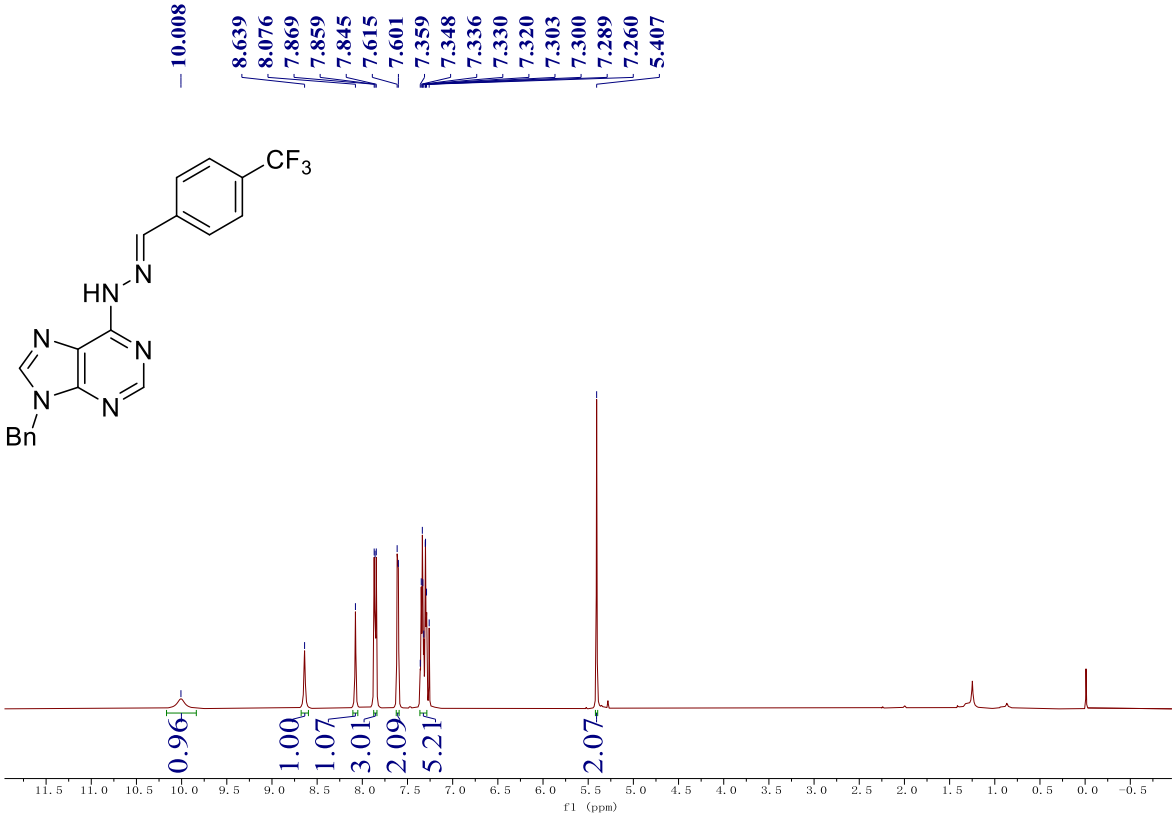
^{1}H NMR Spectrum of 1t at 25 °C (DMSO-d_6, 400 MHz)



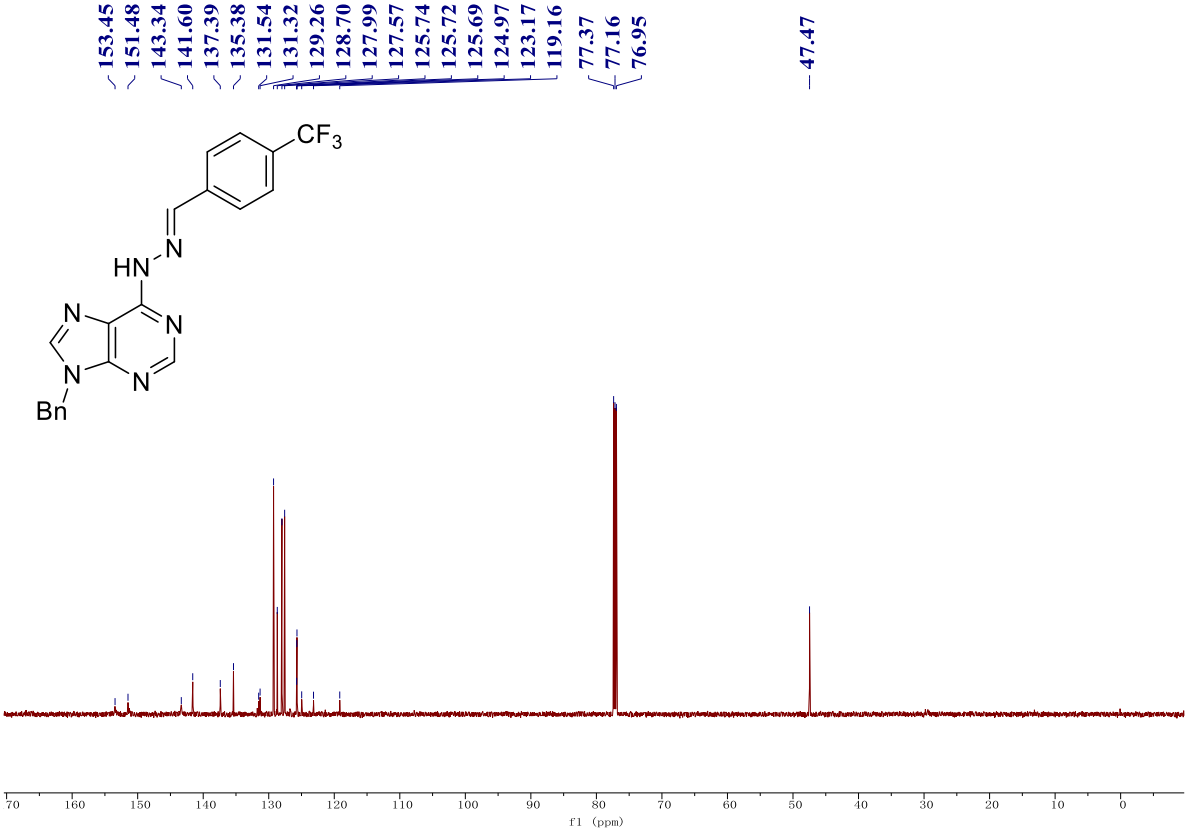
^{13}C NMR Spectrum of 1t at 25 °C (DMSO-d_6, 100 MHz)



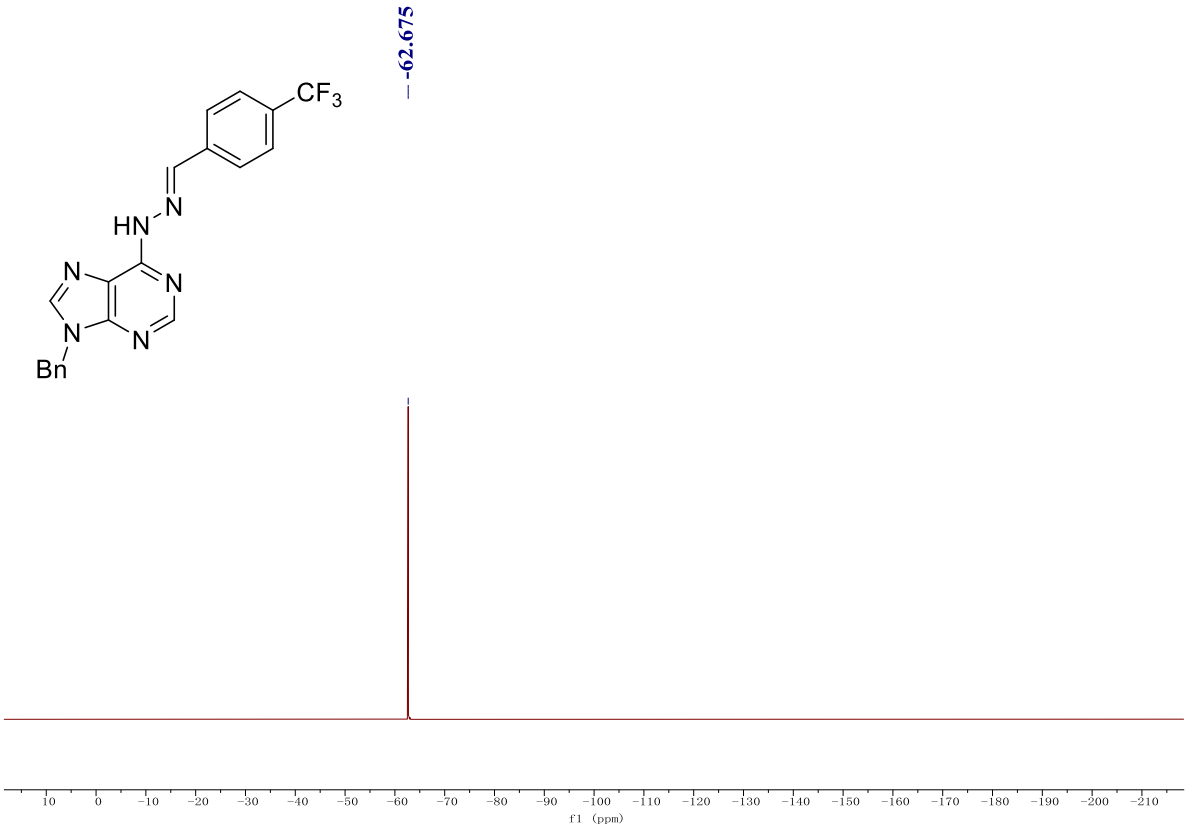
^{1}H NMR Spectrum of 1u at 25 °C ($CDCl_3$, 600 MHz)



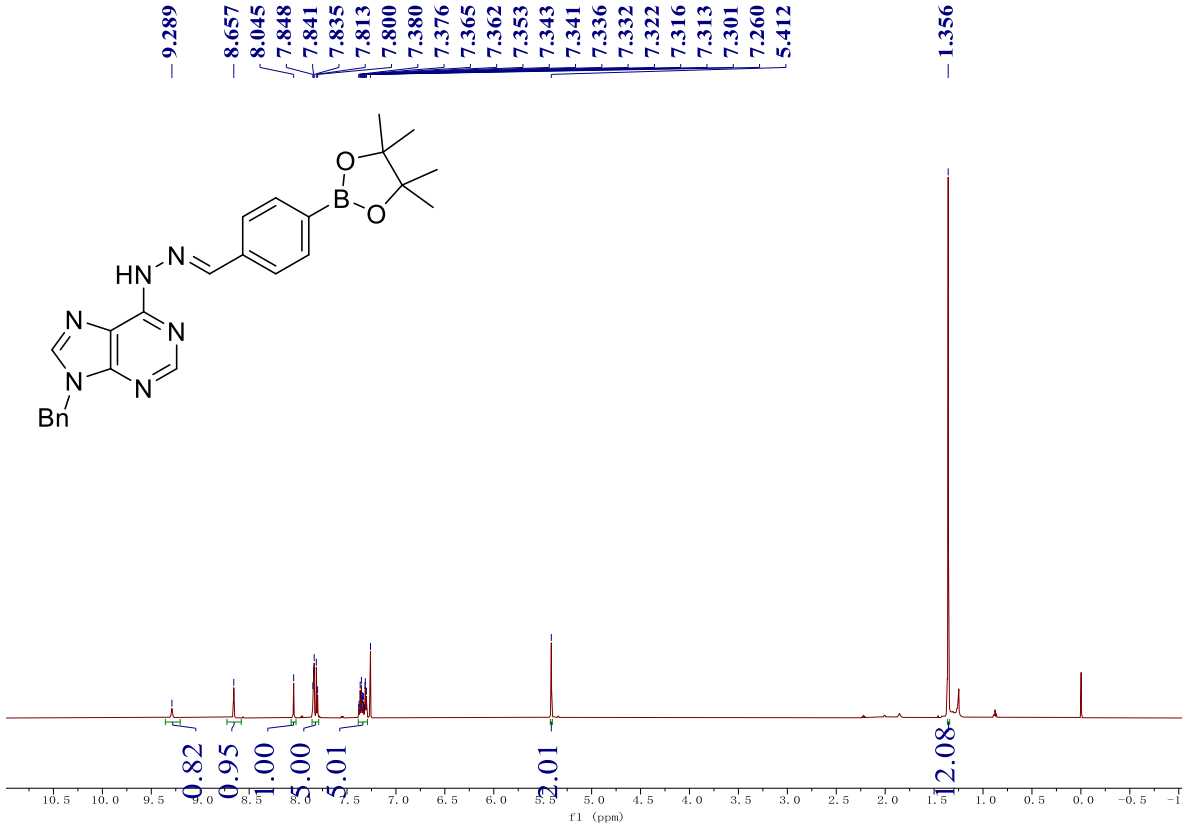
^{13}C NMR Spectrum of 1u at 25 °C ($CDCl_3$, 150 MHz)



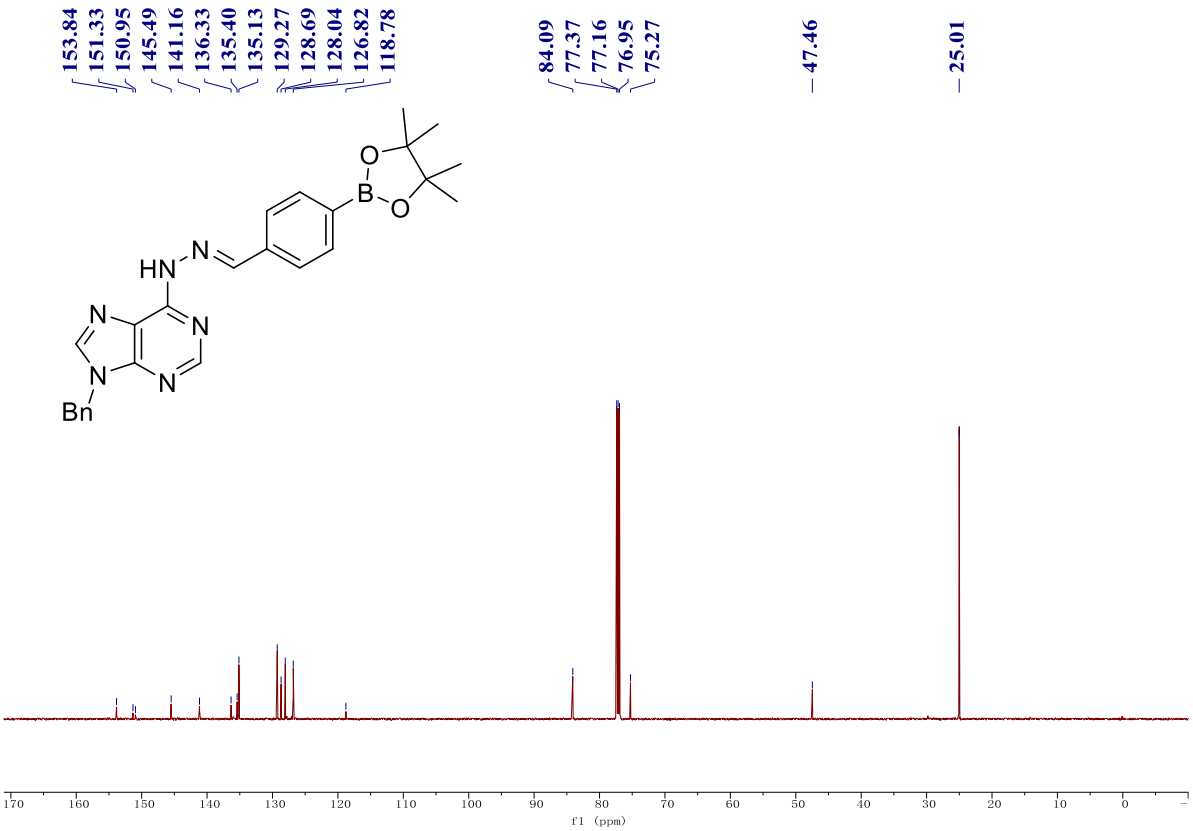
^{19}F NMR Spectrum of 1u at 25 °C ($CDCl_3$, 565 MHz)



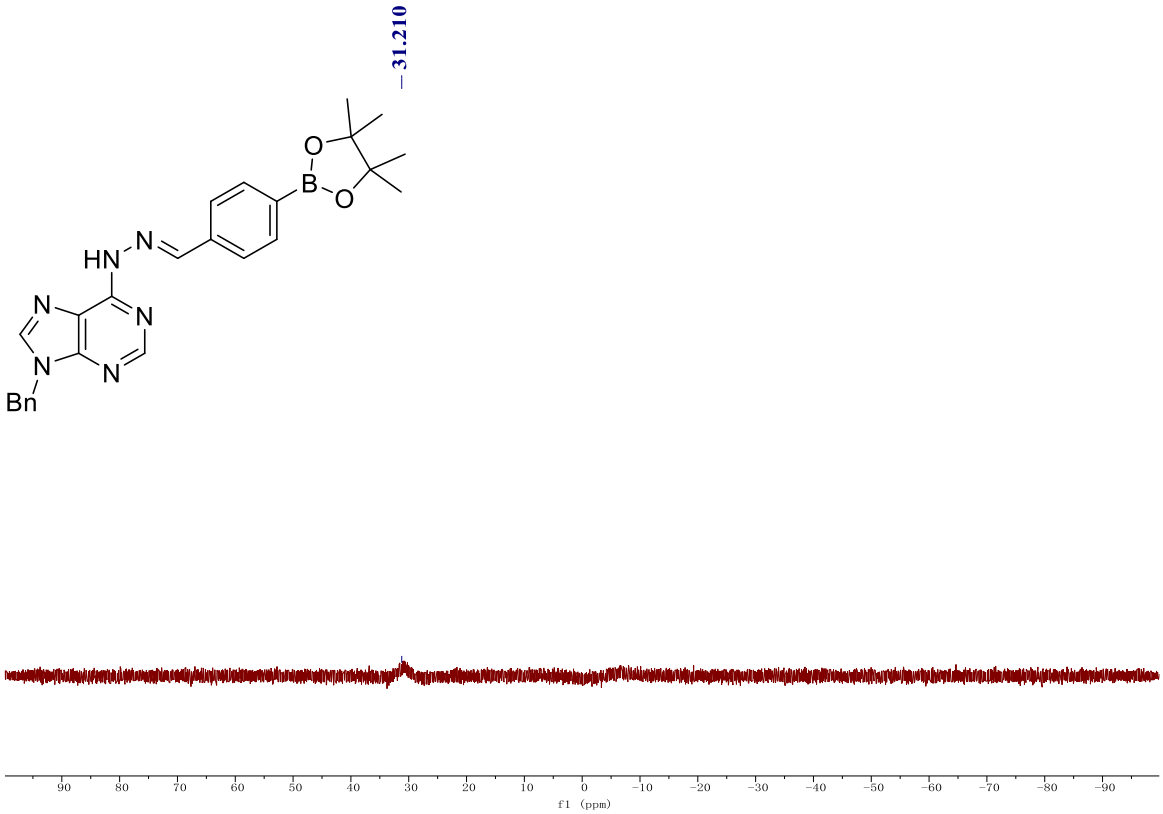
^{1}H NMR Spectrum of 1v at 25 ^{o}C ($CDCl_3$, 600 MHz)



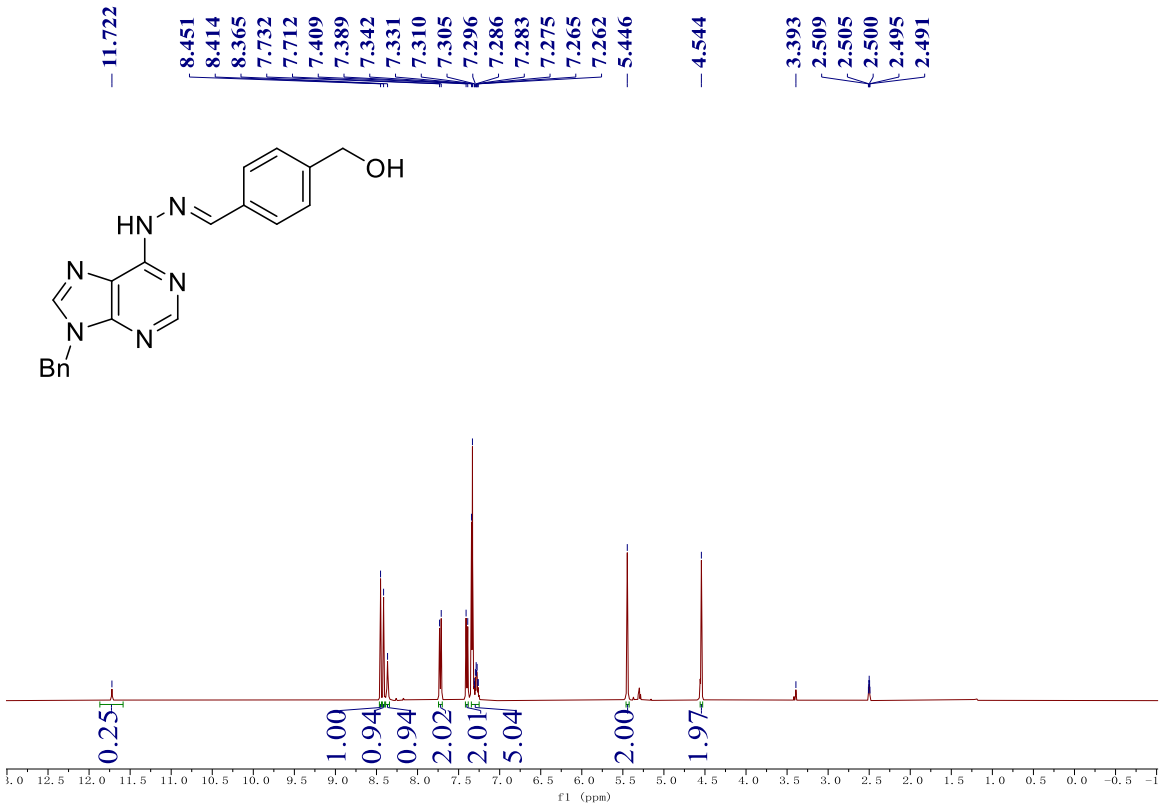
^{13}C NMR Spectrum of 1v at 25 ^{o}C ($CDCl_3$, 150 MHz)



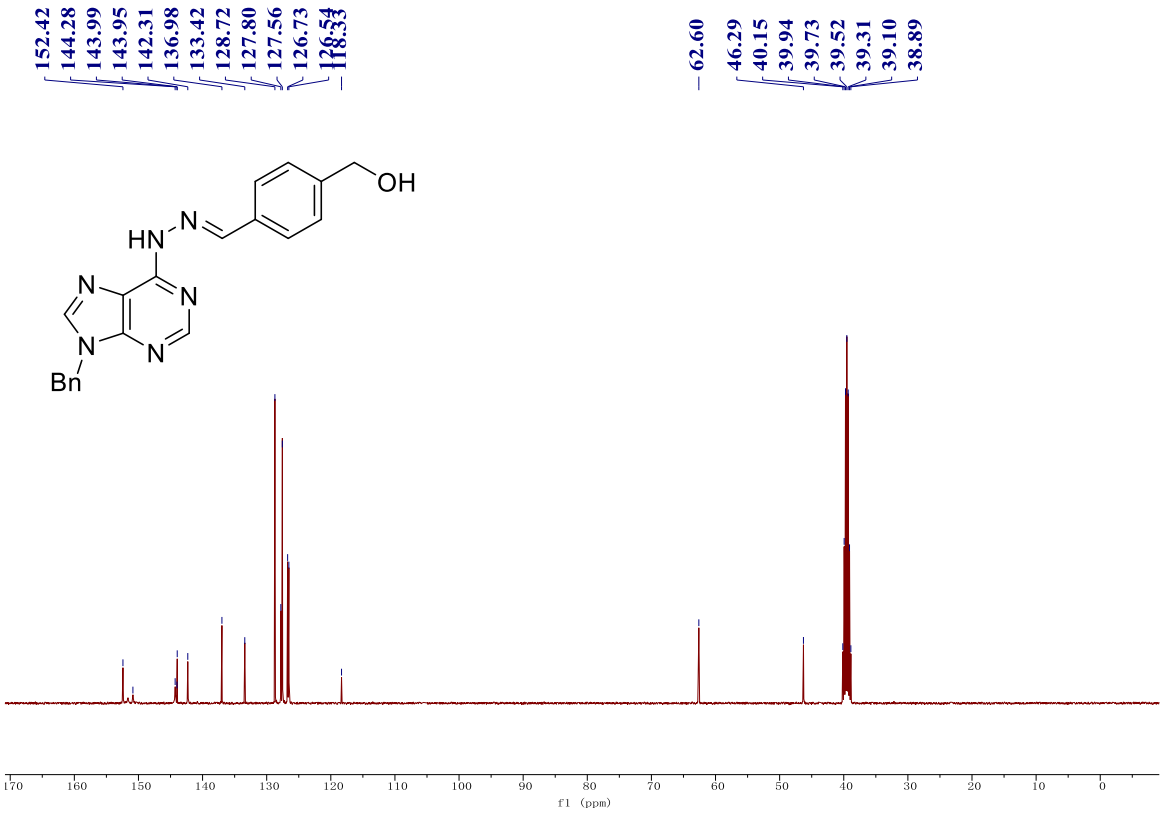
^{11}B NMR Spectrum of 1v at 25 °C ($CDCl_3$, 193 MHz)



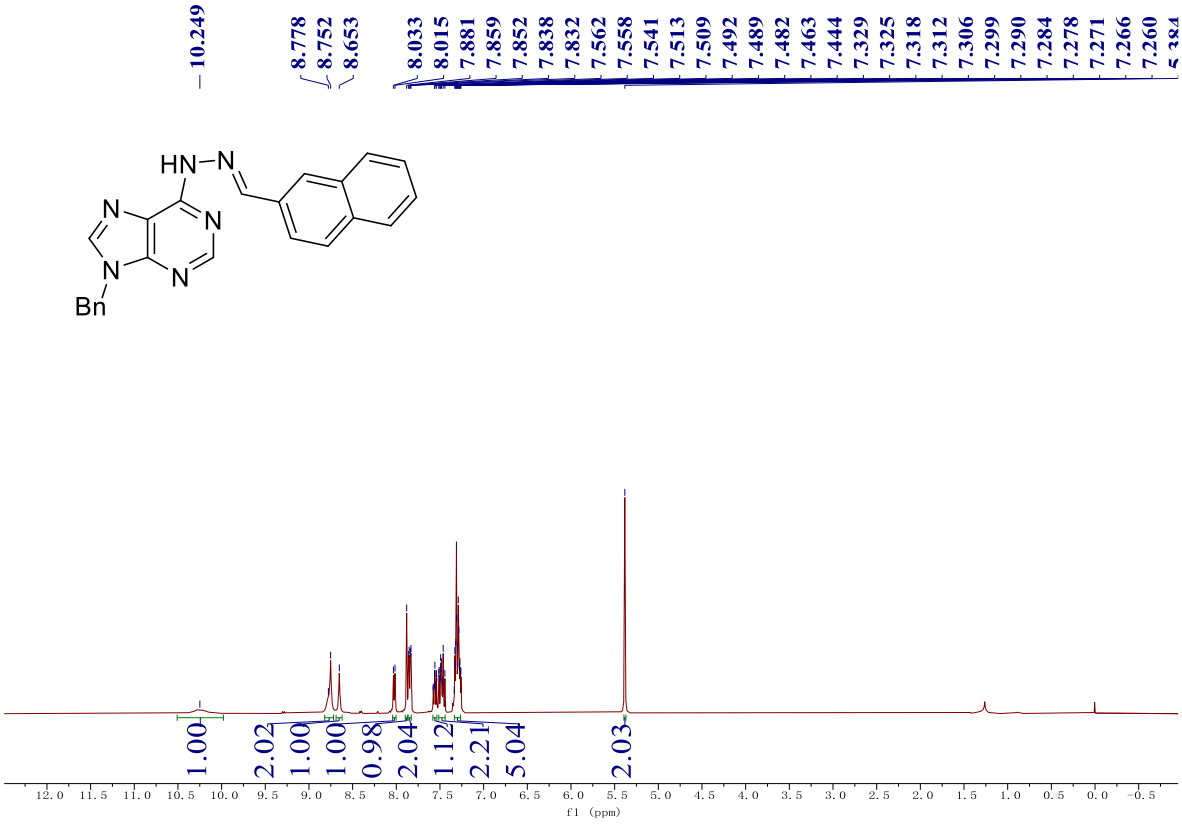
^{1}H NMR Spectrum of 1w at 25 °C (DMSO-d_6, 400 MHz)



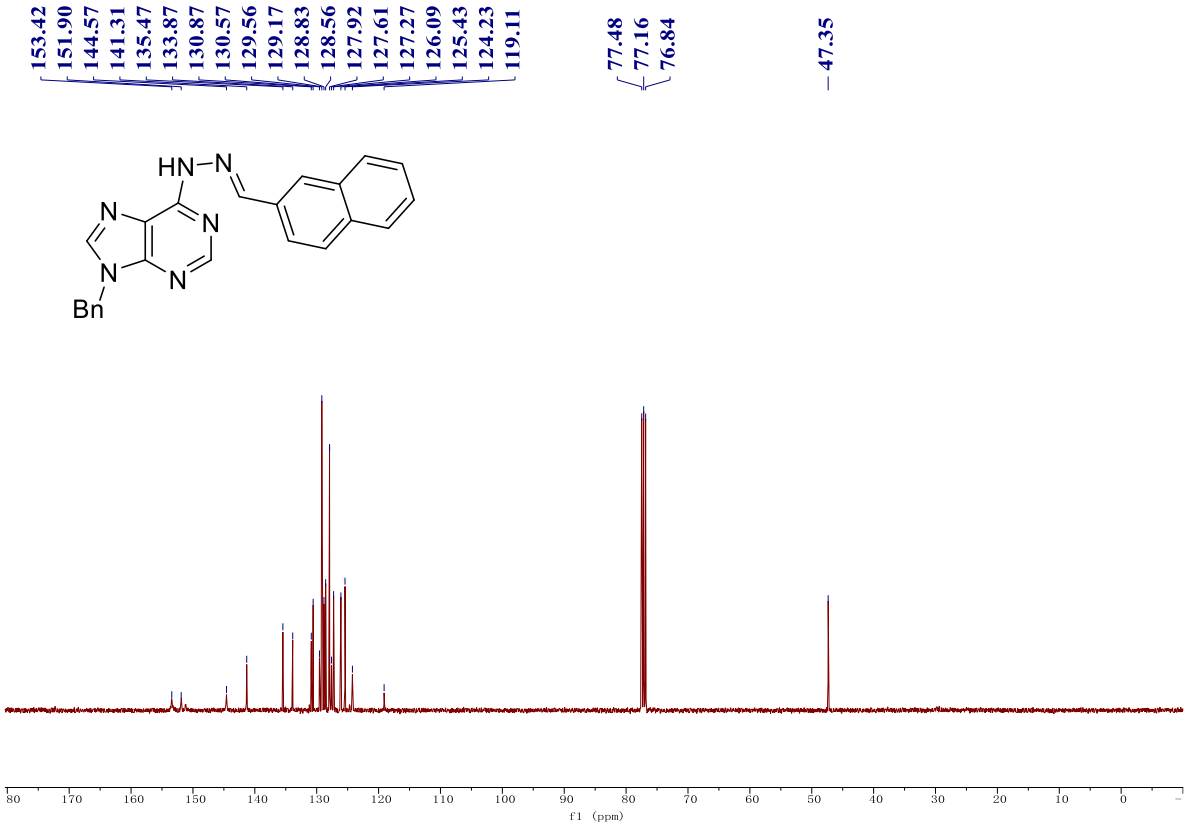
^{13}C NMR Spectrum of 1w at 25 °C (DMSO-d_6, 100 MHz)



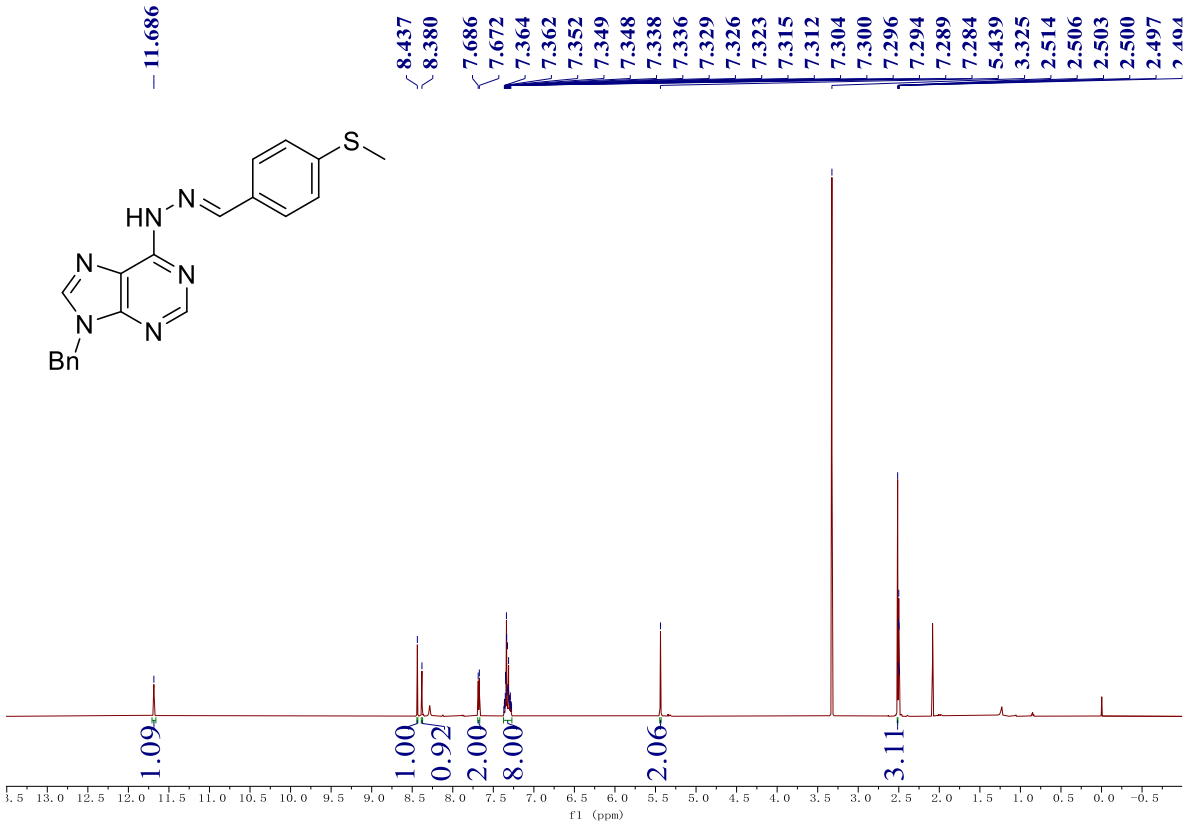
^{1}H NMR Spectrum of 1x at 25 °C ($CDCl_3$, 400 MHz)



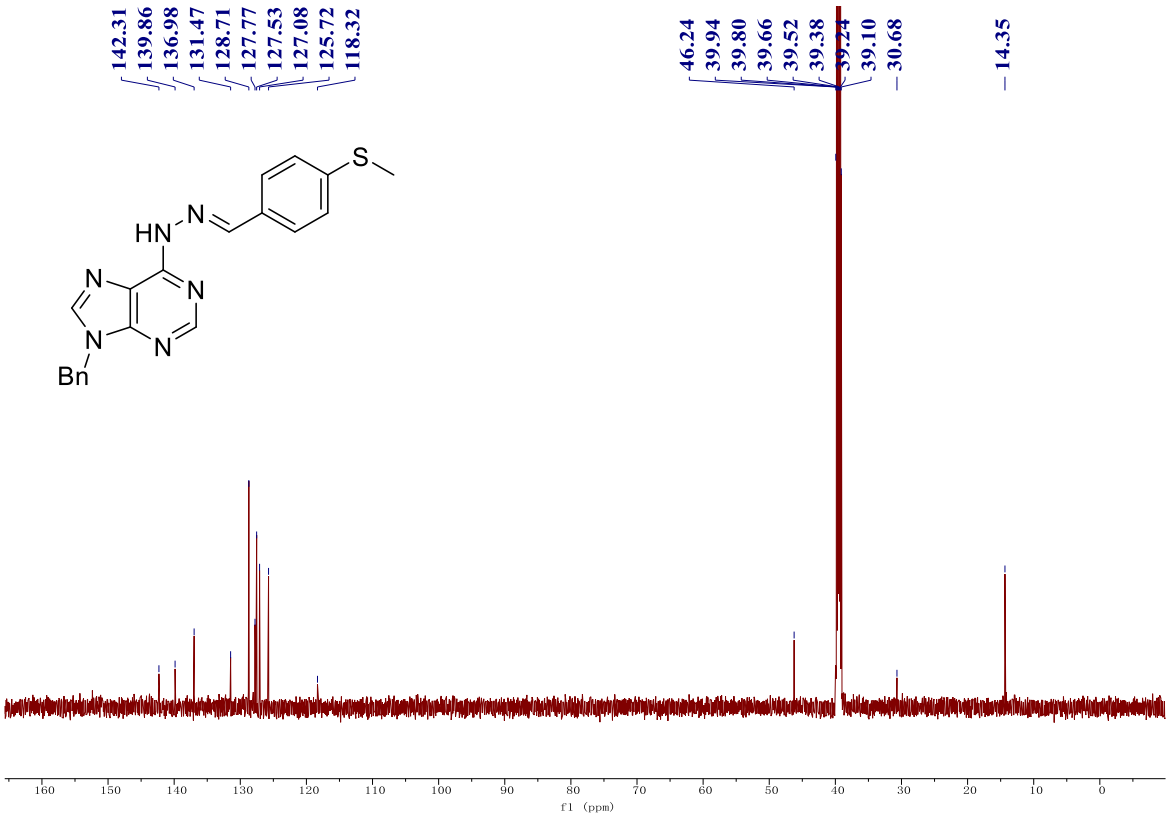
^{13}C NMR Spectrum of 1x at 25 °C ($CDCl_3$, 100 MHz)



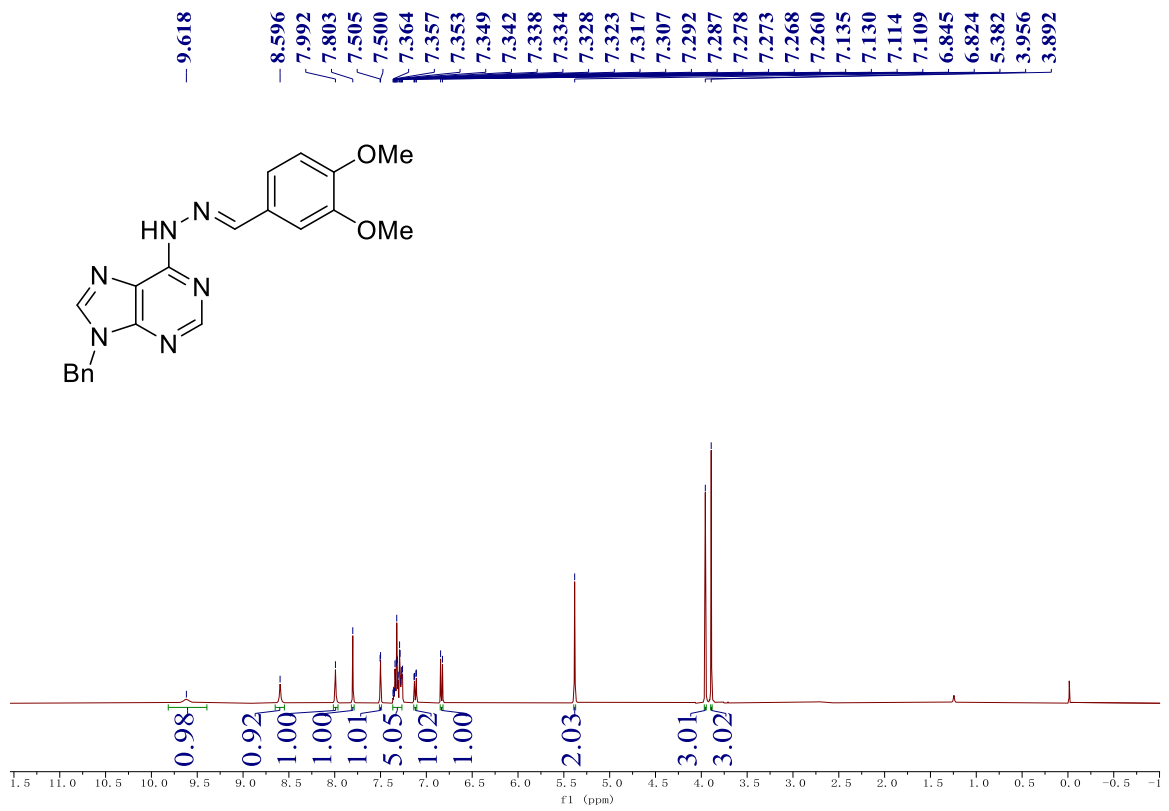
^{1}H NMR Spectrum of 1y at 25 °C (DMSO-d_6, 600 MHz)



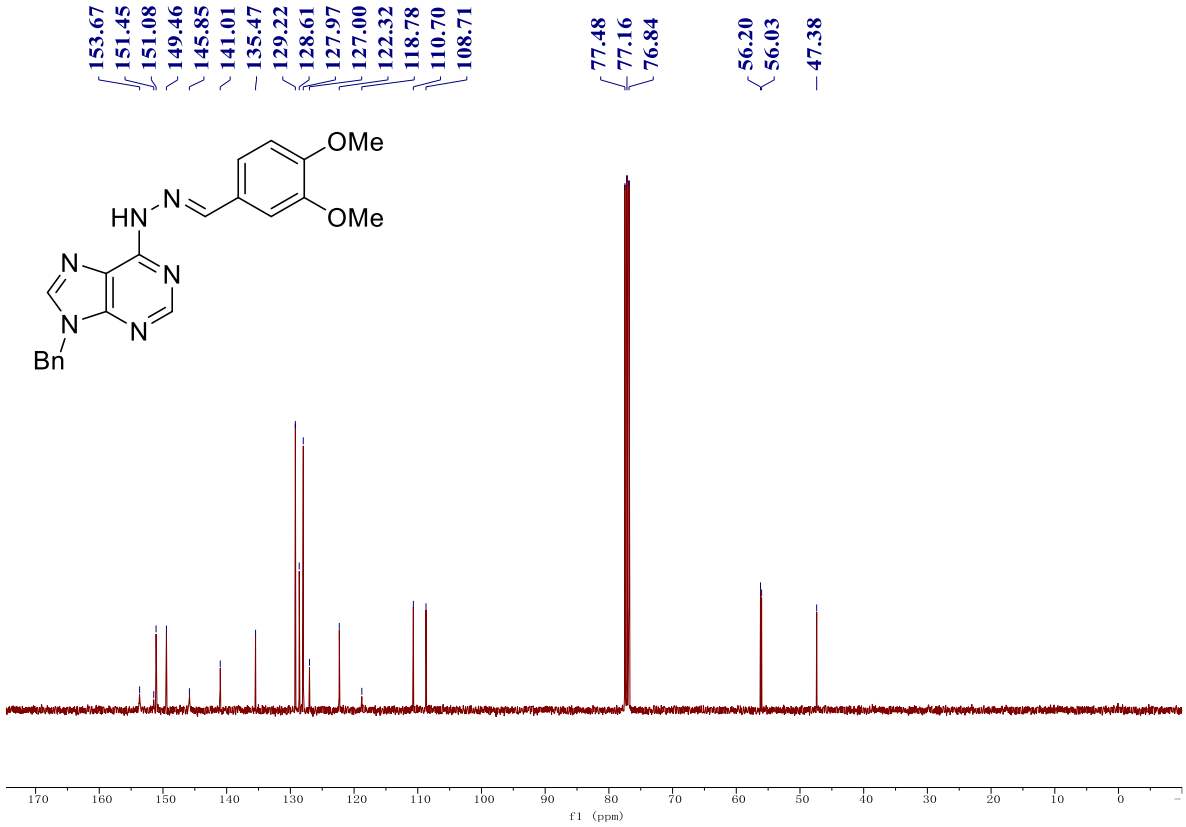
^{13}C NMR Spectrum of 1y at 25 °C (DMSO-d_6, 150 MHz)



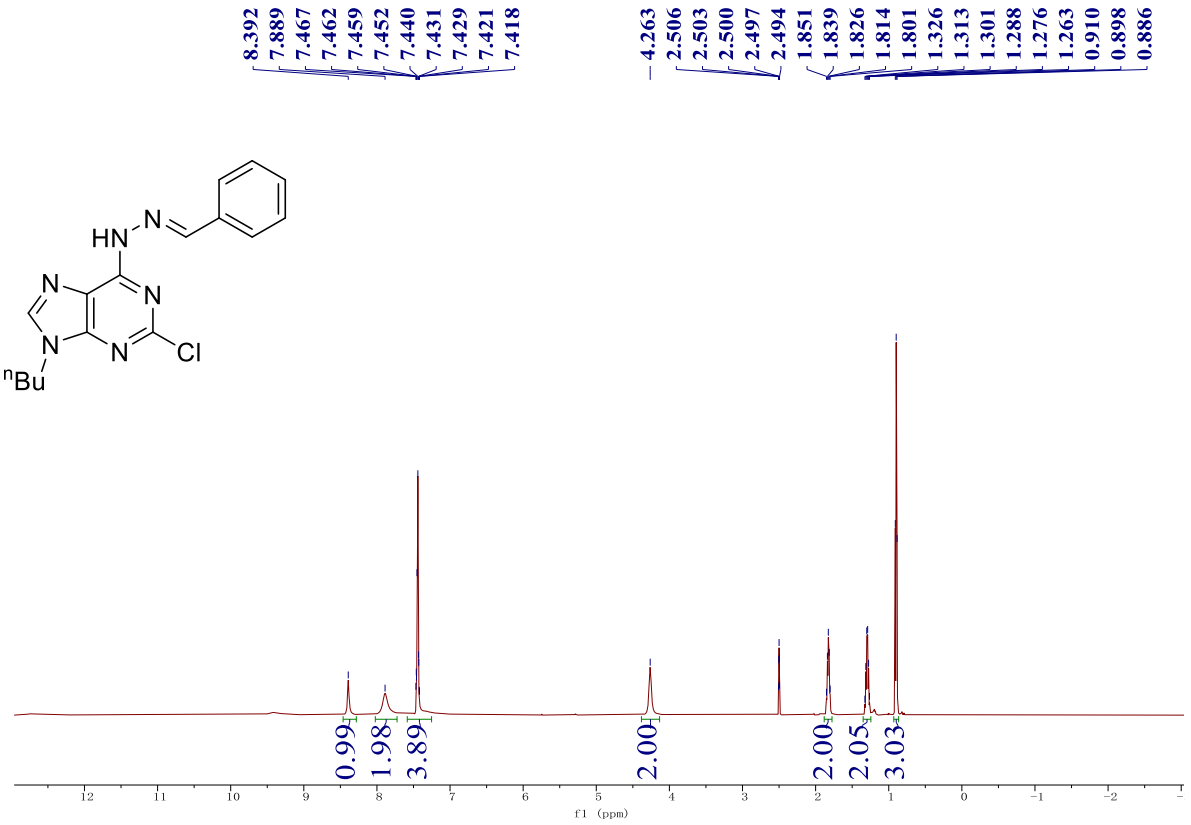
^{1}H NMR Spectrum of 1z at 25 °C ($CDCl_3$, 400 MHz)



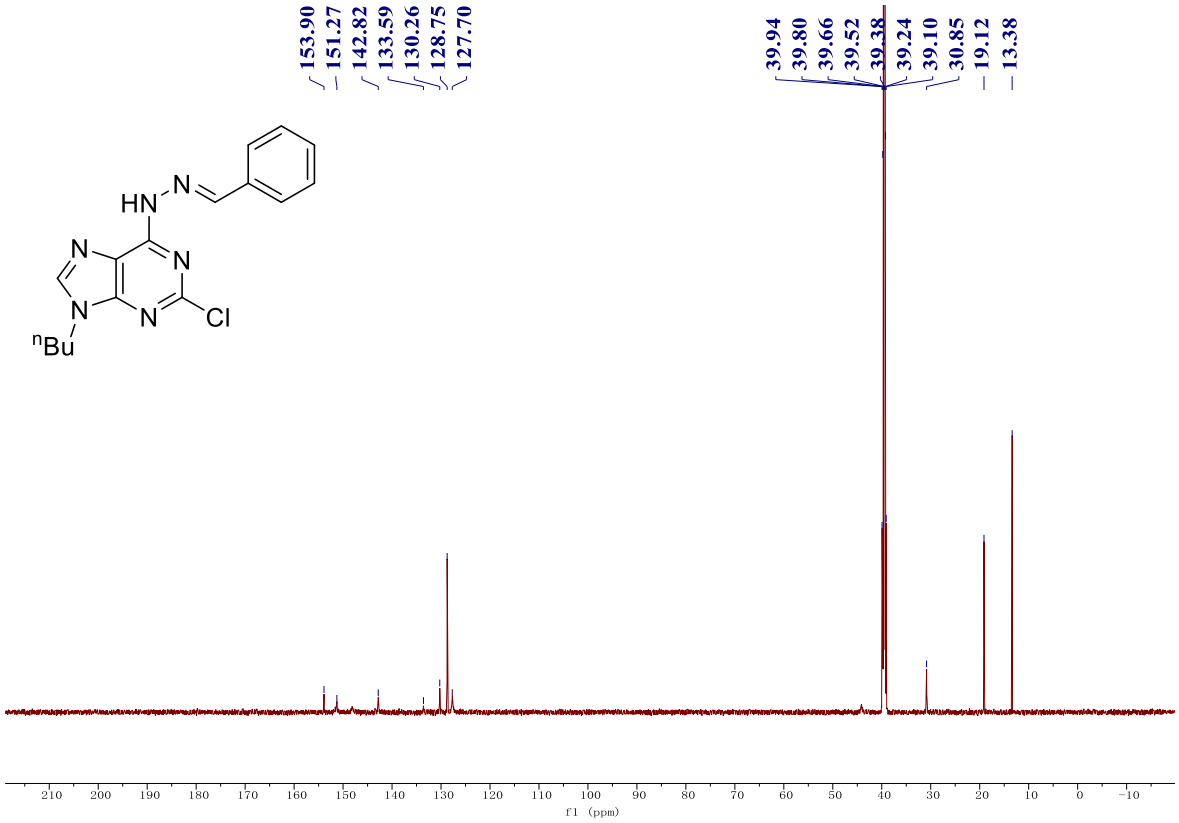
^{13}C NMR Spectrum of 1z at 25 °C ($CDCl_3$, 100 MHz)



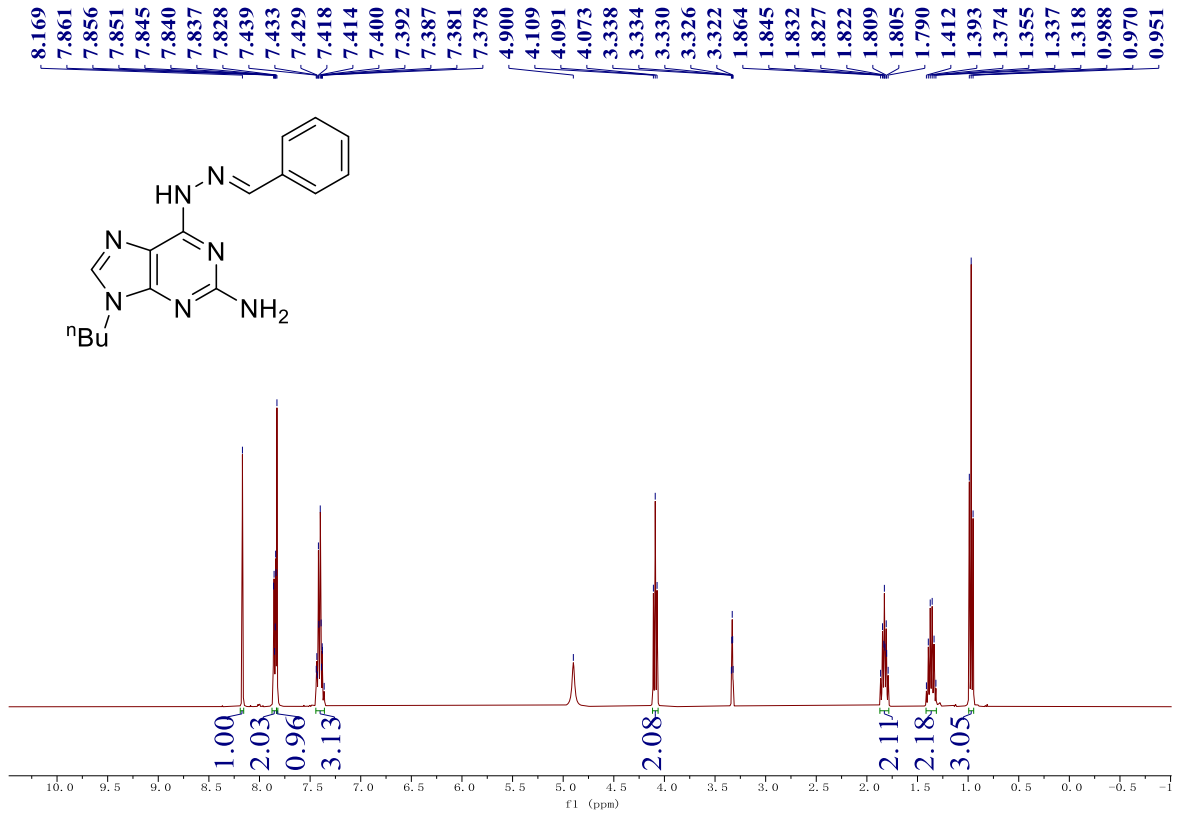
^{1}H NMR Spectrum of 1aa at 25 °C (DMSO-d_6, 600 MHz)



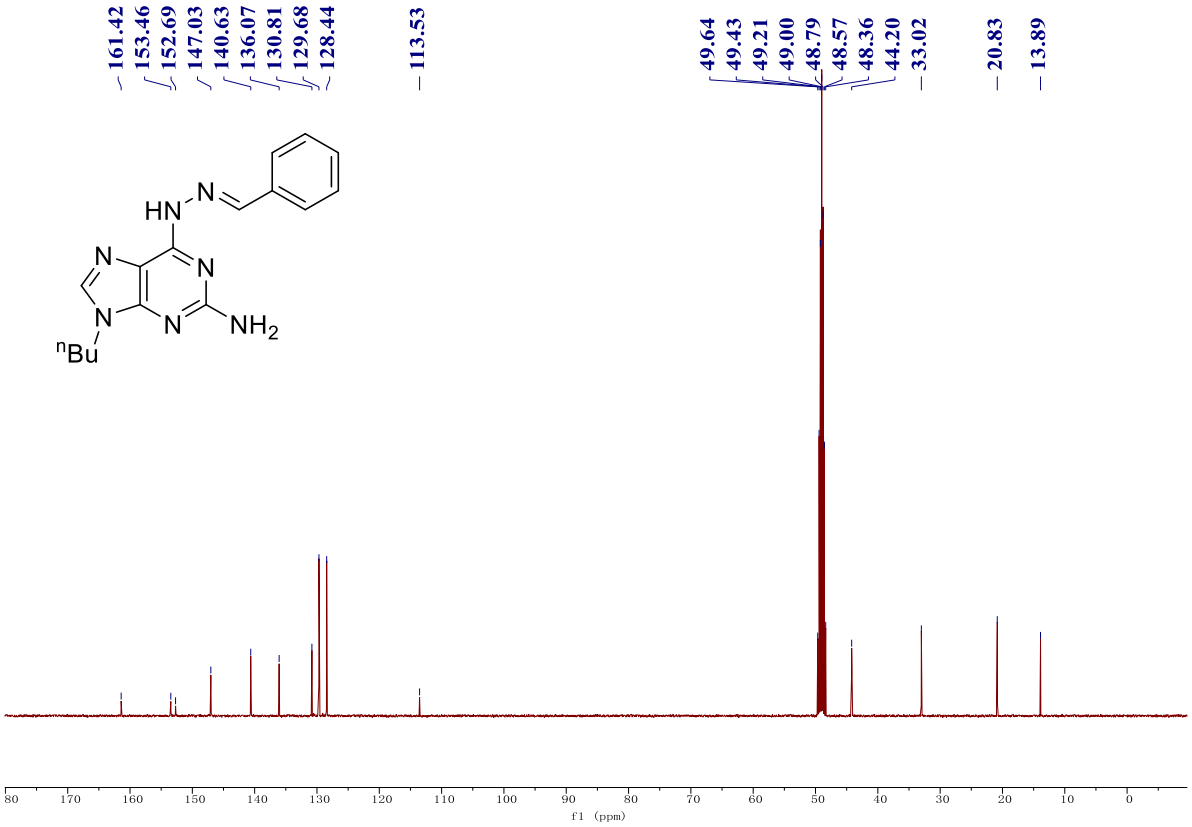
^{13}C NMR Spectrum of 1aa at 25 °C (DMSO-d_6, 150 MHz)



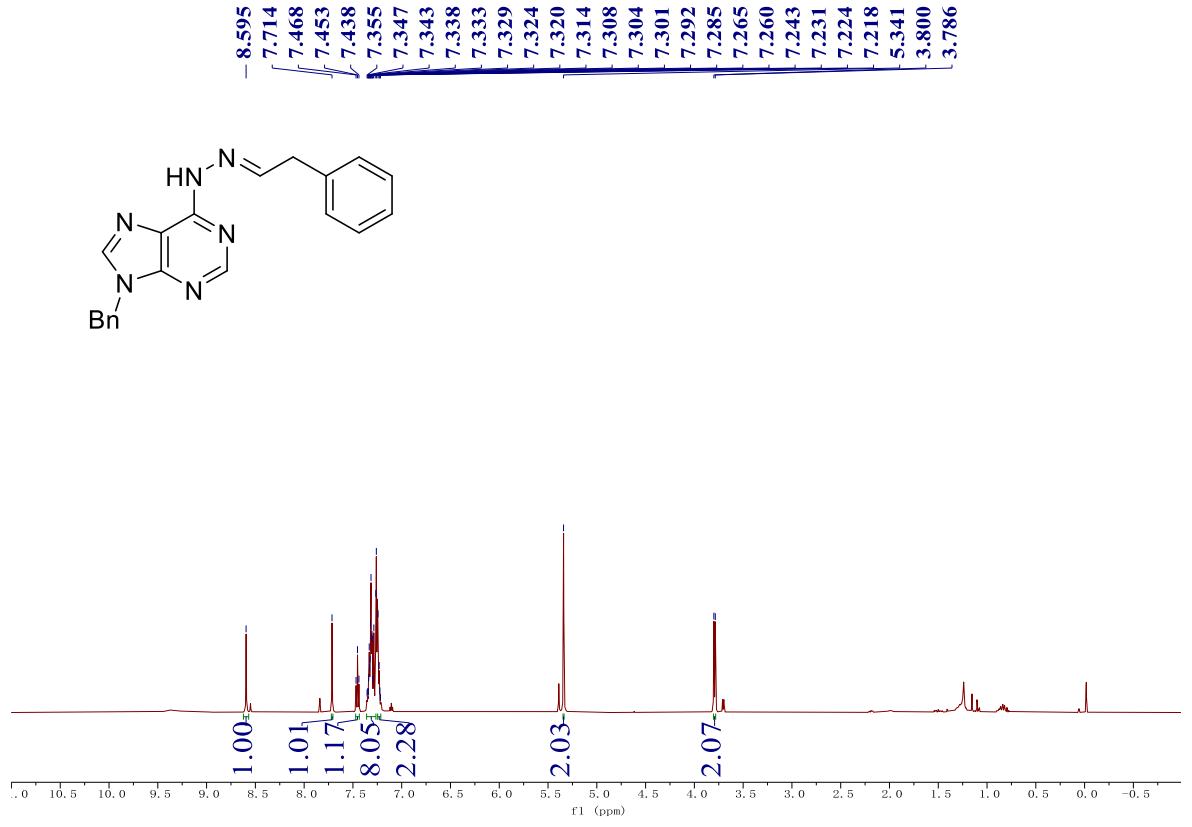
^{1}H NMR Spectrum of 1ab at 25 °C (CD_3OD, 400 MHz)



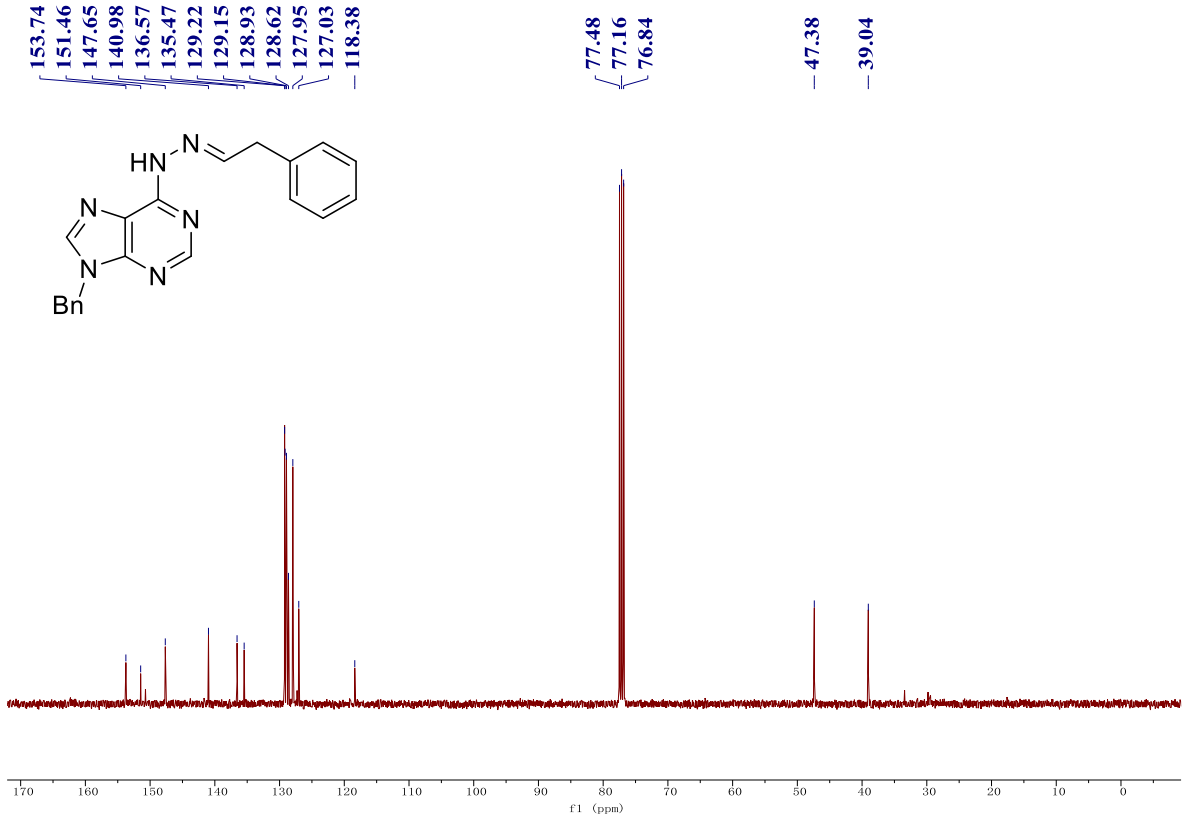
^{13}C NMR Spectrum of 1ab at 25 °C (CD_3OD, 100 MHz)



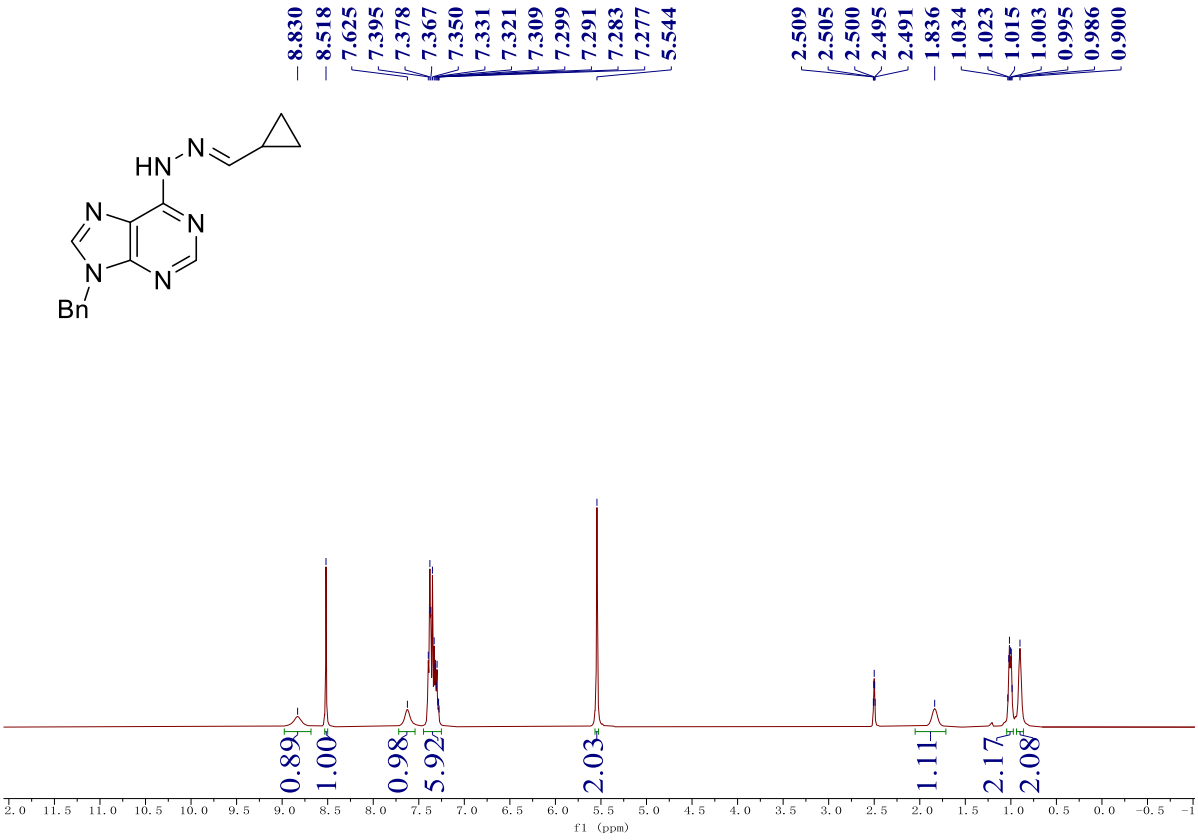
^{1}H NMR Spectrum of 1ac at 25 °C ($CDCl_3$, 400 MHz)



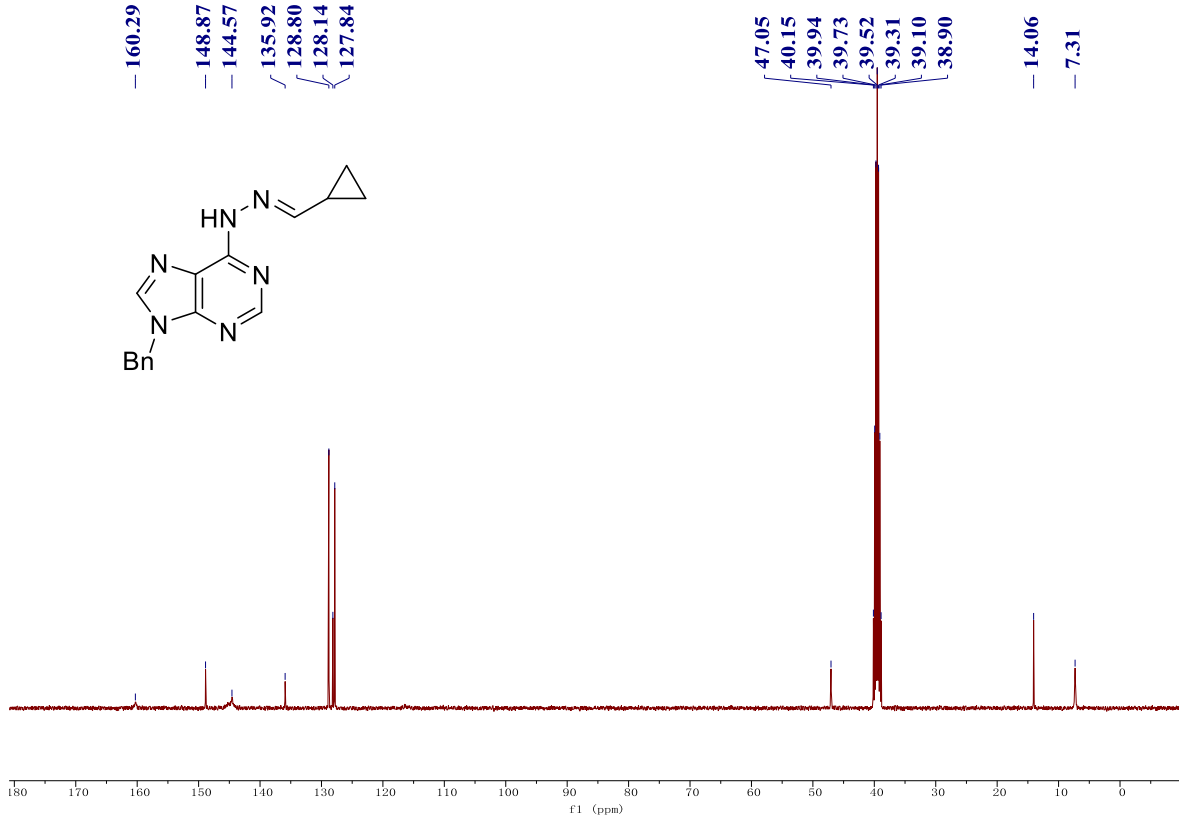
^{13}C NMR Spectrum of 1ac at 25 °C ($CDCl_3$, 100 MHz)



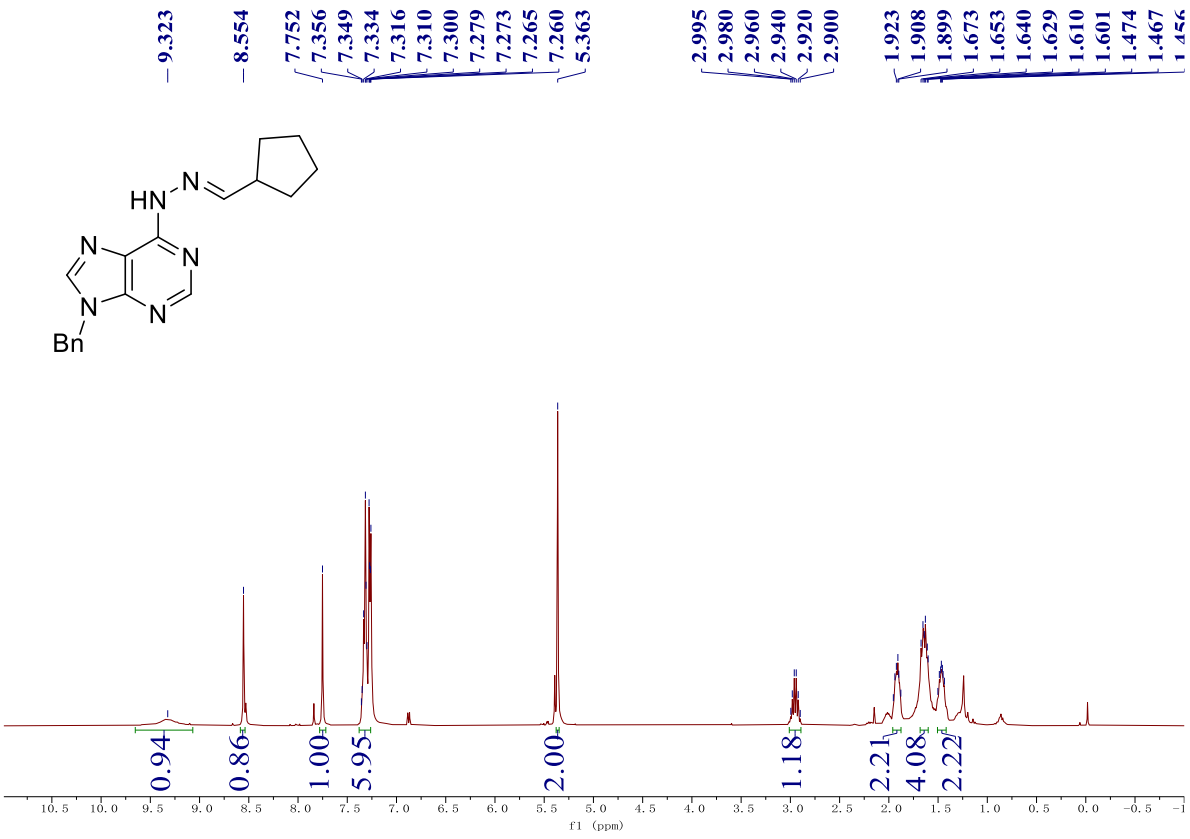
^{1}H NMR Spectrum of 1ad at 25 °C (DMSO-d_6, 400 MHz)



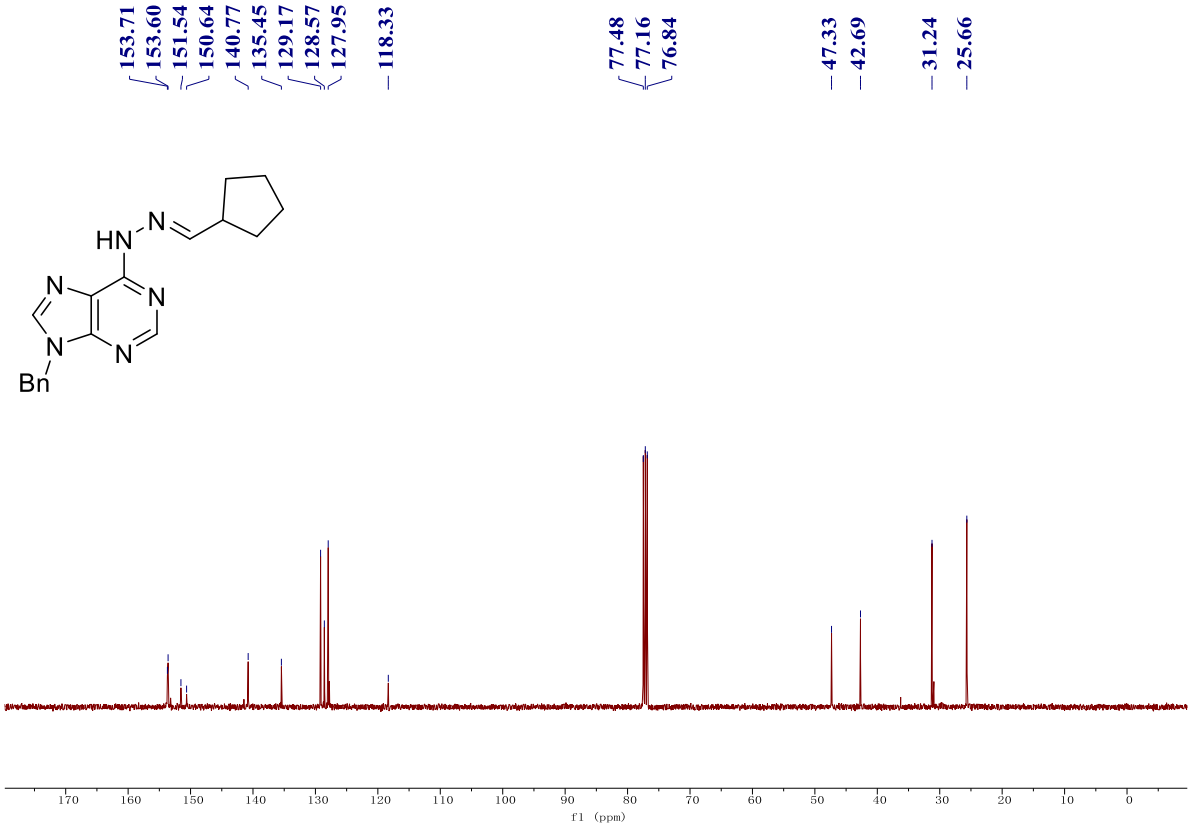
^{13}C NMR Spectrum of 1ad at 25 °C (DMSO-d_6, 100 MHz)



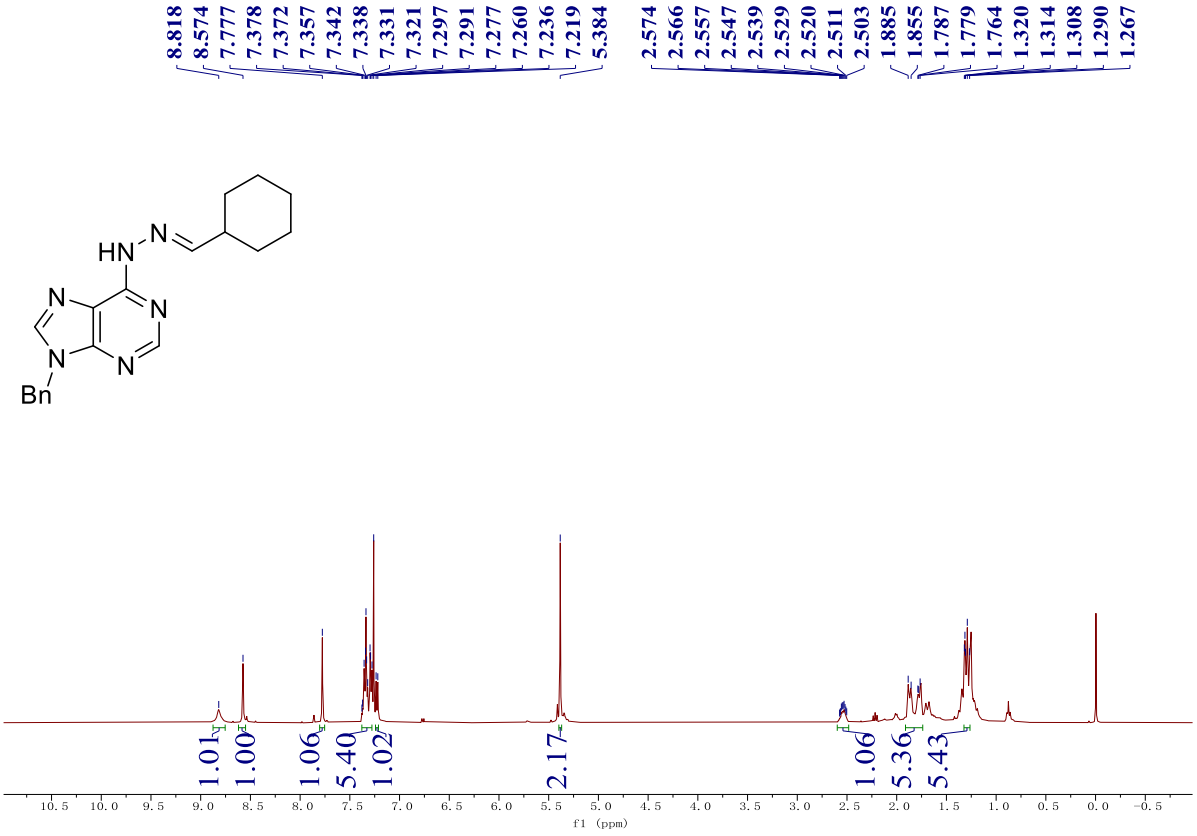
^{1}H NMR Spectrum of 1ae at 25 °C ($CDCl_3$, 400 MHz)



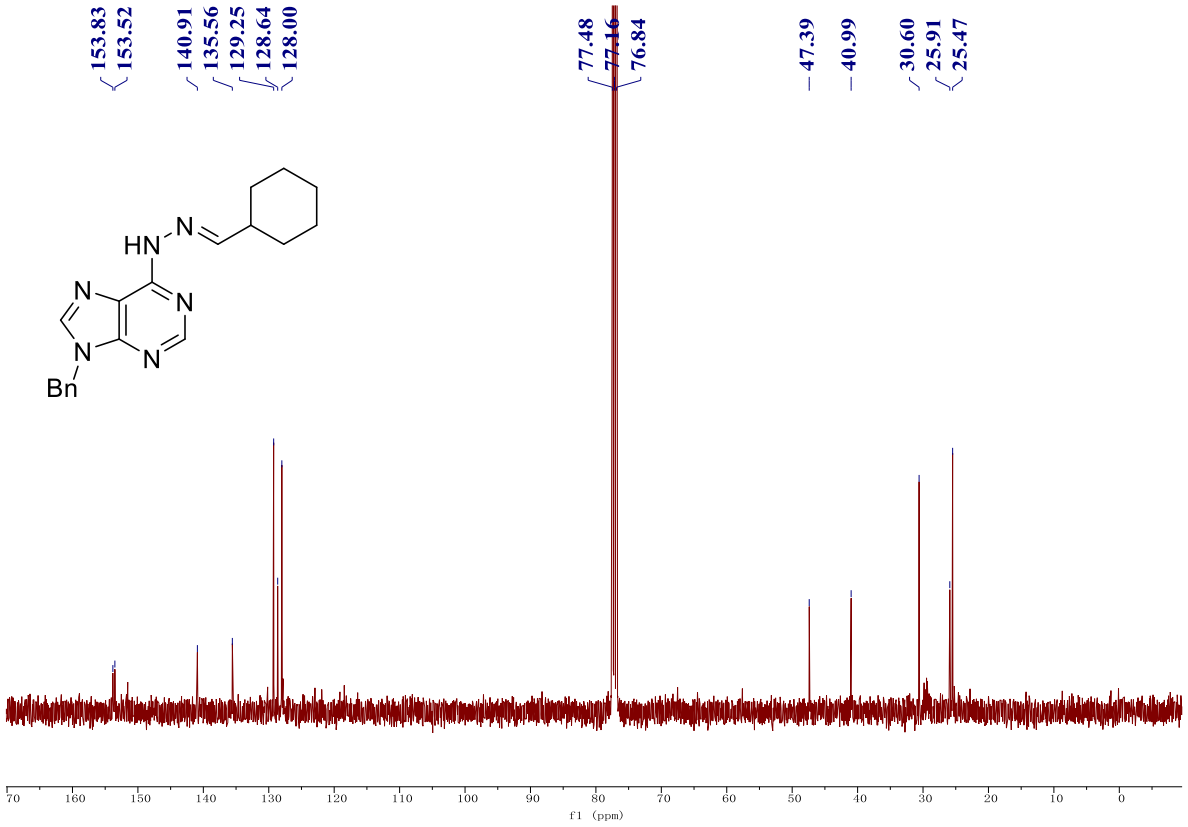
^{13}C NMR Spectrum of 1ae at 25 °C ($CDCl_3$, 100 MHz)



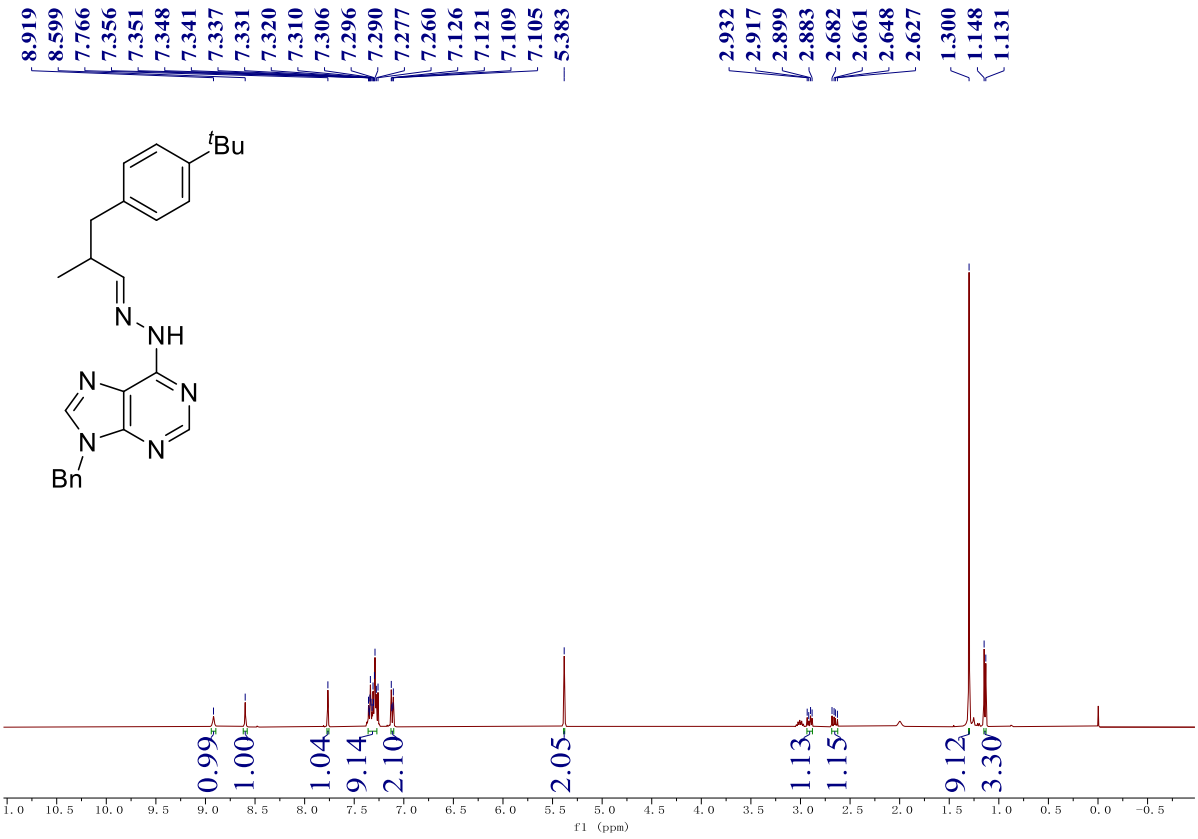
^{1}H NMR Spectrum of 1af at 25 °C ($CDCl_3$, 400 MHz)



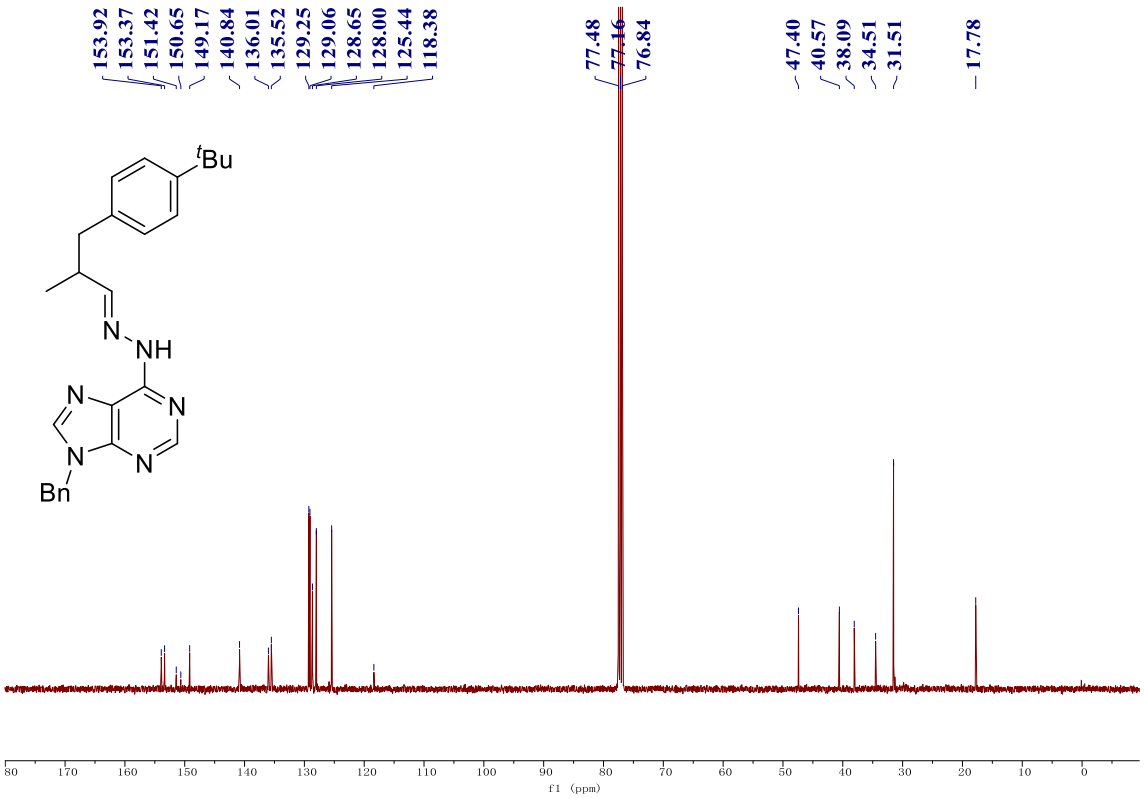
^{13}C NMR Spectrum of 1af at 25 °C ($CDCl_3$, 100 MHz)



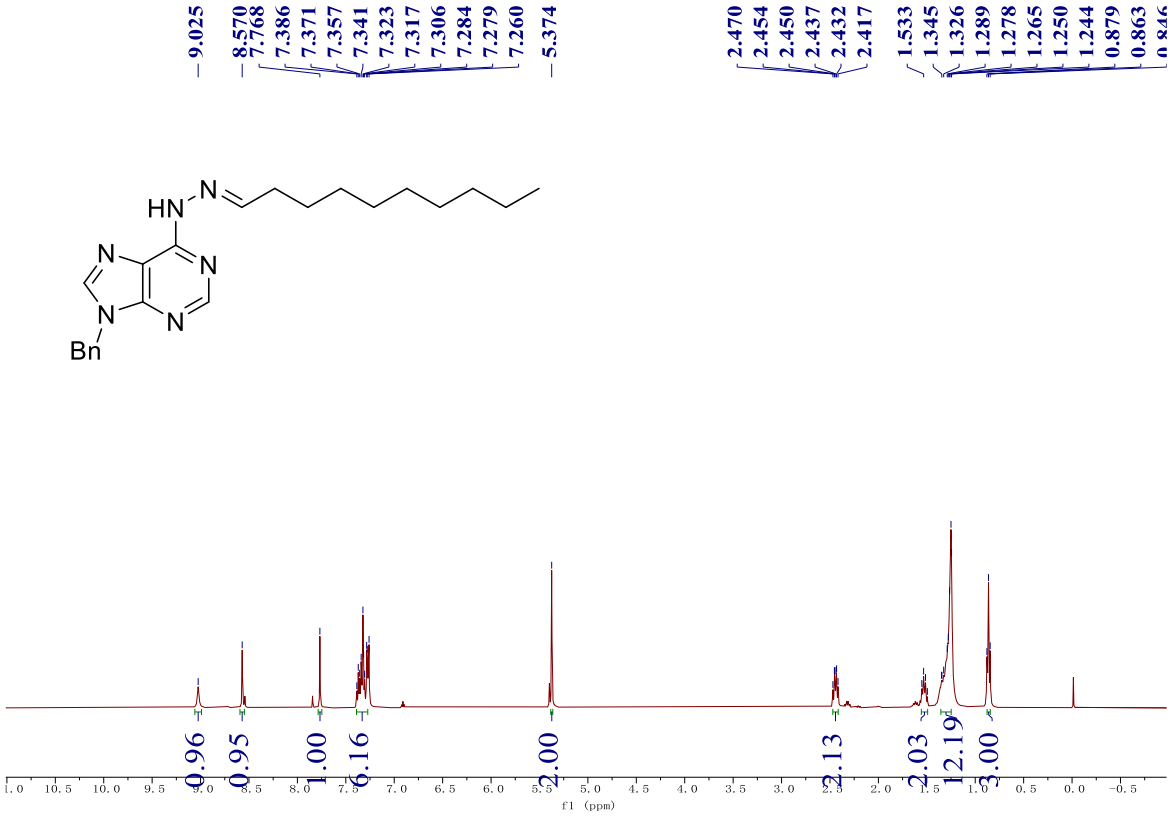
^{1}H NMR Spectrum of 1ag at 25 °C ($CDCl_3$, 400 MHz)



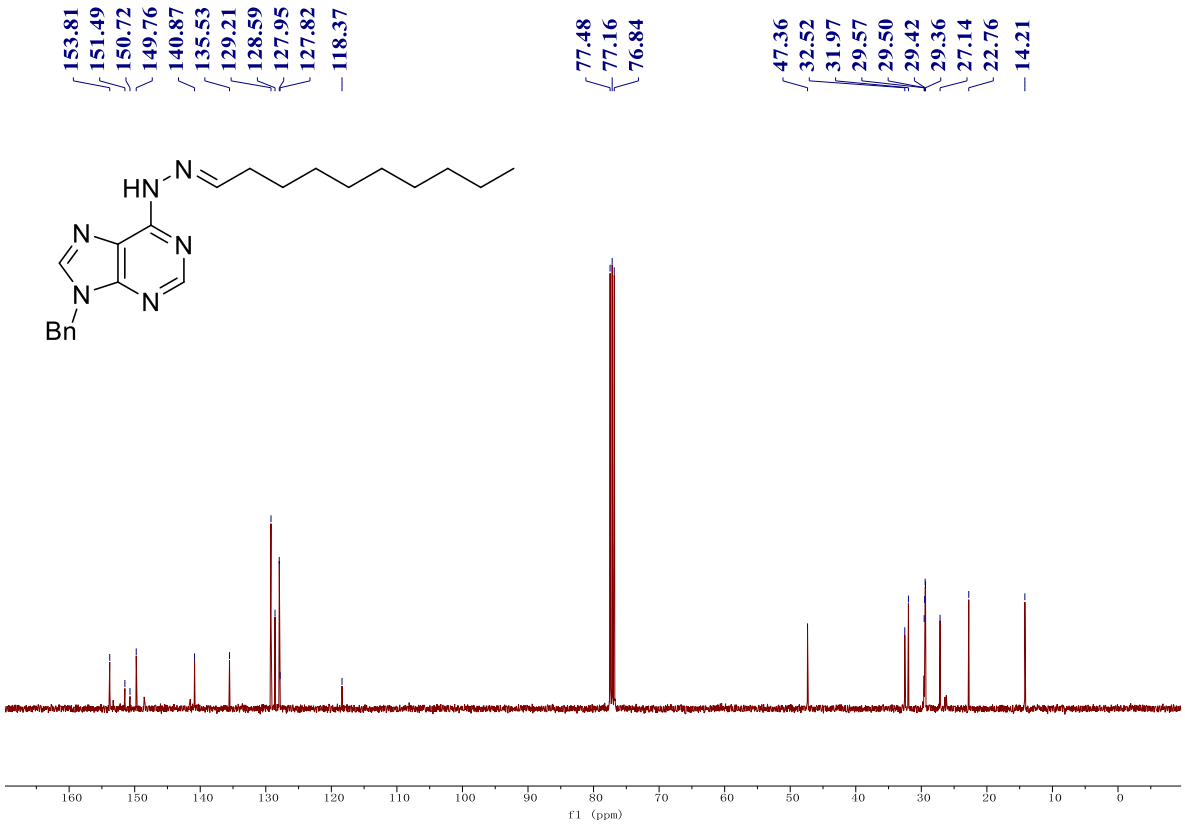
^{13}C NMR Spectrum of 1ag at 25 °C ($CDCl_3$, 100 MHz)



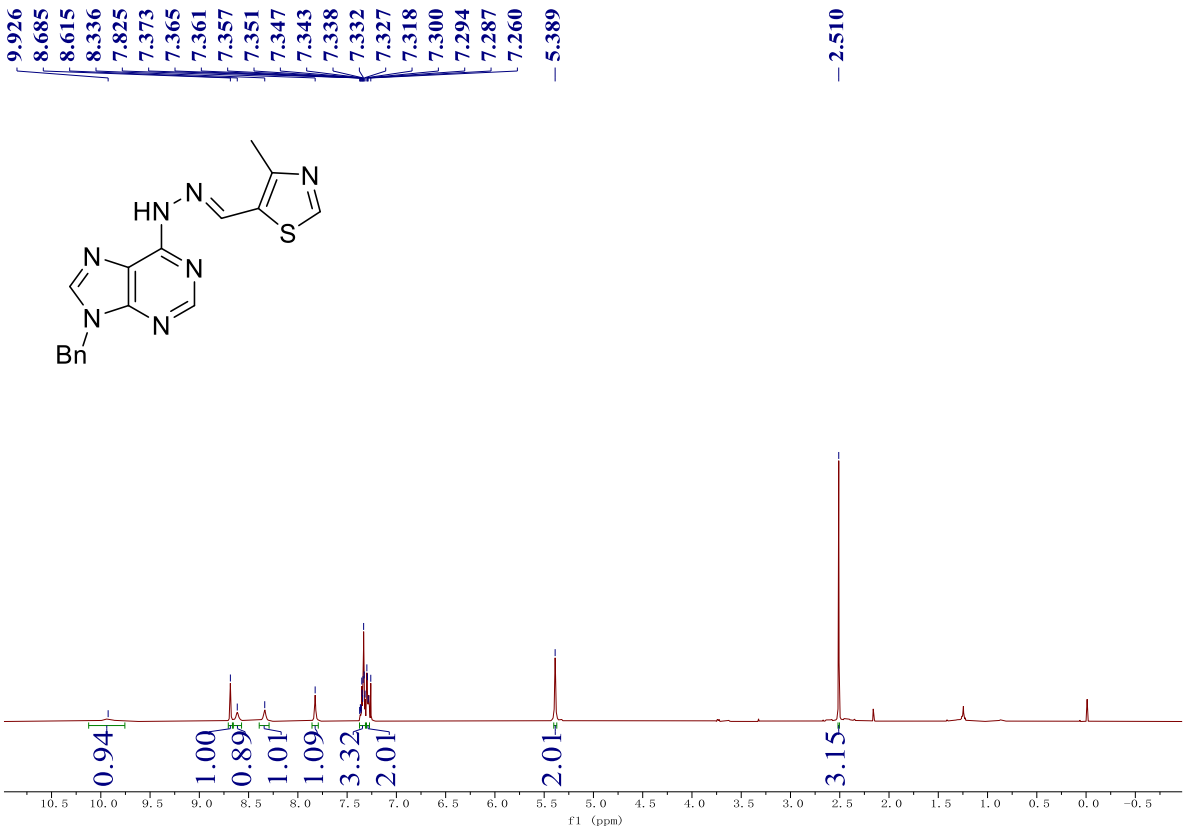
^{1}H NMR Spectrum of 1ah at 25 °C ($CDCl_3$, 400 MHz)



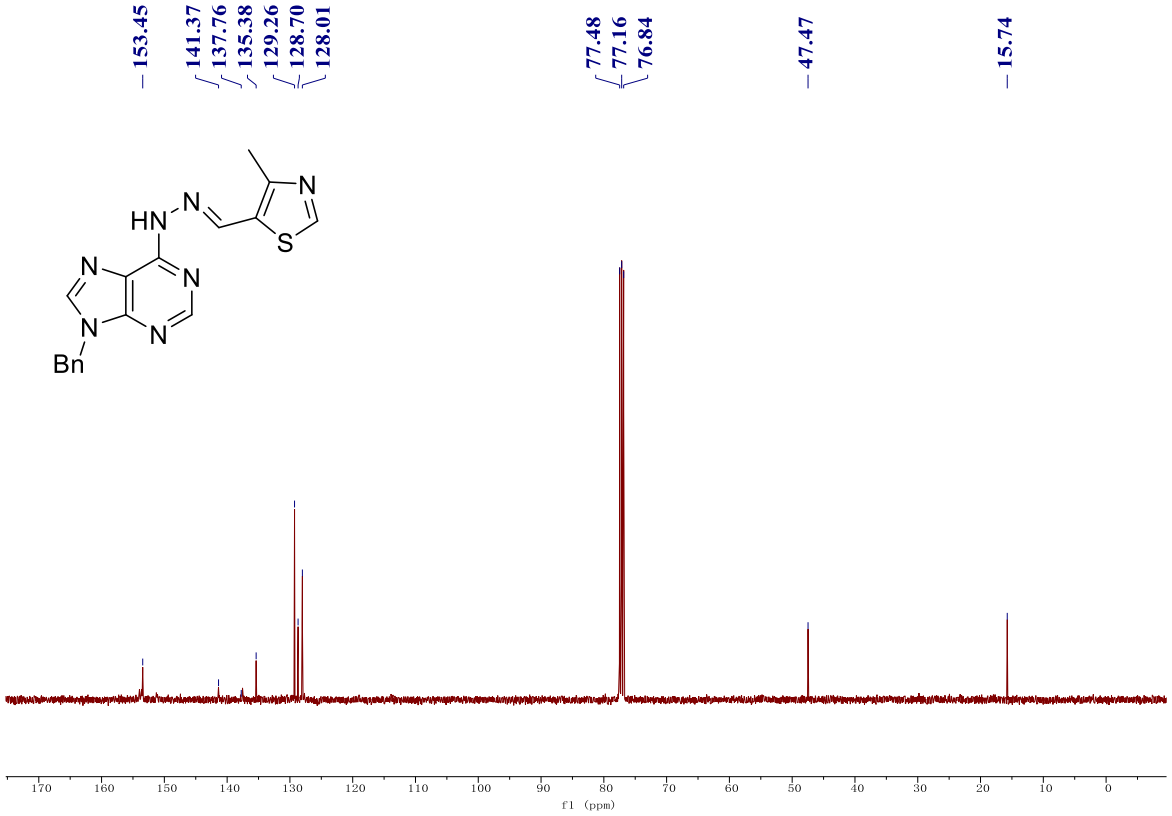
^{13}C NMR Spectrum of 1ah at 25 °C ($CDCl_3$, 100 MHz)



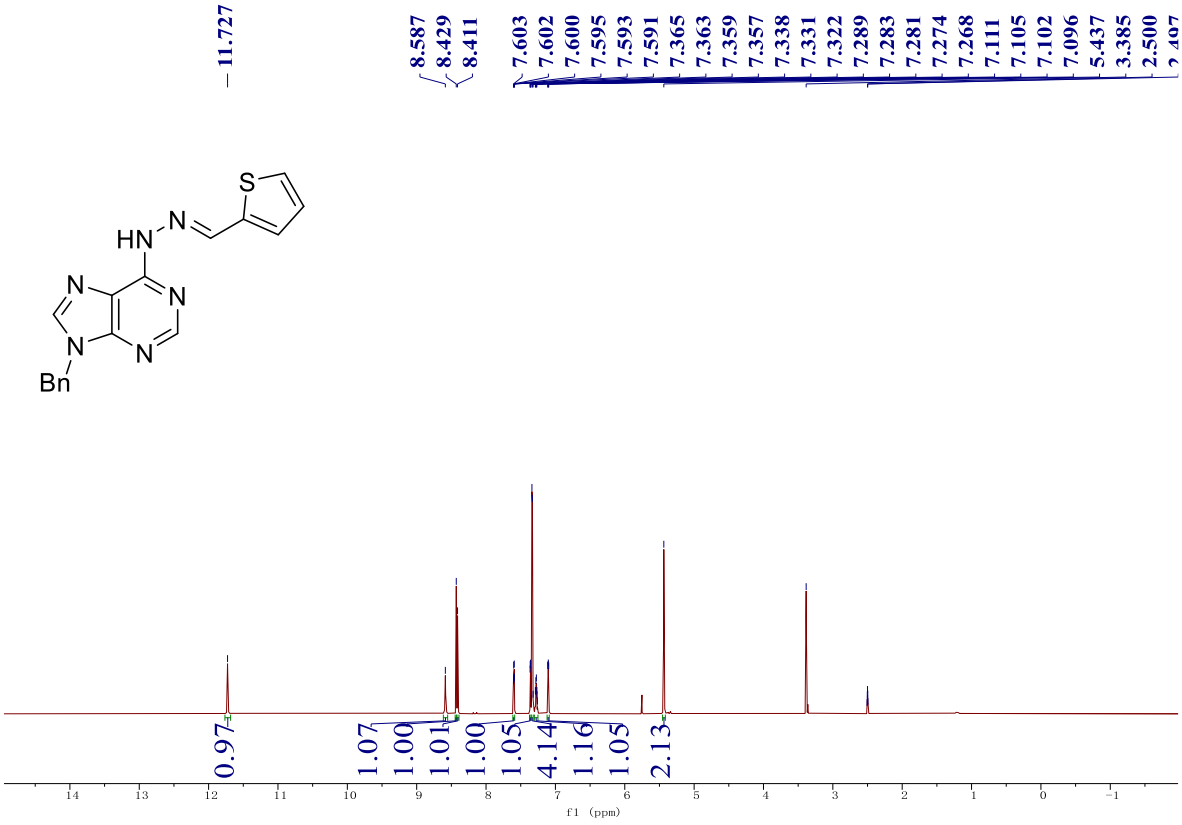
^{1}H NMR Spectrum of 1ai at 25 °C ($CDCl_3$, 400 MHz)



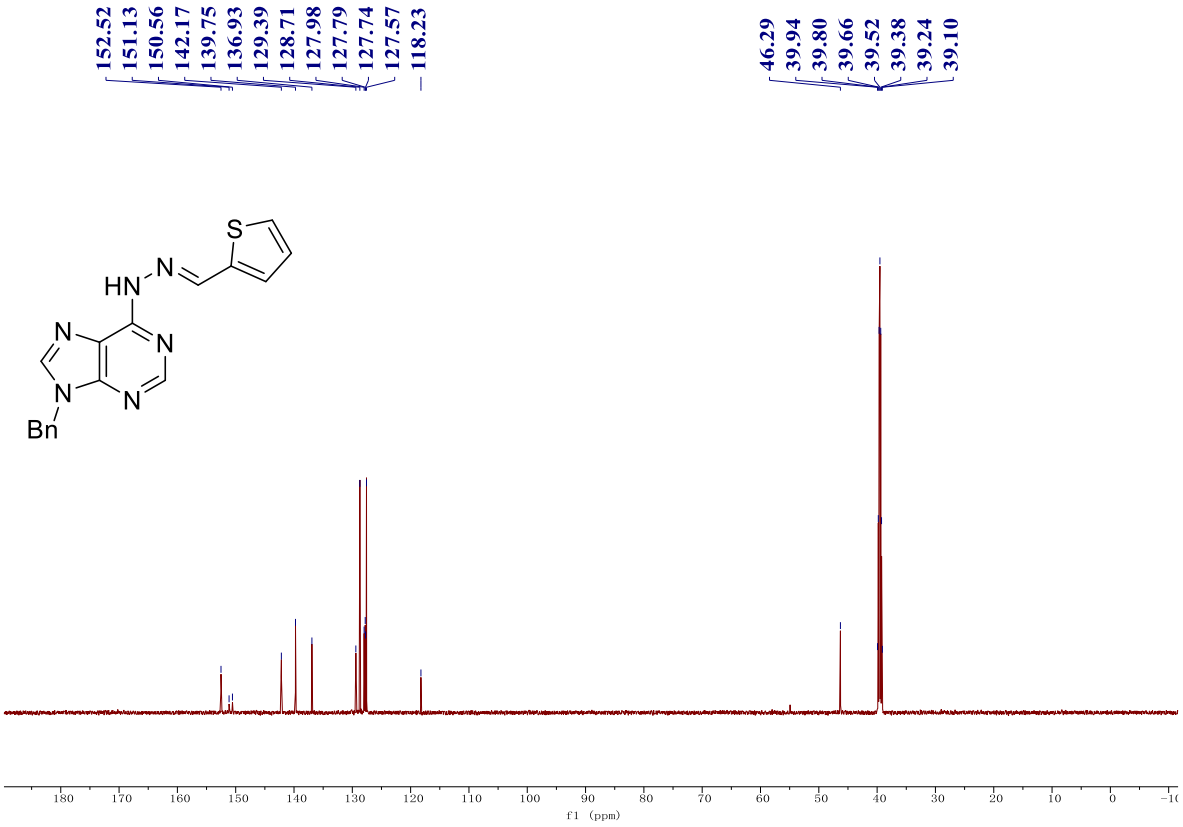
^{13}C NMR Spectrum of 1ai at 25 °C ($CDCl_3$, 100 MHz)



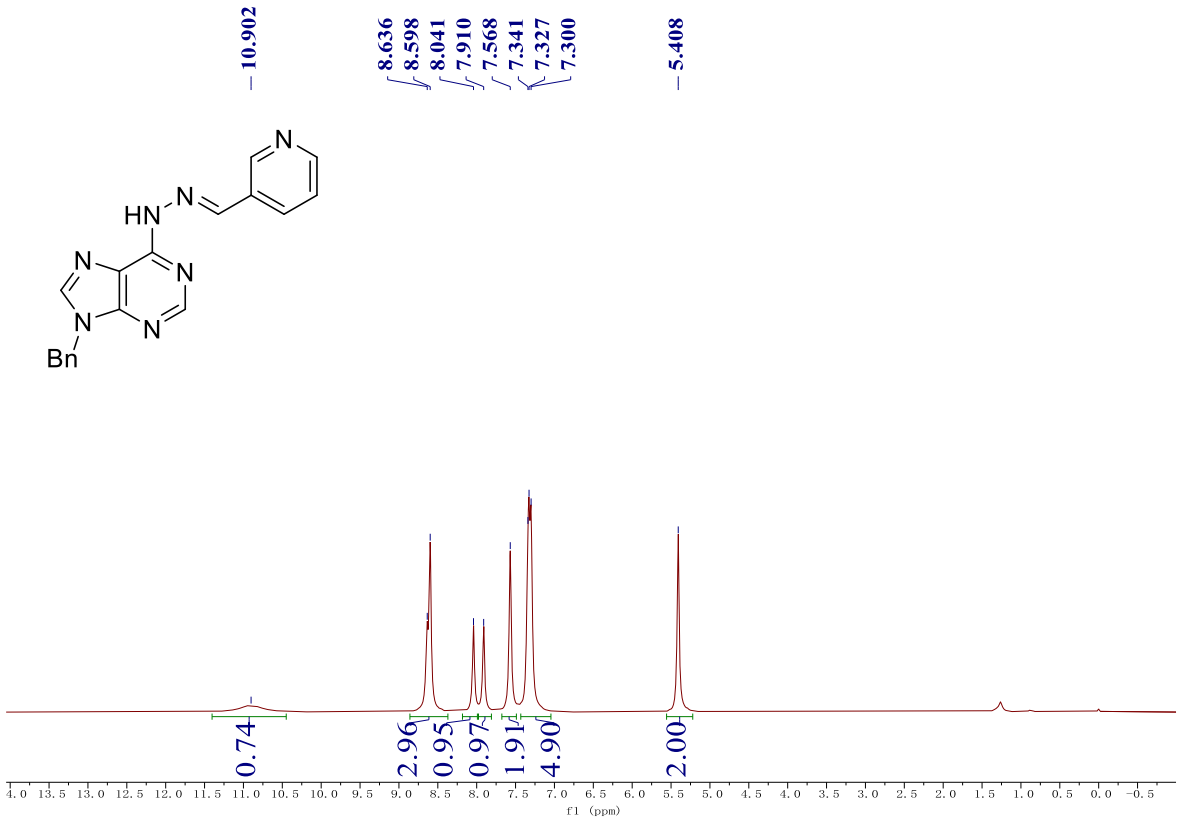
^{1}H NMR Spectrum of 1aj at 25 °C (DMSO-d_6, 600 MHz)



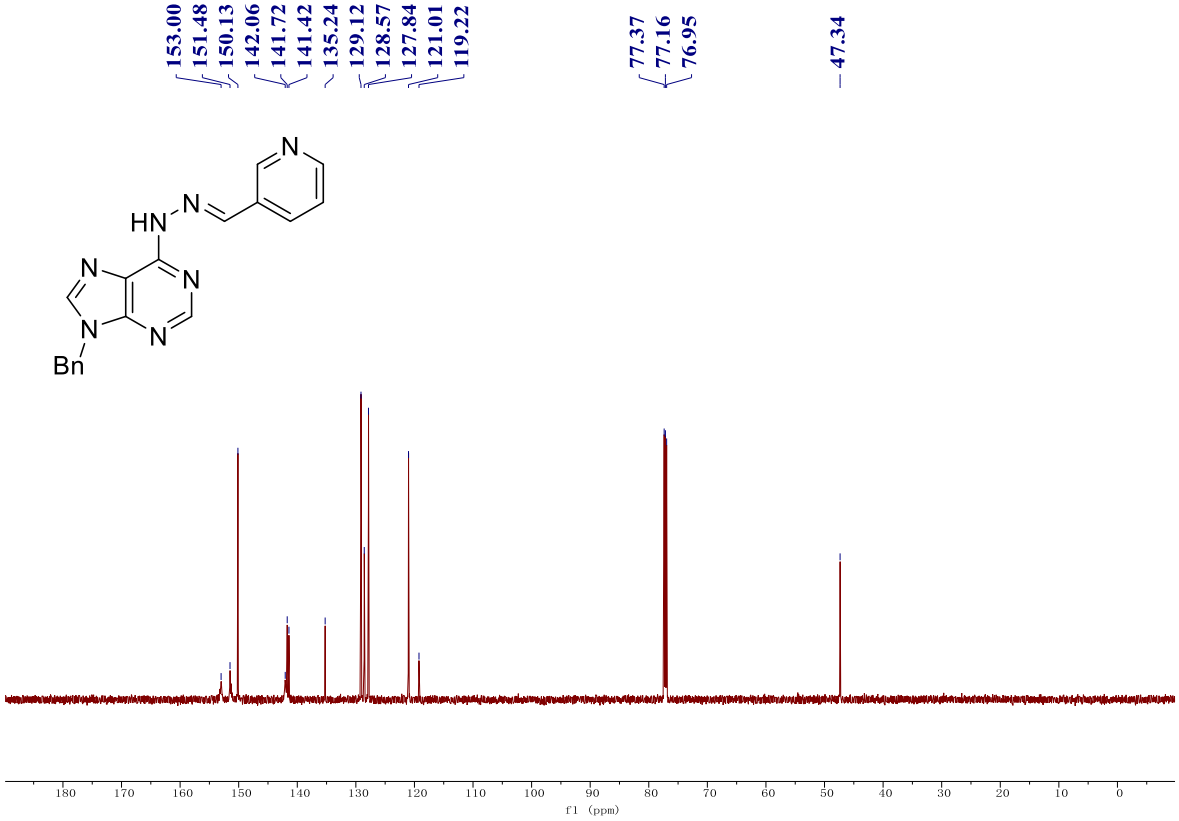
^{13}C NMR Spectrum of 1aj at 25 °C (DMSO-d_6, 150 MHz)



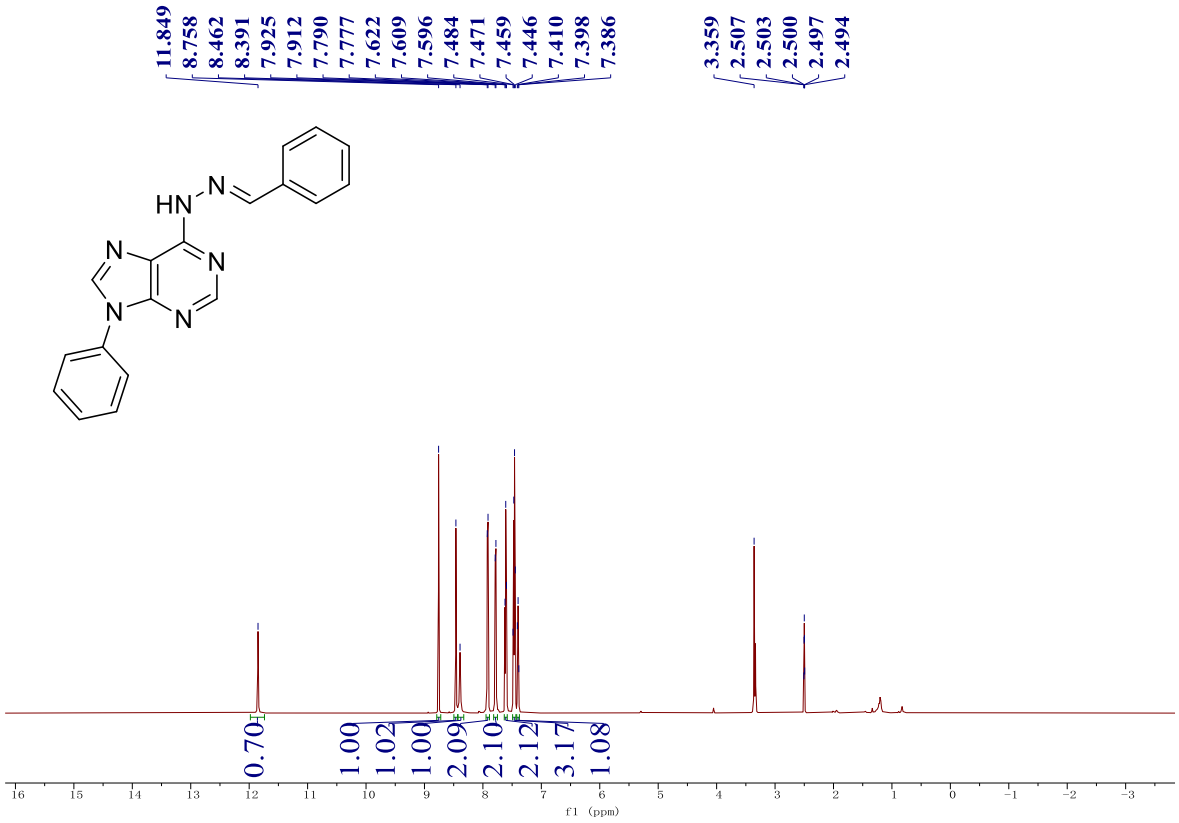
^{1}H NMR Spectrum of 1ak at 25 °C ($CDCl_3$, 600 MHz)



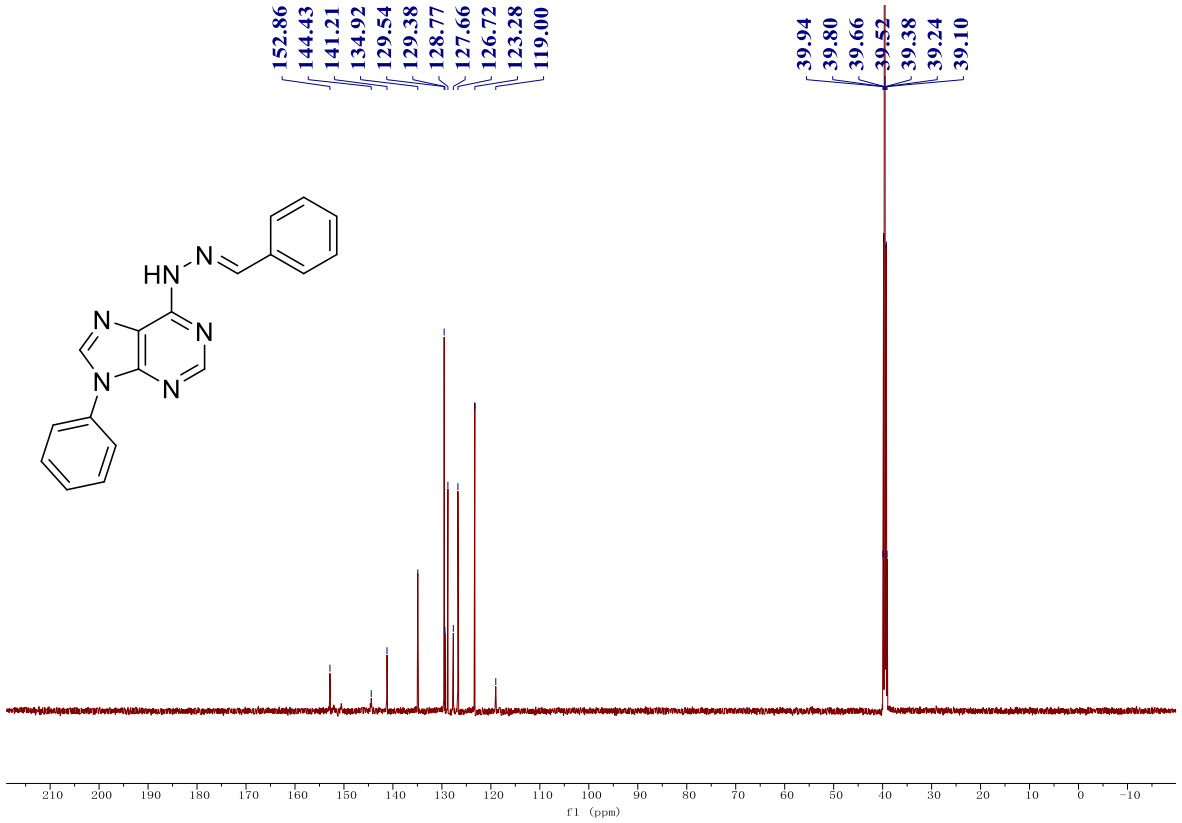
^{13}C NMR Spectrum of 1ak at 25 °C ($CDCl_3$, 150 MHz)



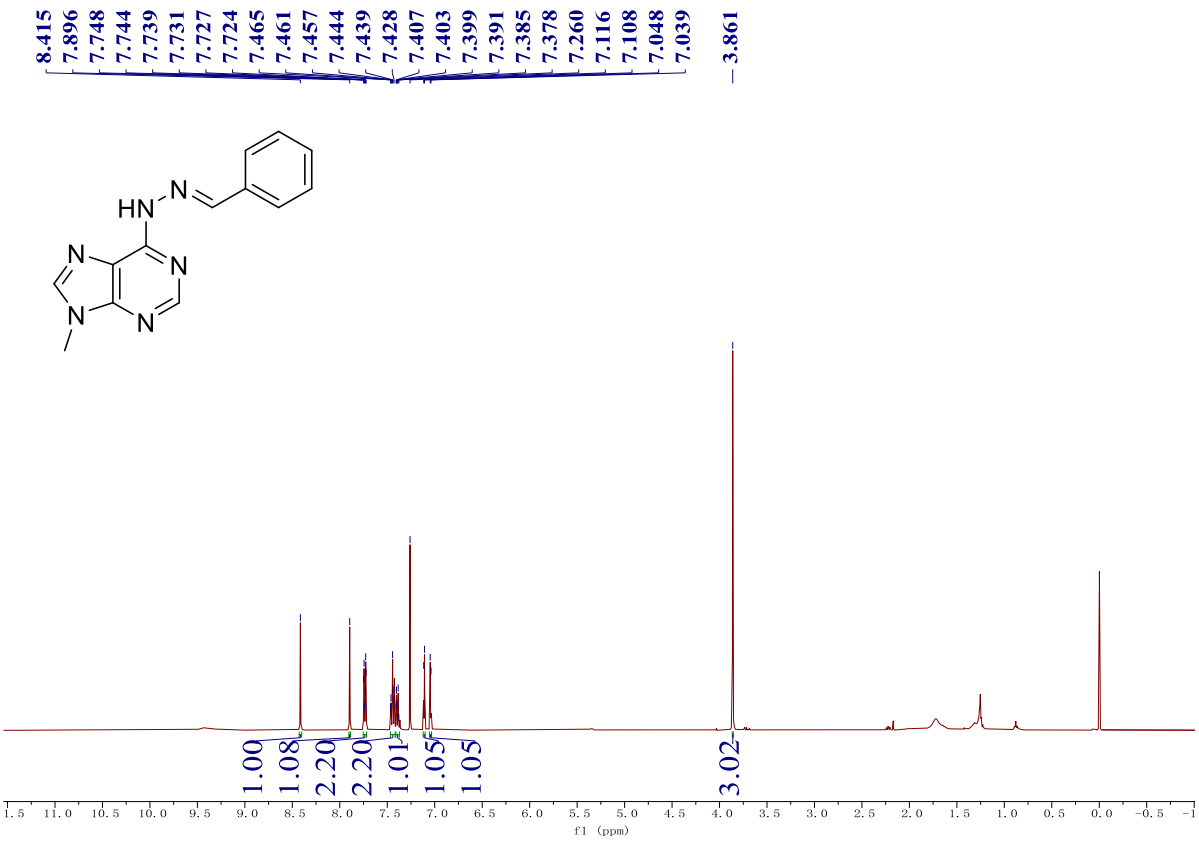
^{1}H NMR Spectrum of 1al at 25 °C (DMSO-d_6, 600 MHz)



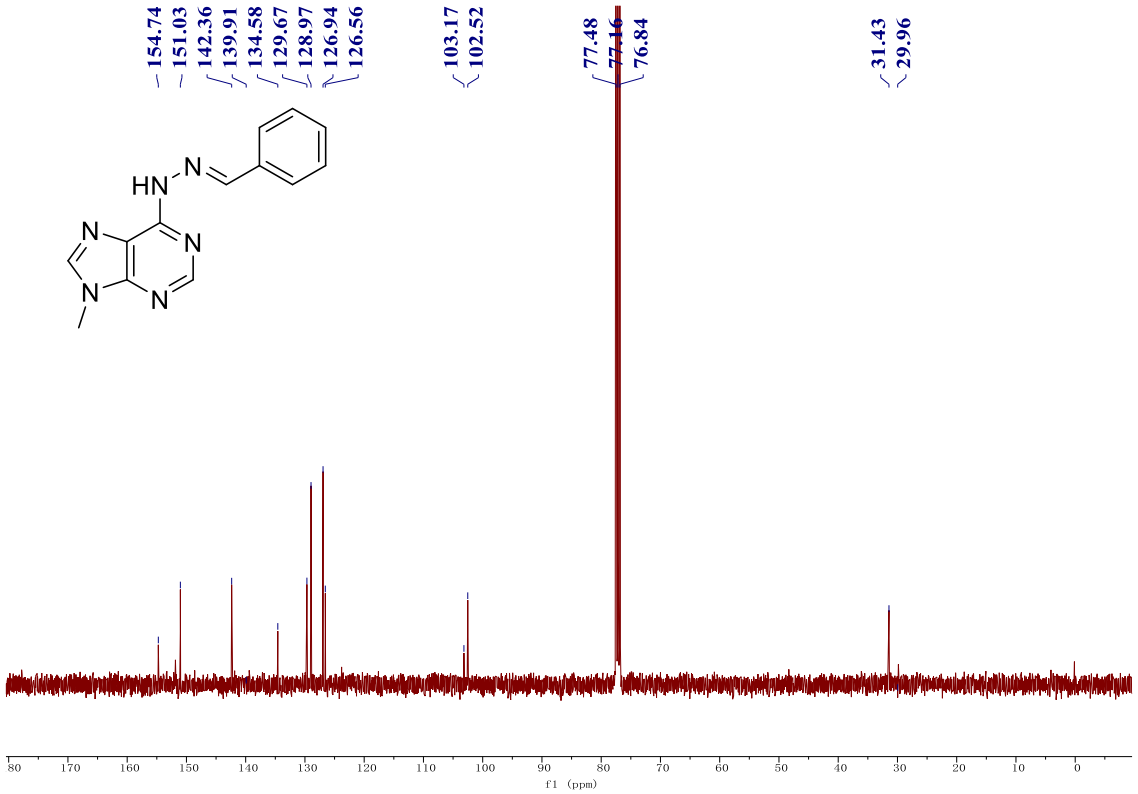
^{13}C NMR Spectrum of 1al at 25 °C (DMSO-d_6, 150 MHz)



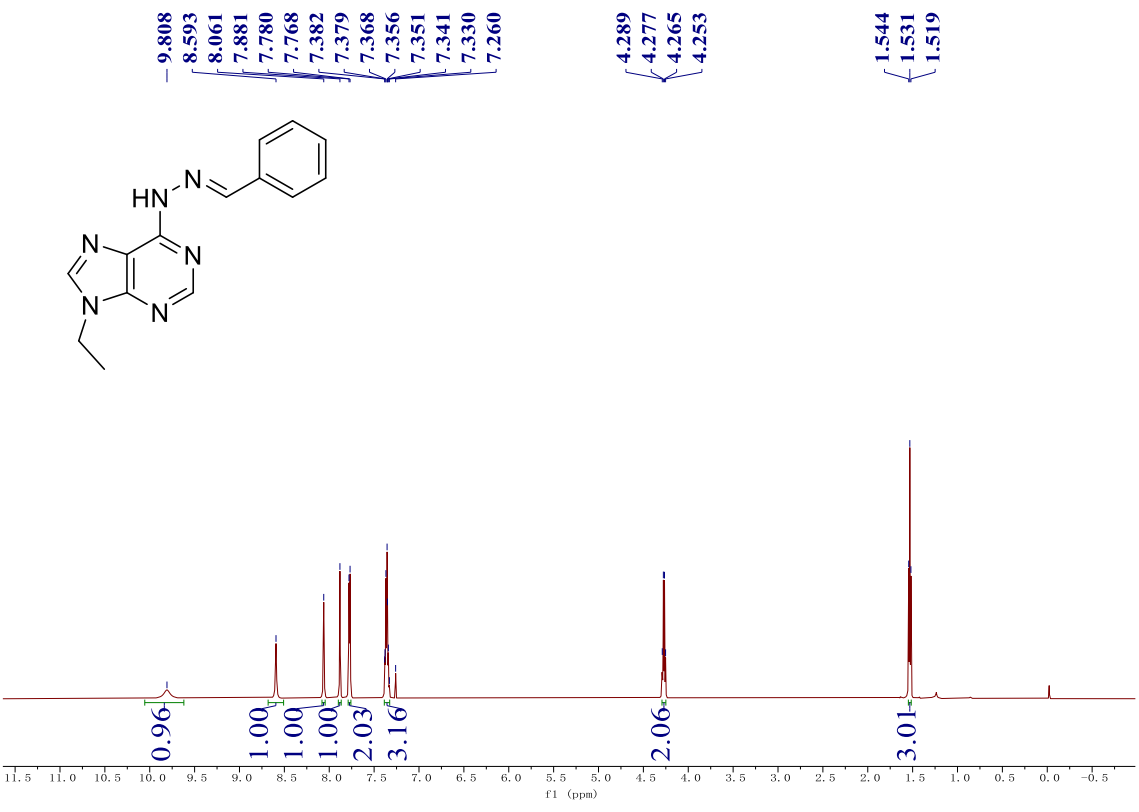
^{1}H NMR Spectrum of 1am at 25 °C ($CDCl_3$, 400 MHz)



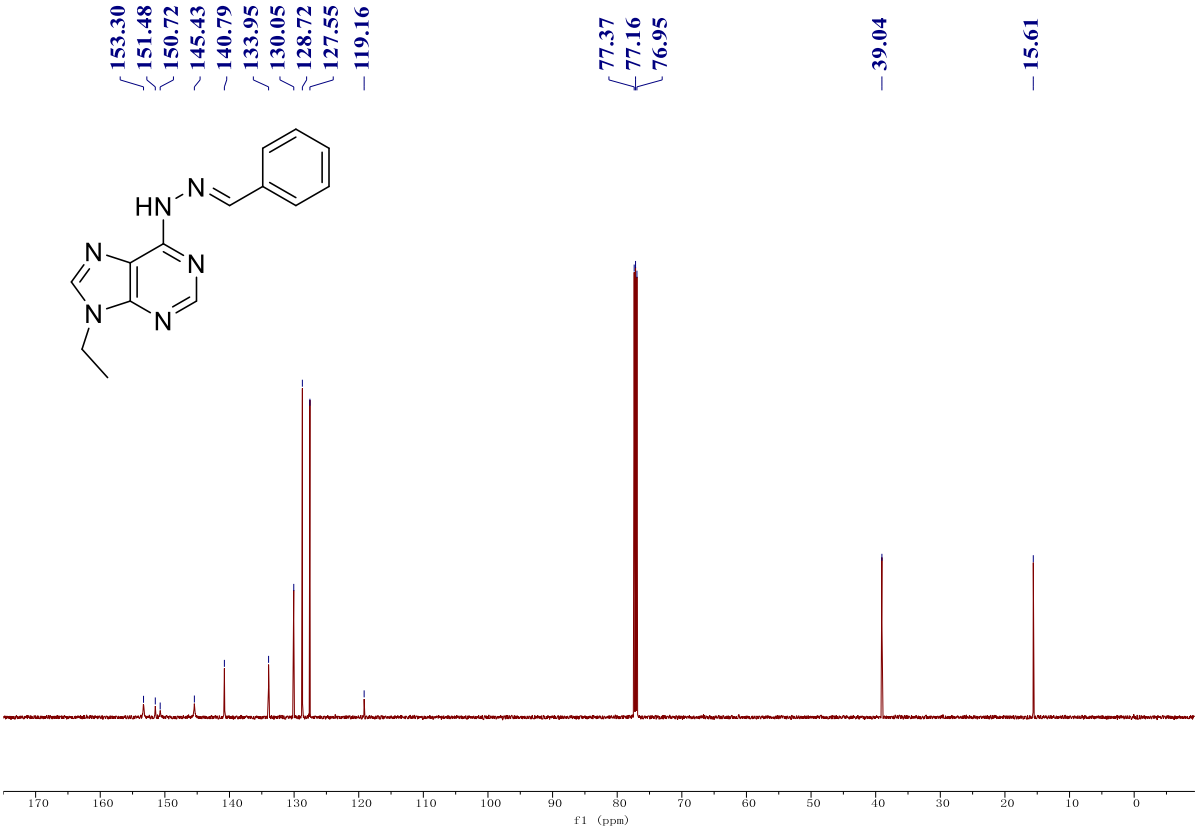
^{13}C NMR Spectrum of 1am at 25 °C ($CDCl_3$, 100 MHz)



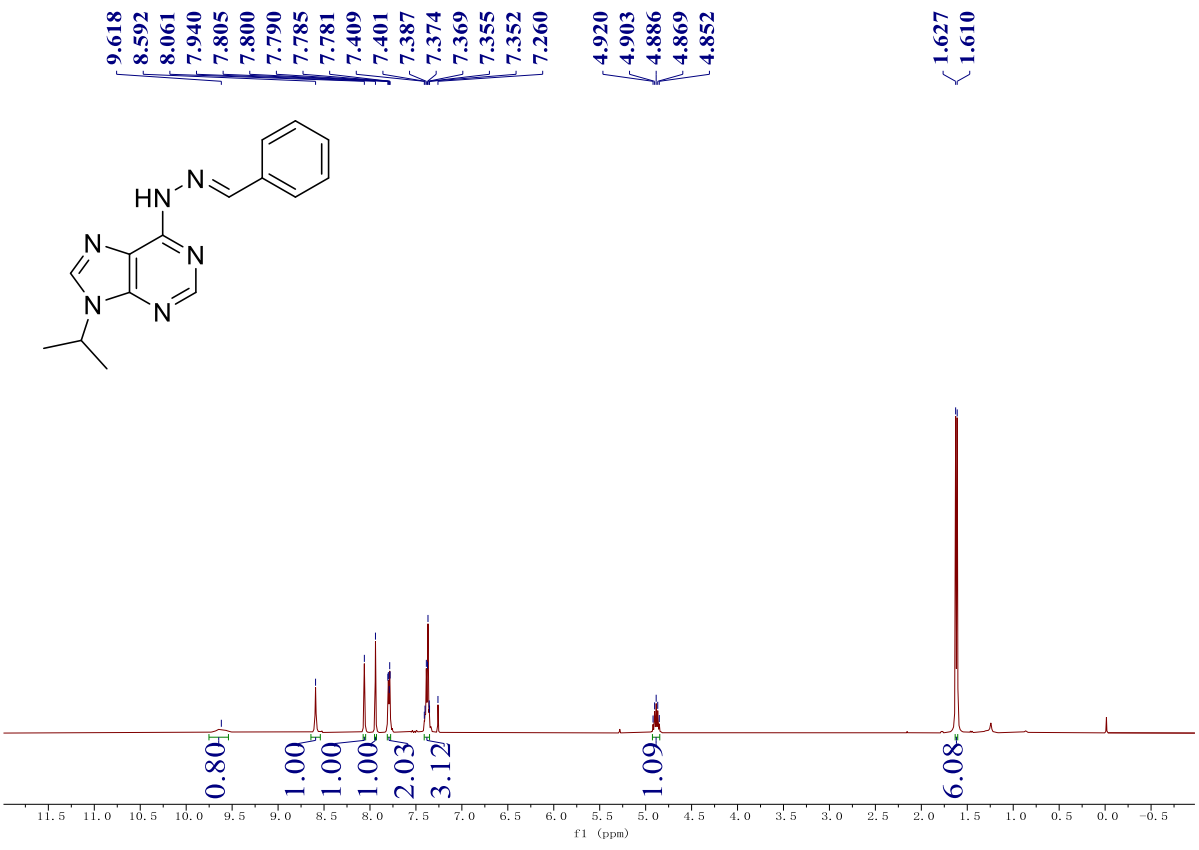
^{1}H NMR Spectrum of 1an at 25 °C ($CDCl_3$, 600 MHz)



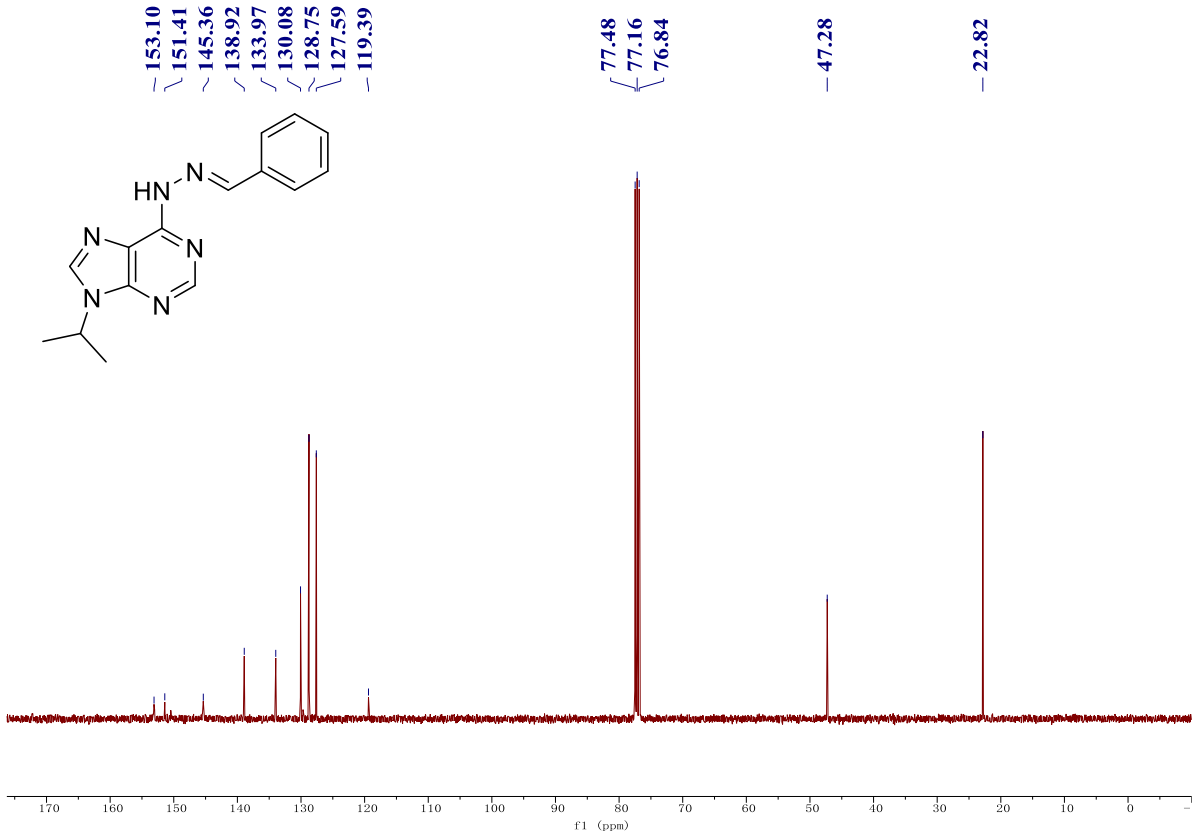
^{13}C NMR Spectrum of 1an at 25 °C ($CDCl_3$, 150 MHz)



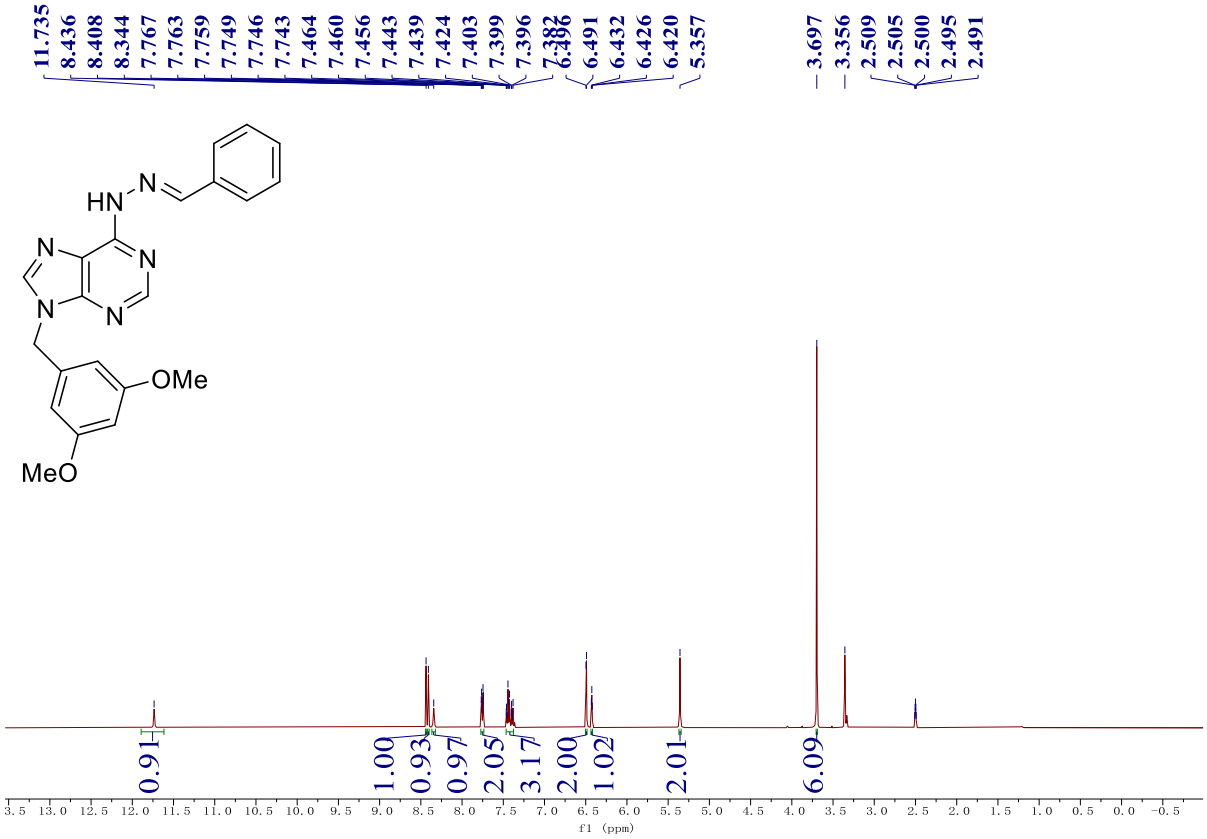
^{1}H NMR Spectrum of 1ao at 25 °C ($CDCl_3$, 400 MHz)



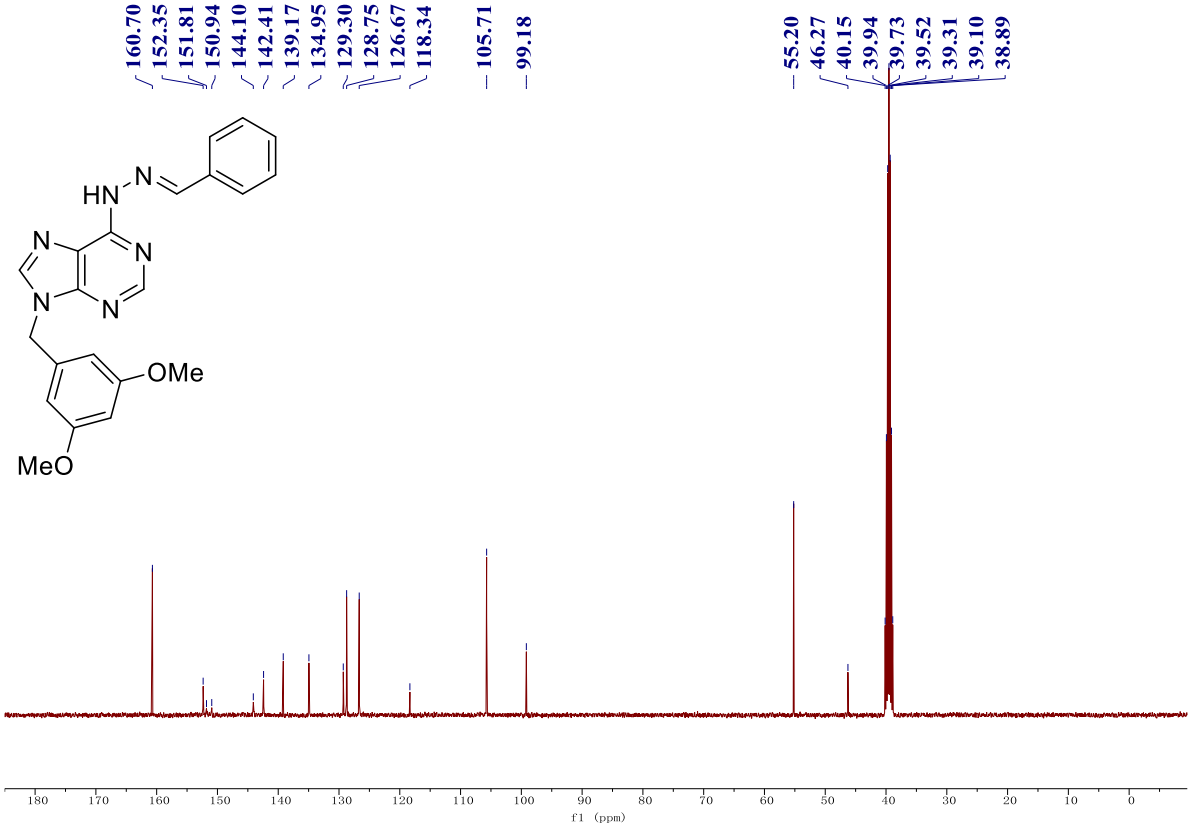
^{13}C NMR Spectrum of 1ao at 25 °C ($CDCl_3$, 100 MHz)



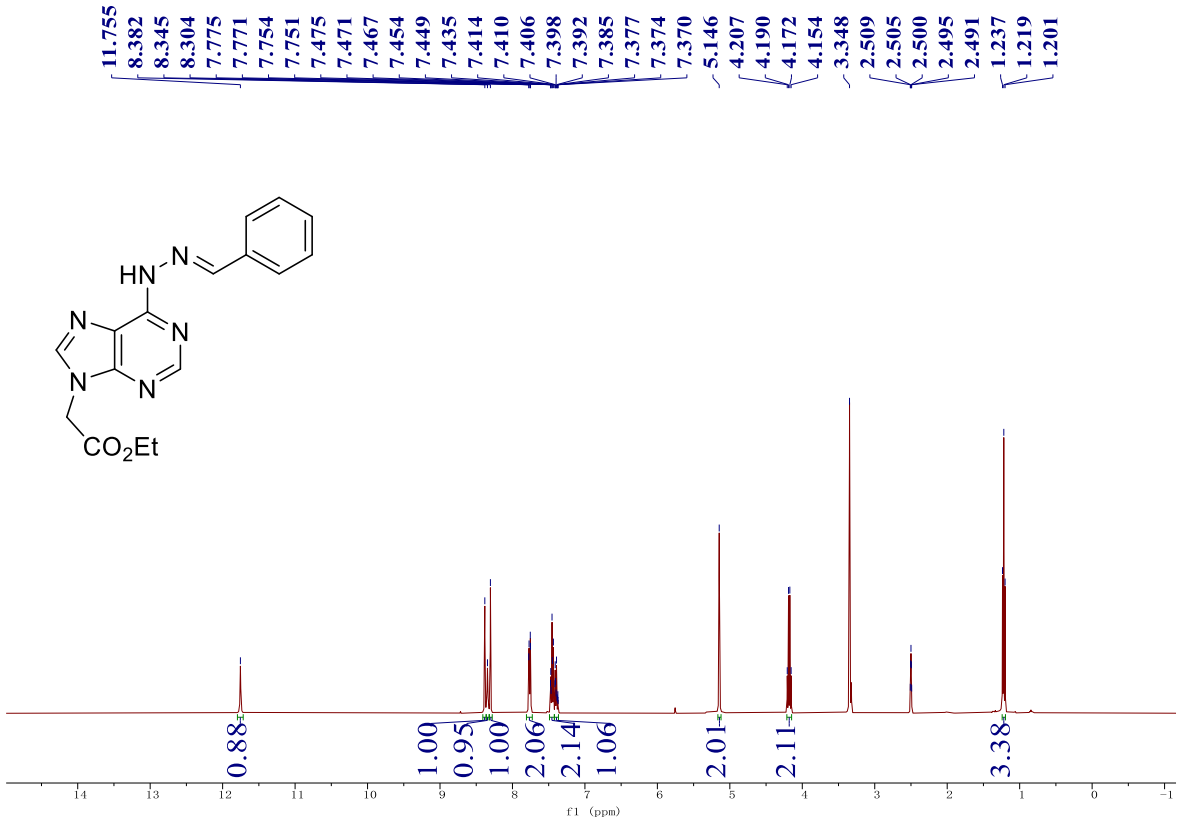
^{1}H NMR Spectrum of 1ap at 25 °C (DMSO-d_6, 400 MHz)



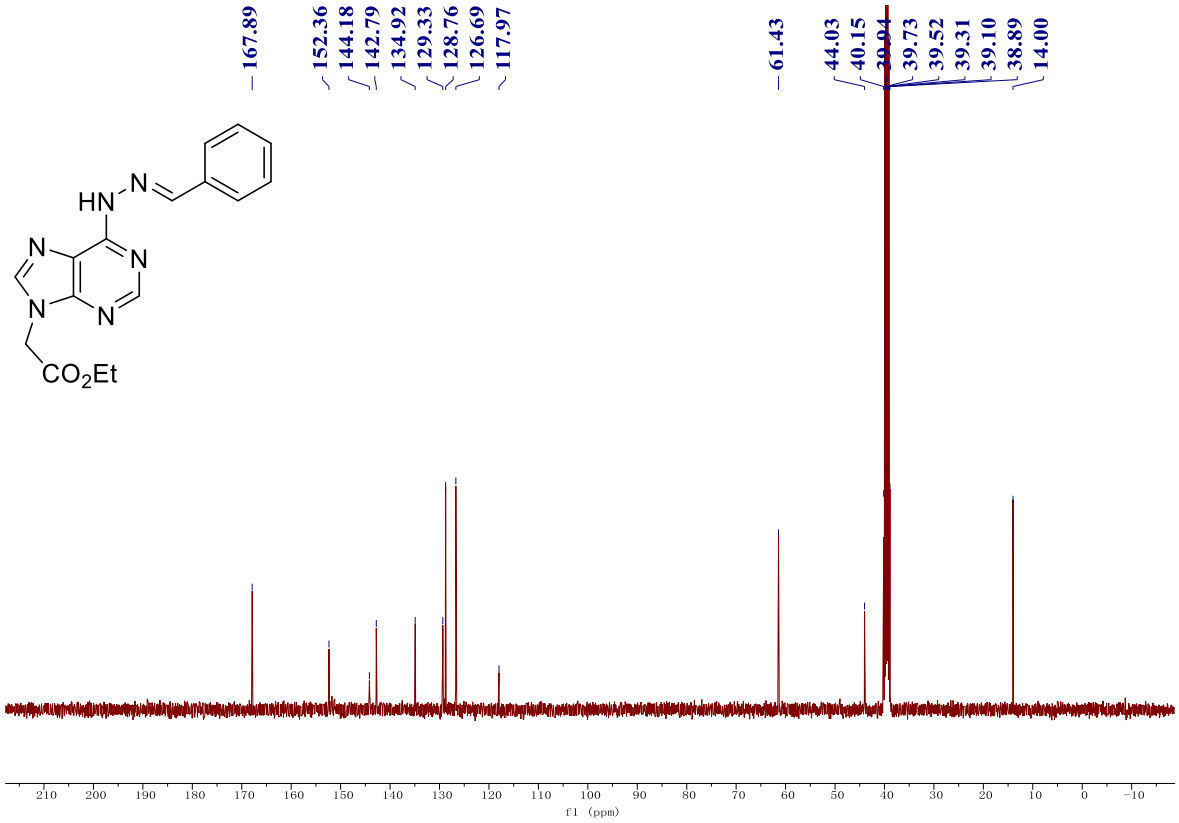
^{13}C NMR Spectrum of 1ap at 25 °C (DMSO-d_6, 100 MHz)



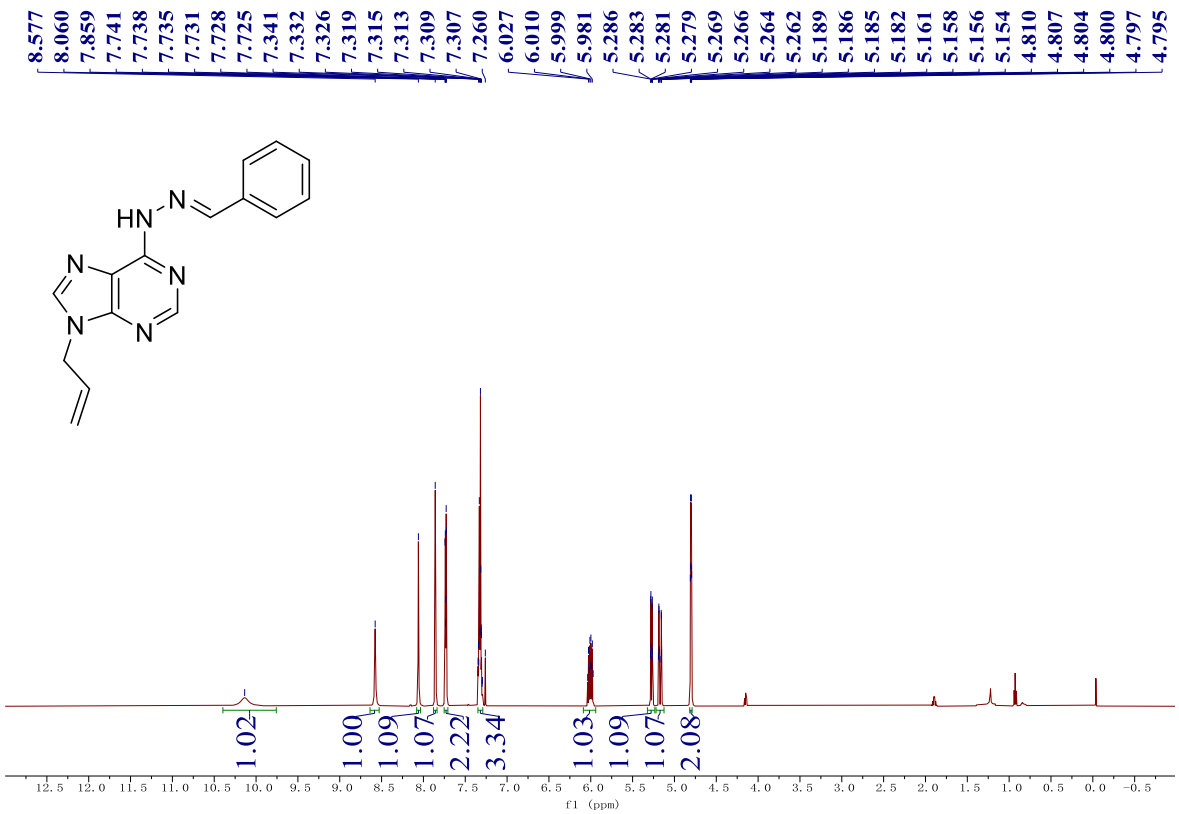
^{1}H NMR Spectrum of 1aq at 25 °C (DMSO-d_6, 400 MHz)



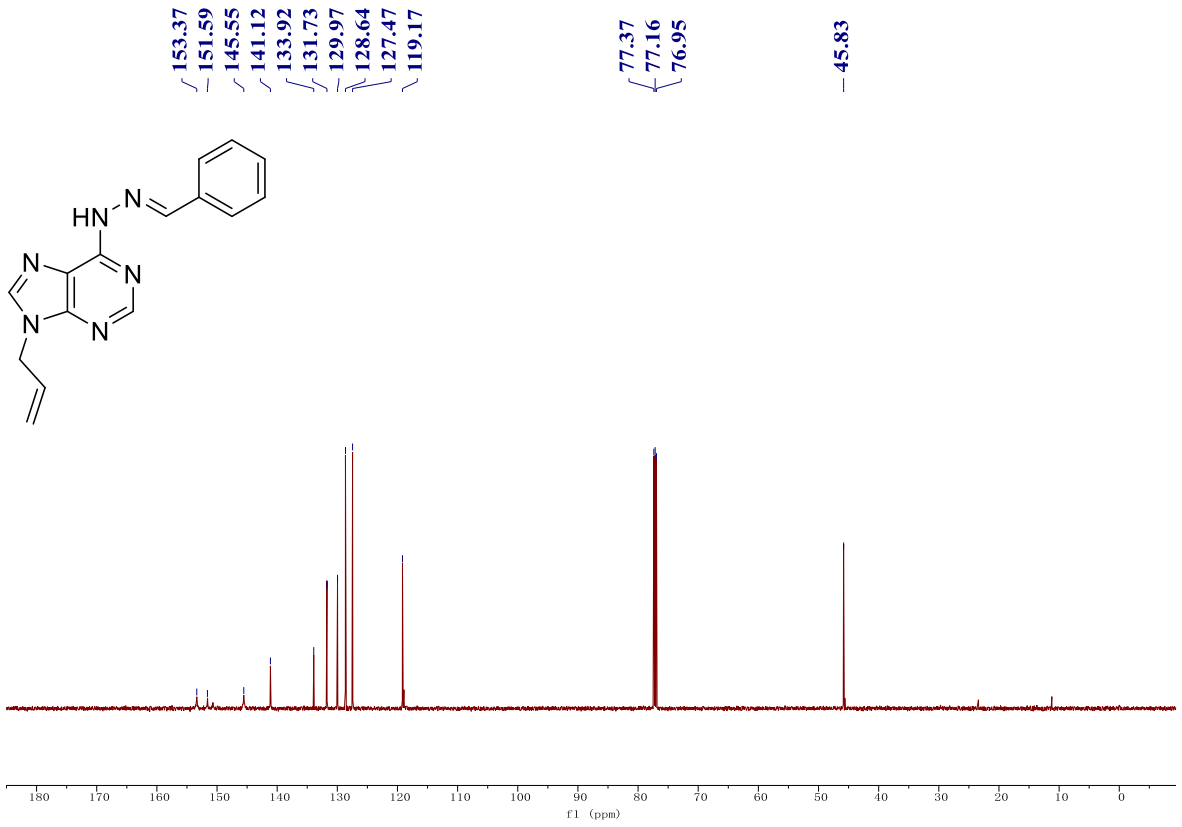
^{13}C NMR Spectrum of 1aq at 25 °C (DMSO-d_6, 100 MHz)



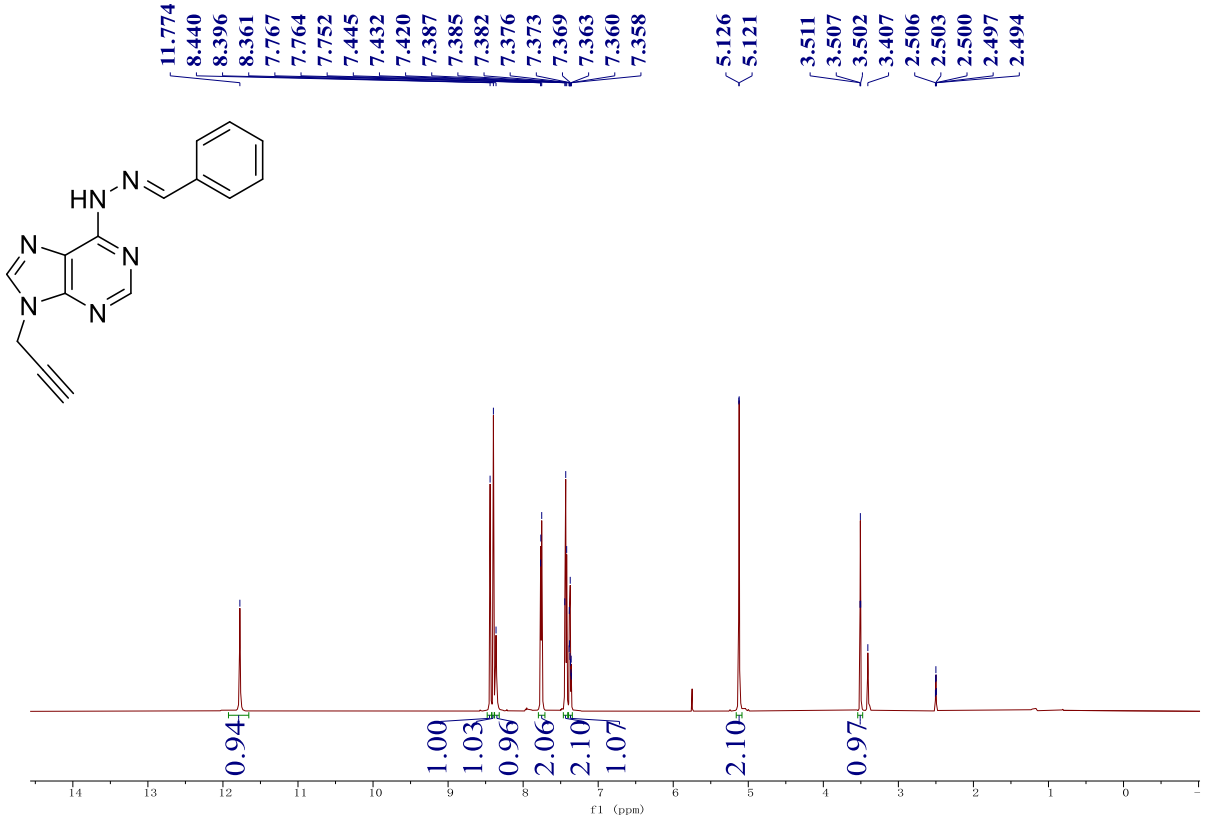
^{1}H NMR Spectrum of 1ar at 25 °C ($CDCl_3$, 600 MHz)



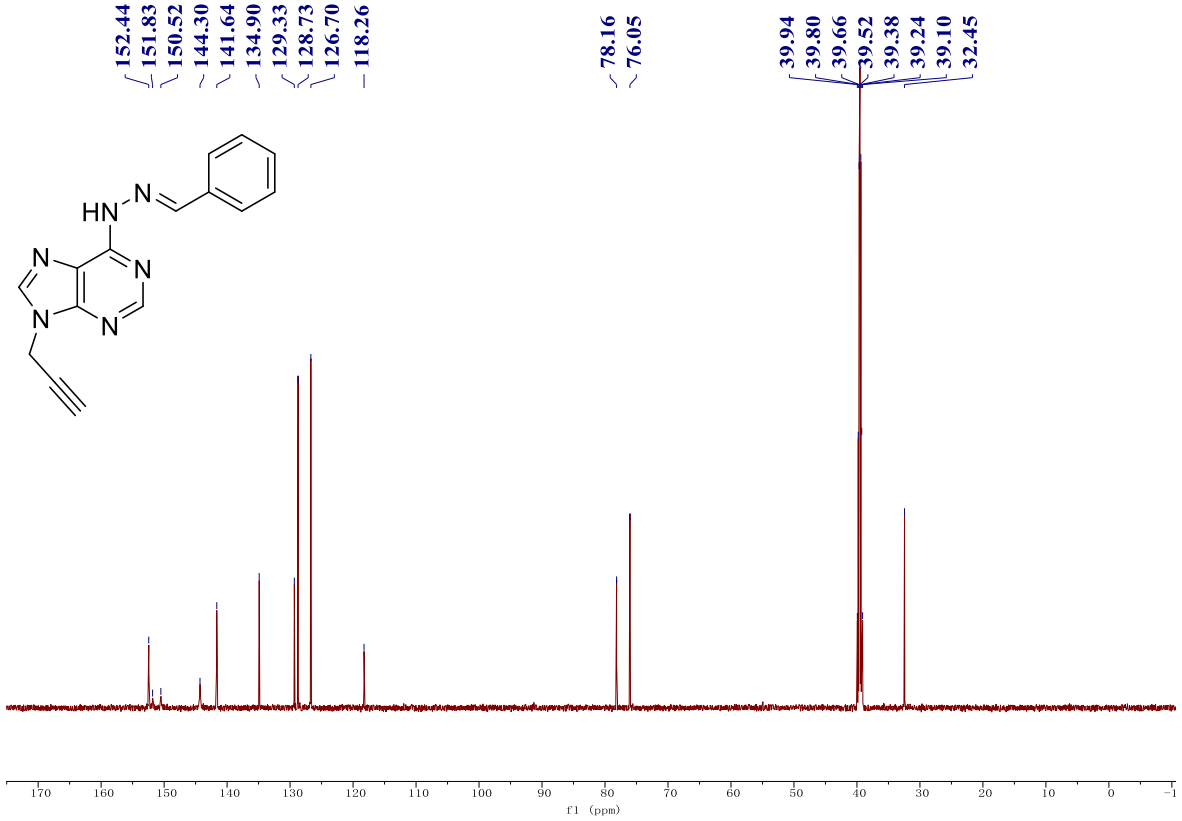
^{13}C NMR Spectrum of 1ar at 25 °C ($CDCl_3$, 150 MHz)



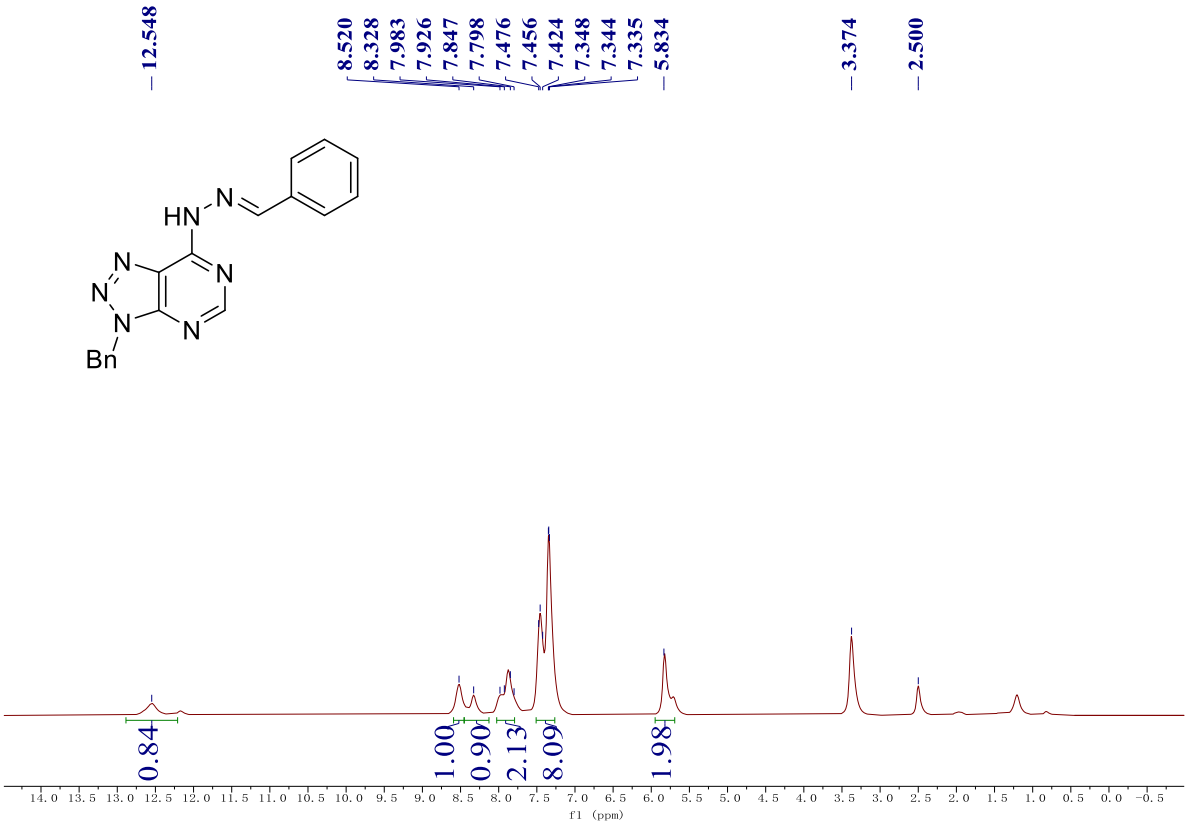
^{1}H NMR Spectrum of 1as at 25 °C (DMSO-d_6, 600 MHz)



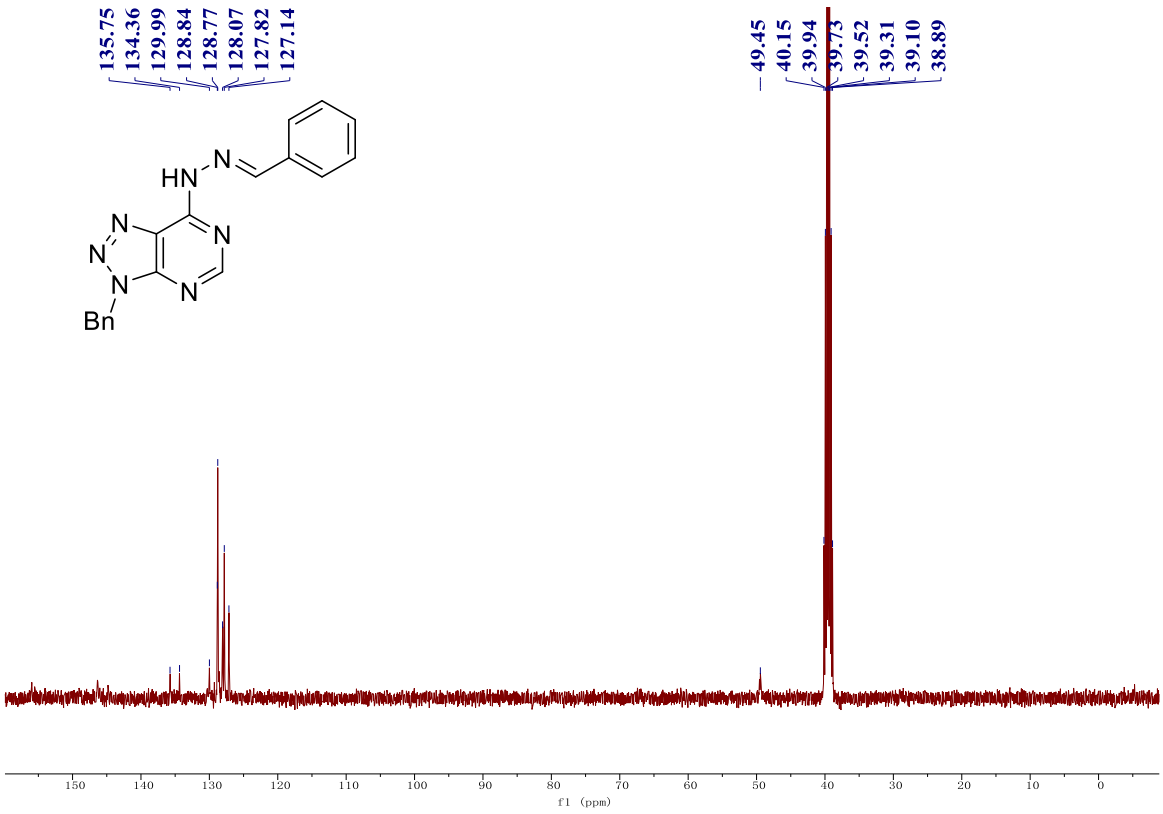
^{13}C NMR Spectrum of 1as at 25 °C (DMSO-d_6, 150 MHz)



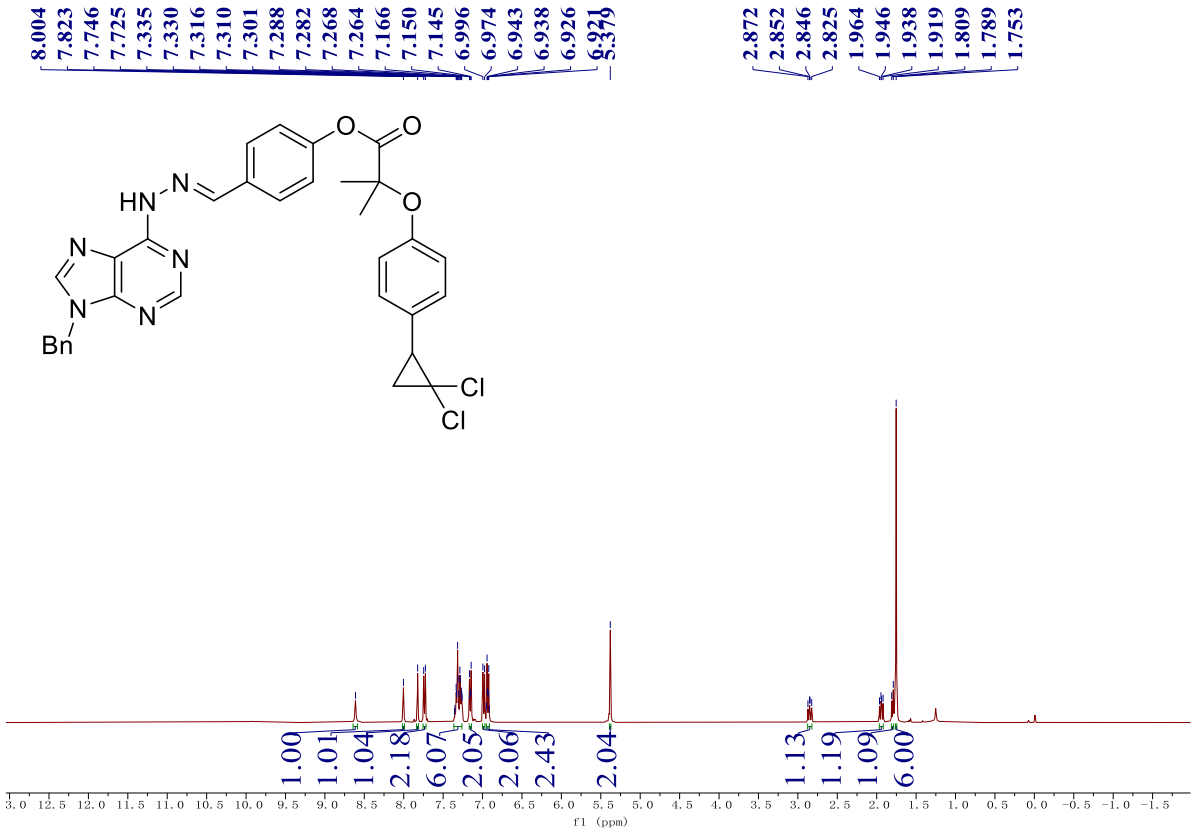
^{1}H NMR Spectrum of 1at at 25 °C (DMSO-d_6, 400 MHz)



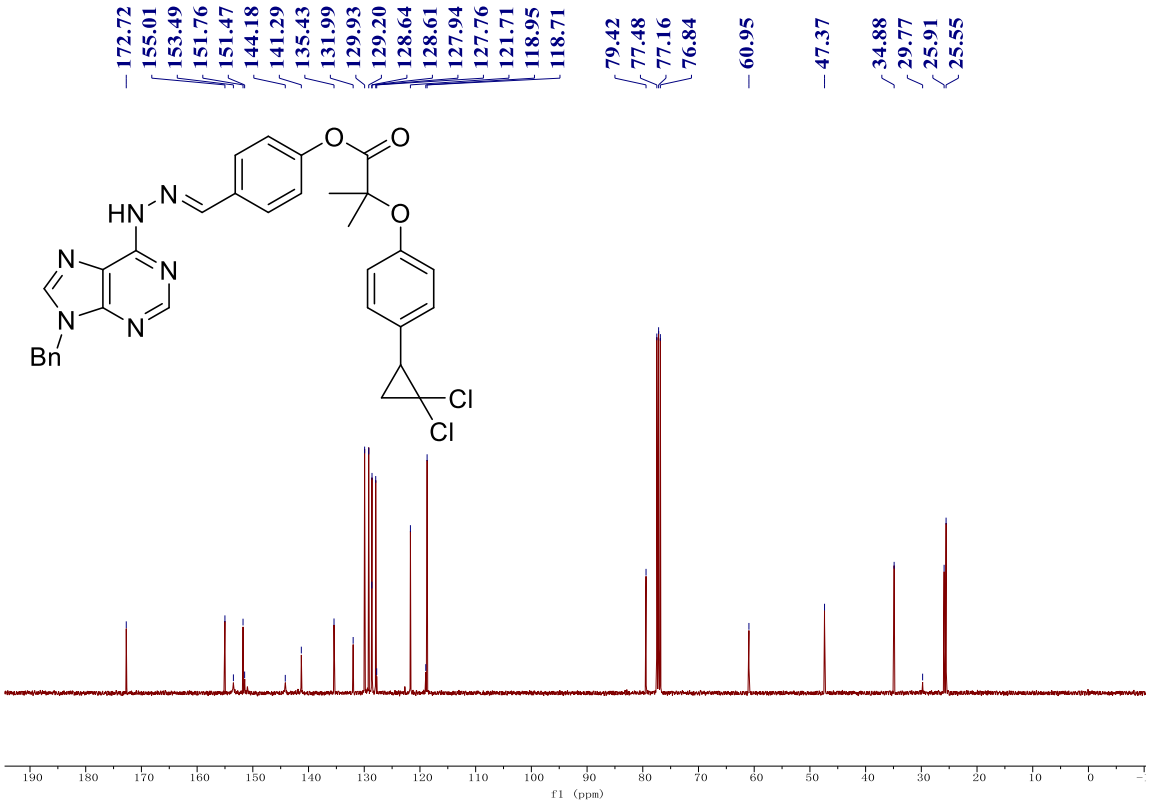
^{13}C NMR Spectrum of 1at at 25 °C (DMSO-d_6, 100 MHz)



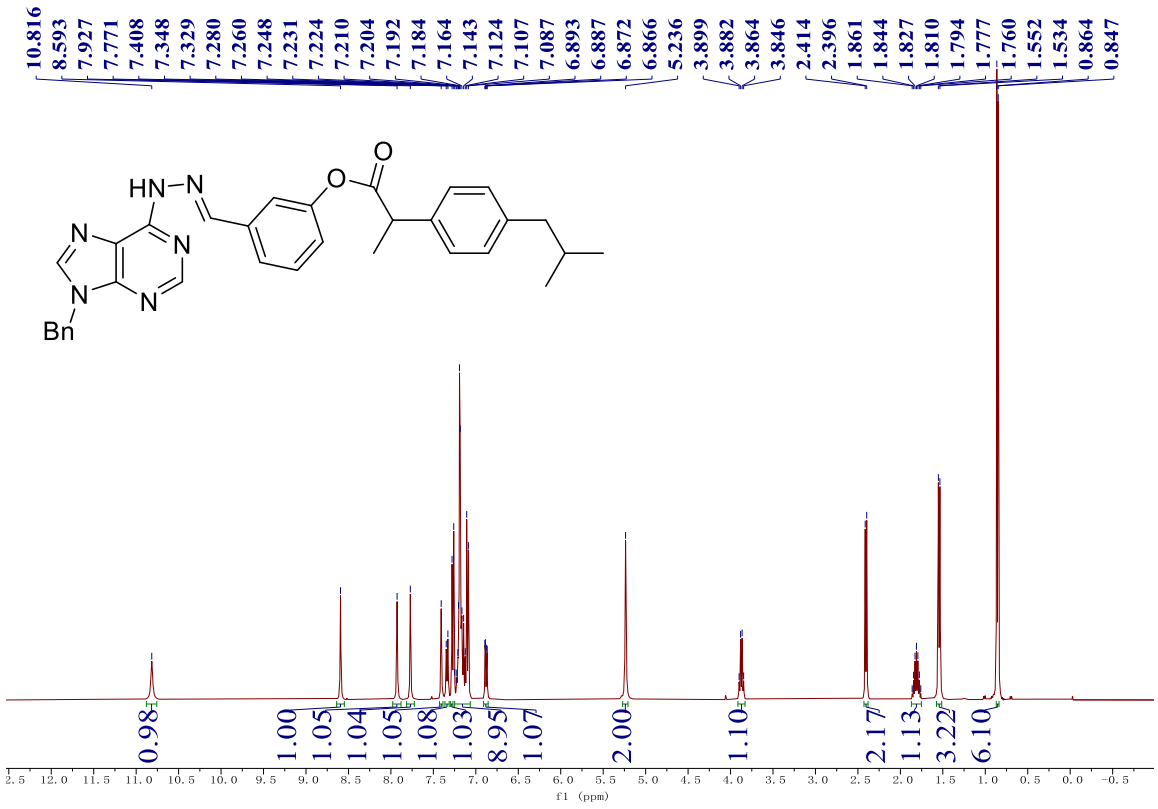
^{1}H NMR Spectrum of 1au at 25 °C ($CDCl_3$, 400 MHz)



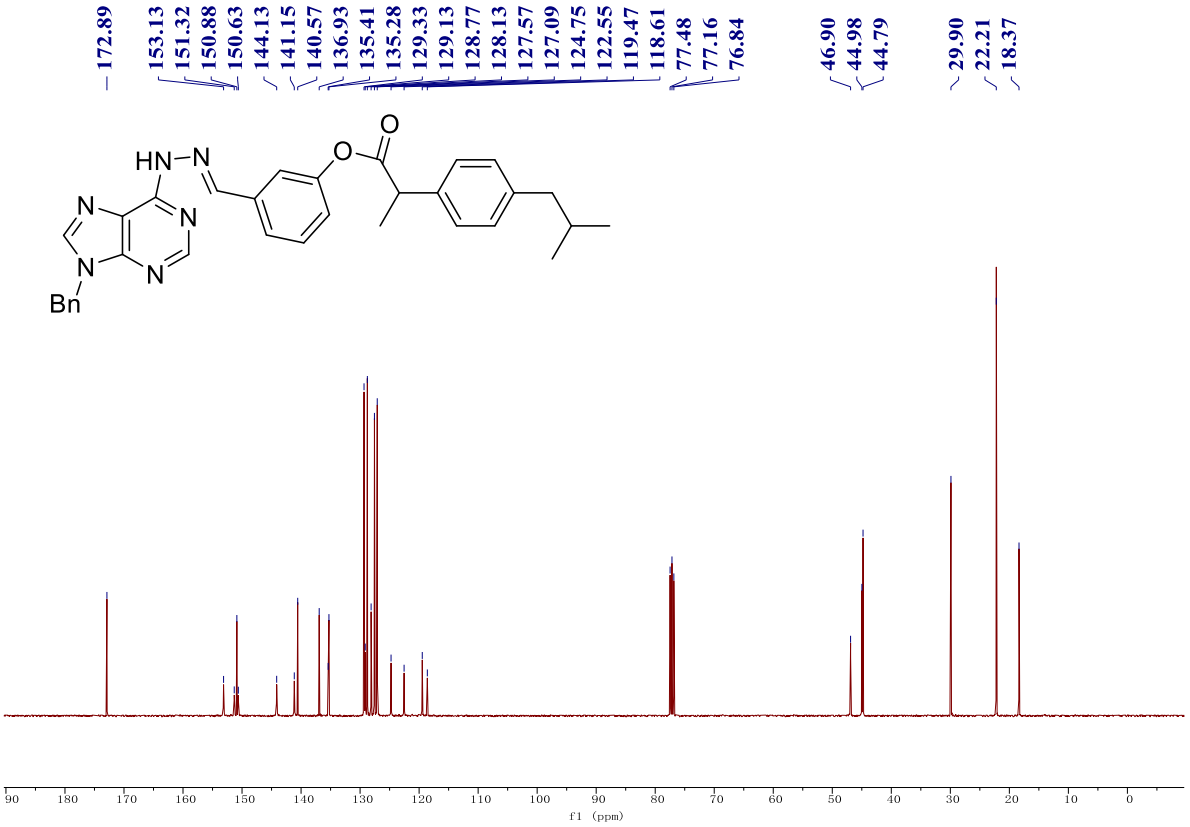
^{13}C NMR Spectrum of 1au at 25 °C ($CDCl_3$, 100 MHz)



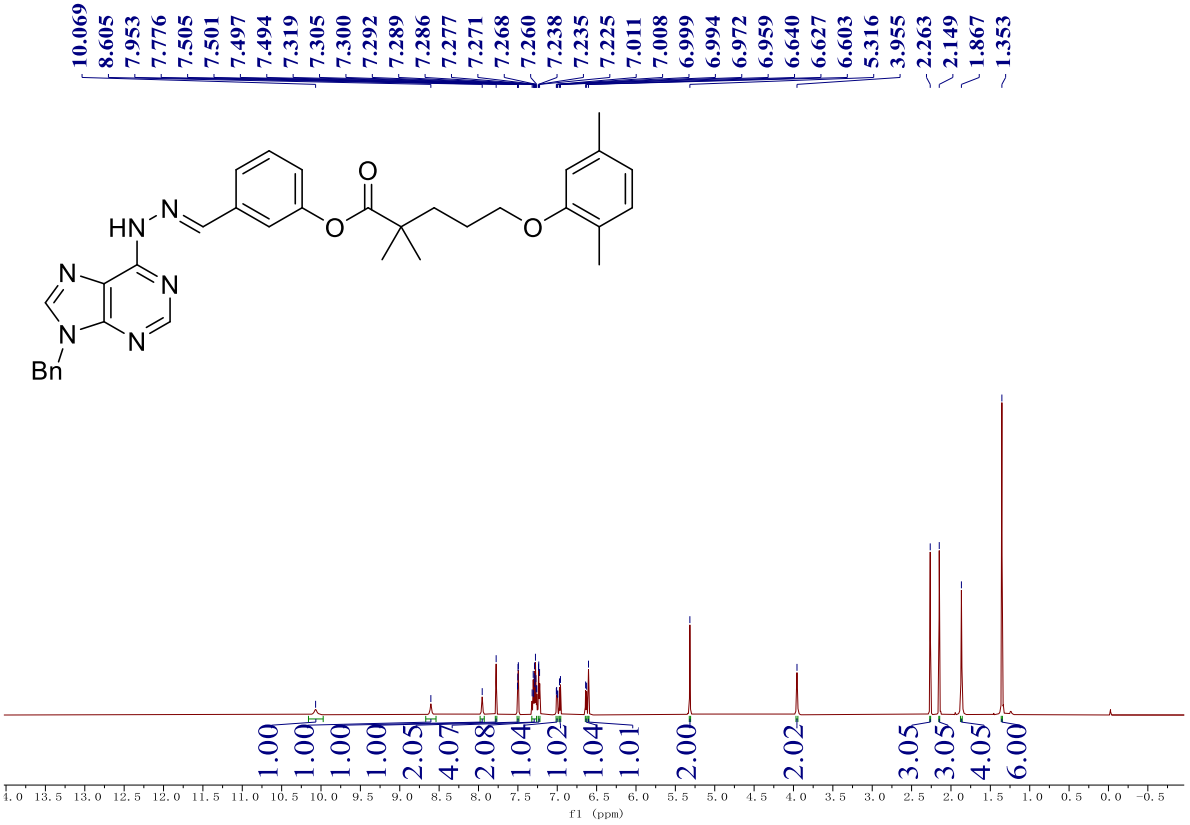
^{1}H NMR Spectrum of 1av at 25 °C ($CDCl_3$, 400 MHz)



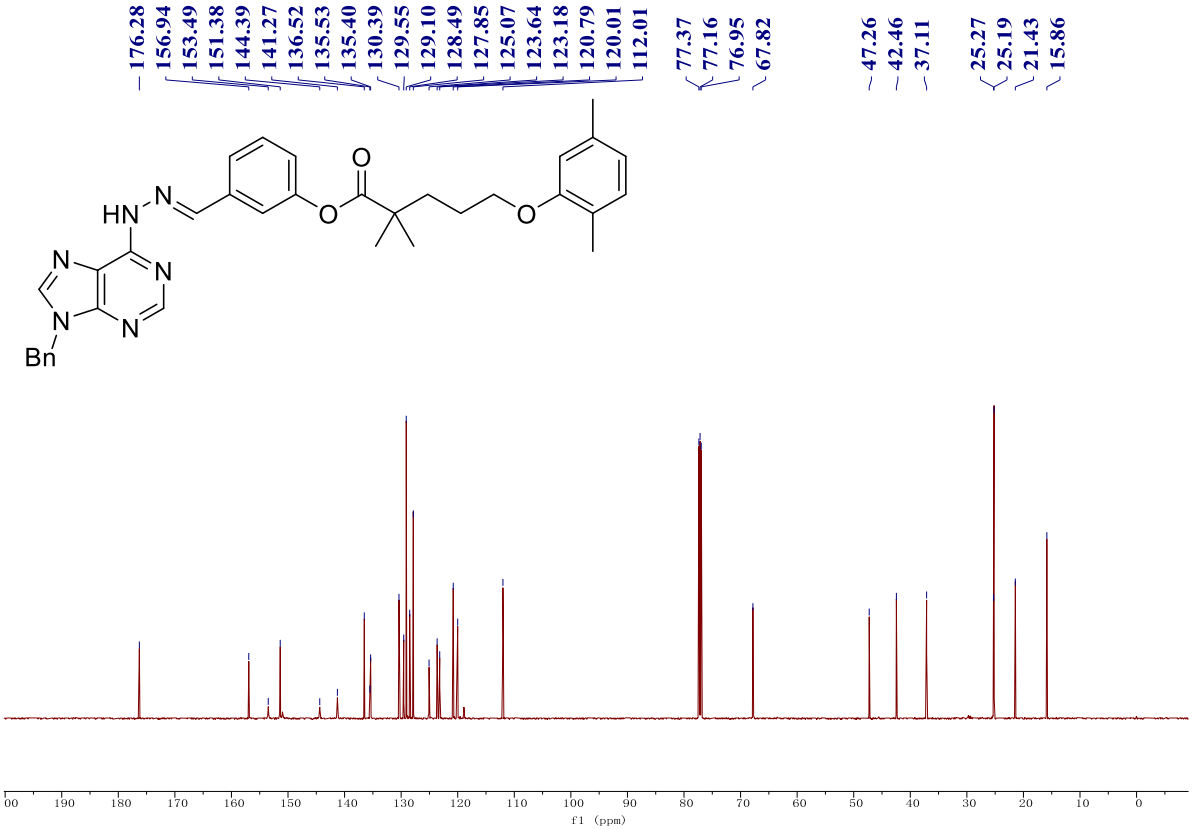
^{13}C NMR Spectrum of 1av at 25 °C ($CDCl_3$, 100 MHz)



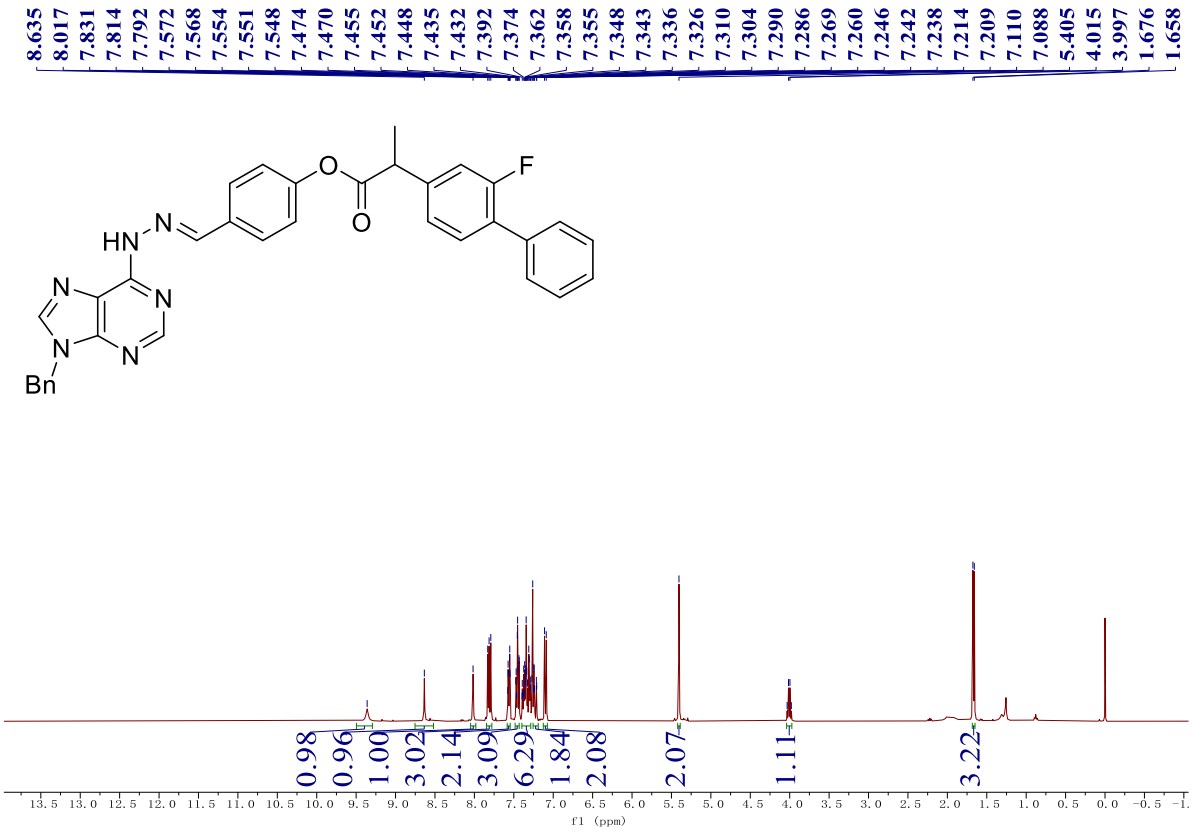
^{1}H NMR Spectrum of 1aw at 25 °C ($CDCl_3$, 600 MHz)



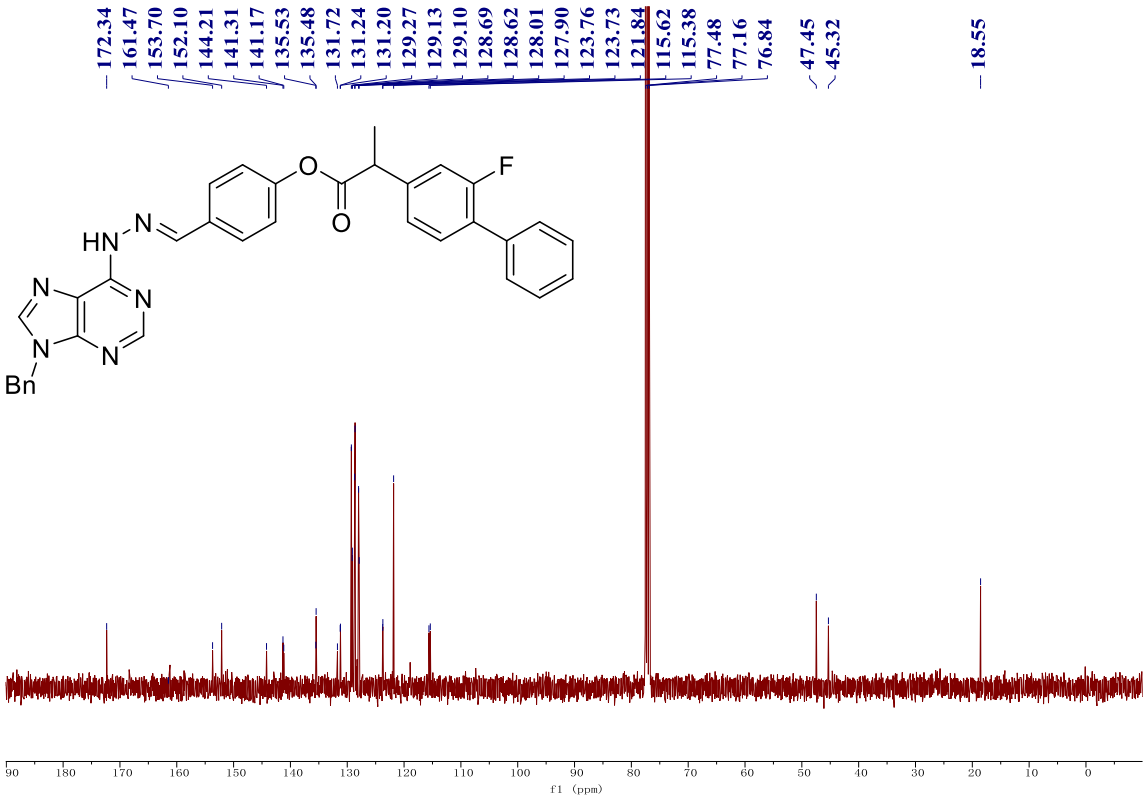
^{13}C NMR Spectrum of 1aw at 25 °C ($CDCl_3$, 150 MHz)



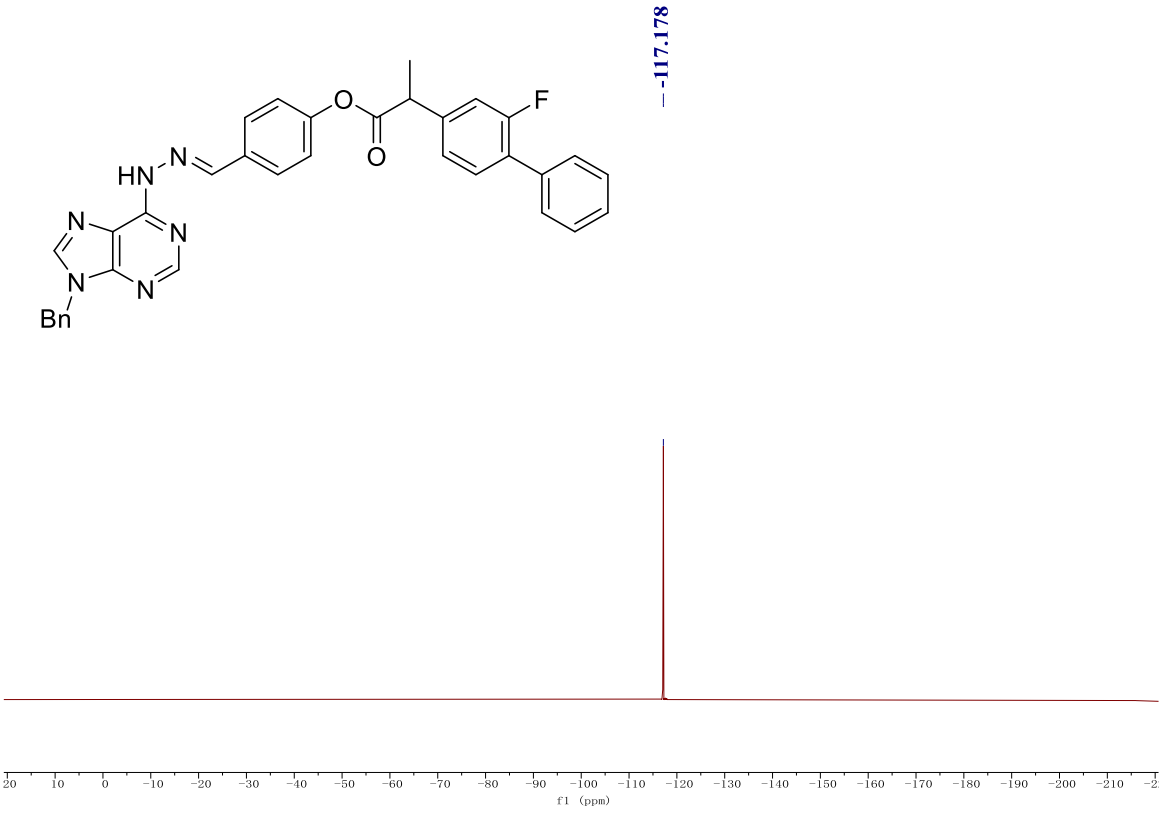
^{1}H NMR Spectrum of 1ax at 25 °C ($CDCl_3$, 400 MHz)



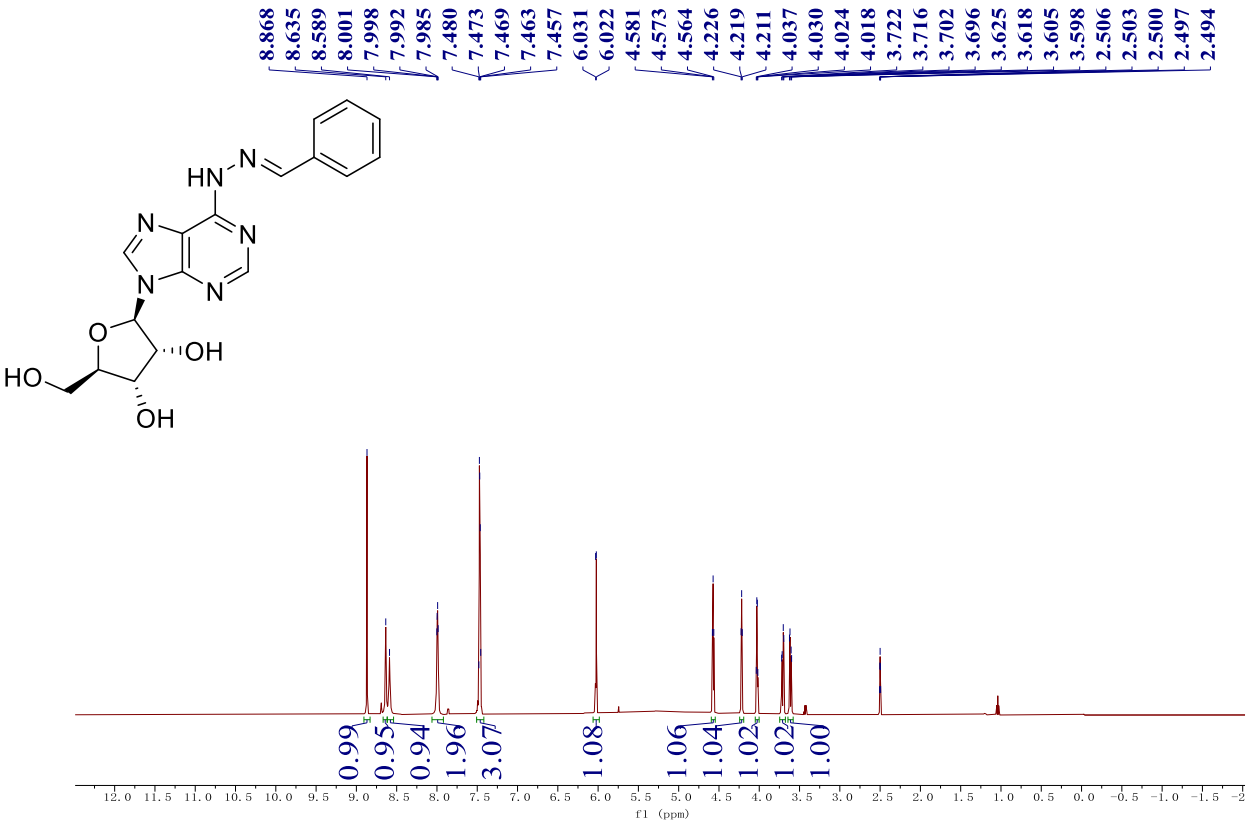
^{13}C NMR Spectrum of 1ax at 25 °C ($CDCl_3$, 100 MHz)



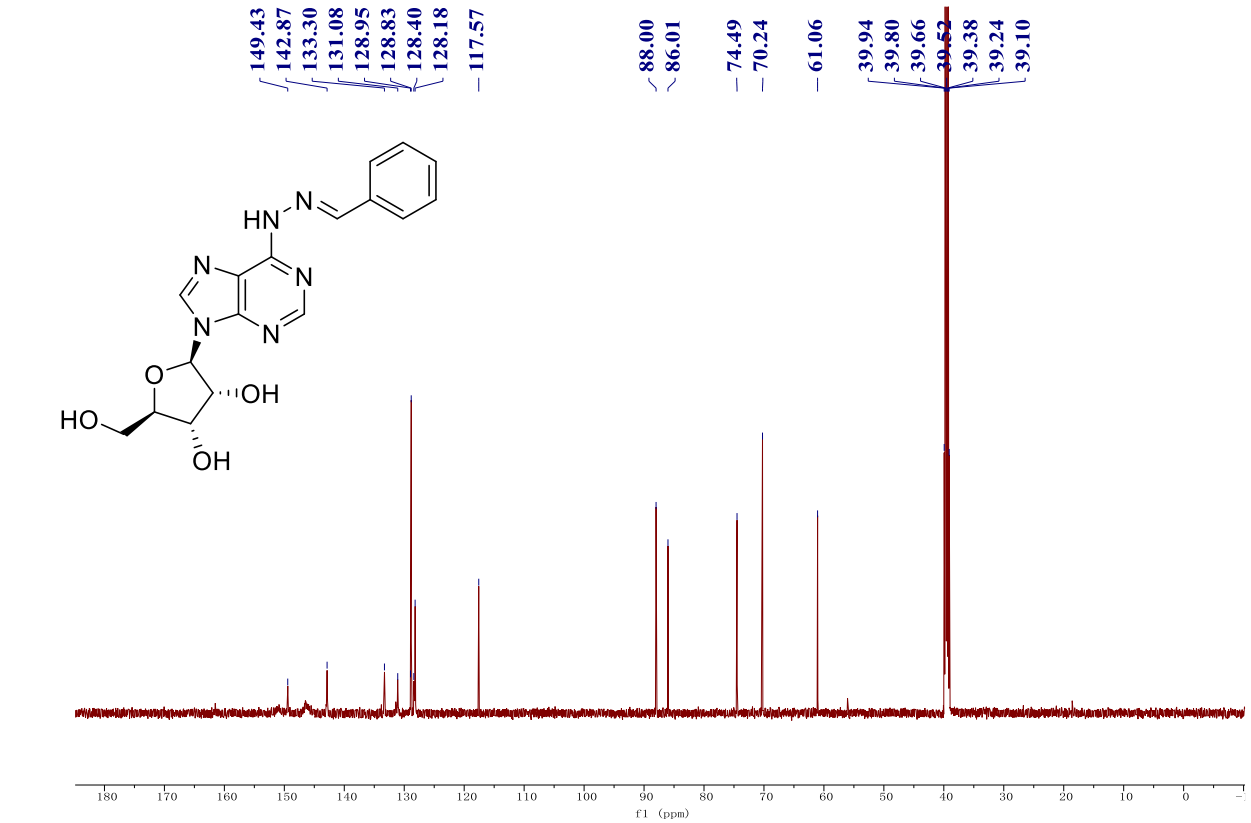
^{19}F NMR Spectrum of 1ax at 25 °C ($CDCl_3$, 400 MHz)



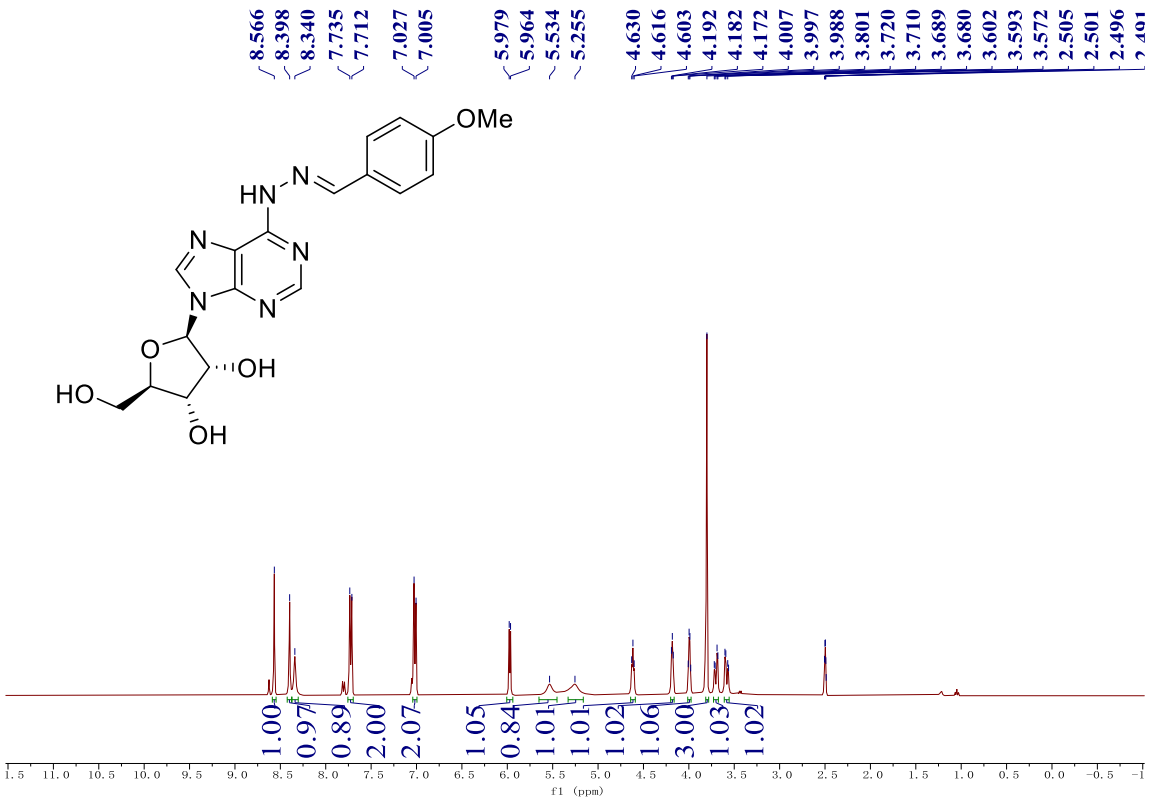
^{1}H NMR Spectrum of 1ay at 25 °C (DMSO-d_6, 600 MHz)



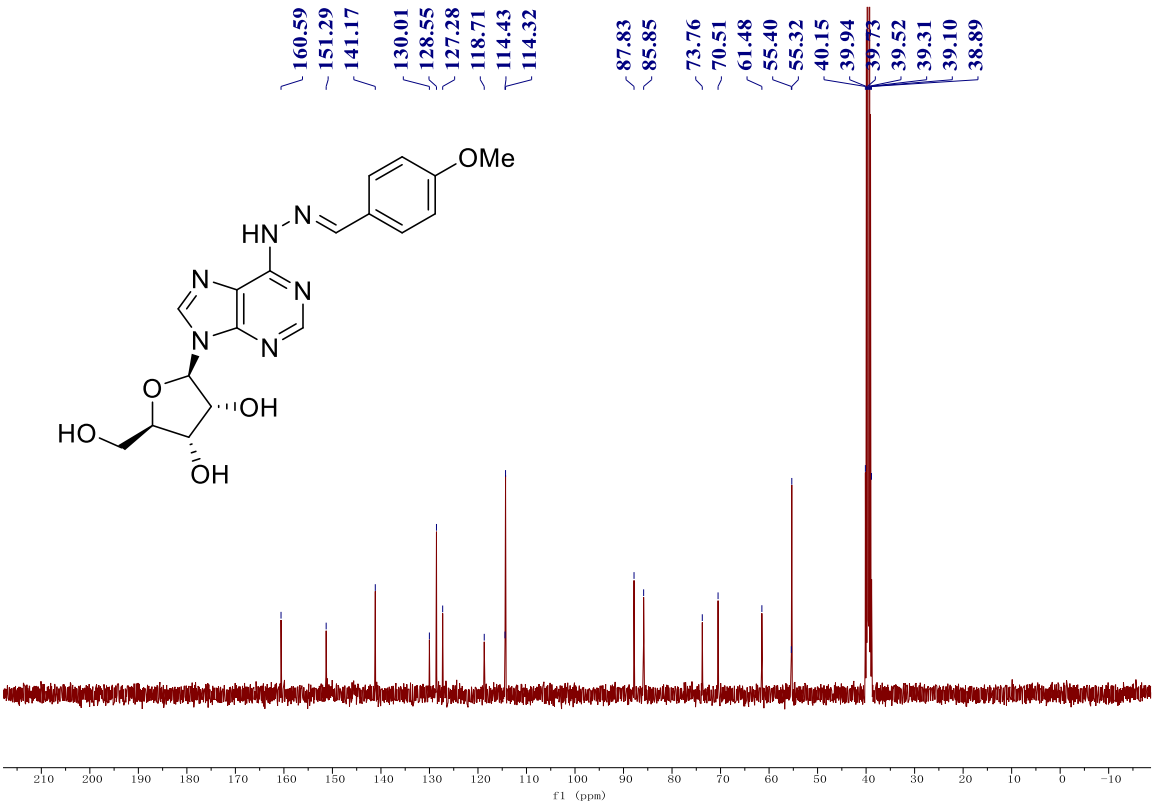
^{13}C NMR Spectrum of 1ay at 25 °C (DMSO-d_6, 150 MHz)



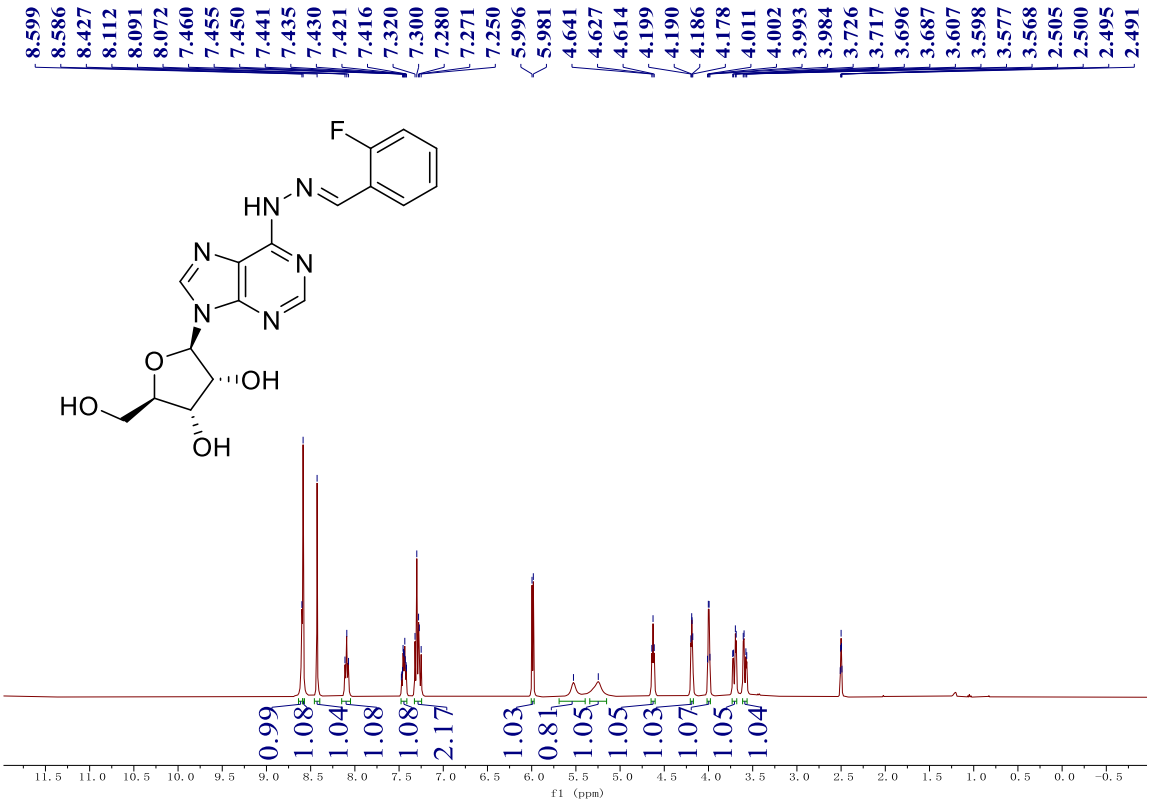
^{1}H NMR Spectrum of 1aaa at 25 °C (DMSO-d_6, 400 MHz)



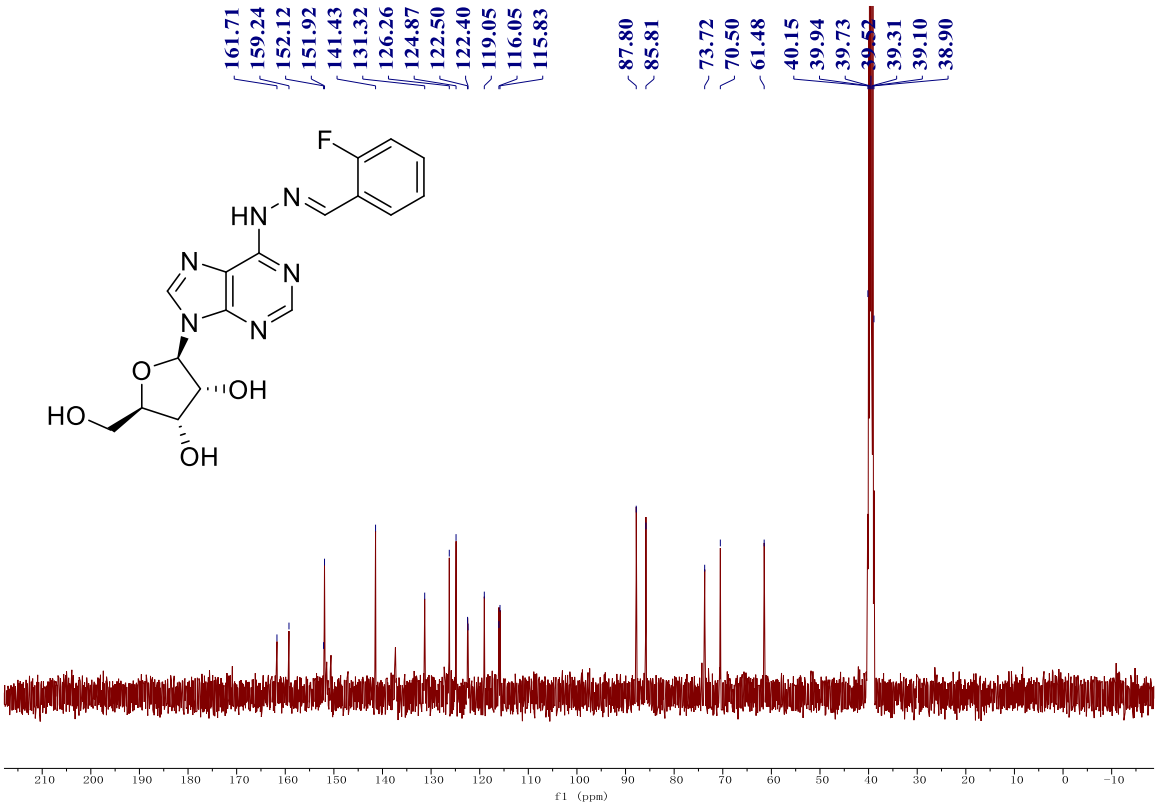
^{13}C NMR Spectrum of 1aaa at 25 °C (DMSO-d_6, 100 MHz)



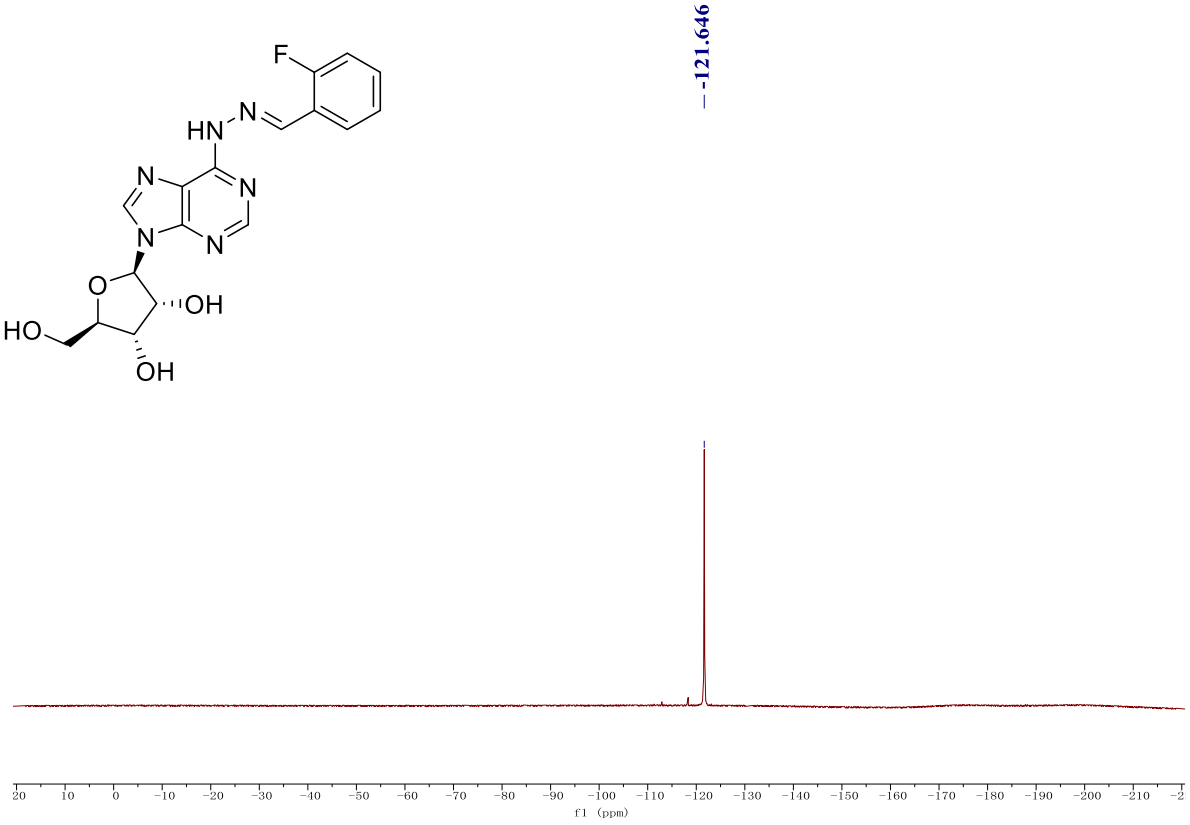
^{1}H NMR Spectrum of 1aab at 25 °C (DMSO-d_6, 400 MHz)



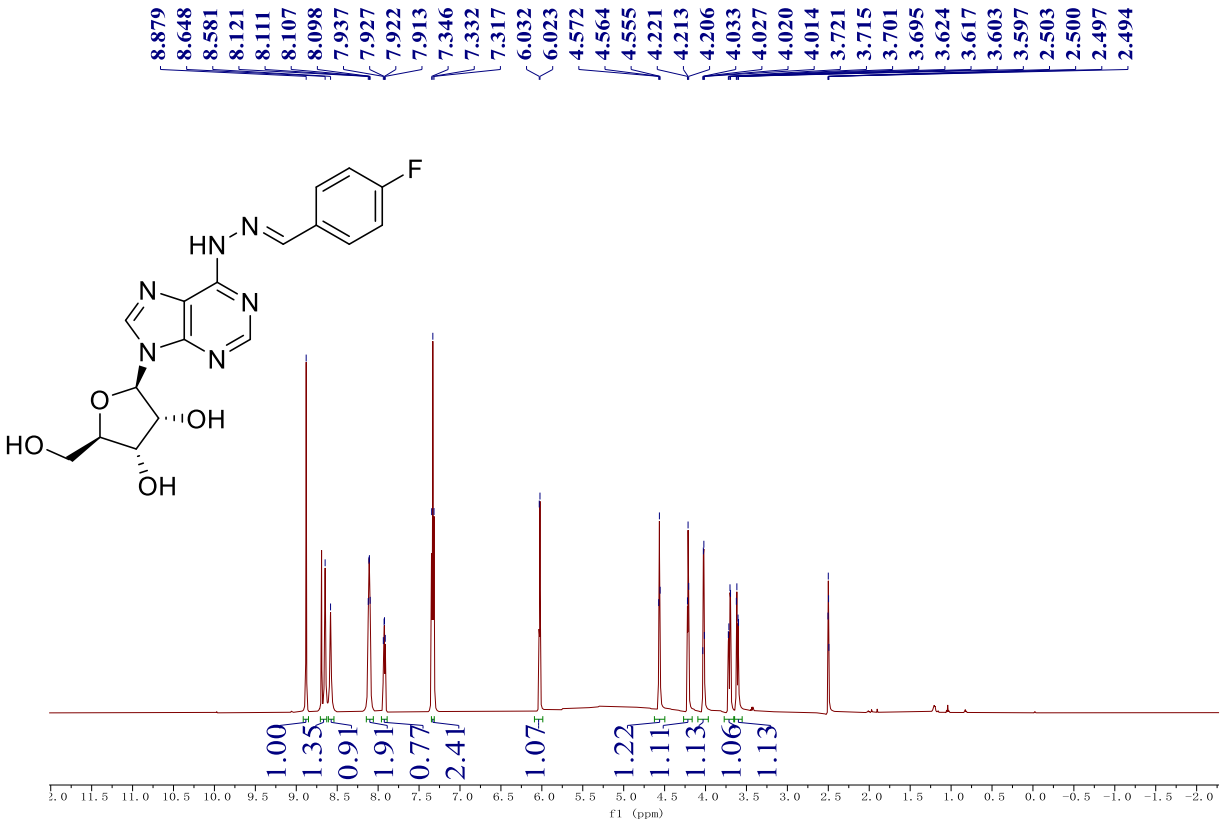
^{13}C NMR Spectrum of 1aab at 25 °C (DMSO-d_6, 100 MHz)



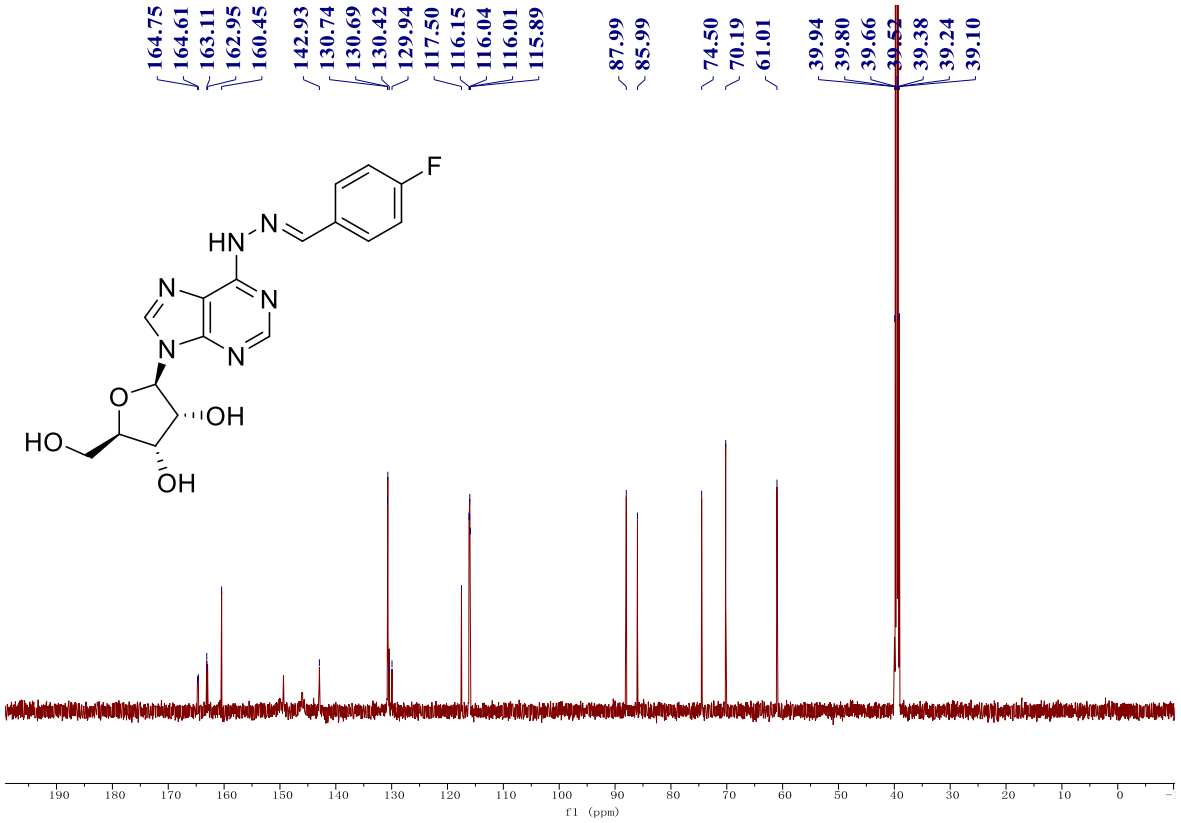
^{19}F NMR Spectrum of 1aab at 25 °C (DMSO-d_6, 376 MHz)



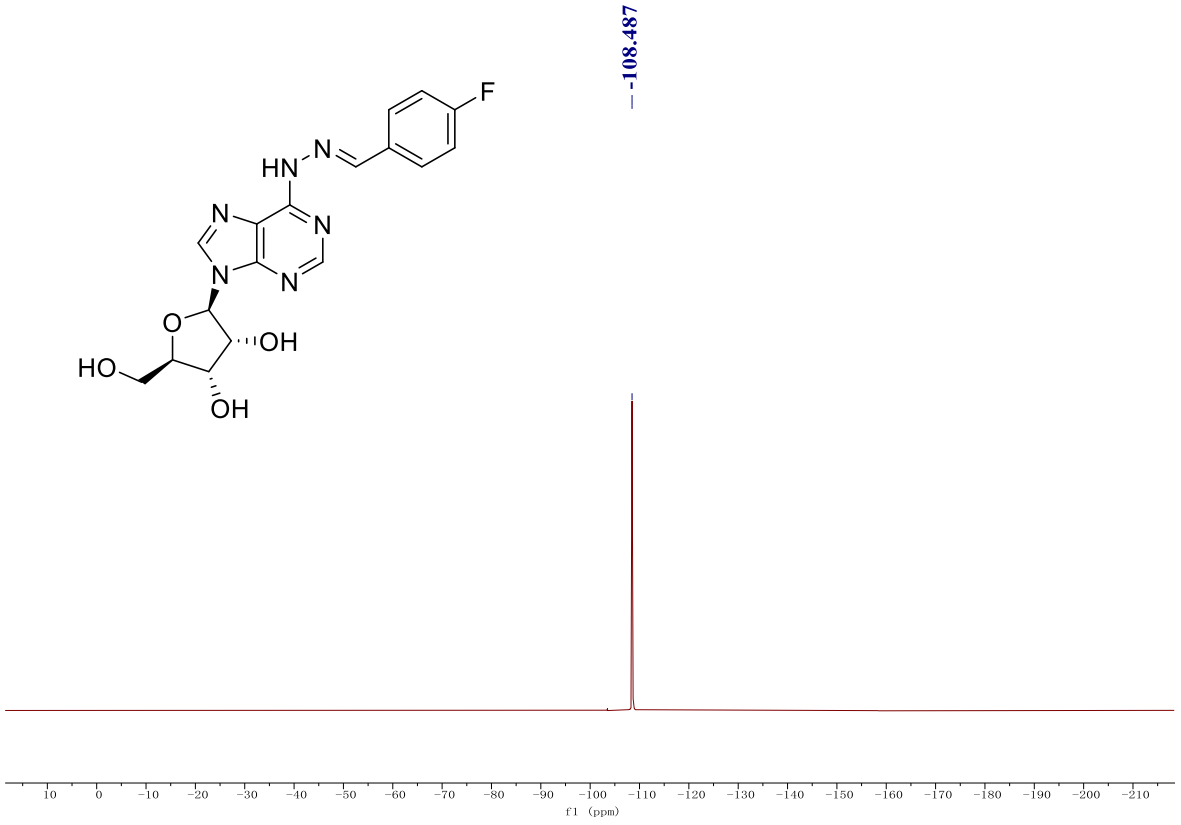
^{1}H NMR Spectrum of 1aac at 25 °C (DMSO-d_6, 600 MHz)



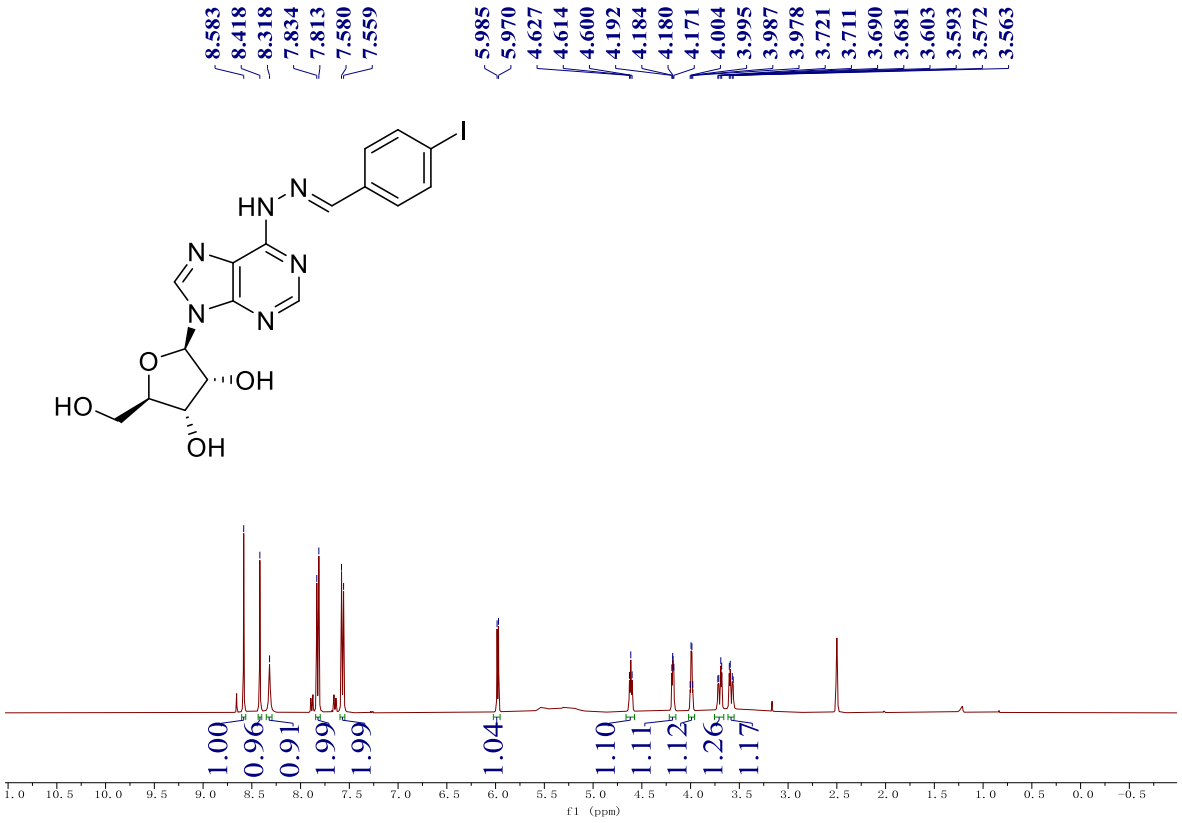
^{13}C NMR Spectrum of 1aac at 25 °C (DMSO-d_6, 100 MHz)



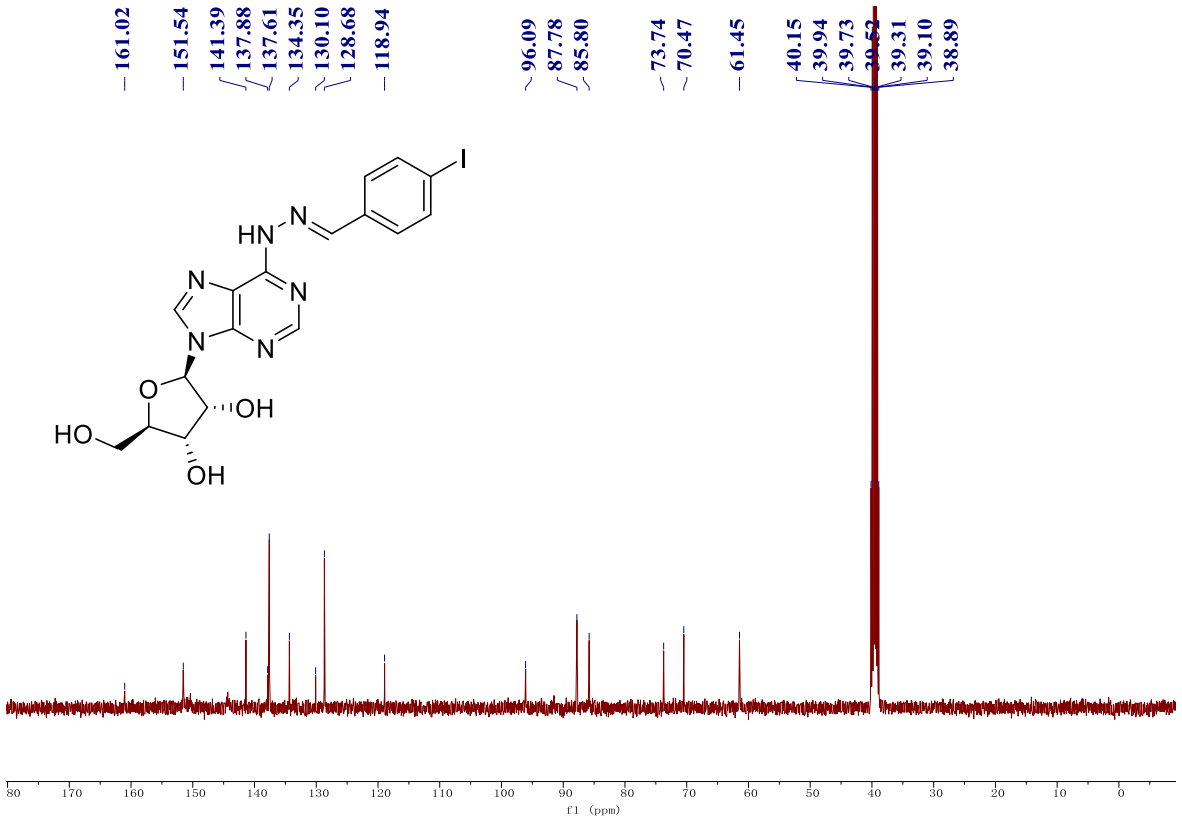
^{19}F NMR Spectrum of 1aac at 25 °C (DMSO-d_6, 565 MHz)



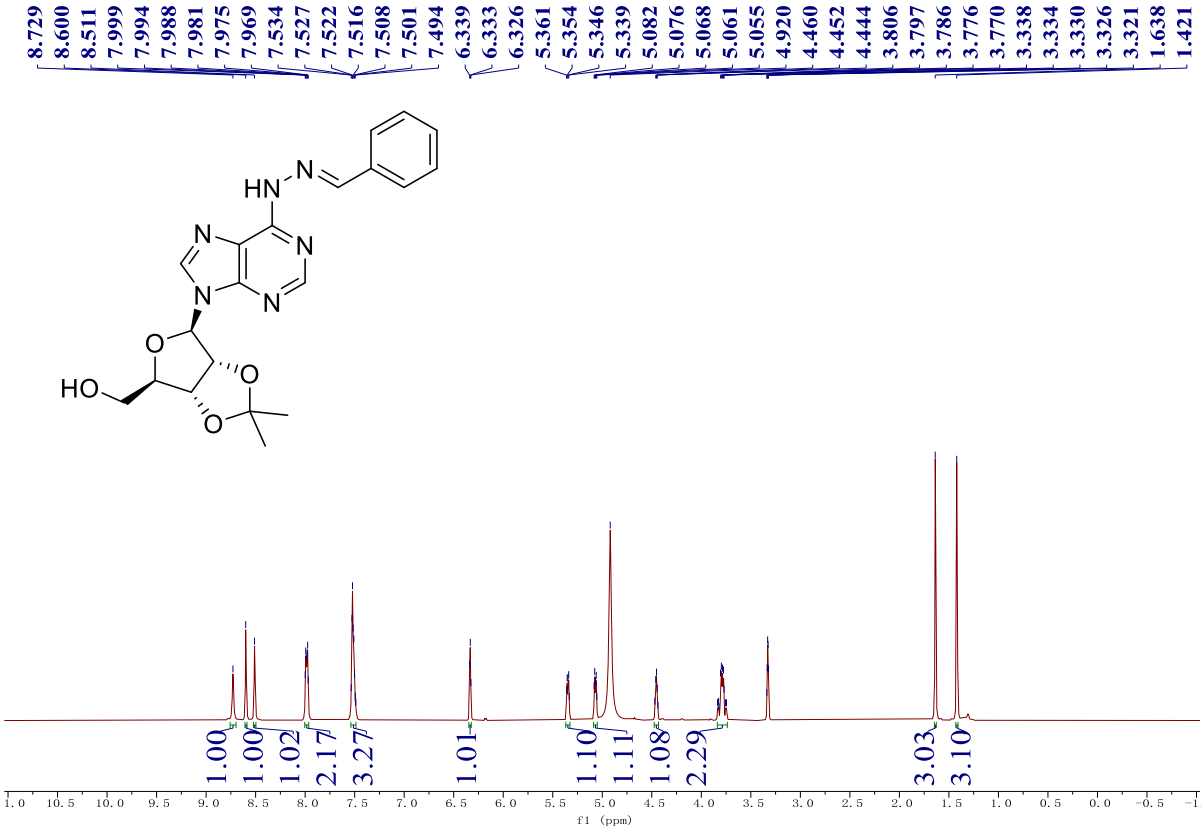
^{1}H NMR Spectrum of 1aad at 25 °C (DMSO-d_6, 400 MHz)



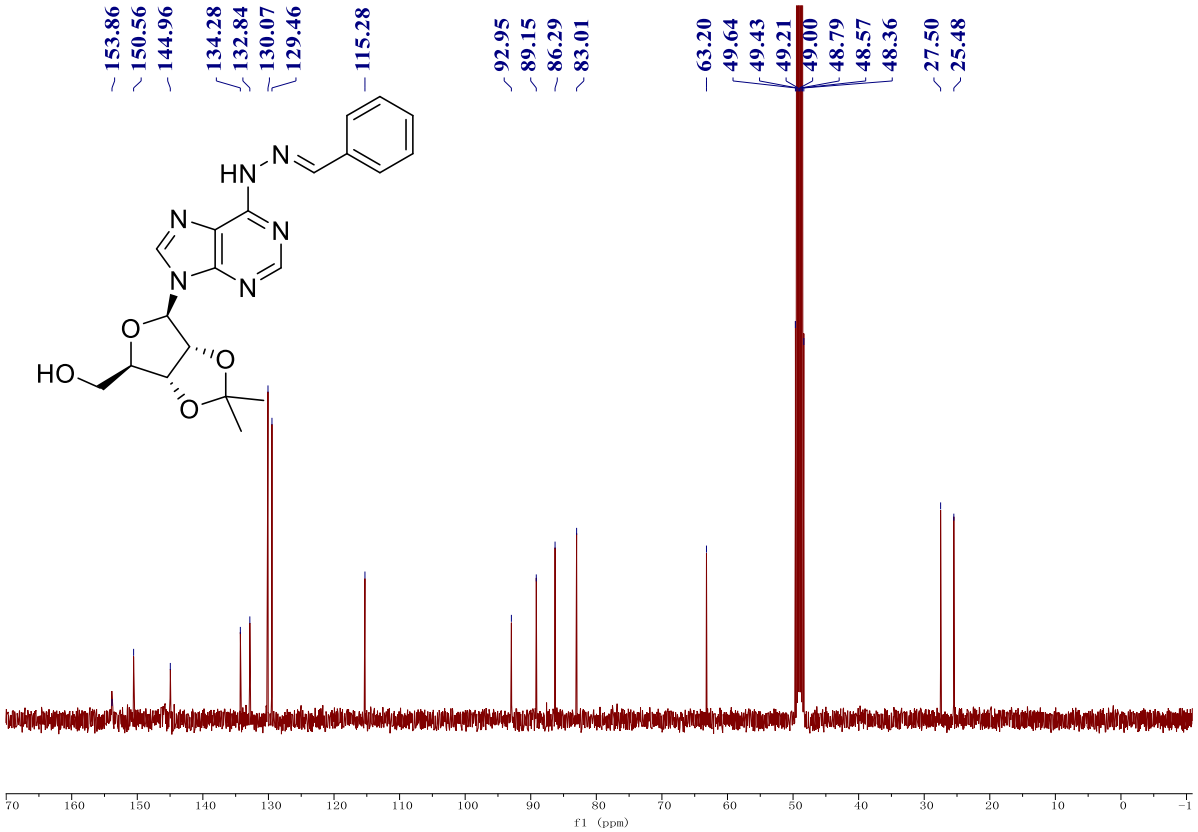
^{13}C NMR Spectrum of 1aad at 25 °C (DMSO-d_6, 100 MHz)



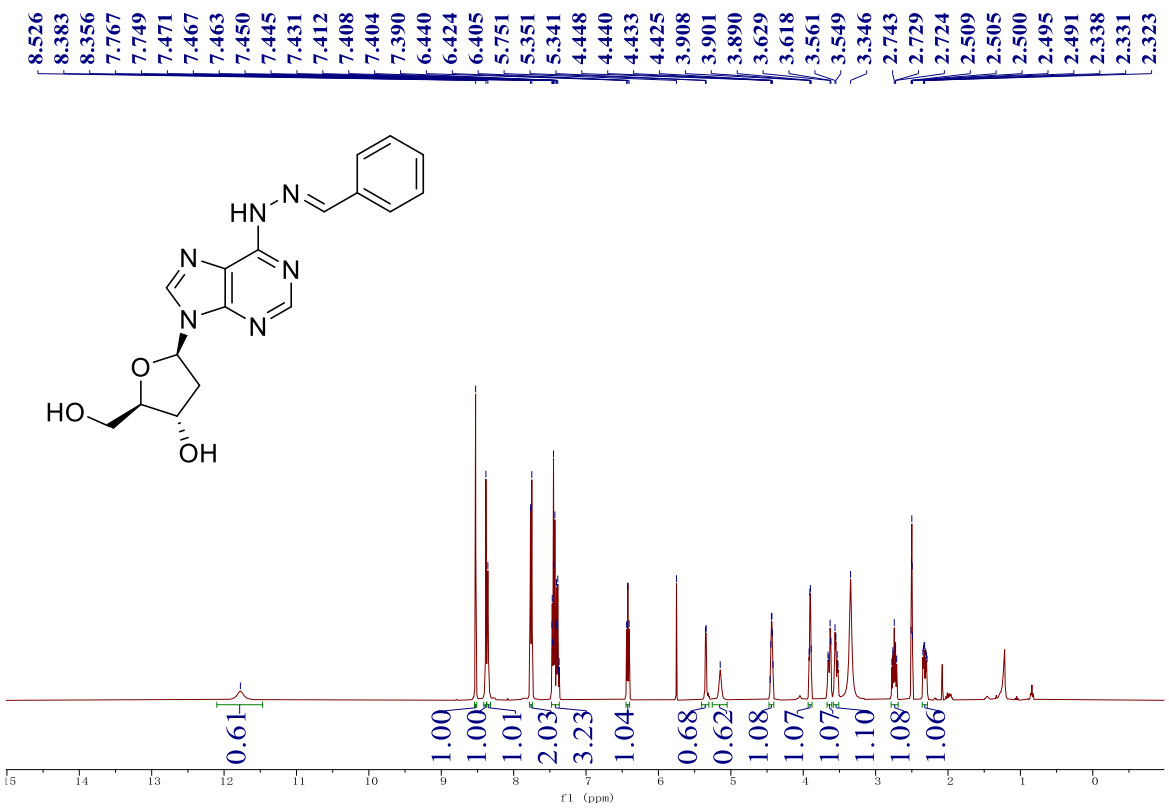
^{1}H NMR Spectrum of 1aaf at 25 °C (CD_3OD, 400 MHz)



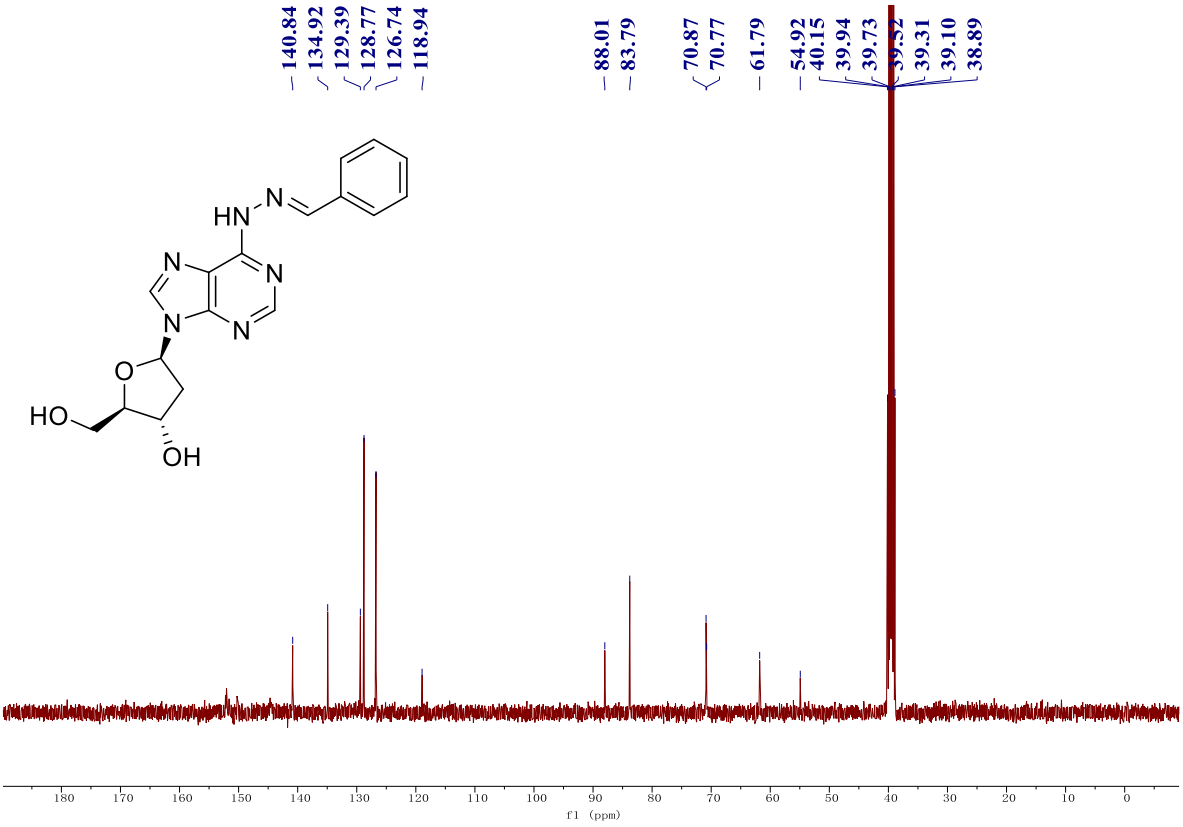
^{13}C NMR Spectrum of 1aaf at 25 °C (CD_3OD, 100 MHz)



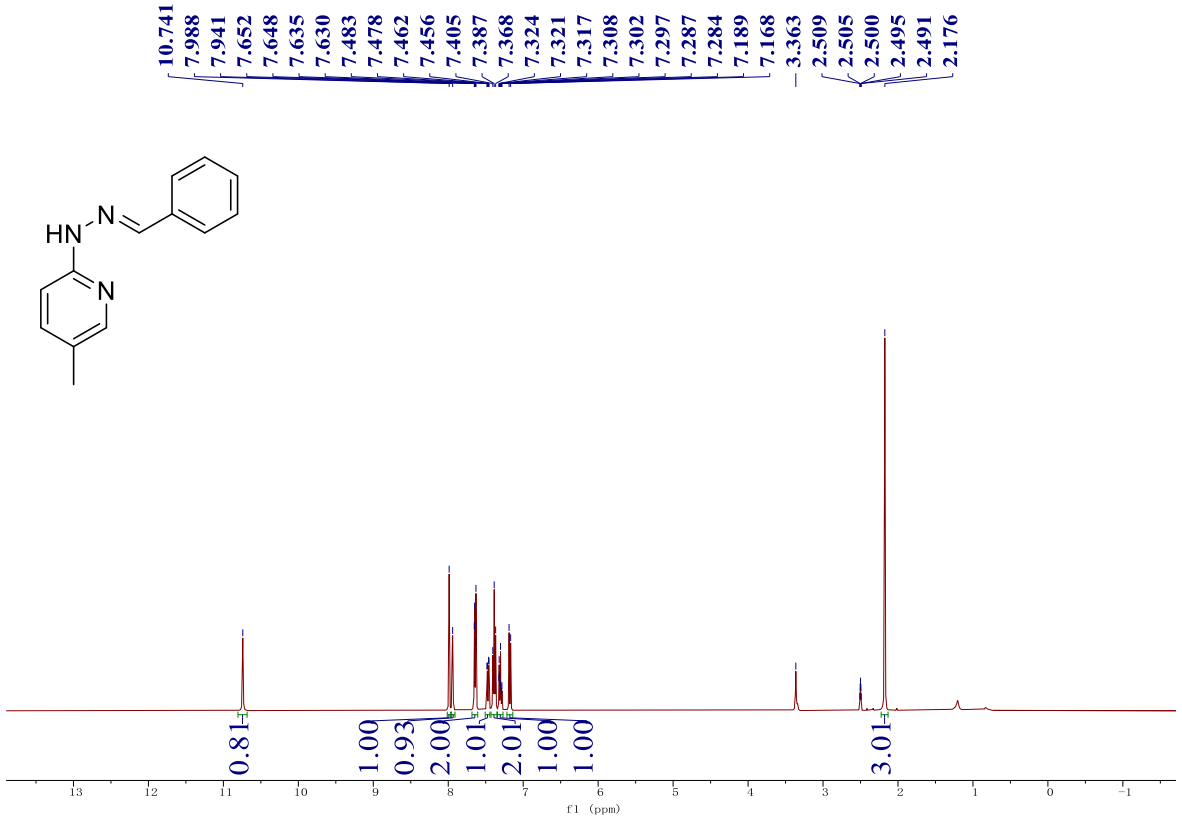
^{1}H NMR Spectrum of 1aag at 25 °C (DMSO-d_6, 400 MHz)



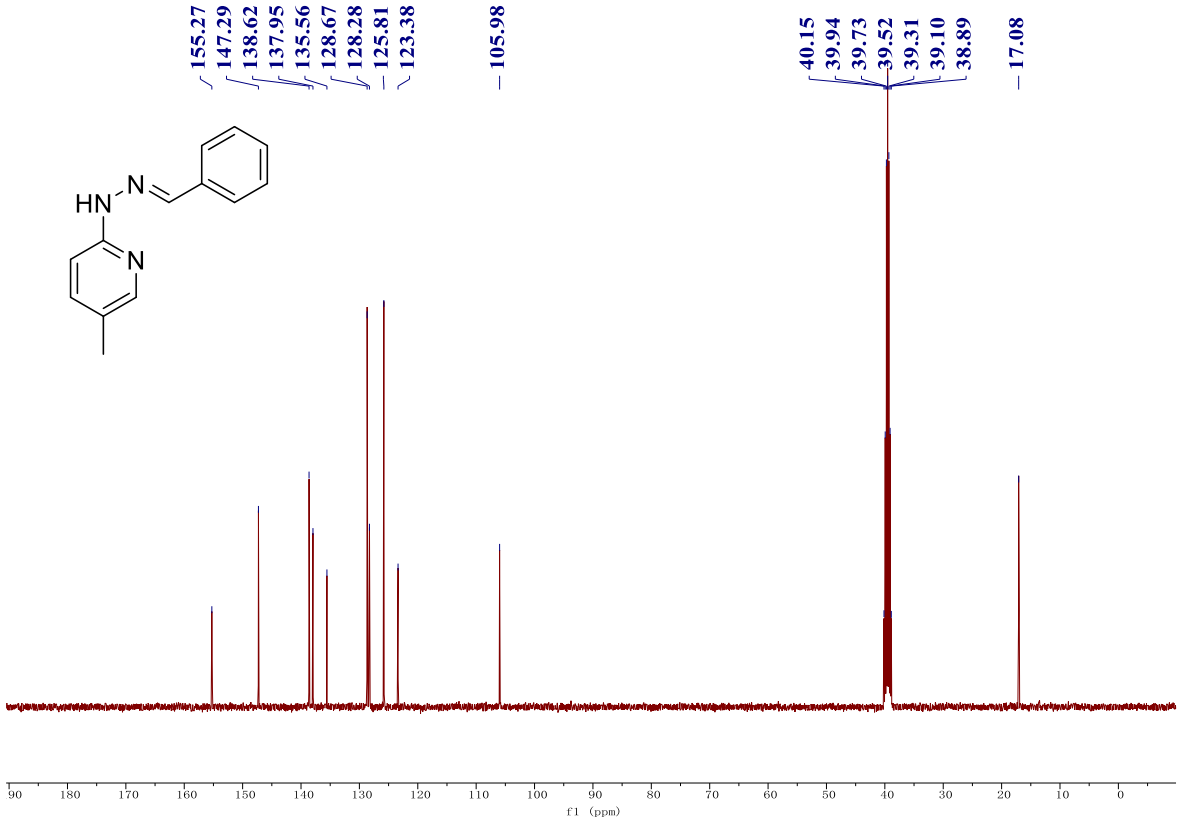
^{13}C NMR Spectrum of 1aag at 25 °C (DMSO-d_6, 100 MHz)



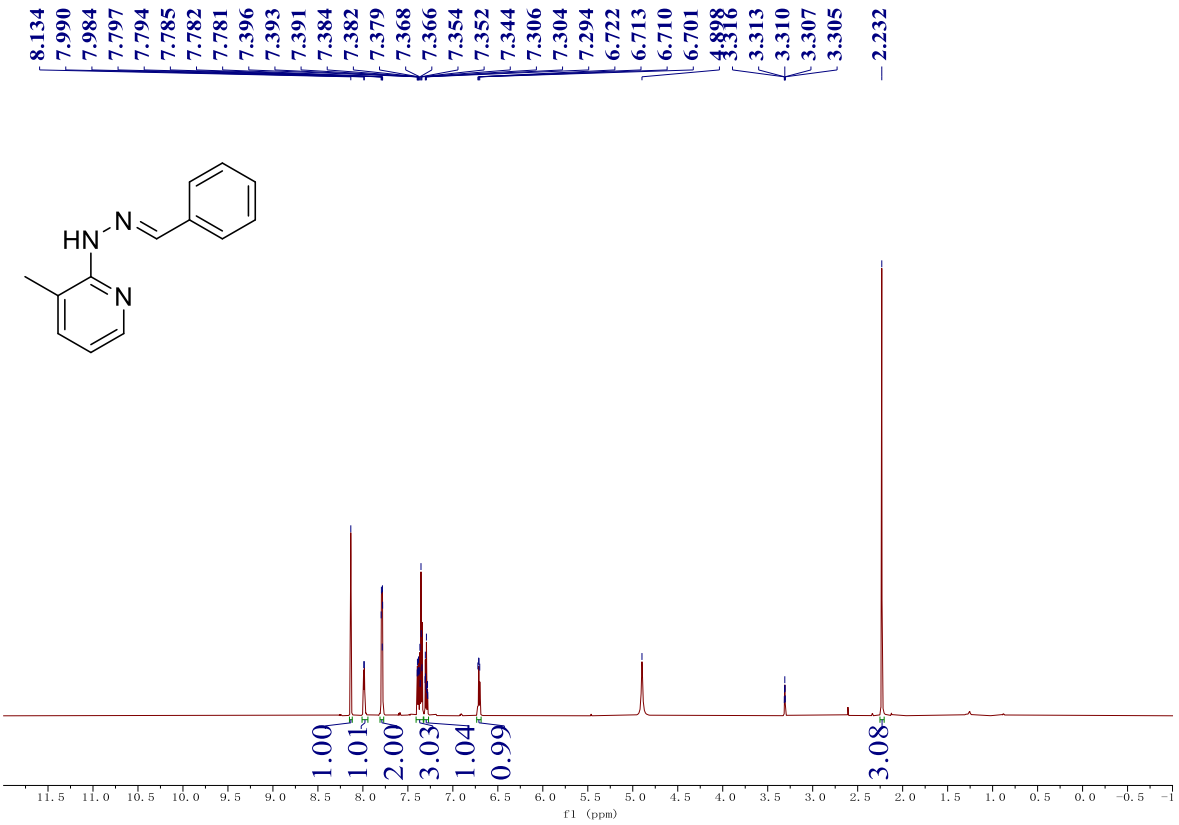
^{1}H NMR Spectrum of 1aah at 25 ^{o}C ($CDCl_3$, 400 MHz)



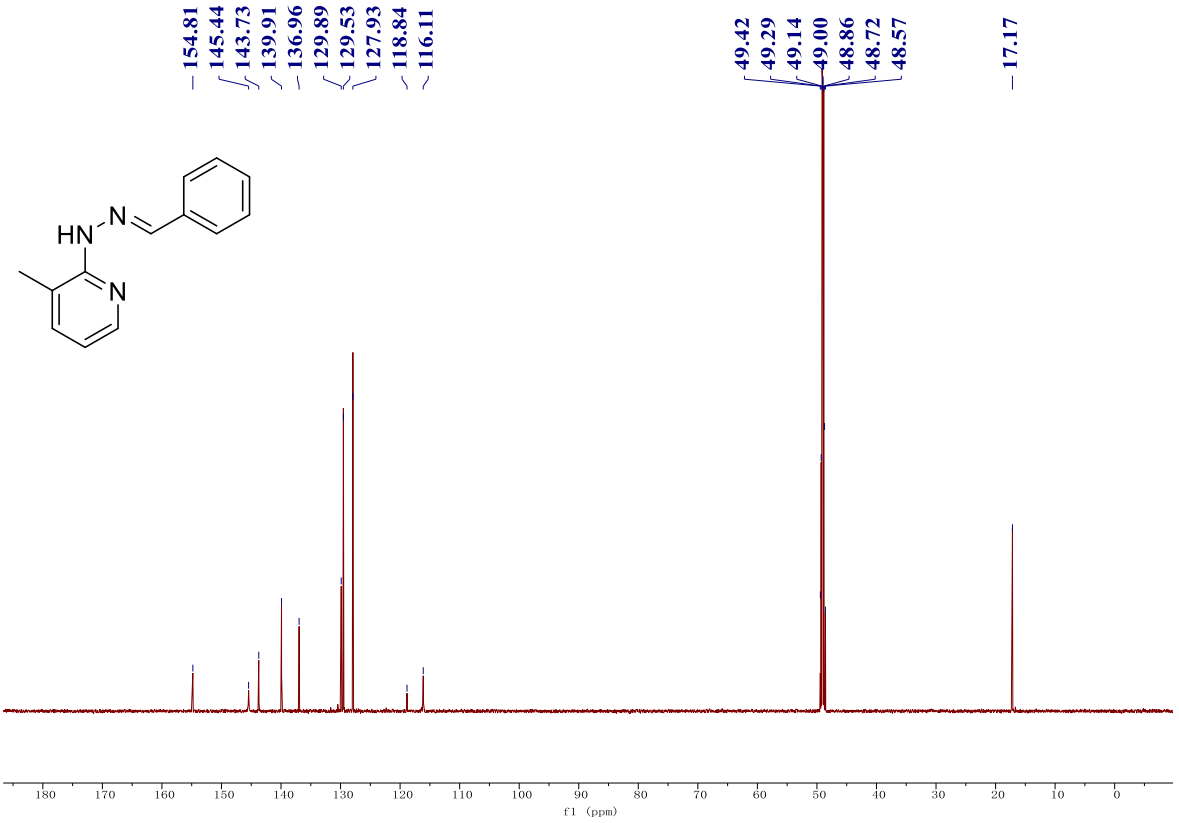
^{13}C NMR Spectrum of 1aah at 25 ^{o}C ($CDCl_3$, 100 MHz)



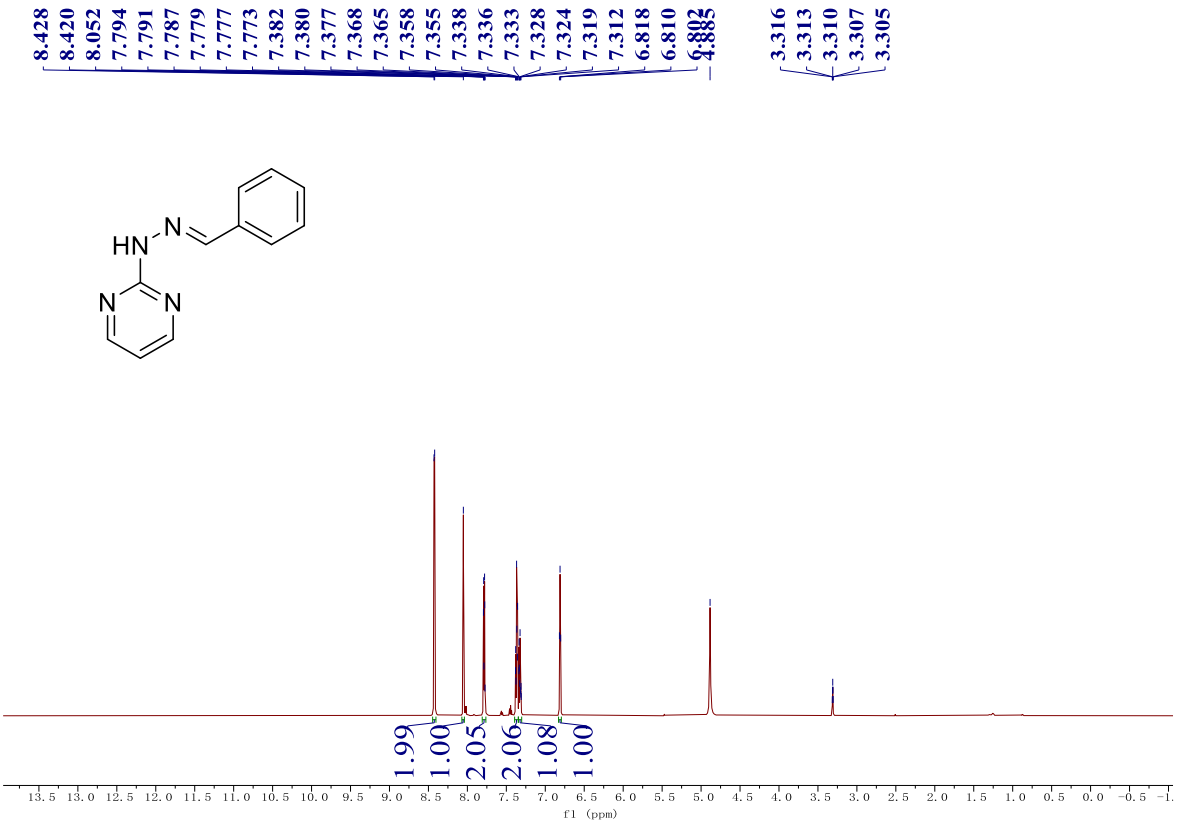
^{1}H NMR Spectrum of 1aai at 25 °C (CD_3OD, 600 MHz)



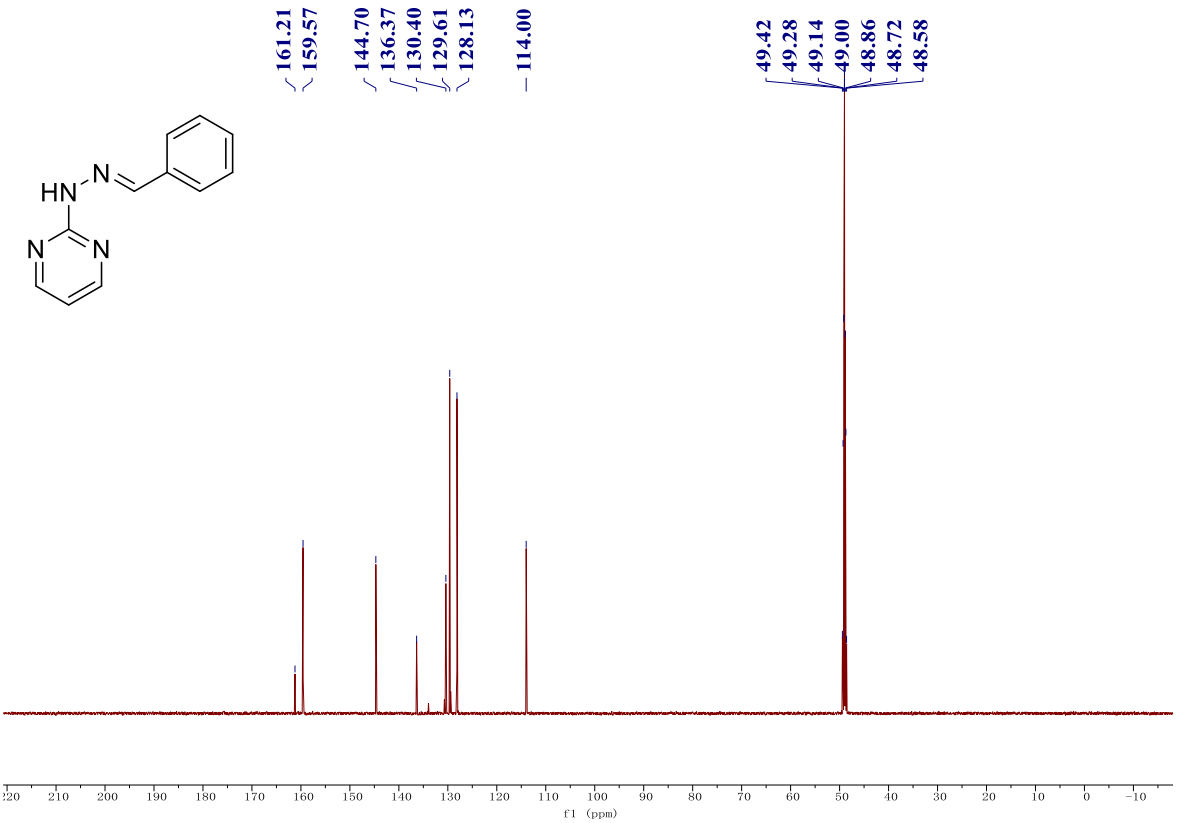
^{13}C NMR Spectrum of 1aai at 25 °C (CD_3OD, 150 MHz)



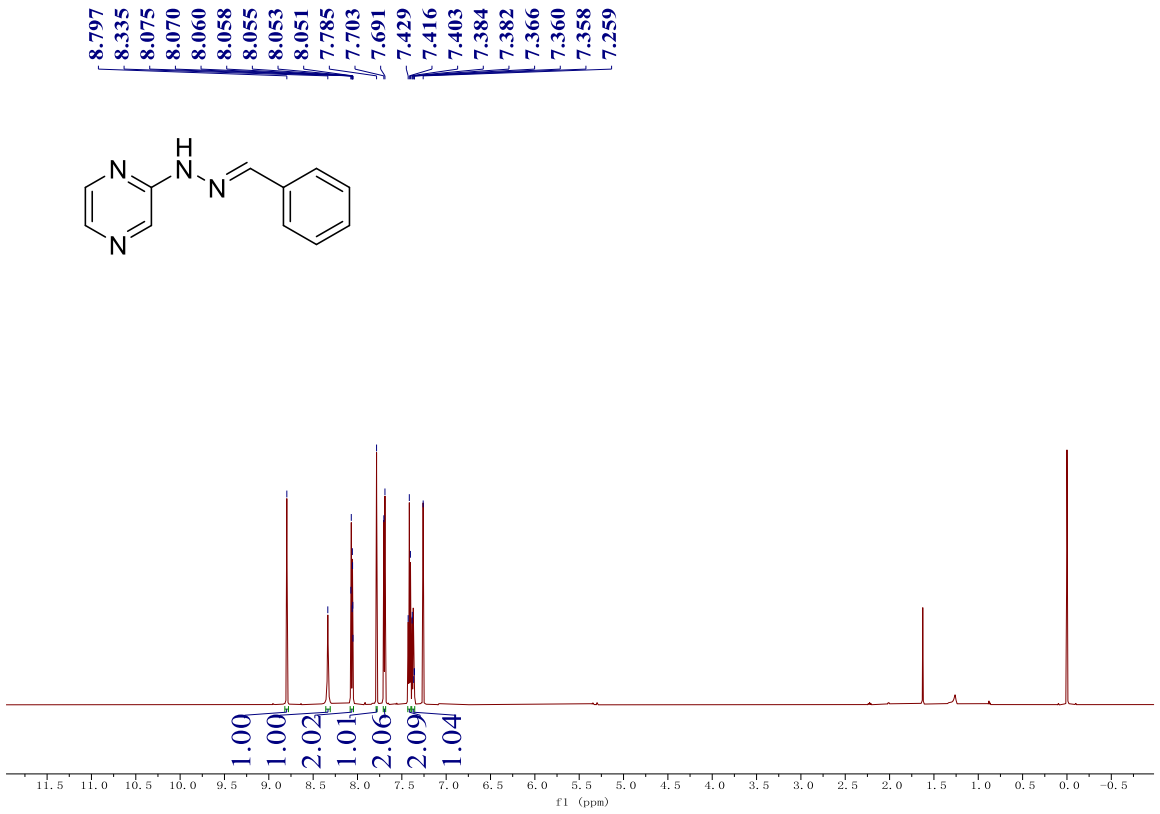
^{1}H NMR Spectrum of 1aaj at 25 °C (CD_3OD, 600 MHz)



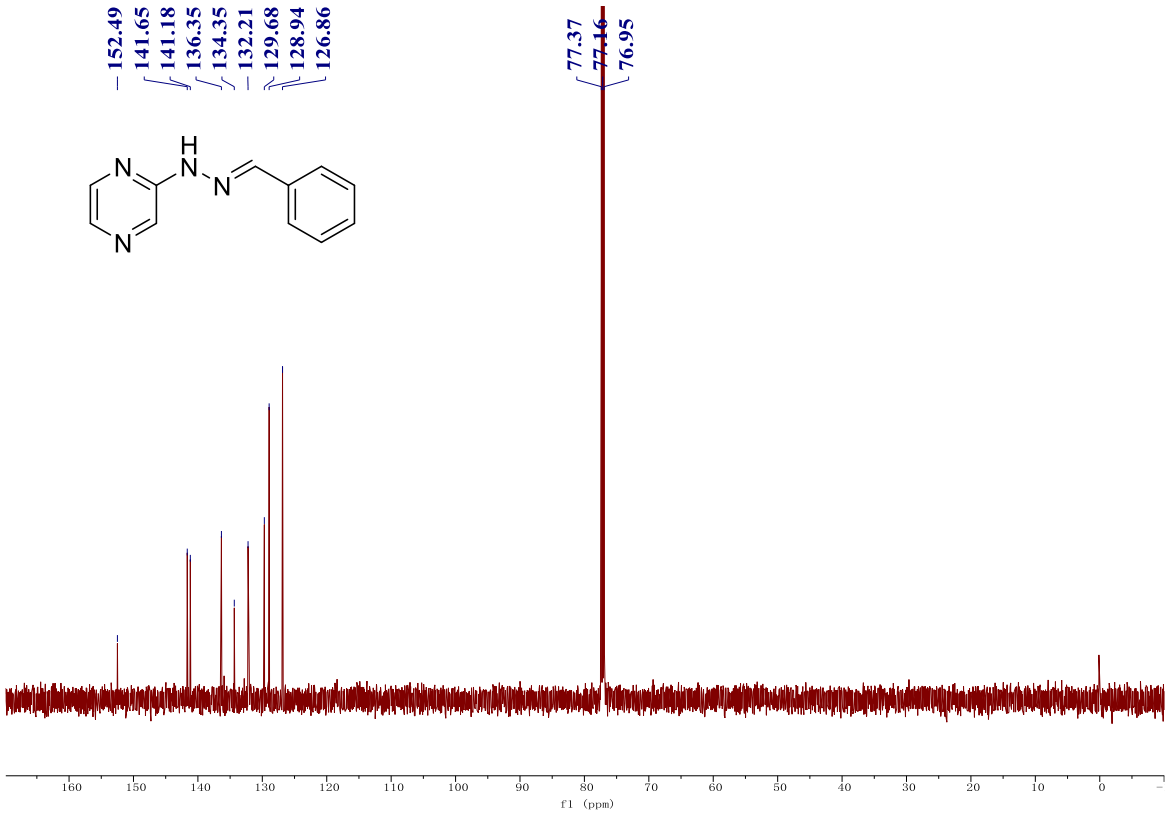
^{13}C NMR Spectrum of 1aaj at 25 °C (CD_3OD, 150 MHz)



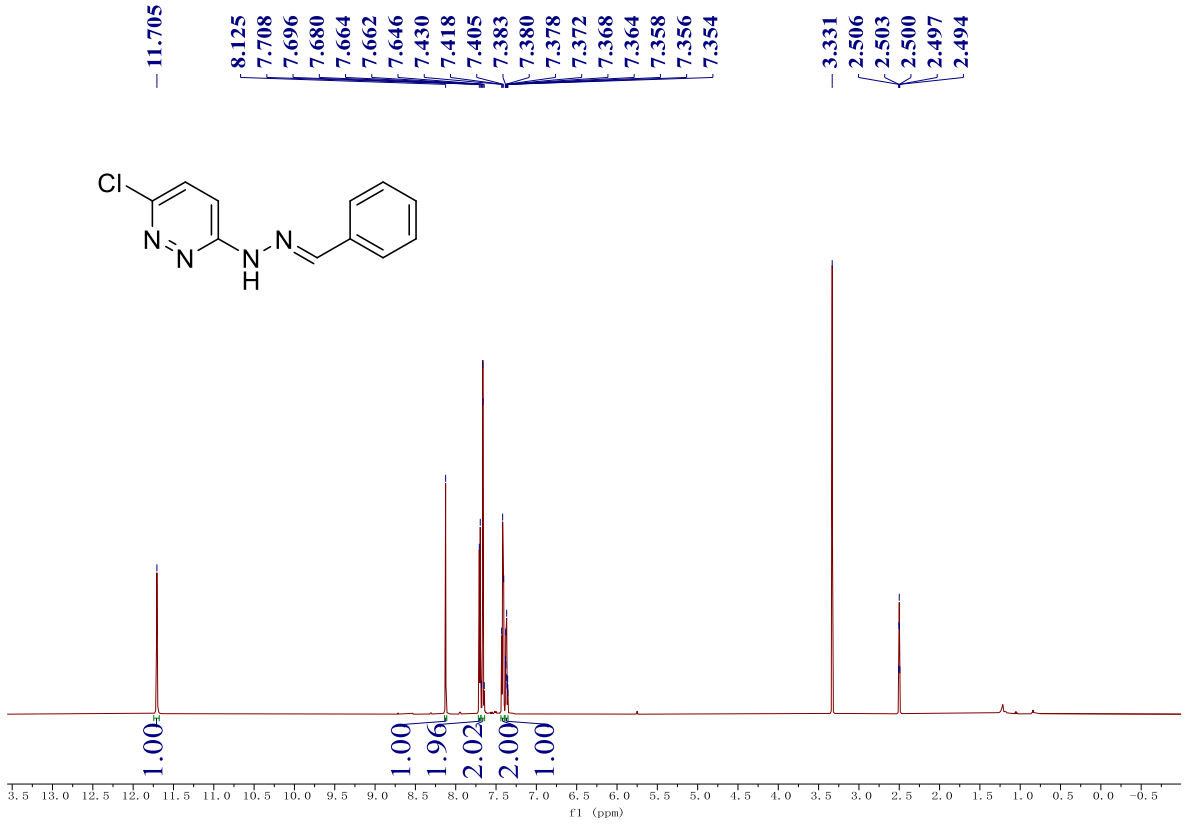
^{1}H NMR Spectrum of 1aak at 25 ^{o}C ($CDCl_3$, 600 MHz)



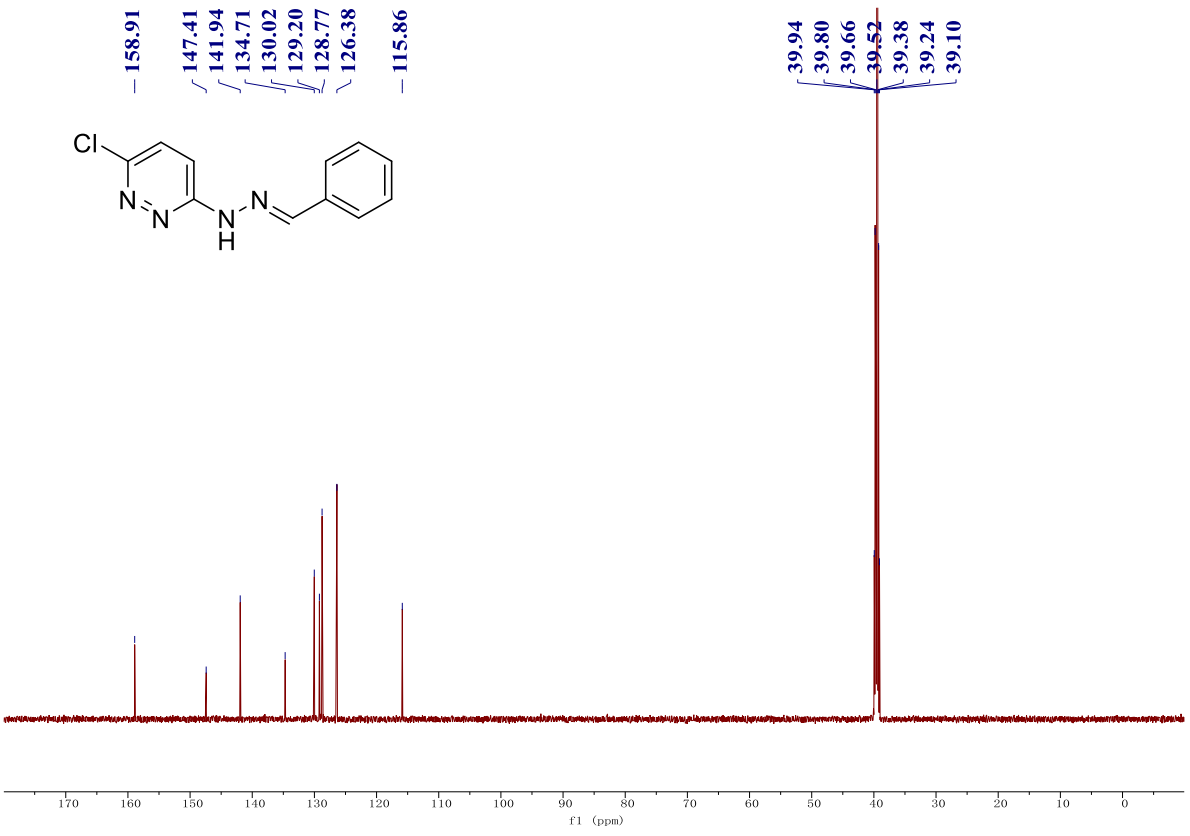
^{13}C NMR Spectrum of 1aak at 25 ^{o}C ($CDCl_3$, 150 MHz)



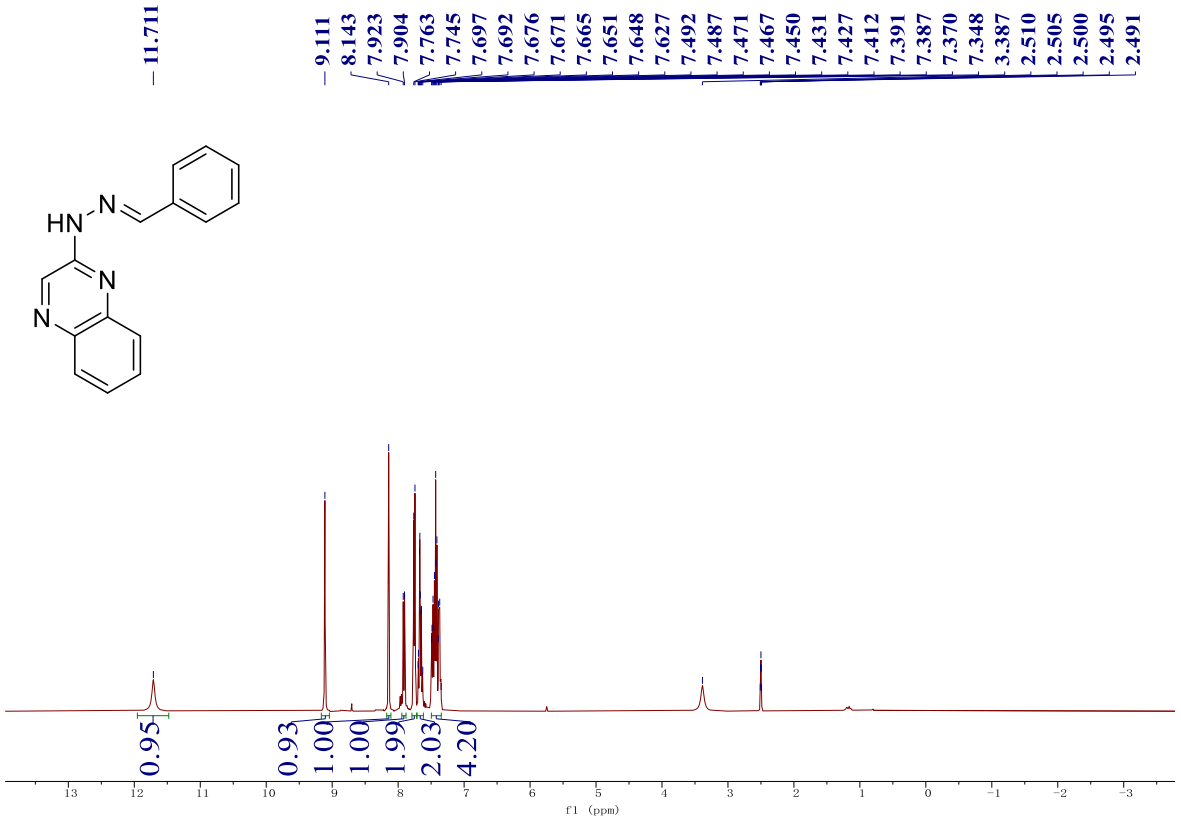
^{1}H NMR Spectrum of 1aal at 25 °C (DMSO-d_6, 600 MHz)



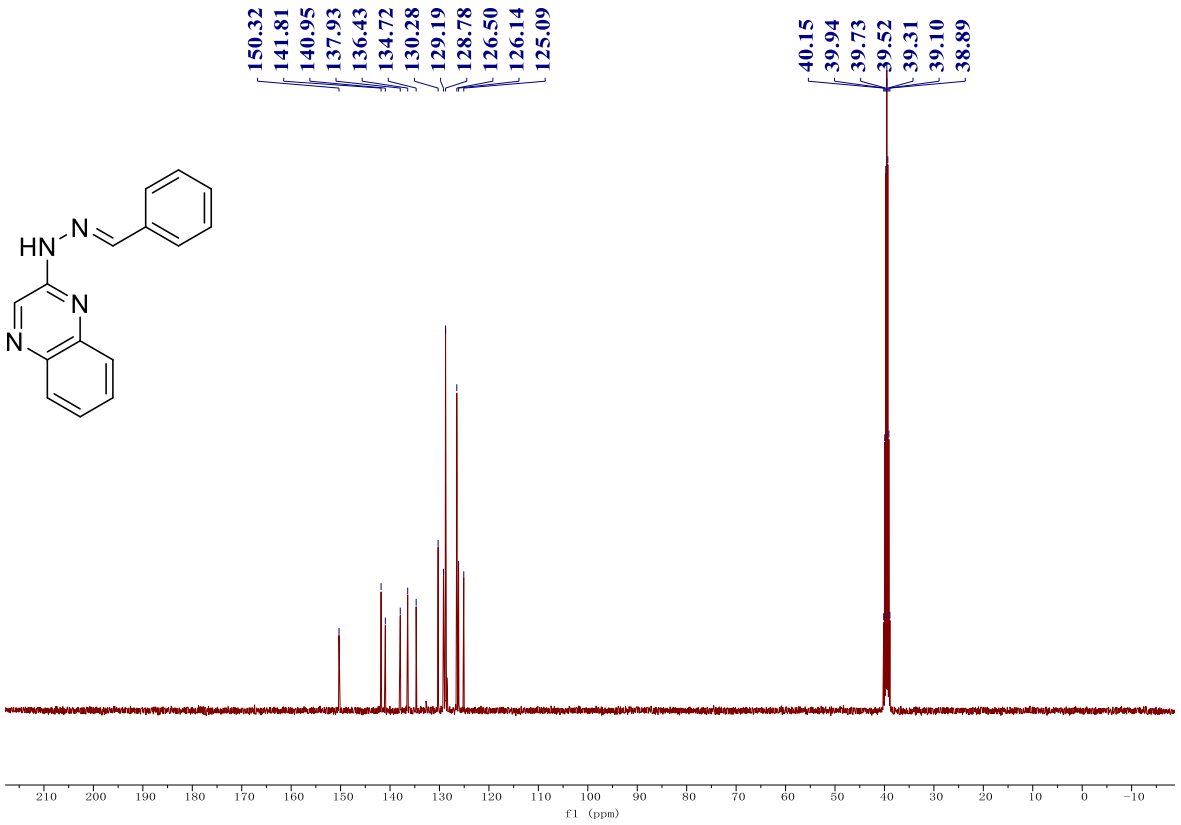
^{13}C NMR Spectrum of 1aal at 25 °C (DMSO-d_6, 150 MHz)



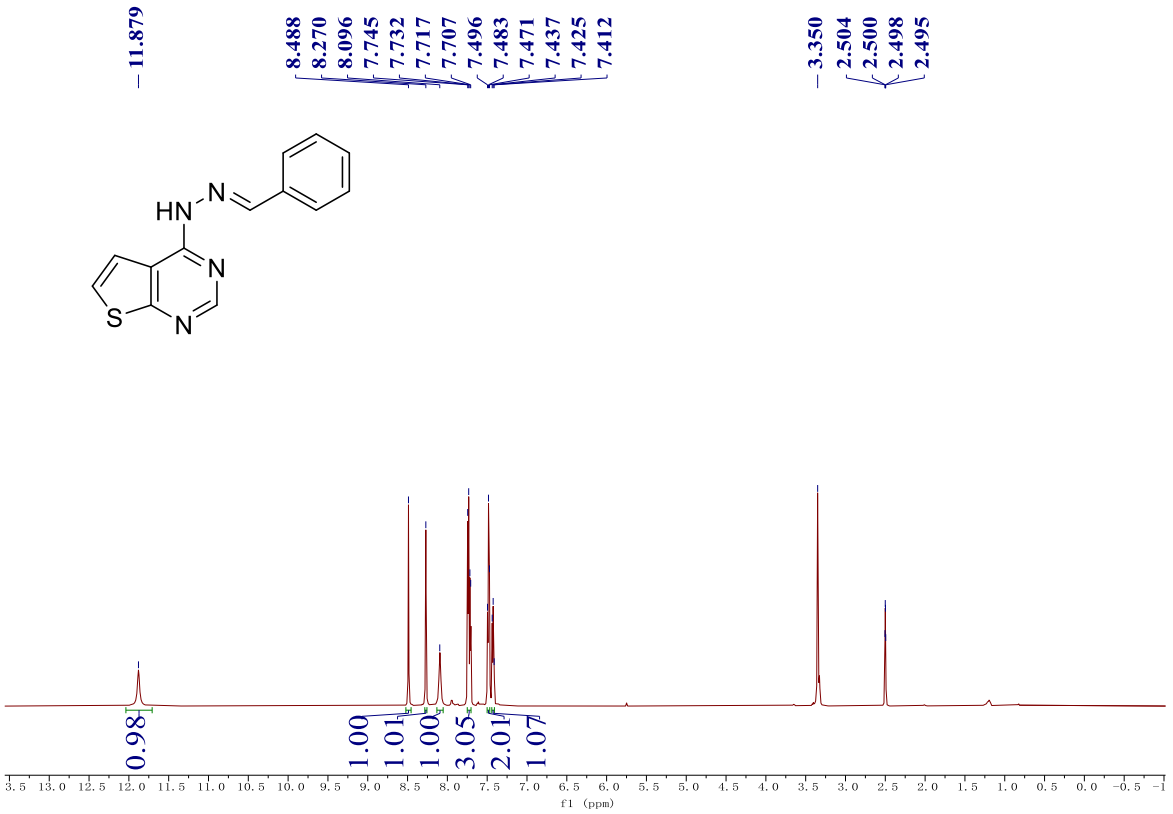
^{1}H NMR Spectrum of 1aan at 25 °C (DMSO-d_6, 400 MHz)



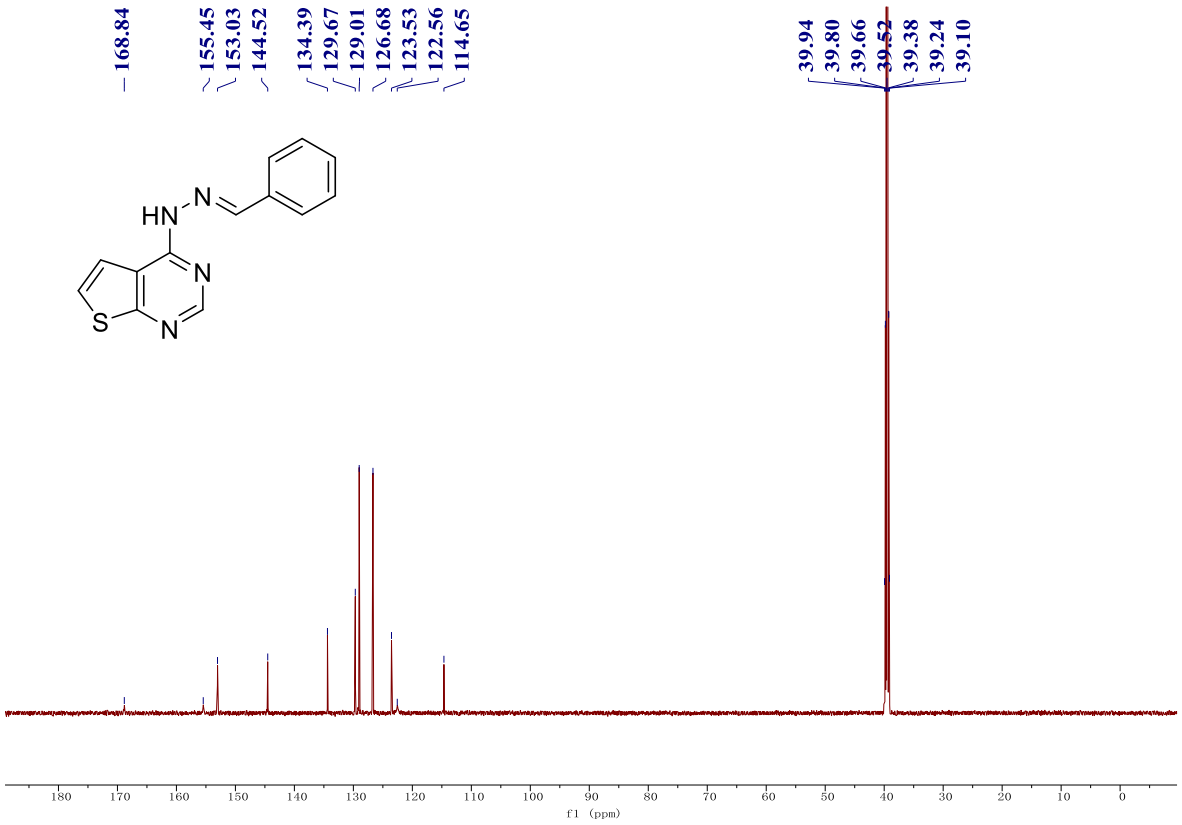
^{13}C NMR Spectrum of 1aan at 25 °C (DMSO-d_6, 100 MHz)



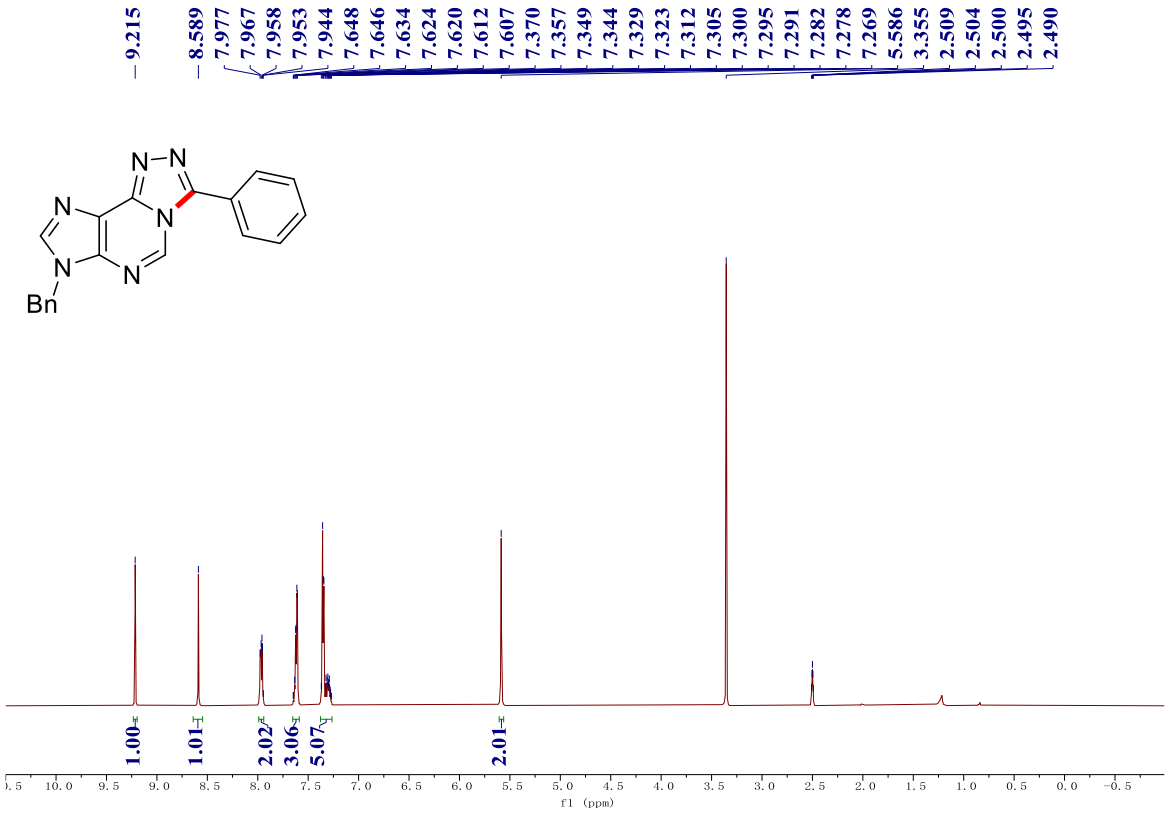
^{1}H NMR Spectrum of 1aao at 25 °C (DMSO-d_6, 600 MHz)



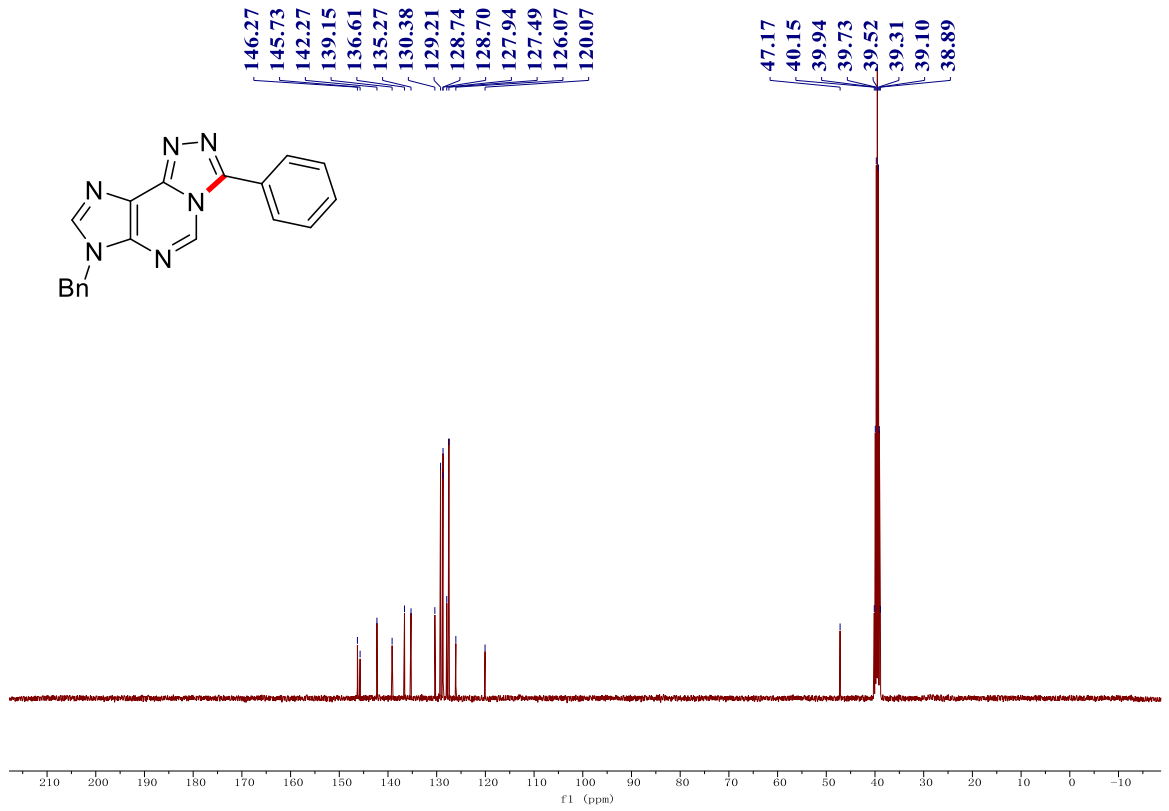
^{13}C NMR Spectrum of 1aao at 25 °C (DMSO-d_6, 150 MHz)



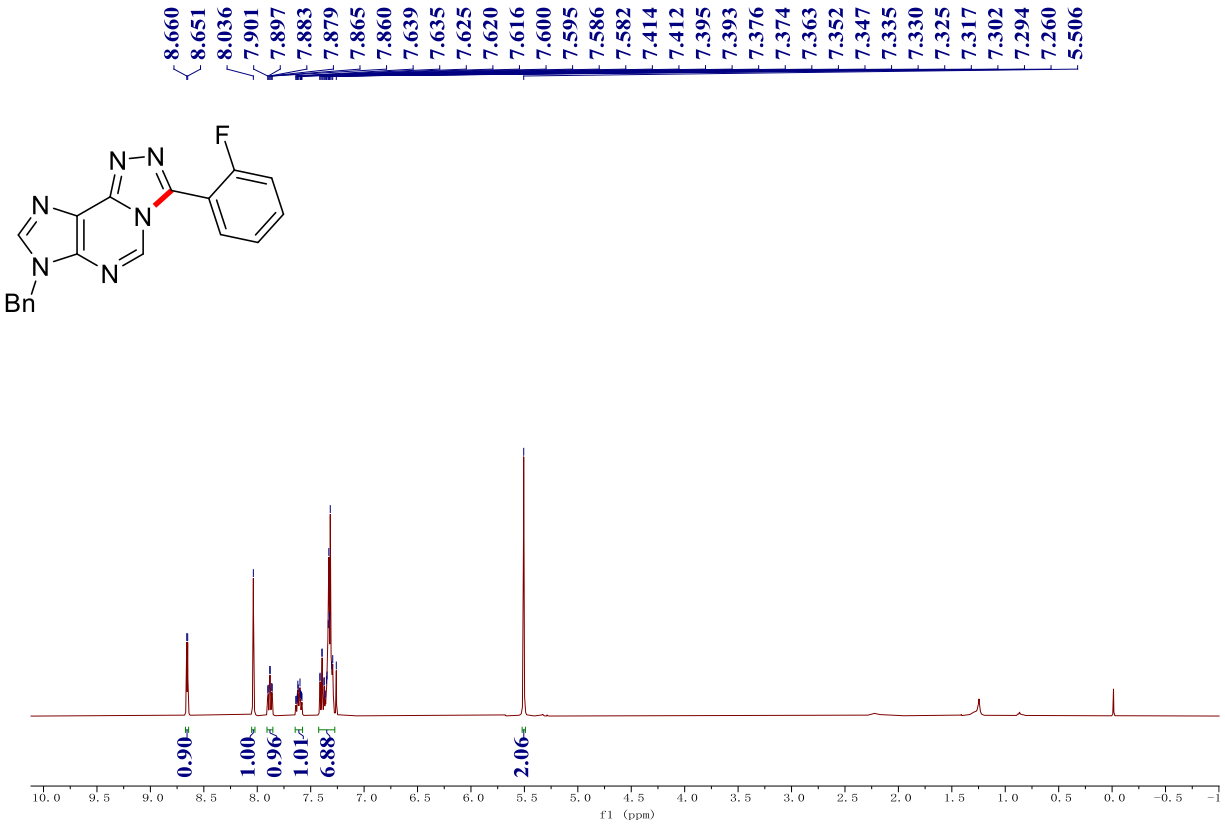
^{1}H NMR Spectrum of 2a at 25 °C (DMSO-d_6, 400 MHz)



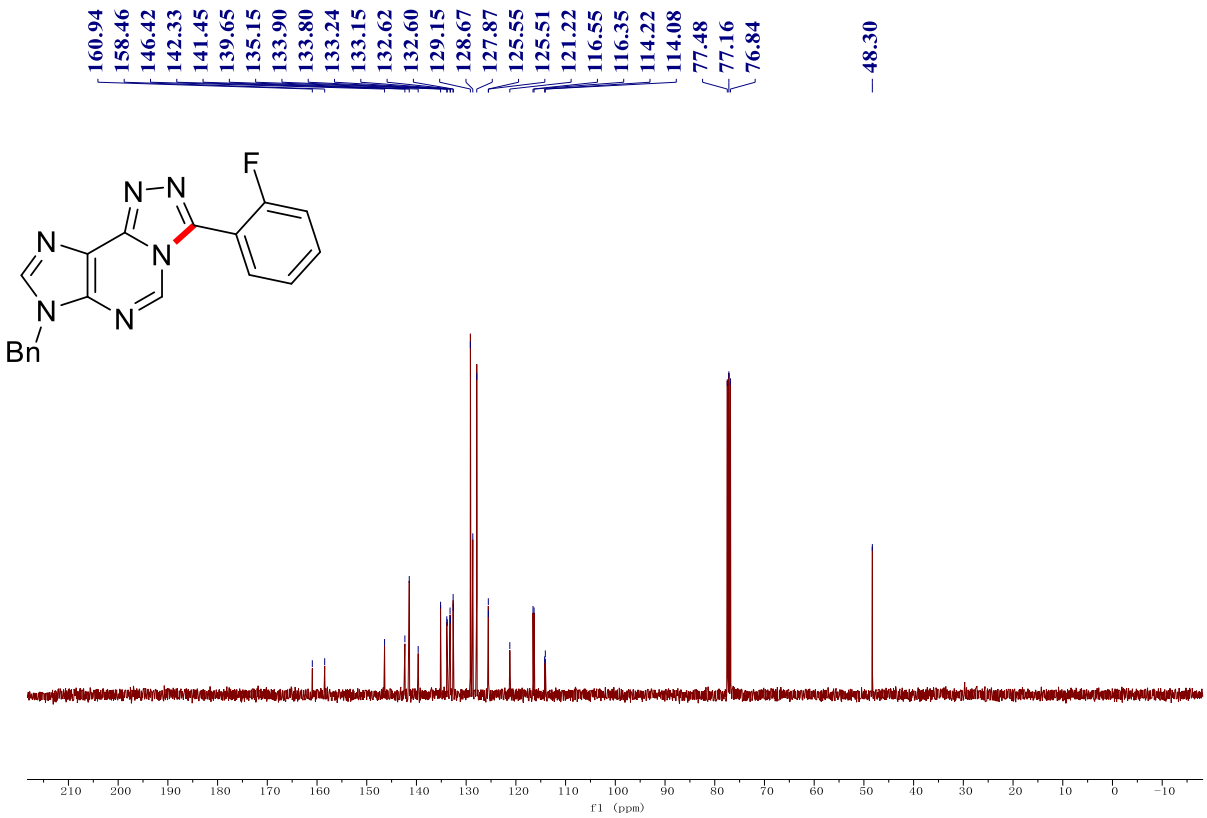
^{13}C NMR Spectrum of 2a at 25 °C (DMSO-d_6, 100 MHz)



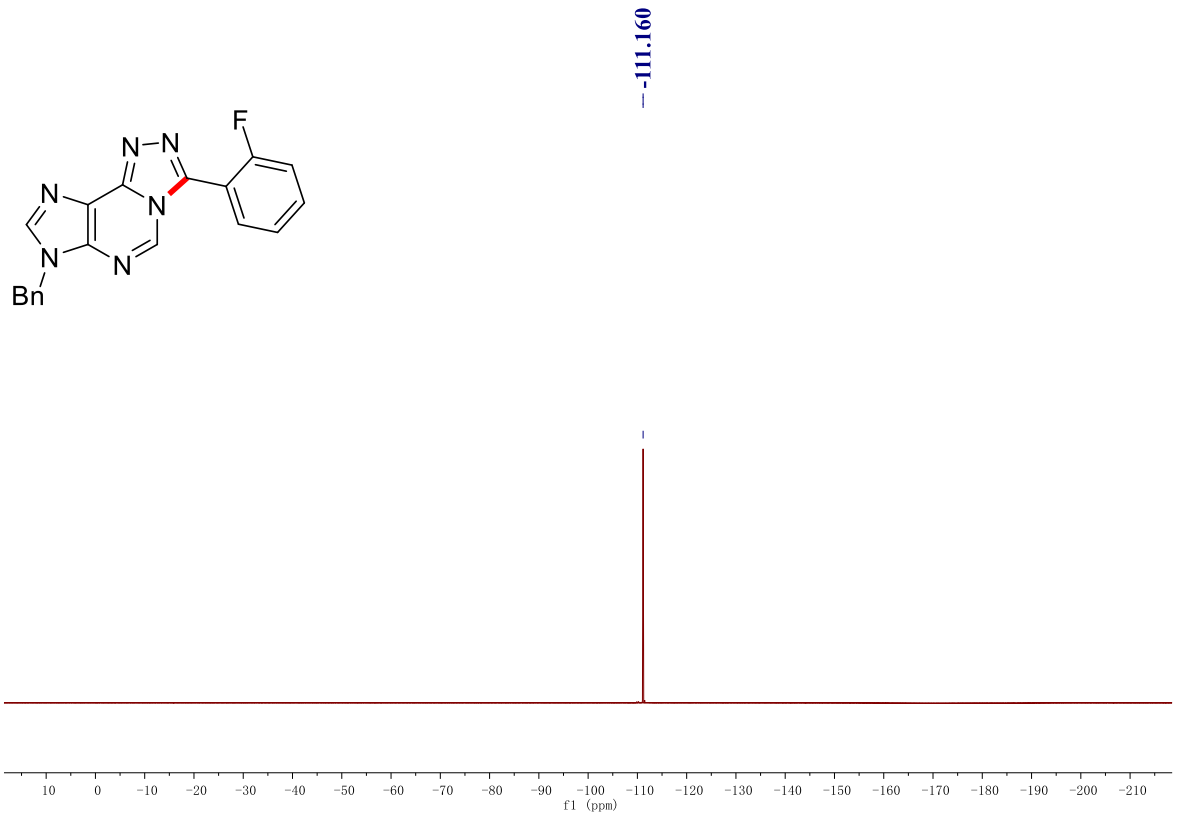
^{1}H NMR Spectrum of 2b at 25 °C ($CDCl_3$, 400 MHz)



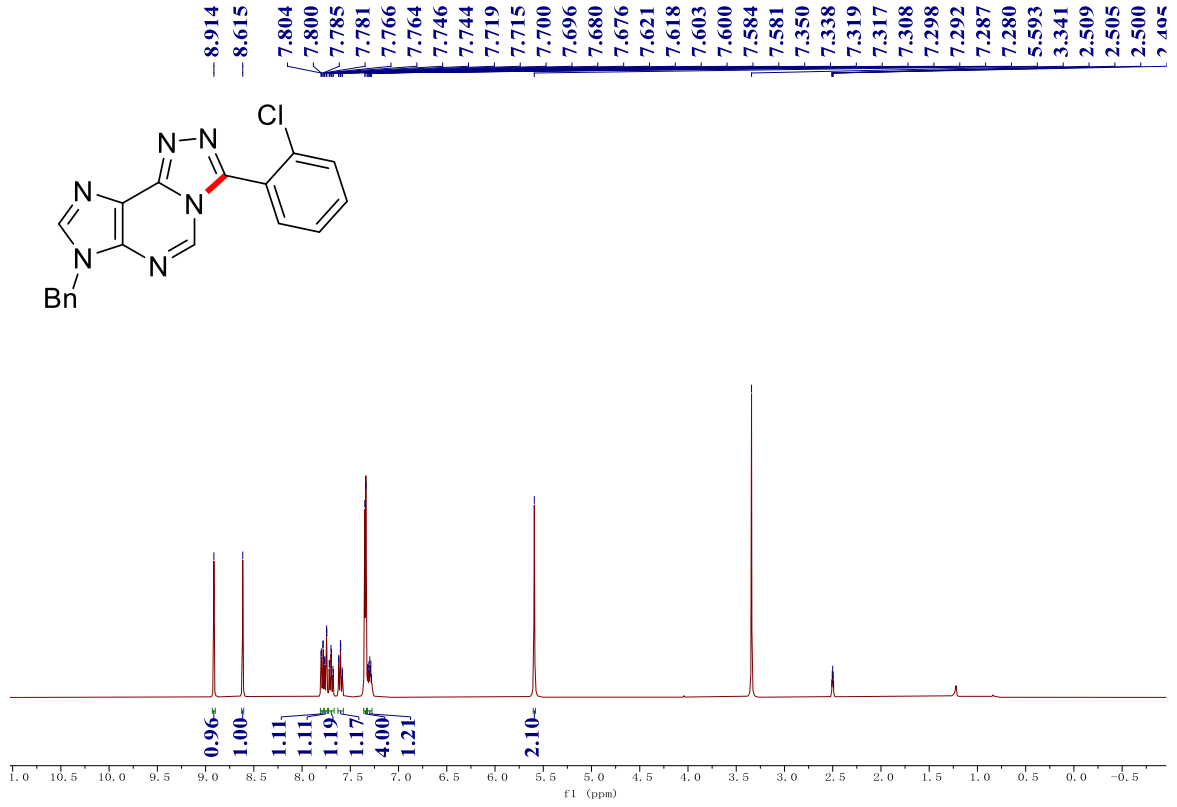
^{13}C NMR Spectrum of 2b at 25 °C ($CDCl_3$, 100 MHz)



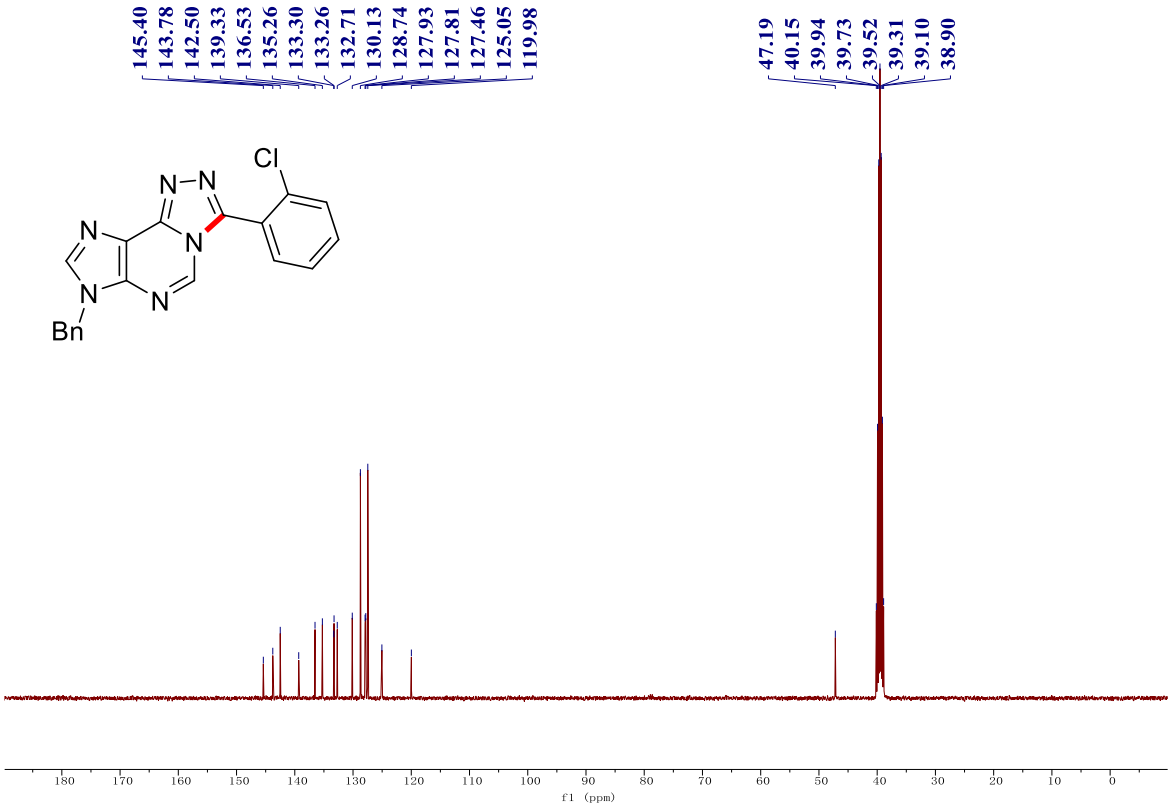
^{19}F NMR Spectrum of 2b at 25 °C ($CDCl_3$, 376 MHz)



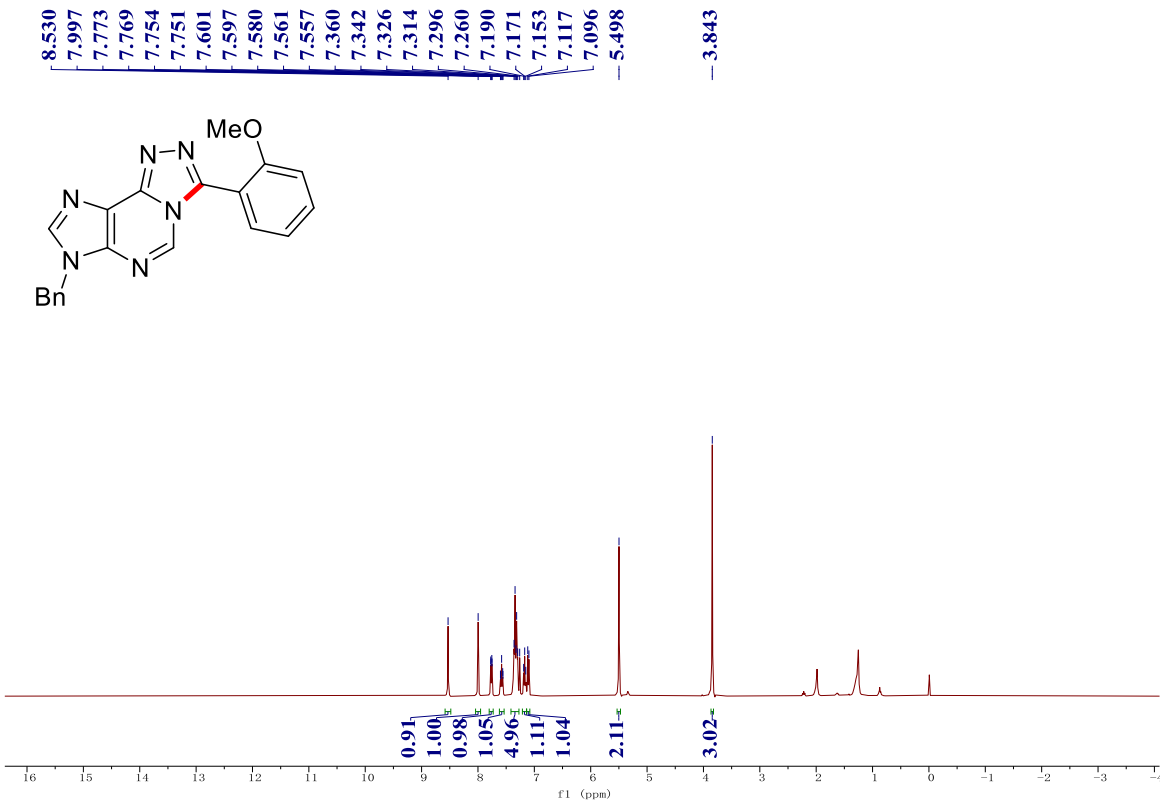
^{1}H NMR Spectrum of 2c at 25 °C (DMSO-d_6, 400 MHz)



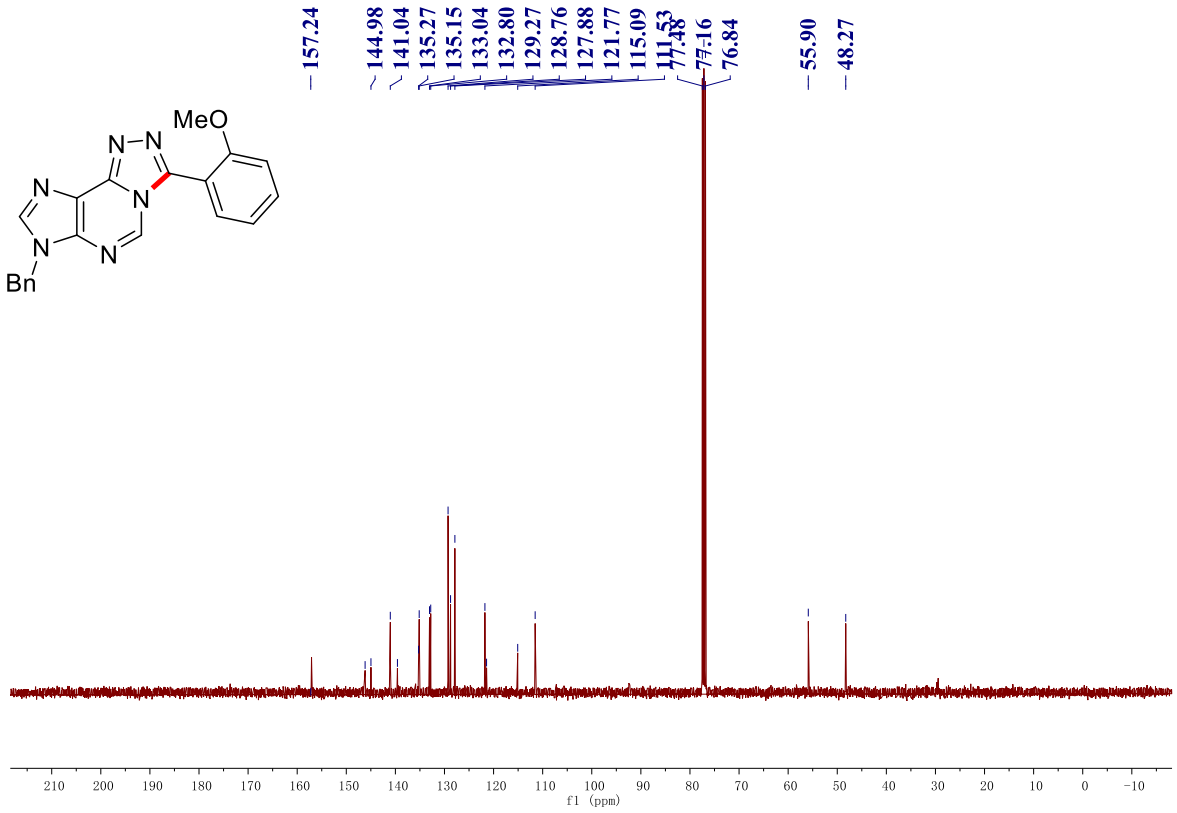
^{13}C NMR Spectrum of 2c at 25 °C (DMSO-d_6, 100 MHz)



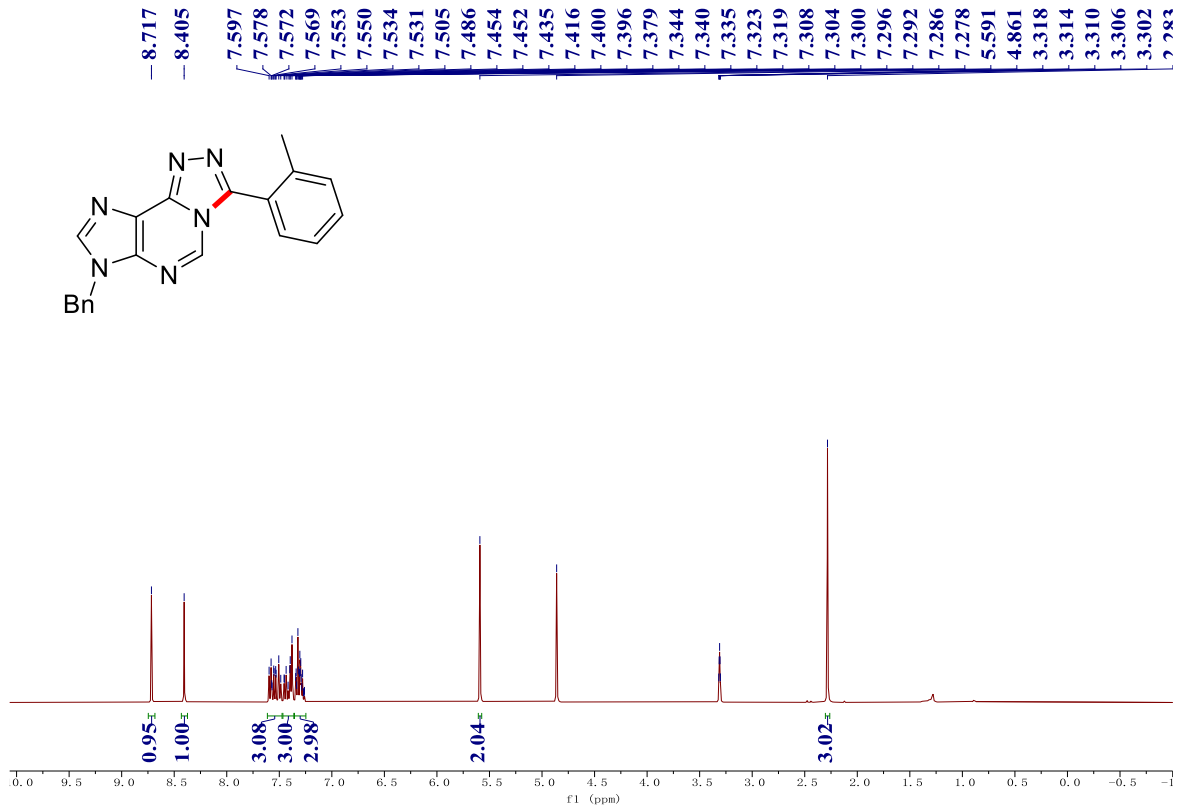
^{1}H NMR Spectrum of 2d at 25 °C ($CDCl_3$, 400 MHz)



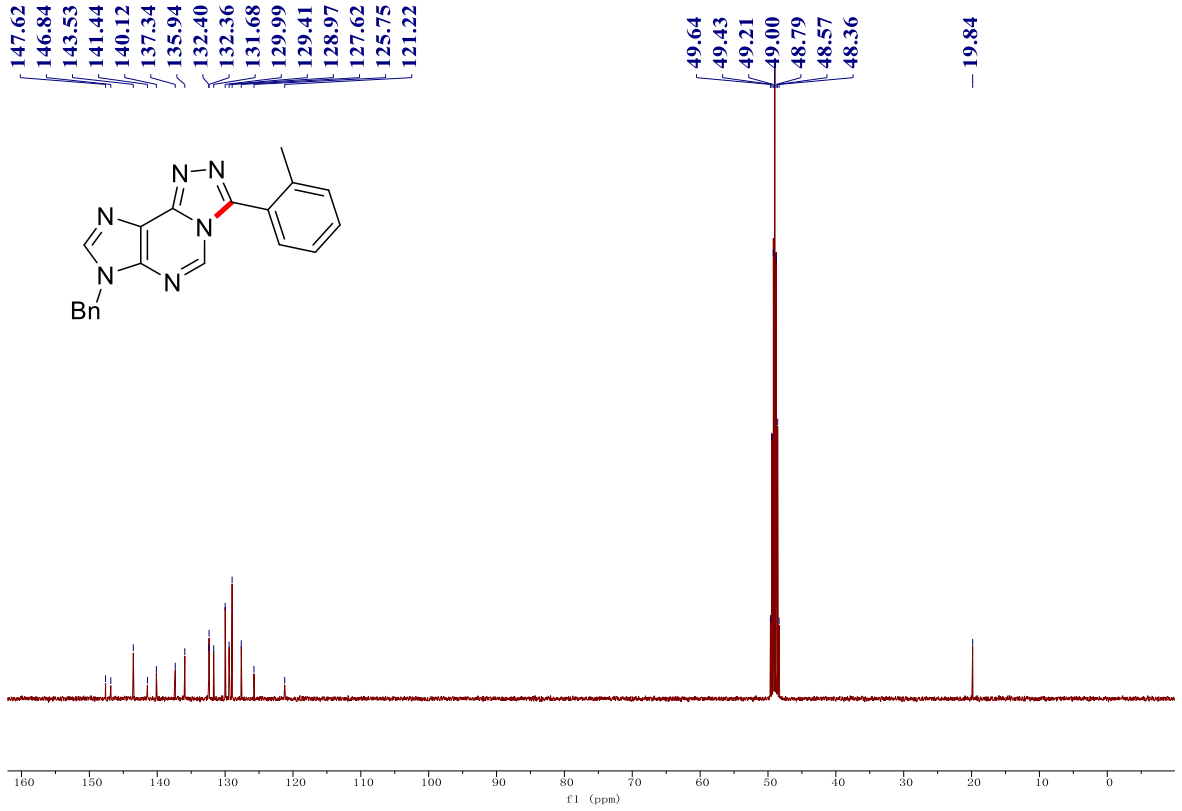
^{13}C NMR Spectrum of 2d at 25 °C ($CDCl_3$, 100 MHz)



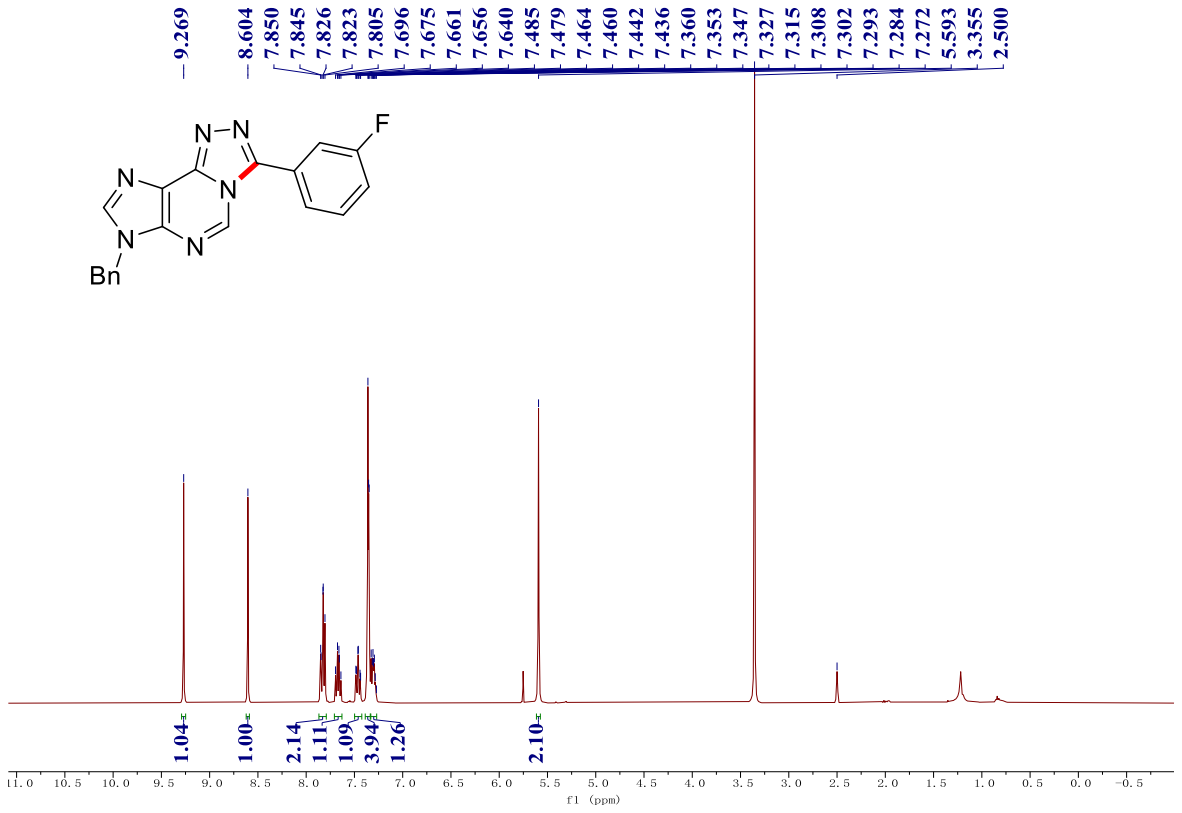
^{1}H NMR Spectrum of 2e at 25 °C (CD_3OD, 400 MHz)



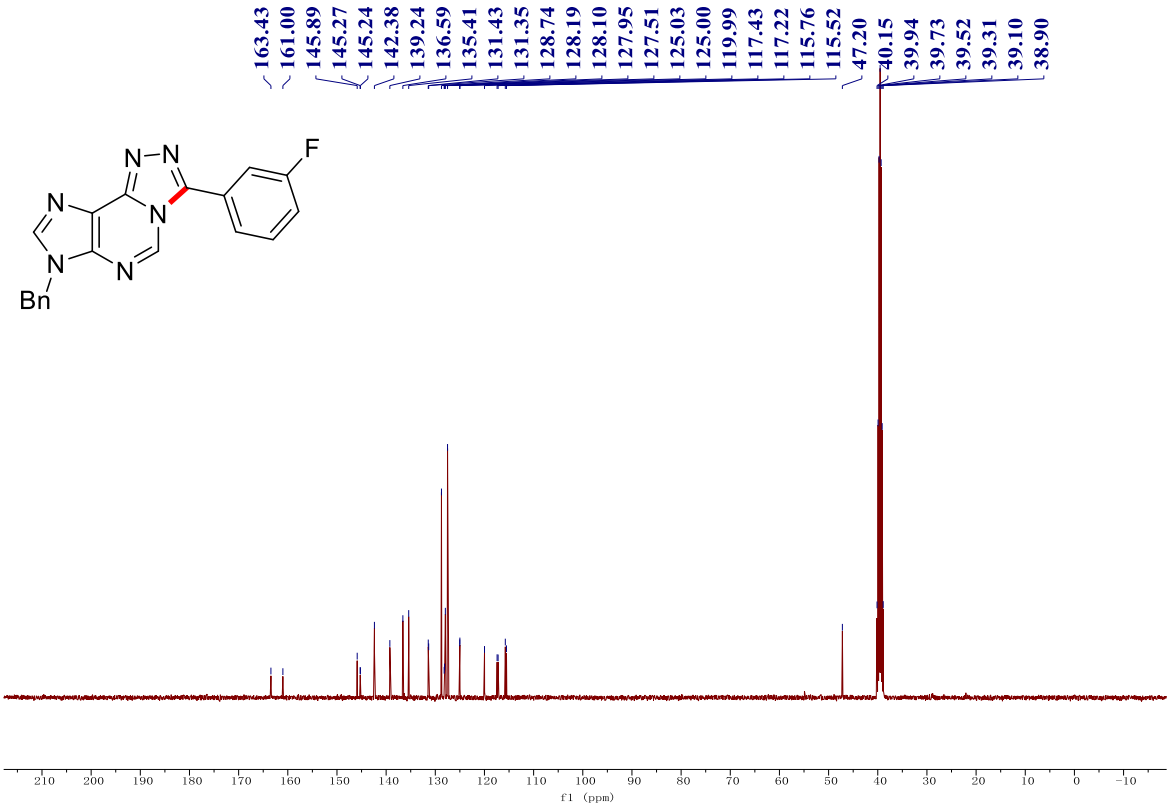
^{13}C NMR Spectrum of 2e at 25 °C (CD_3OD, 100 MHz)



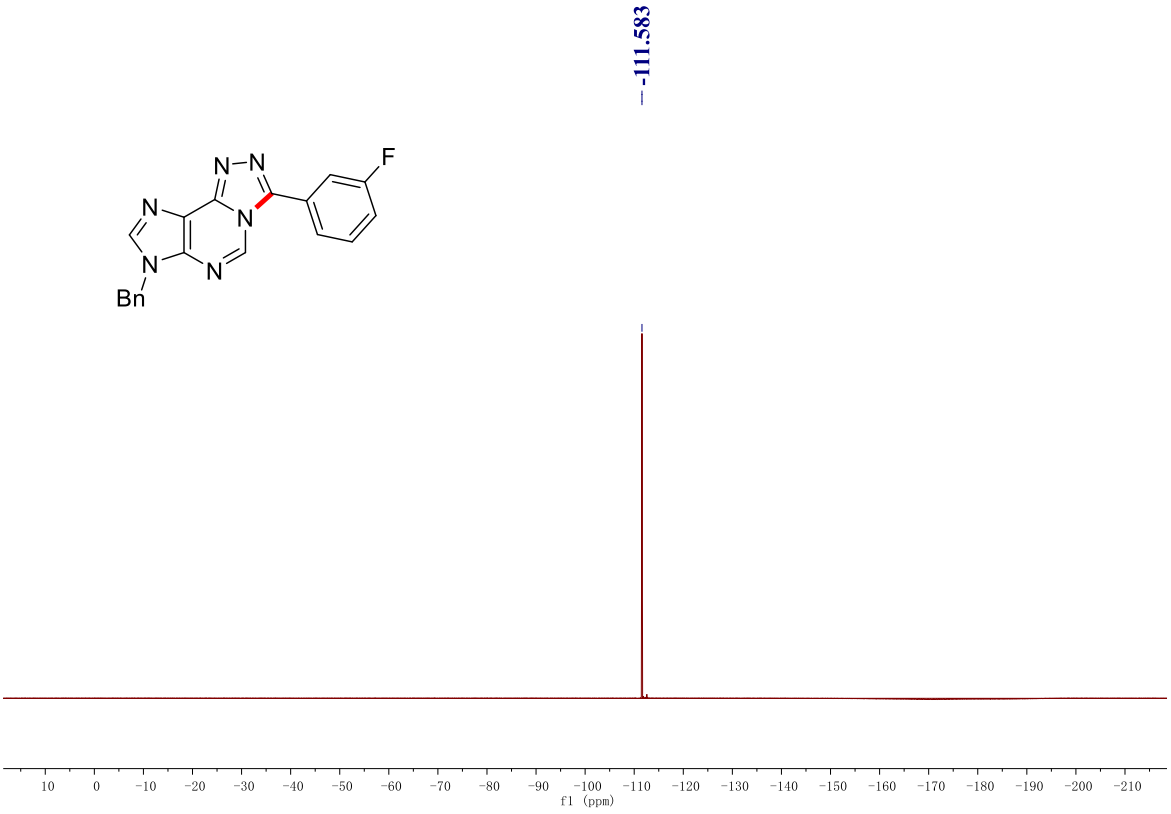
^{1}H NMR Spectrum of 2f at 25 °C (DMSO-d_6, 400 MHz)



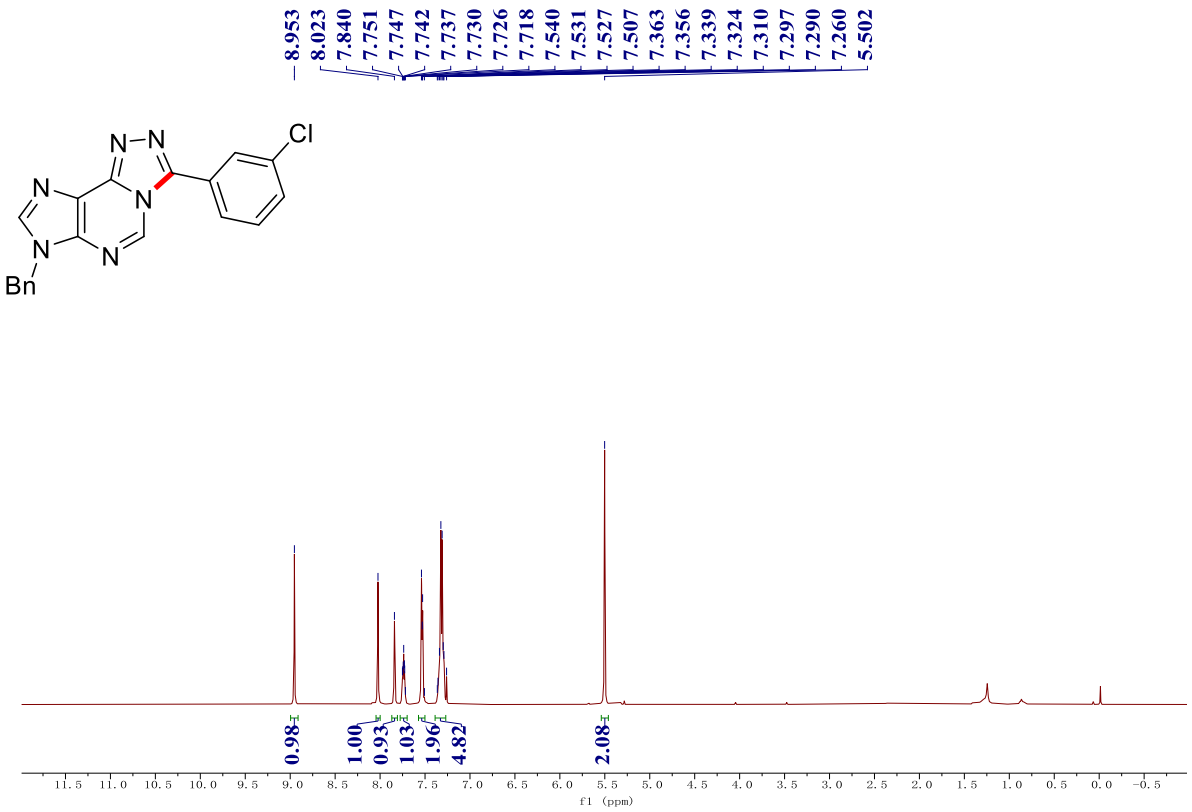
^{13}C NMR Spectrum of 2f at 25 °C (DMSO-d_6, 100 MHz)



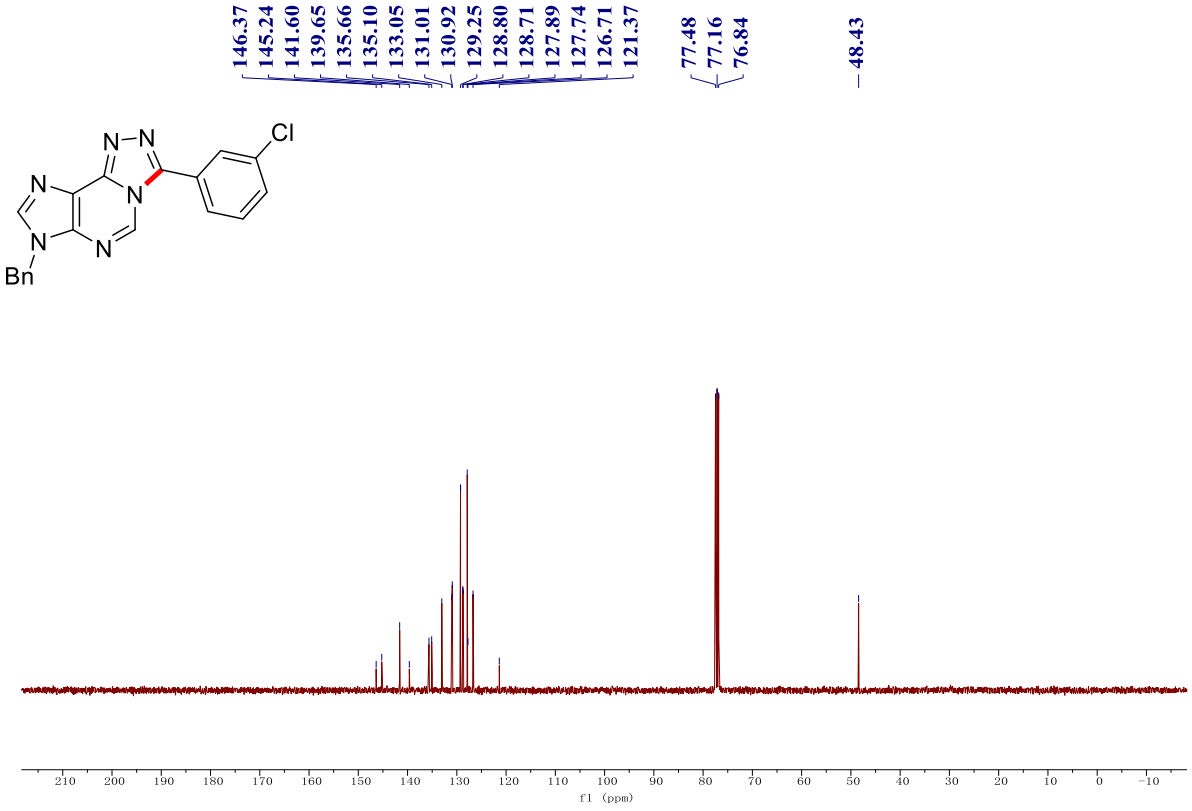
^{19}F NMR Spectrum of 2f at 25 °C (DMSO-d_6, 376 MHz)



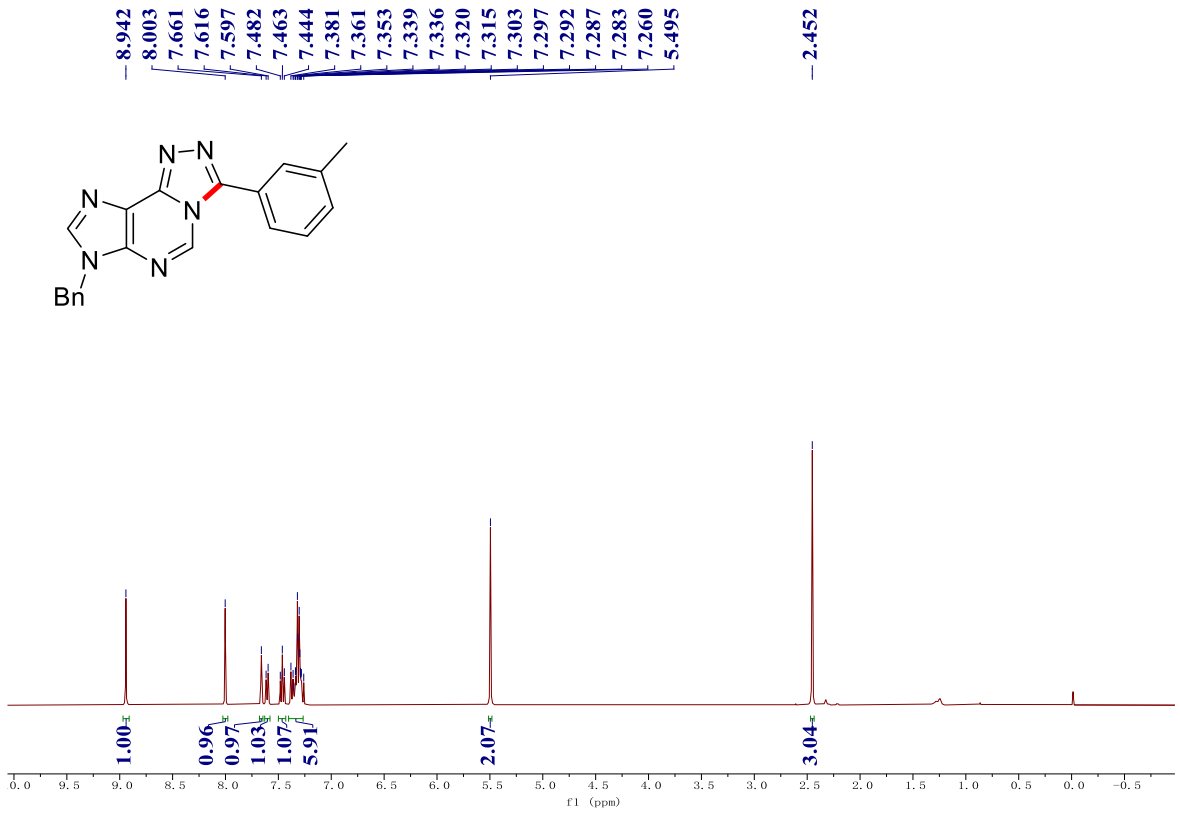
^{1}H NMR Spectrum of 2g at 25 ^{o}C ($CDCl_3$, 400 MHz)



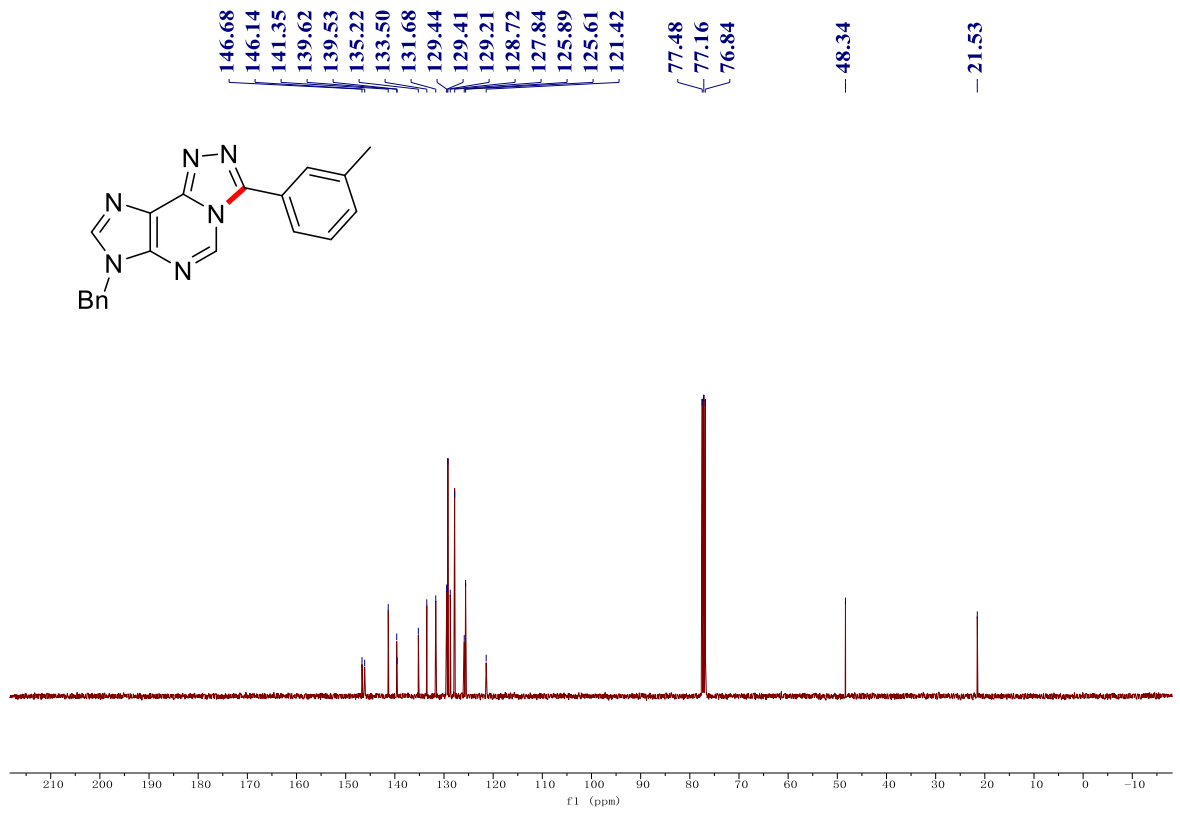
^{13}C NMR Spectrum of 2g at 25 ^{o}C ($CDCl_3$, 100 MHz)



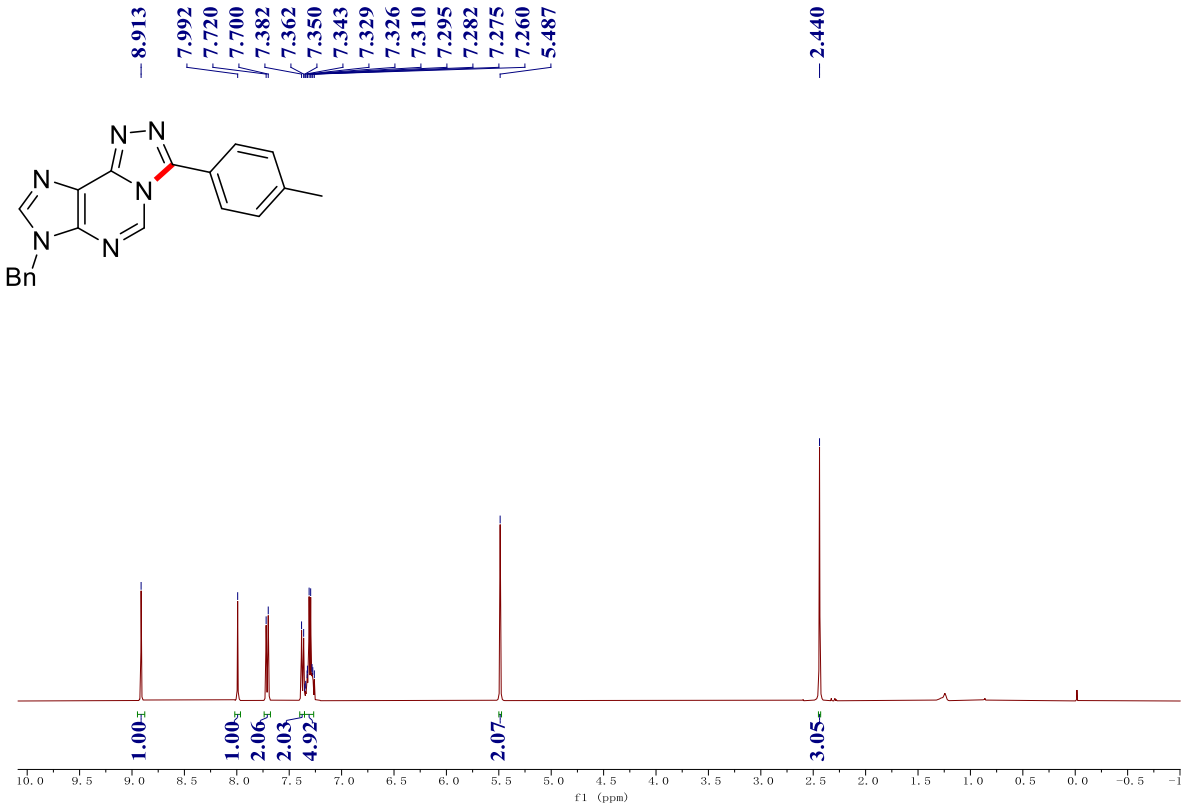
^{1}H NMR Spectrum of 2h at 25 ^{o}C ($CDCl_3$, 400 MHz)



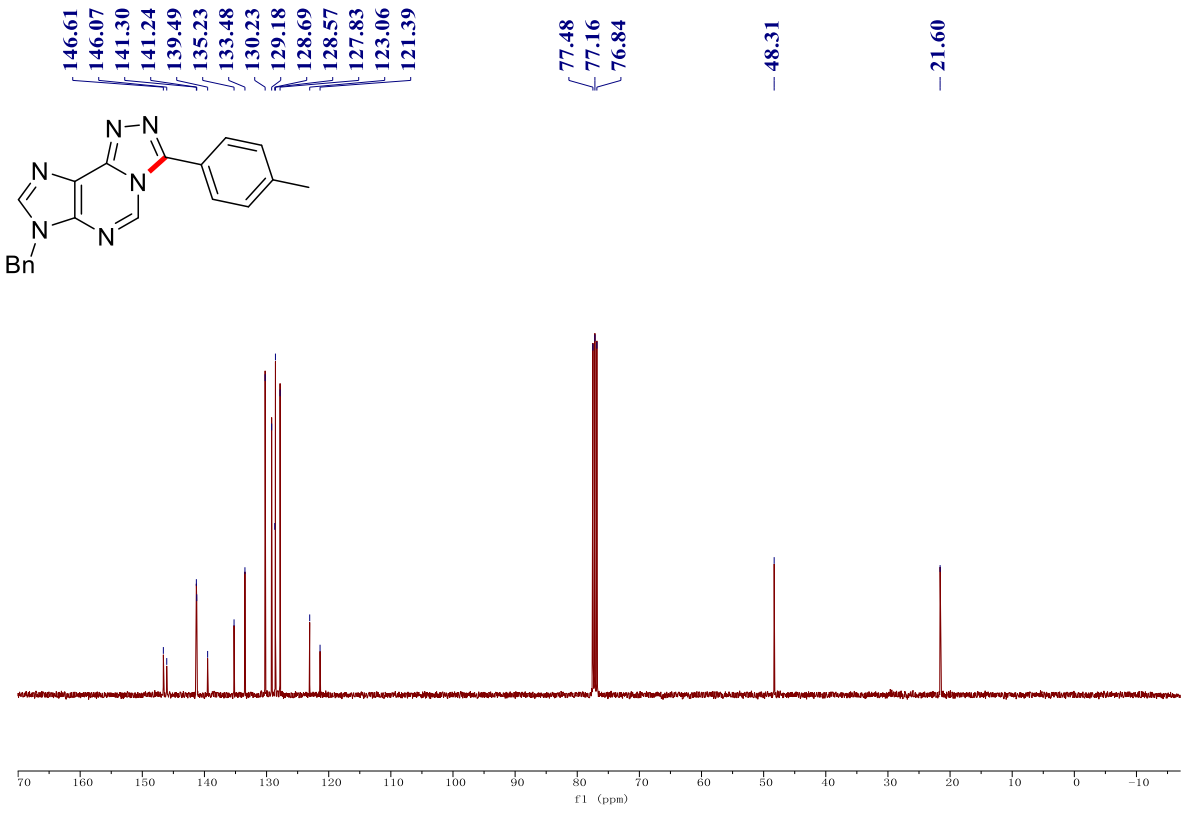
^{13}C NMR Spectrum of 2h at 25 ^{o}C ($CDCl_3$, 100 MHz)



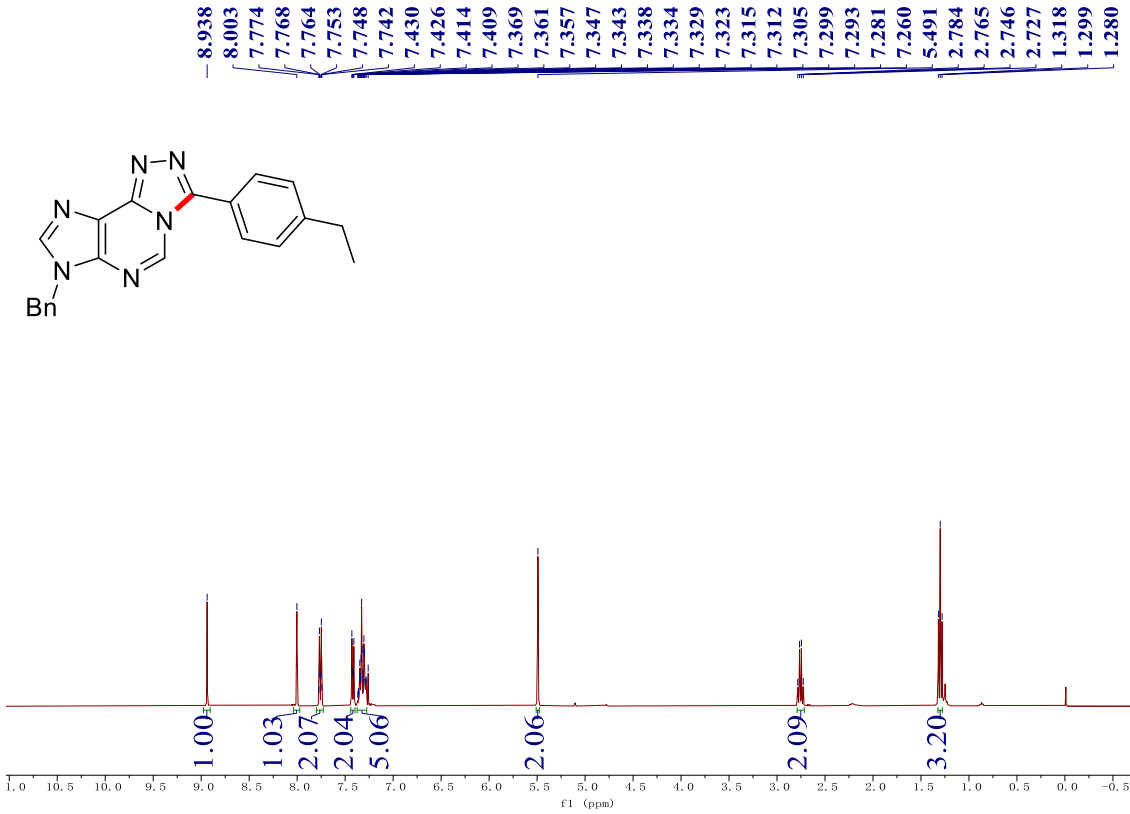
^{1}H NMR Spectrum of 2i at 25 ^{o}C ($CDCl_3$, 400 MHz)



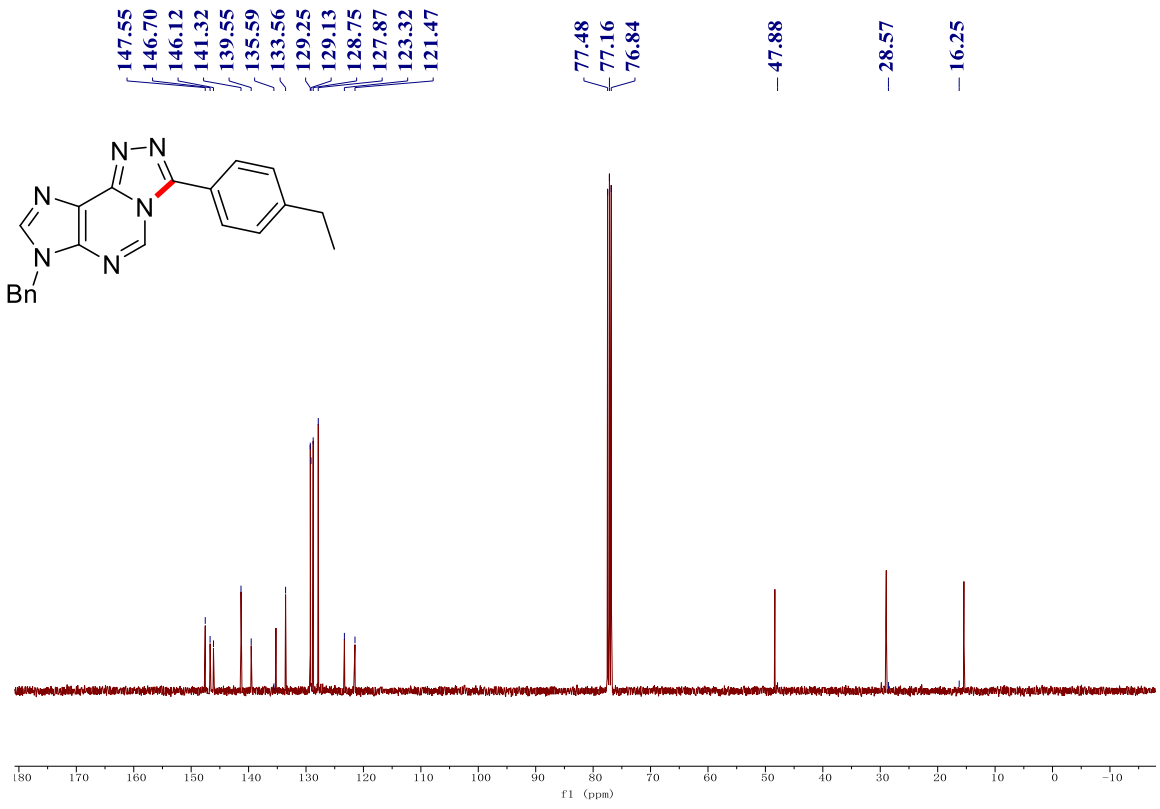
^{13}C NMR Spectrum of 2i at 25 ^{o}C ($CDCl_3$, 100 MHz)



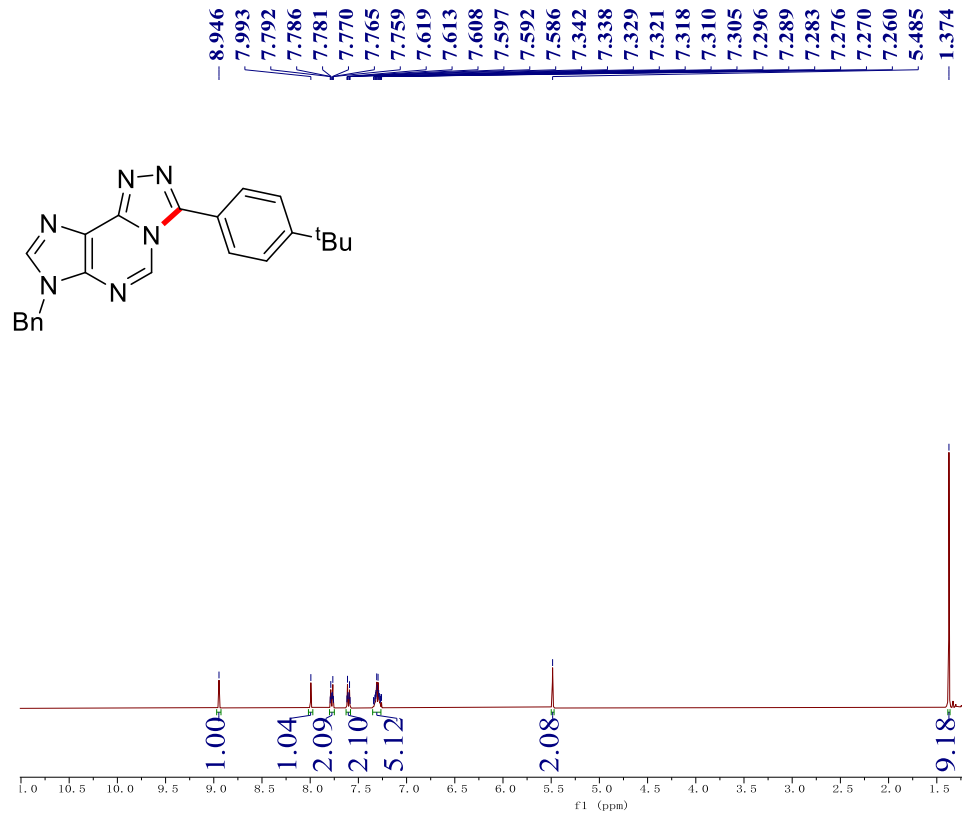
^{1}H NMR Spectrum of 2j at 25 °C ($CDCl_3$, 400 MHz)



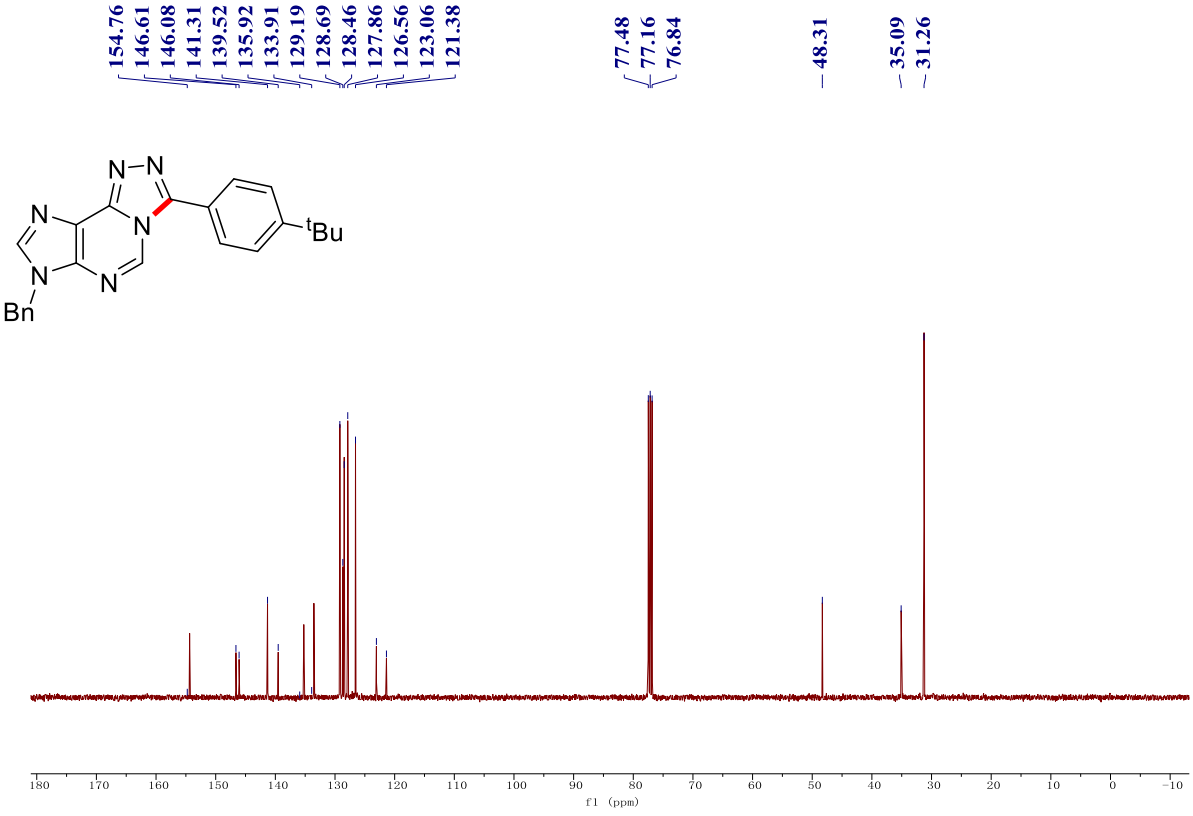
^{13}C NMR Spectrum of 2j at 25 °C ($CDCl_3$, 100 MHz)



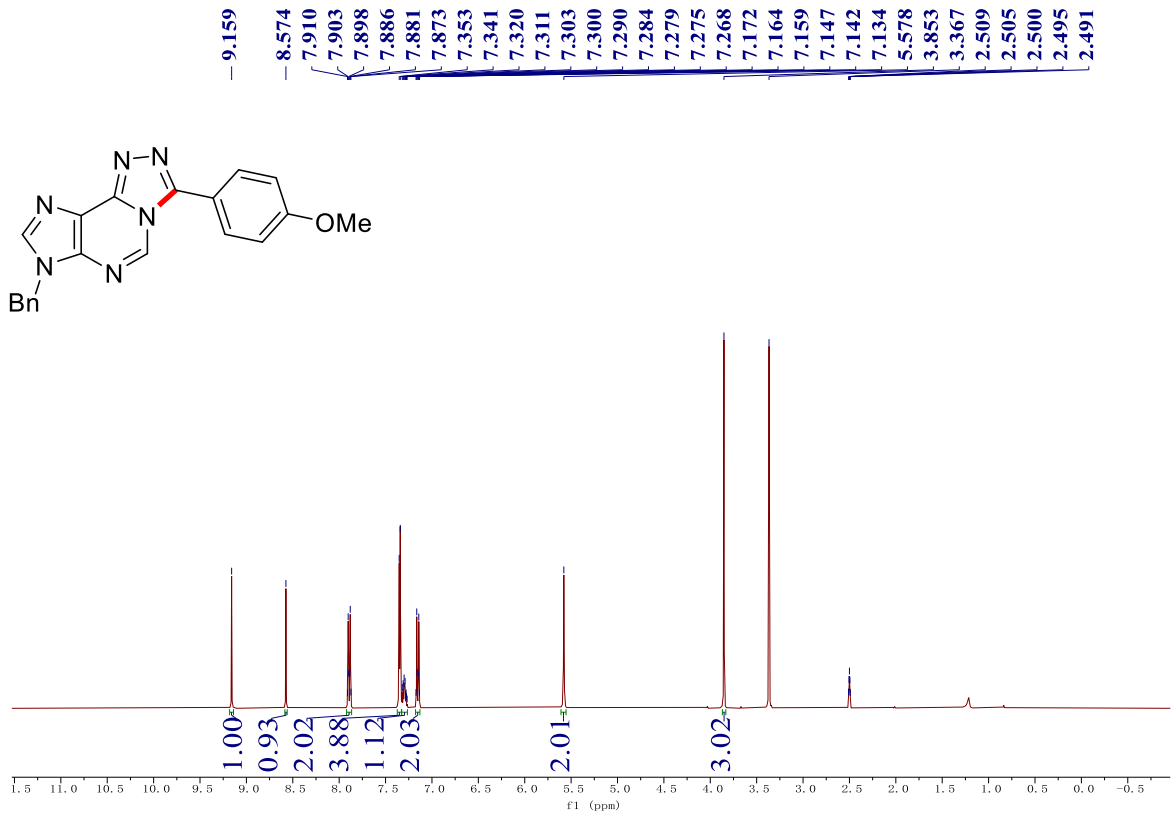
^{1}H NMR Spectrum of 2k at 25 °C ($CDCl_3$, 400 MHz)



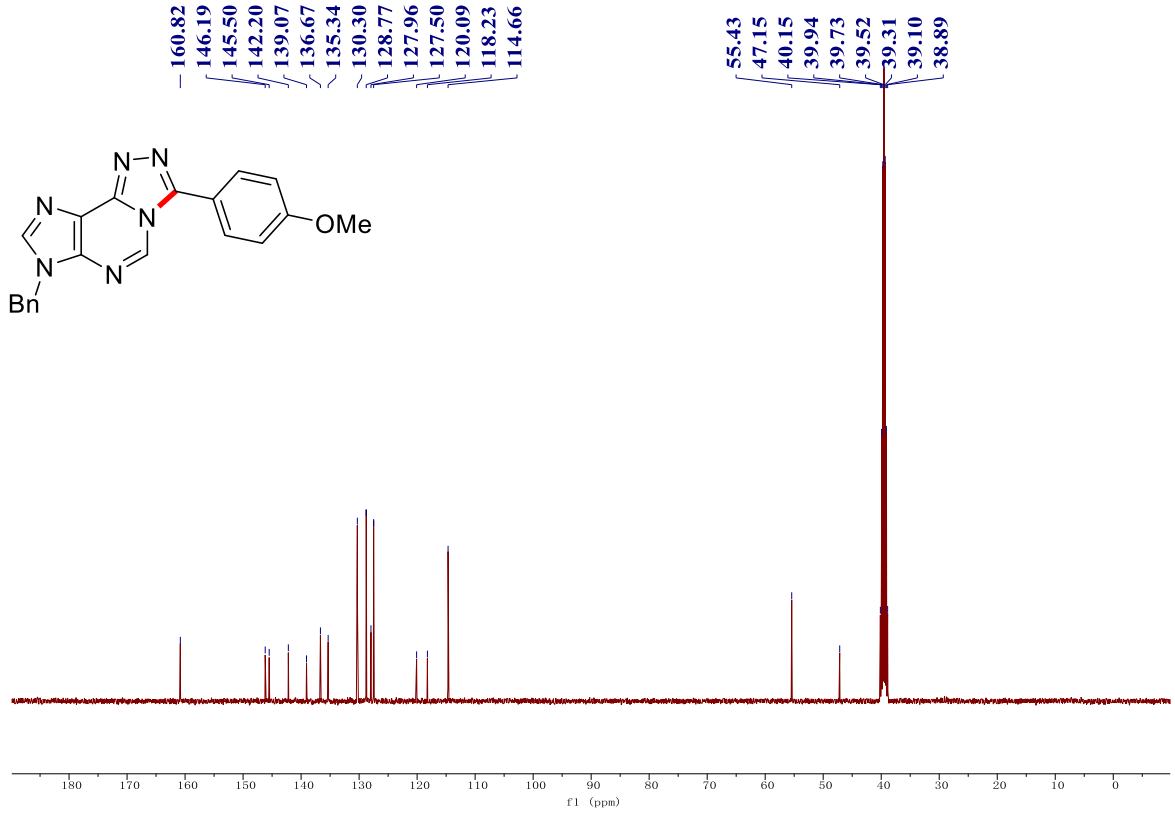
^{13}C NMR Spectrum of 2k at 25 °C ($CDCl_3$, 100 MHz)



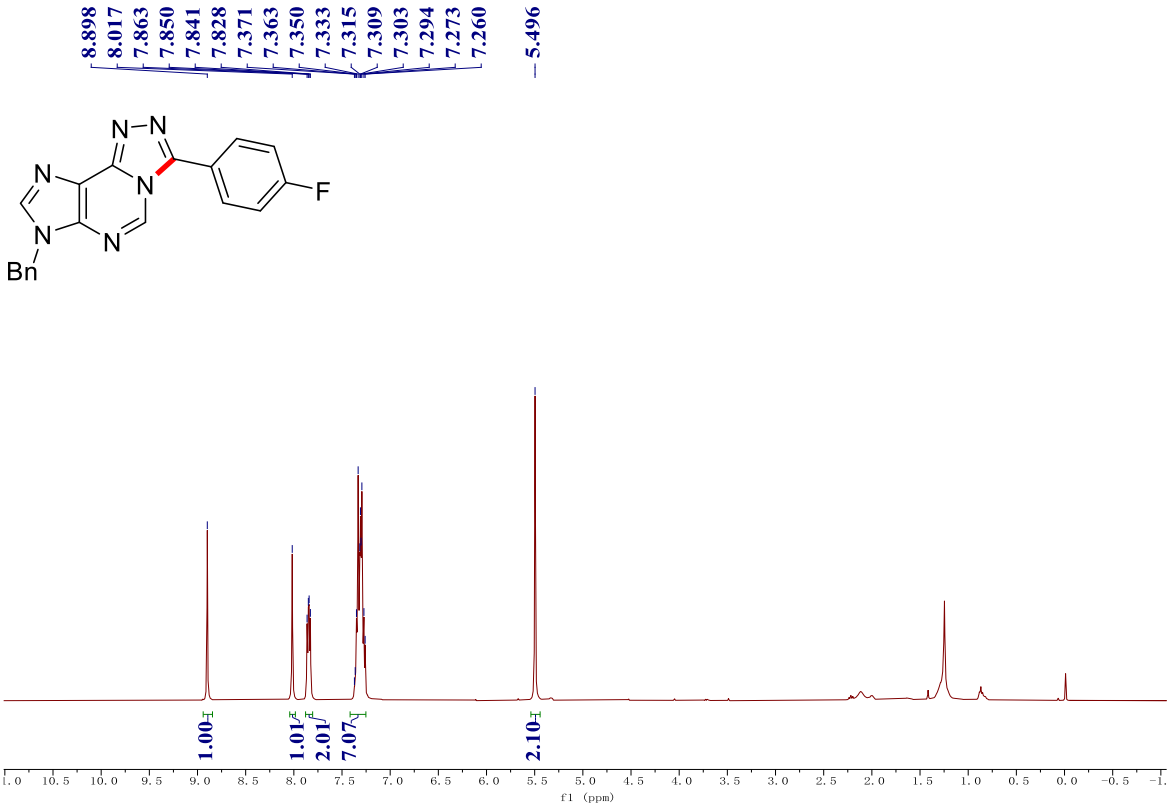
^{1}H NMR Spectrum of 2l at 25 °C (DMSO-d_6, 400 MHz)



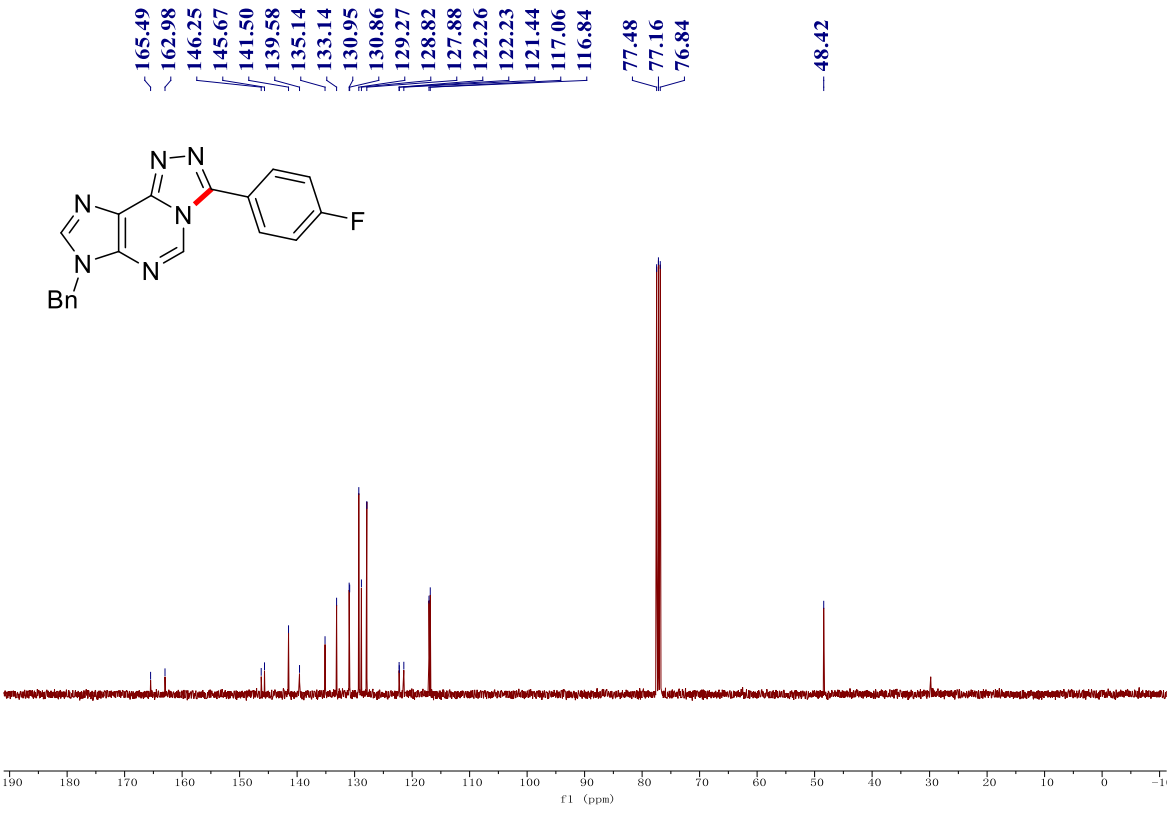
^{13}C NMR Spectrum of 2l at 25 °C (DMSO-d_6, 100 MHz)



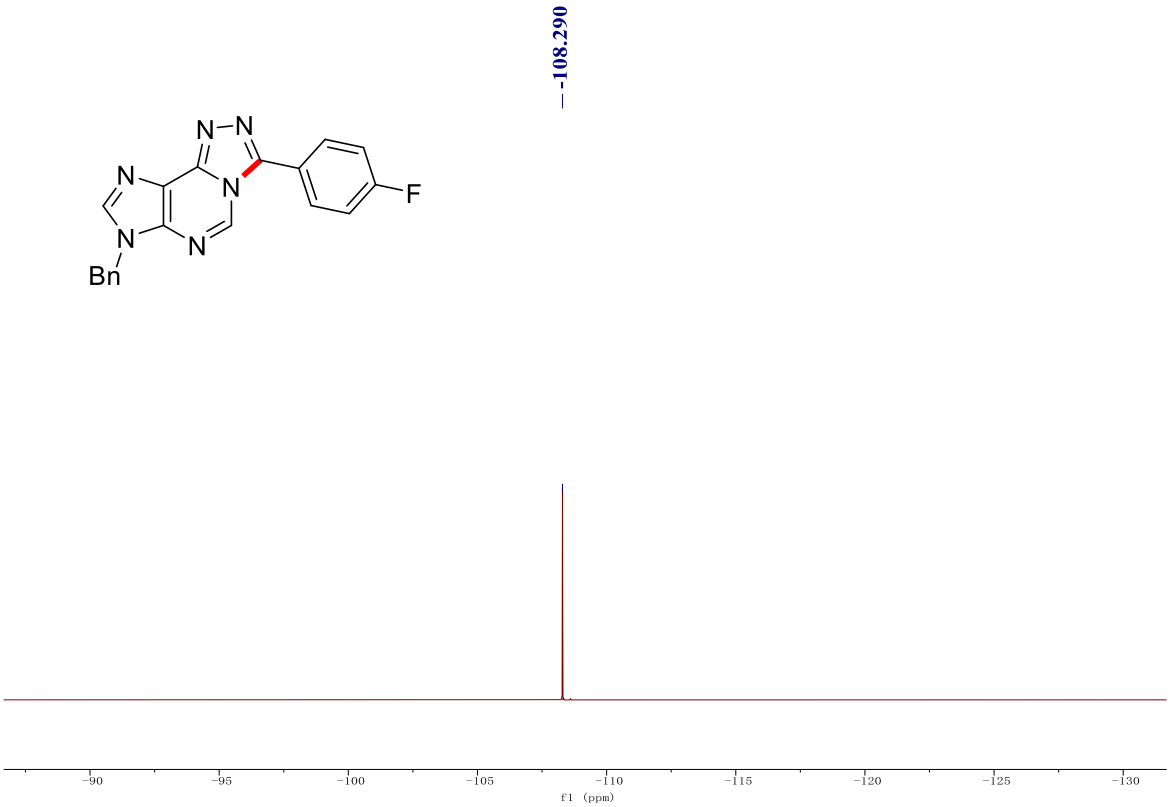
^{1}H NMR Spectrum of 2m at 25 ^{o}C ($CDCl_3$, 400 MHz)



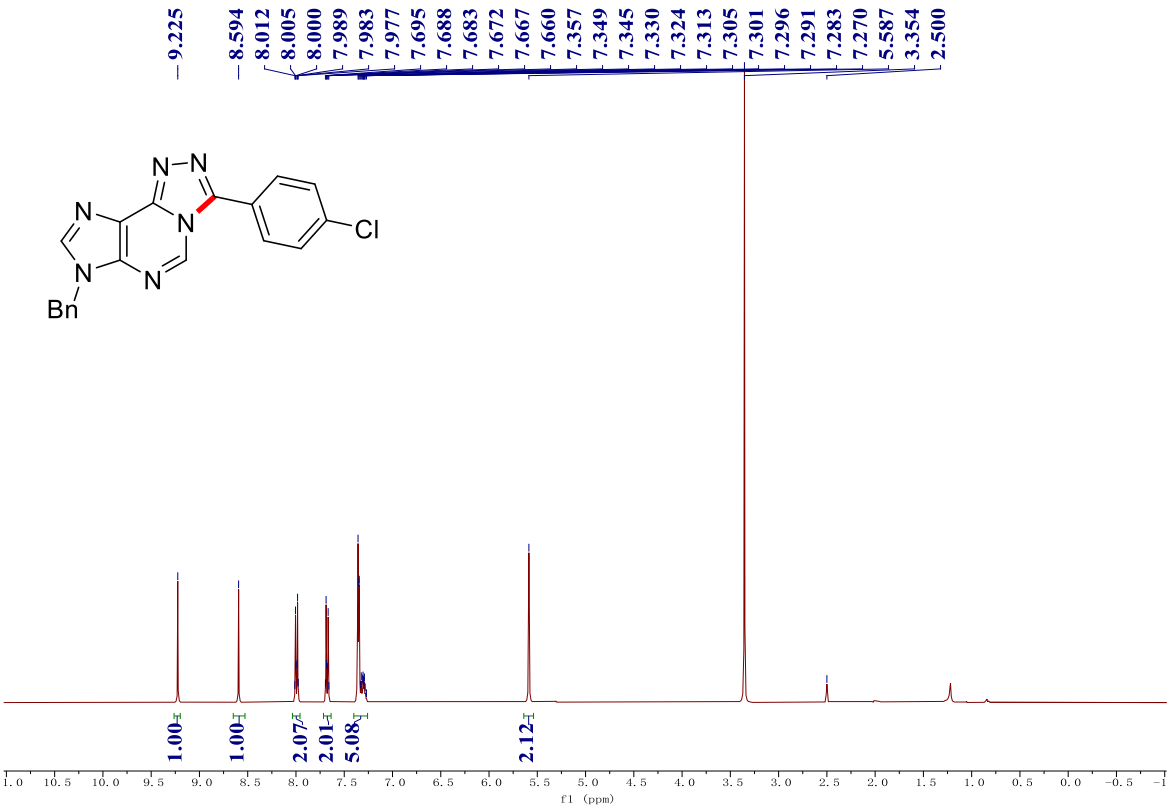
^{13}C NMR Spectrum of 2m at 25 ^{o}C ($CDCl_3$, 100 MHz)



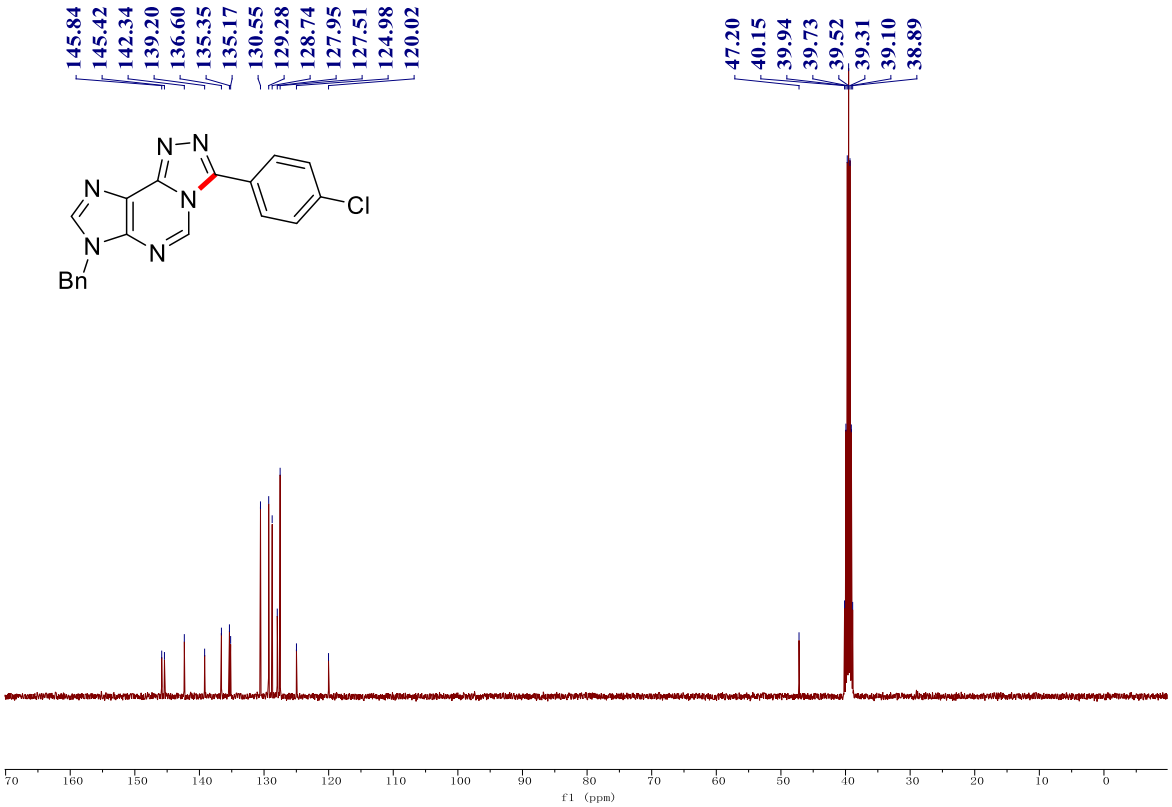
^{13}F NMR Spectrum of 2m at 25 °C ($CDCl_3$, 376 MHz)



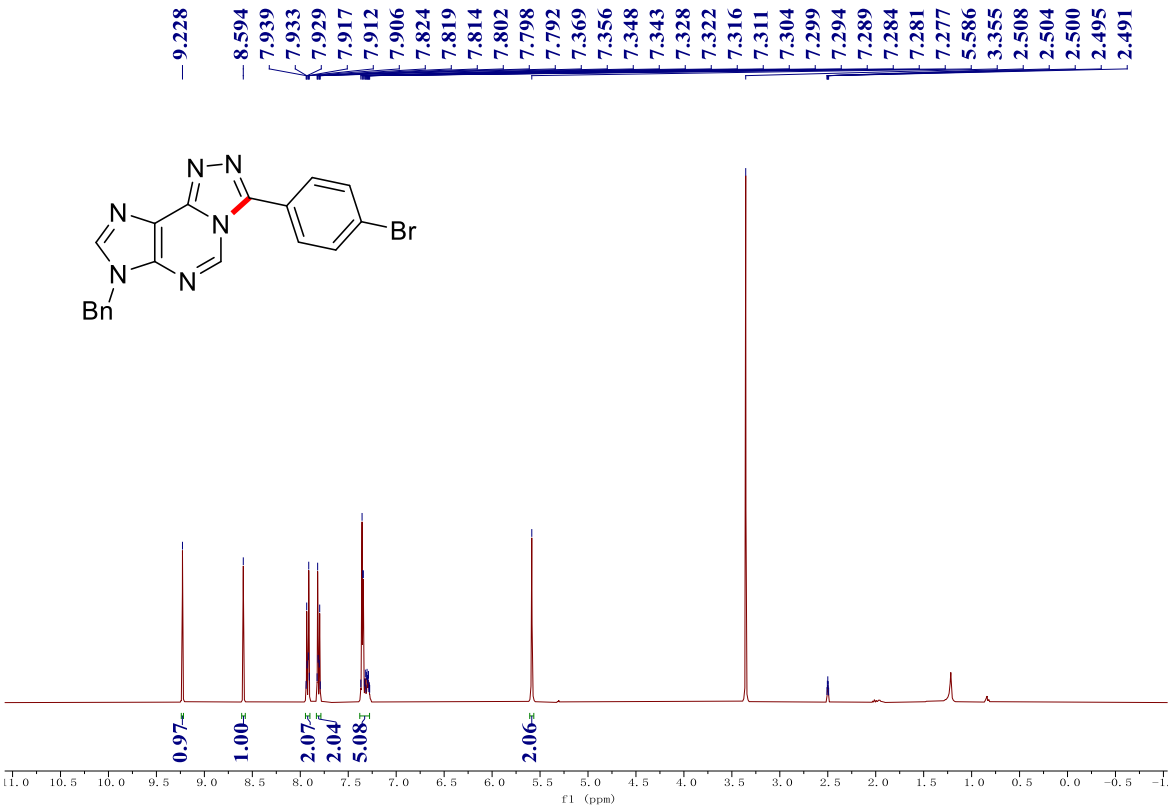
^{1}H NMR Spectrum of 2n at 25 °C (DMSO-d_6, 400 MHz)



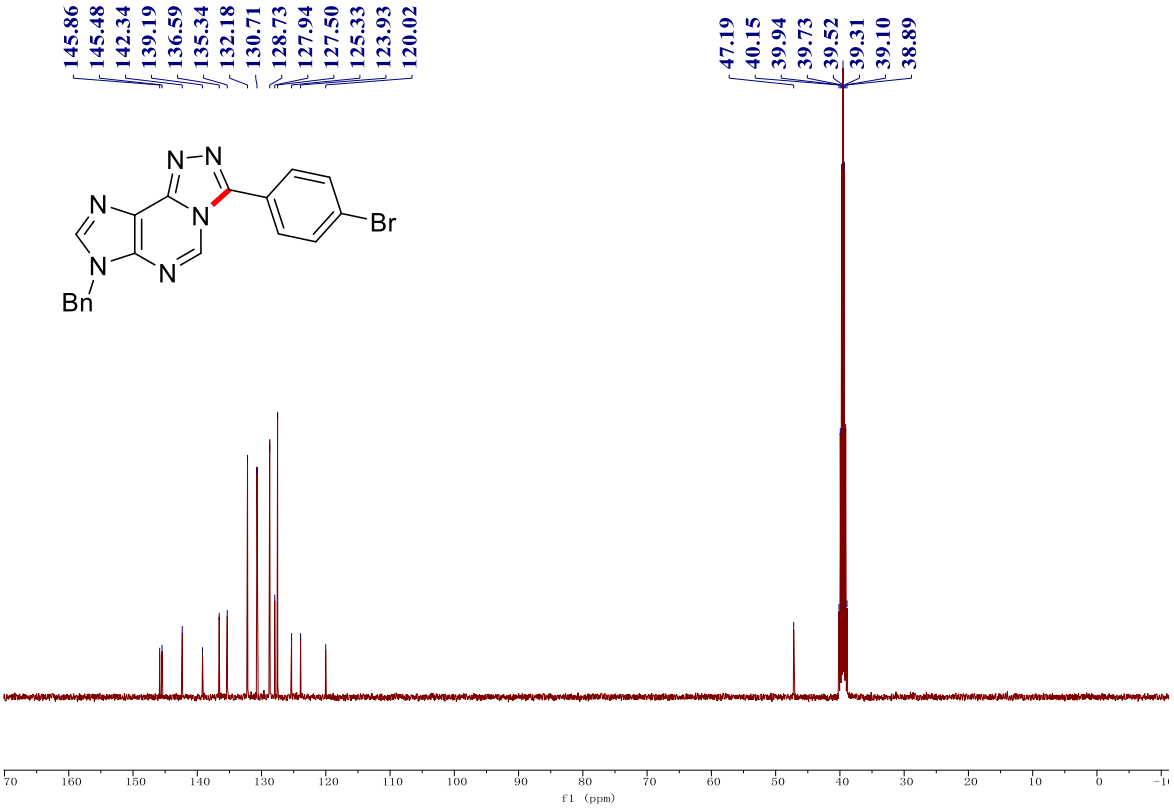
^{13}C NMR Spectrum of 2n at 25 °C (DMSO-d_6, 100 MHz)



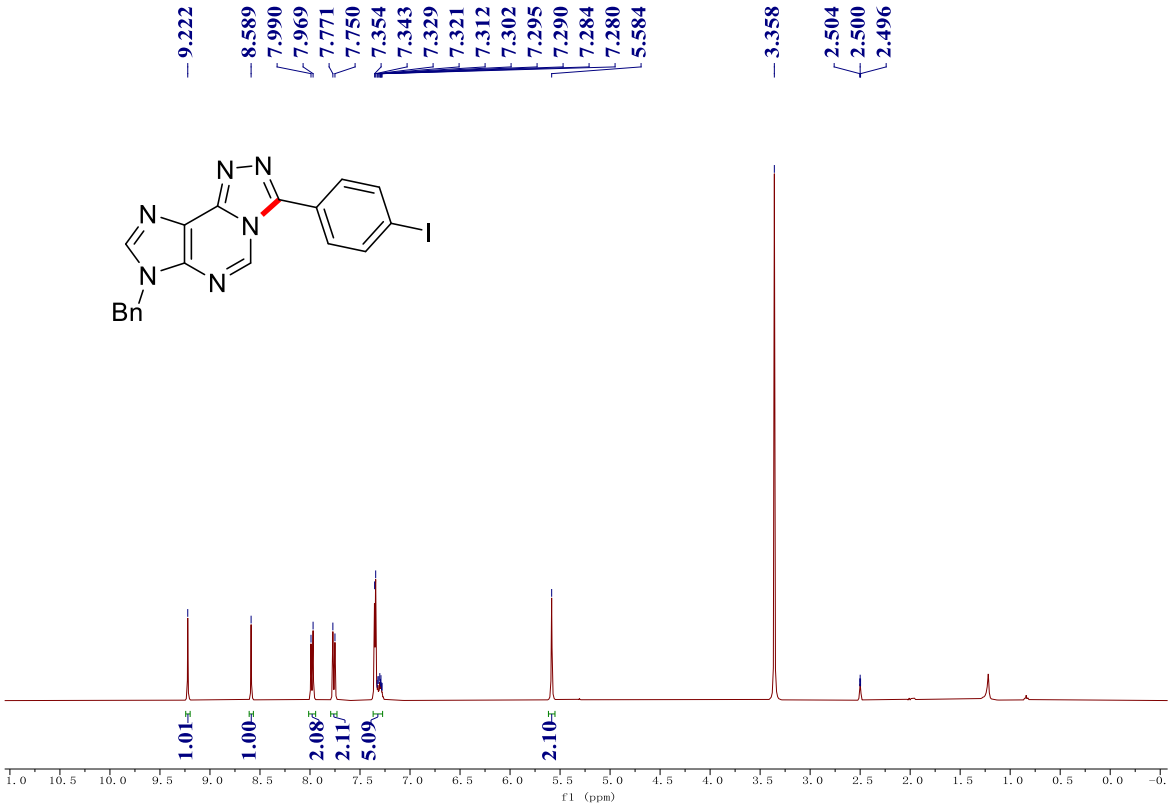
^{1}H NMR Spectrum of 2o at 25 °C (DMSO-d_6, 400 MHz)



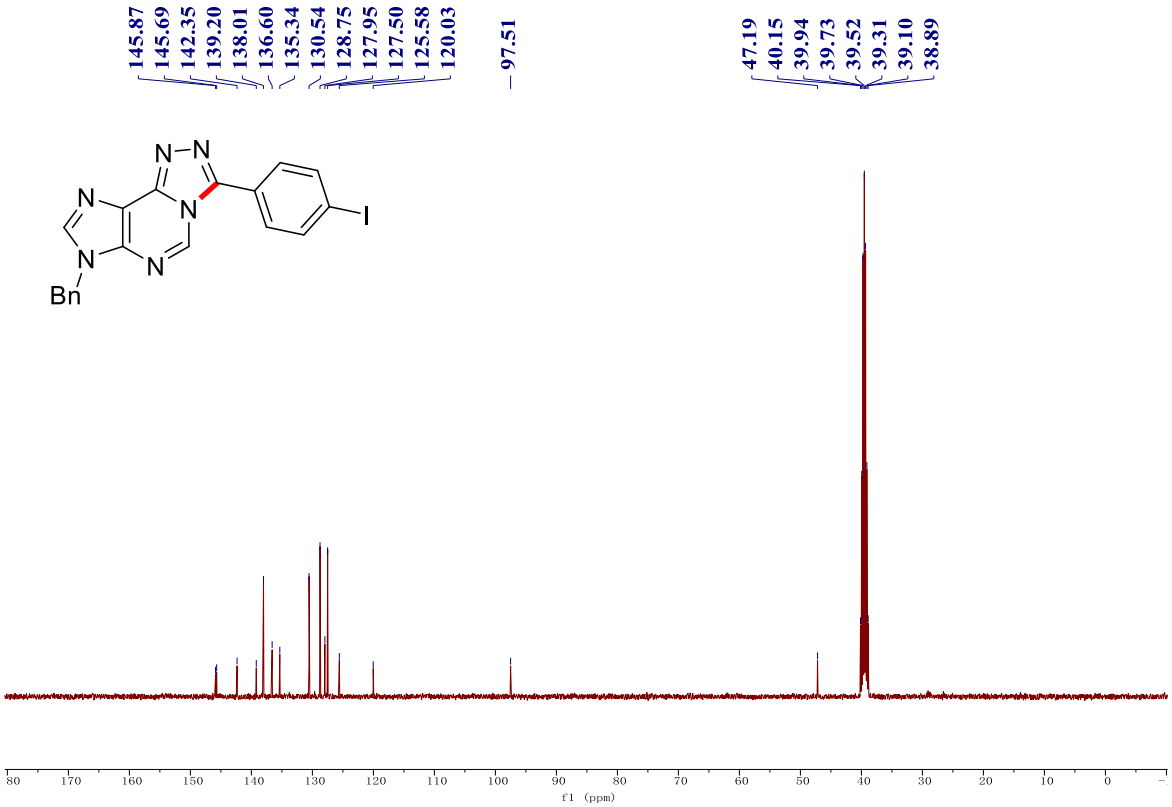
^{13}C NMR Spectrum of 2o at 25 °C (DMSO-d_6, 100 MHz)



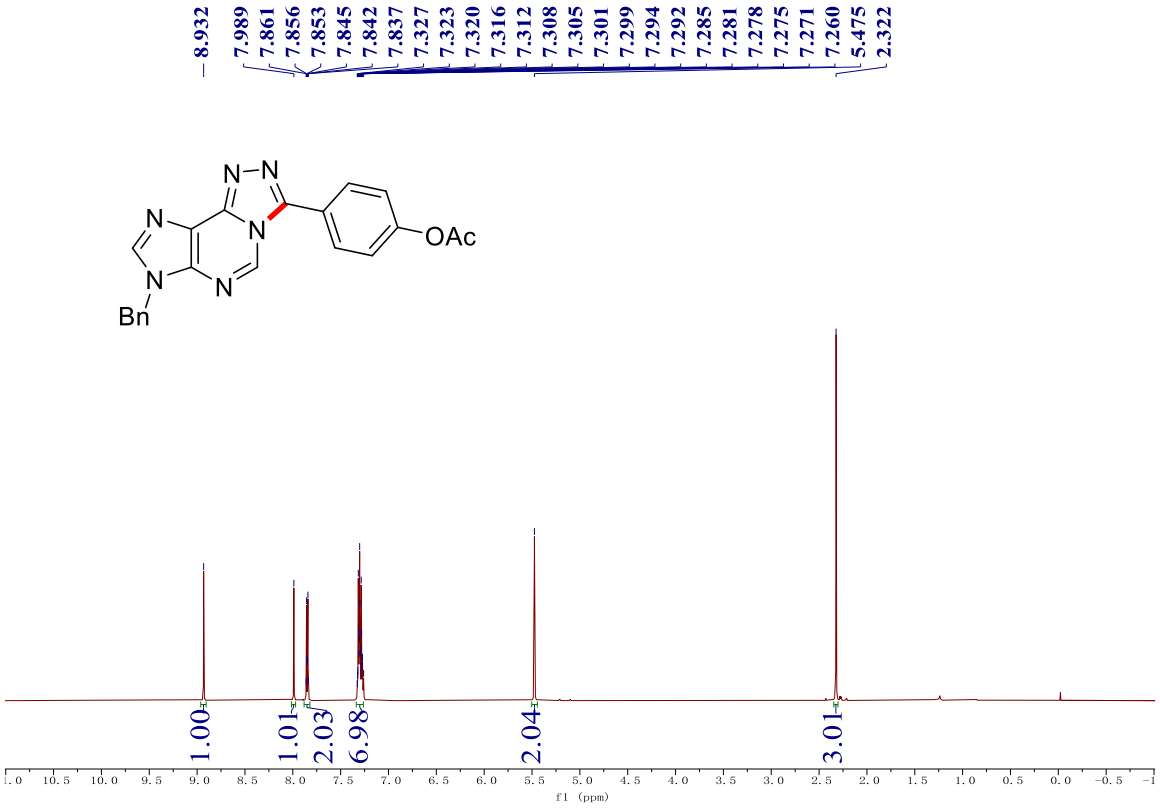
^{1}H NMR Spectrum of 2p at 25 °C (DMSO-d_6, 400 MHz)



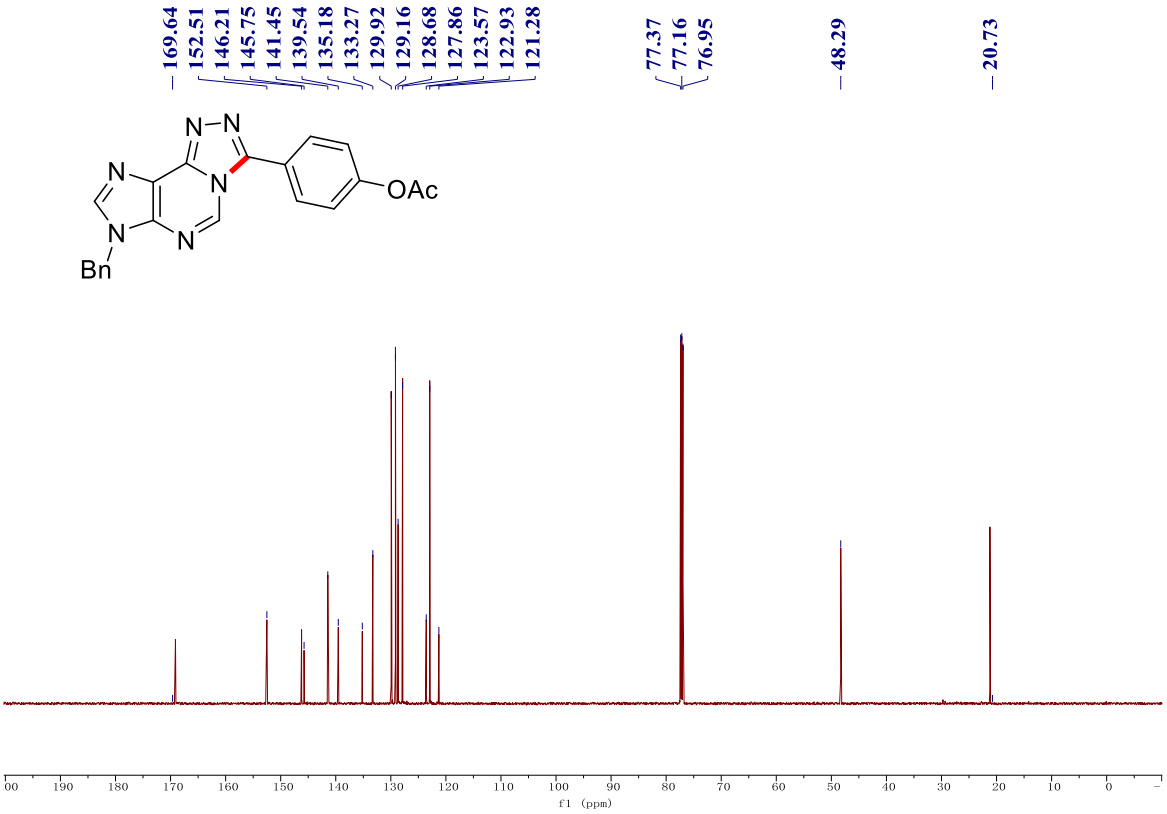
^{13}C NMR Spectrum of 2p at 25 °C (DMSO-d_6, 100 MHz)



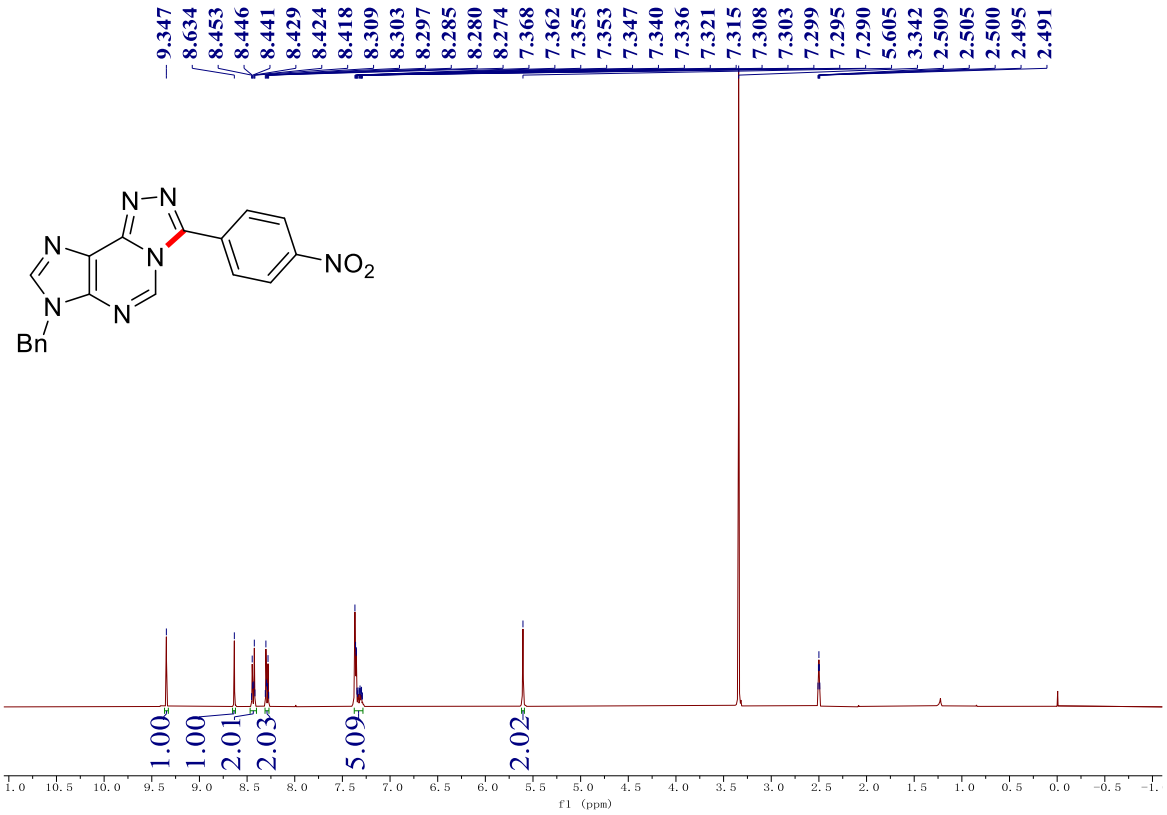
^{1}H NMR Spectrum of 2q at 25 °C ($CDCl_3$, 600 MHz)



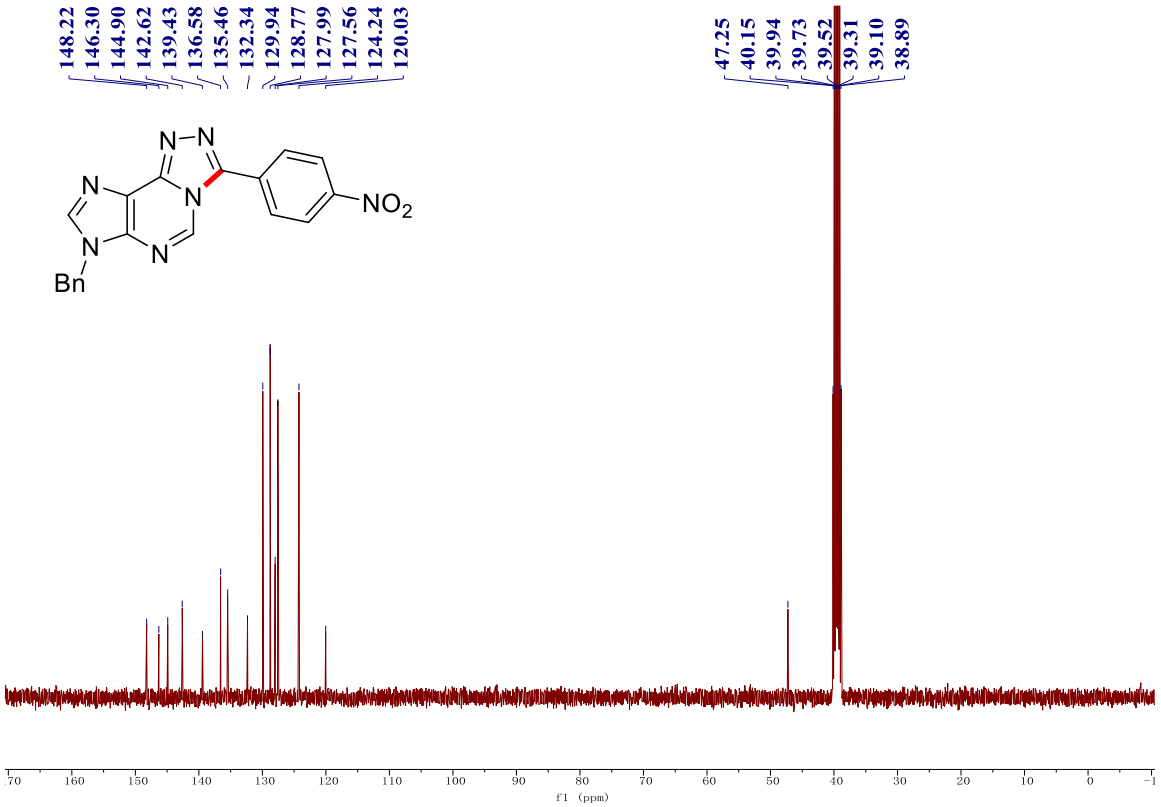
^{13}C NMR Spectrum of 2q at 25 °C ($CDCl_3$, 150 MHz)



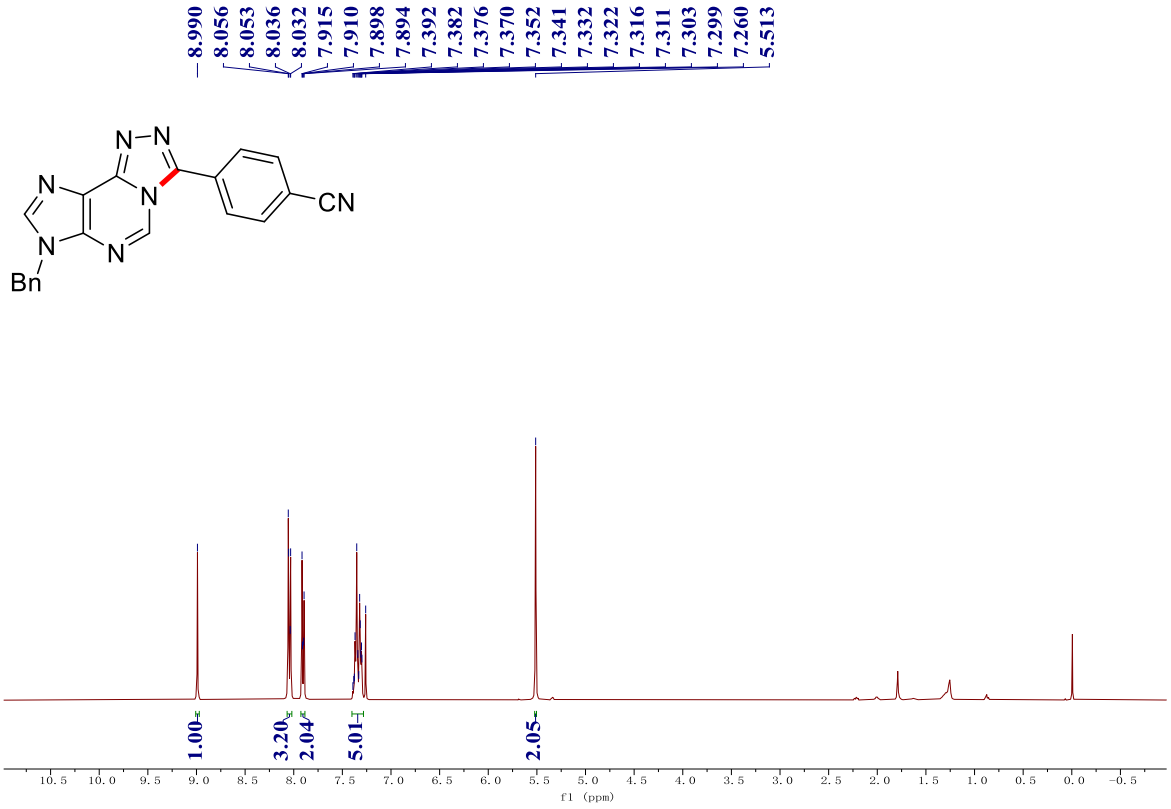
^{1}H NMR Spectrum of 2r at 25 °C (DMSO-d_6, 400 MHz)



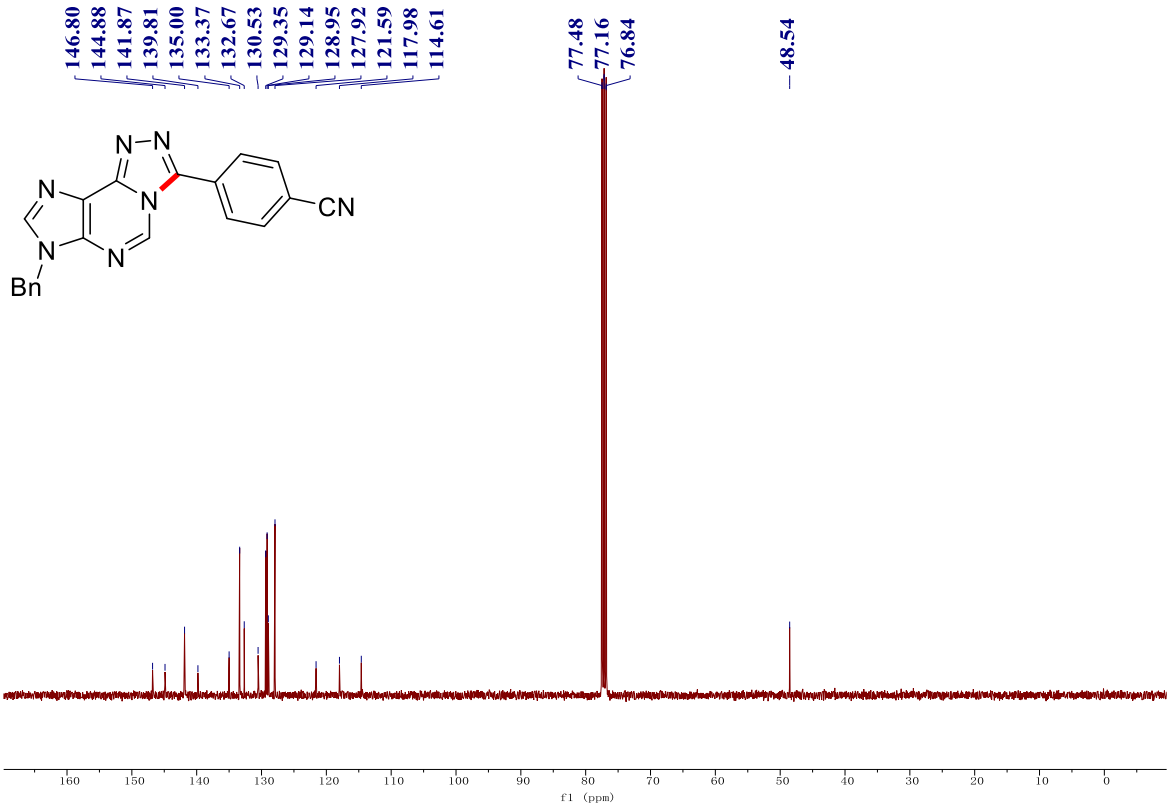
^{13}C NMR Spectrum of 2r at 25 °C (DMSO-d_6, 100 MHz)



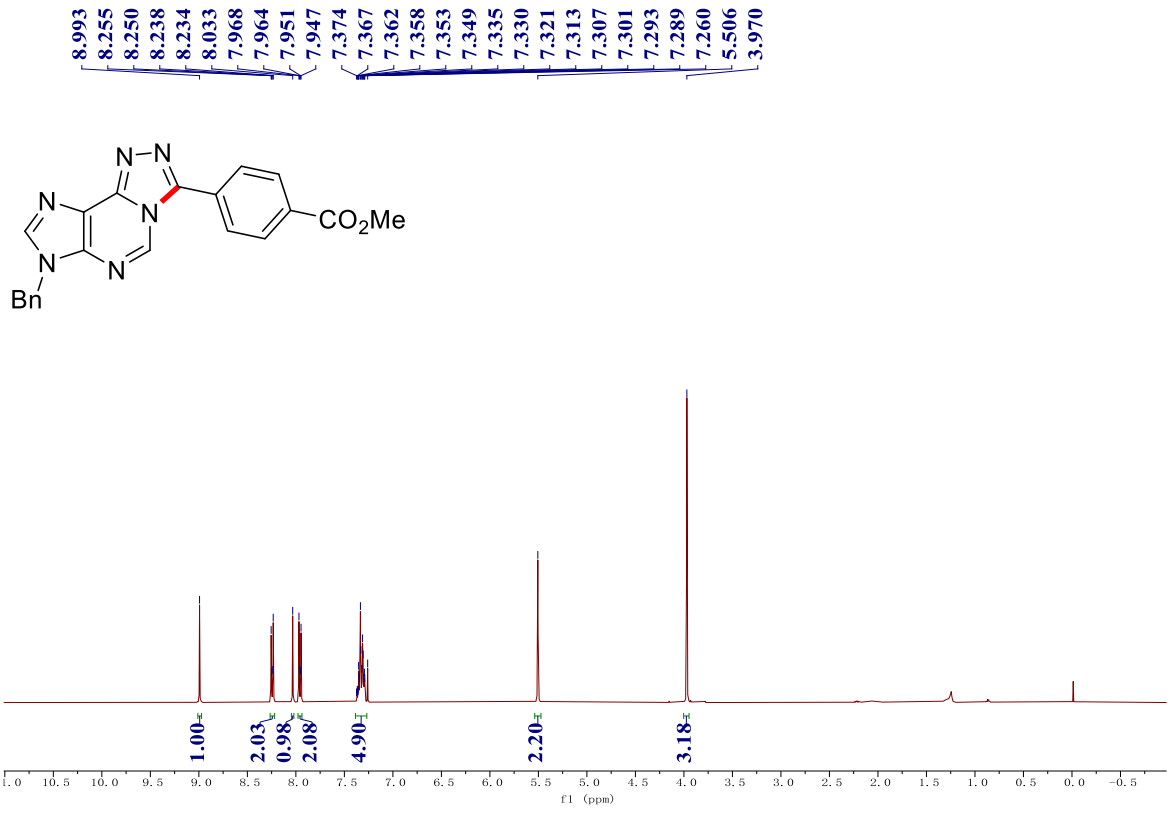
^{1}H NMR Spectrum of 2s at 25 °C ($CDCl_3$, 400 MHz)



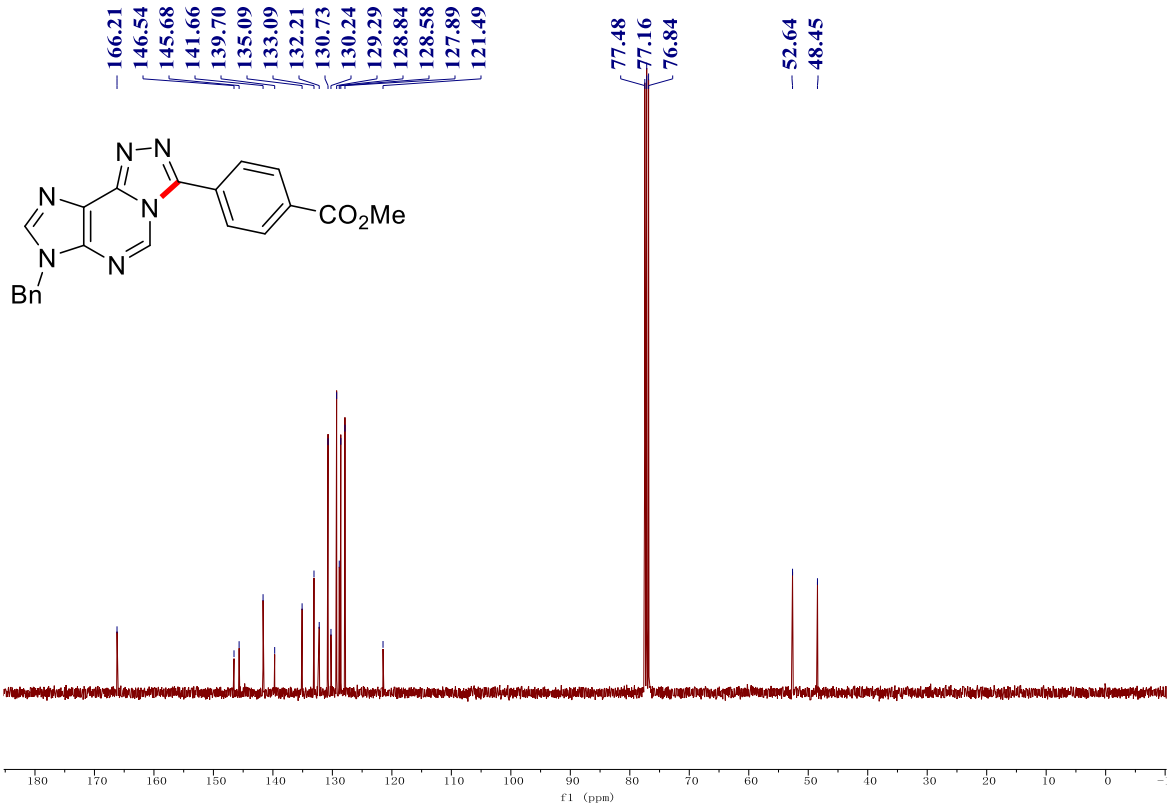
^{13}C NMR Spectrum of 2s at 25 °C ($CDCl_3$, 100 MHz)



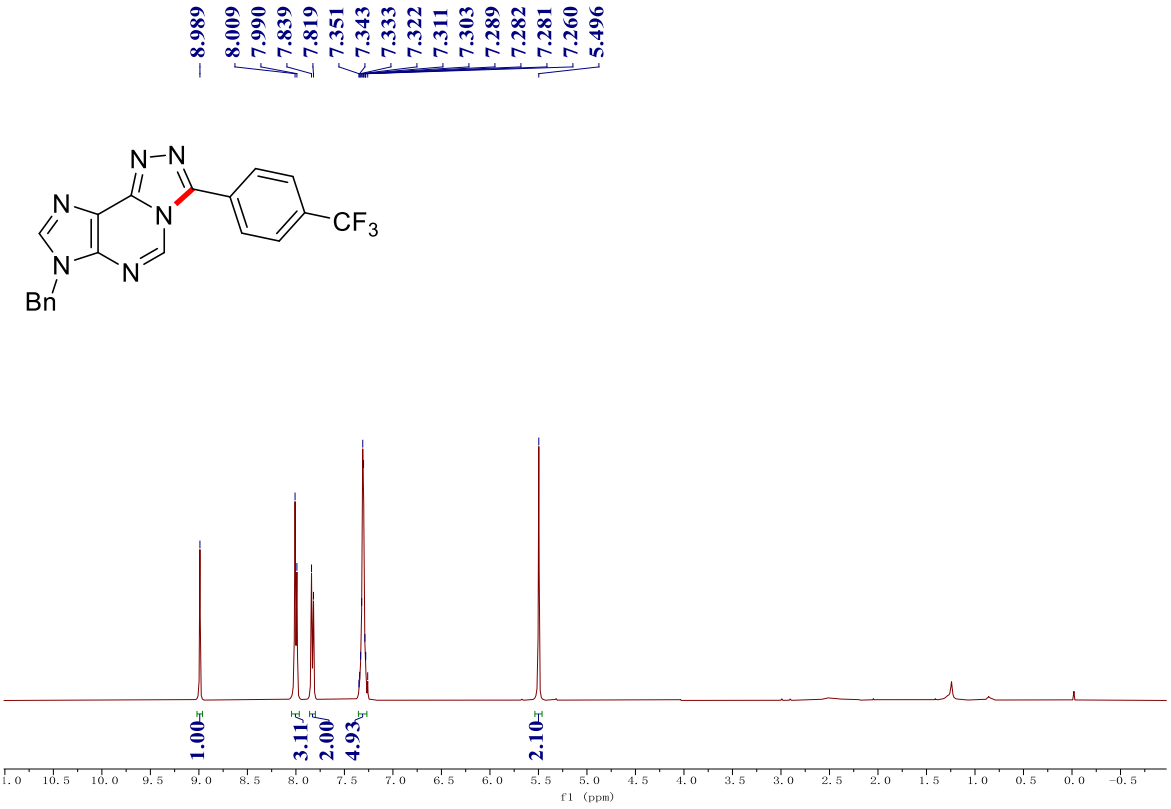
^{1}H NMR Spectrum of 2t at 25 °C ($CDCl_3$, 400 MHz)



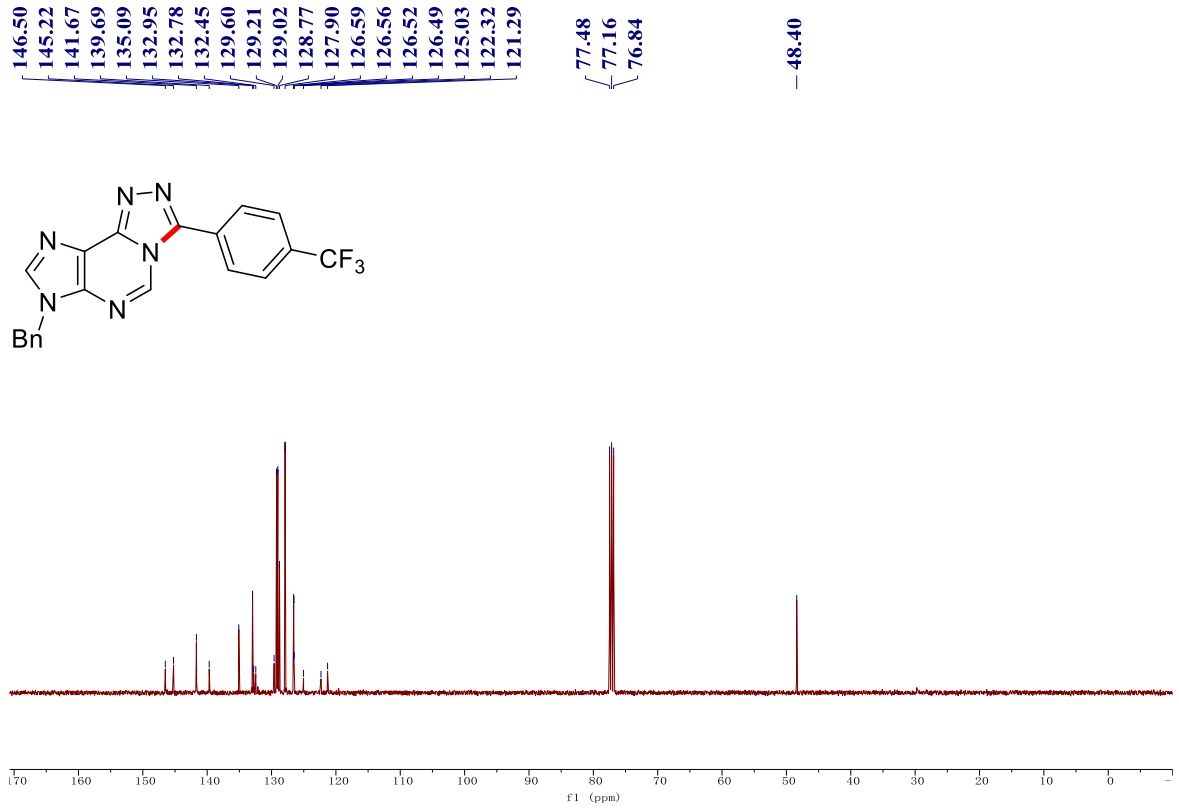
^{13}C NMR Spectrum of 2t at 25 °C ($CDCl_3$, 100 MHz)



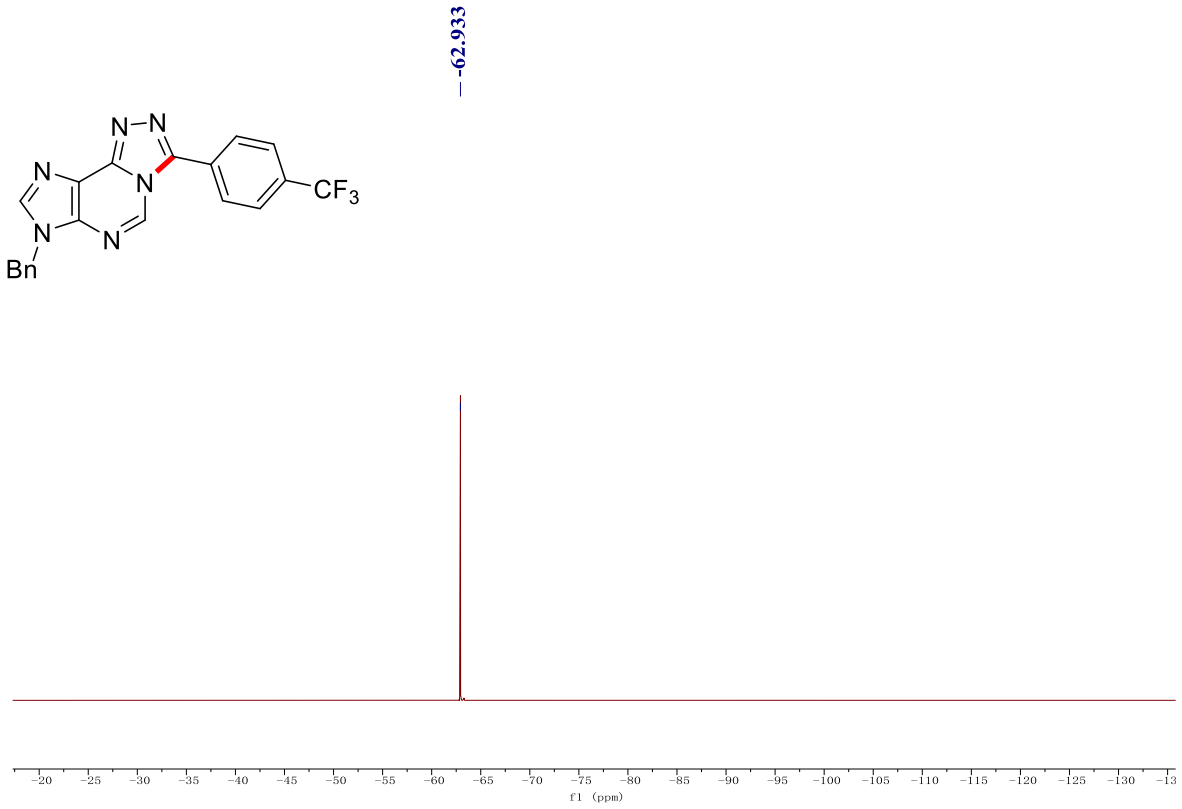
^{1}H NMR Spectrum of 2u at 25 °C ($CDCl_3$, 400 MHz)



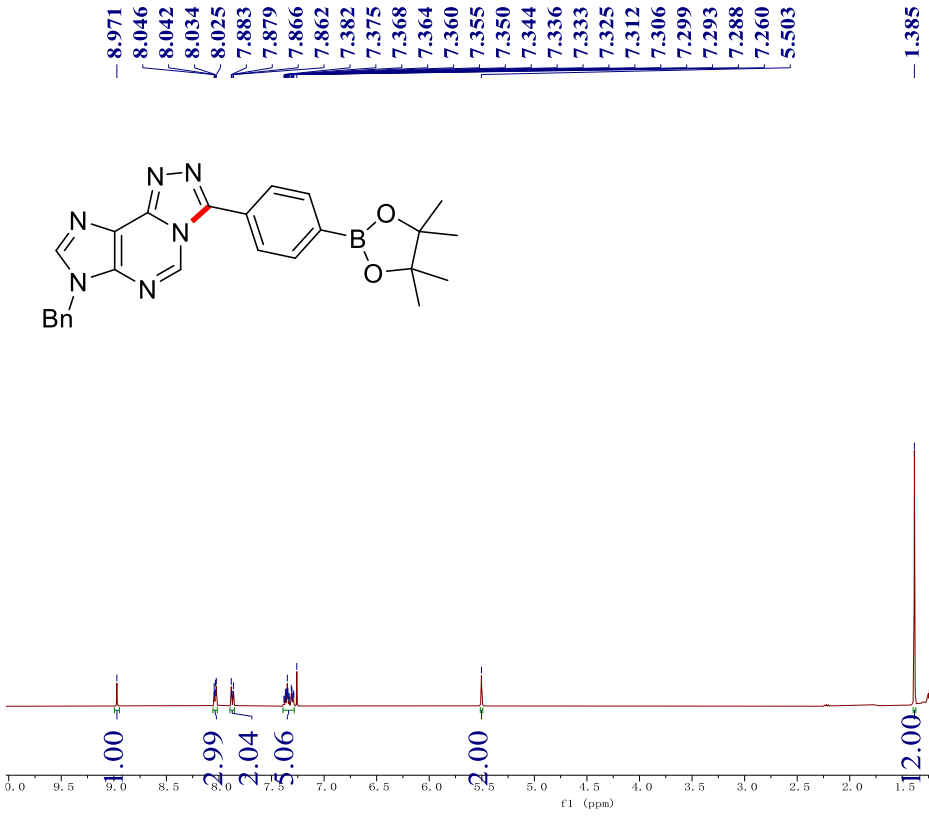
^{13}C NMR Spectrum of 2u at 25 °C ($CDCl_3$, 100 MHz)



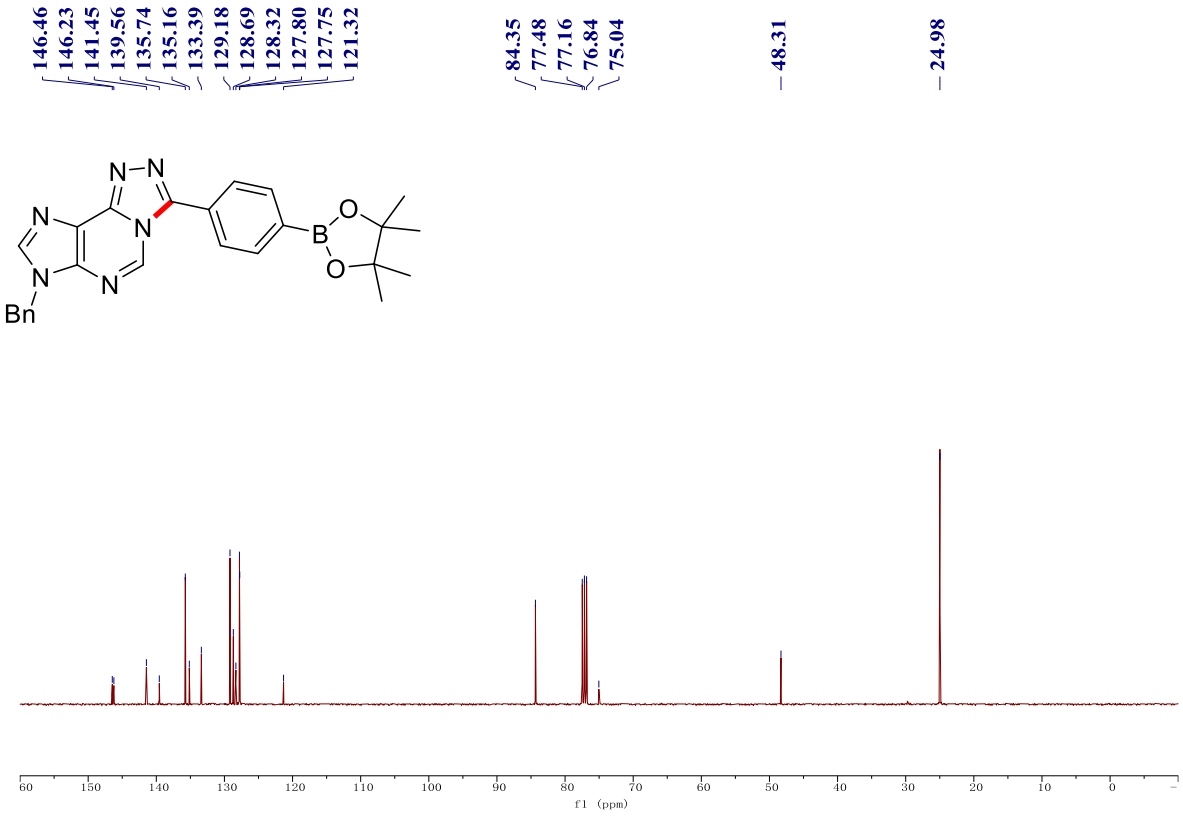
^{19}F NMR Spectrum of 2u at 25 °C ($CDCl_3$, 376 MHz)



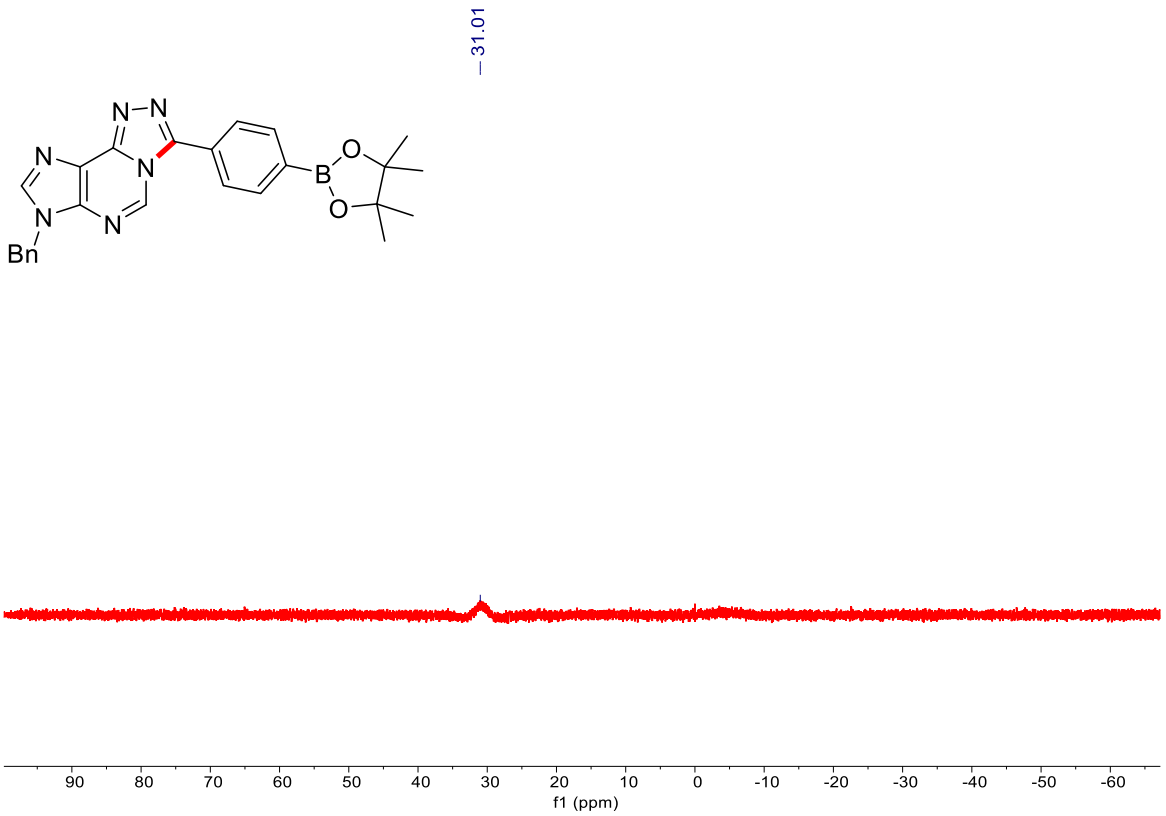
^{1}H NMR Spectrum of 2v at 25 °C ($CDCl_3$, 400 MHz)



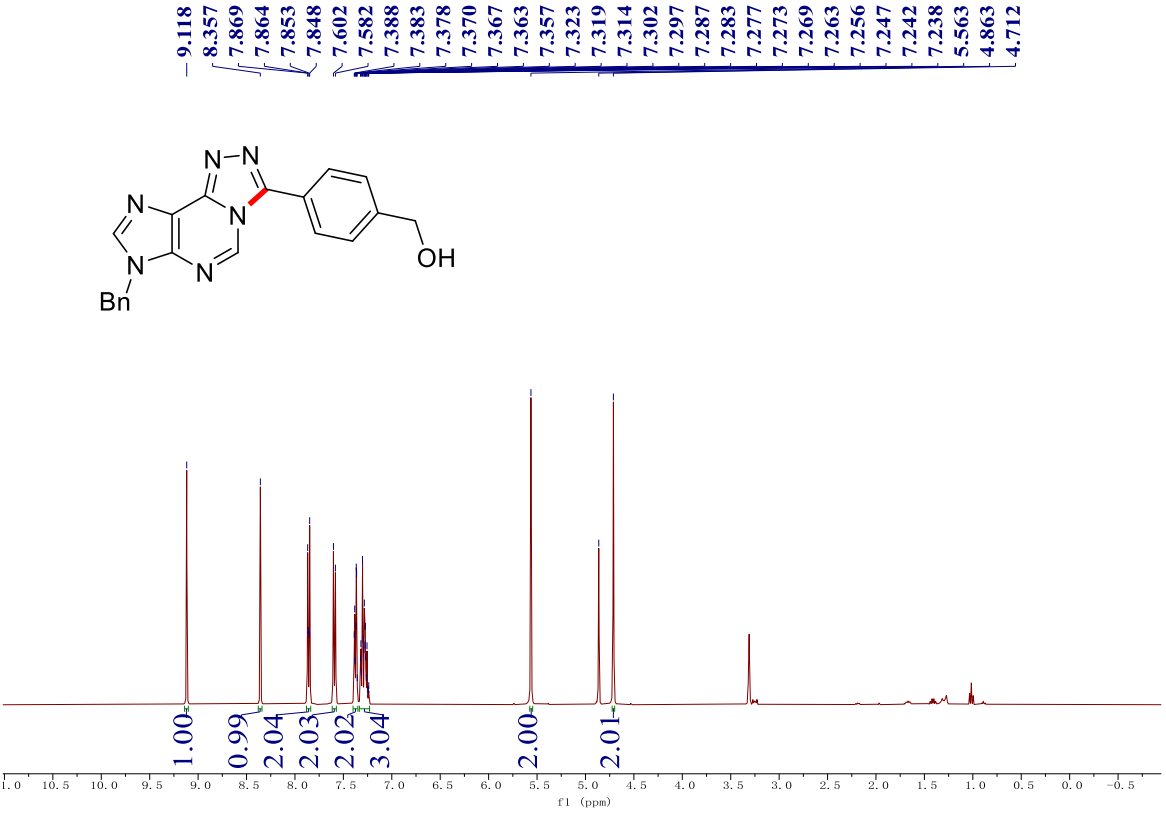
^{13}C NMR Spectrum of 2v at 25 °C ($CDCl_3$, 100 MHz)



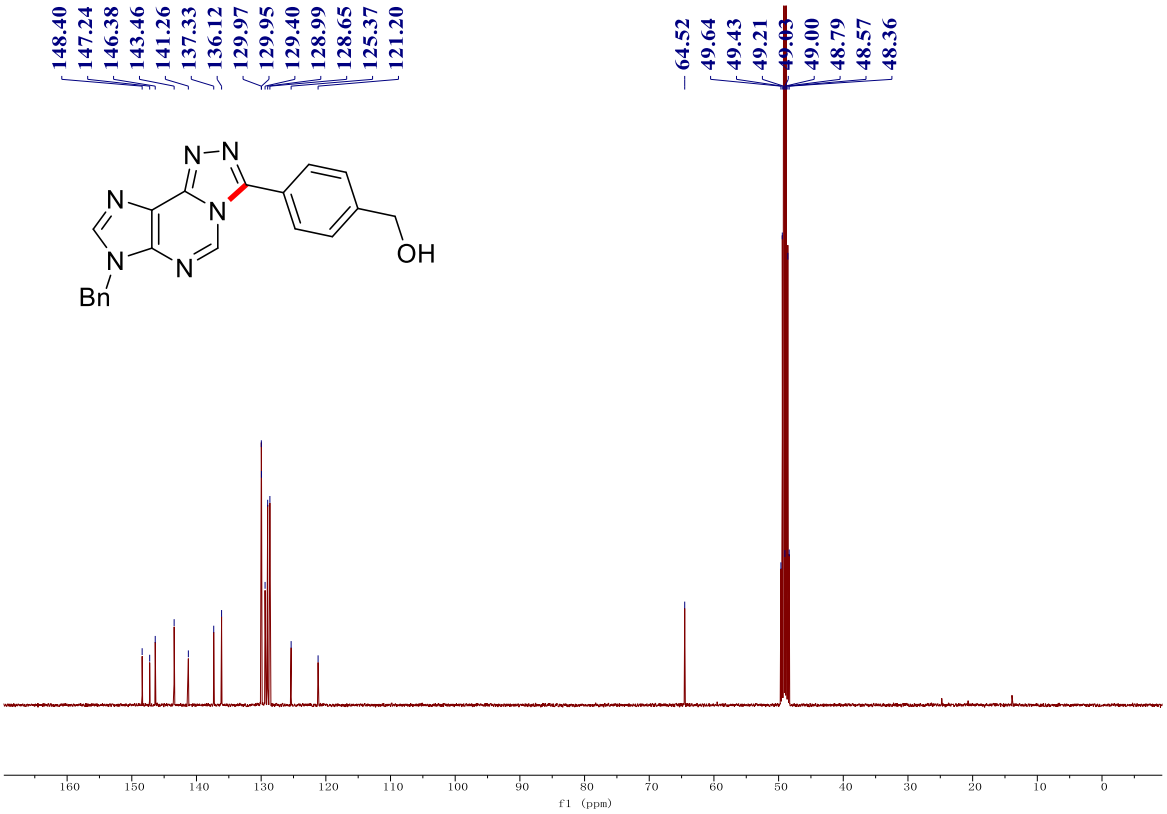
^{1}B NMR Spectrum of 2v at 25 °C ($CDCl_3$, 193 MHz)



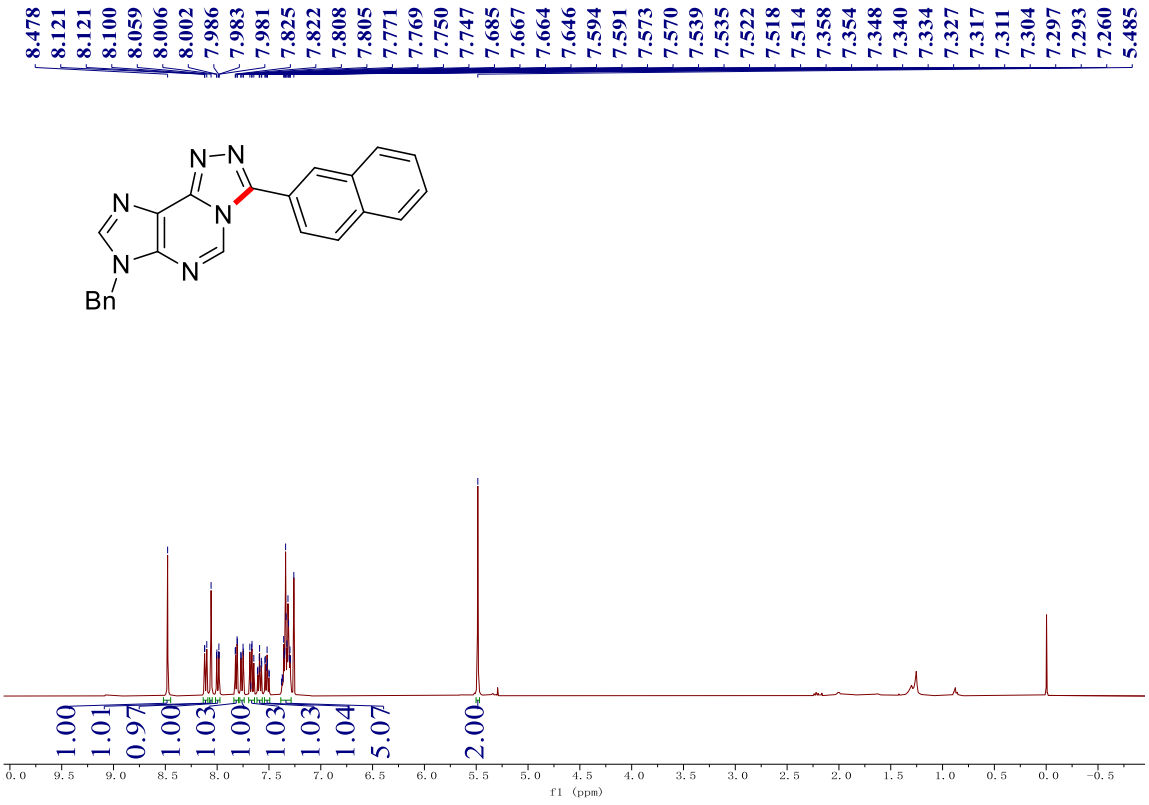
^{1}H NMR Spectrum of 2w at 25 °C (CD_3OD, 400 MHz)



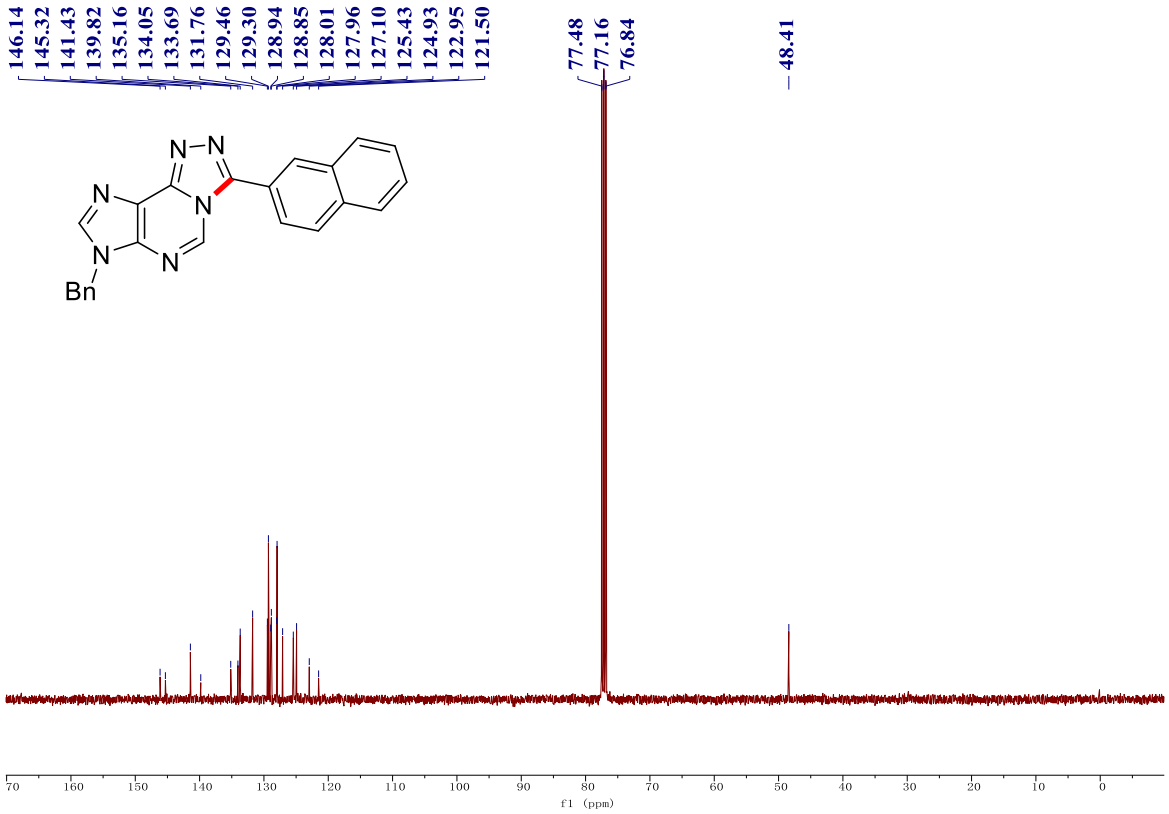
^{13}C NMR Spectrum of 2w at 25 °C (CD_3OD, 100 MHz)



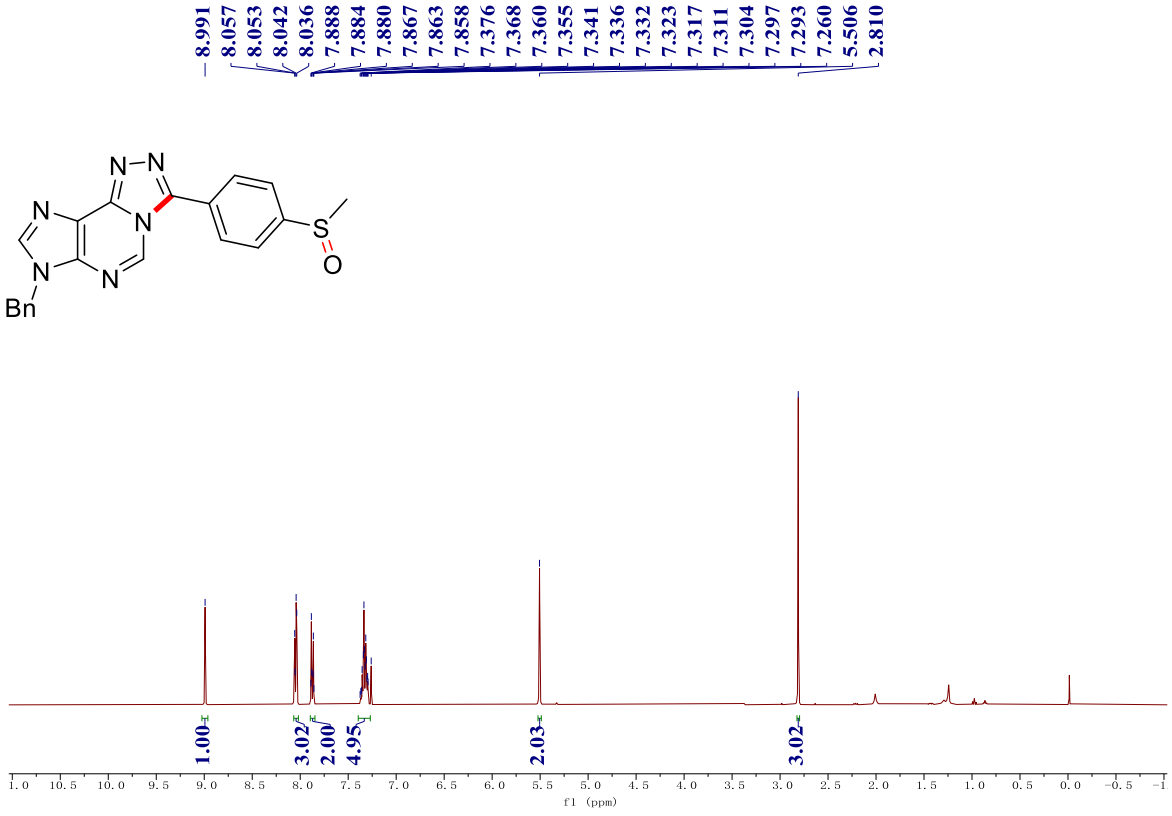
^{1}H NMR Spectrum of 2x at 25 °C ($CDCl_3$, 400 MHz)



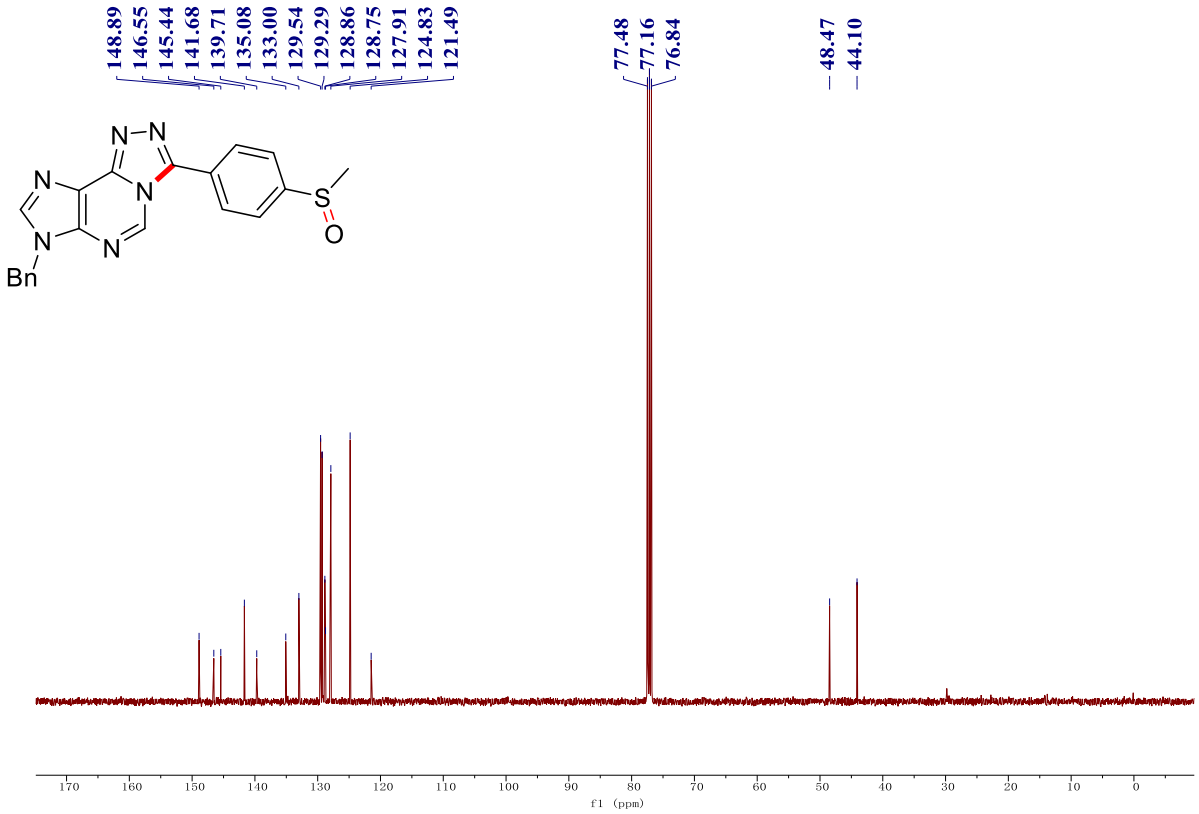
^{13}C NMR Spectrum of 2x at 25 °C ($CDCl_3$, 100 MHz)



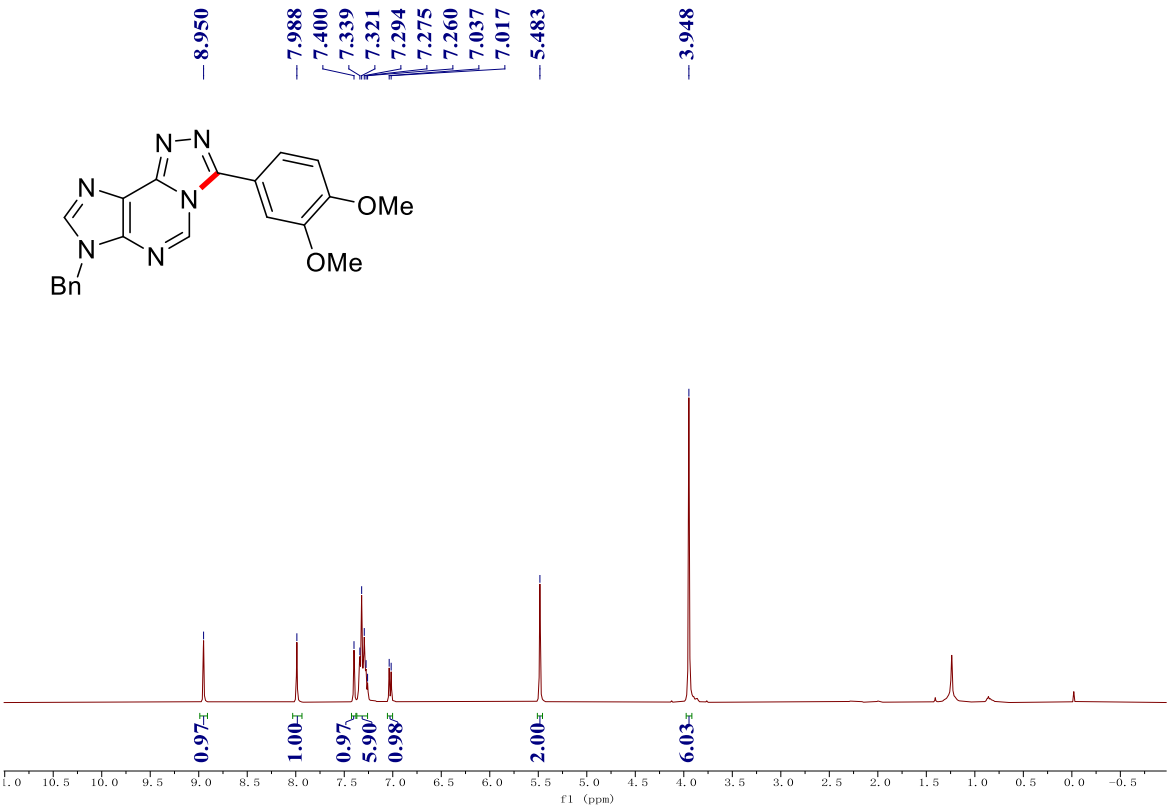
^{1}H NMR Spectrum of 2y at 25 °C ($CDCl_3$, 400 MHz)



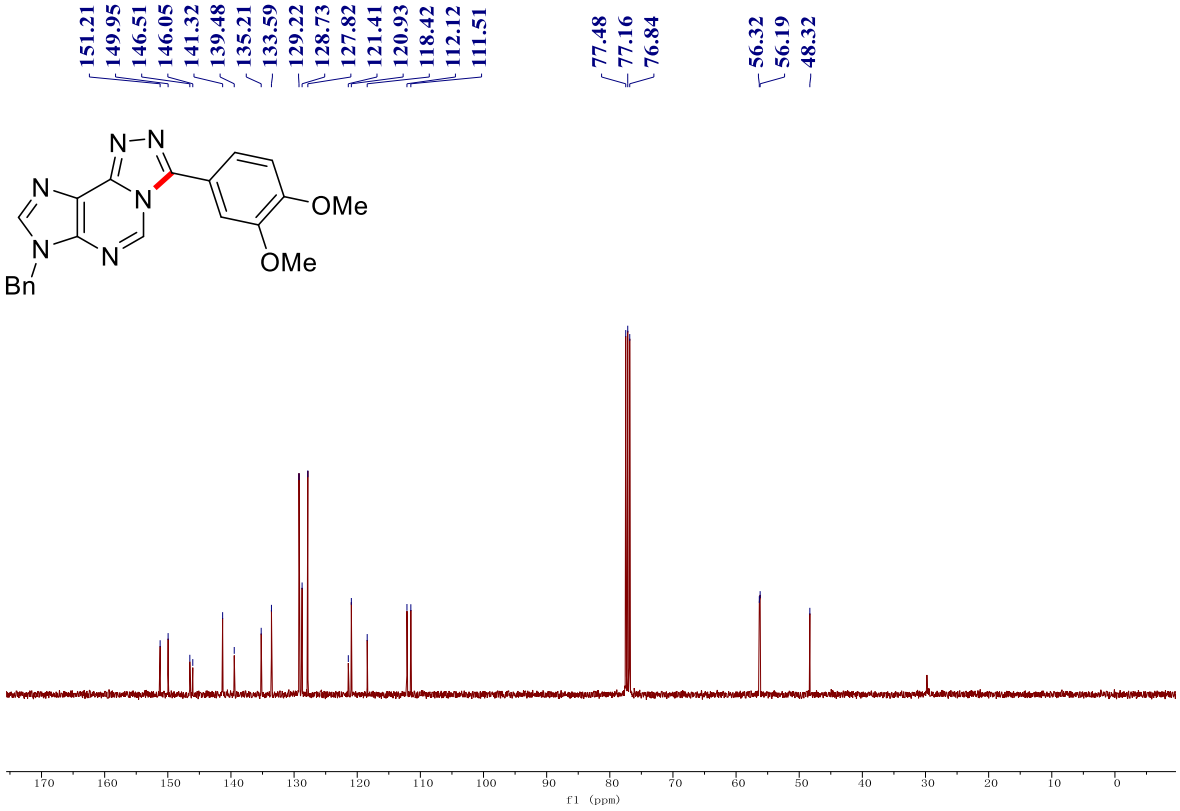
^{13}C NMR Spectrum of 2y at 25 °C ($CDCl_3$, 100 MHz)



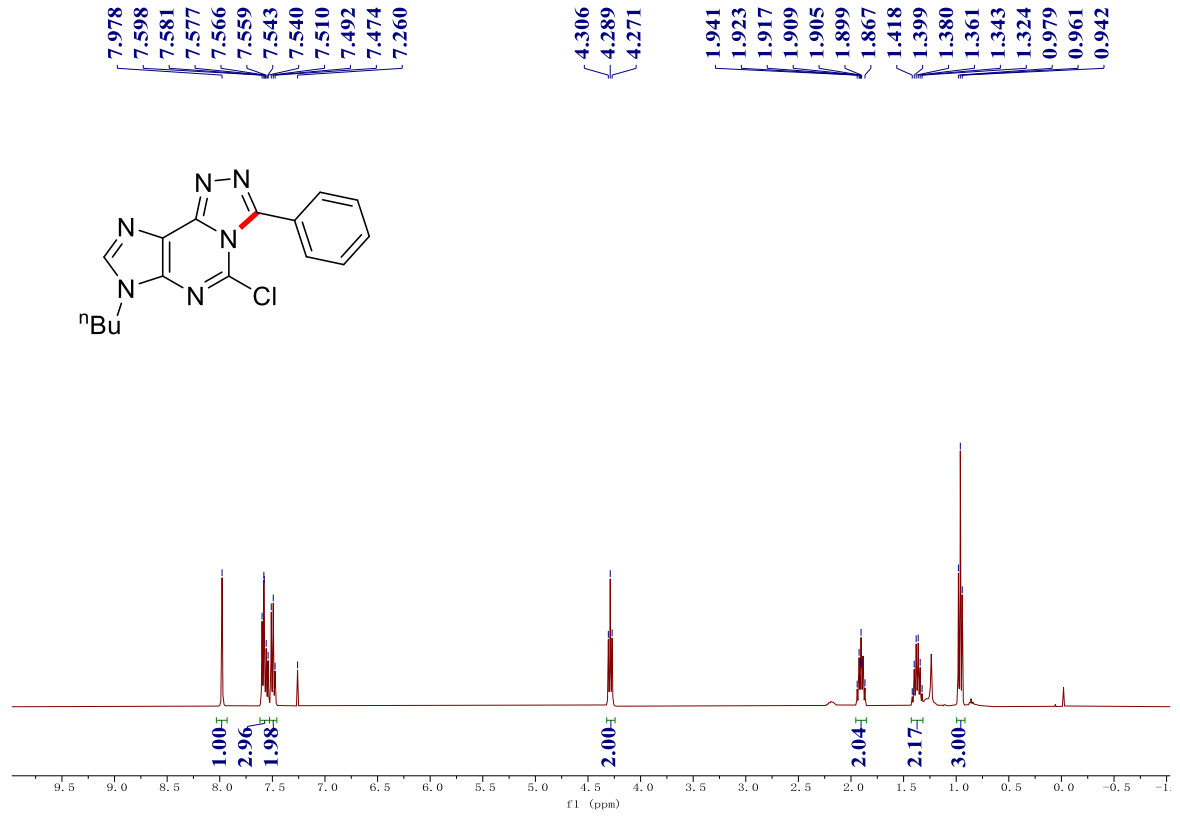
^{1}H NMR Spectrum of 2z at 25 °C ($CDCl_3$, 400 MHz)



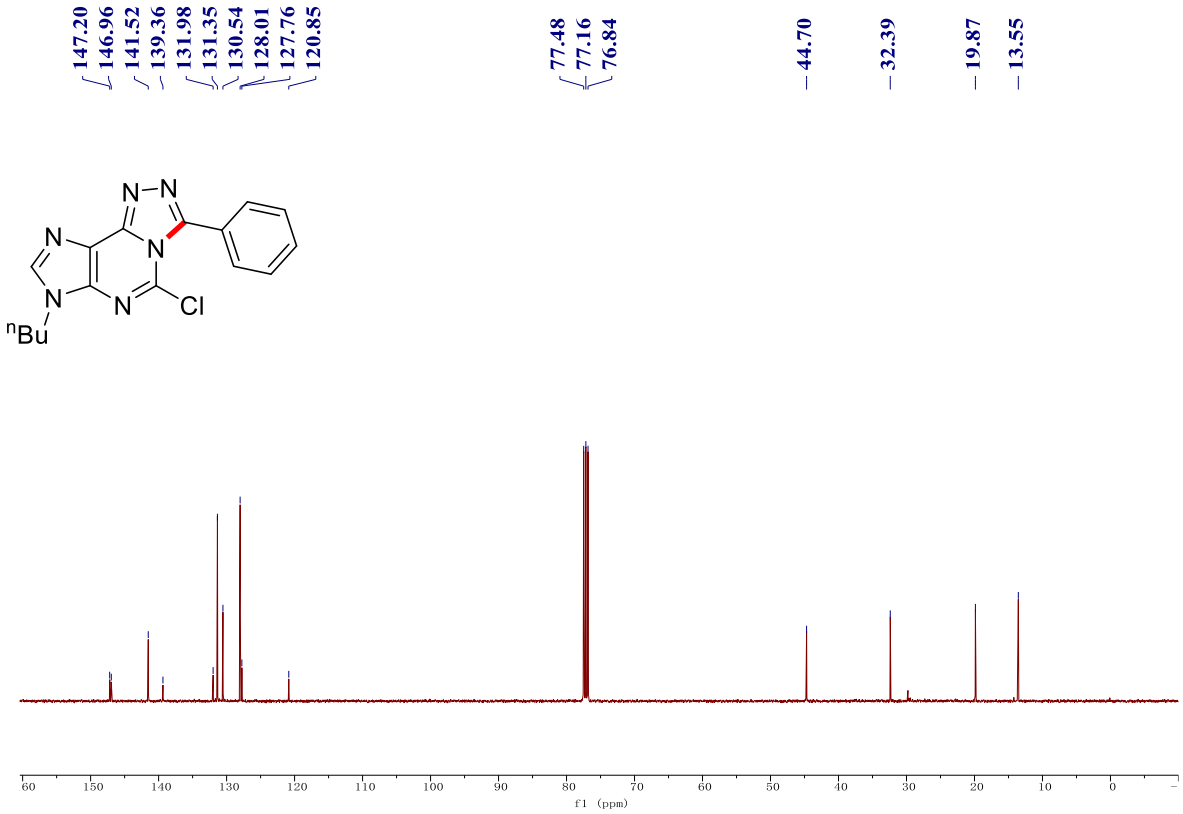
^{13}C NMR Spectrum of 2z at 25 °C ($CDCl_3$, 100 MHz)



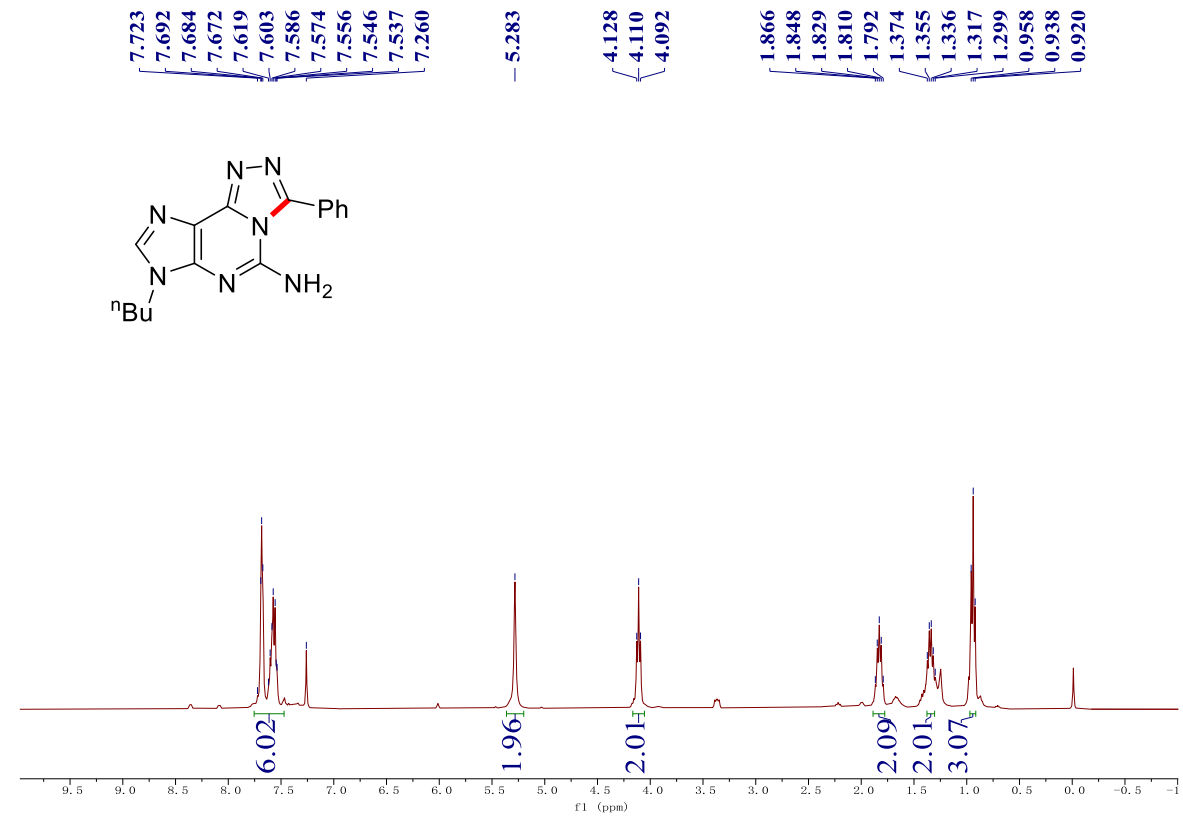
^{1}H NMR Spectrum of 2aa at 25 °C ($CDCl_3$, 400 MHz)



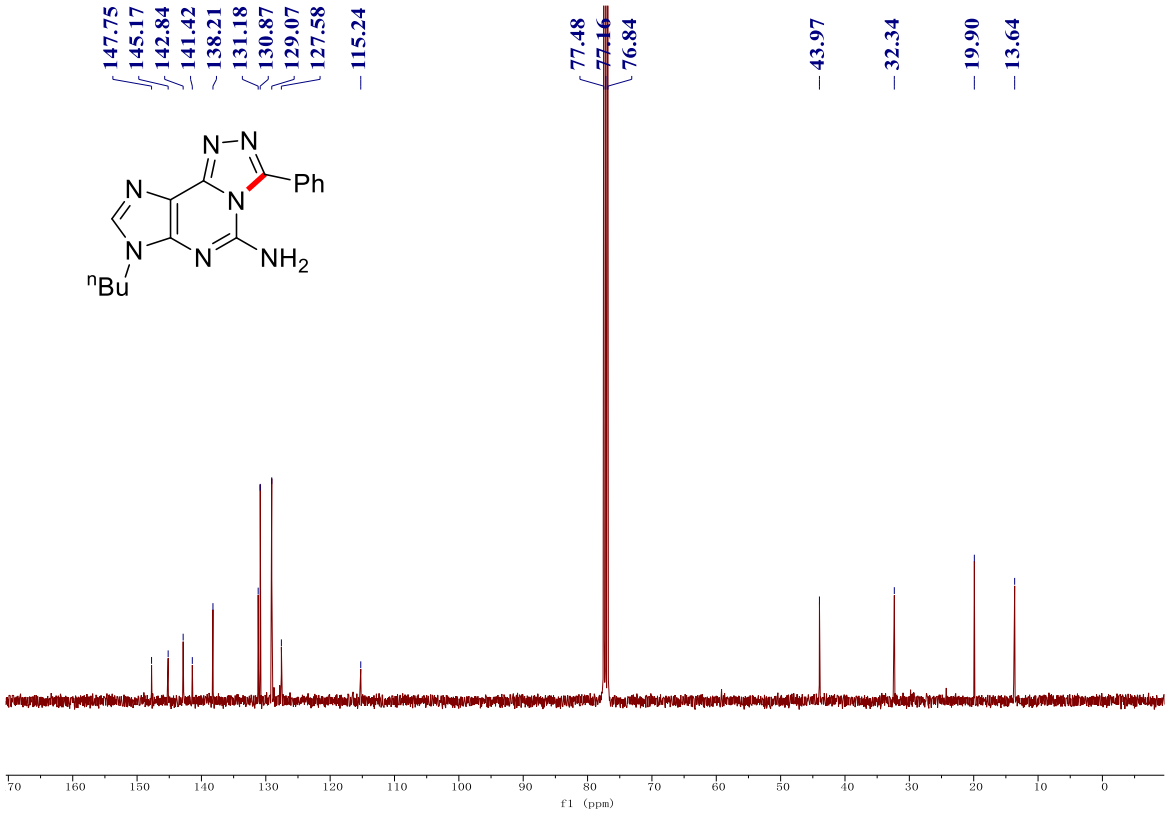
^{13}C NMR Spectrum of 2aa at 25 °C ($CDCl_3$, 100 MHz)



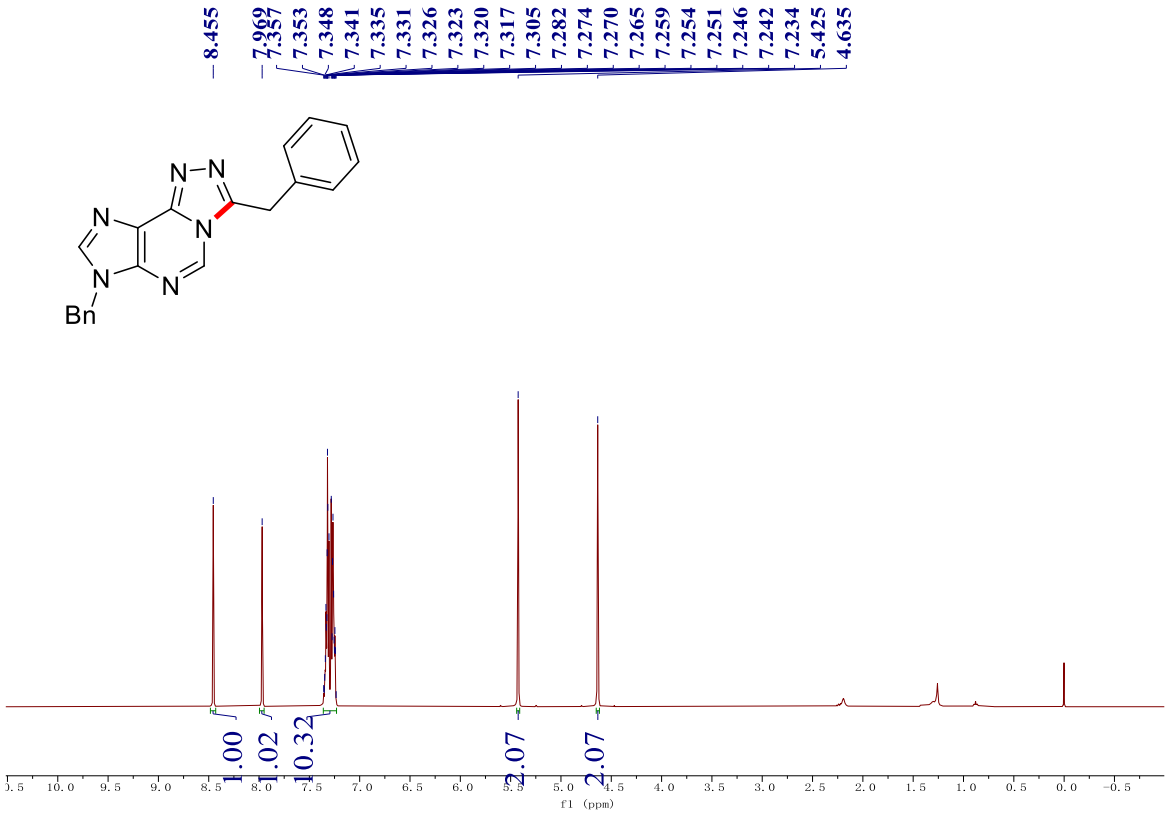
^{1}H NMR Spectrum of 2ab at 25 °C ($CDCl_3$, 400 MHz)



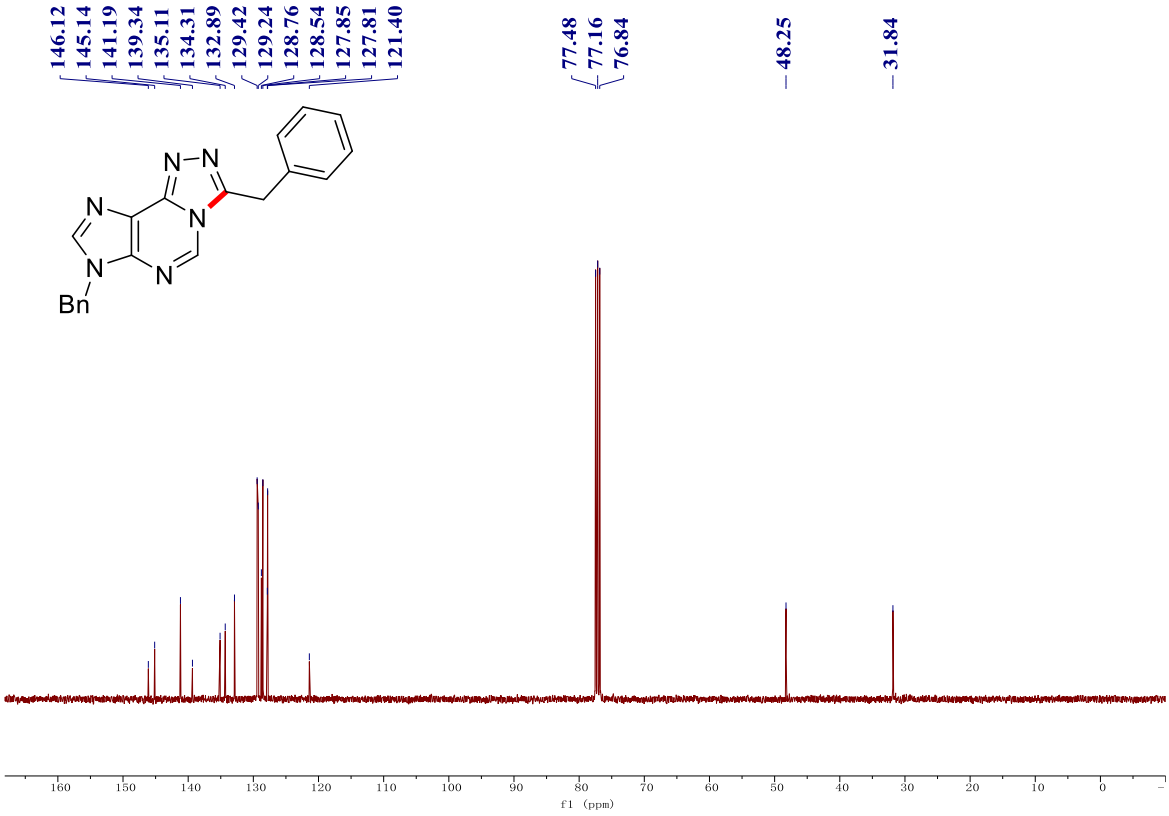
^{13}C NMR Spectrum of 2ab at 25 °C ($CDCl_3$, 100 MHz)



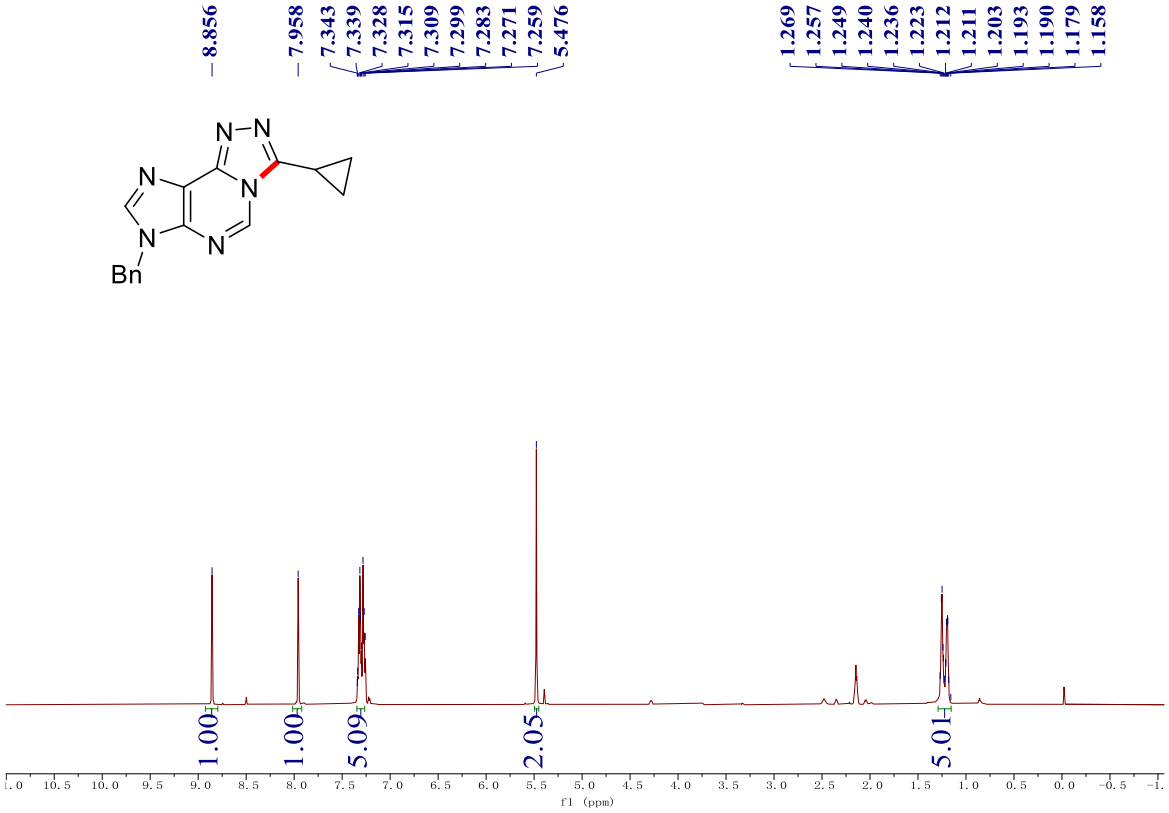
^{1}H NMR Spectrum of 2ac at 25 °C ($CDCl_3$, 400 MHz)



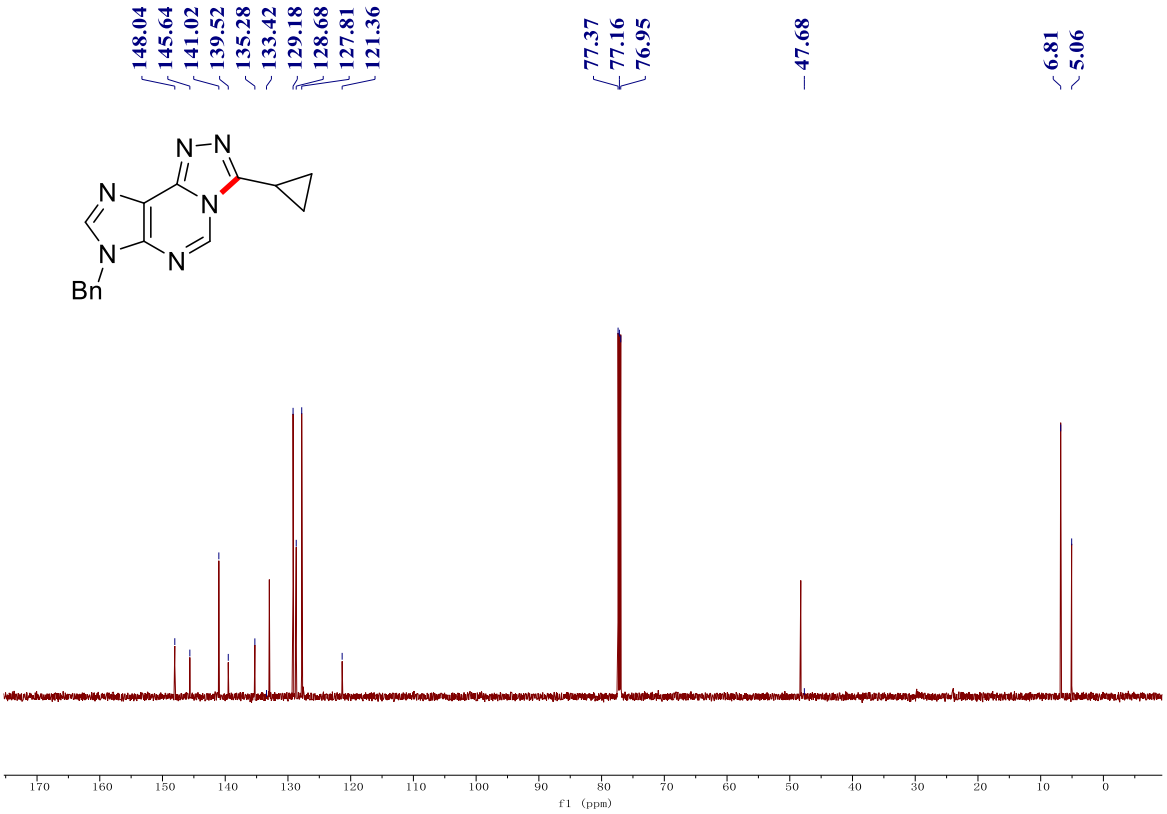
^{13}C NMR Spectrum of 2ac at 25 °C ($CDCl_3$, 100 MHz)



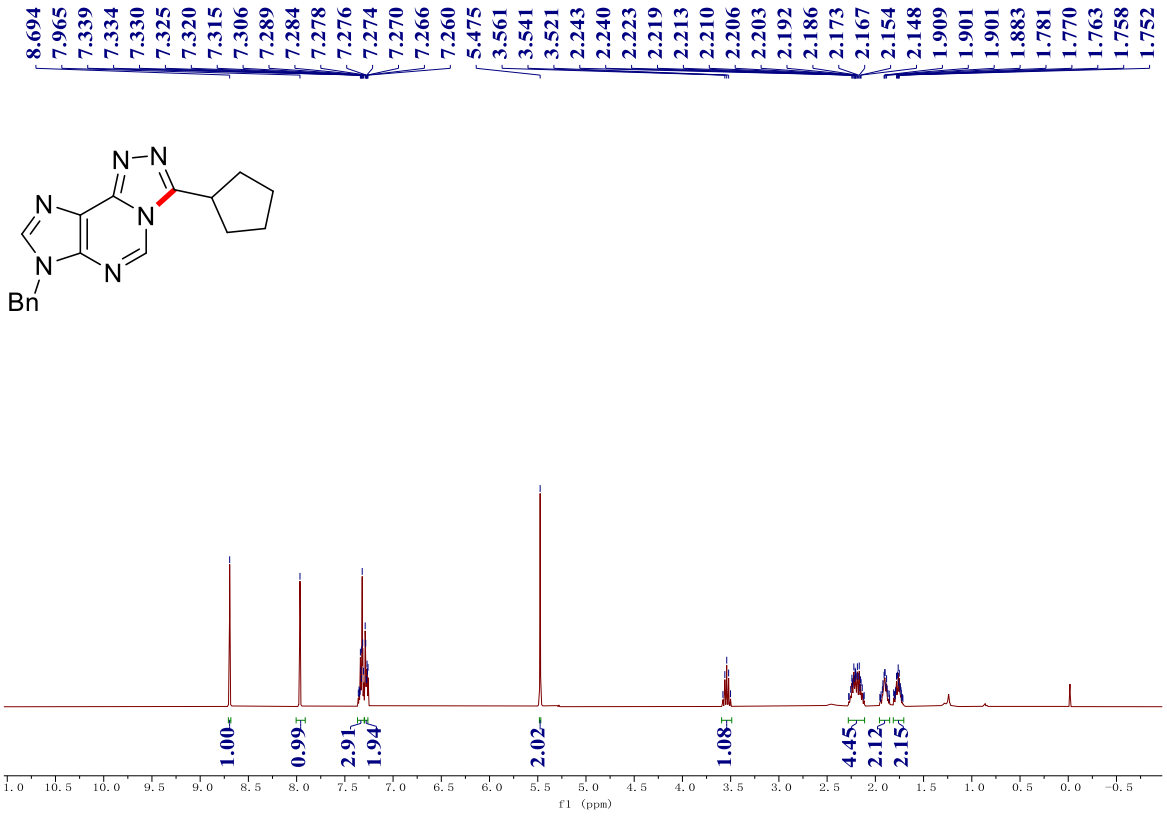
^{1}H NMR Spectrum of 2ad at 25 °C ($CDCl_3$, 600 MHz)



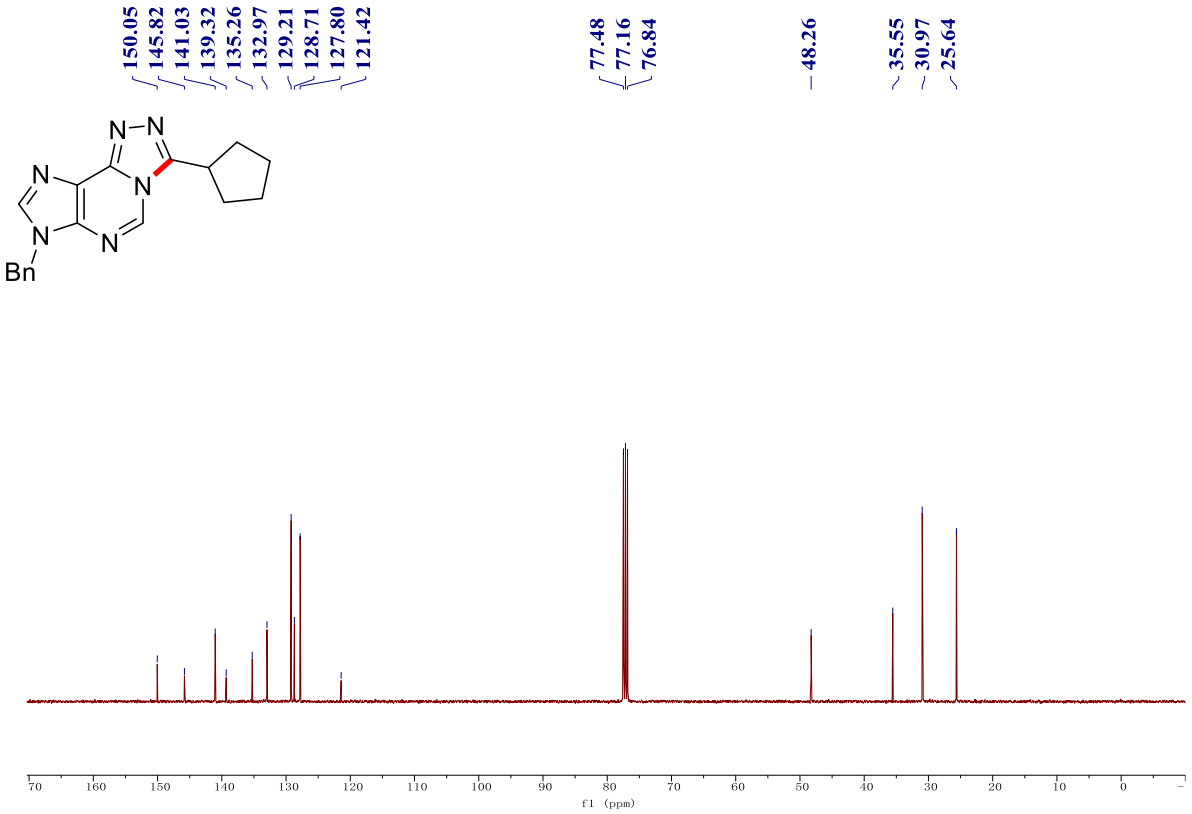
^{13}C NMR Spectrum of 2ad at 25 °C ($CDCl_3$, 150 MHz)



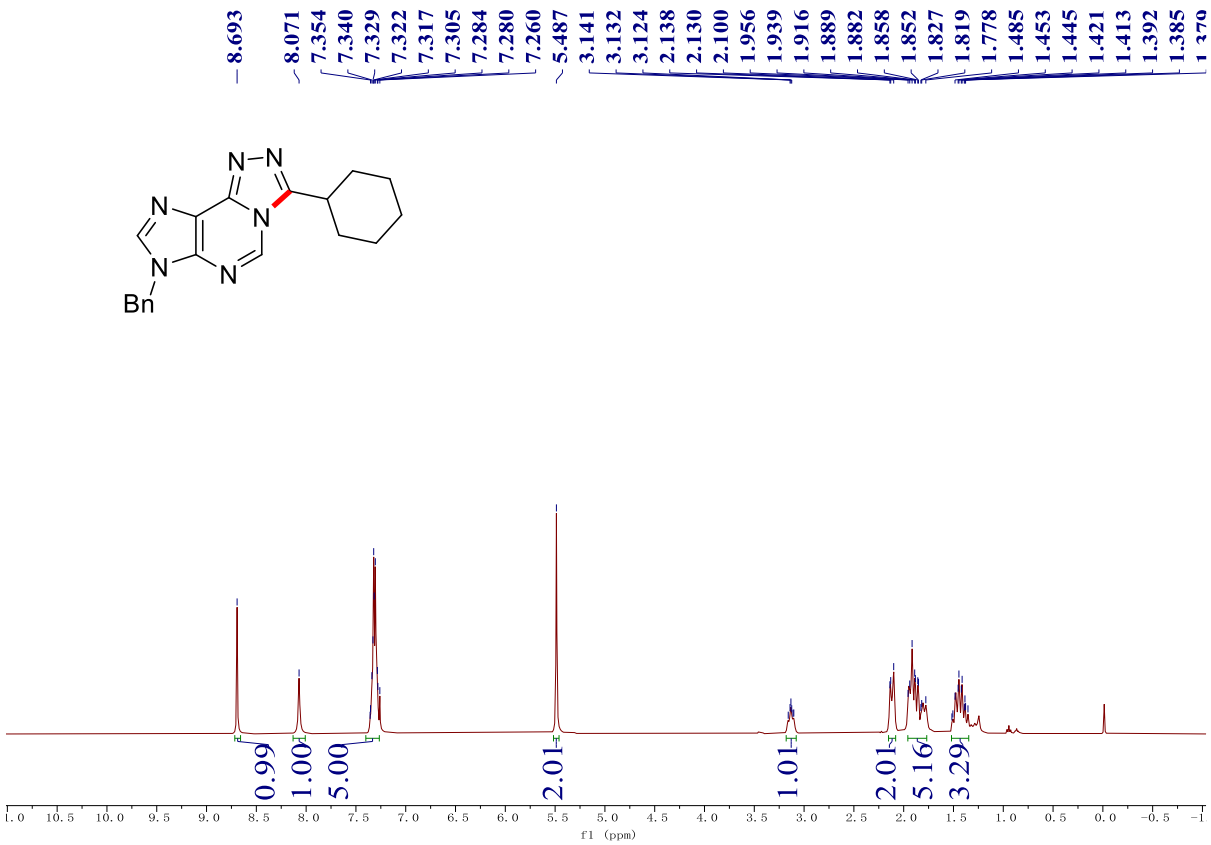
^{1}H NMR Spectrum of 2ae at 25 °C ($CDCl_3$, 400 MHz)



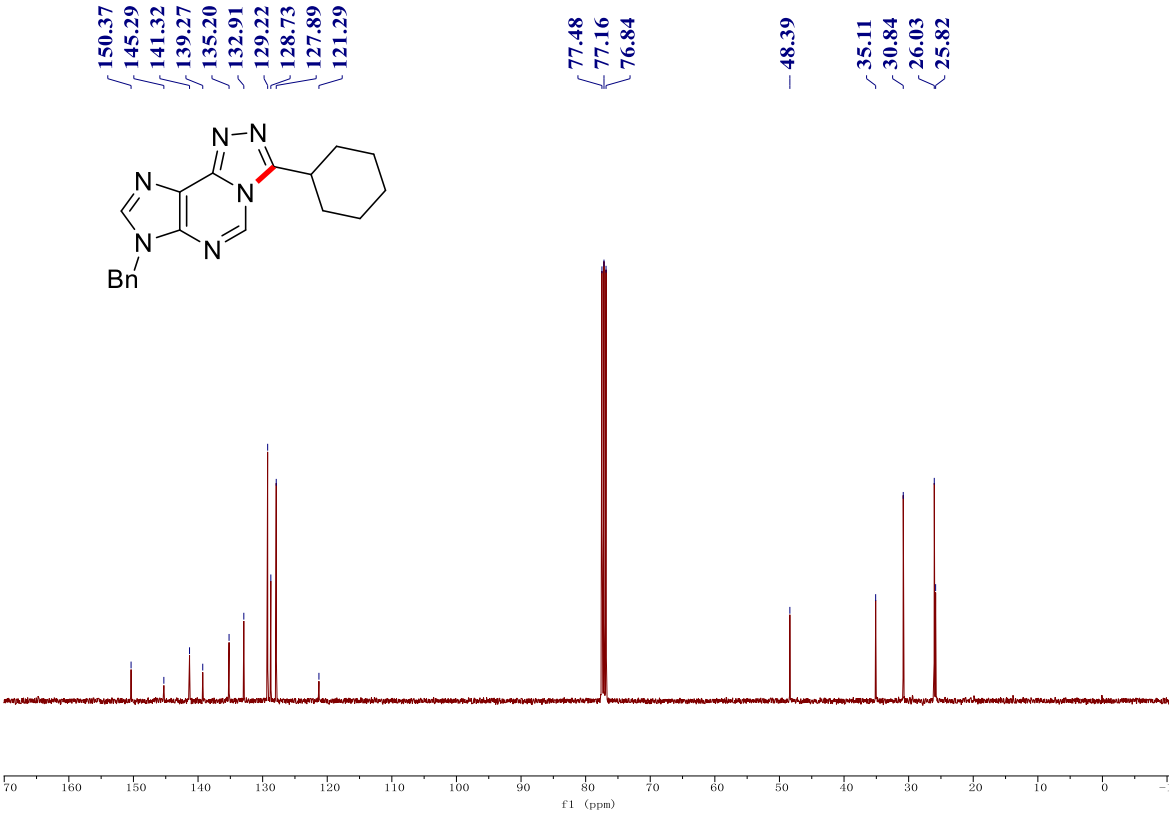
^{13}C NMR Spectrum of 2ae at 25 °C ($CDCl_3$, 100 MHz)



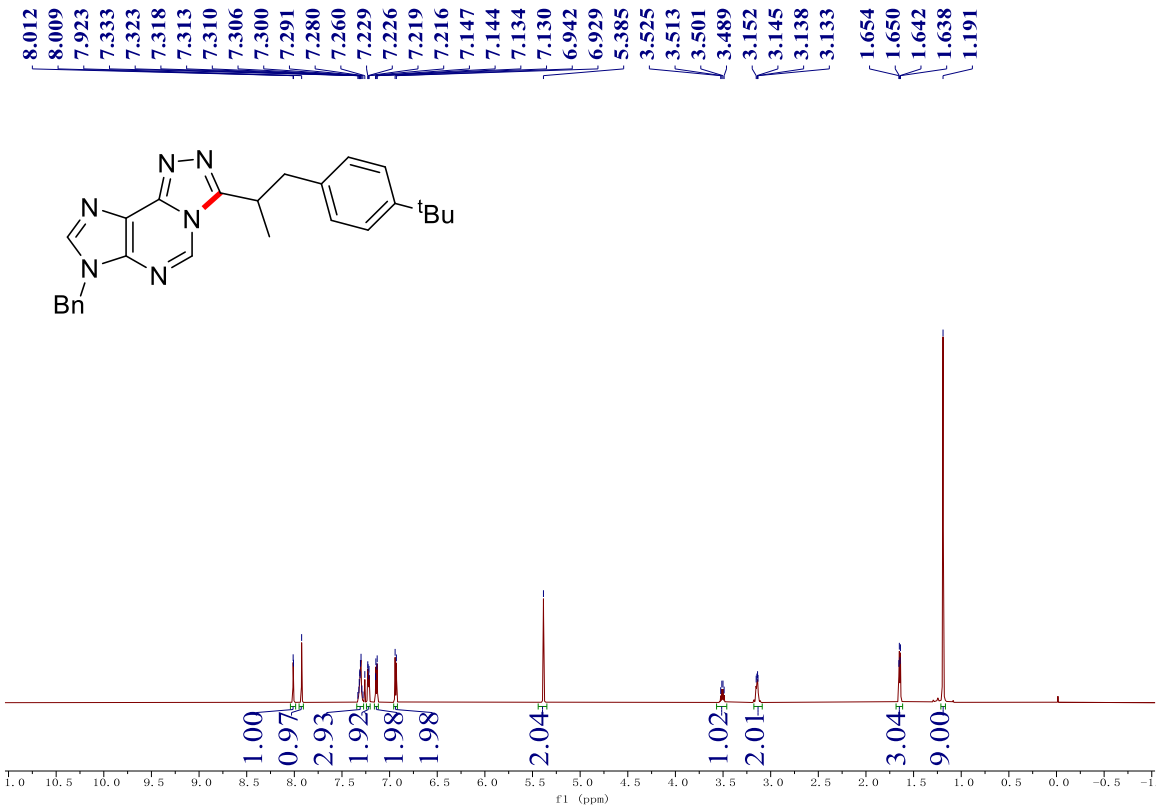
^{1}H NMR Spectrum of 2af at 25 °C ($CDCl_3$, 400 MHz)



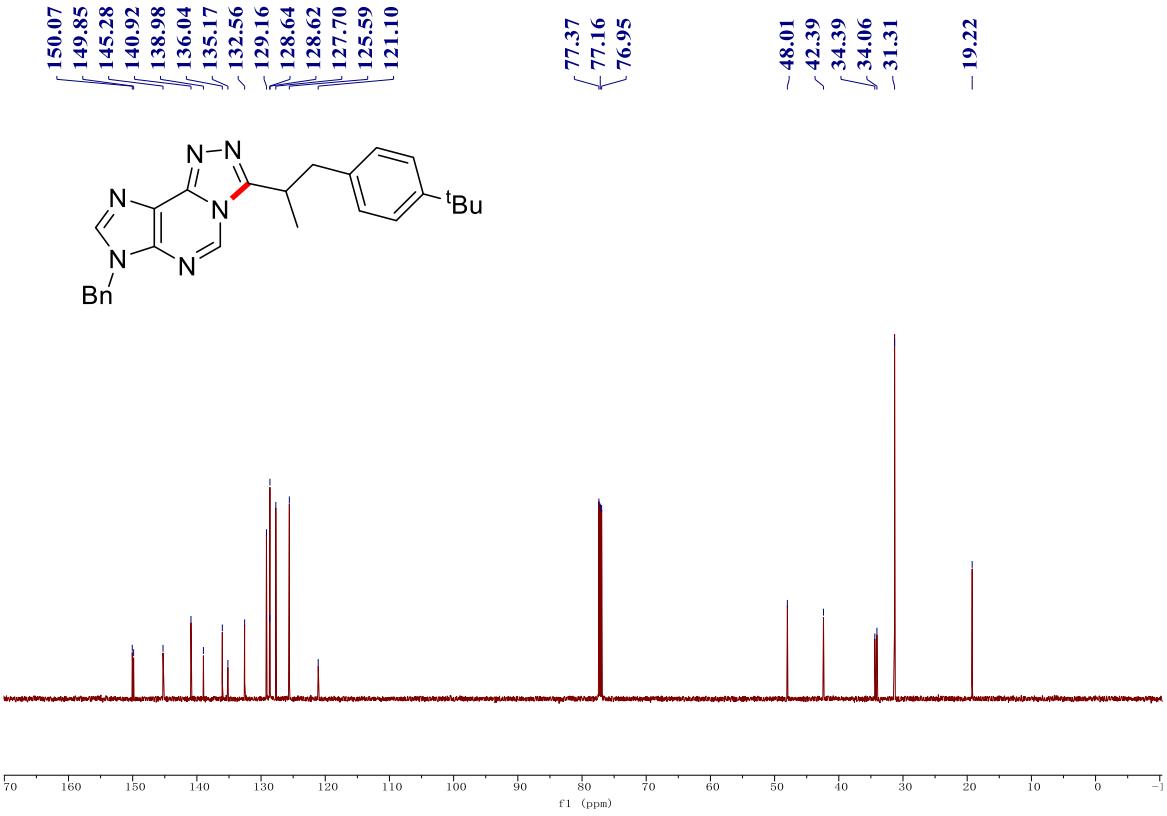
^{13}C NMR Spectrum of 2af at 25 °C ($CDCl_3$, 100 MHz)



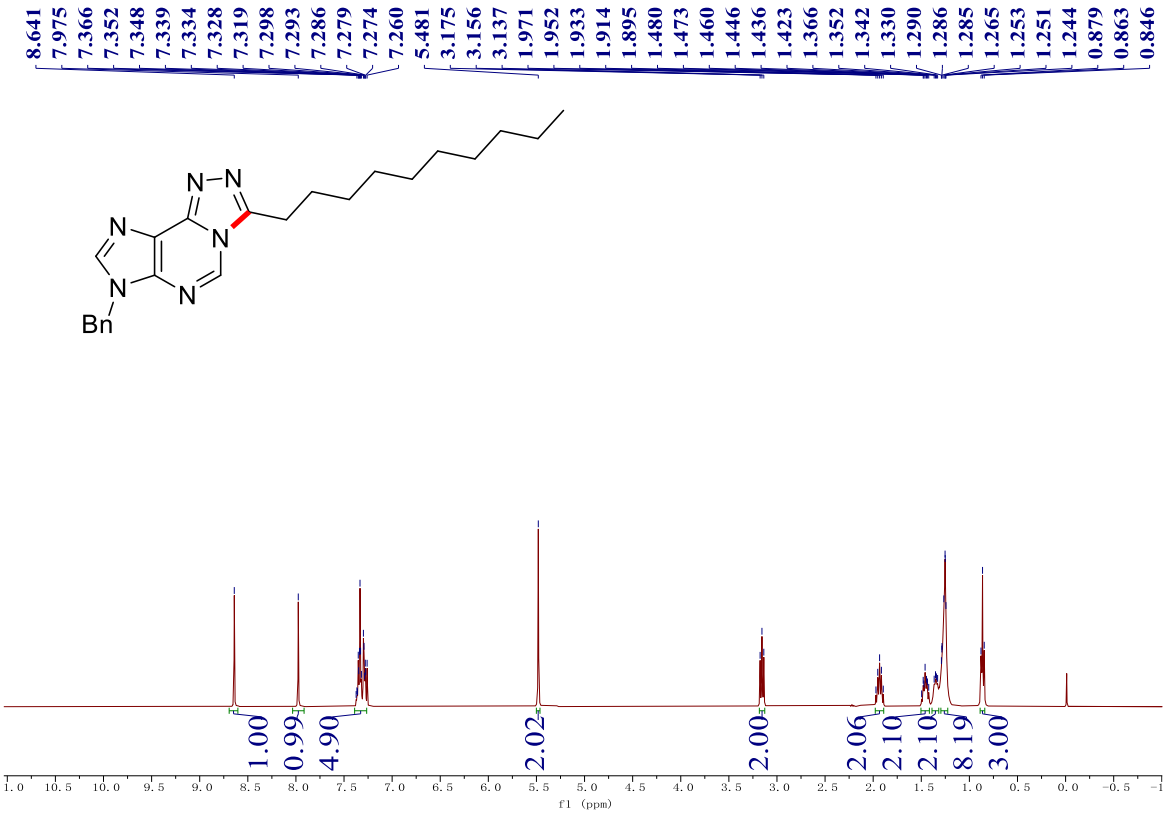
^{1}H NMR Spectrum of 2ag at 25 °C ($CDCl_3$, 600 MHz)



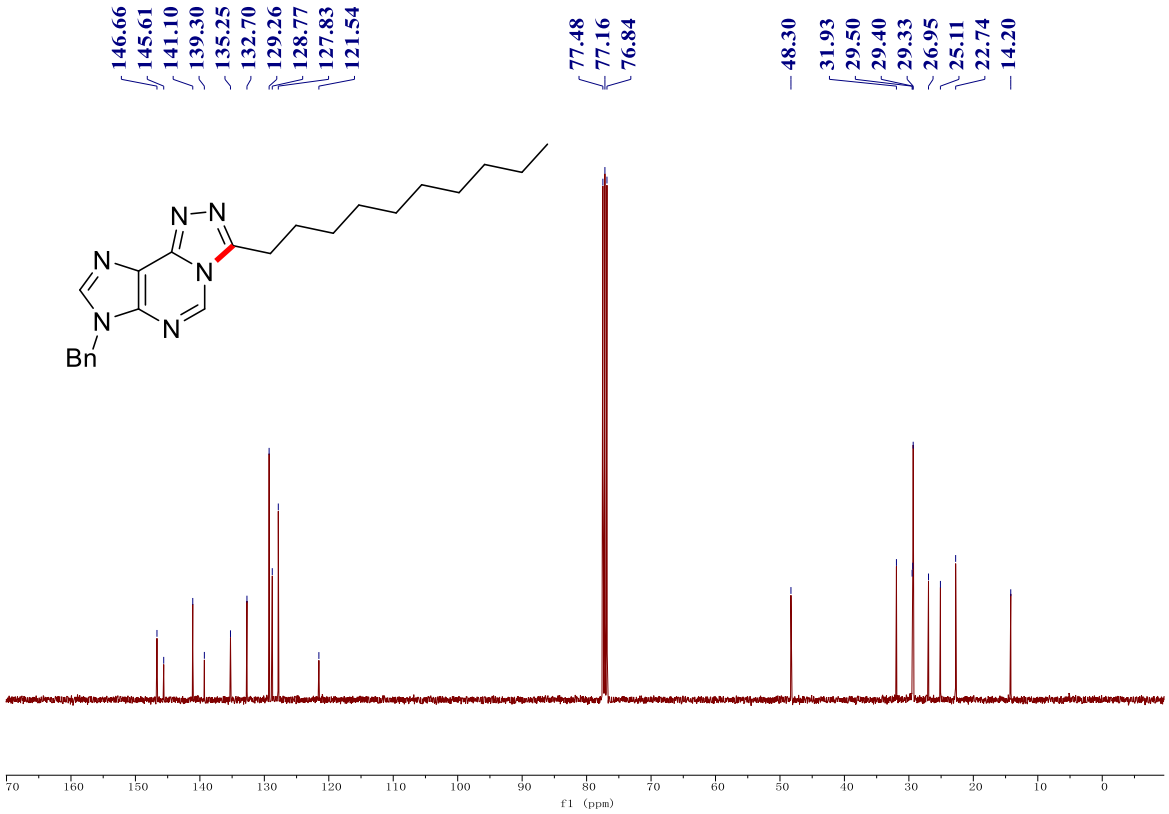
^{13}C NMR Spectrum of 2ag at 25 °C ($CDCl_3$, 150 MHz)



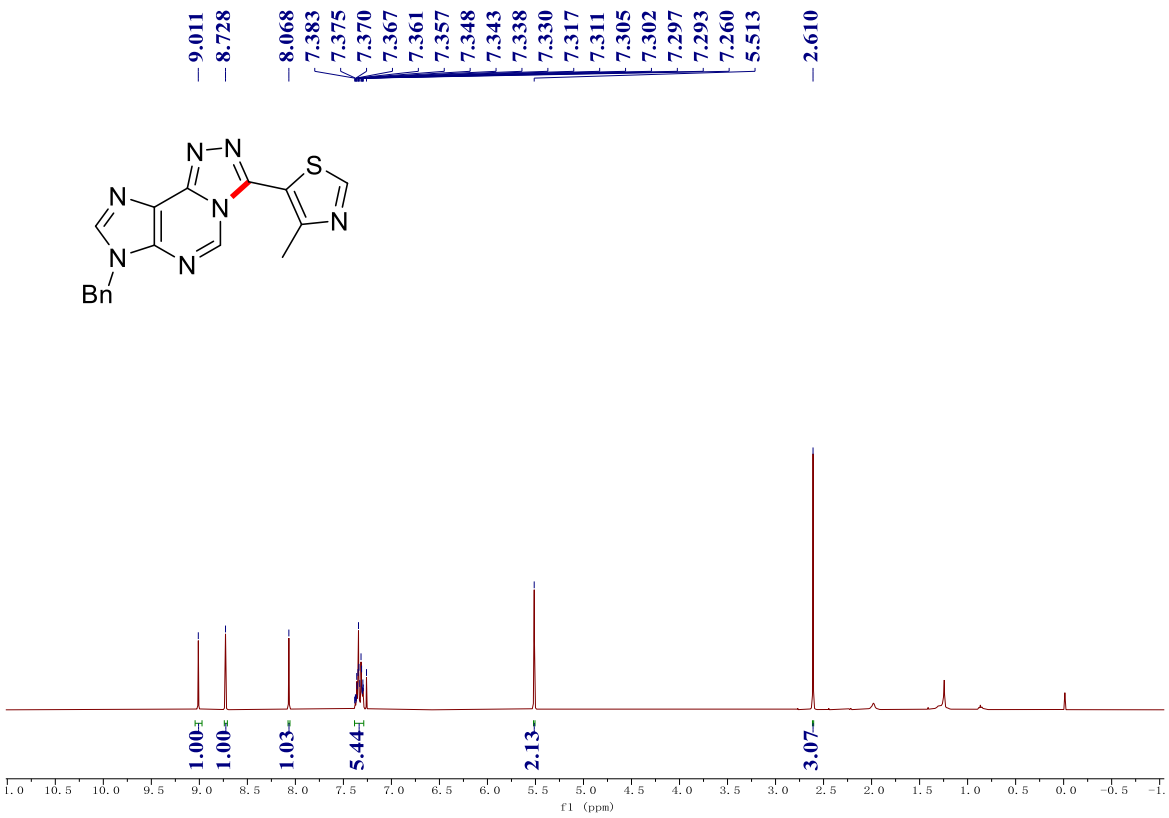
^{1}H NMR Spectrum of 2ah at 25 °C ($CDCl_3$, 400 MHz)



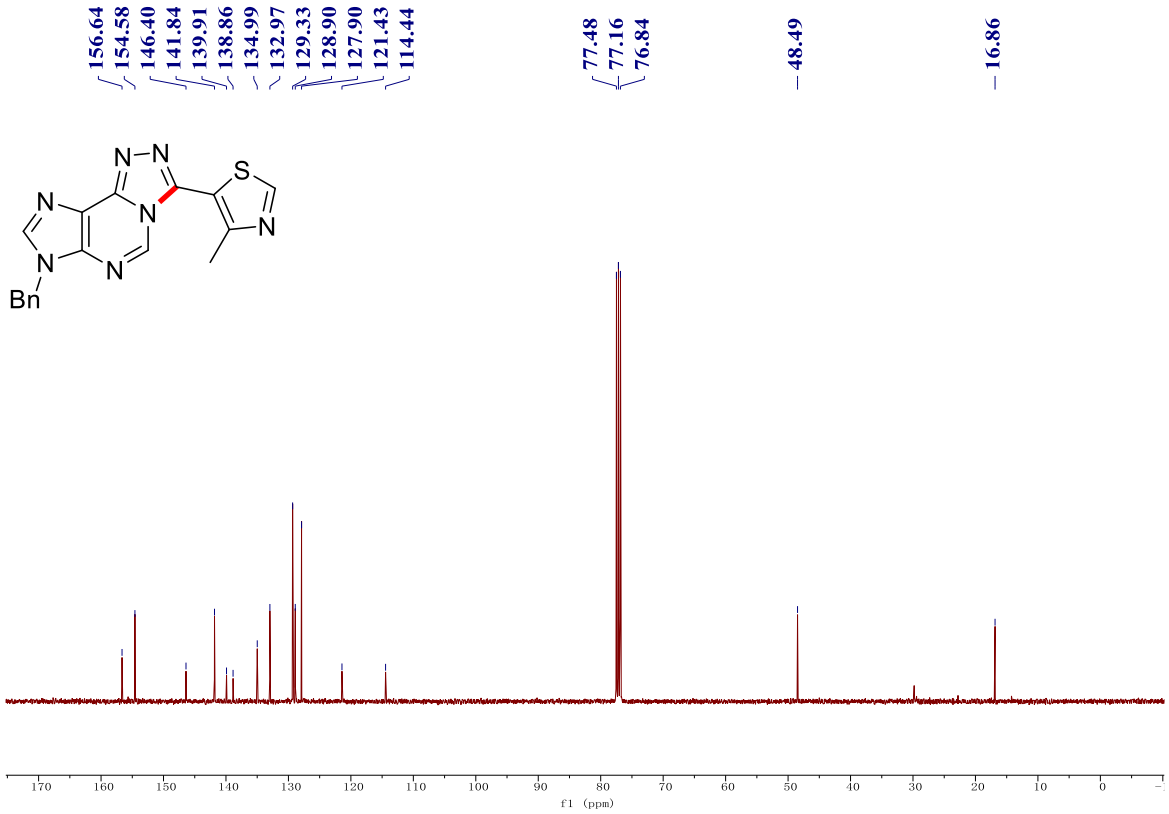
^{13}C NMR Spectrum of 2ah at 25 °C ($CDCl_3$, 100 MHz)



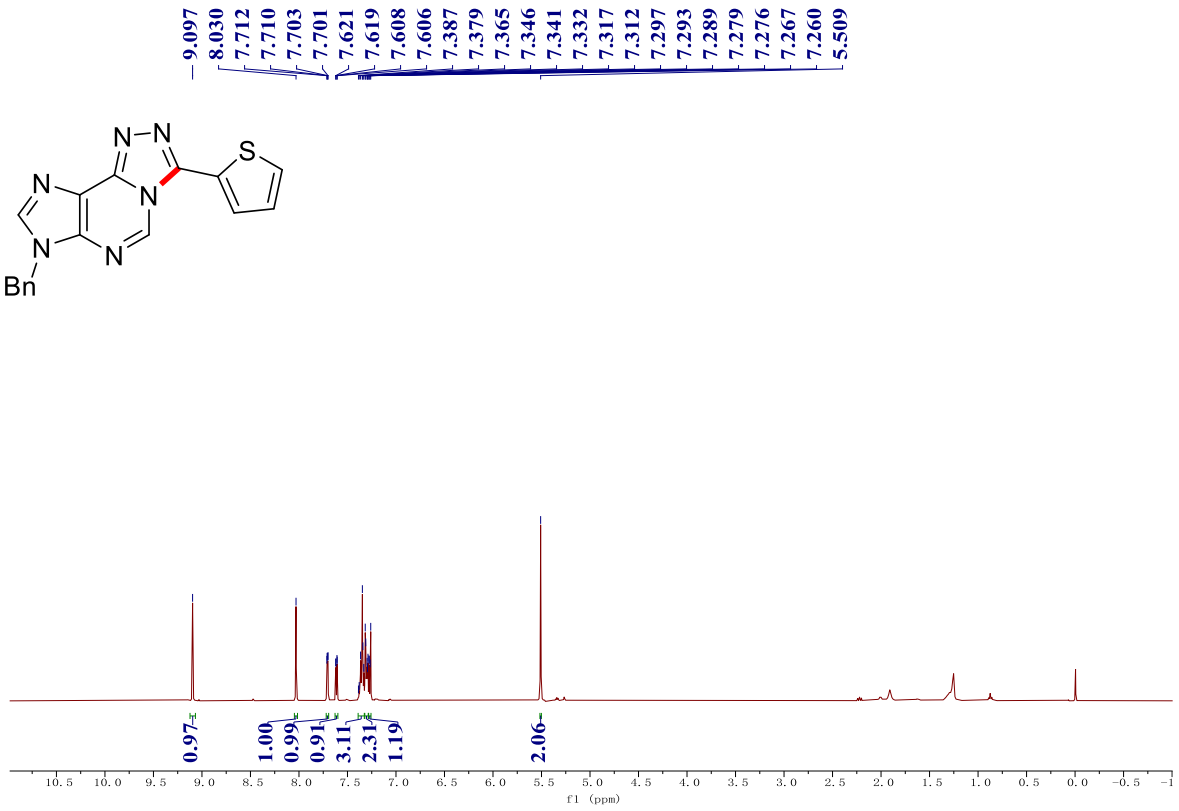
^{1}H NMR Spectrum of 2ai at 25 °C ($CDCl_3$, 400 MHz)



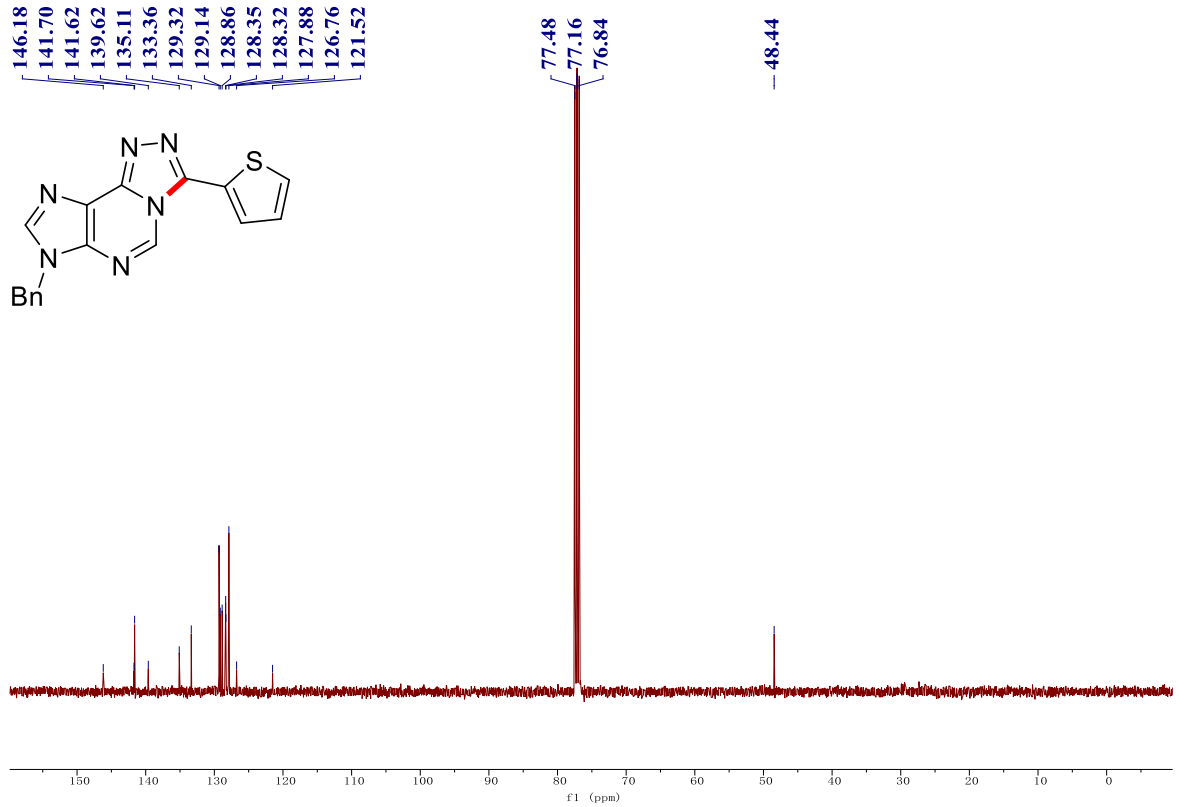
^{13}C NMR Spectrum of 2ai at 25 °C ($CDCl_3$, 100 MHz)



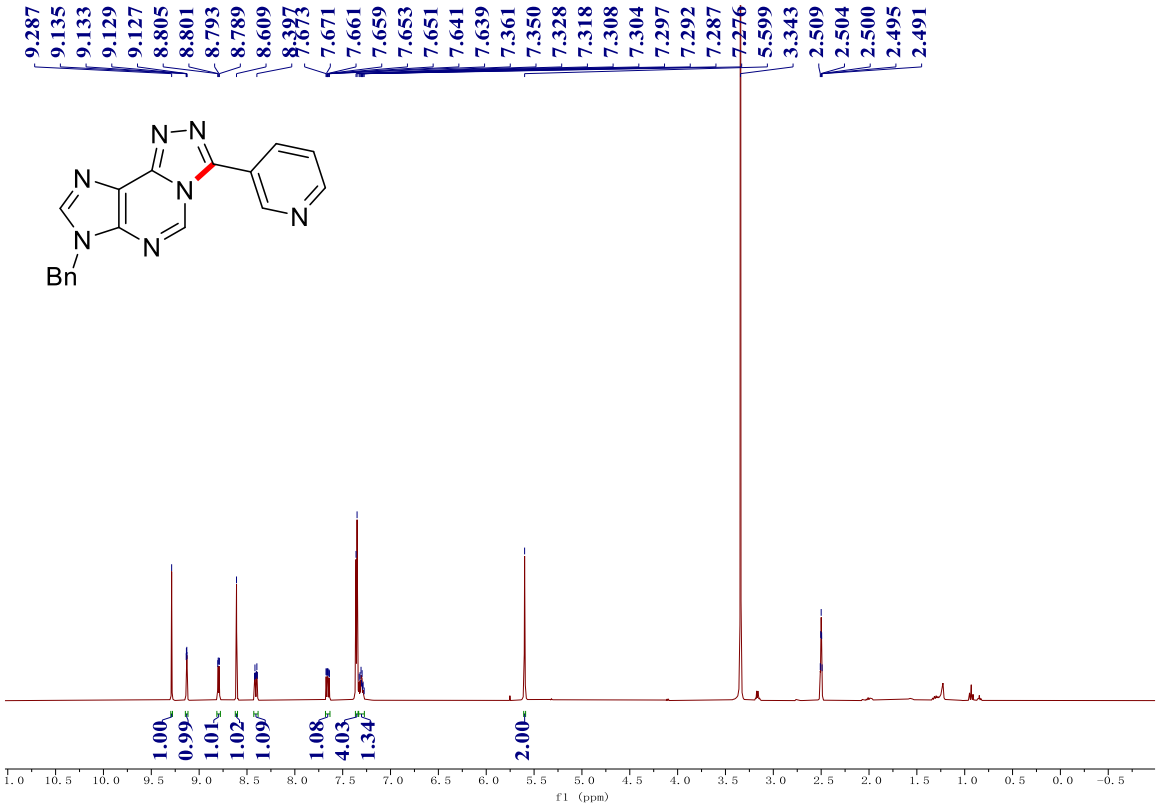
^{1}H NMR Spectrum of 2aj at 25 °C ($CDCl_3$, 400 MHz)



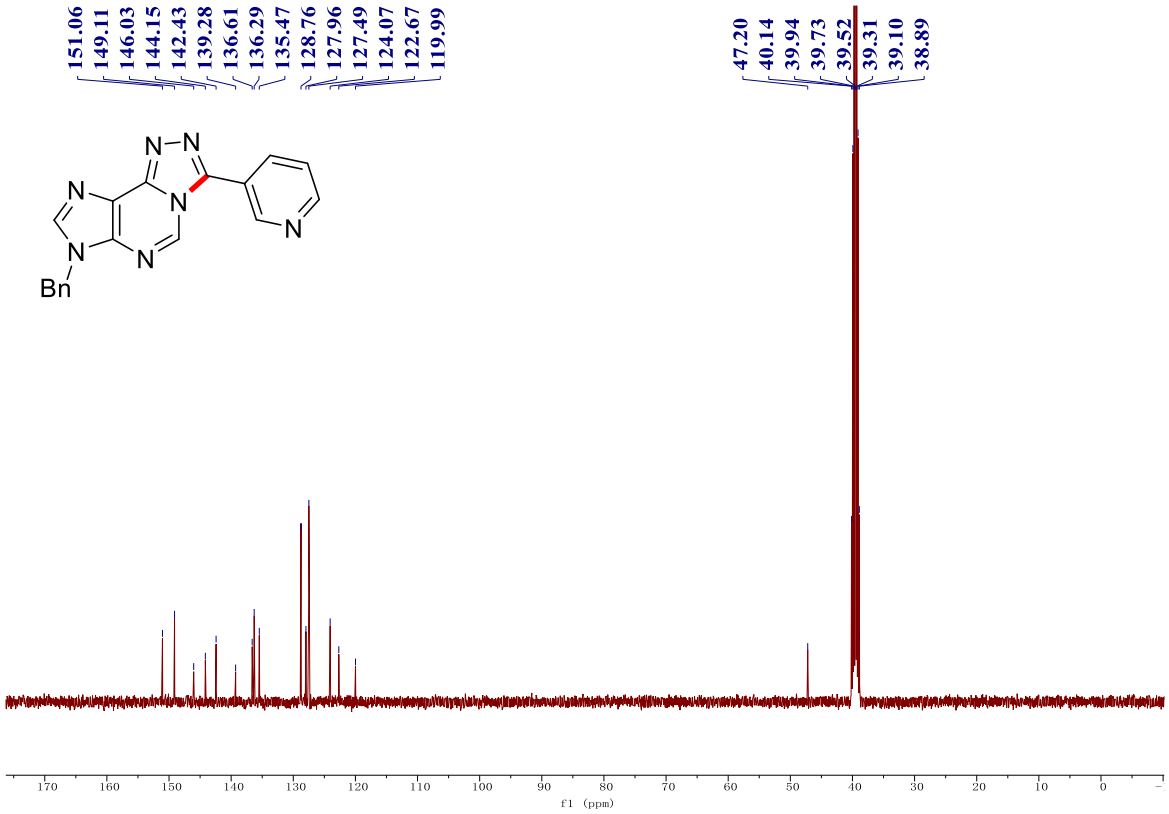
^{13}C NMR Spectrum of 2aj at 25 °C ($CDCl_3$, 100 MHz)



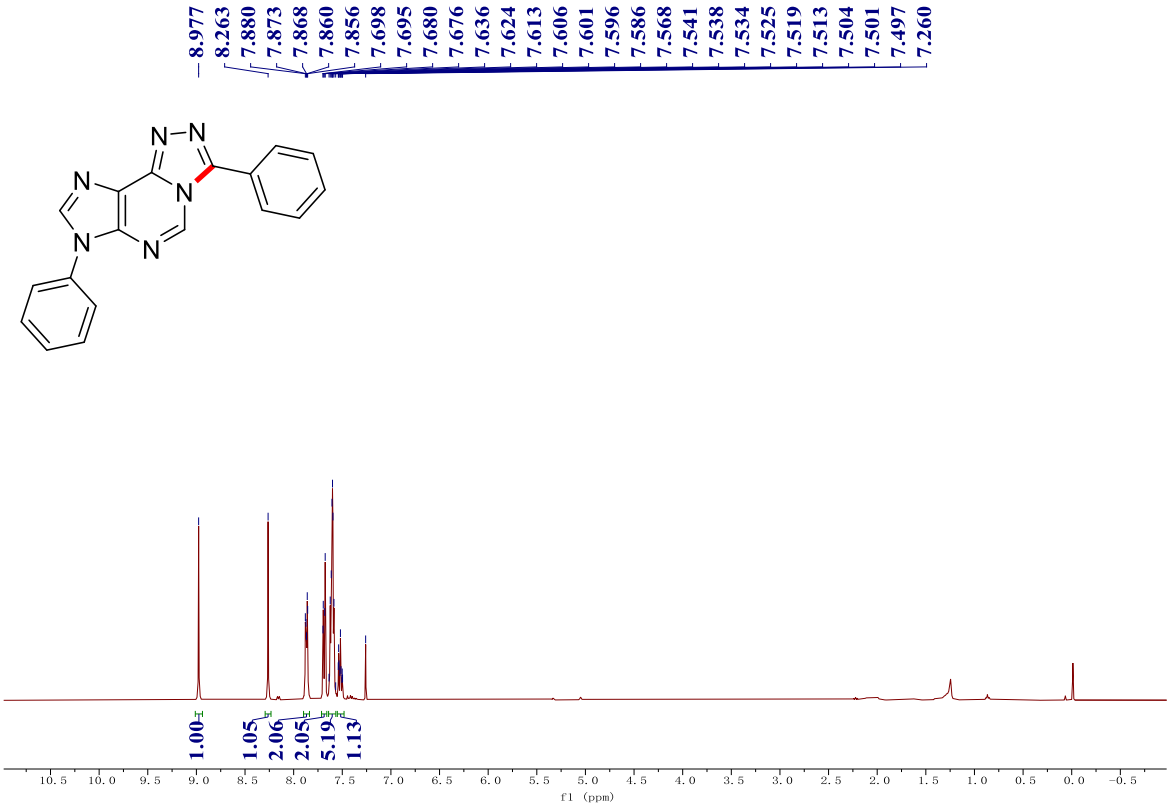
^{1}H NMR Spectrum of 2ak at 25 °C (DMSO-d_6, 400 MHz)



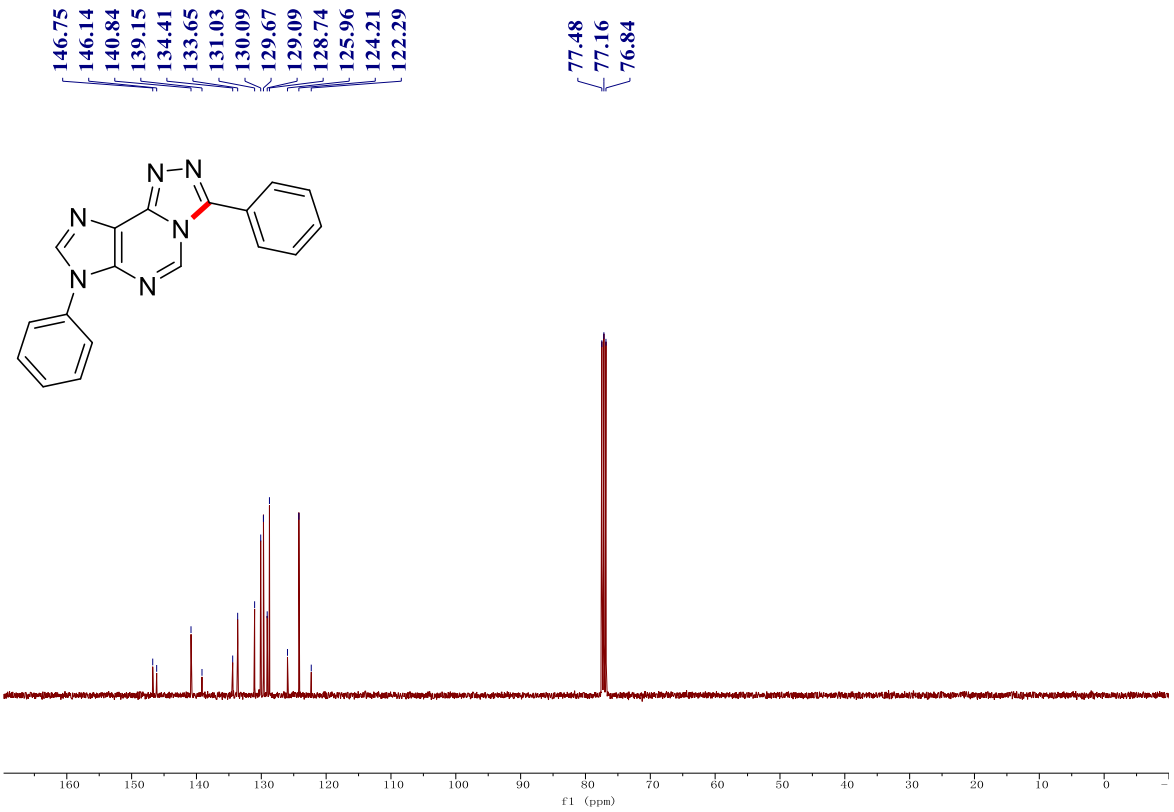
^{13}C NMR Spectrum of 2ak at 25 °C (DMSO-d_6, 100 MHz)



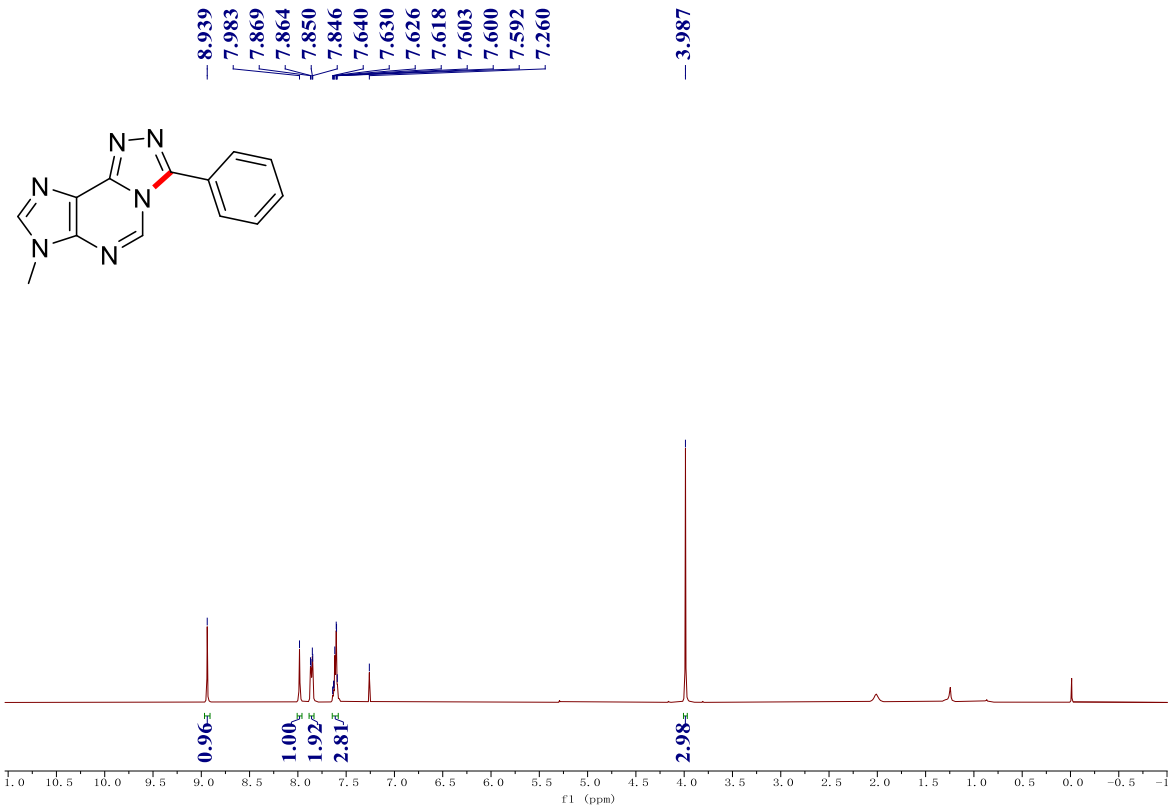
^{1}H NMR Spectrum of 2al at 25 °C ($CDCl_3$, 400 MHz)



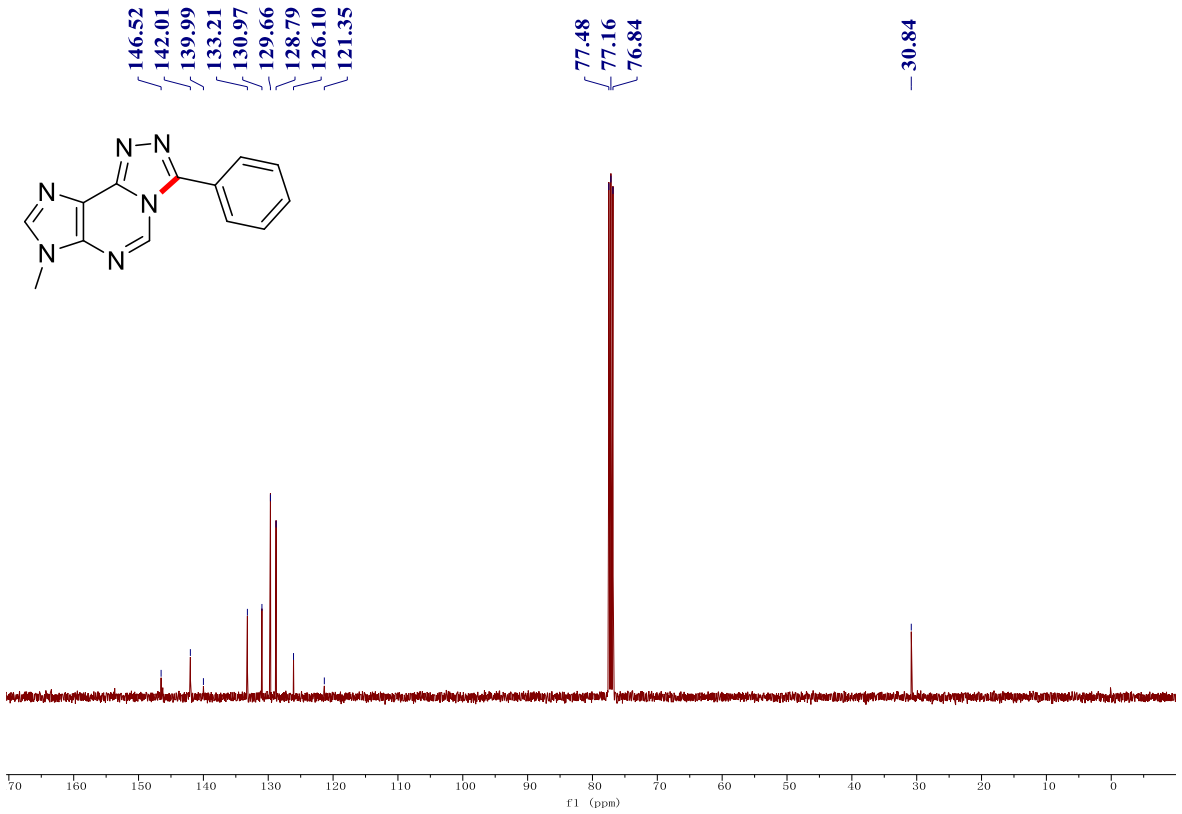
^{13}C NMR Spectrum of 2al at 25 °C ($CDCl_3$, 100 MHz)



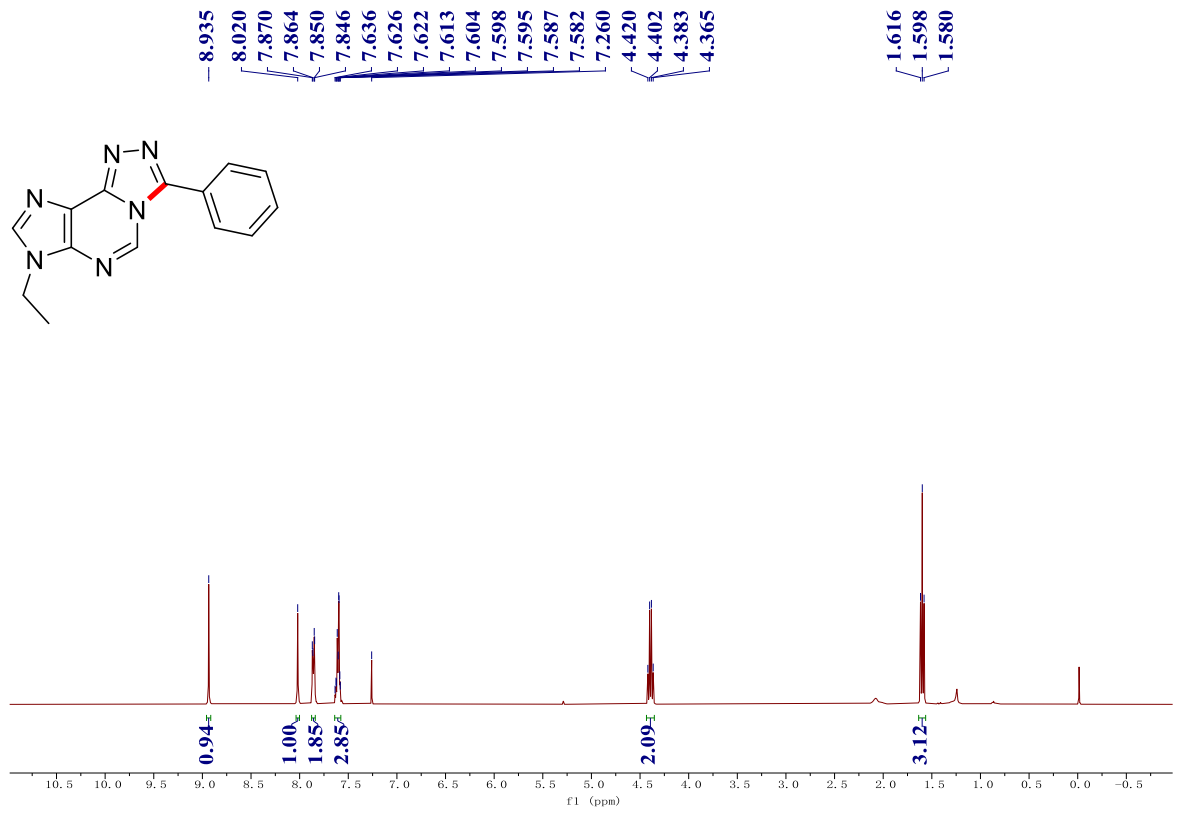
^{1}H NMR Spectrum of 2am at 25 °C ($CDCl_3$, 400 MHz)



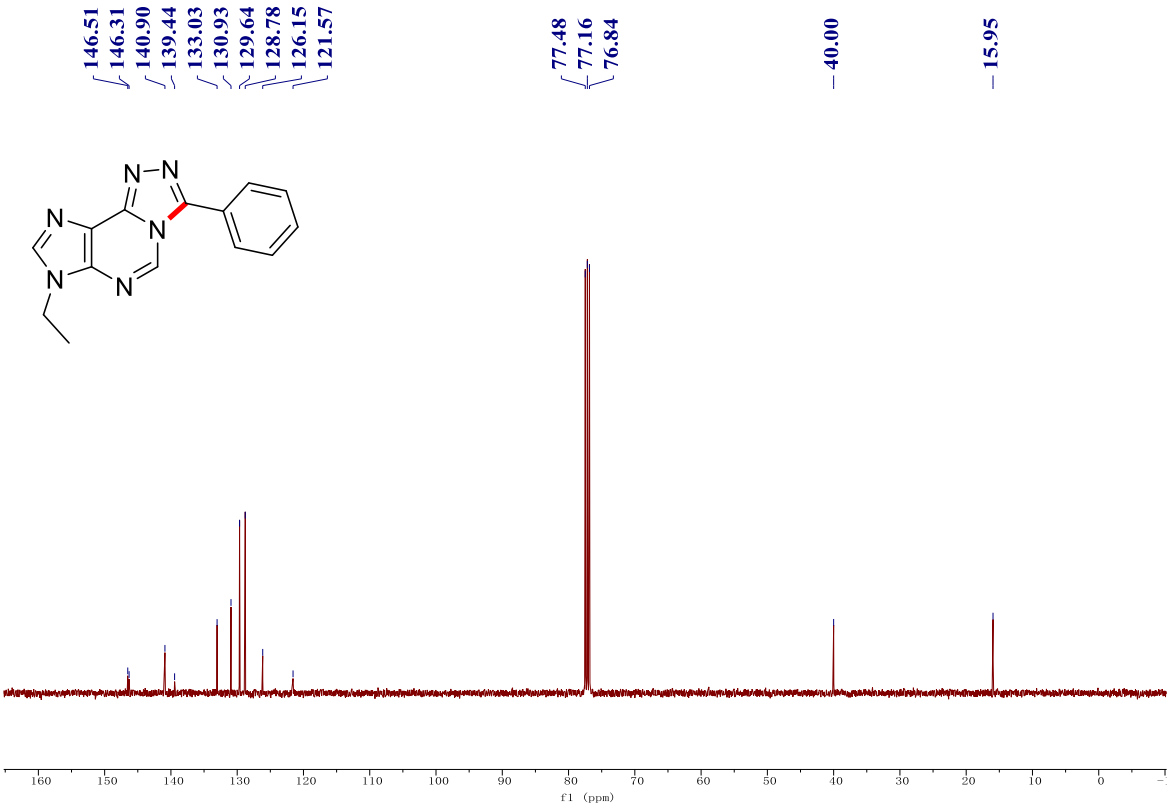
^{13}C NMR Spectrum of 2am at 25 °C ($CDCl_3$, 100 MHz)



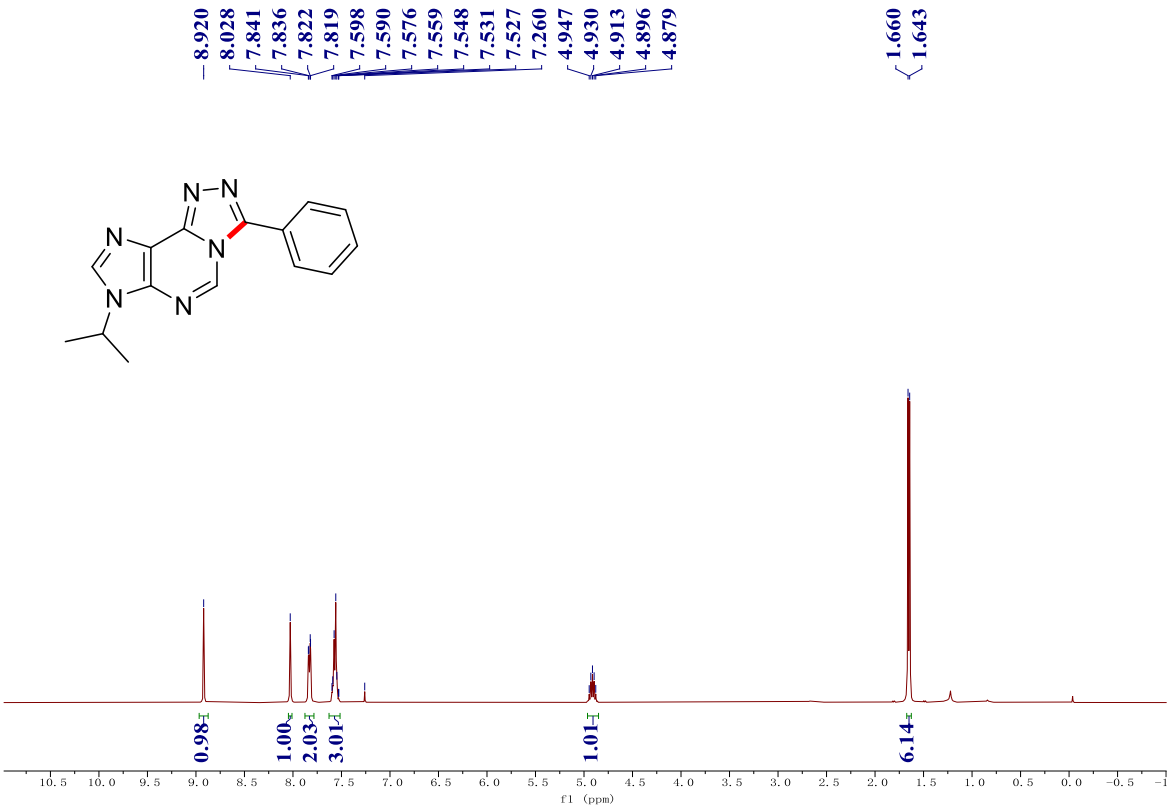
^{1}H NMR Spectrum of 2an at 25 °C ($CDCl_3$, 400 MHz)



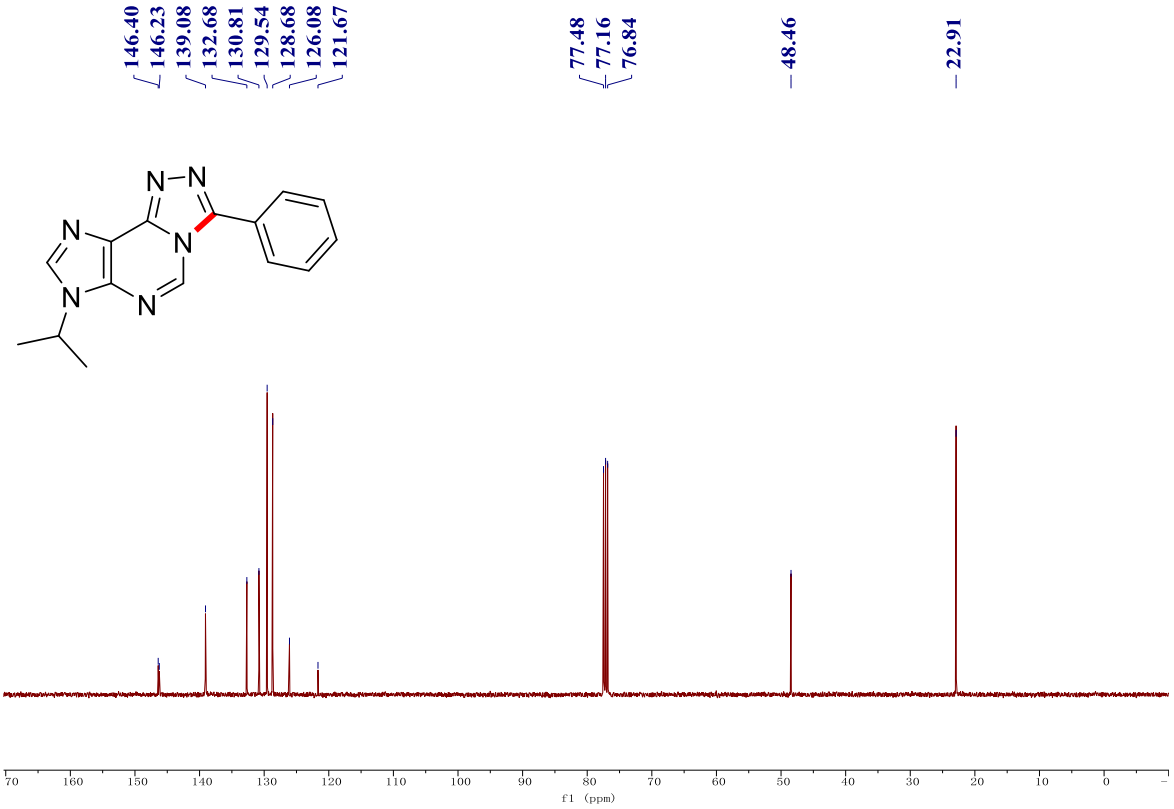
^{13}C NMR Spectrum of 2an at 25 °C ($CDCl_3$, 100 MHz)



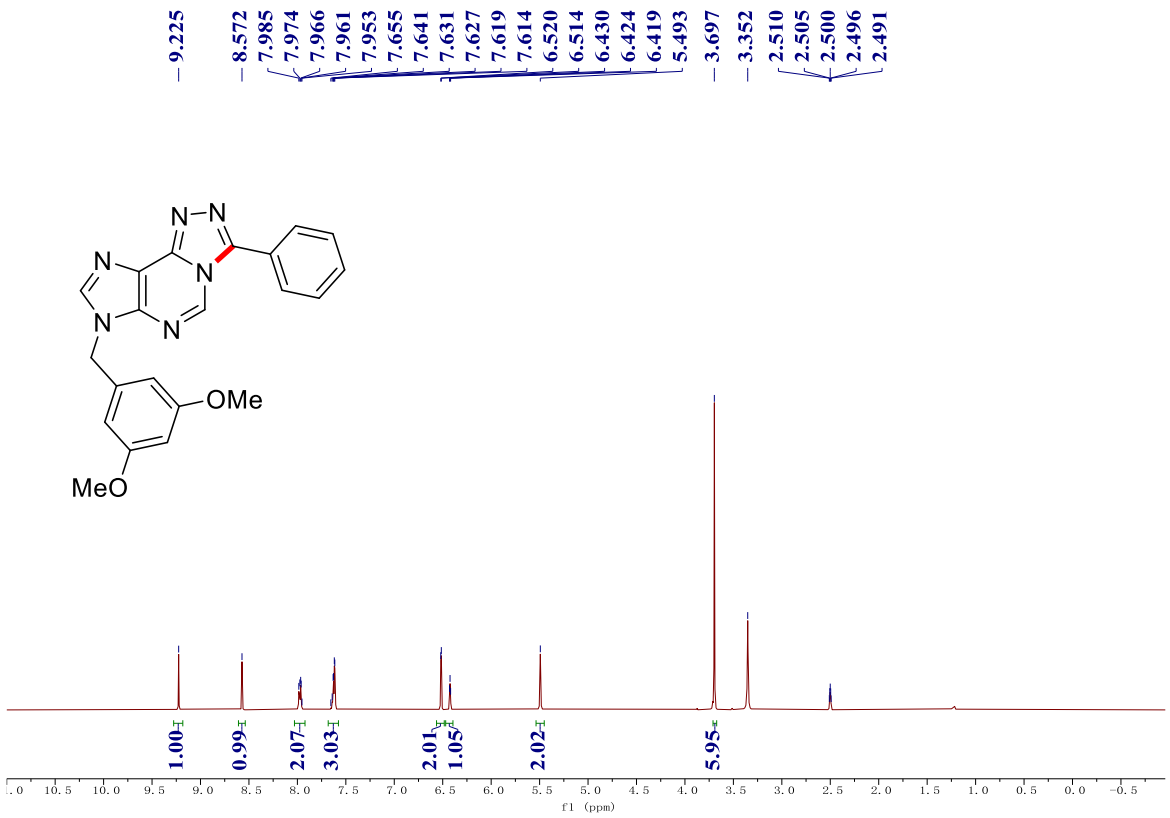
^{1}H NMR Spectrum of 2ao at 25 °C ($CDCl_3$, 400 MHz)



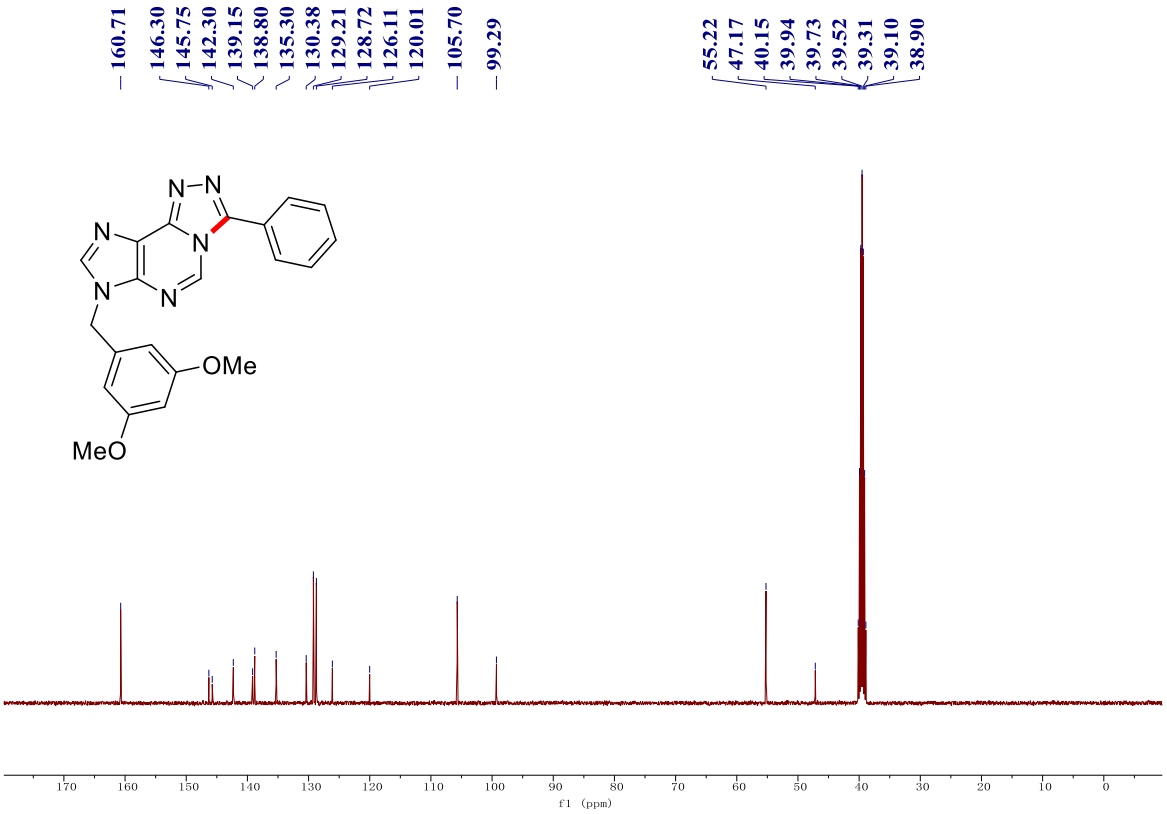
^{13}C NMR Spectrum of 2ao at 25 °C ($CDCl_3$, 100 MHz)



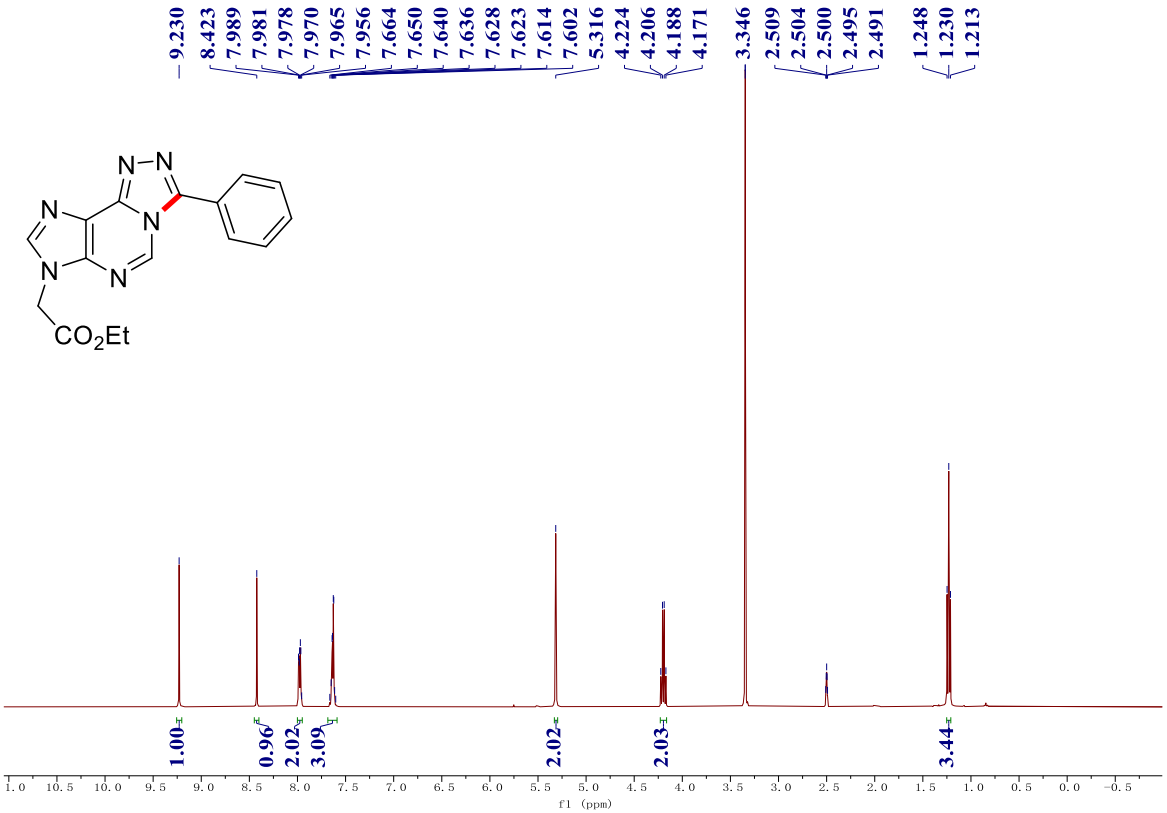
^{1}H NMR Spectrum of 2ap at 25 °C (DMSO-d_6, 400 MHz)



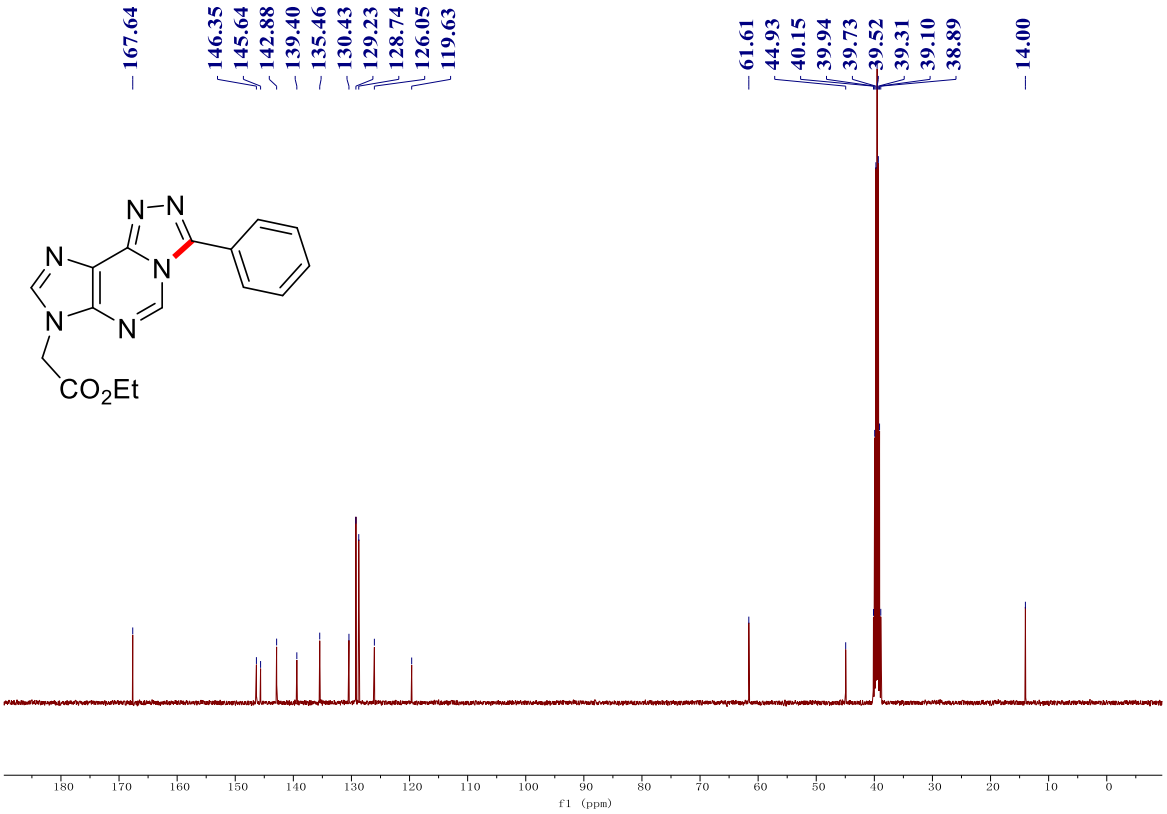
^{13}C NMR Spectrum of 2ap at 25 °C (DMSO-d_6, 100 MHz)



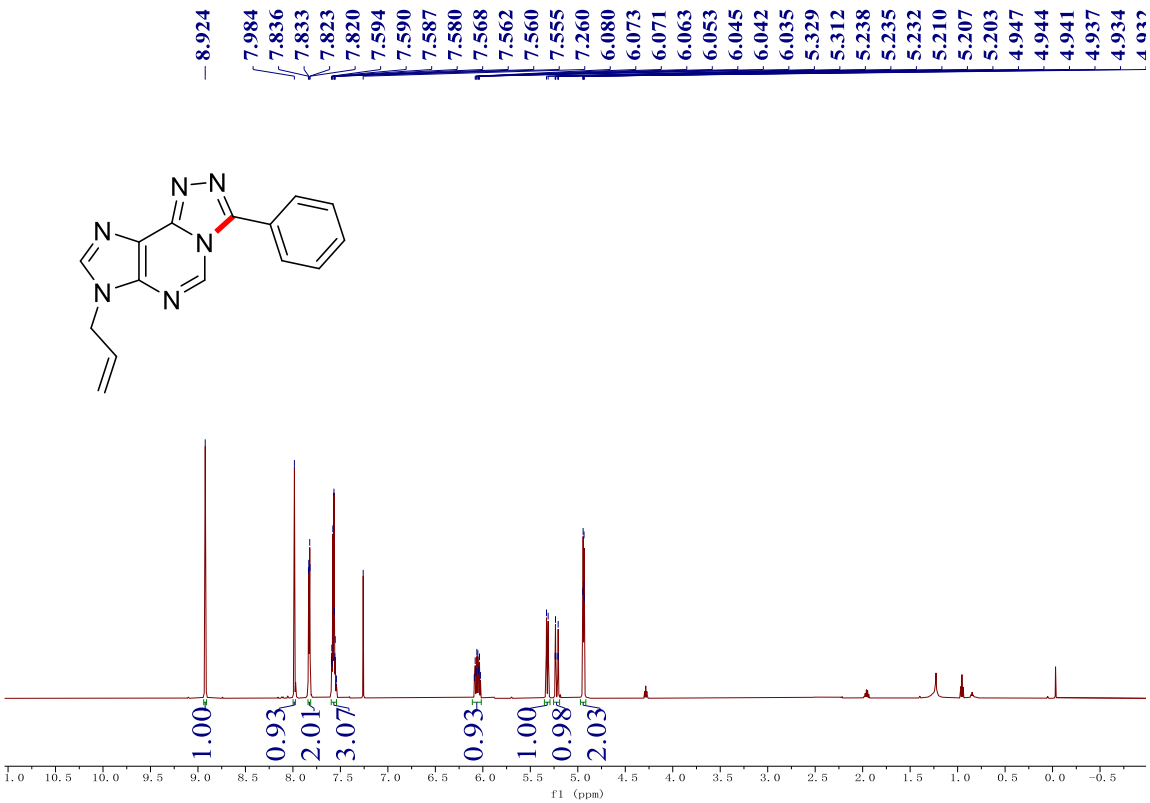
^{1}H NMR Spectrum of 2aq at 25 °C (DMSO-d_6, 400 MHz)



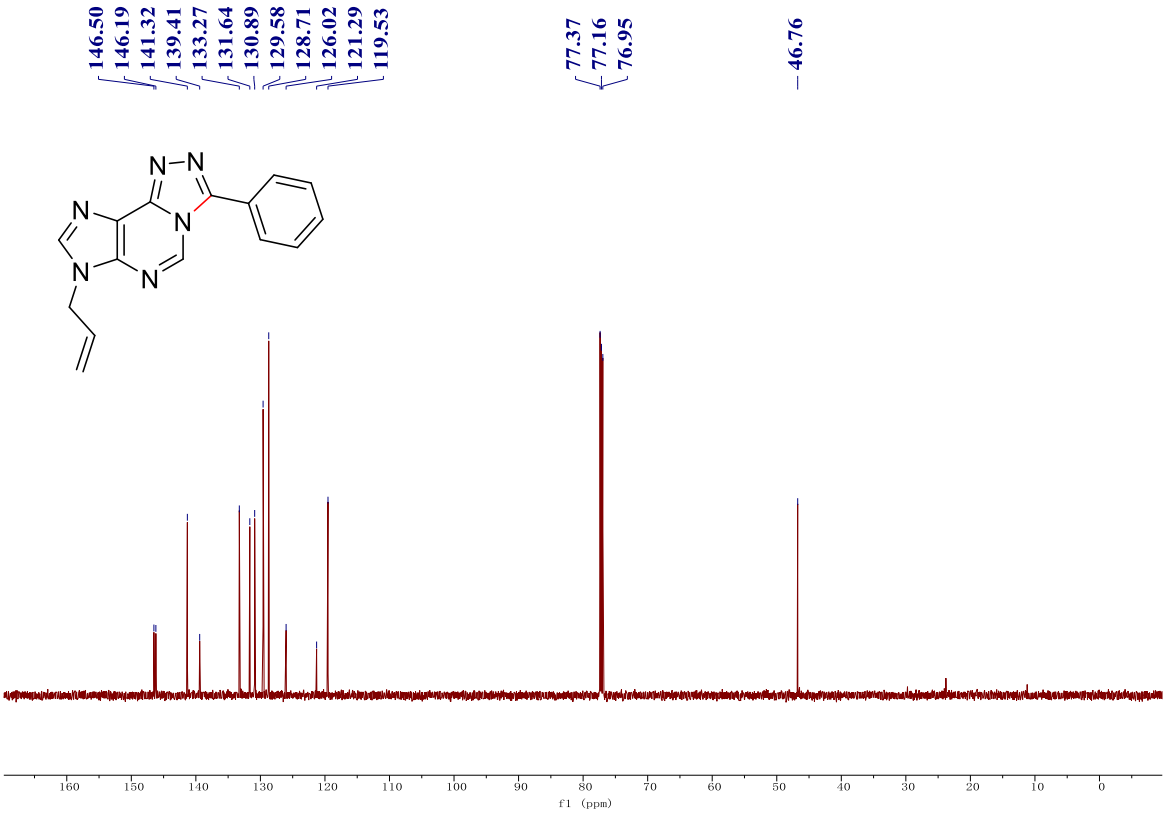
^{13}C NMR Spectrum of 2aq at 25 °C (DMSO-d_6, 100 MHz)



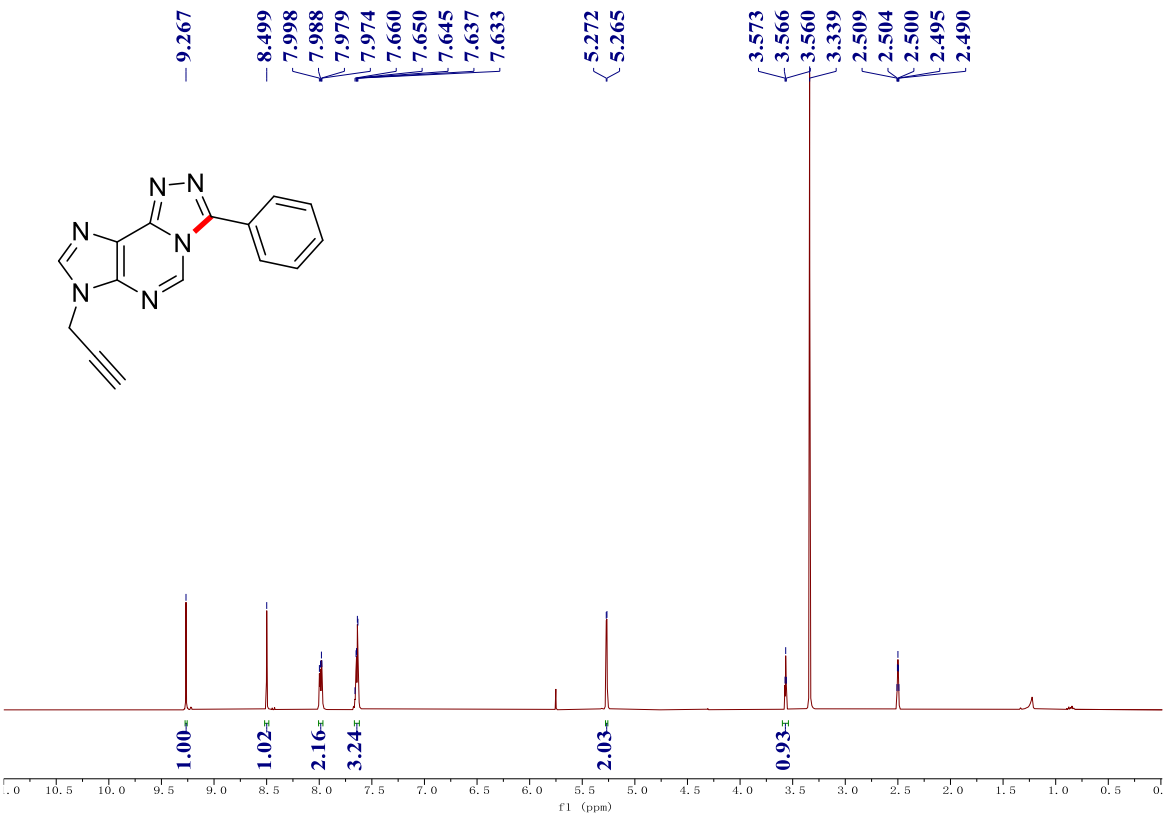
^{1}H NMR Spectrum of 2ar at 25 °C ($CDCl_3$, 600 MHz)



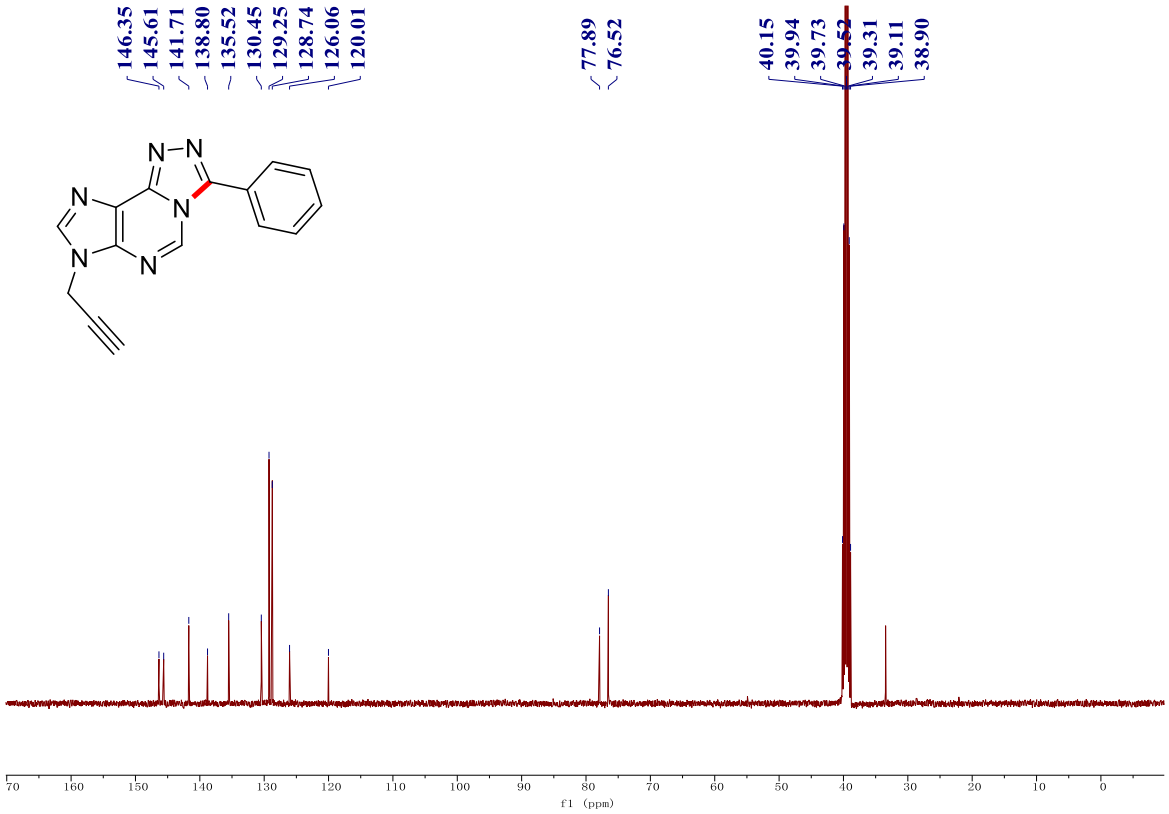
^{13}C NMR Spectrum of 2ar at 25 °C ($CDCl_3$, 150 MHz)



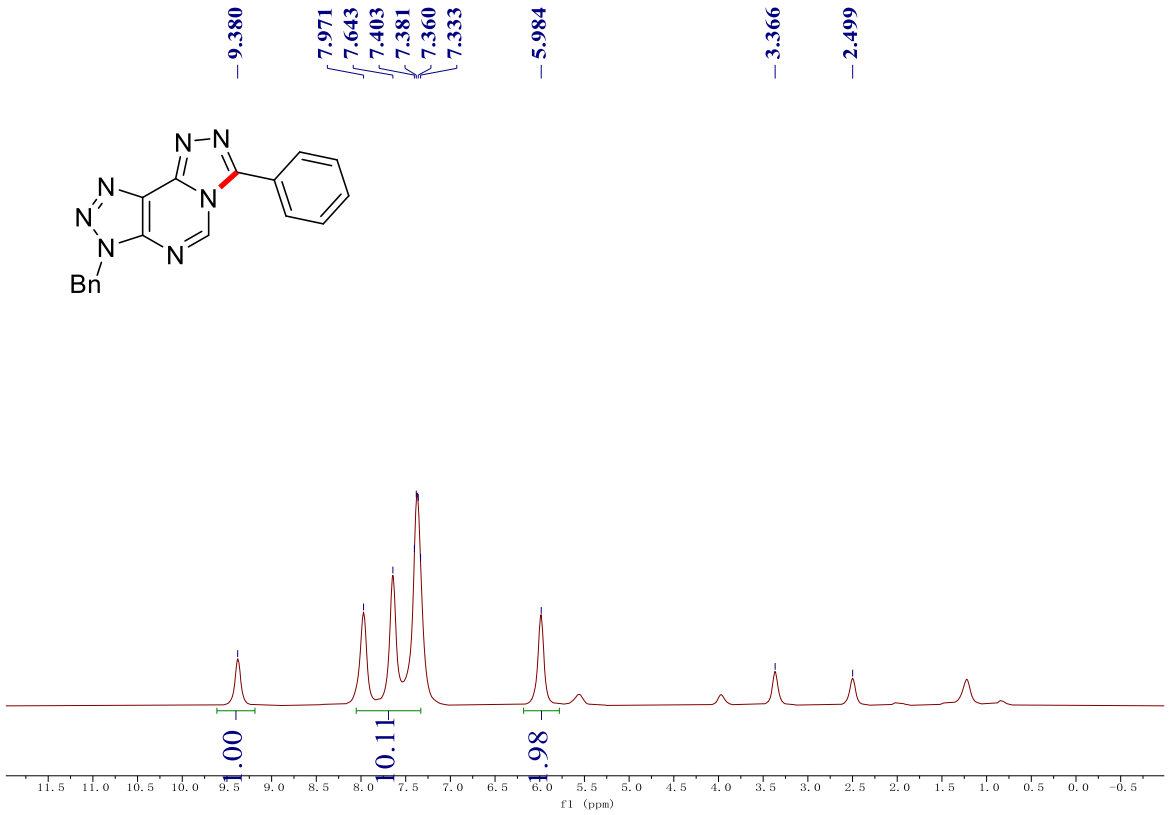
^{1}H NMR Spectrum of 2as at 25 °C (DMSO-d_6, 400 MHz)



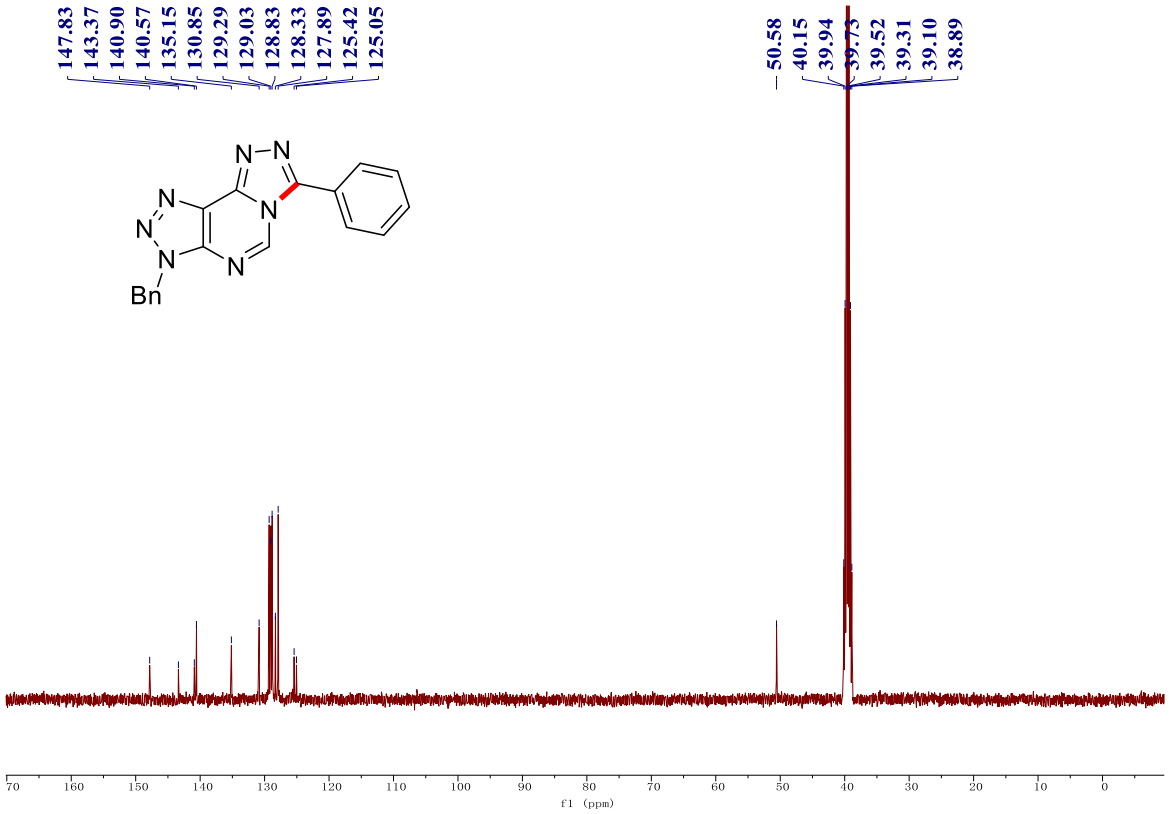
^{13}C NMR Spectrum of 2as at 25 °C (DMSO-d_6, 100 MHz)



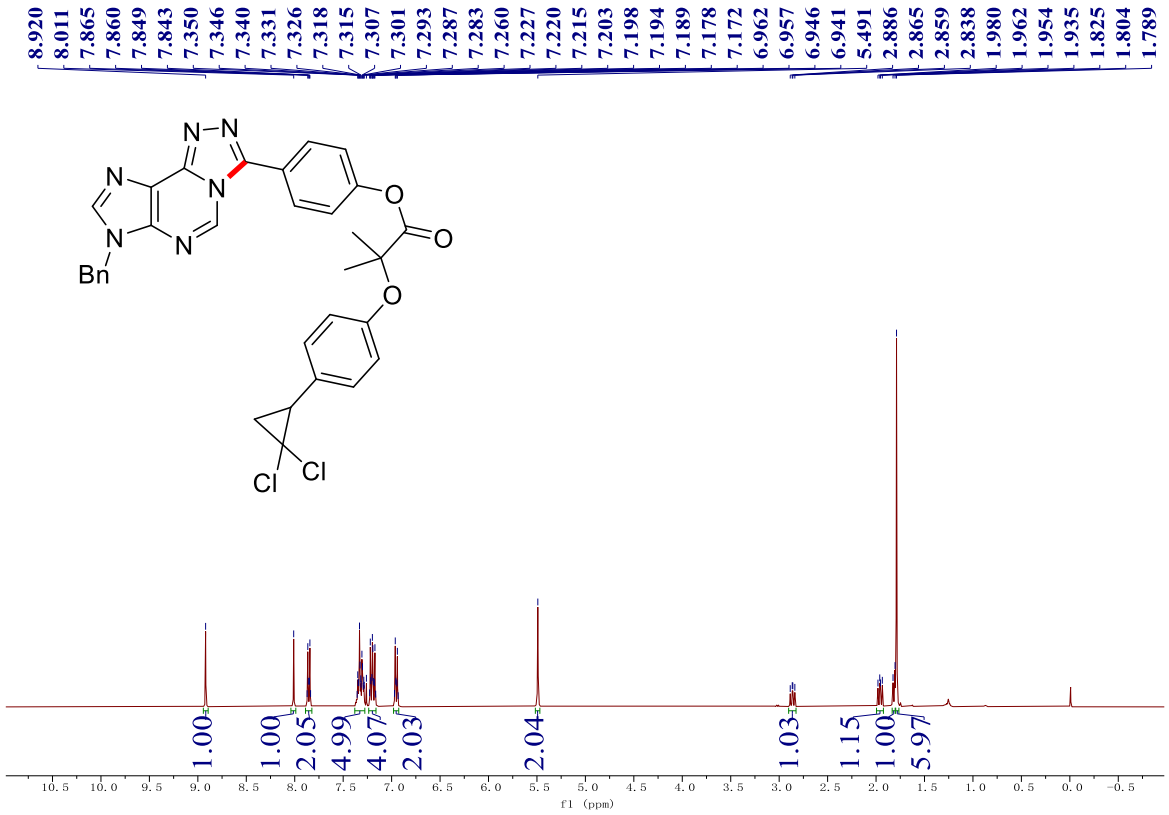
^{1}H NMR Spectrum of 2at at 25 °C (DMSO-d_6, 400 MHz)



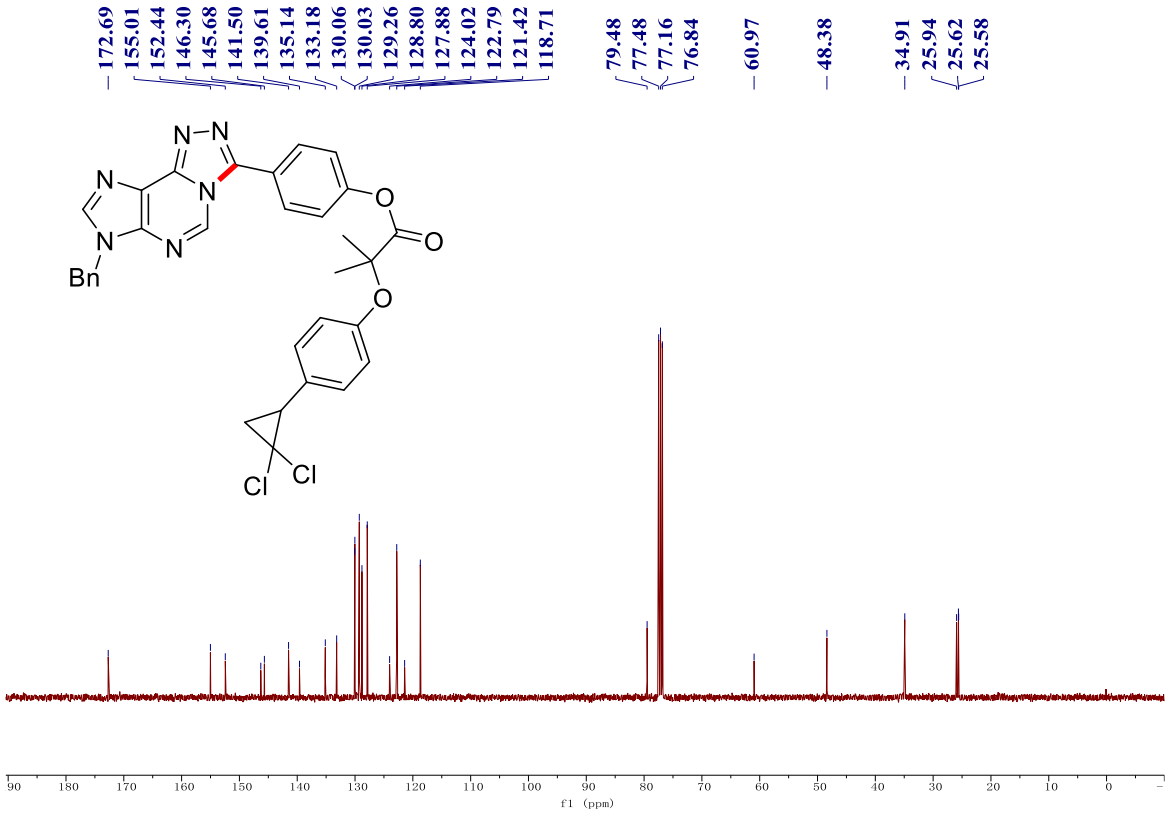
^{13}C NMR Spectrum of 2at at 25 °C (DMSO-d_6, 100 MHz)



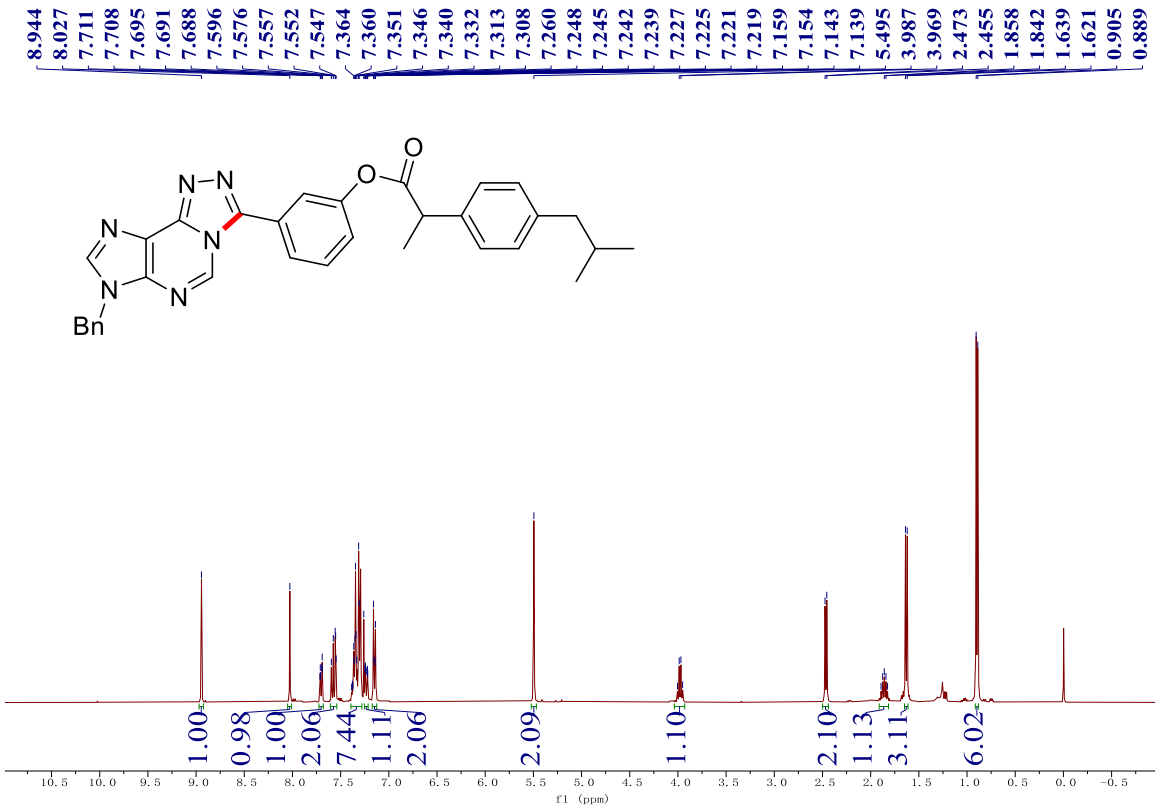
^{1}H NMR Spectrum of 2au at 25 °C ($CDCl_3$, 400 MHz)



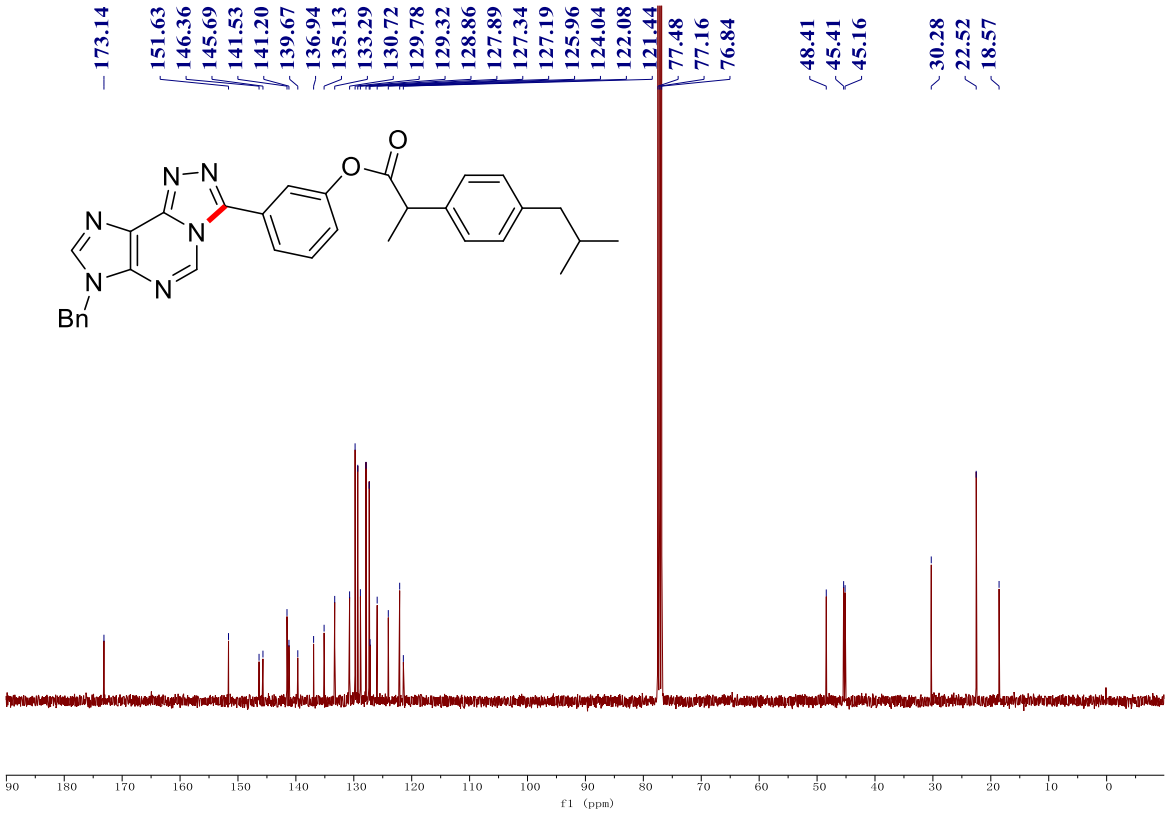
^{13}C NMR Spectrum of 2au at 25 °C ($CDCl_3$, 100 MHz)



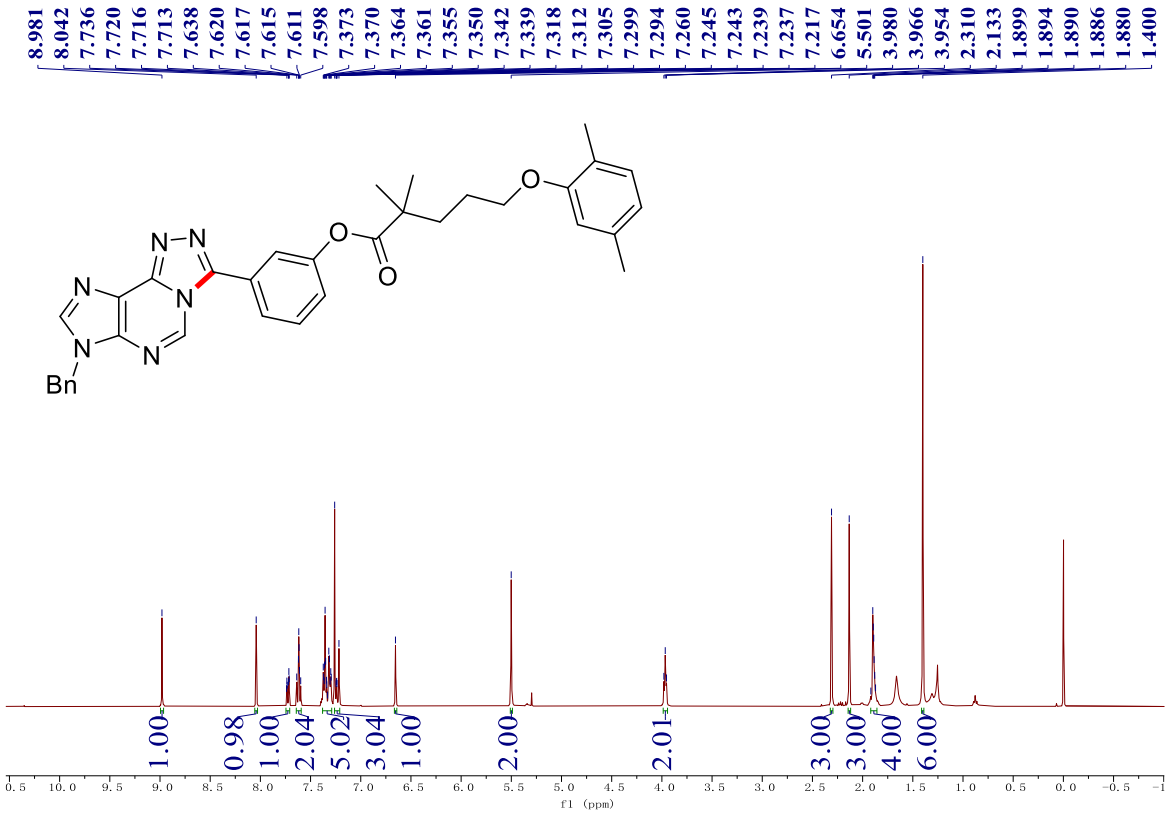
^{1}H NMR Spectrum of 2av at 25 °C ($CDCl_3$, 400 MHz)



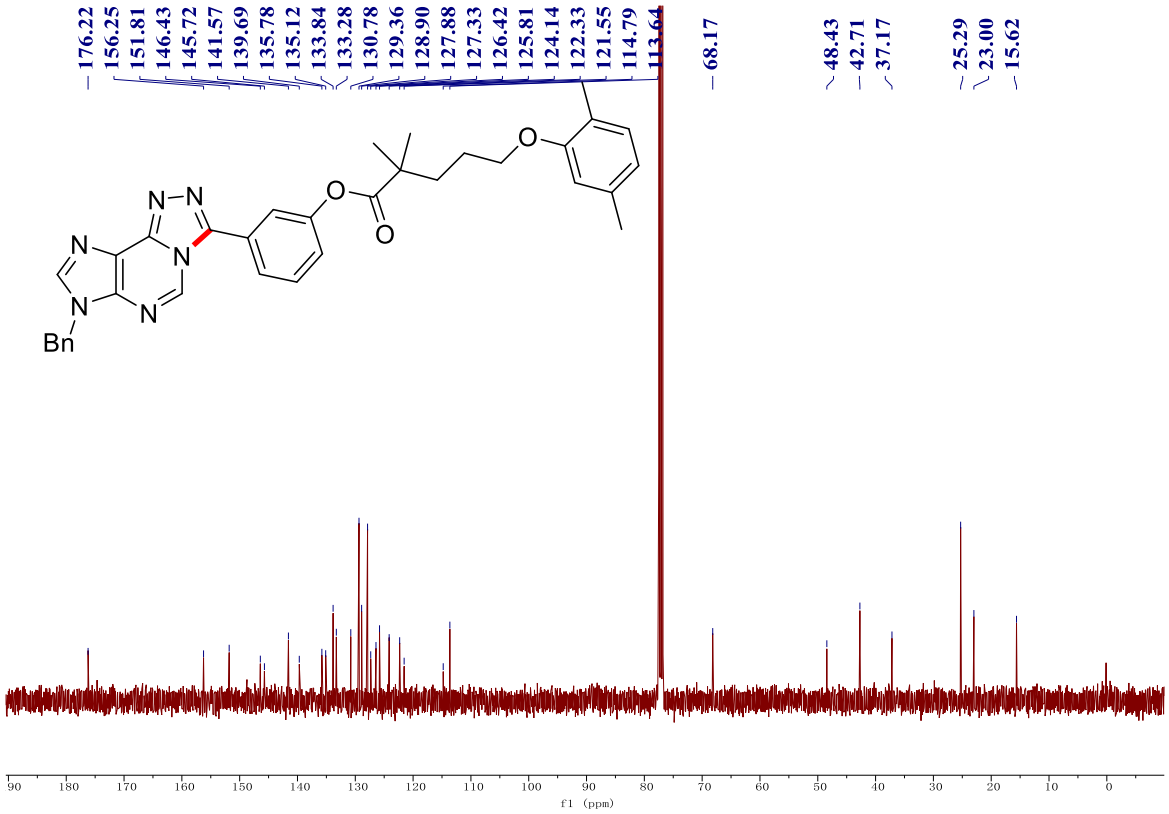
^{13}C NMR Spectrum of 2av at 25 °C ($CDCl_3$, 100 MHz)



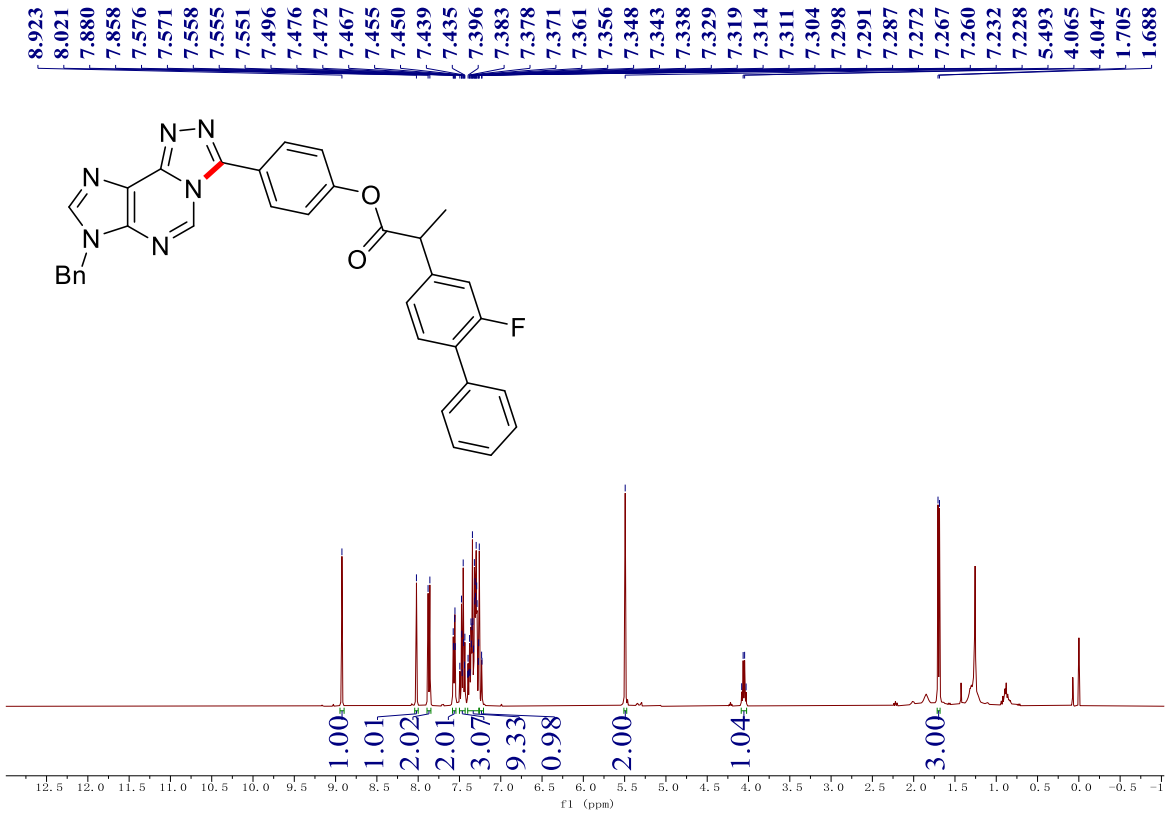
^{1}H NMR Spectrum of 2aw at 25 °C ($CDCl_3$, 400 MHz)



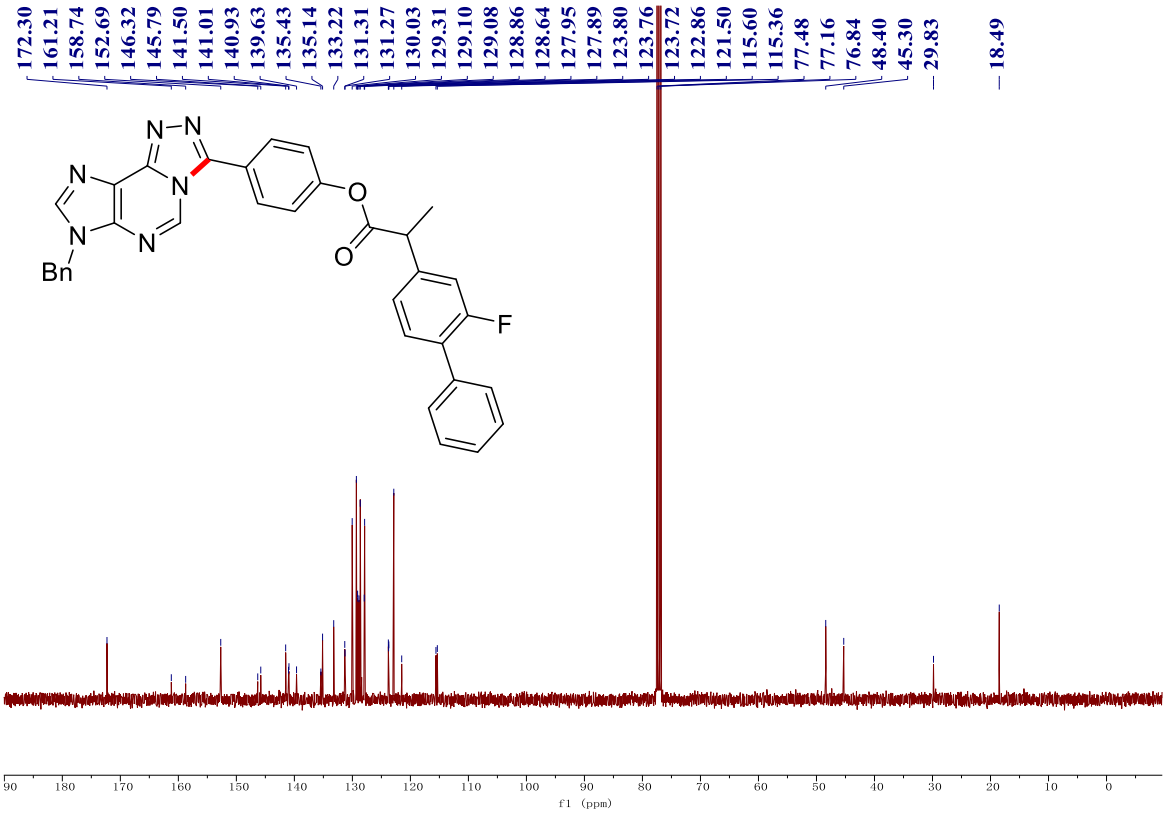
^{13}C NMR Spectrum of 2aw at 25 °C ($CDCl_3$, 100 MHz)



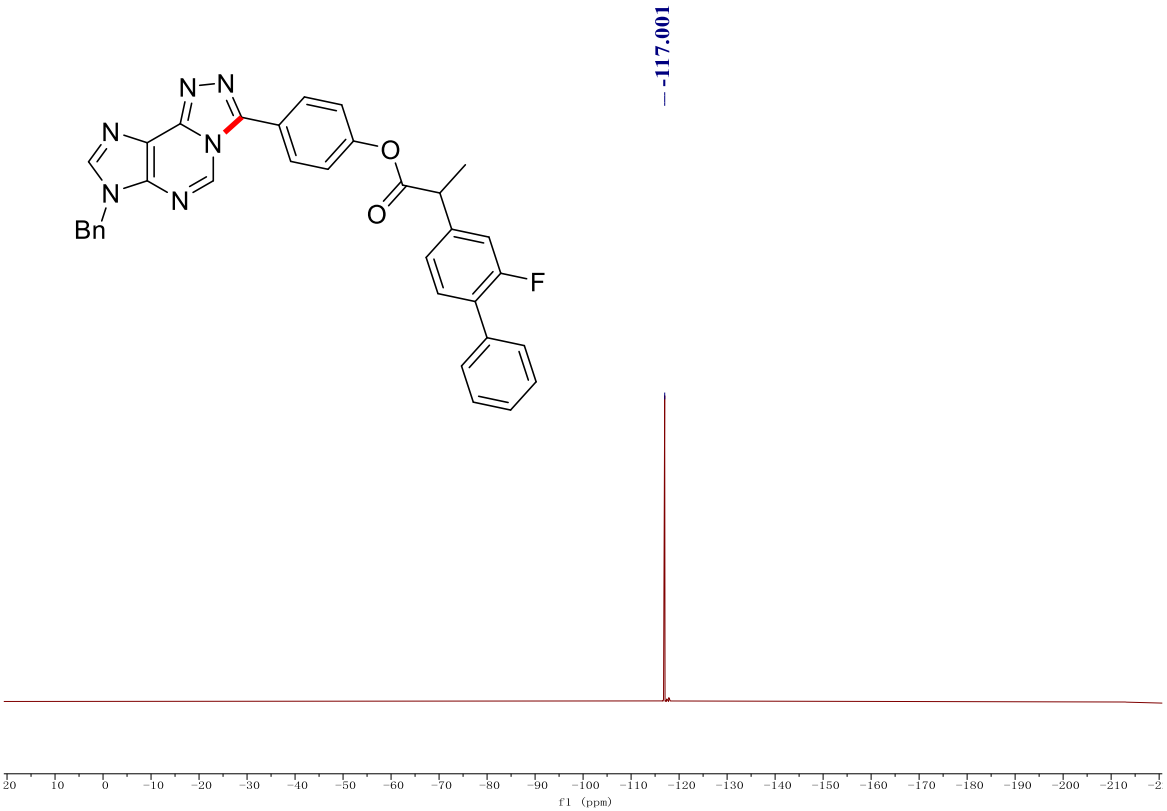
^{1}H NMR Spectrum of 2ax at 25 °C ($CDCl_3$, 400 MHz)



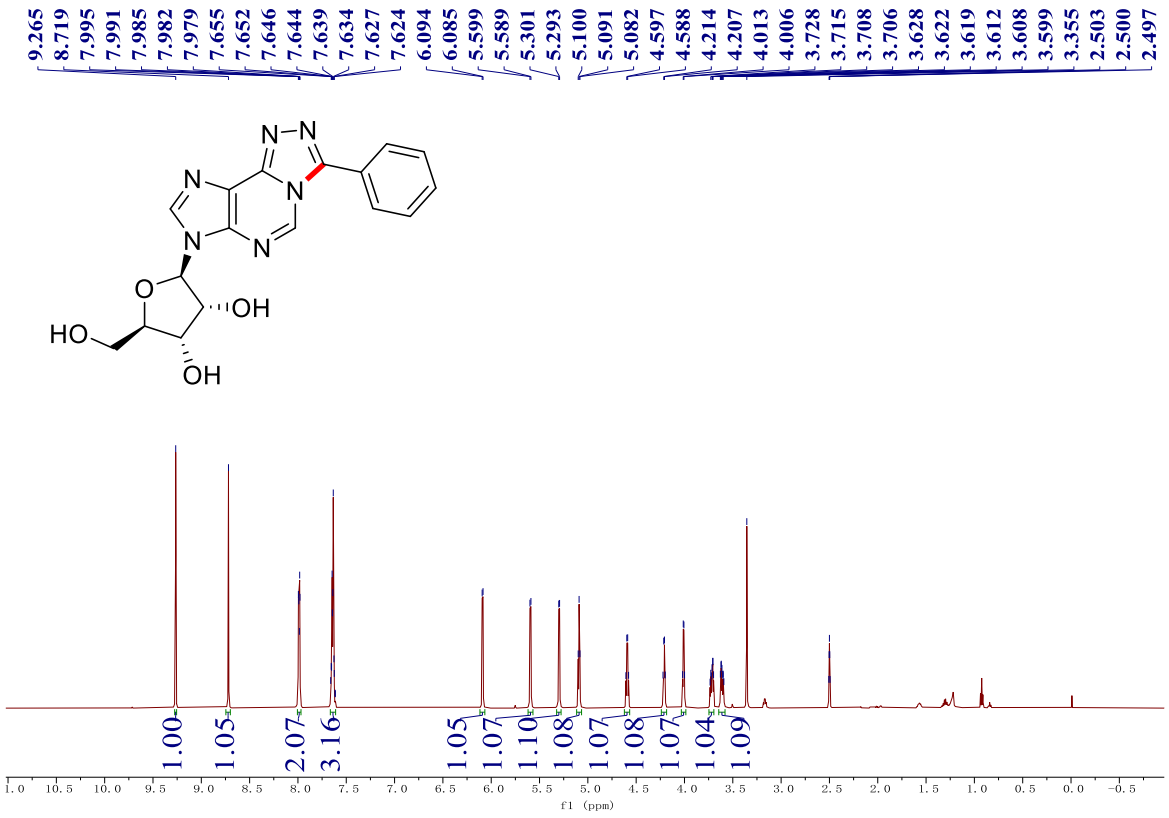
^{13}C NMR Spectrum of 2ax at 25 °C ($CDCl_3$, 100 MHz)



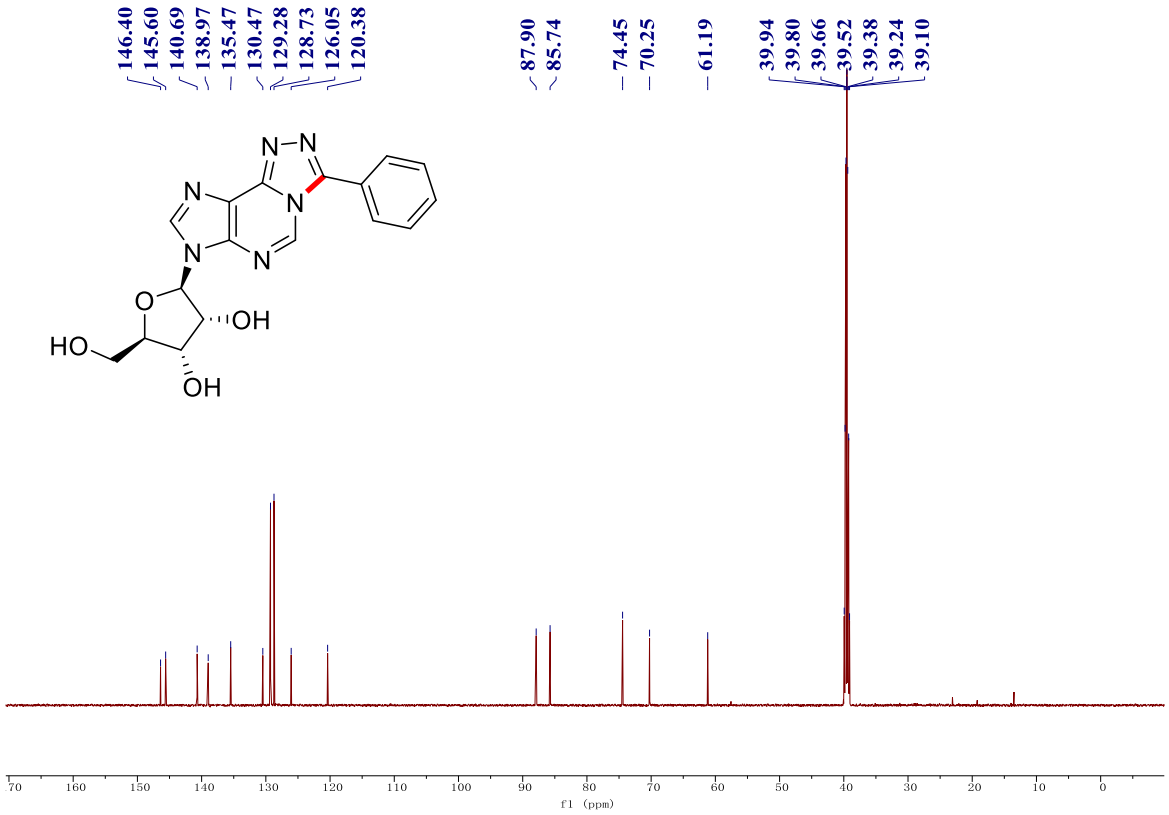
^{19}F NMR Spectrum of 2ax at 25 °C ($CDCl_3$, 376 MHz)



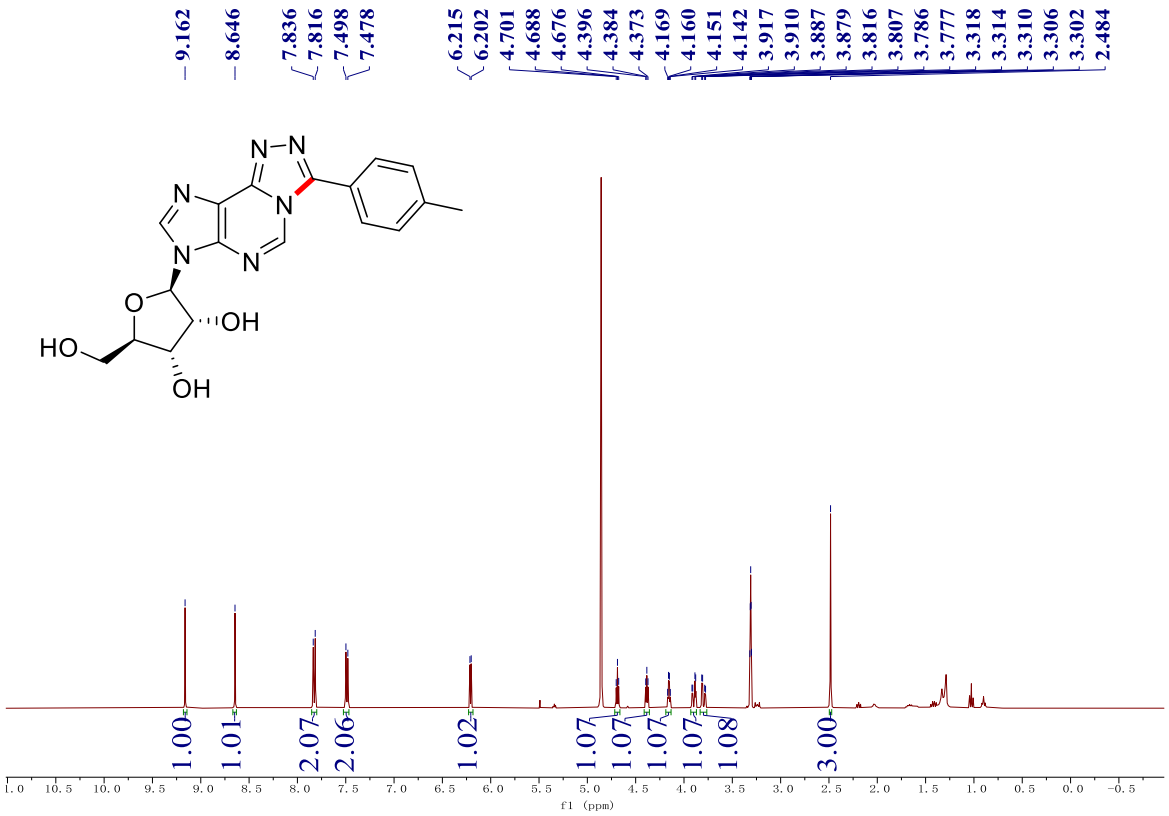
^{1}H NMR Spectrum of 2ay at 25 °C (DMSO-d_6, 600 MHz)



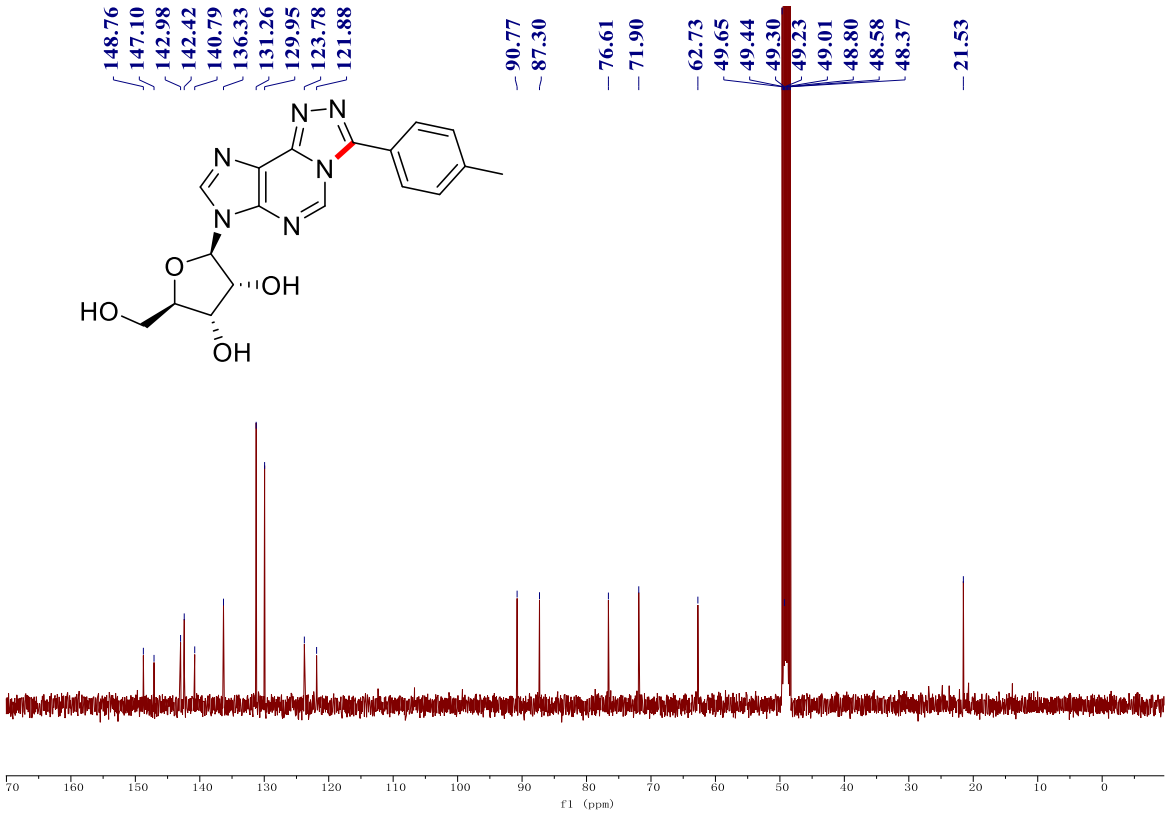
^{13}C NMR Spectrum of 2ay at 25 °C (DMSO-d_6, 150 MHz)



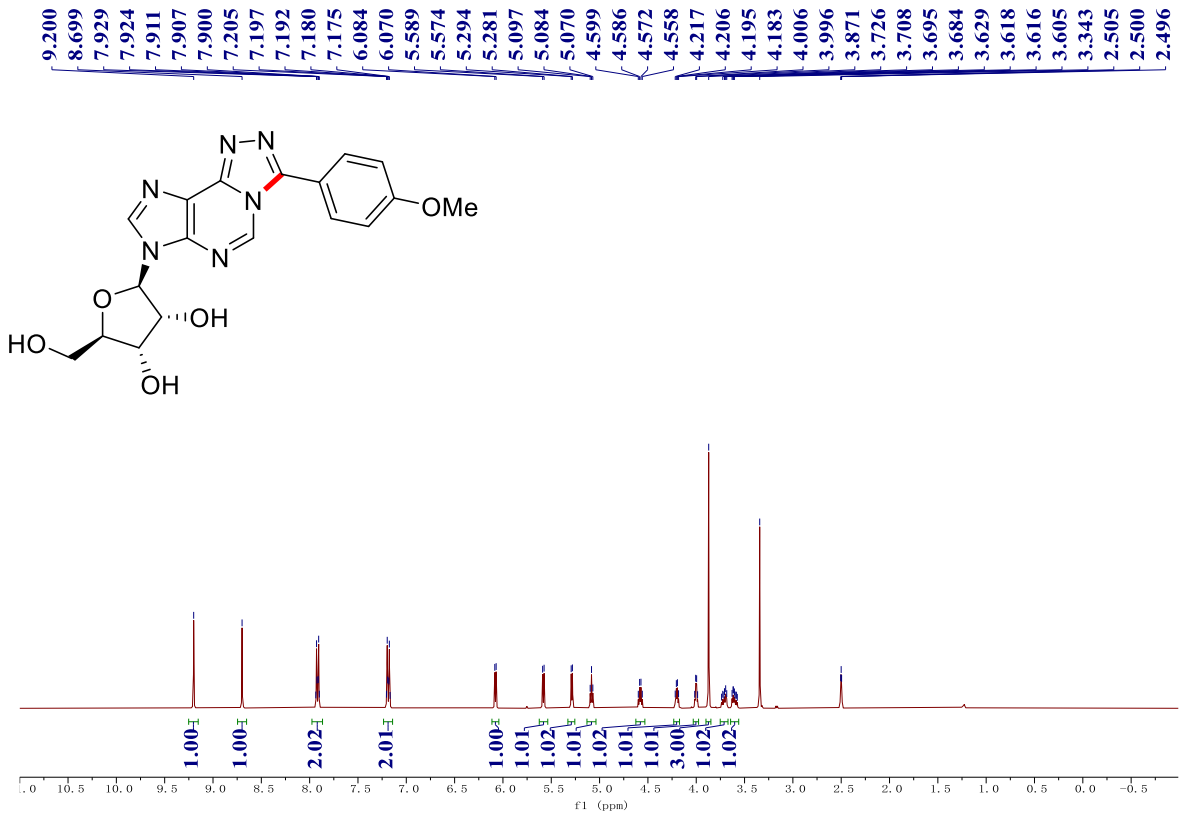
^{1}H NMR Spectrum of 2az at 25 °C (CD_3OD, 400 MHz)



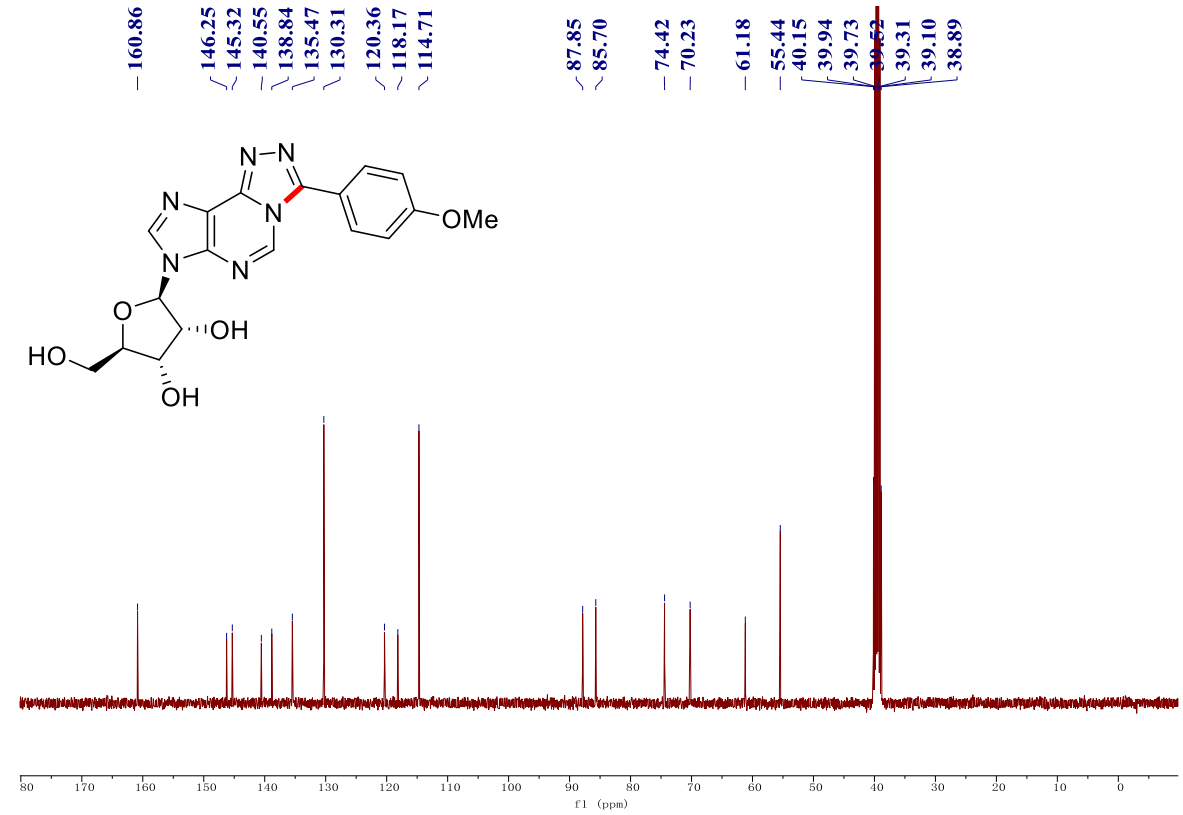
^{13}C NMR Spectrum of 2az at 25 °C (CD_3OD, 100 MHz)



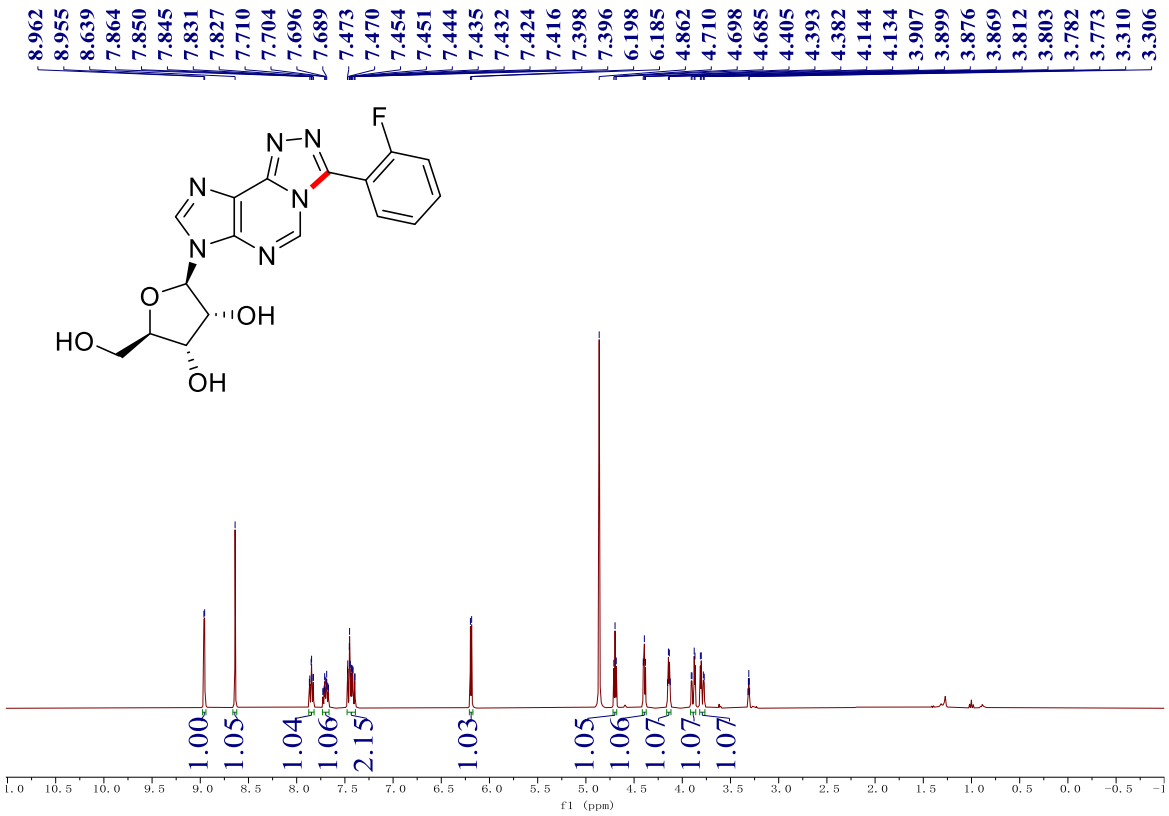
^{1}H NMR Spectrum of 2aaa at 25 °C (DMSO-d_6, 400 MHz)



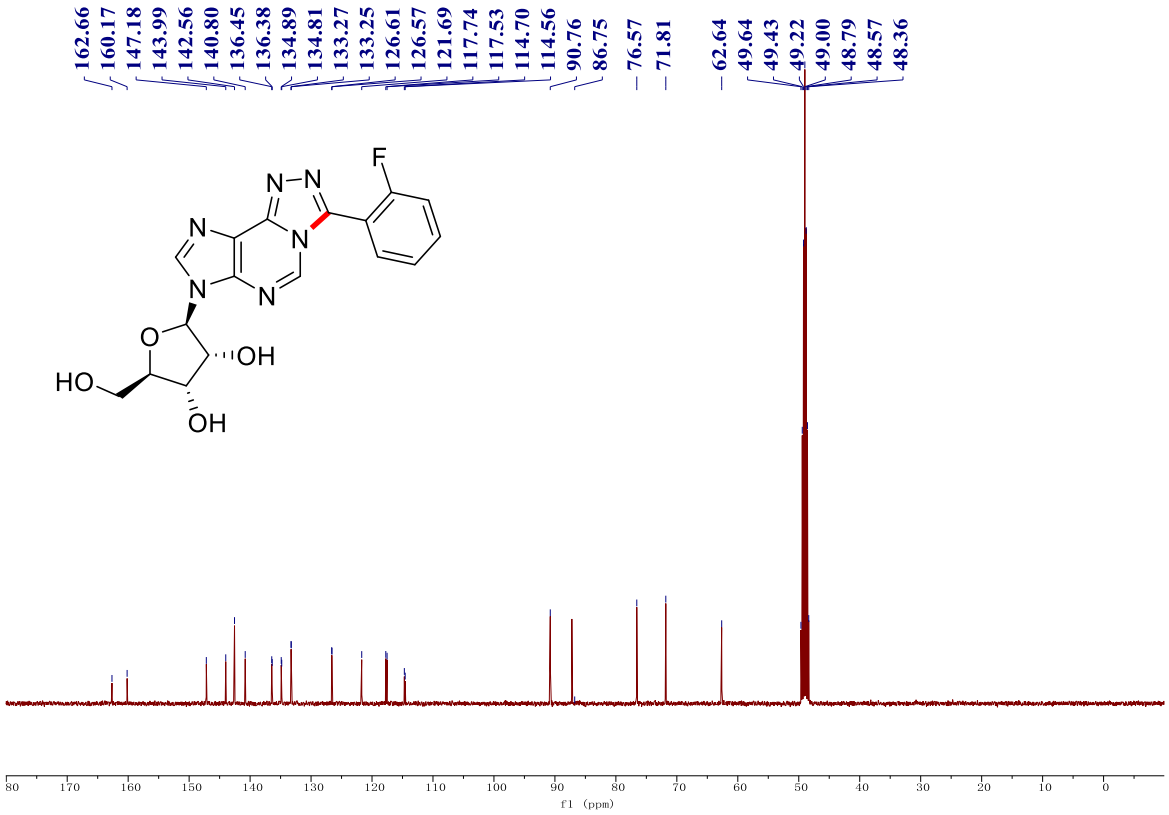
^{13}C NMR Spectrum of 2aaa at 25 °C (DMSO-d_6, 100 MHz)



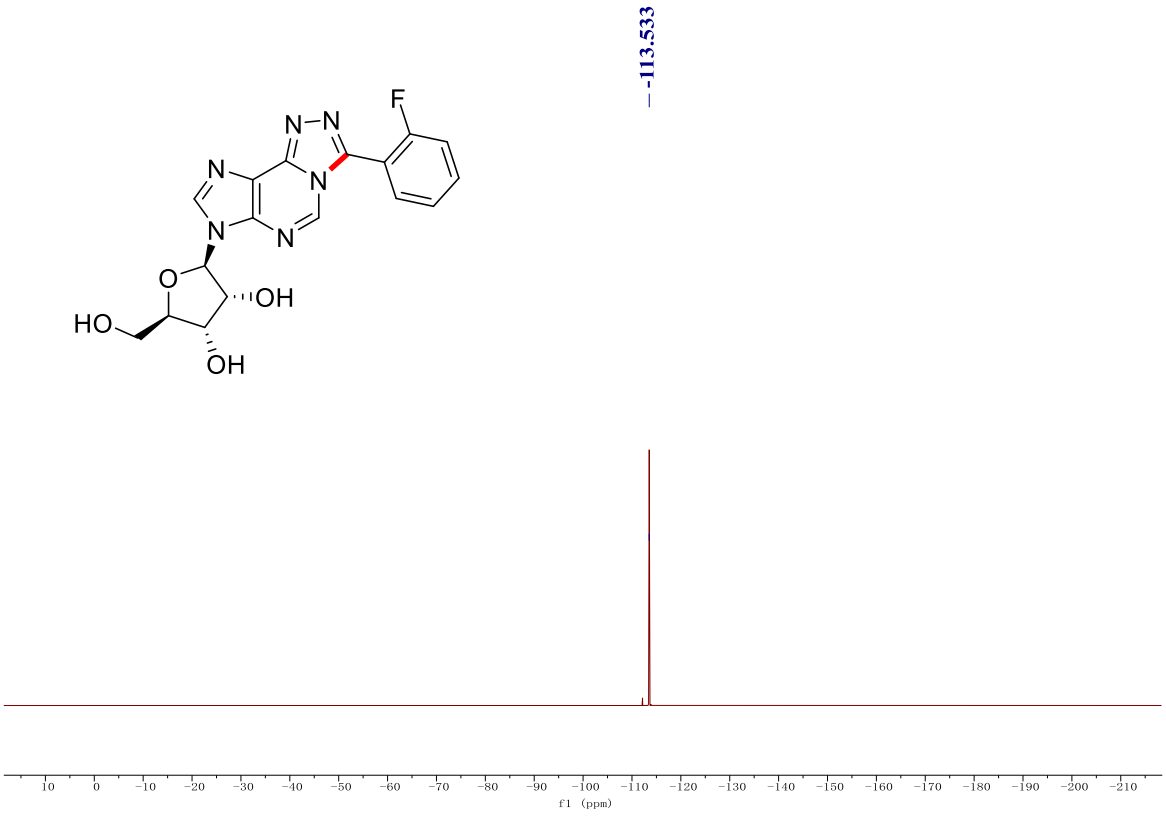
^{1}H NMR Spectrum of 2aab at 25 °C (CD_3OD, 400 MHz)



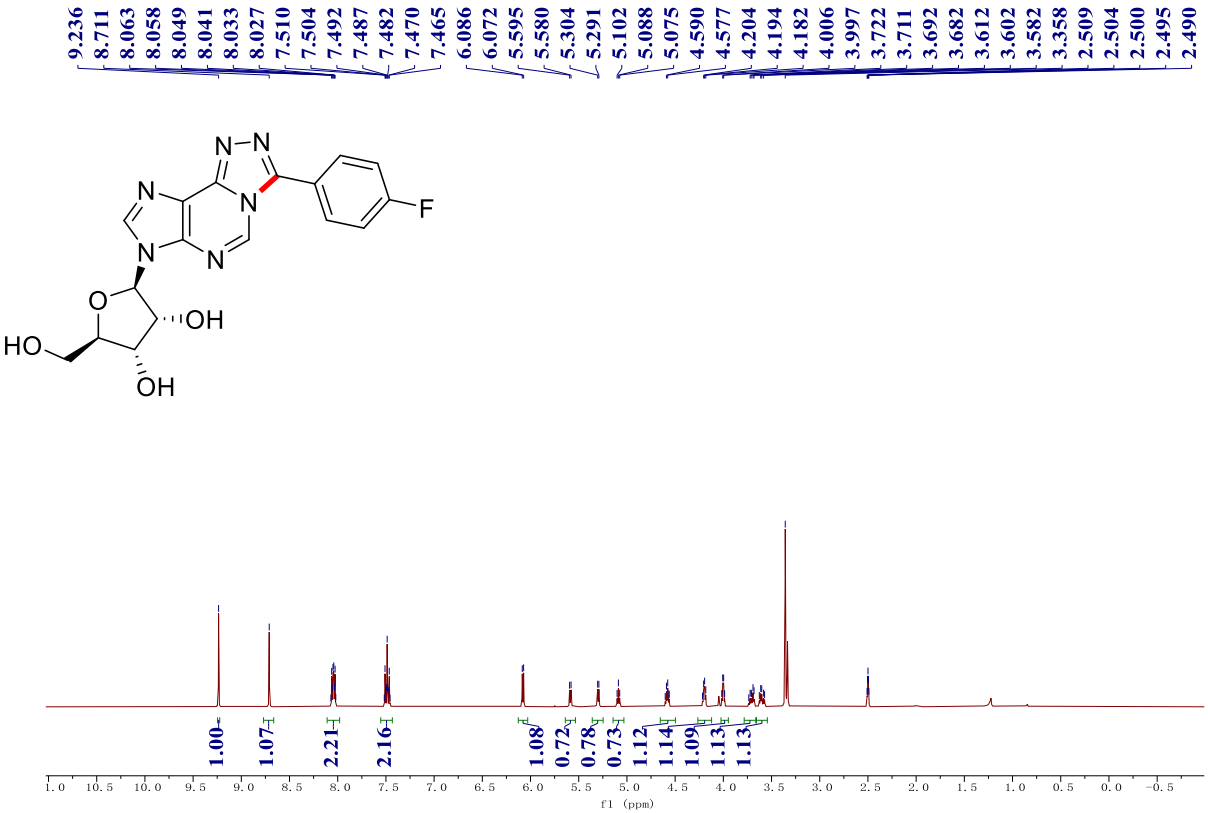
^{13}C NMR Spectrum of 2aab at 25 °C (CD_3OD, 100 MHz)



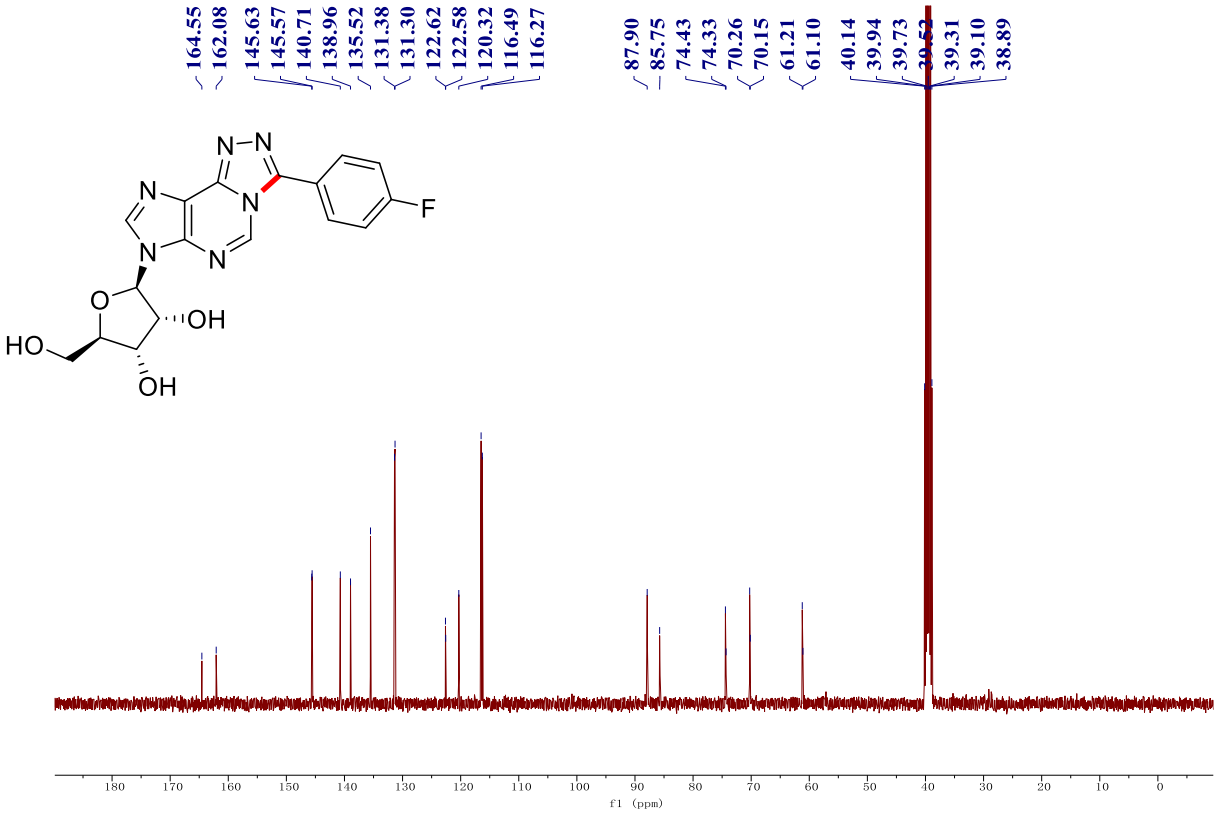
^{1}F NMR Spectrum of 2aab at 25 °C (CD_3OD, 376 MHz)



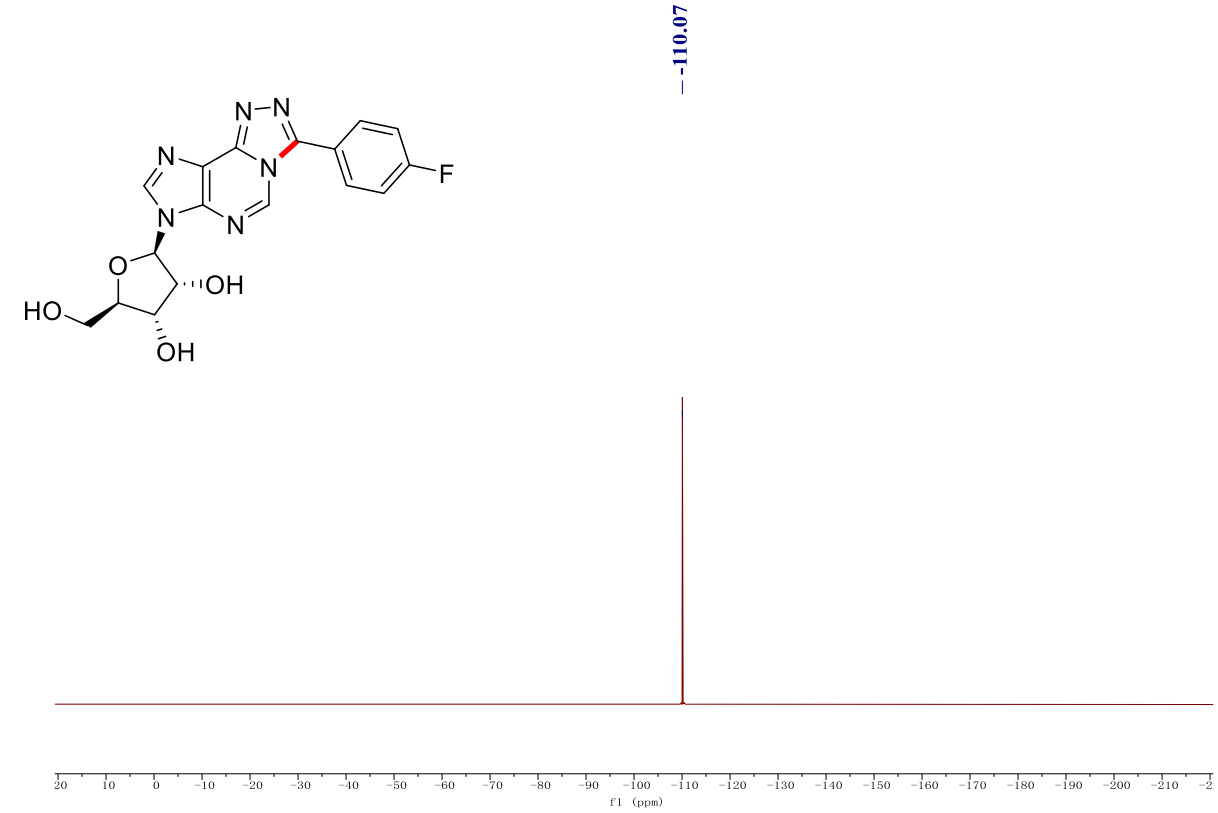
^{1}H NMR Spectrum of 2aac at 25 °C (DMSO-d_6, 400 MHz)



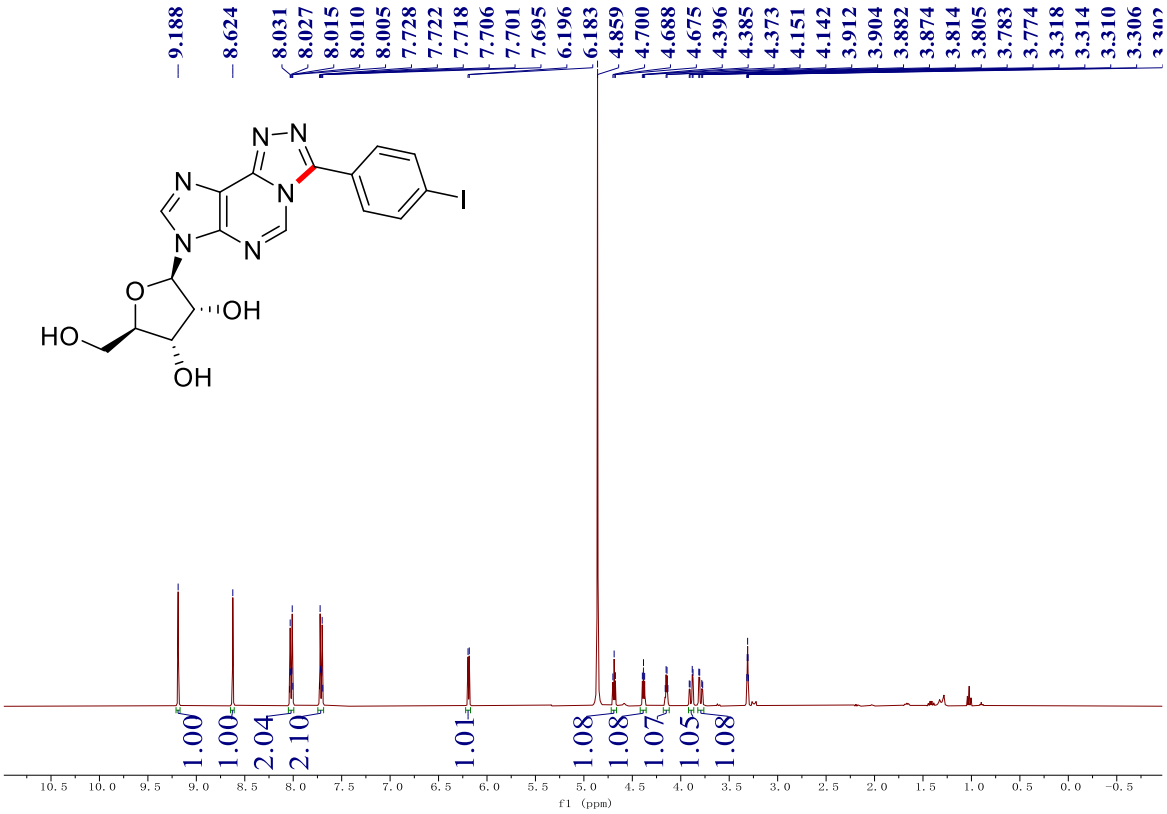
^{13}C NMR Spectrum of 2aac at 25 °C (DMSO-d_6, 100 MHz)



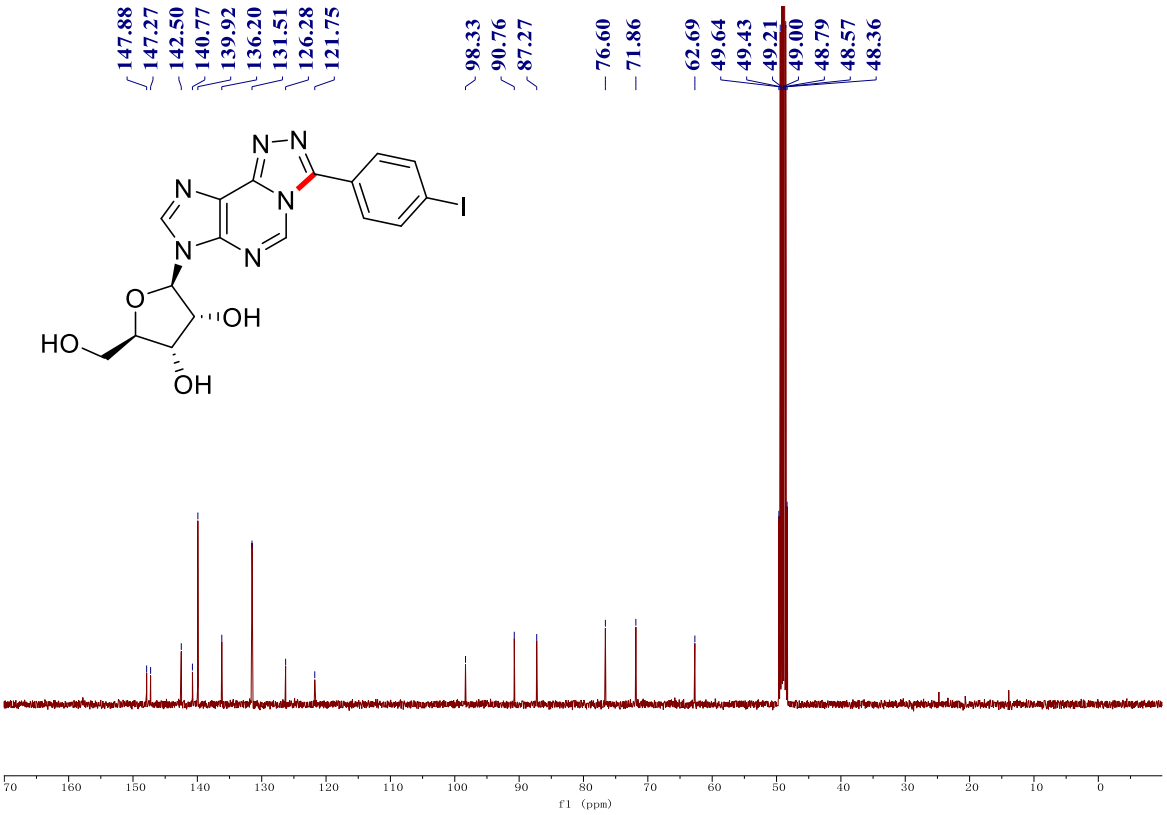
^{19}F NMR Spectrum of 2aac at 25 °C (DMSO-d_6, 376 MHz)



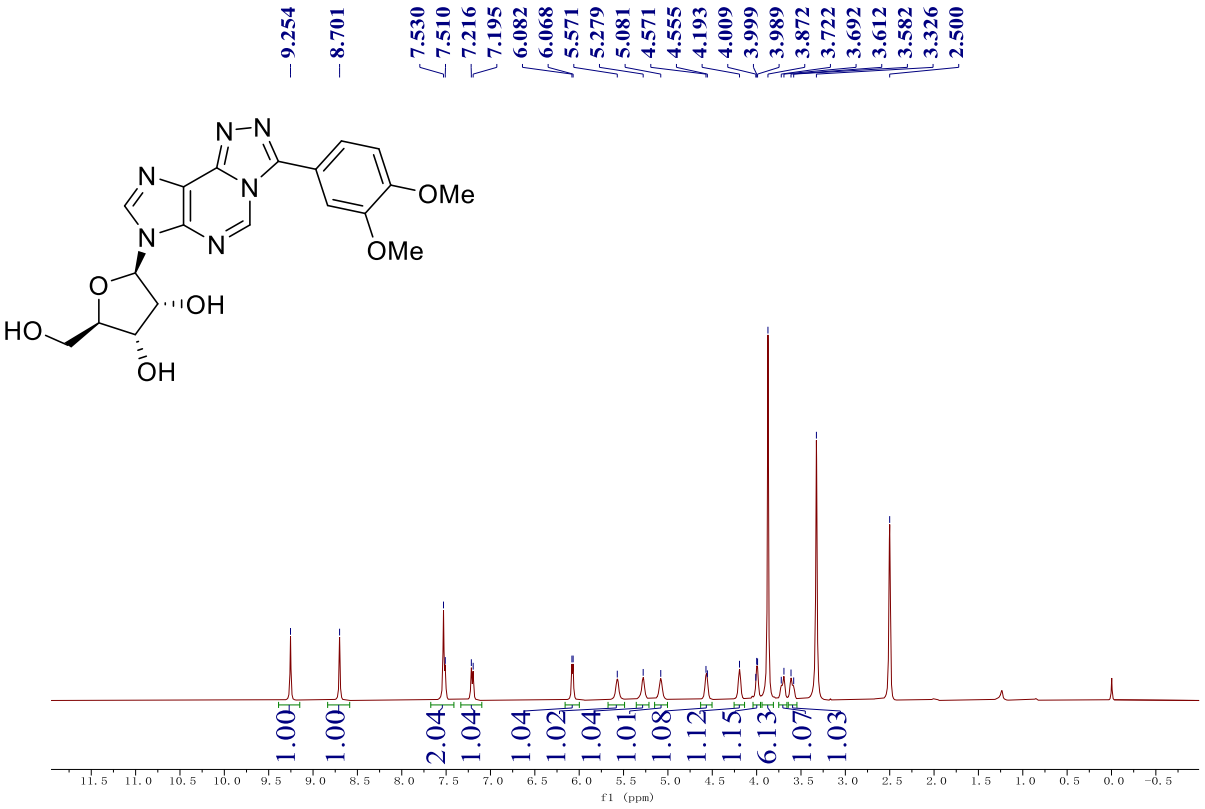
^{1}H NMR Spectrum of 2aad at 25 ^{o}C (CD_3OD, 400 MHz)



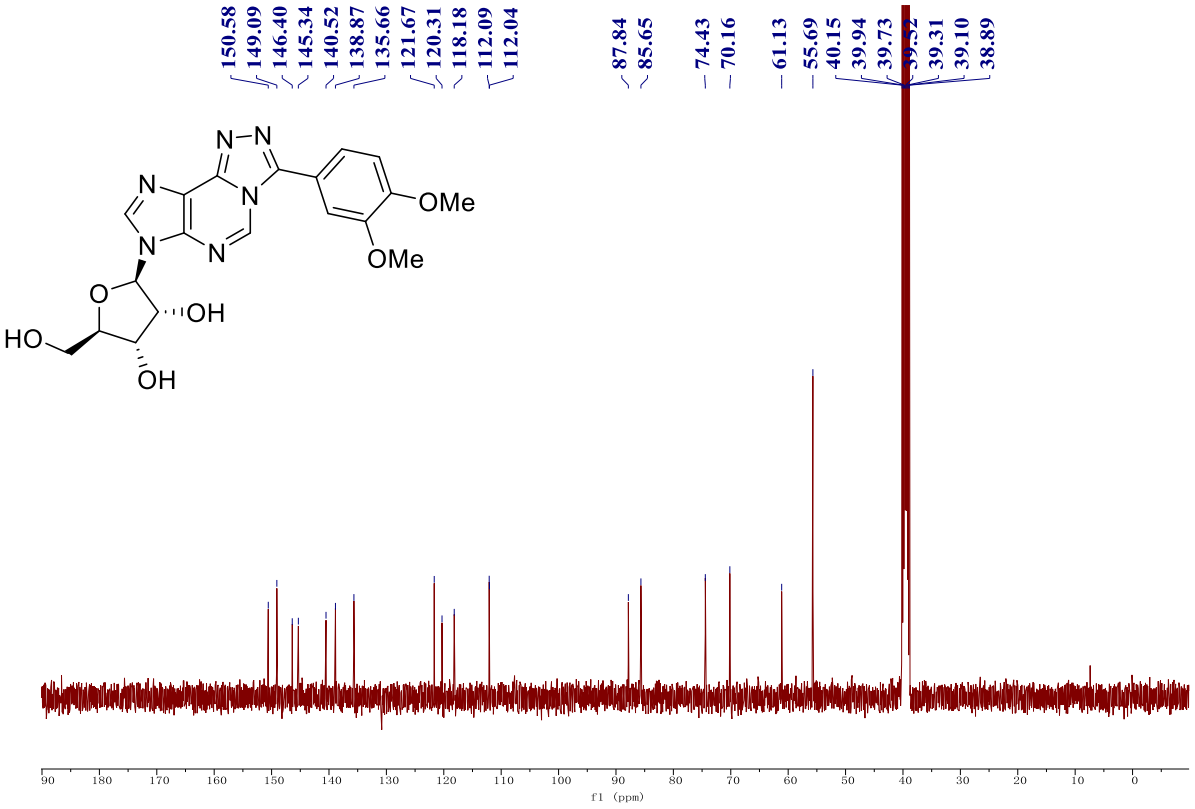
^{13}C NMR Spectrum of 2aad at 25 ^{o}C (CD_3OD, 100 MHz)



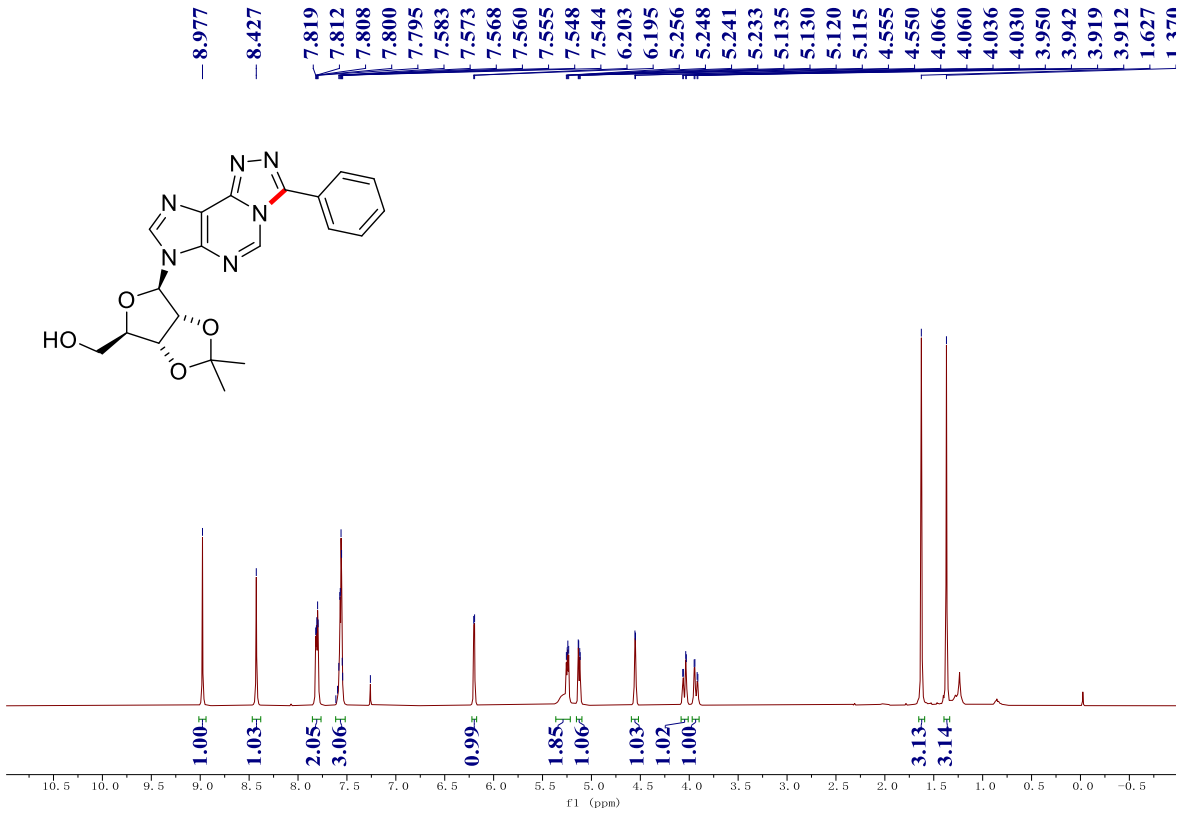
^{1}H NMR Spectrum of 2aae at 25 °C (DMSO-d_6, 400 MHz)



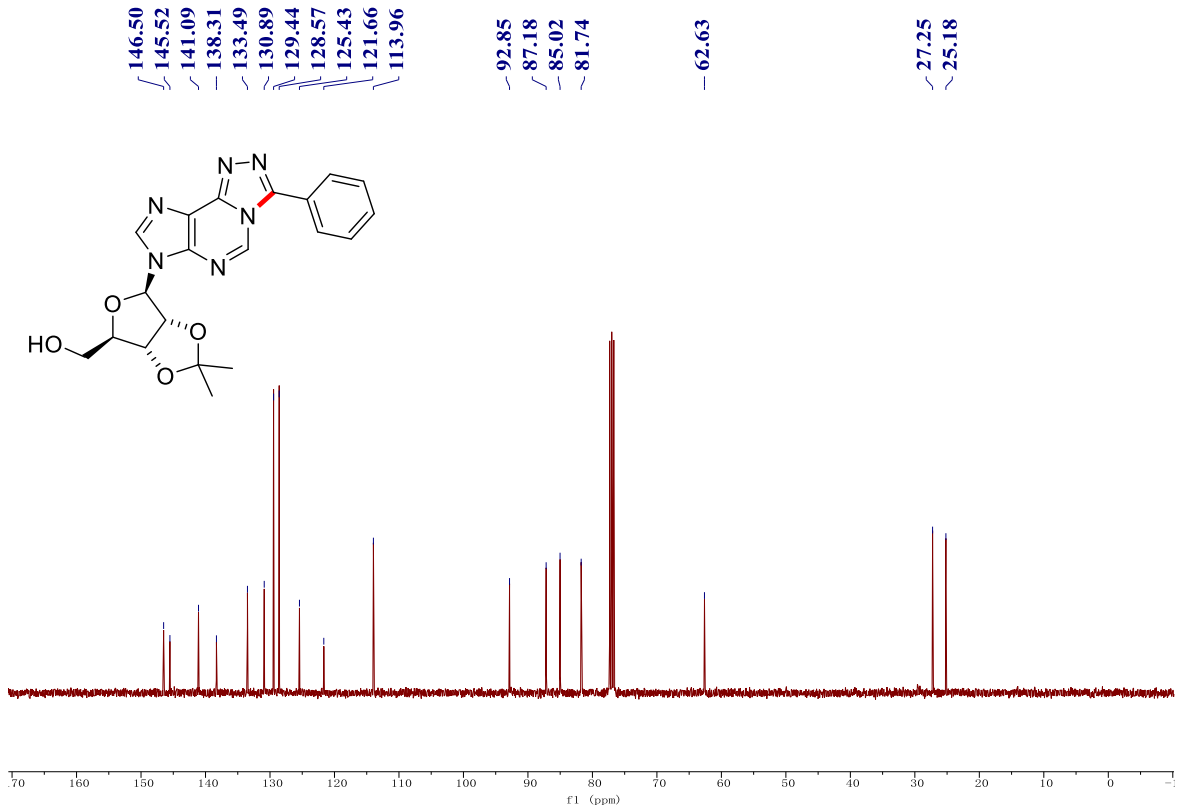
^{13}C NMR Spectrum of 2aae at 25 °C (DMSO-d_6, 100 MHz)



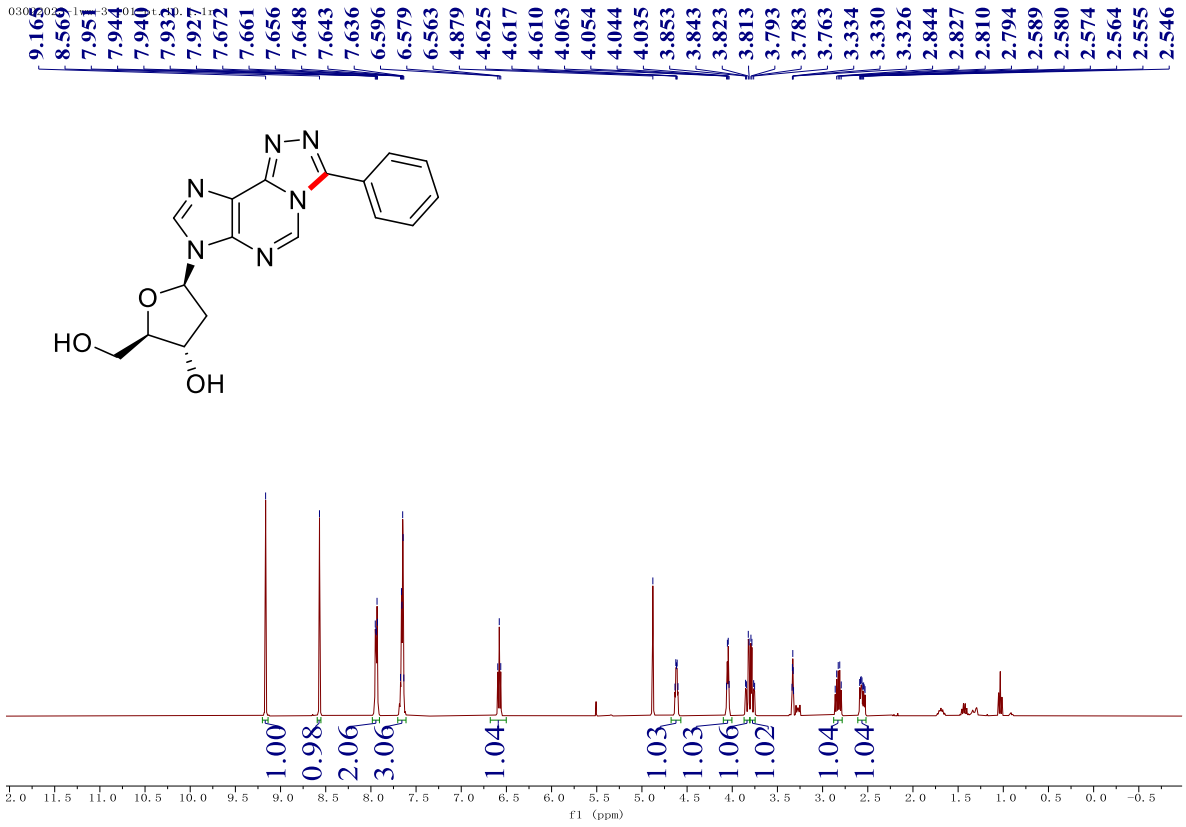
^{1}H NMR Spectrum of 2aaf at 25 °C ($CDCl_3$, 400 MHz)



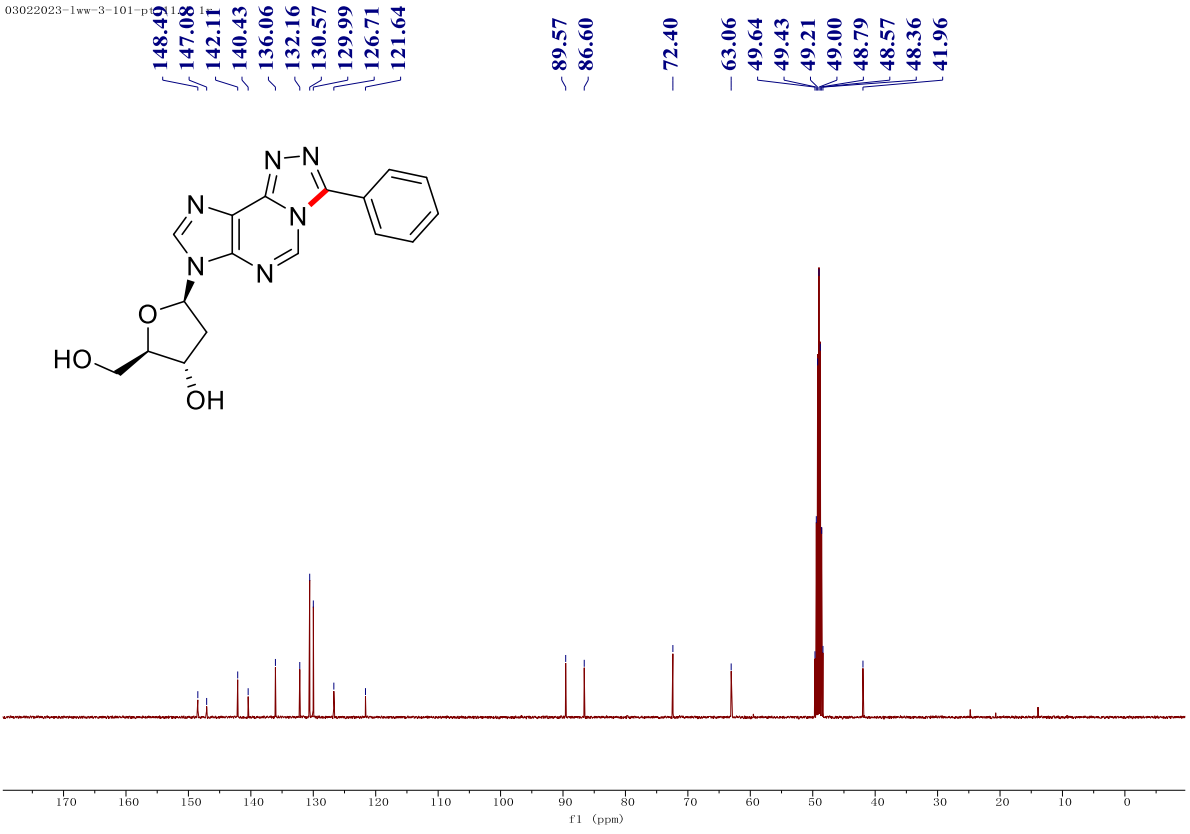
^{13}C NMR Spectrum of 2aaf at 25 °C ($CDCl_3$, 100 MHz)



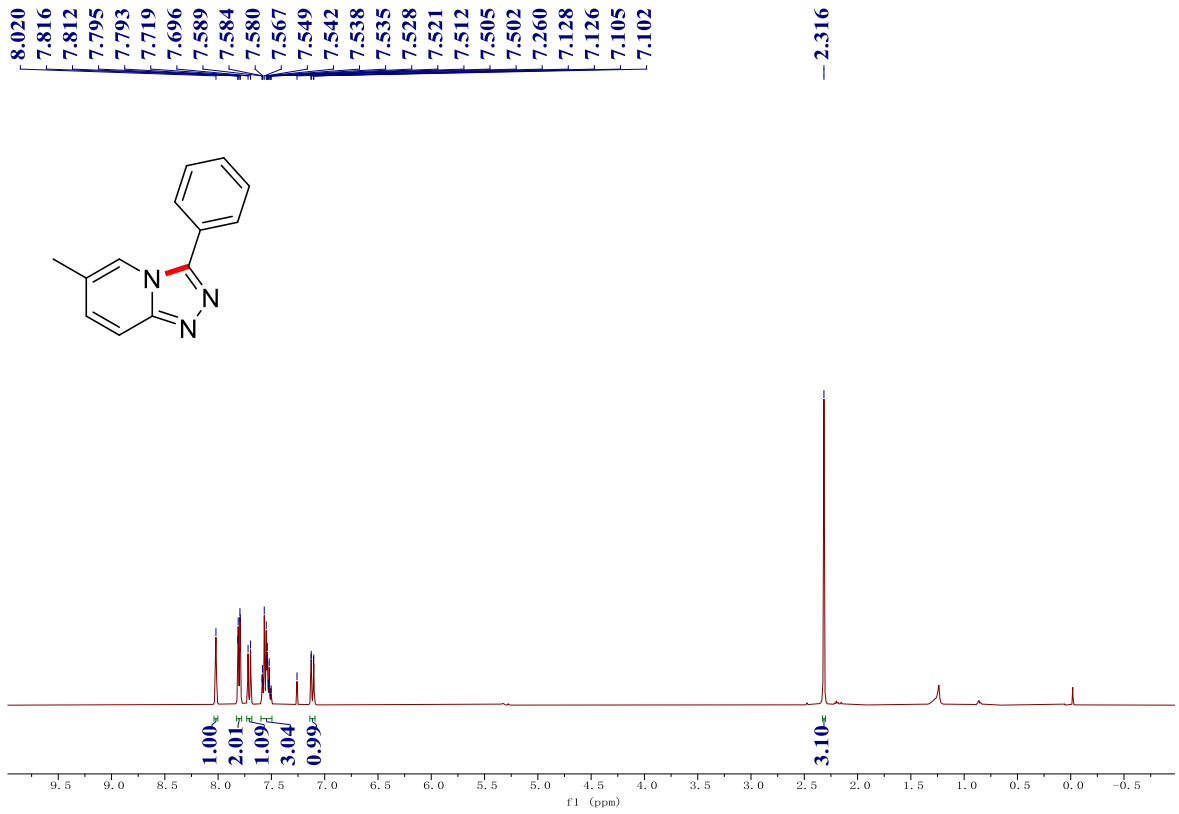
^{1}H NMR Spectrum of 2aag at 25 °C (CD_3OD, 400 MHz)



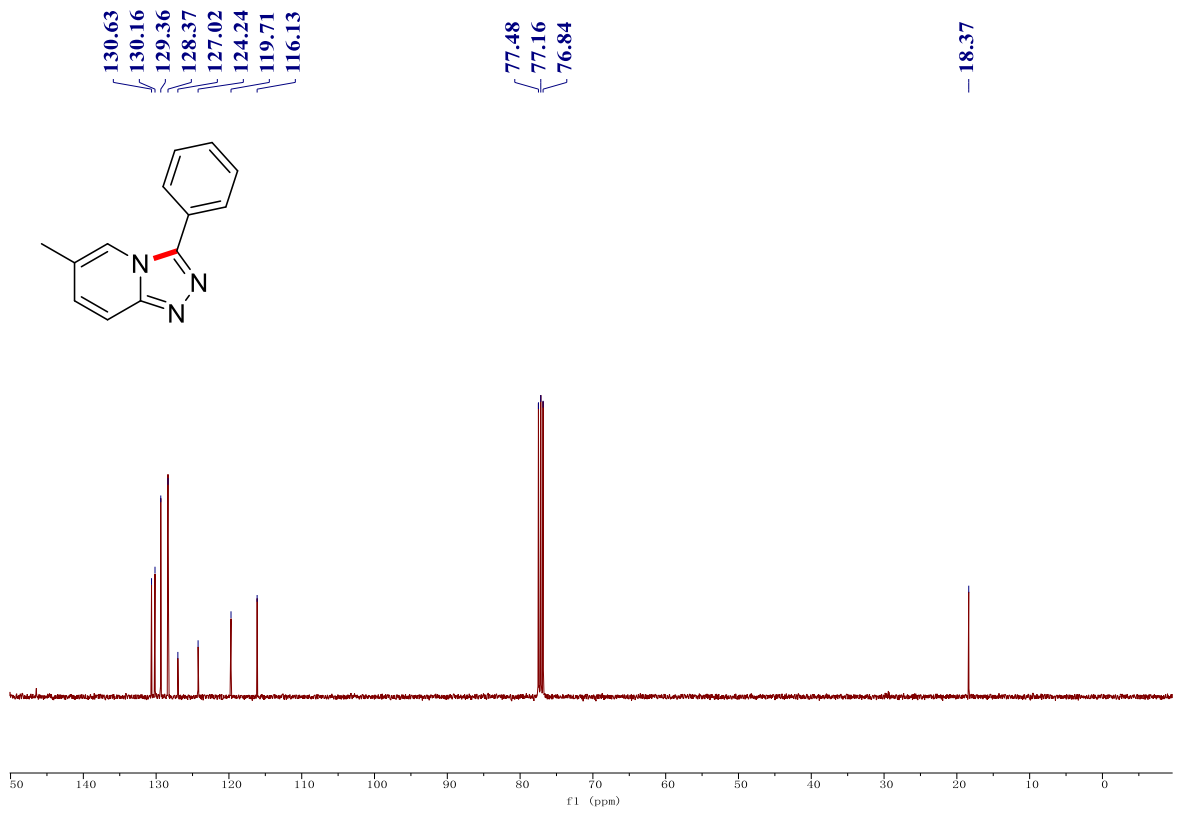
^{13}C NMR Spectrum of 2aag at 25 °C (CD_3OD, 100 MHz)



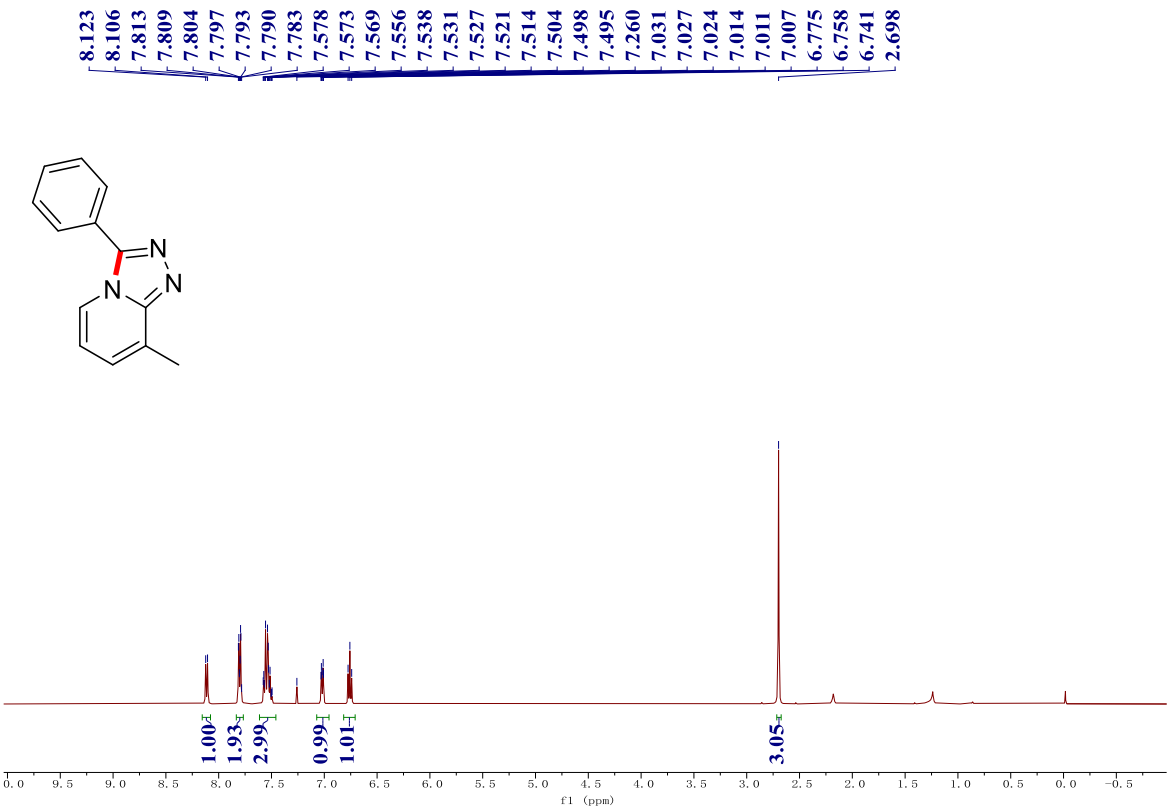
^{1}H NMR Spectrum of 2aah at 25 ^{o}C ($CDCl_3$, 400 MHz)



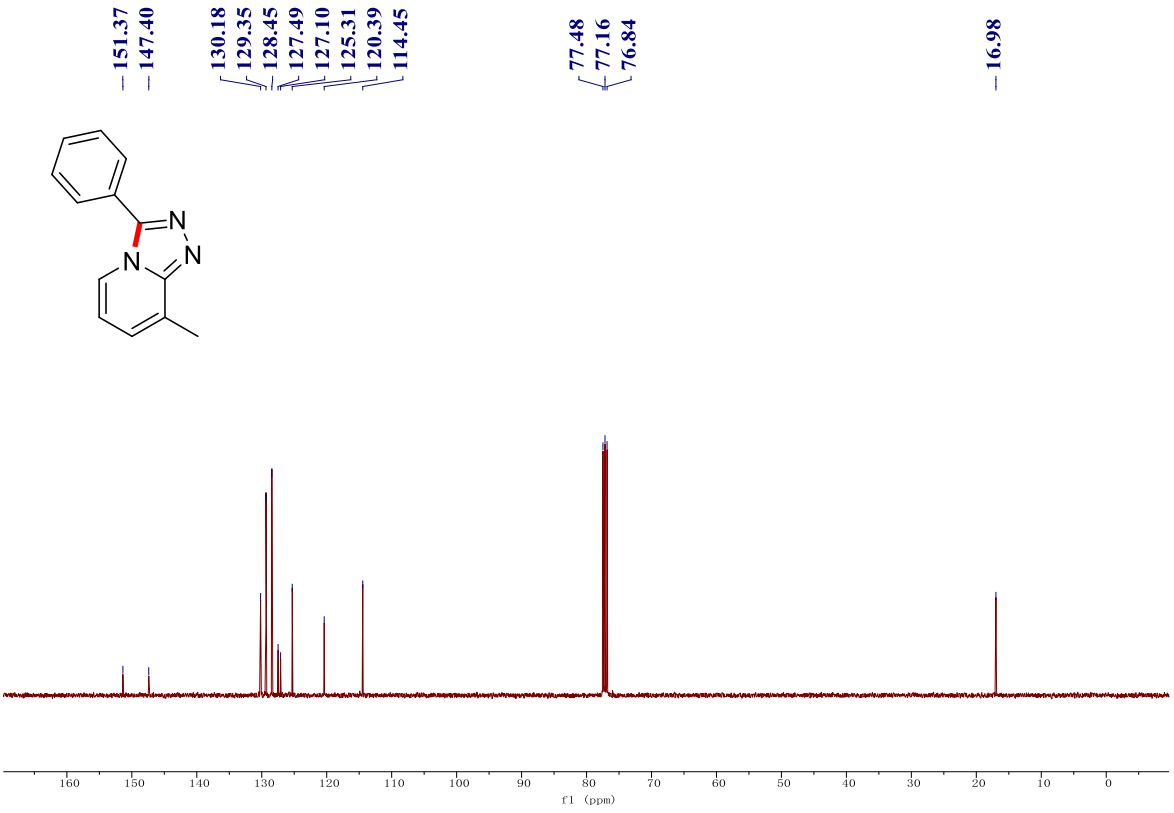
^{13}C NMR Spectrum of 2aah at 25 ^{o}C ($CDCl_3$, 100 MHz)



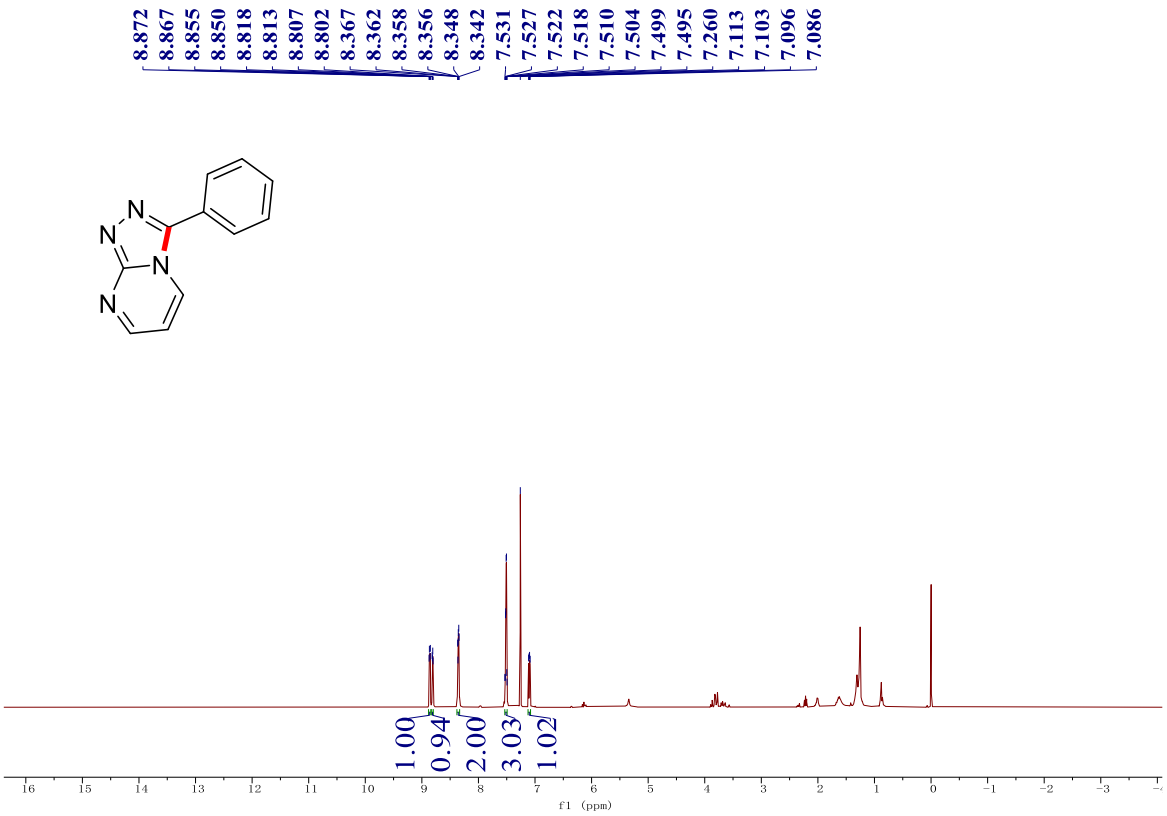
^{1}H NMR Spectrum of 2aai at 25 °C ($CDCl_3$, 400 MHz)



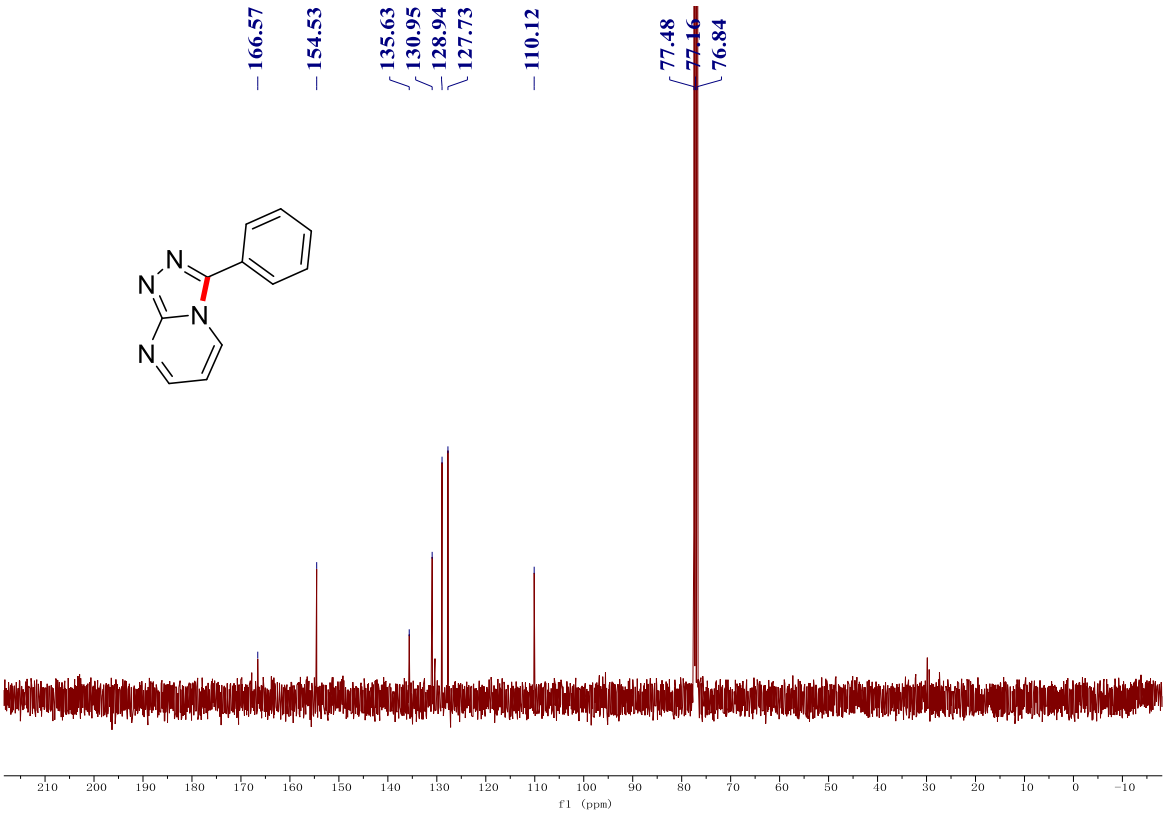
^{13}C NMR Spectrum of 2aai at 25 °C ($CDCl_3$, 100 MHz)



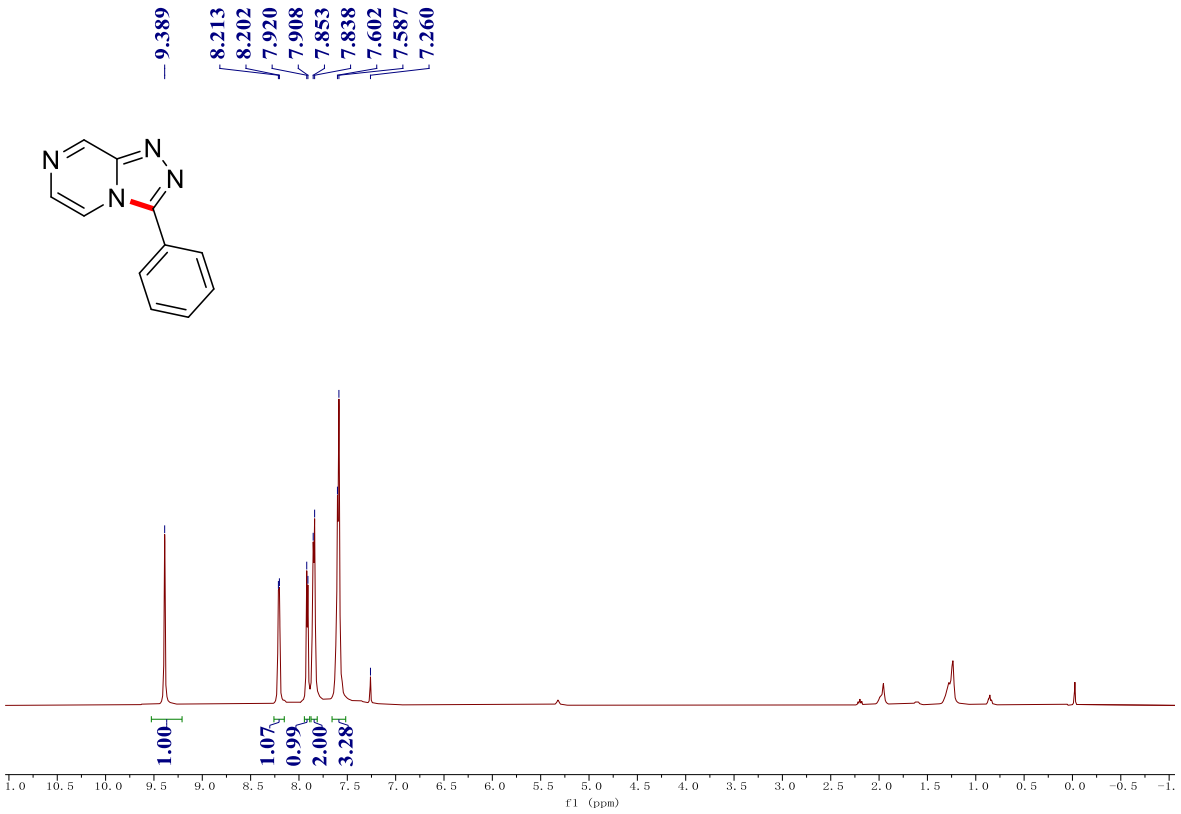
^{1}H NMR Spectrum of 2aaj at 25 ^{o}C ($CDCl_3$, 400 MHz)



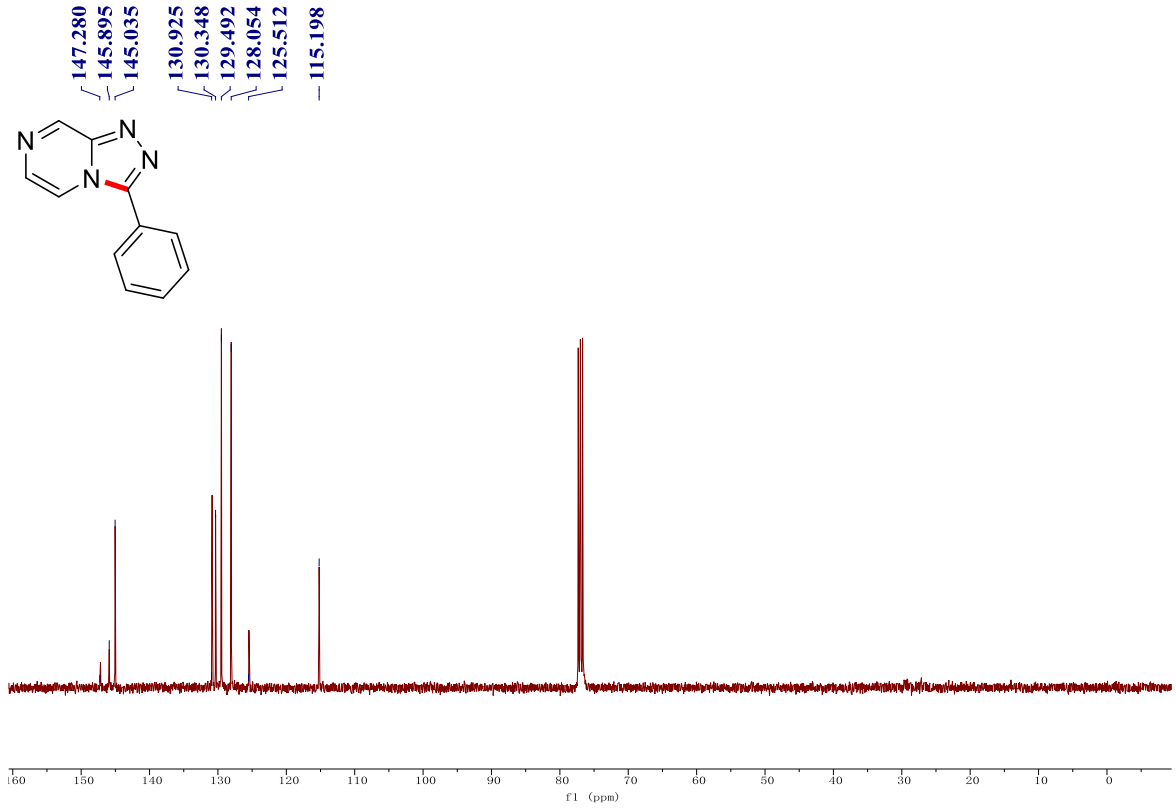
^{13}C NMR Spectrum of 2aaj at 25 ^{o}C ($CDCl_3$, 100 MHz)



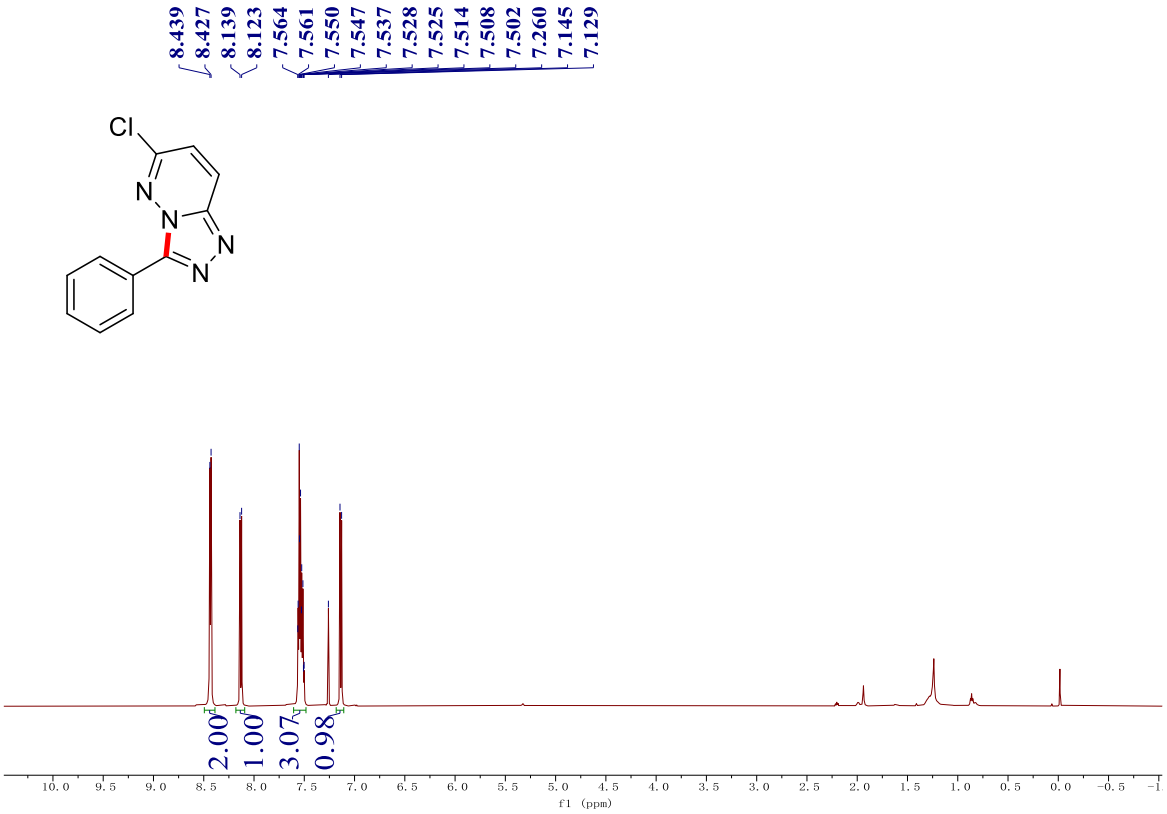
^{1}H NMR Spectrum of 2aak at 25 ^{o}C ($CDCl_3$, 400 MHz)



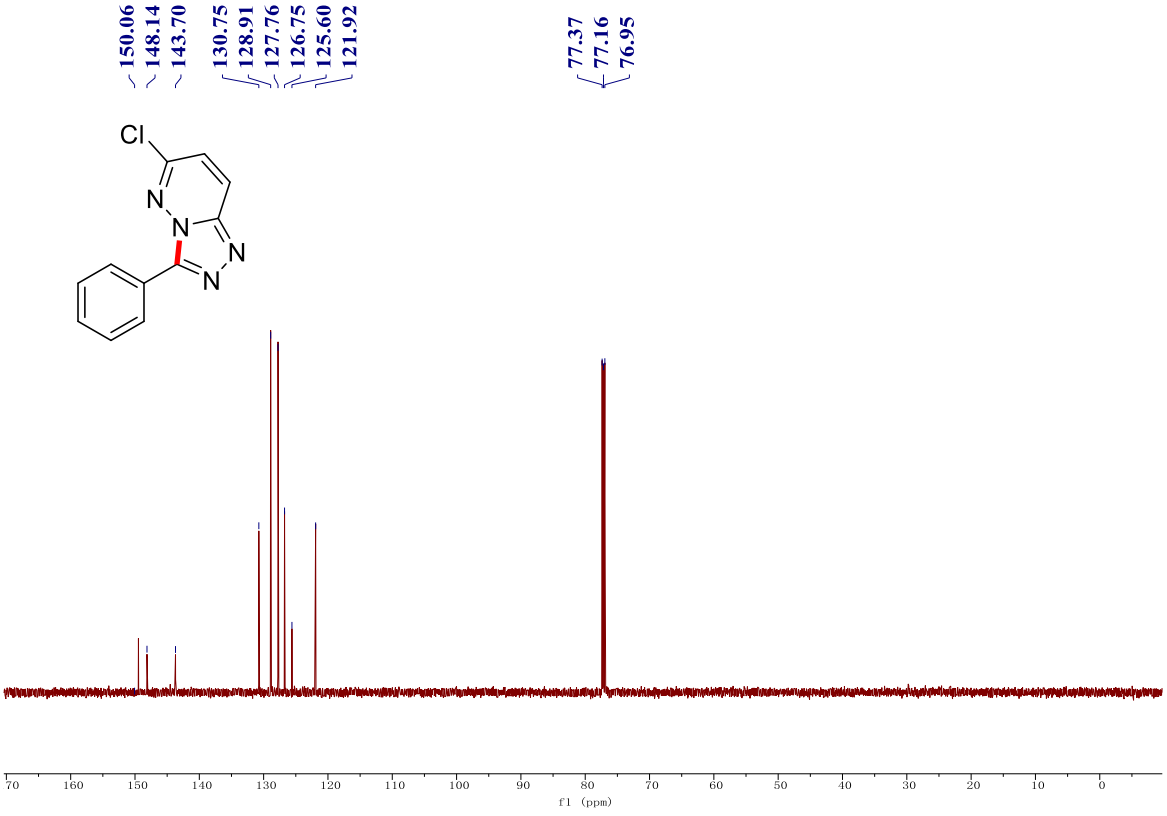
^{13}C NMR Spectrum of 2aak at 25 ^{o}C ($CDCl_3$, 100 MHz)



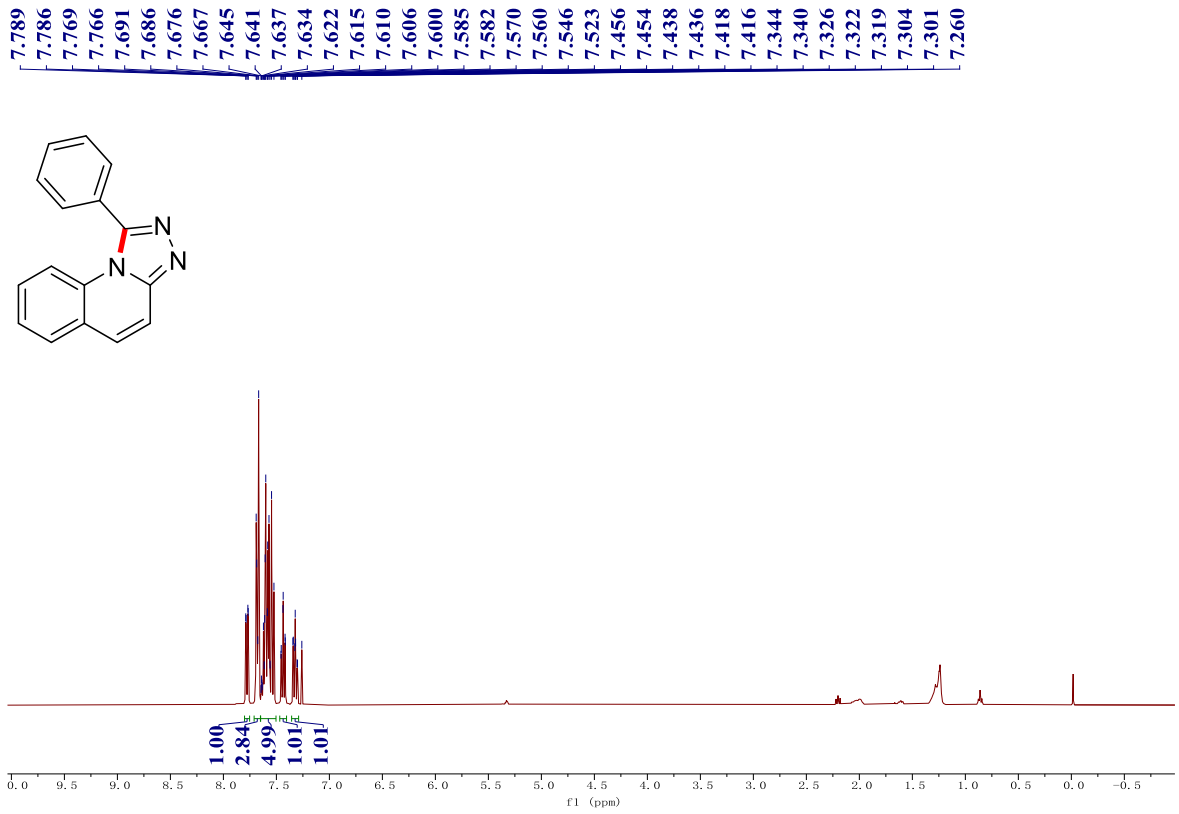
^{1}H NMR Spectrum of 2aal at 25 °C ($CDCl_3$, 600 MHz)



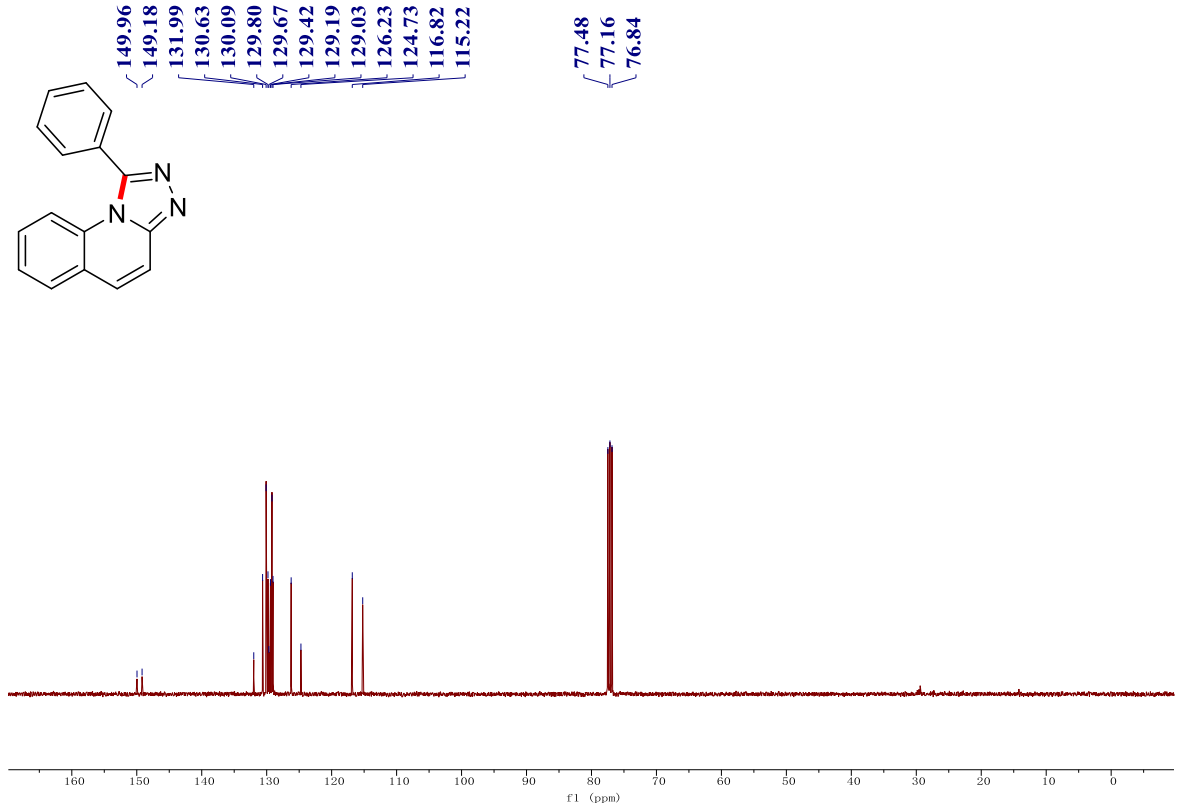
^{13}C NMR Spectrum of 2aal at 25 °C ($CDCl_3$, 150 MHz)



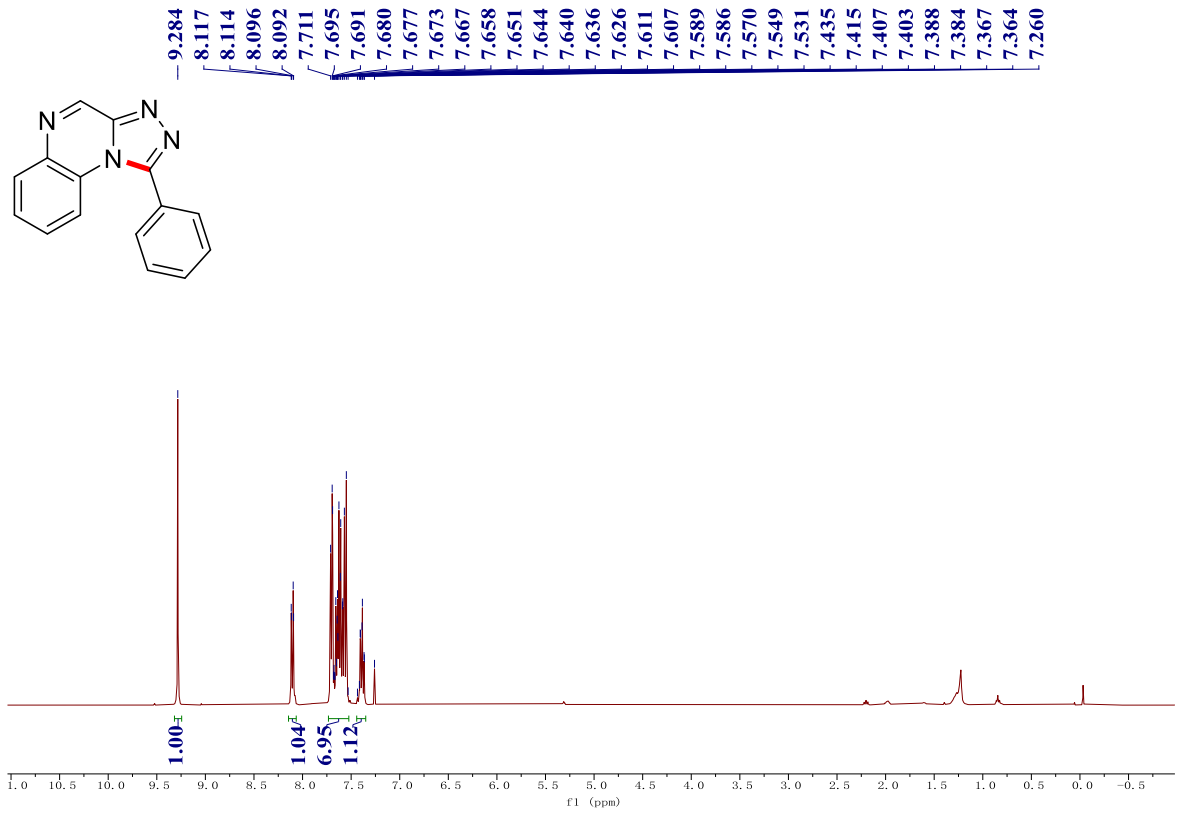
^{1}H NMR Spectrum of 2aam at 25 ^{o}C ($CDCl_3$, 400 MHz)



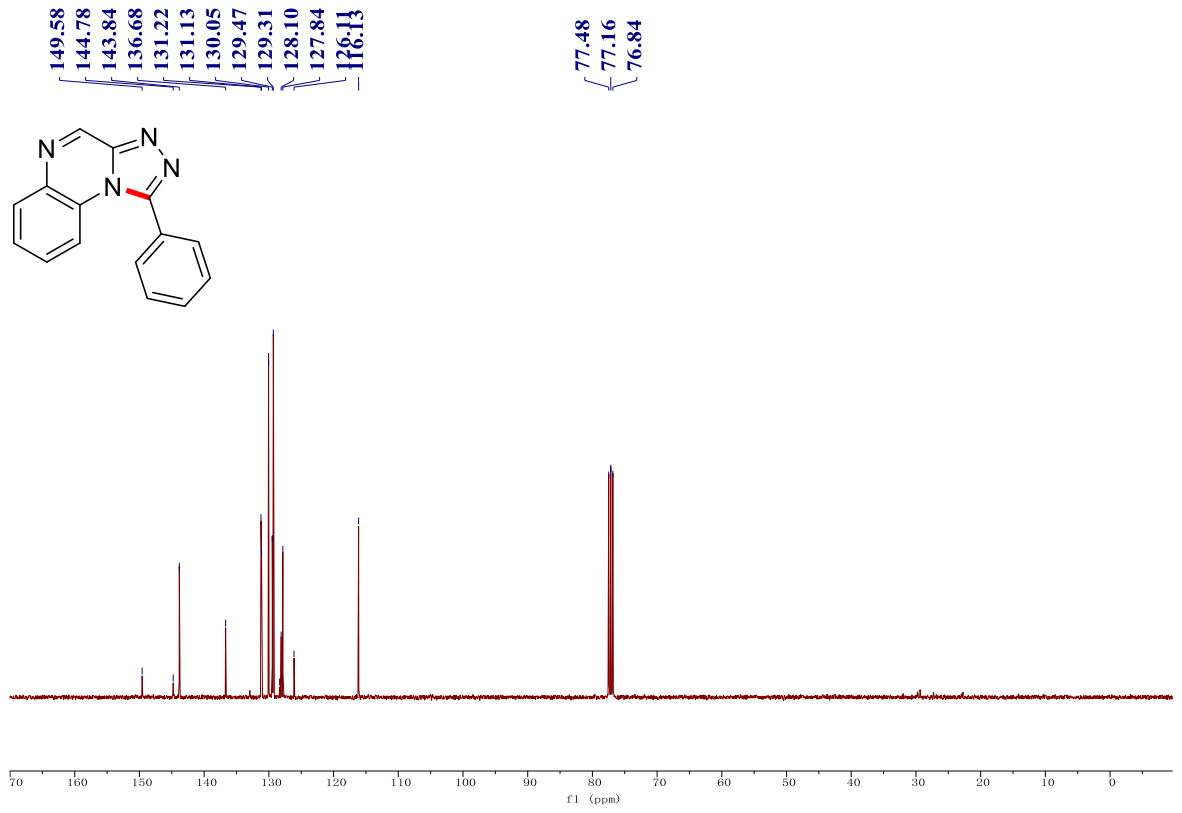
^{13}C NMR Spectrum of 2aam at 25 ^{o}C ($CDCl_3$, 100 MHz)



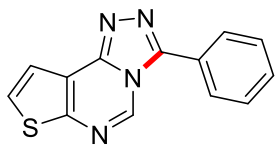
^{1}H NMR Spectrum of 2aan at 25 ^{o}C ($CDCl_3$, 400 MHz)

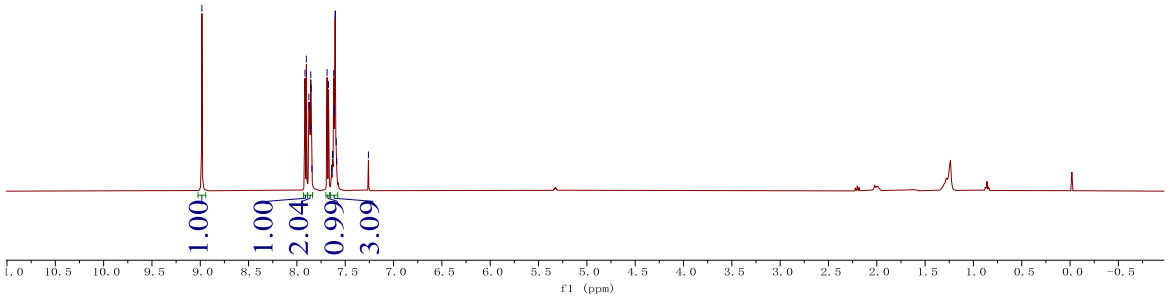


^{13}C NMR Spectrum of 2aan at 25 ^{o}C ($CDCl_3$, 100 MHz)

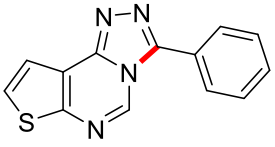


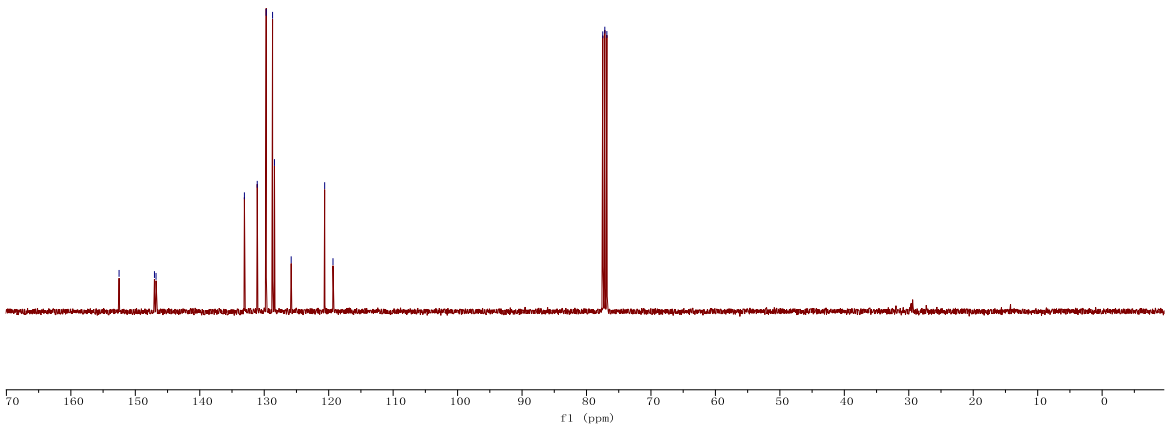
^{1}H NMR Spectrum of 2aao at 25 °C ($CDCl_3$, 400 MHz)



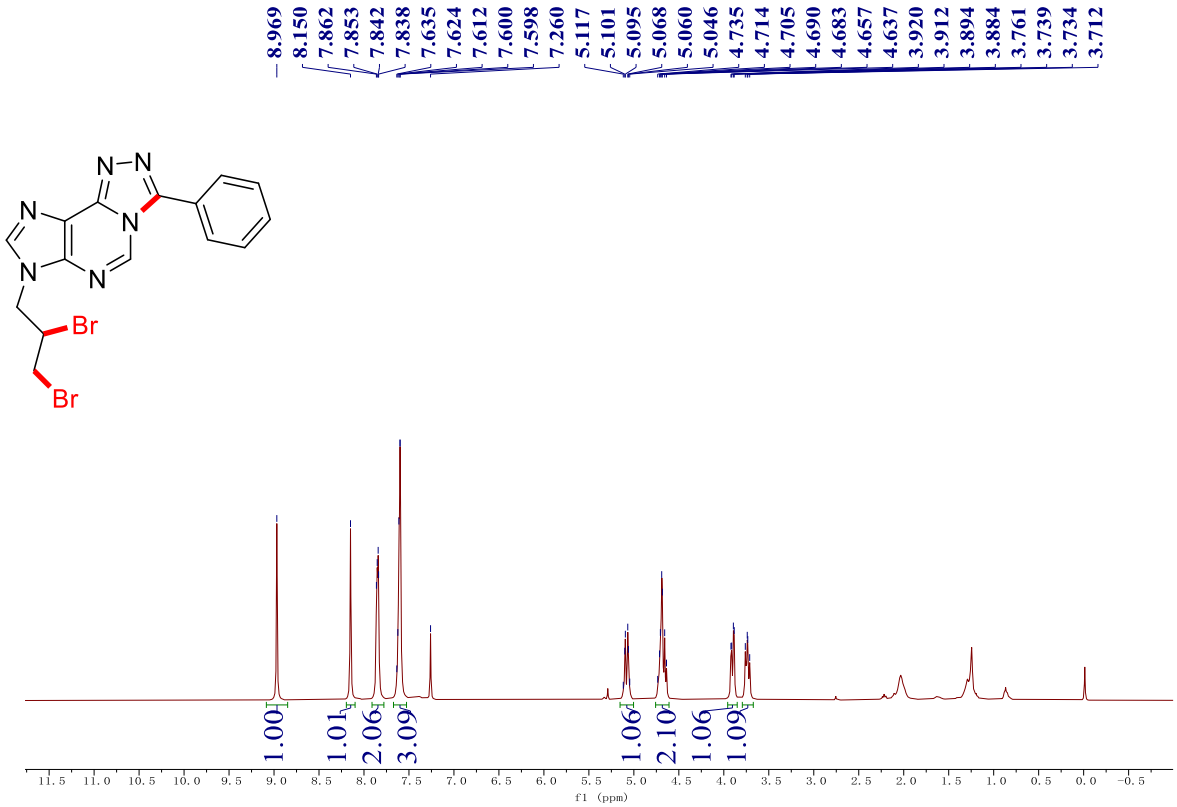


^{13}C NMR Spectrum of 2aao at 25 °C ($CDCl_3$, 100 MHz)

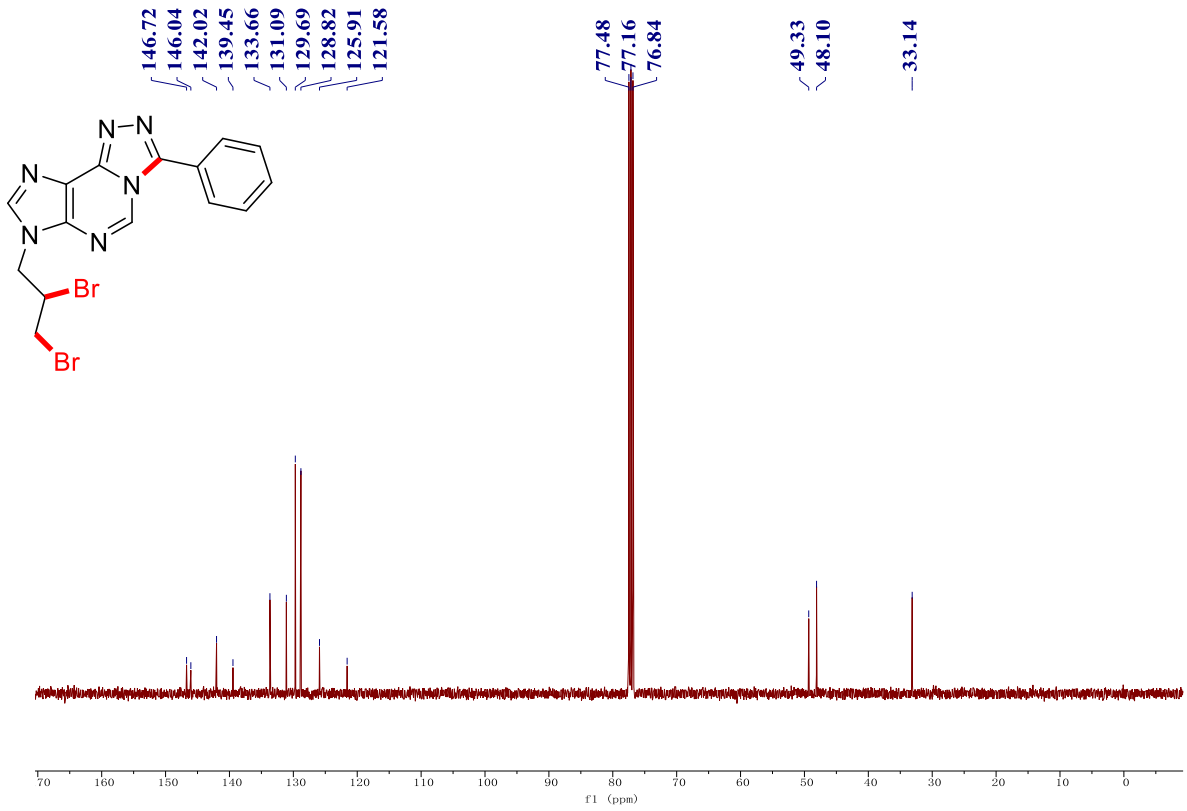




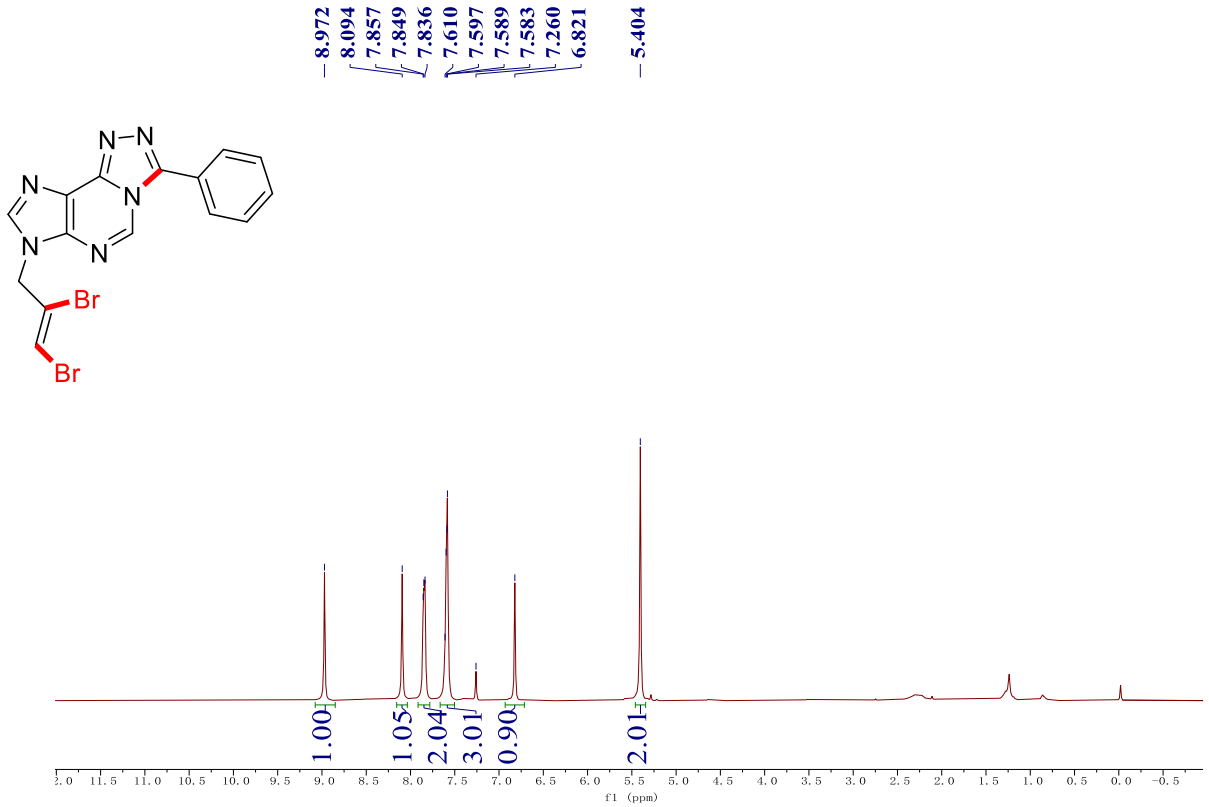
^{1}H NMR Spectrum of 2ar' at 25 °C ($CDCl_3$, 400 MHz)



^{13}C NMR Spectrum of 2ar' at 25 °C ($CDCl_3$, 100 MHz)



^{1}H NMR Spectrum of 2as' at 25 °C ($CDCl_3$, 400 MHz)



^{13}C NMR Spectrum of 2as' at 25 °C ($CDCl_3$, 100 MHz)