

Supporting Information

Catalyst-free multicomponent formation of novel acylpyrrole-containing 6-iminohexahydro-1*H*-pyrido[1,2-*a*]pyrimidine derivatives

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General information :

Microwave reaction was carried out in WF-4000C model microwave system and all reactions were investigated under the power of 200W. ^1H and ^{13}C NMR (400 MHz) spectra were recorded in DMSO- d_6 at room temperature using a Bruker Ultra Shield Plus 400 MHz instrument with reference tetramethylsilane (TMS). Melting points (m.p.) were taken on a Gal-lenkamp apparatus. FTIR spectra were measured using a Germany Bruker Vertex 80v FT-IR spectrometer by incorporating samples in KBr disks. High resolution mass spectra were operated on Agilent 6550 iFunnel Q-TOF. All of the reagents were purchased from chemical reagent Crop. in China and used without further purification

Experimental :

Typical procedure for the Preparation of 3-(1-methyl-1H-pyrrol-2-yl)-3-oxopropanenitrileb (4):

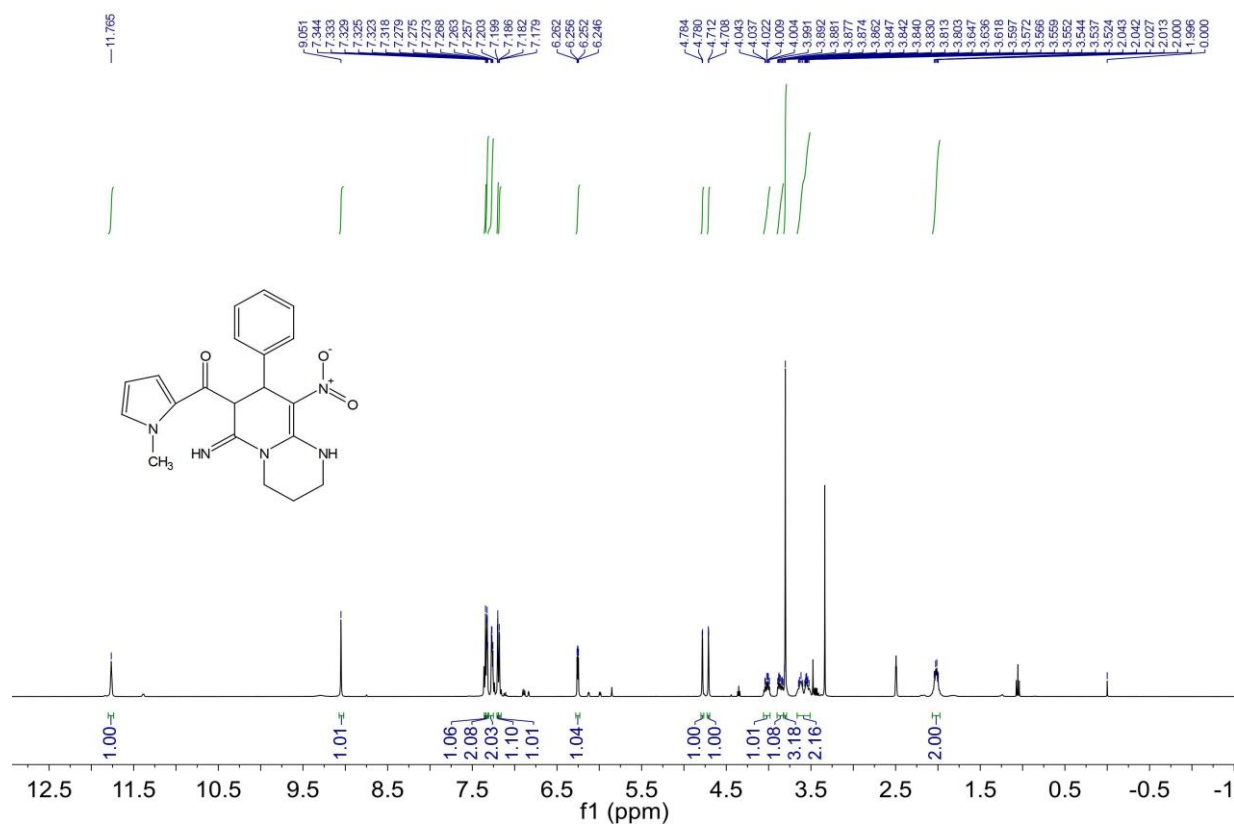
A mixture of cyanoacetic acid (1.7 g, 20 mmol) and acetic anhydride (2.04 g, 20 mmol) was heated at 100 °C for 30 min and then the resulting mixture was mixed with N-methylpyrrole (1.62 g, 20 mmol) and the reaction mixture was heated at 100 °C again for 1.5 h. At last cooling it to 20 °C, the reaction mixture was poured over water and crude solid product was collected by filtration and recrystallized (EtOH) to give the compound **4** as pale yellow crystals.

(Conventional heating) synthesis of acylpyrrole-containing pyrido[1,2-a]pyrimidine **5b**:

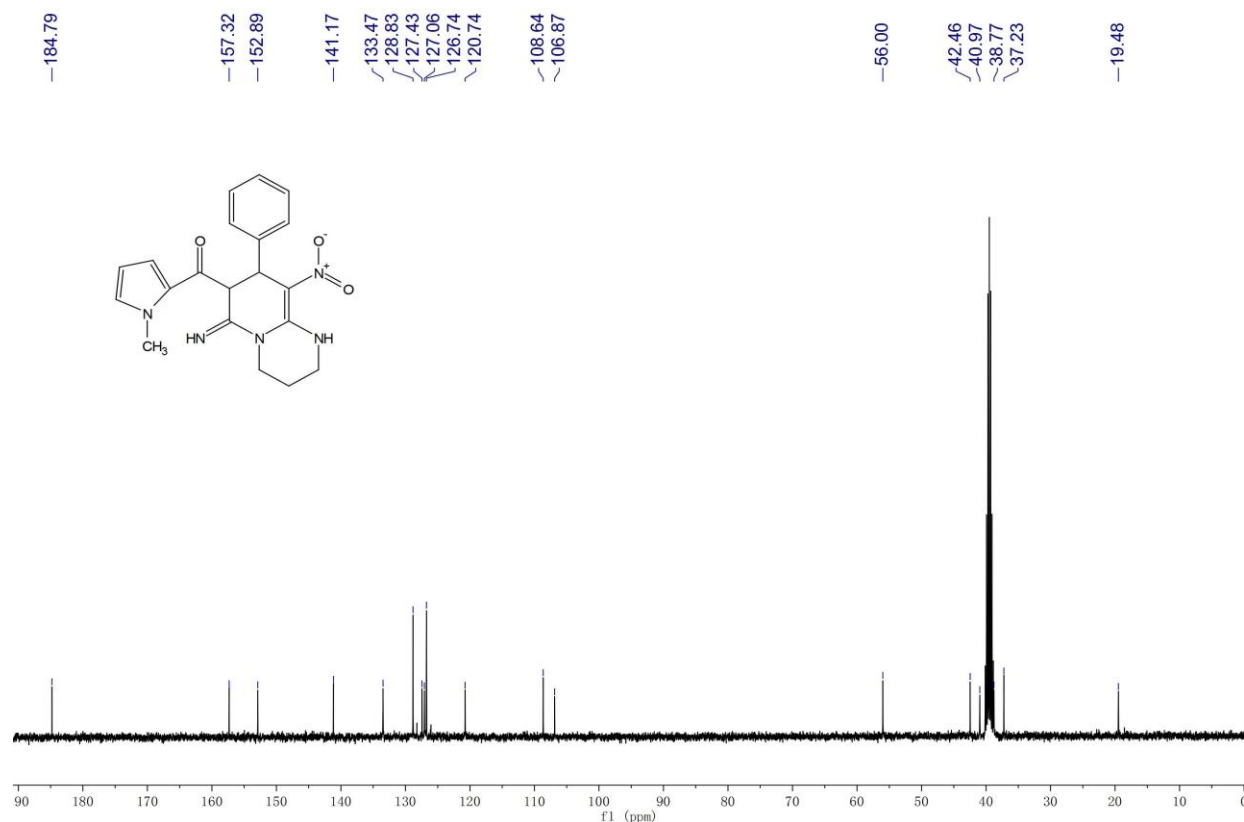
A mixture of propane-1,3-diamine **1** (0.074 g, 1 mmol), 1,1-bis(methylthio)-2-nitroethene **2** (1 mmol, 0.165 g) and 5 mL EtOH in a 50 mL flask was stirred at reflux for 6 h. After completion of the reaction (monitored by TLC), 3-(1-methyl-1H-pyrrol-2-yl)-3-oxopropanenitrileb **4** (0.148 g, 1 mmol), 2,4-dichlorobenzaldehyde **3b** (0.174 g, 1 mmol) were added to the reaction mixture, and it was stirred at reflux for 1h. Then, the reaction mixture was allowed to cool to room temperature, and the precipitates were filtered and washed with ethanol to give product **5b** in yield of 78% (as showed in Table 1).

(Microwave irradiation) synthesis of acylpyrrole-containing pyrido[1,2-a]pyrimidines **5b**:

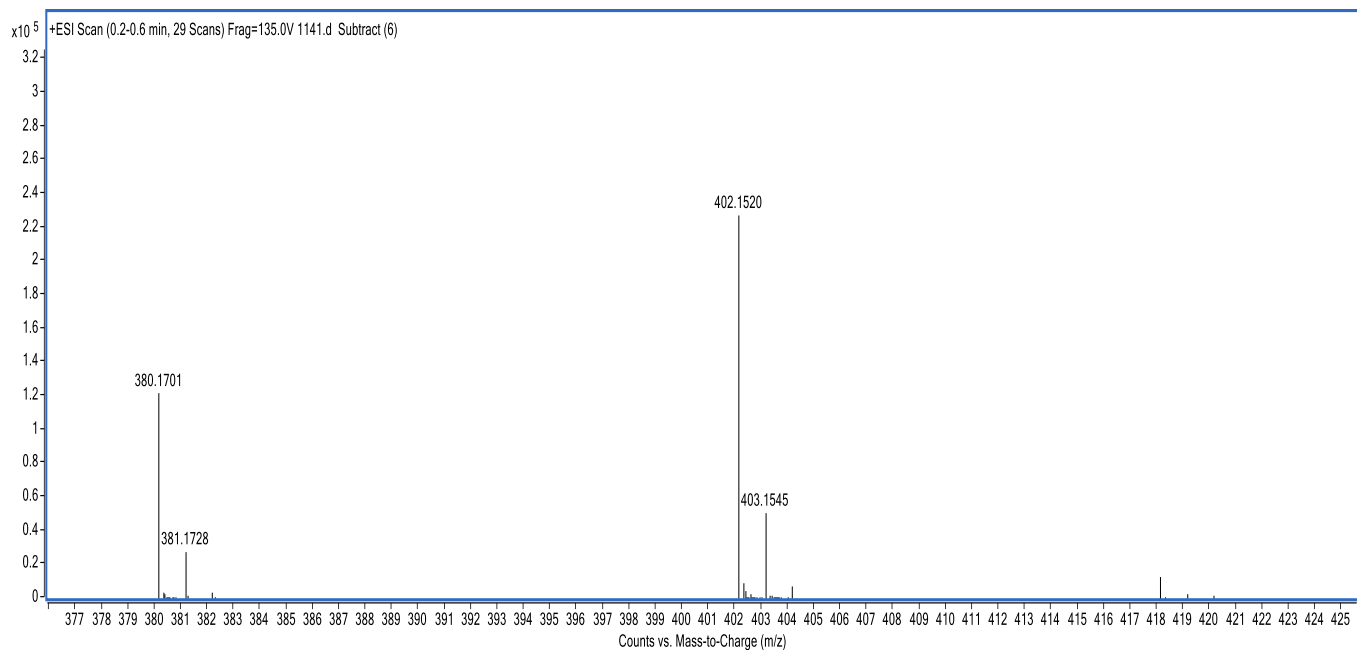
In a 20 ml reaction vessel, a mixture of 1,1-bis(methylthio)-2-nitroethene **2** (0.165 g, 1mmol), propane-1,3-diamine **1** (0.074 g, 1 mmol) and 5 mL EtOH were heated at 80°C under microwave irradiation (200W) for 1 h. After completion of the reaction (confirmed by TLC), aromatic aldehydes **3** (1 mmol) and 3-(1-methyl-1H-pyrrol-2-yl)-3-oxopropanenitrile **4** (0.148 g, 1 mmol) were added to the reaction mixture for 10-15 min at 80°C under microwave irradiation (200W). Then, the reaction mixture was cooled to room temperature and the precipitated solid was filtered by Buchner filtration, afterward washed with little cold EtOH and dried under vacuum to acquire product **5** in yield of 76-89% (as showed in Table 1).



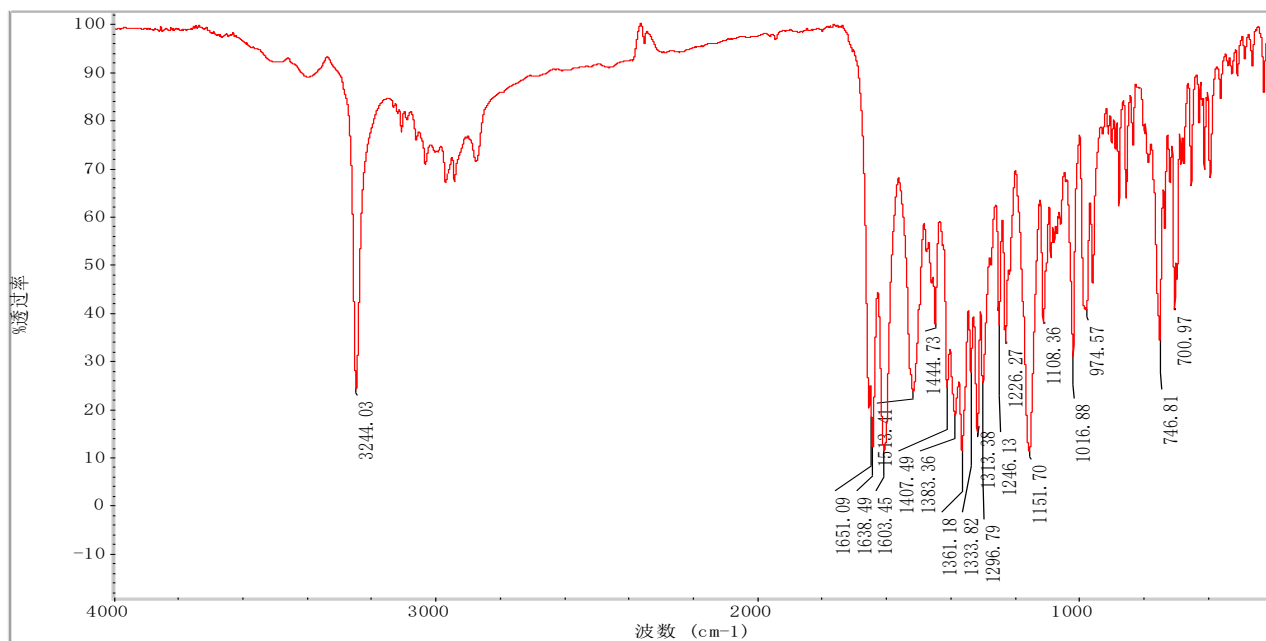
¹H-NMR Spectrum of Compound 5a



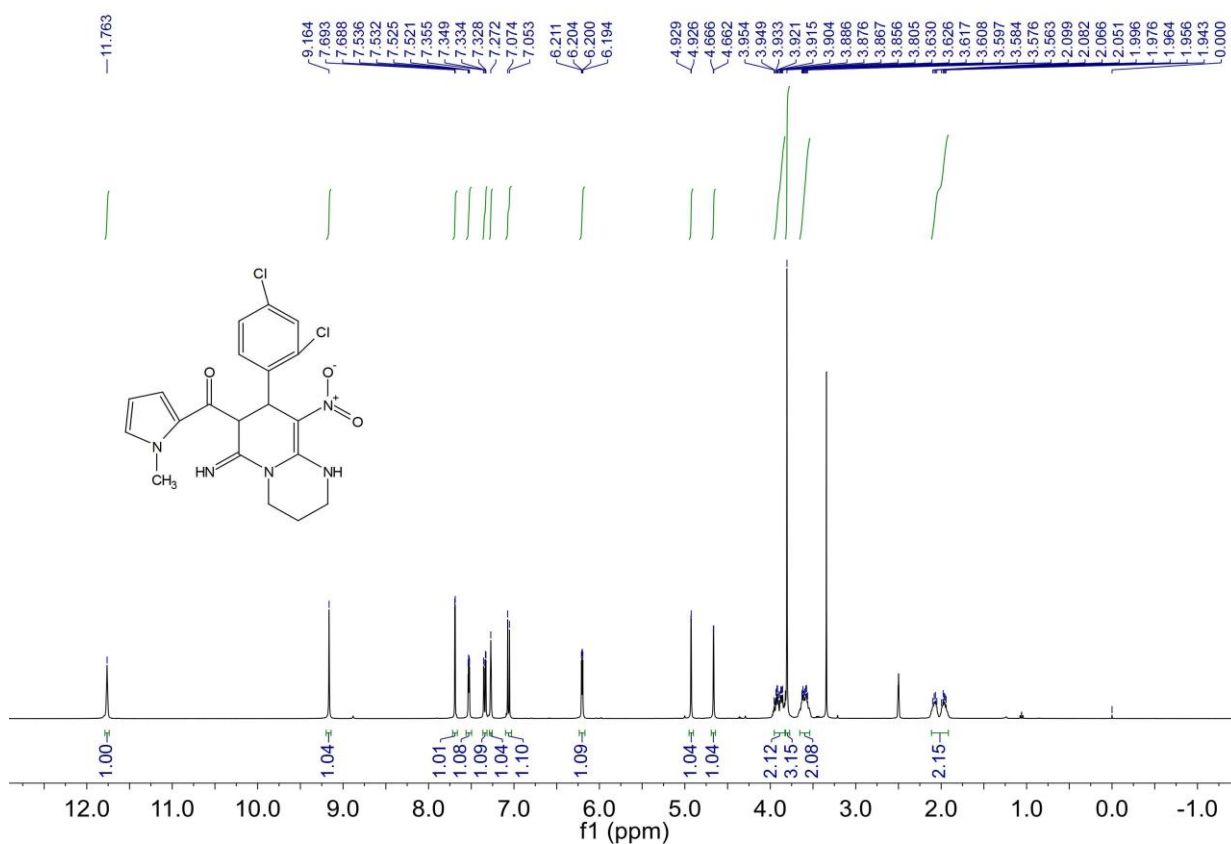
¹³C-NMR Spectrum of Compound 5a



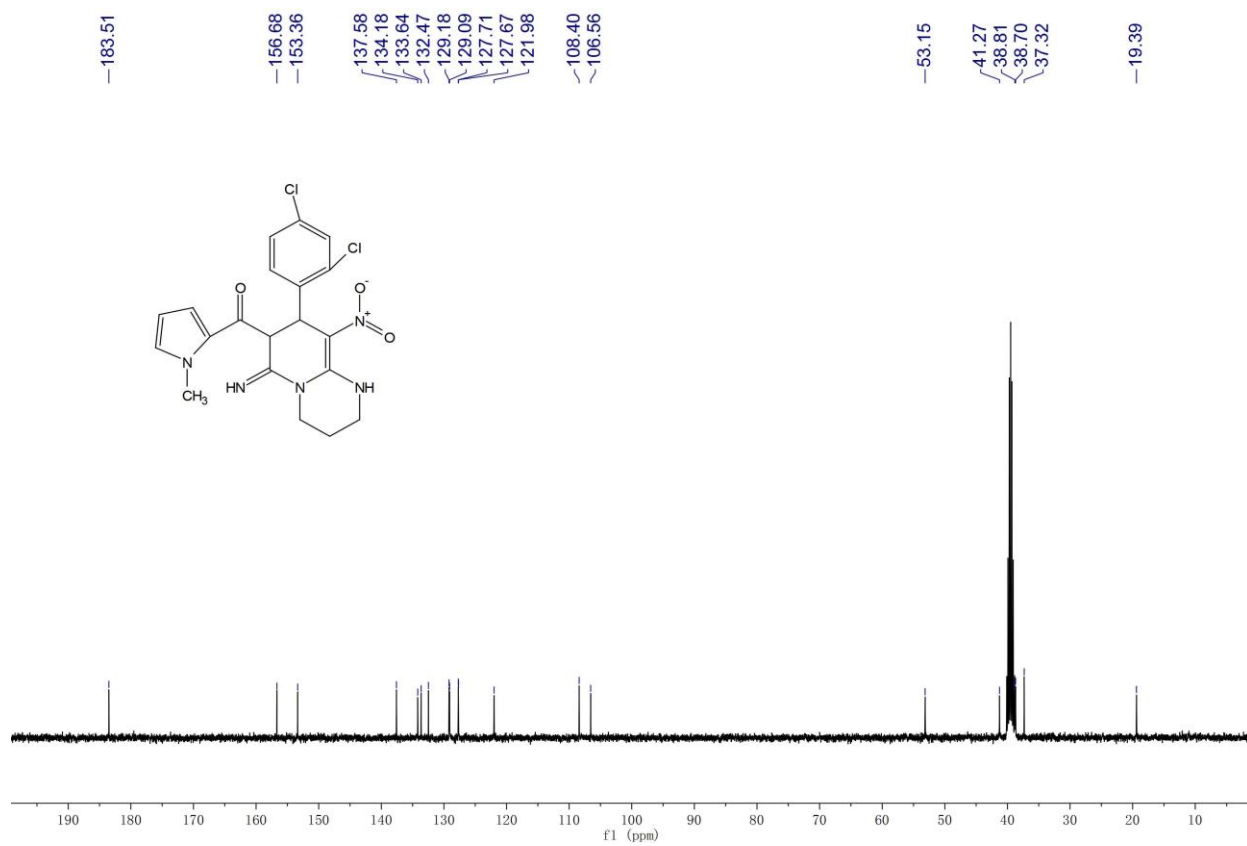
HRMS Spectrum of Compound 5a



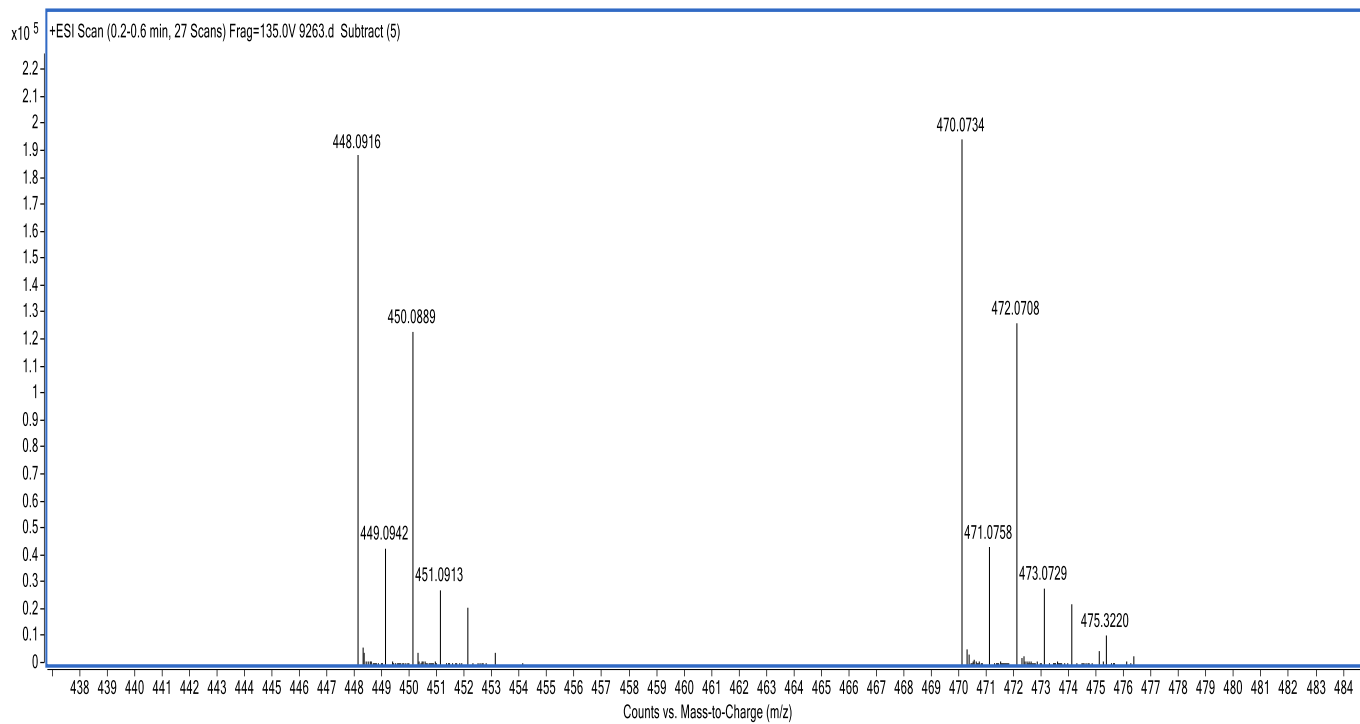
IR Spectrum of Compound 5a



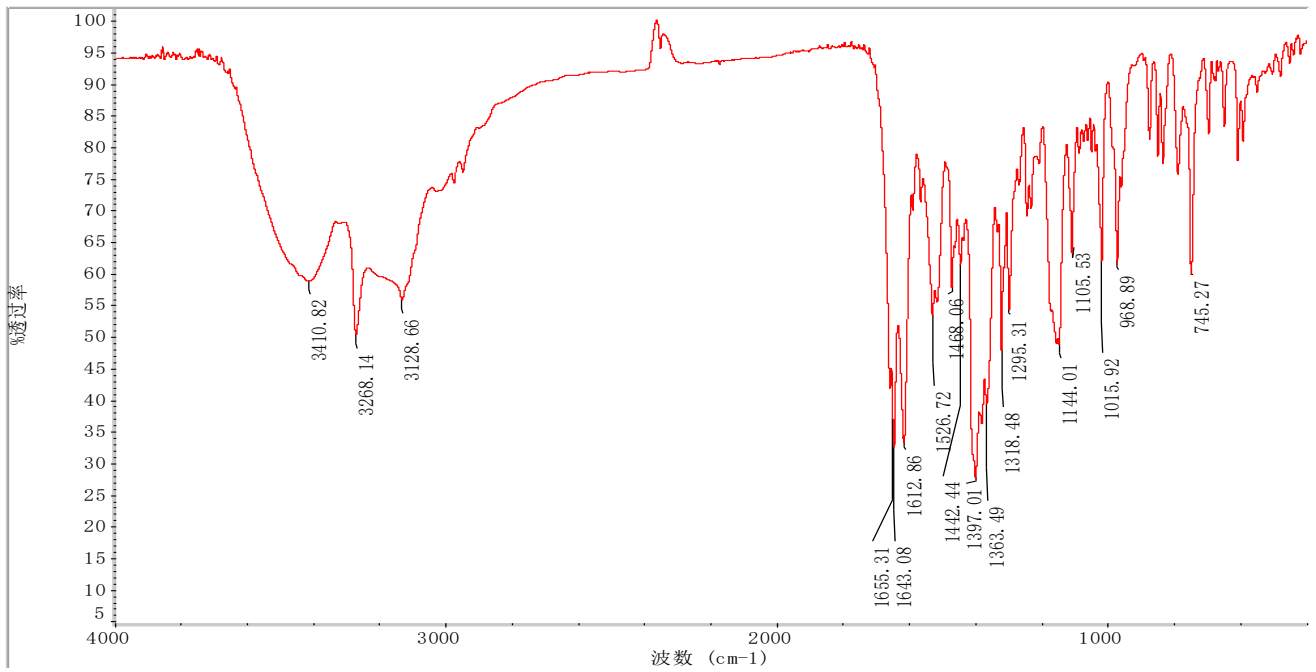
¹H-NMR Spectrum of Compound 4b



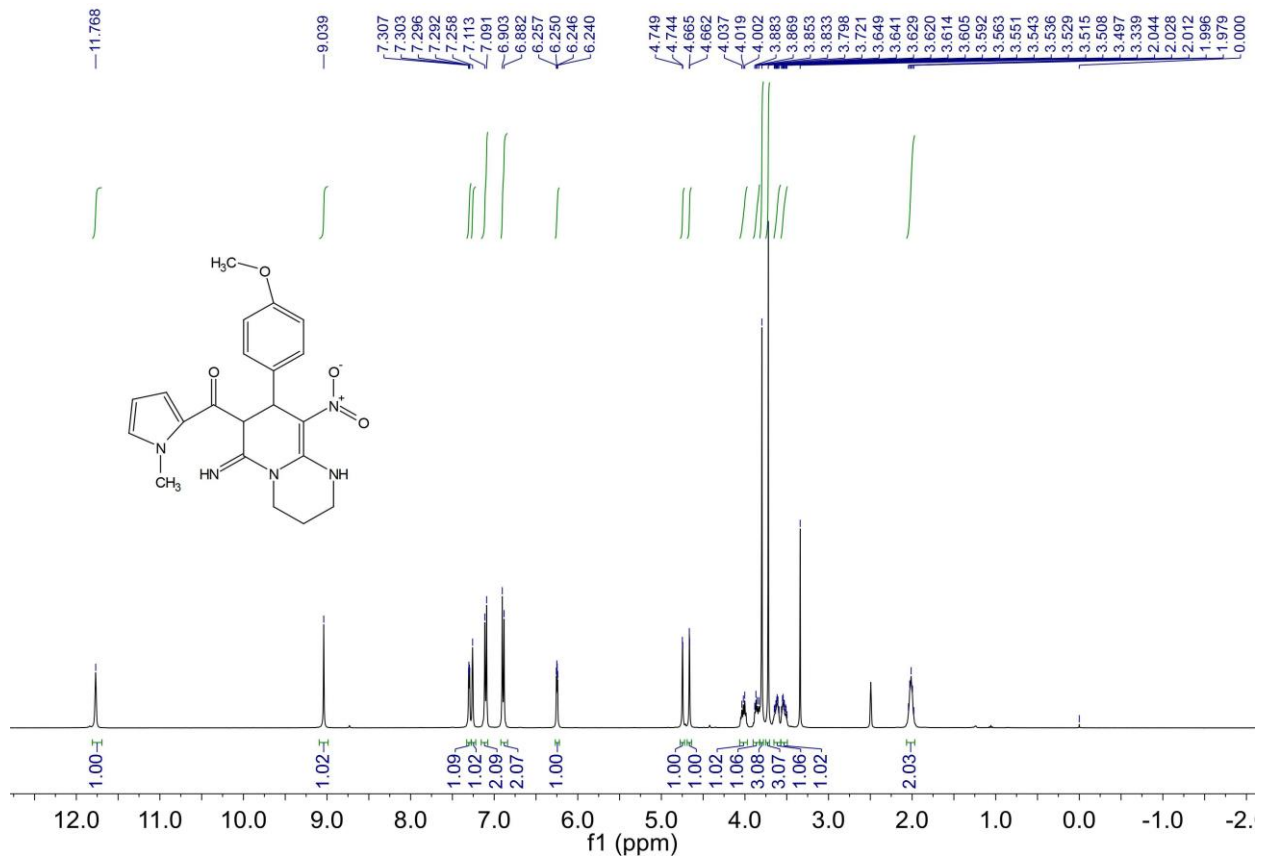
¹³C-NMR Spectrum of Compound 5a



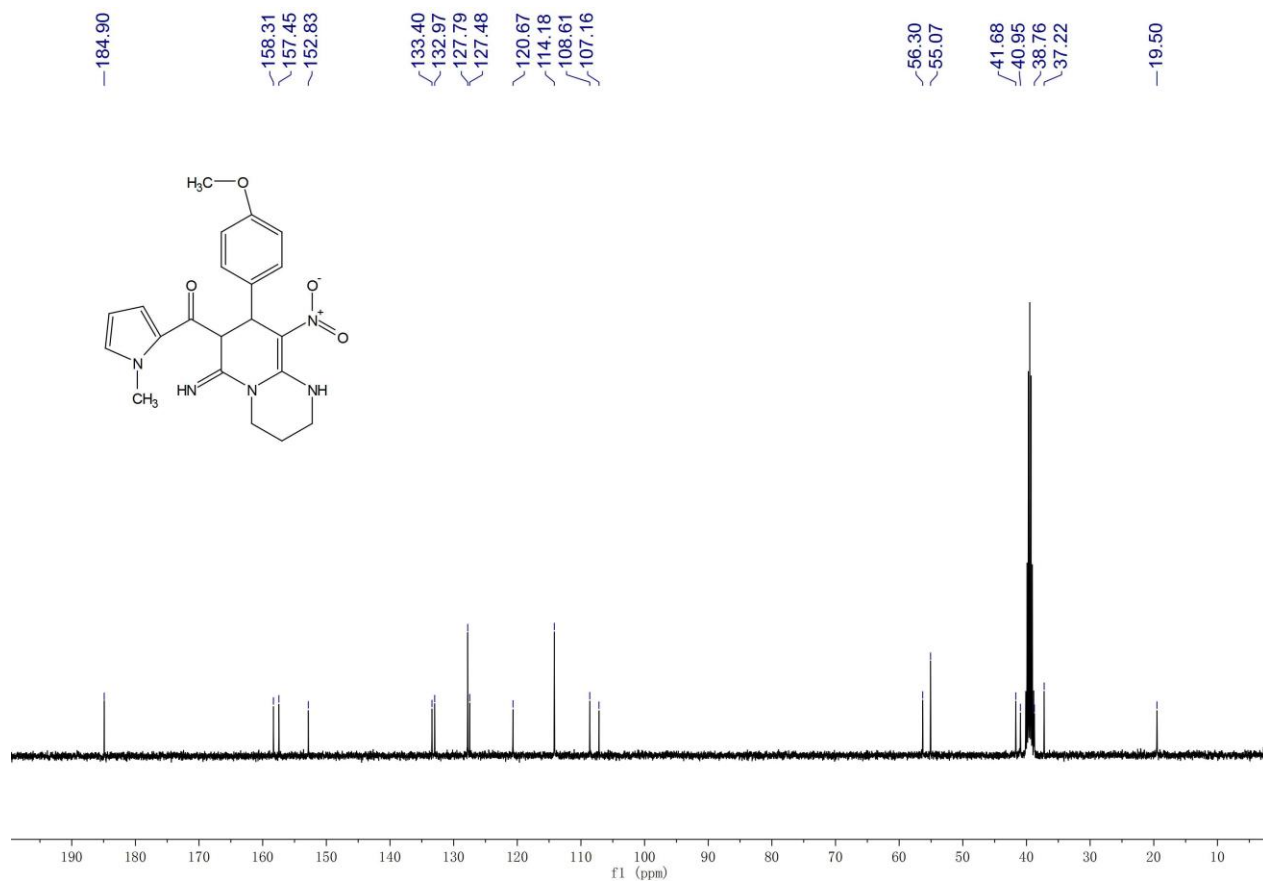
HRMS Spectrum of Compound 5b



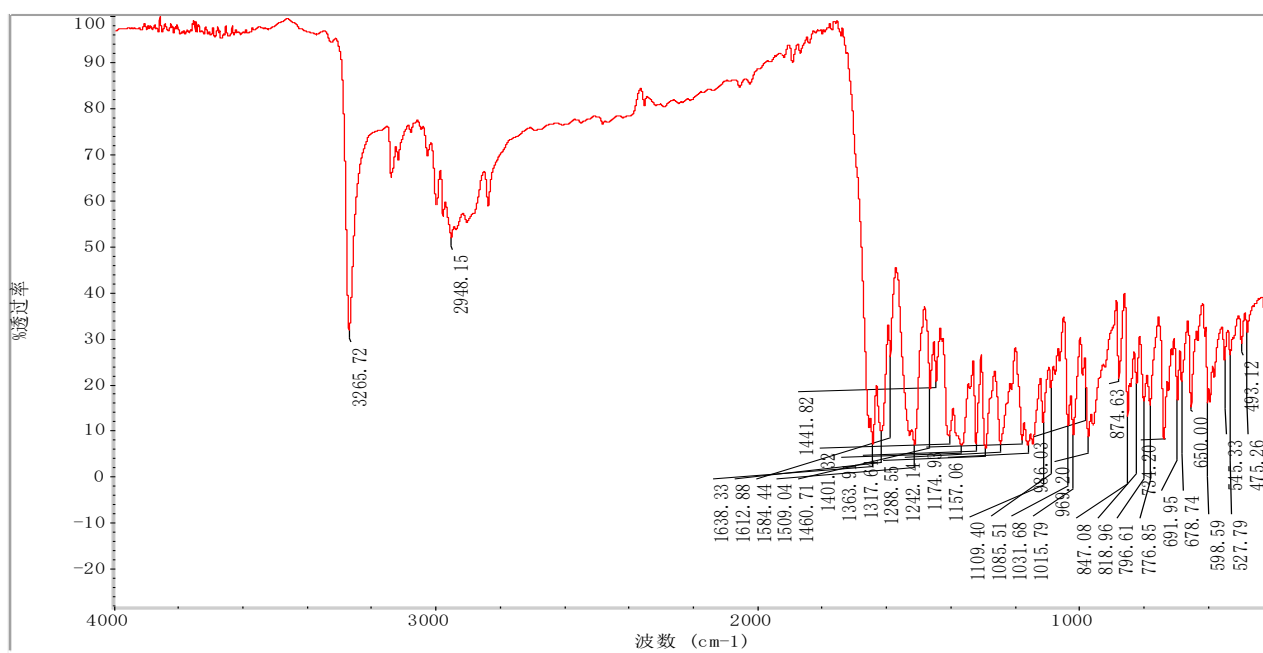
IR Spectrum of Compound 5b



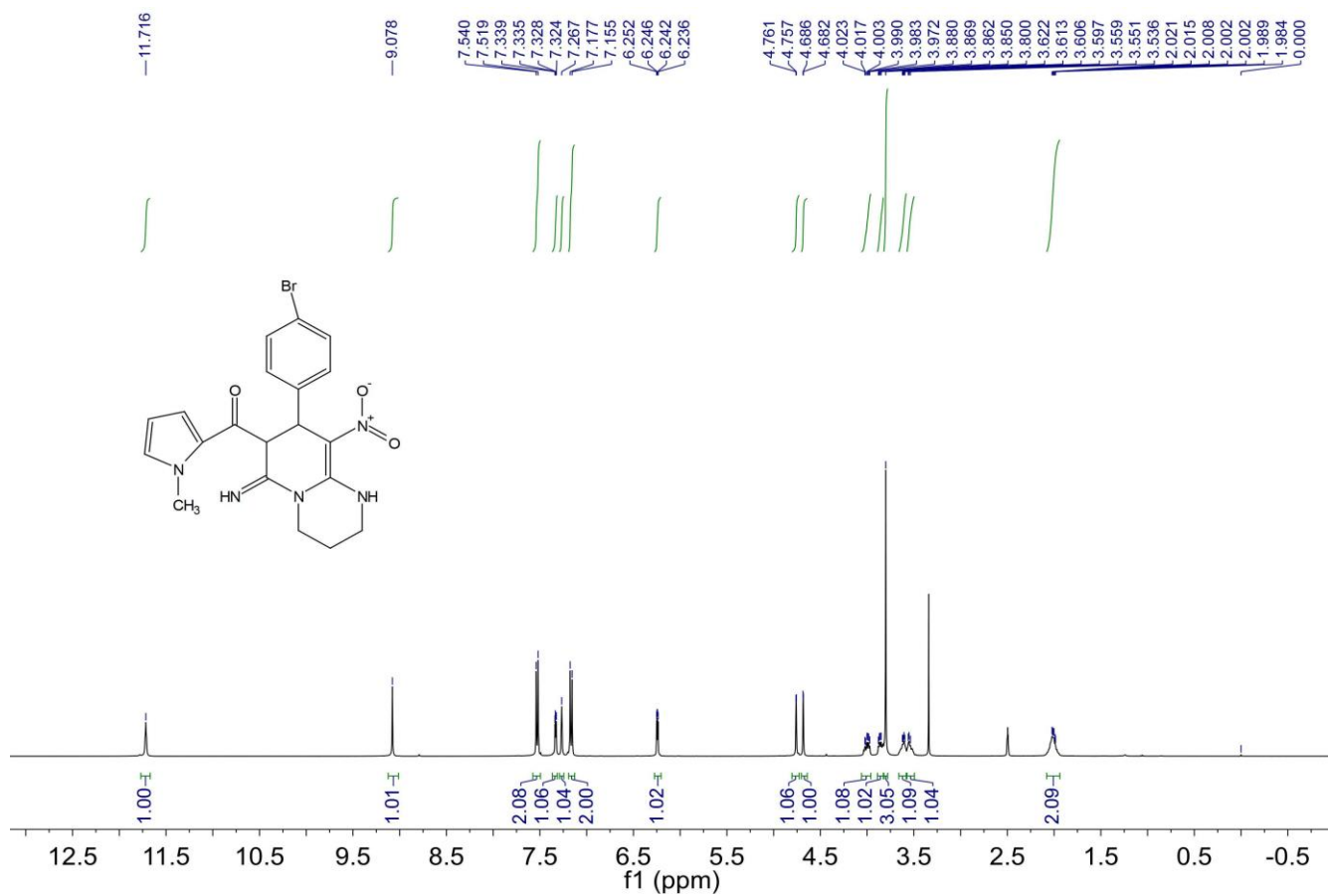
¹H-NMR Spectrum of Compound 5c



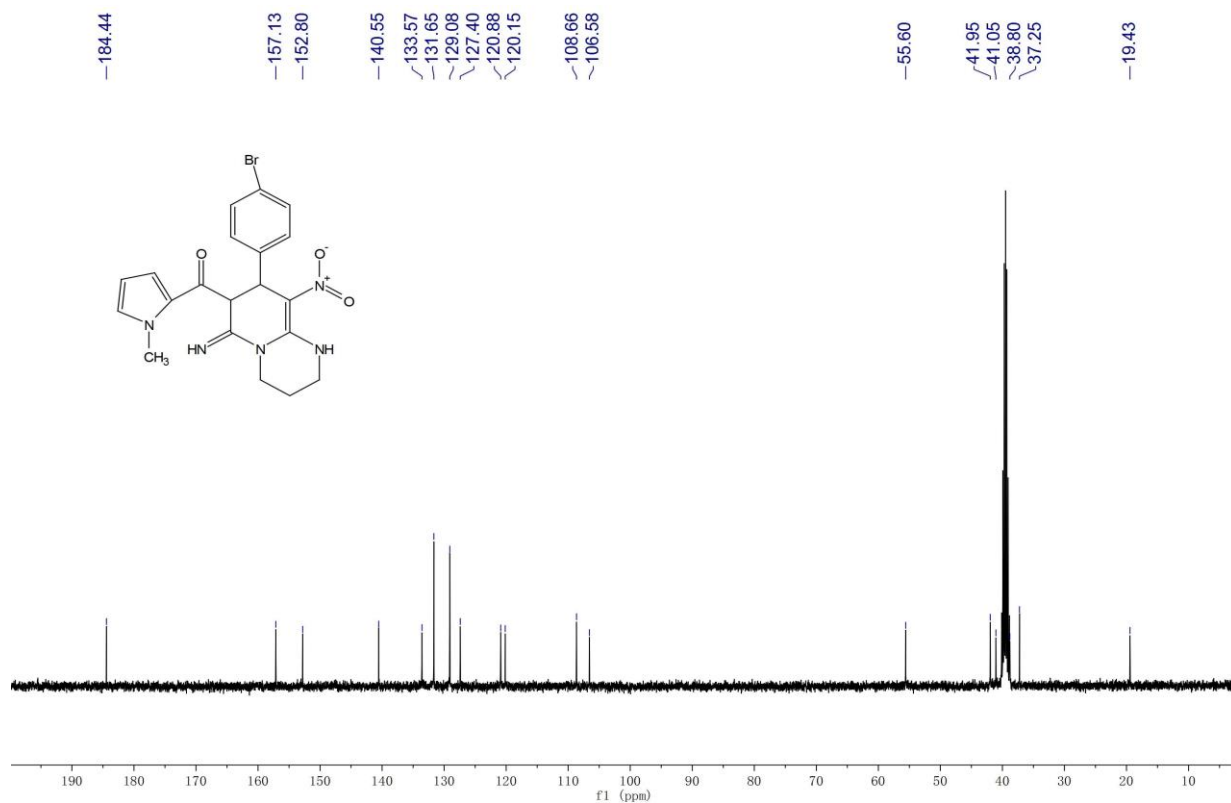
$^{13}\text{C-NMR}$ Spectrum of Compound 5c



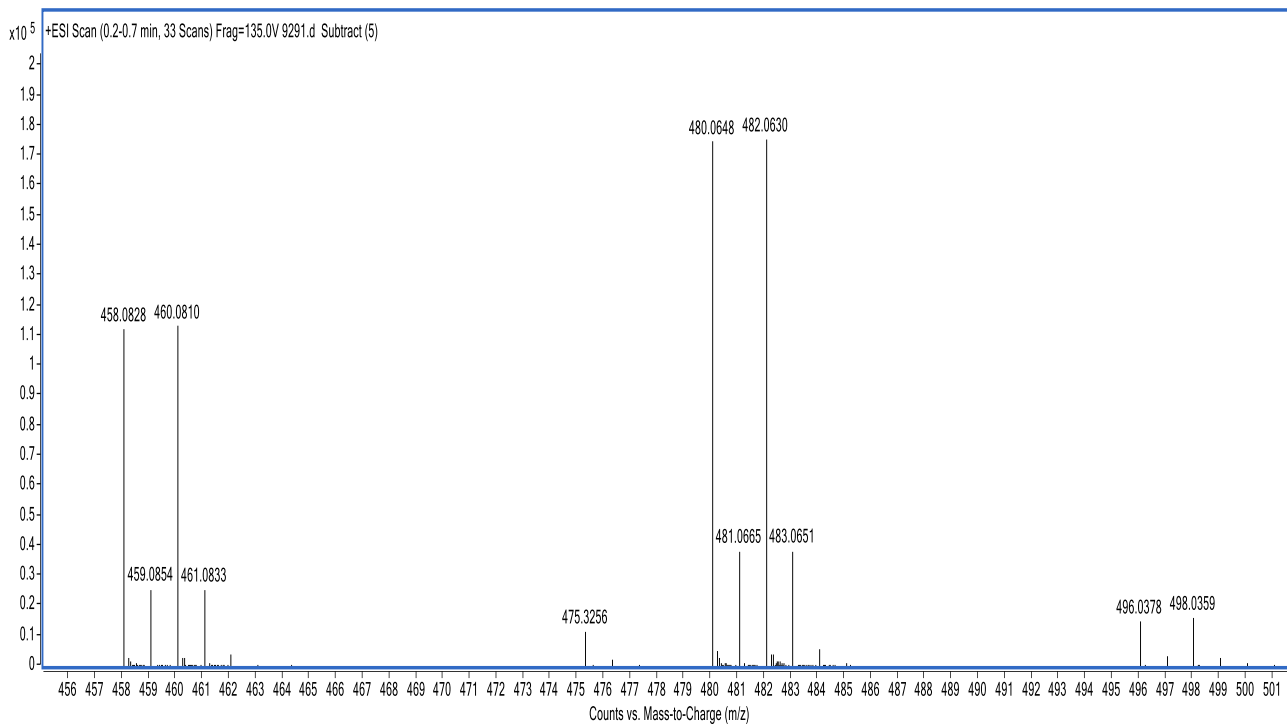
IR Spectrum of Compound 5c



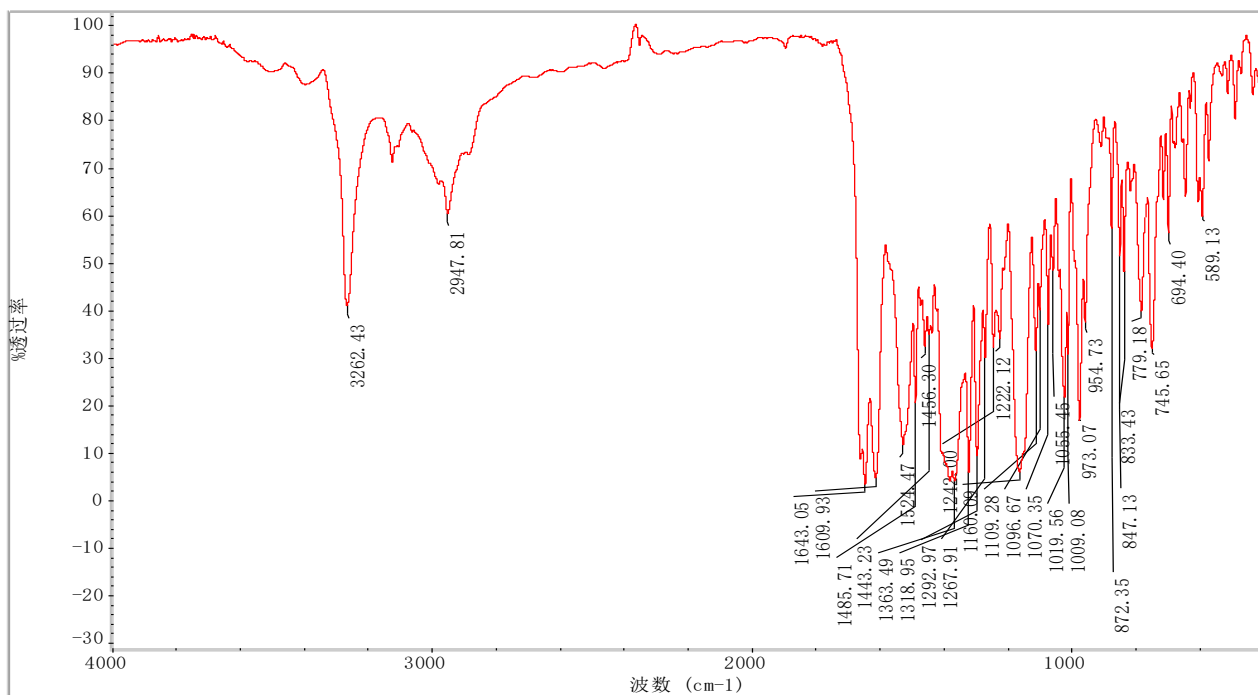
¹H-NMR Spectrum of Compound 5d



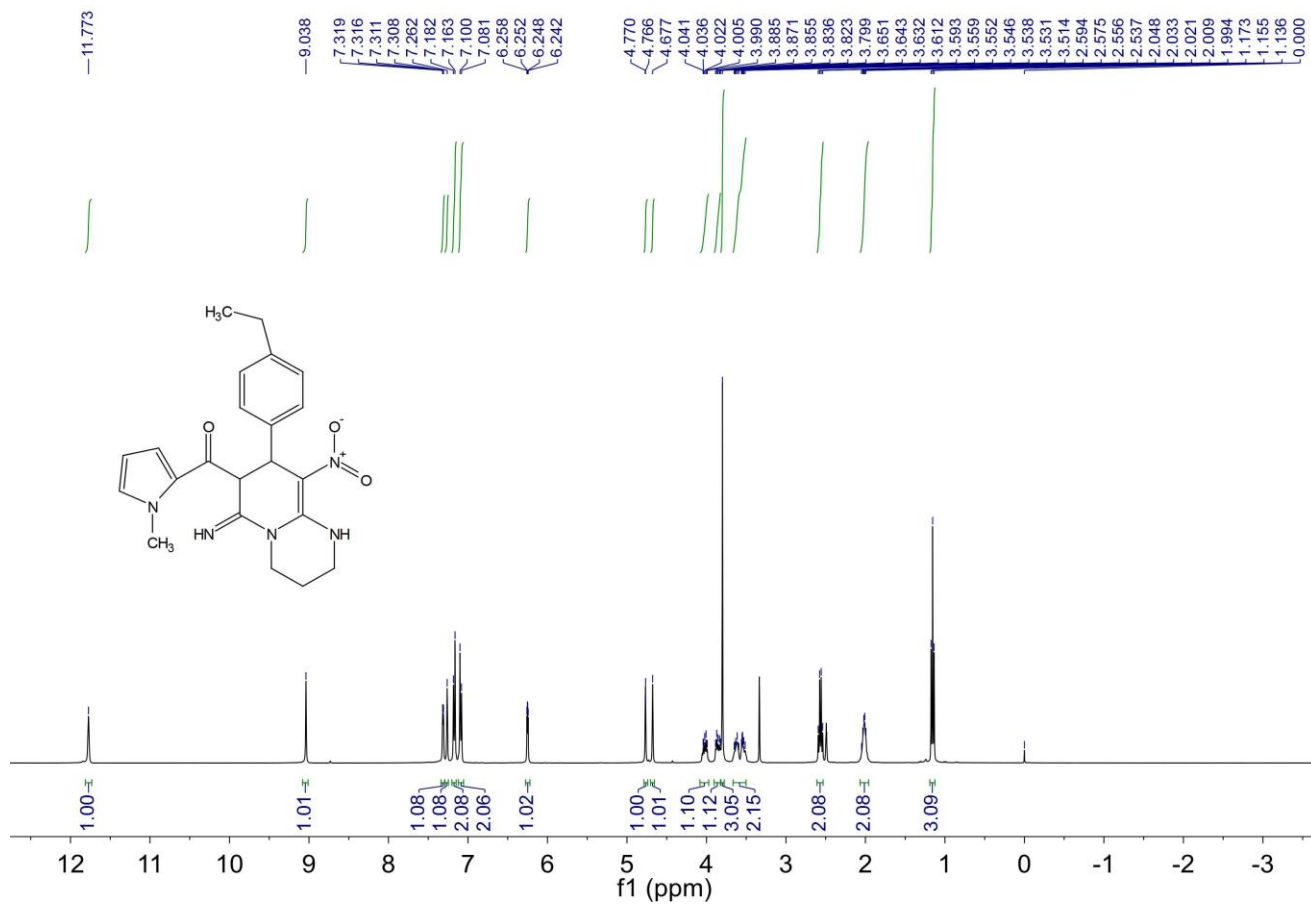
¹³C-NMR Spectrum of Compound 5d



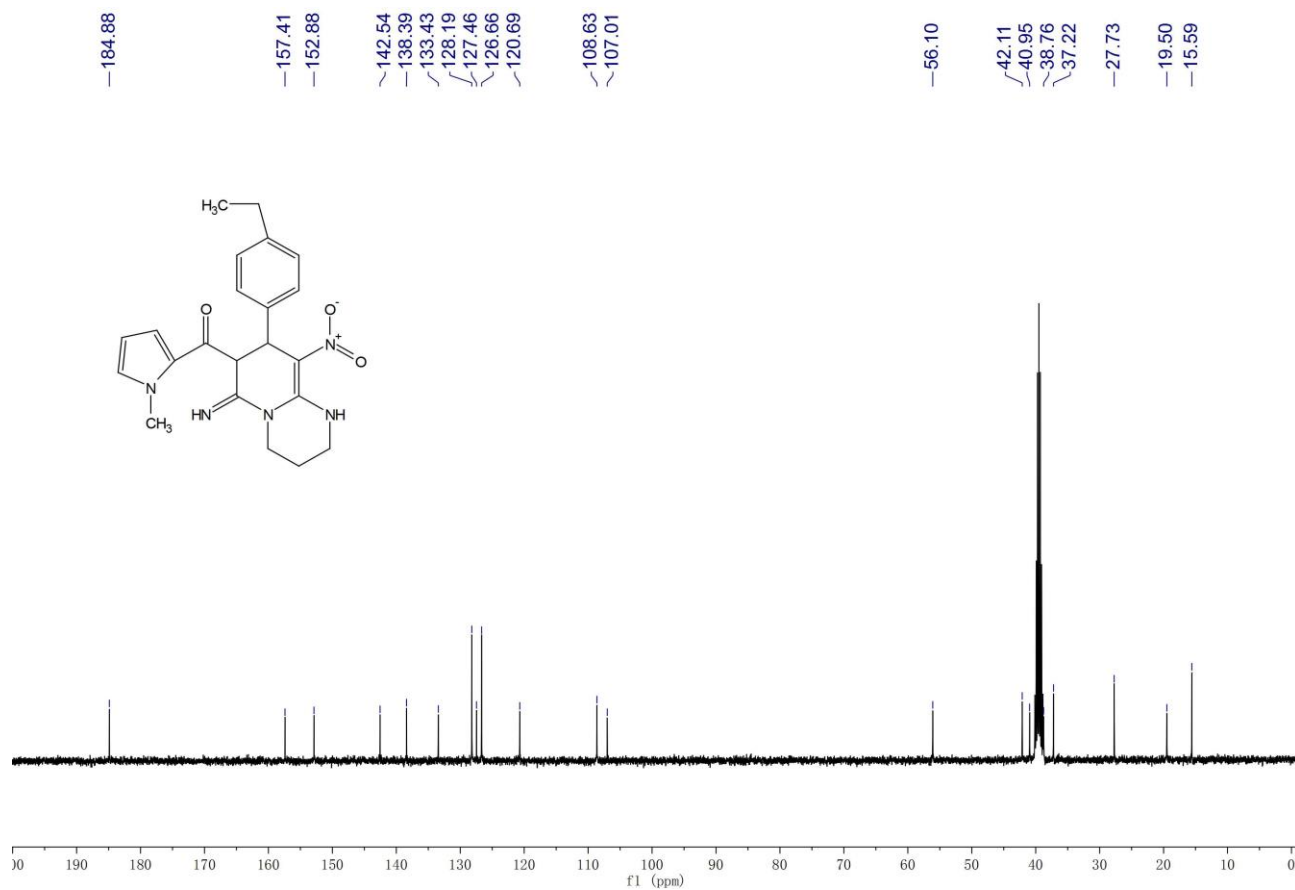
HRMS Spectrum of Compound 5d



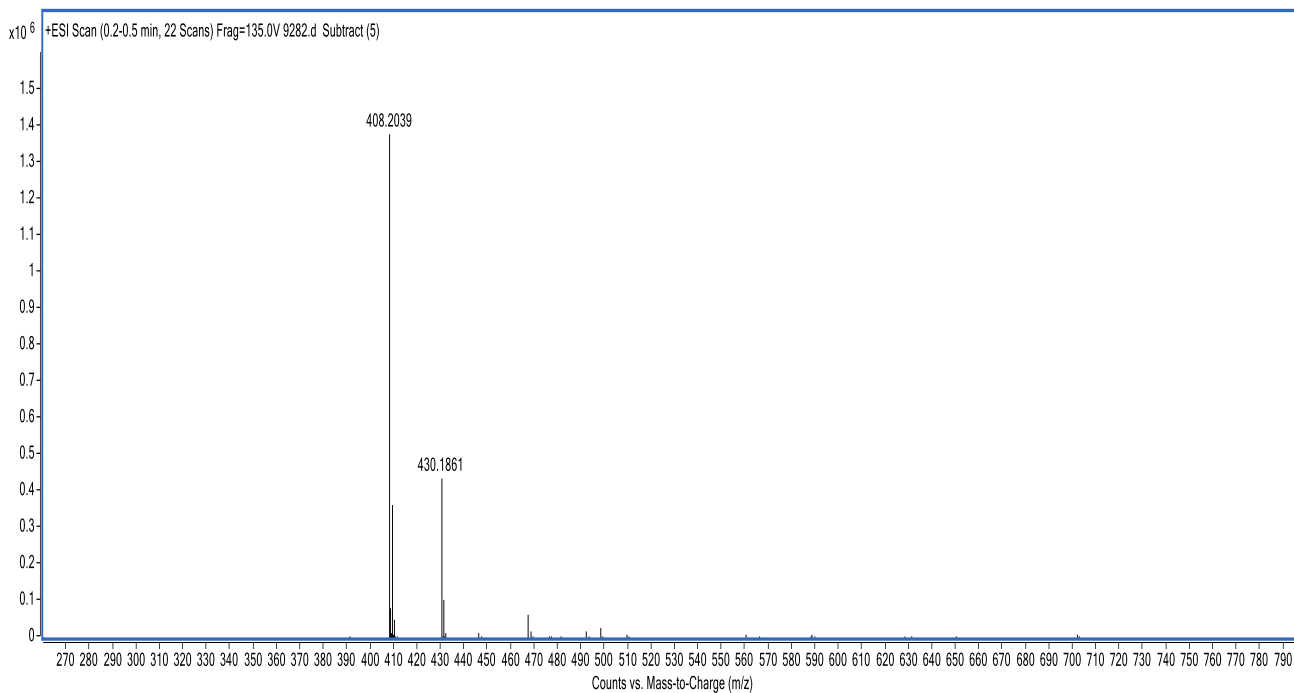
IR Spectrum of Compound 5d



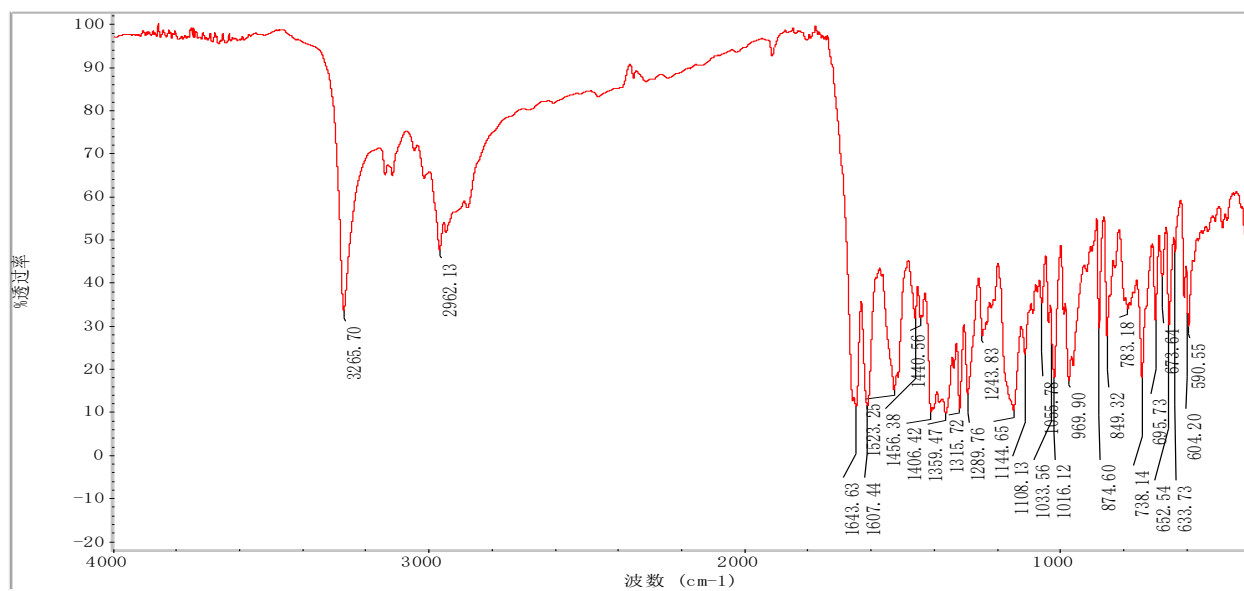
¹H-NMR Spectrum of Compound 5e



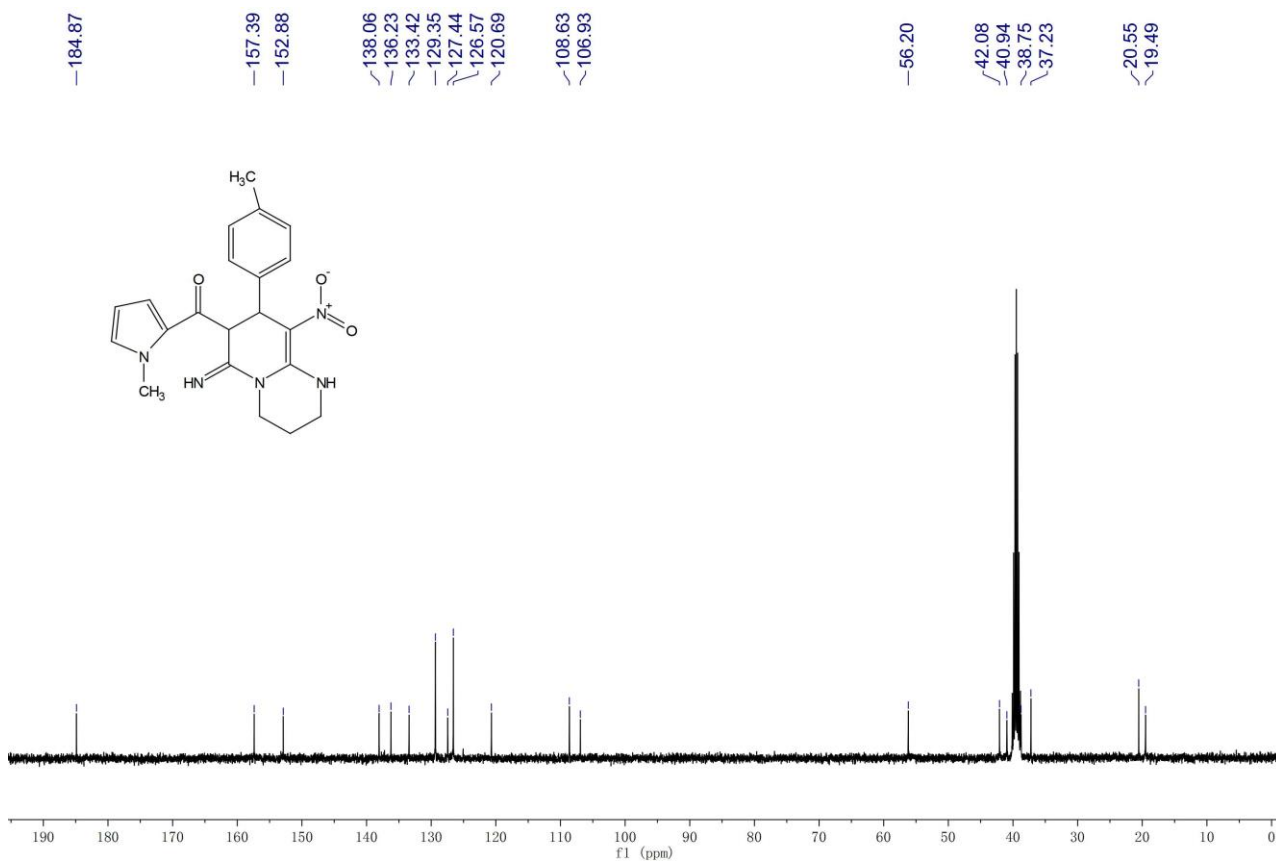
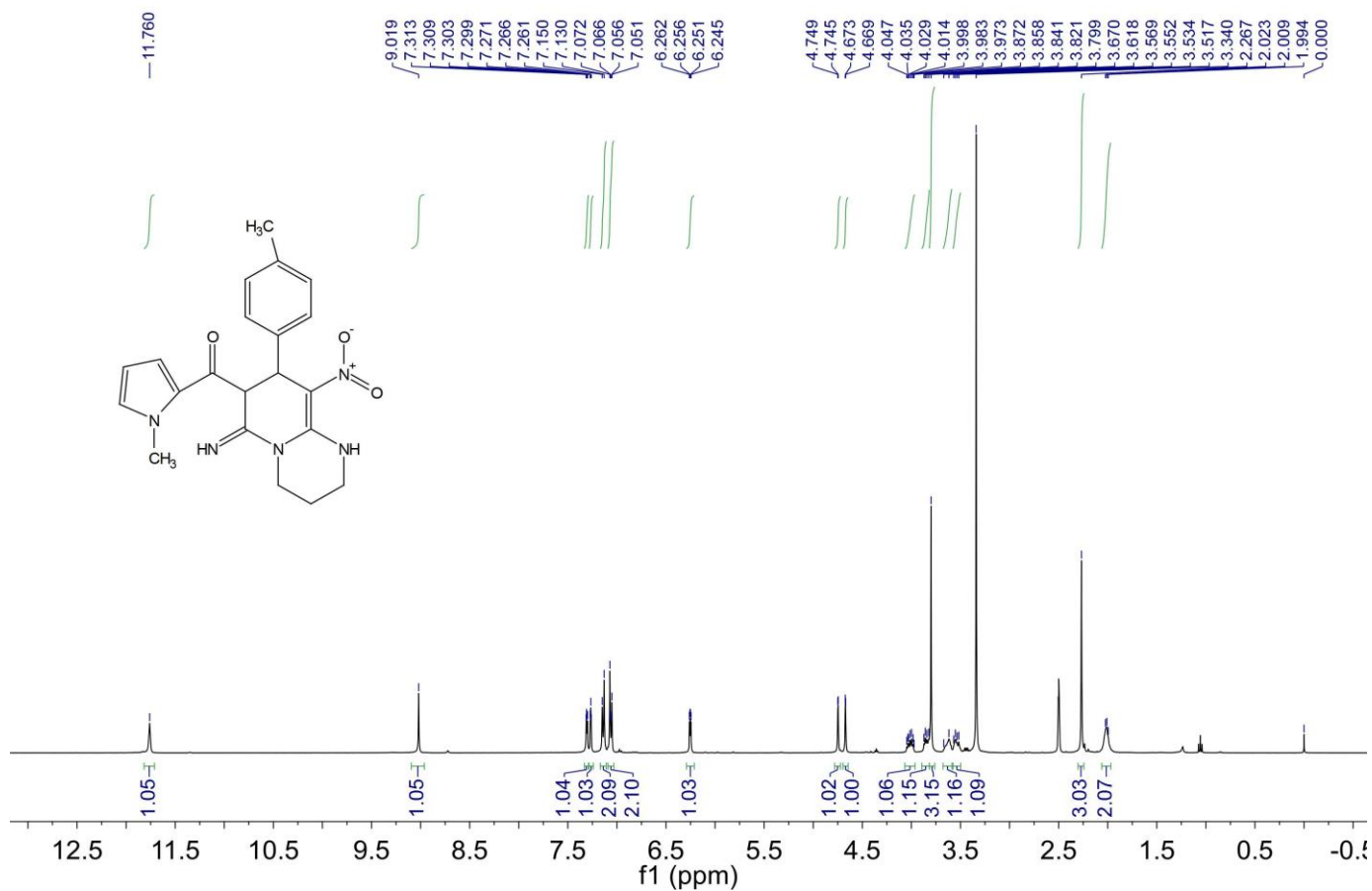
¹³C-NMR Spectrum of Compound 5e

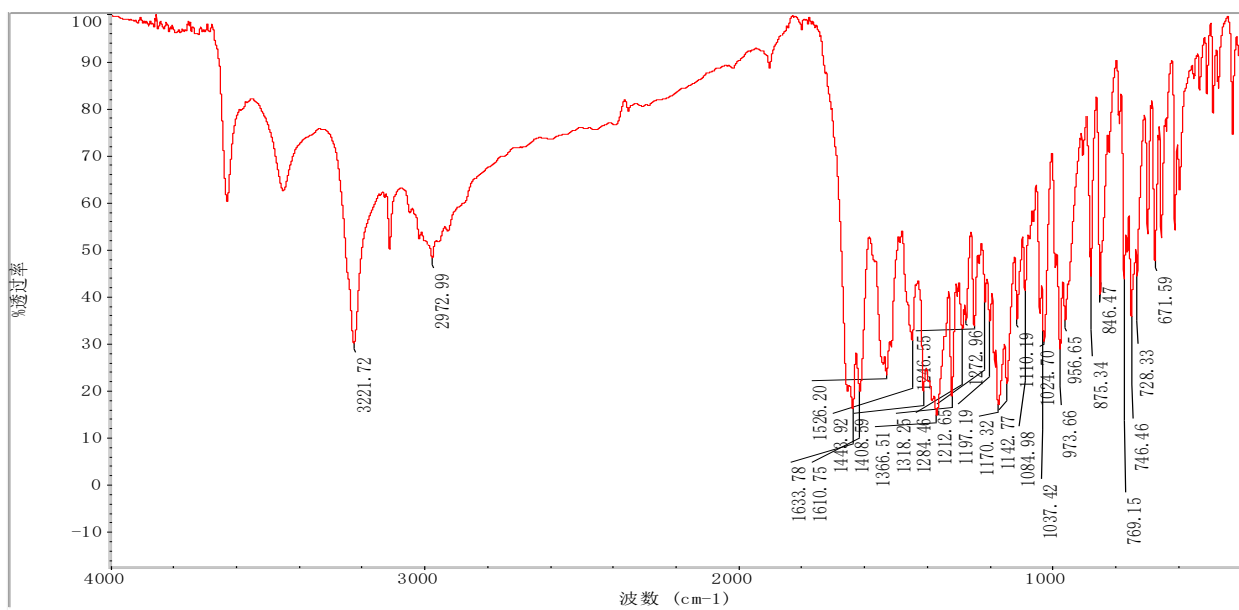


HRMS Spectrum of Compound 5e

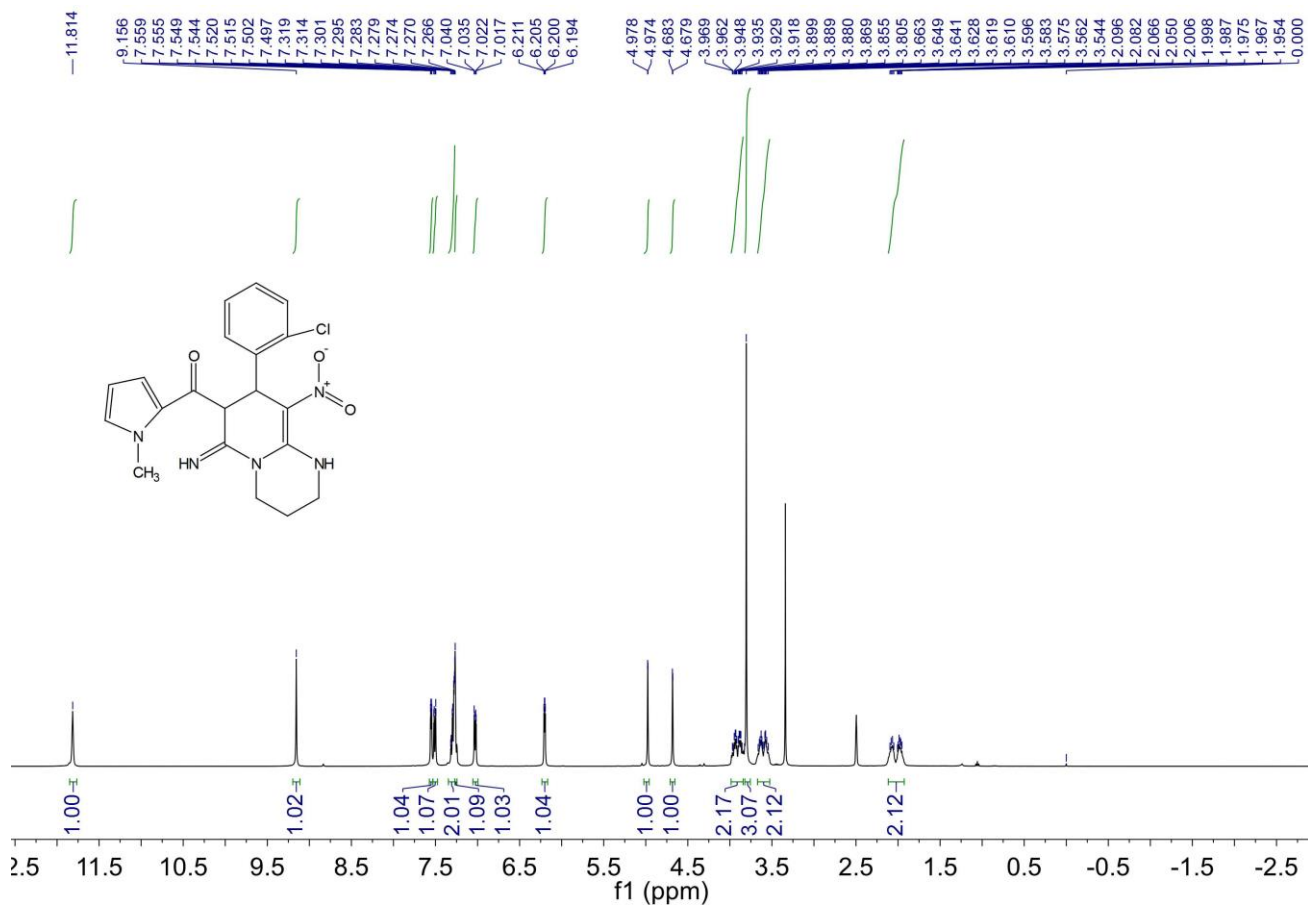


IR Spectrum of Compound 5e

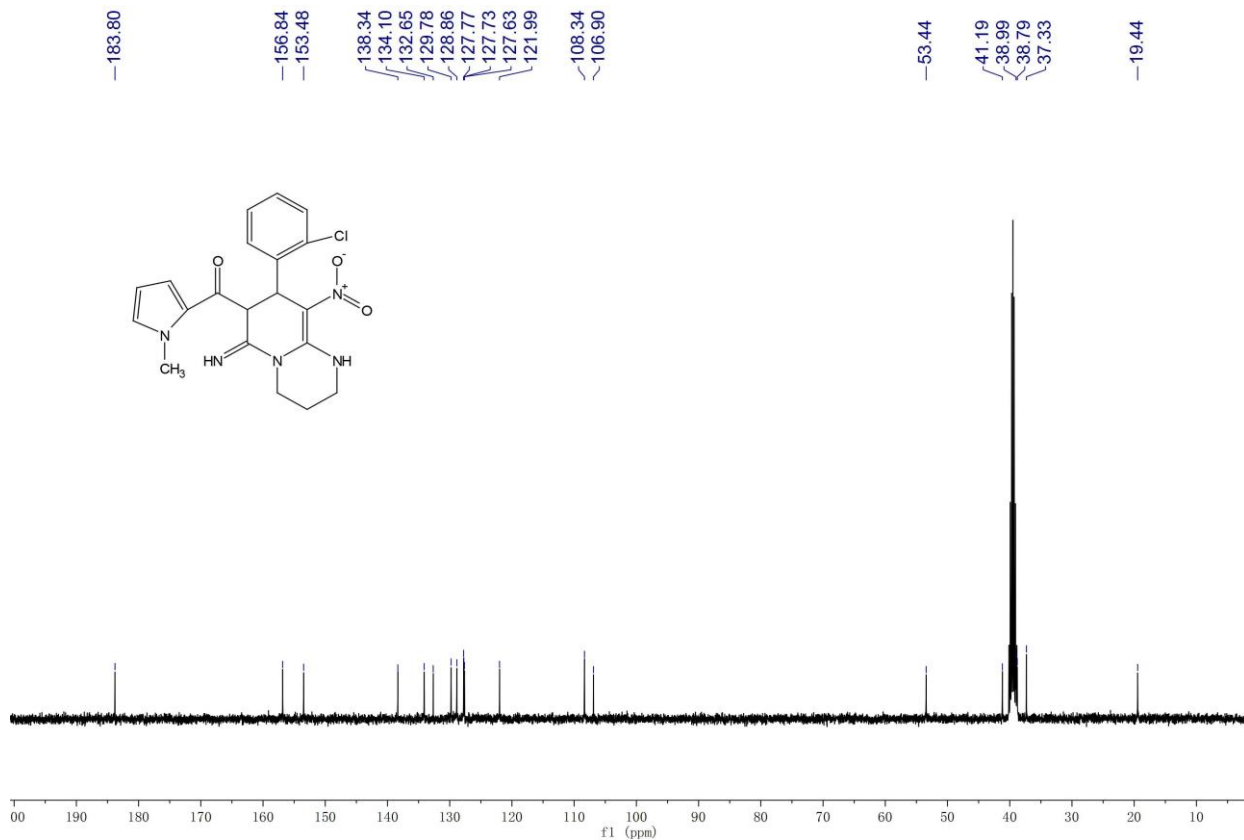




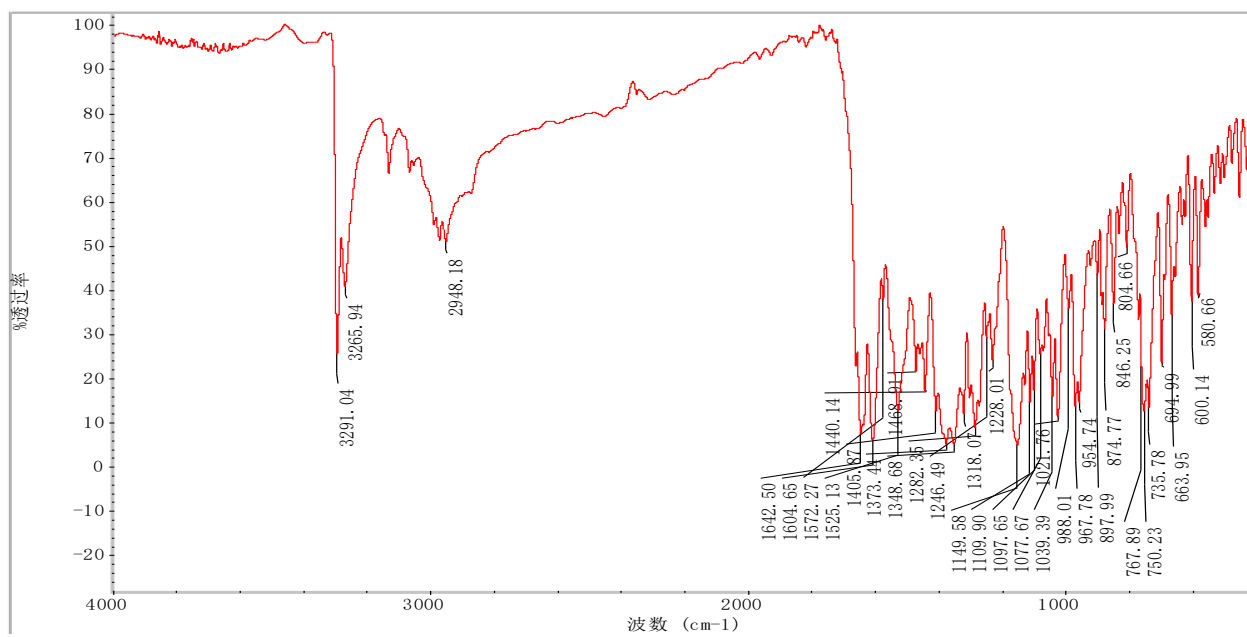
IR Spectrum of Compound 5f



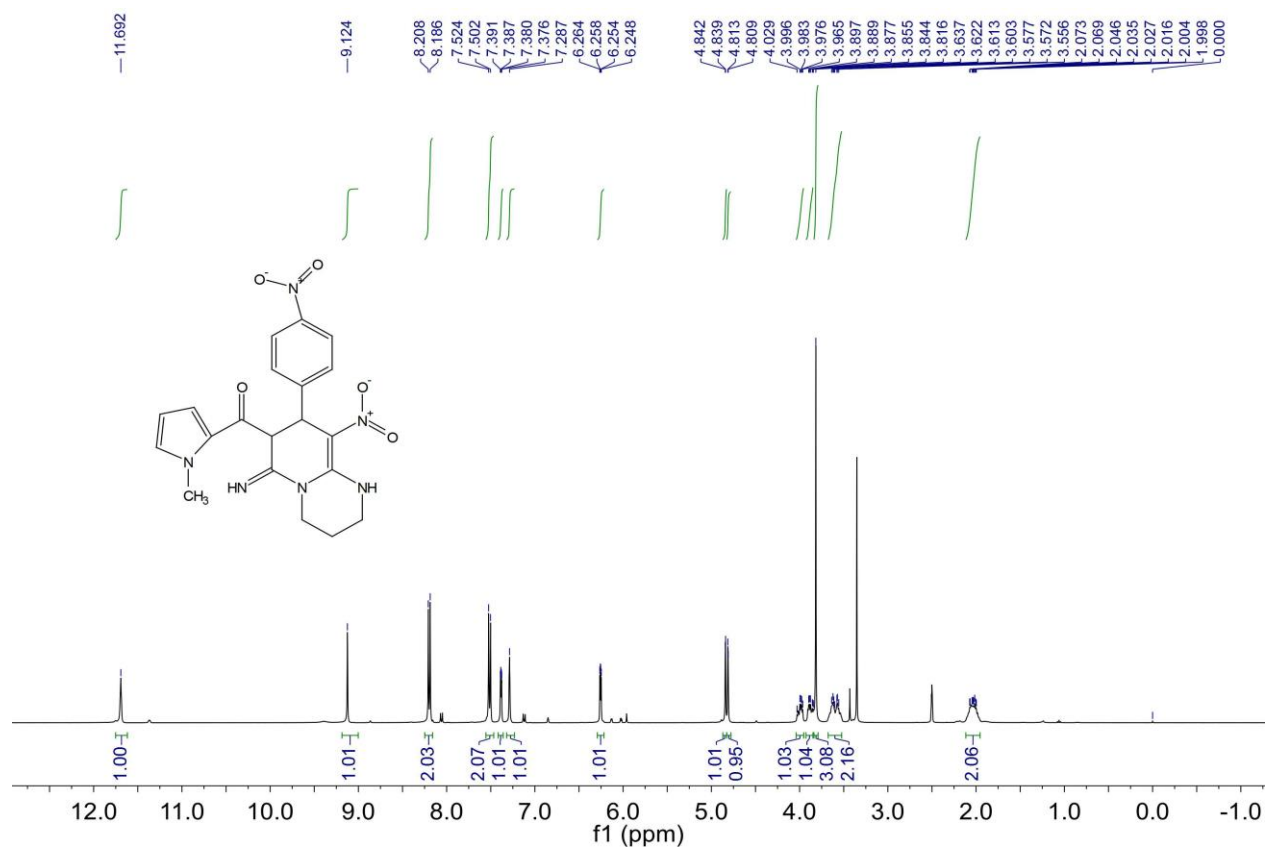
¹H-NMR Spectrum of Compound 5g



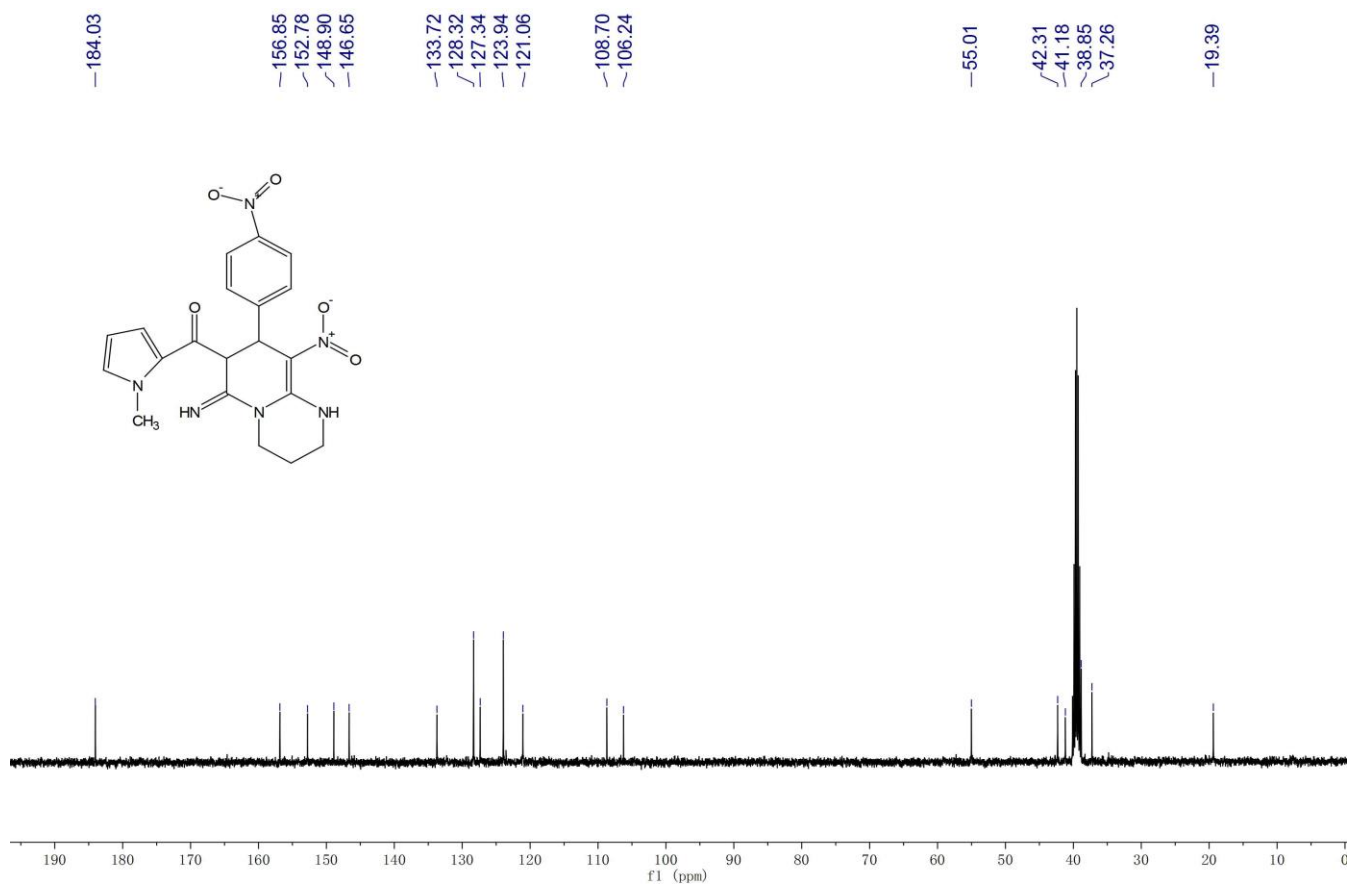
$^{13}\text{C-NMR}$ Spectrum of Compound 5g



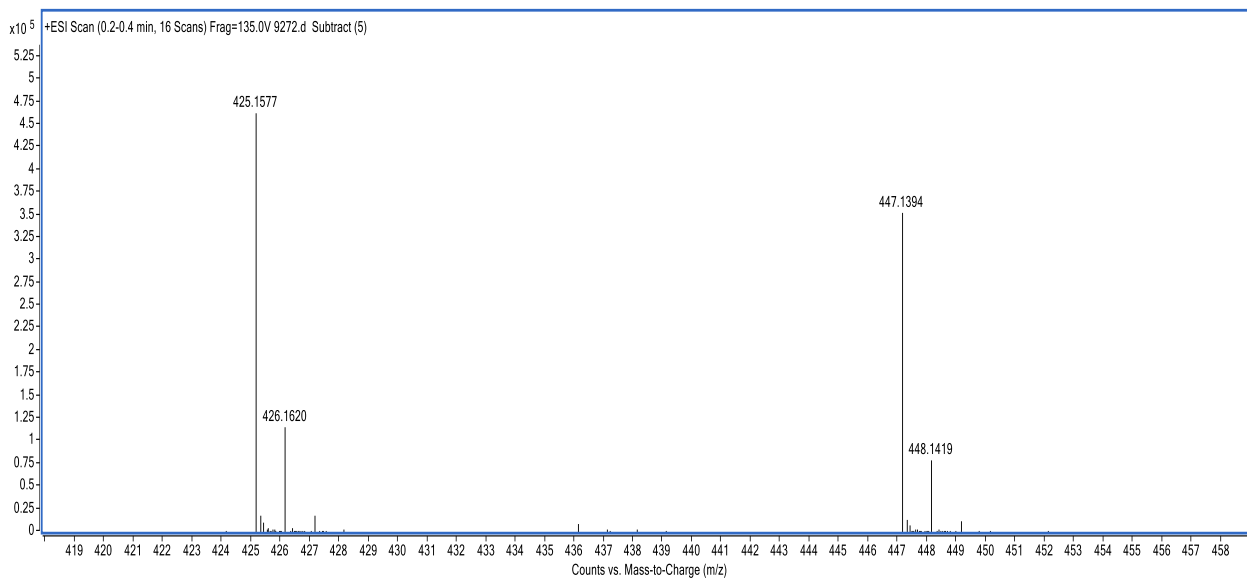
IR Spectrum of Compound 5g



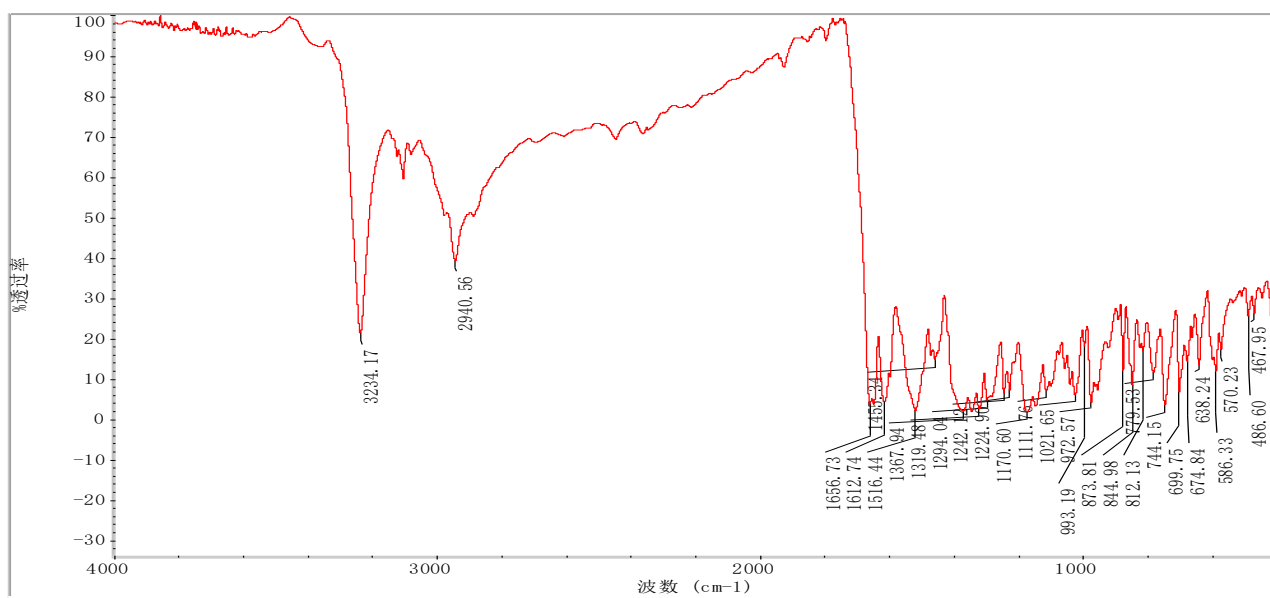
¹H-NMR Spectrum of Compound 5h



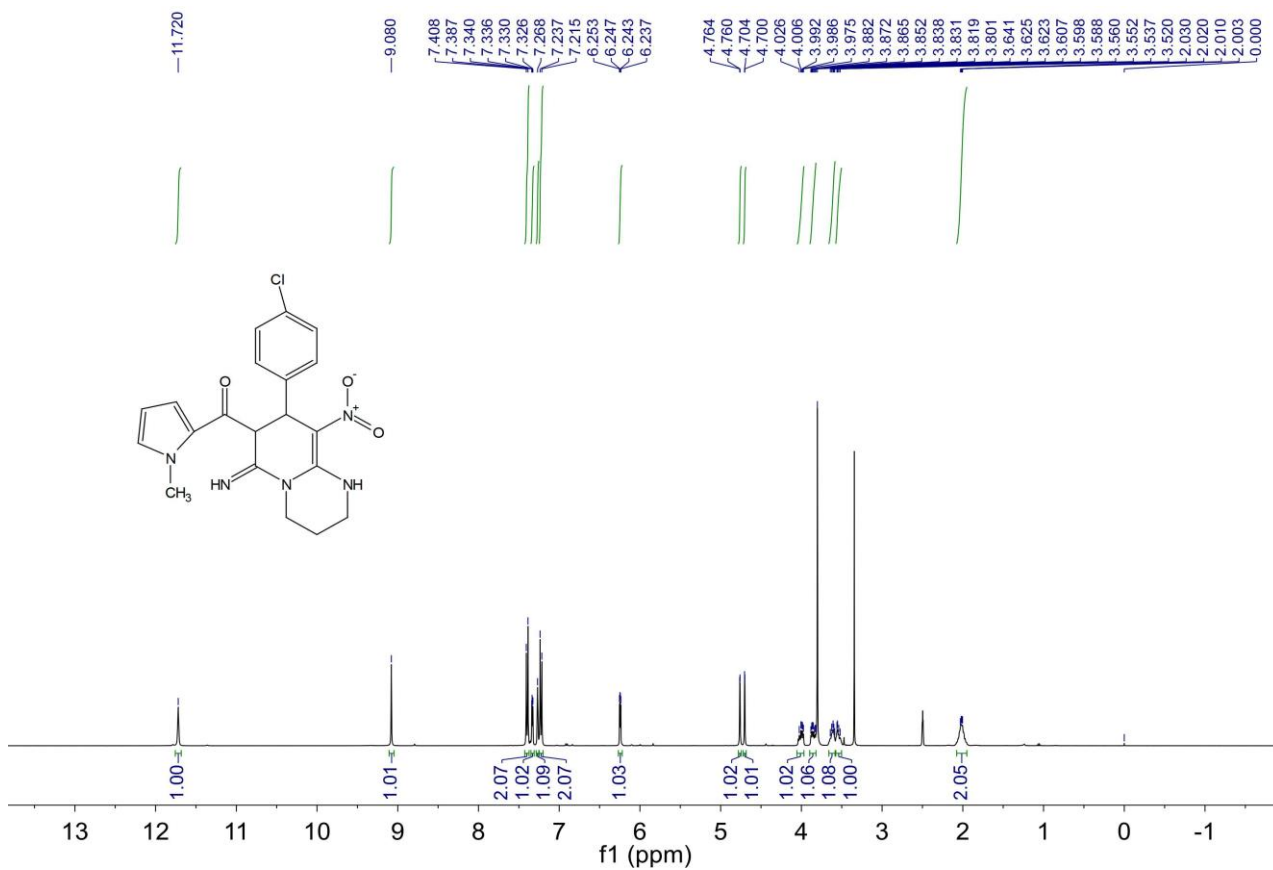
¹³C-NMR Spectrum of Compound 5h



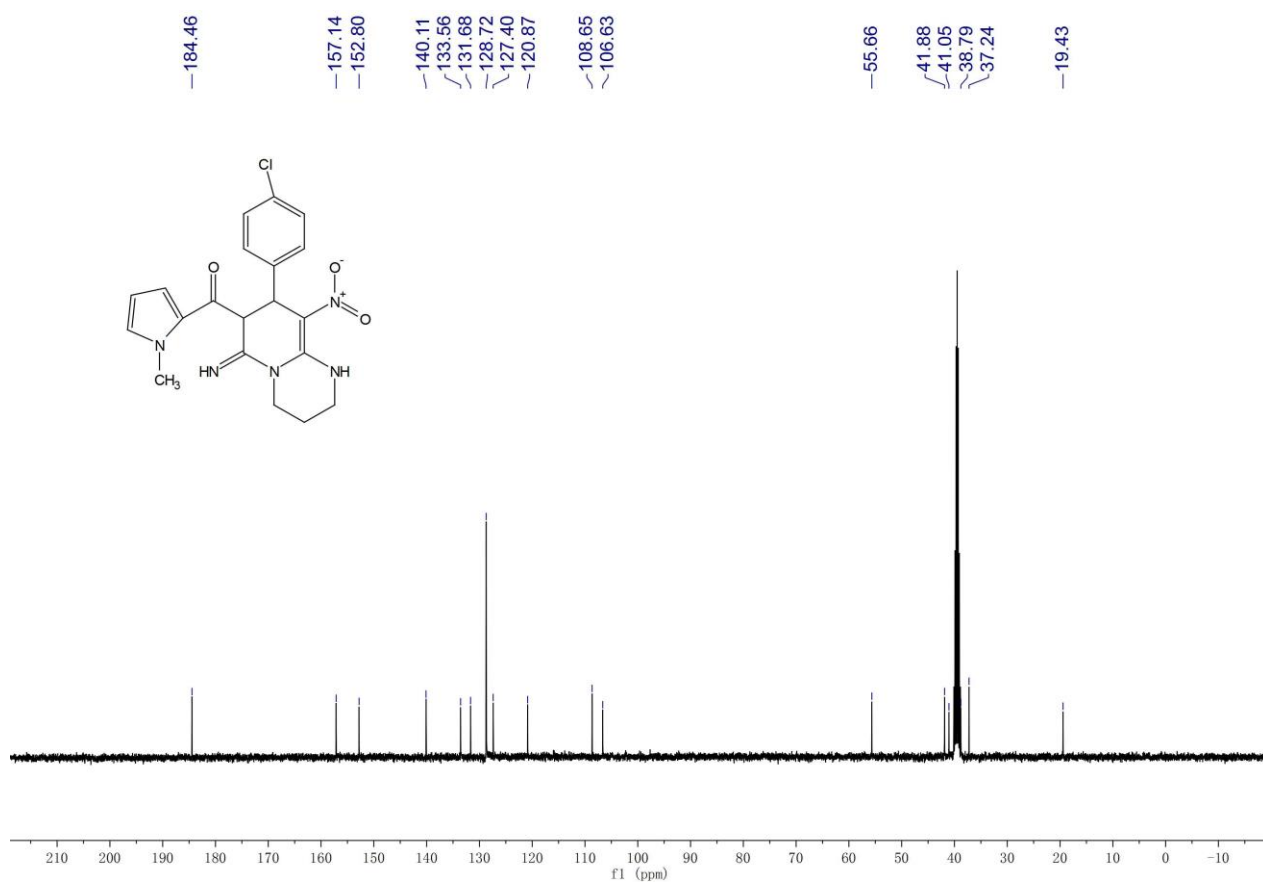
HRMS Spectrum of Compound 5h



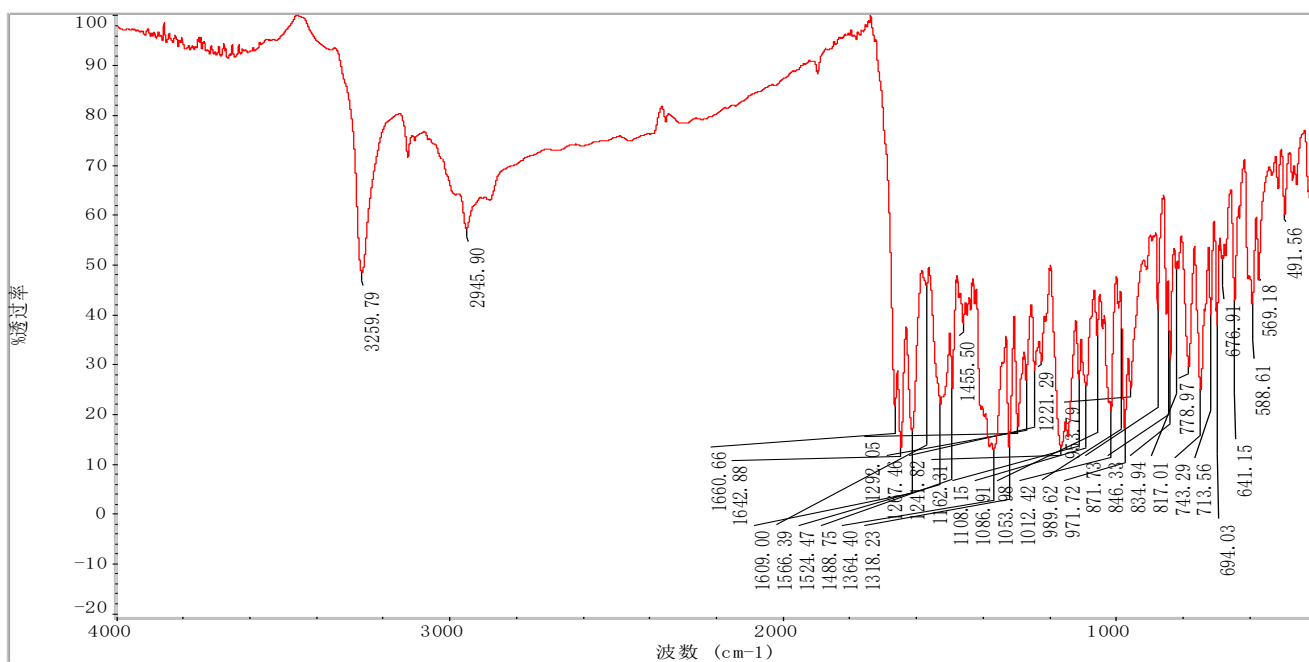
IR Spectrum of Compound 5h



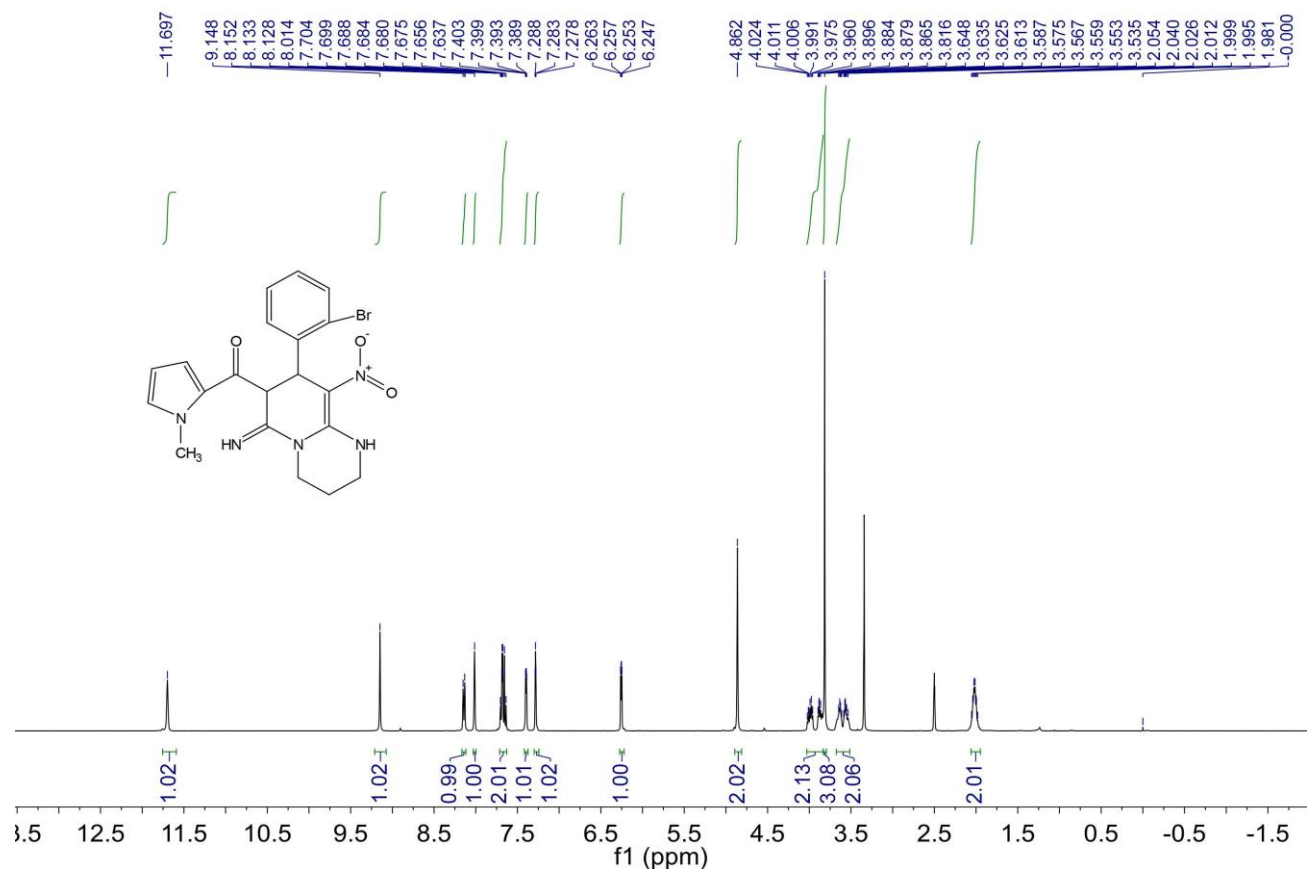
¹H-NMR Spectrum of Compound 5i



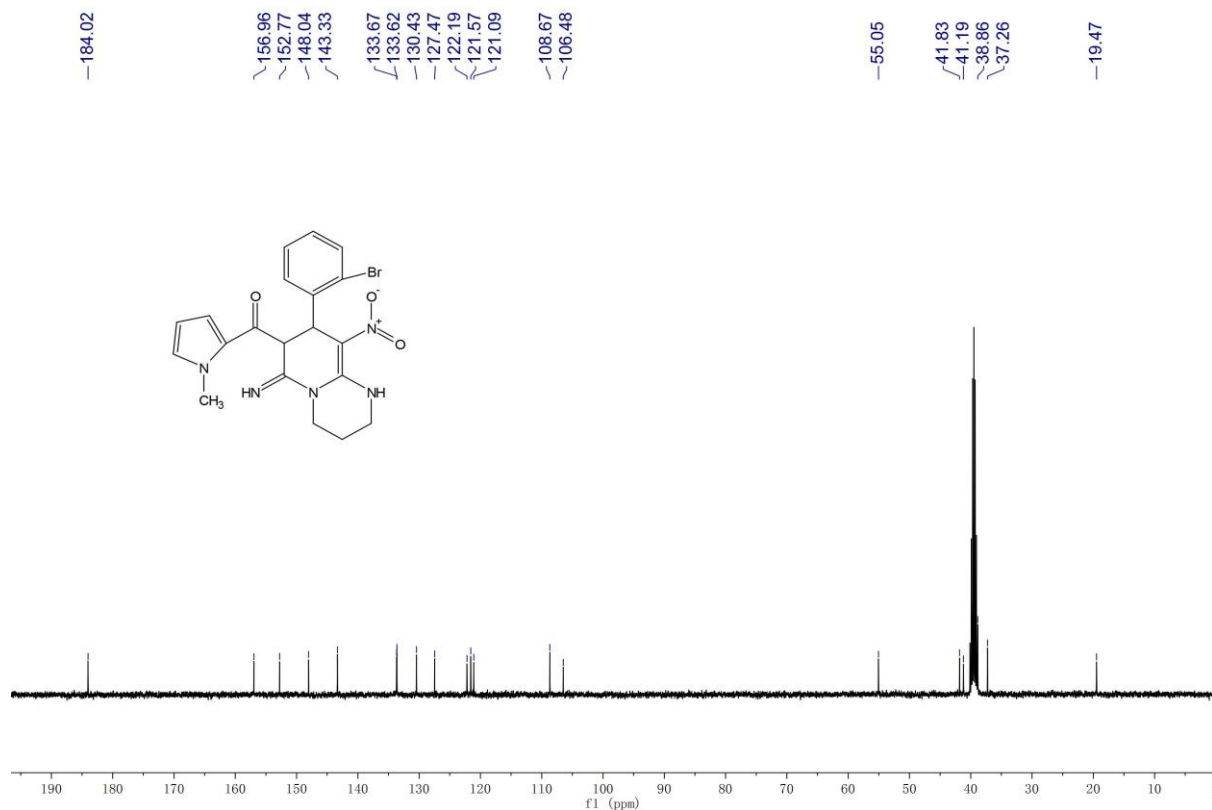
¹³C-NMR Spectrum of Compound 5i



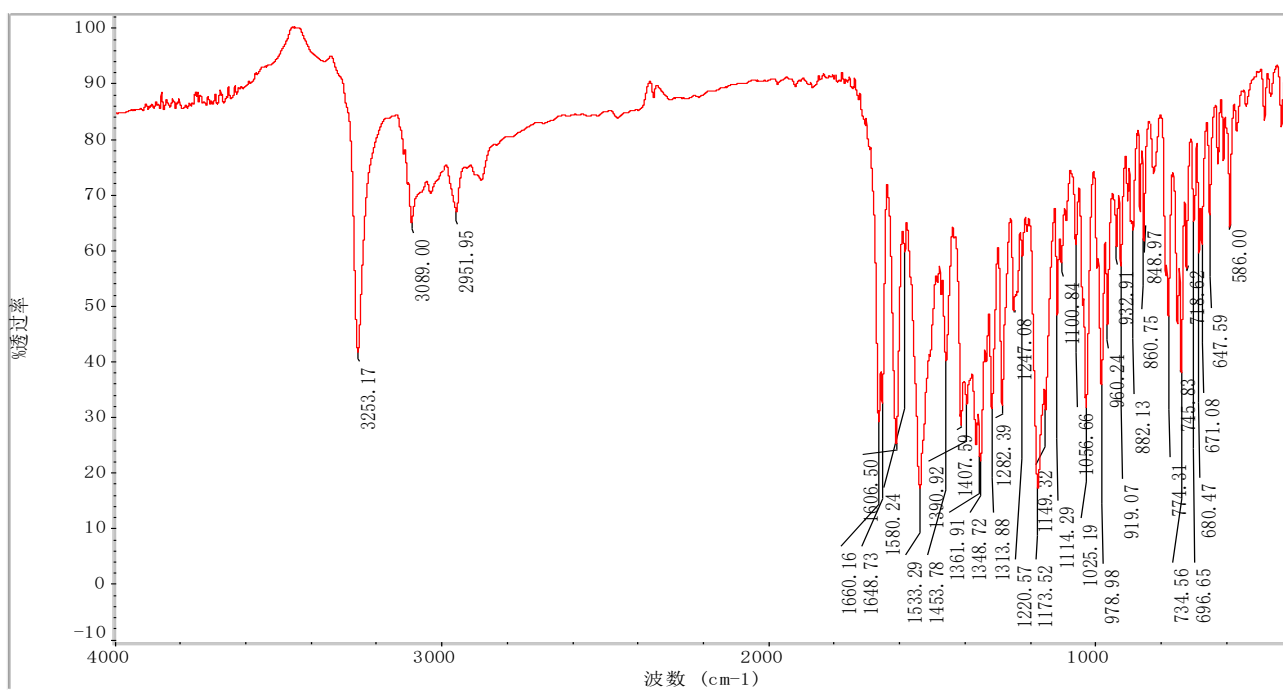
IR Spectrum of Compound 5i



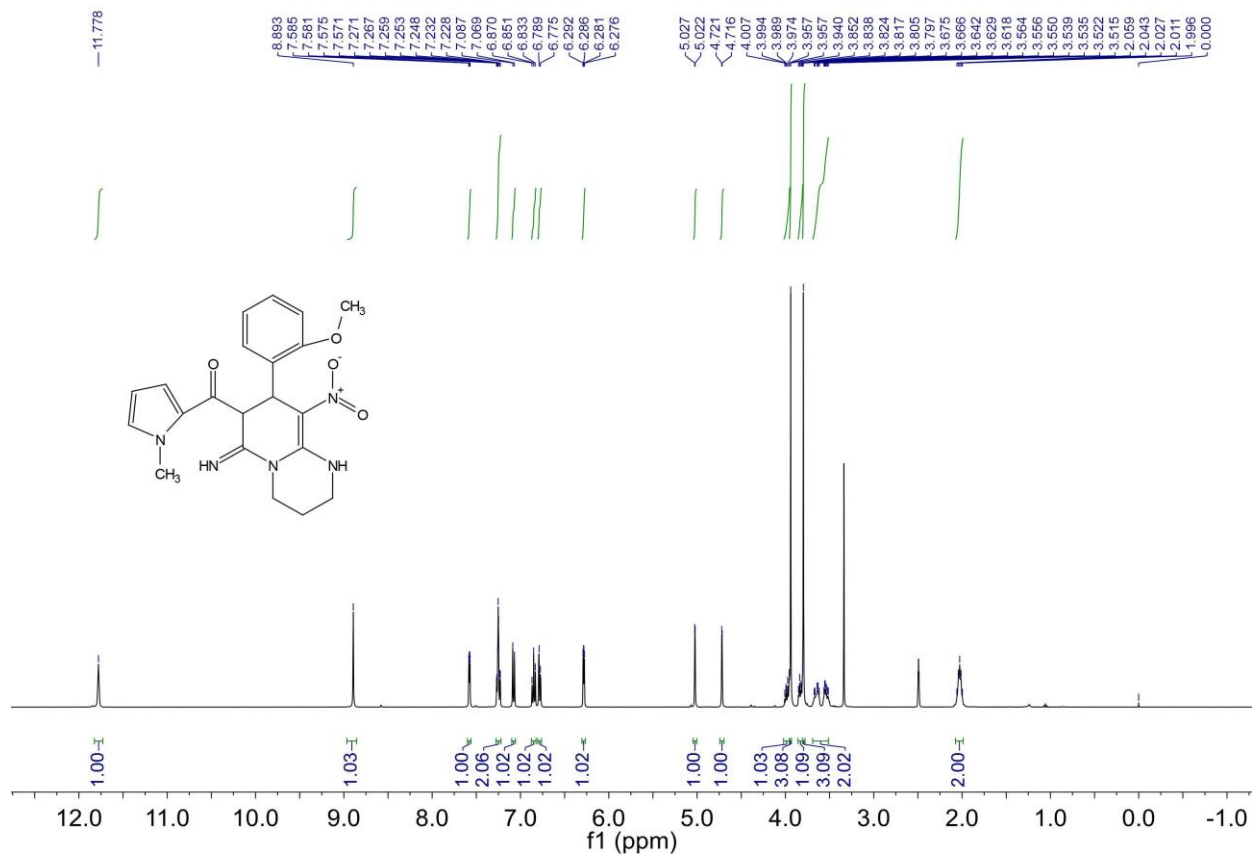
¹H-NMR Spectrum of Compound 5j



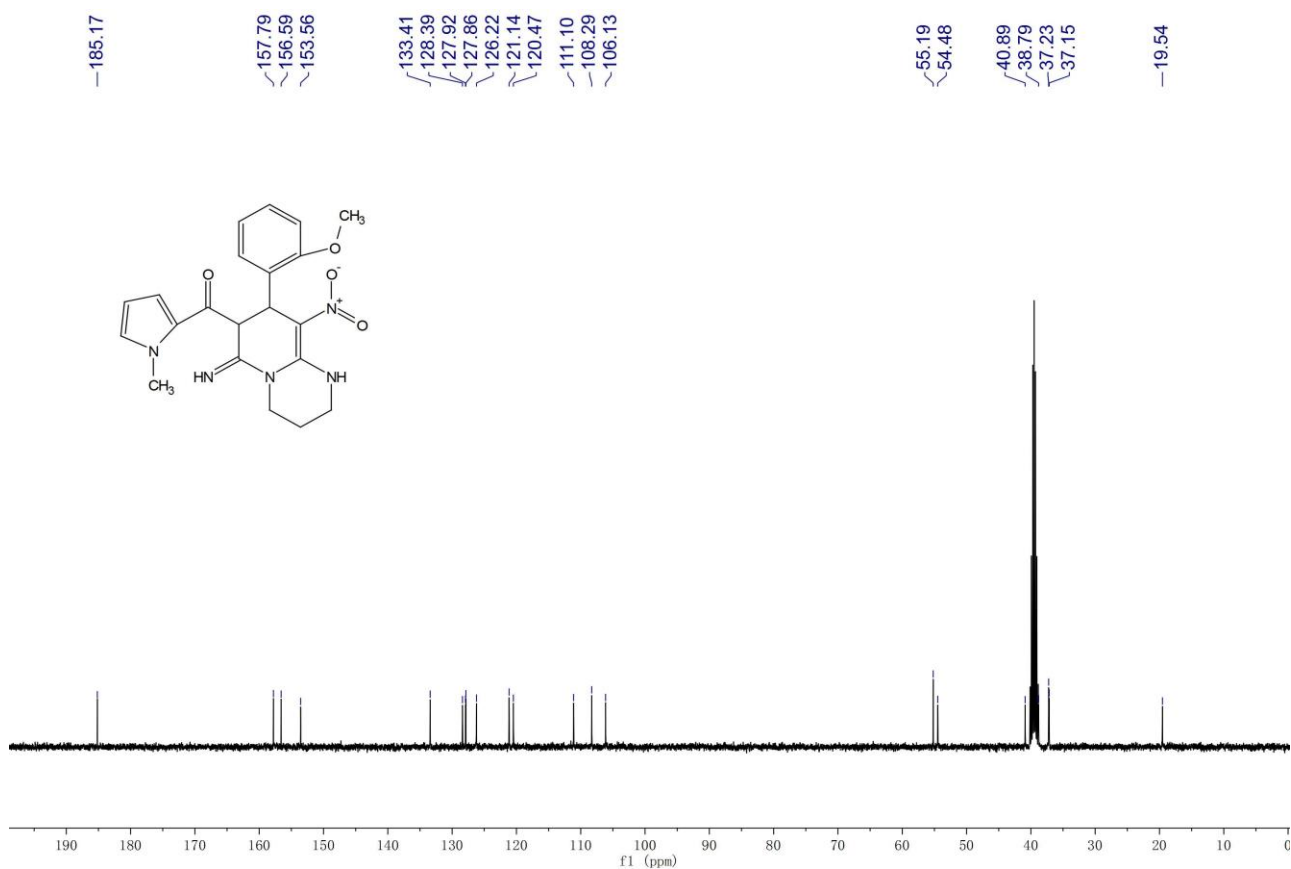
¹³C-NMR Spectrum of Compound 5j



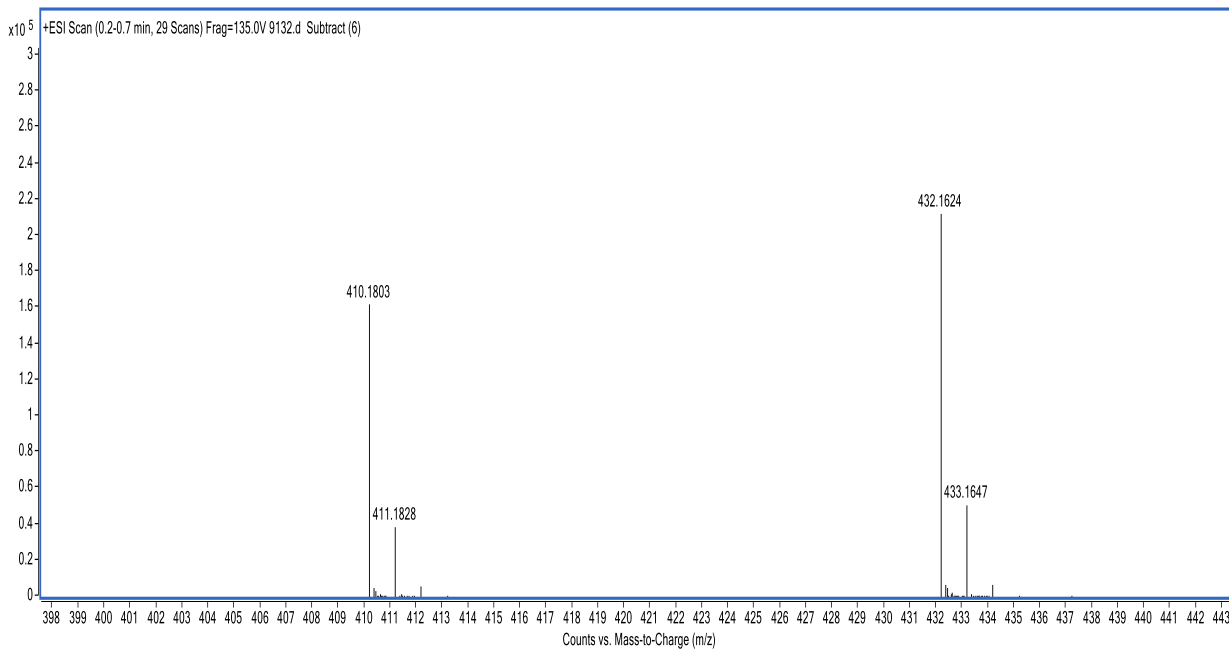
IR Spectrum of Compound 5j



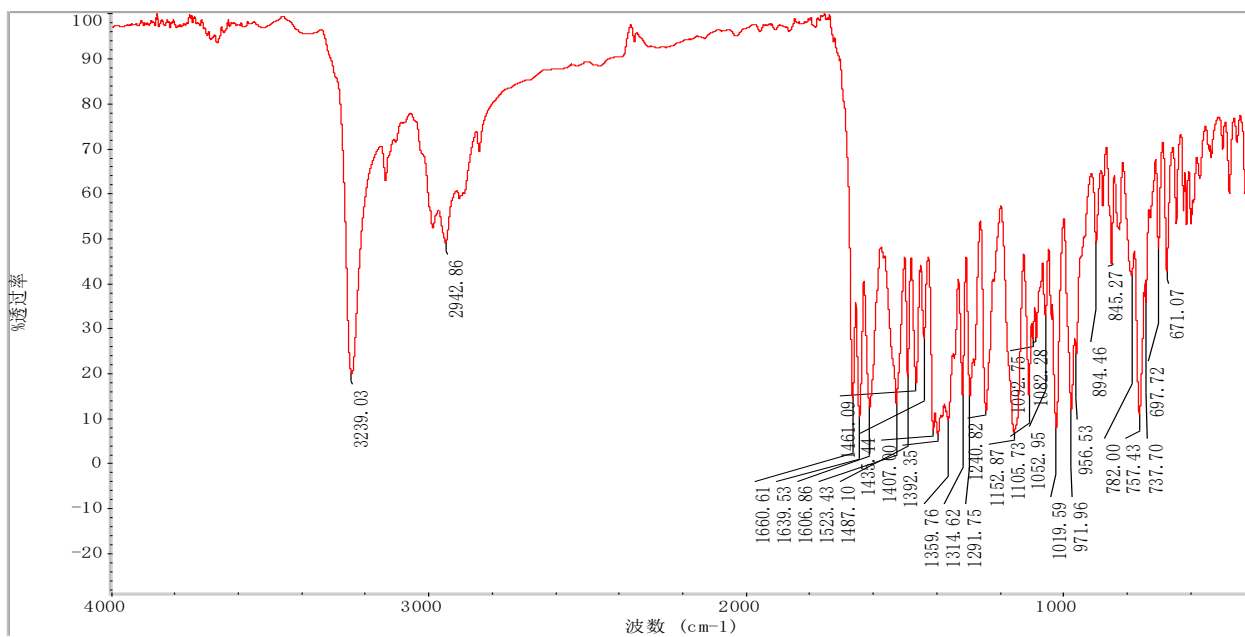
¹H-NMR Spectrum of Compound 5k



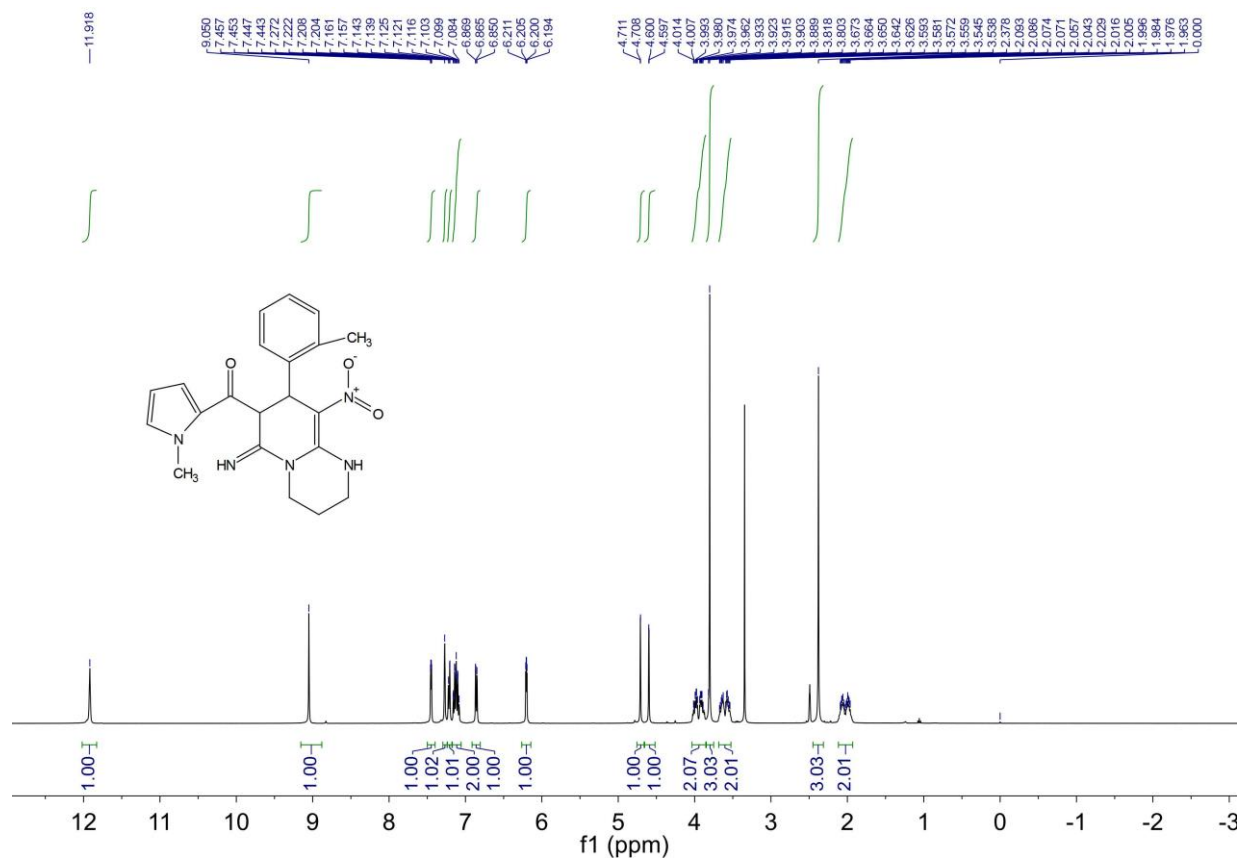
¹³C-NMR Spectrum of Compound 5k



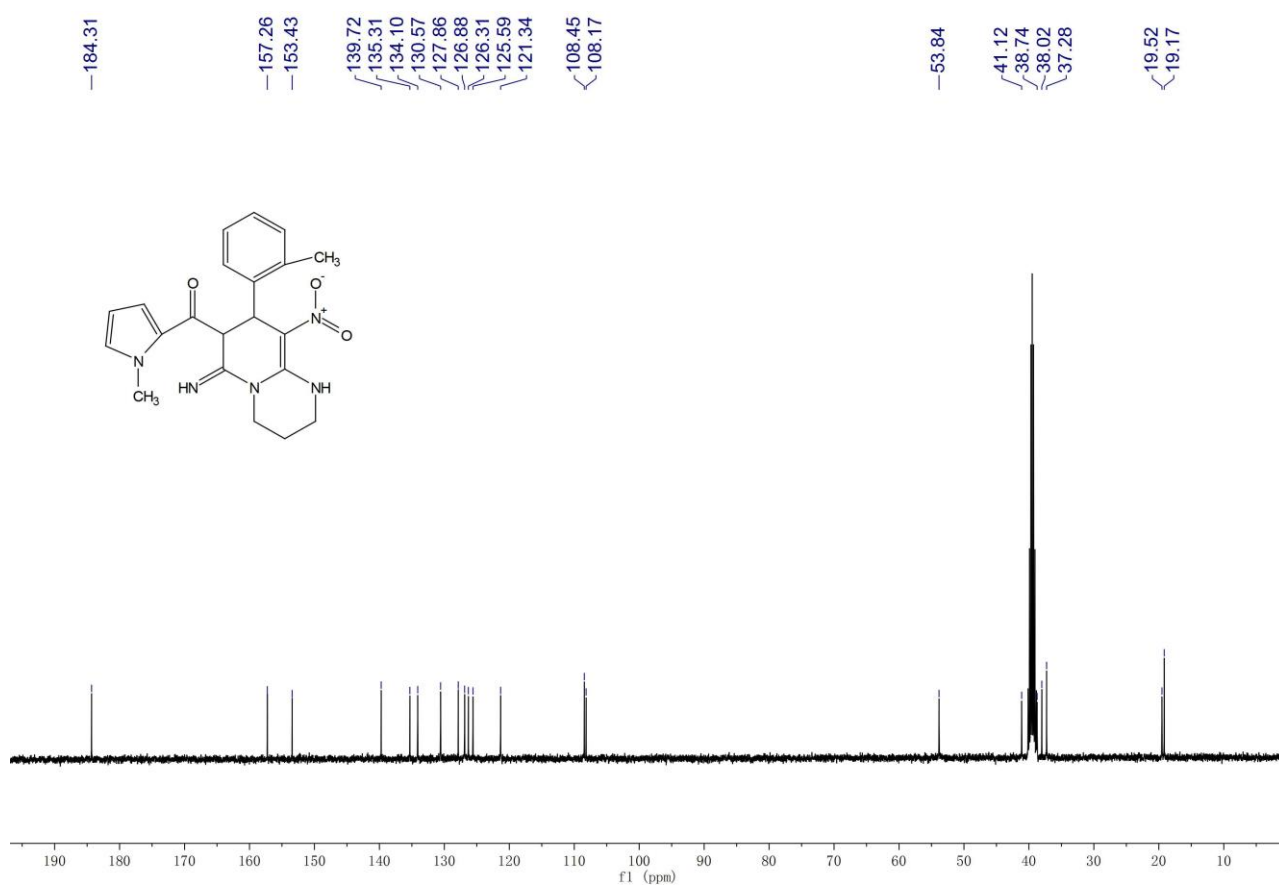
HRMS Spectrum of Compound 5k



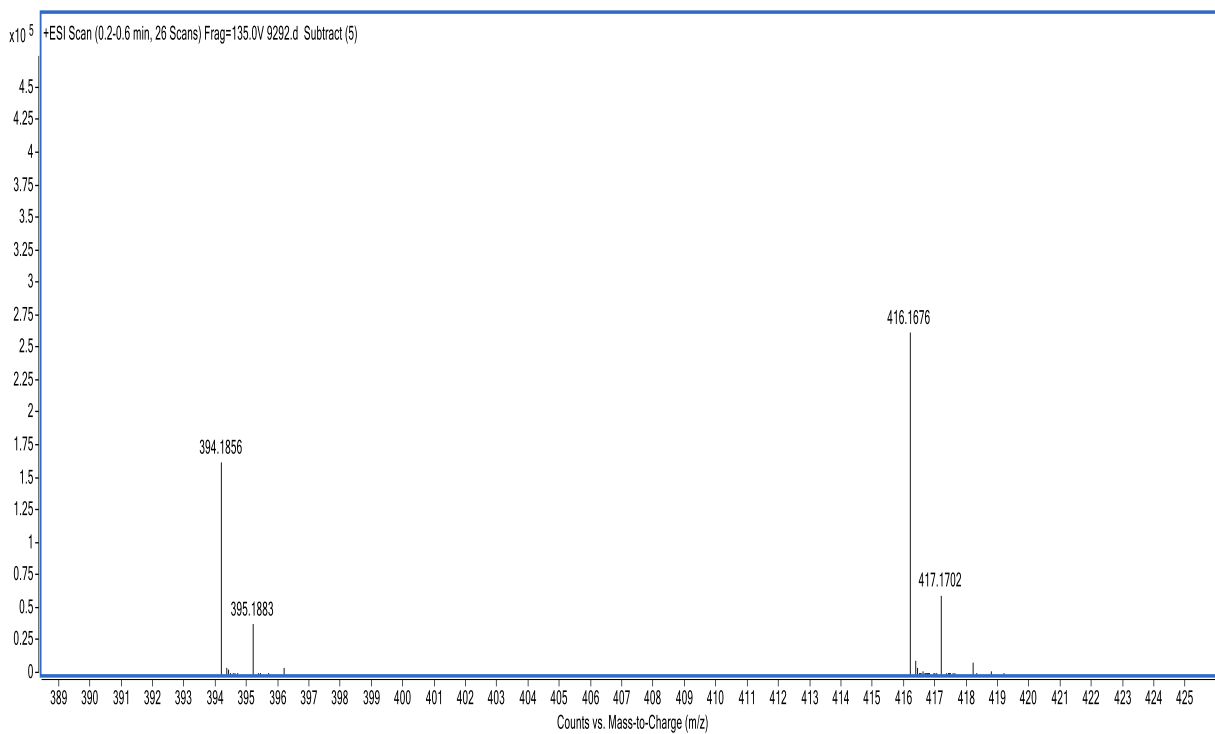
IR Spectrum of Compound 5k



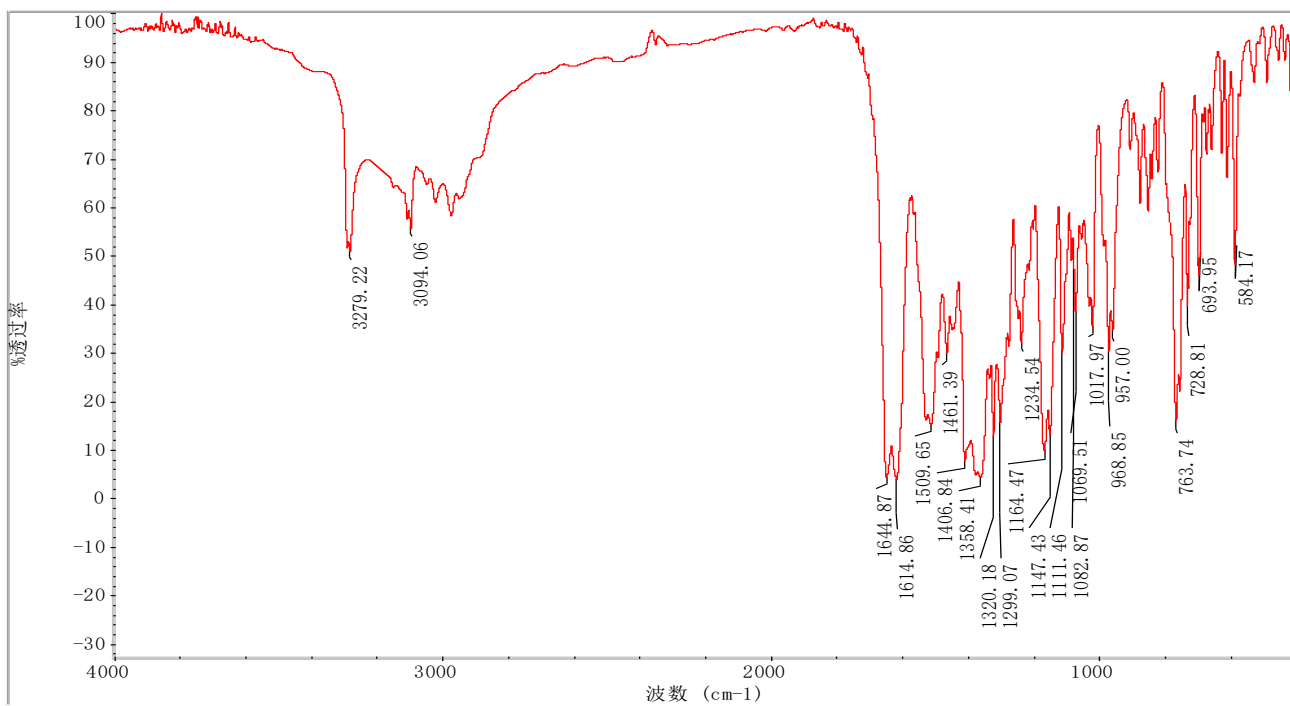
¹H-NMR Spectrum of Compound 51



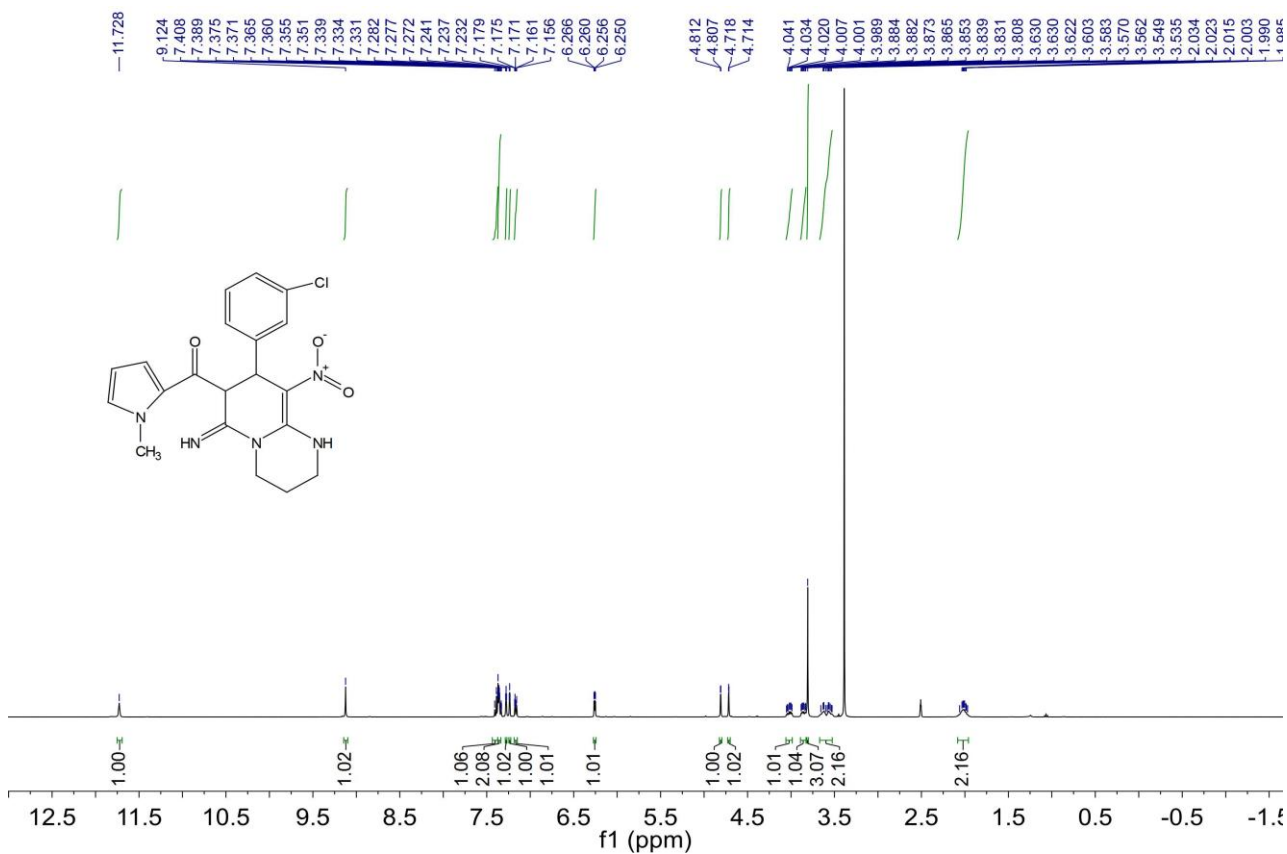
¹³C-NMR Spectrum of Compound 51



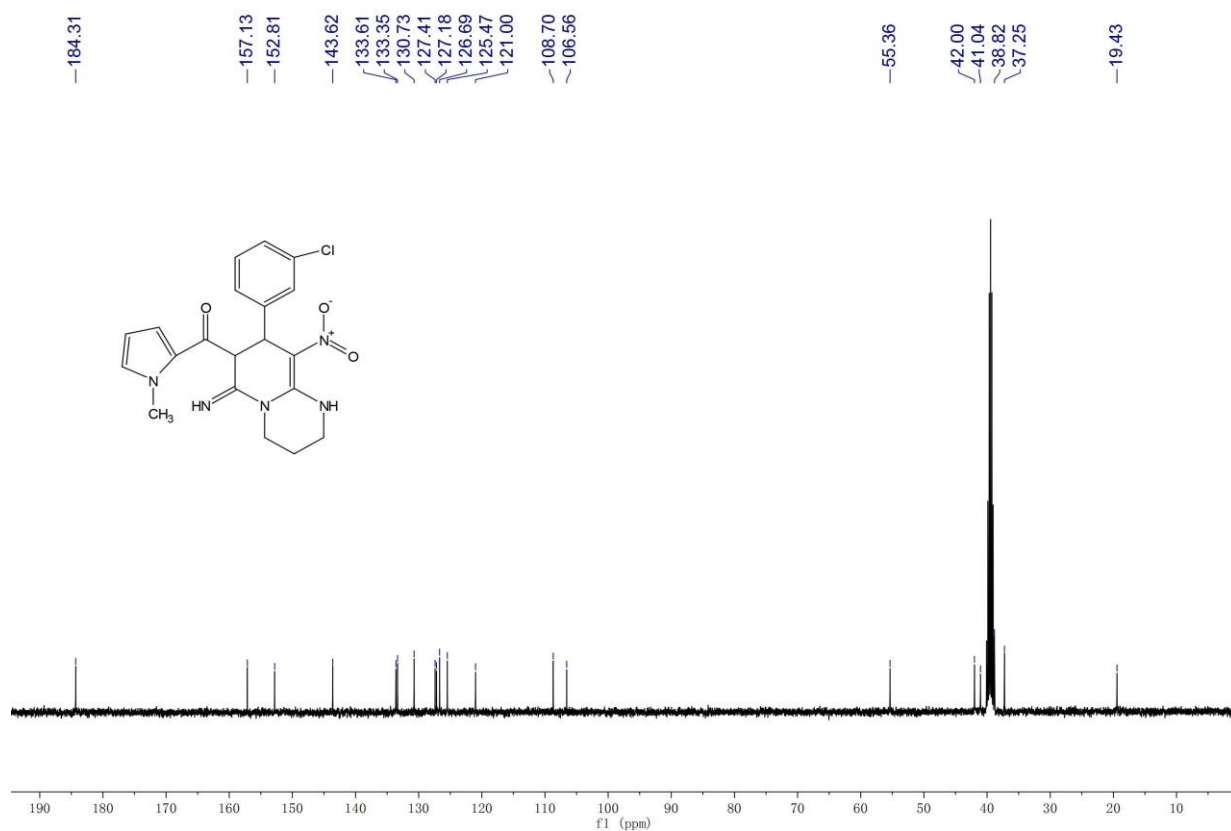
HRMS Spectrum of Compound 51



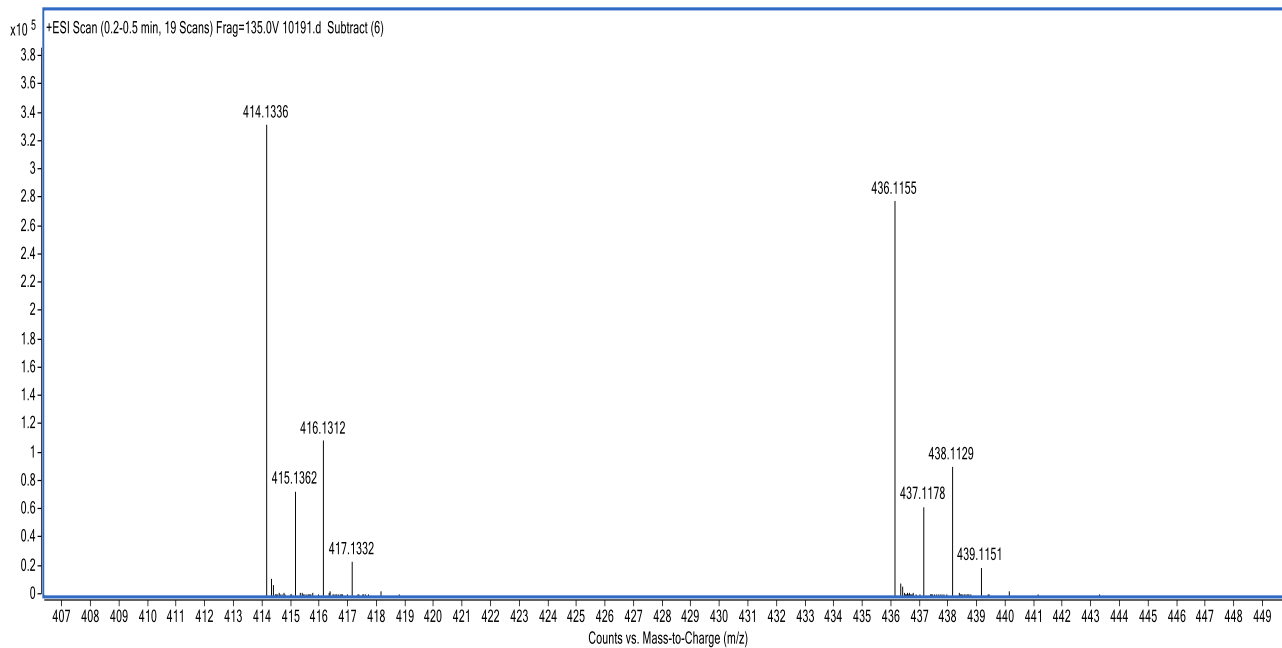
IR Spectrum of Compound 51



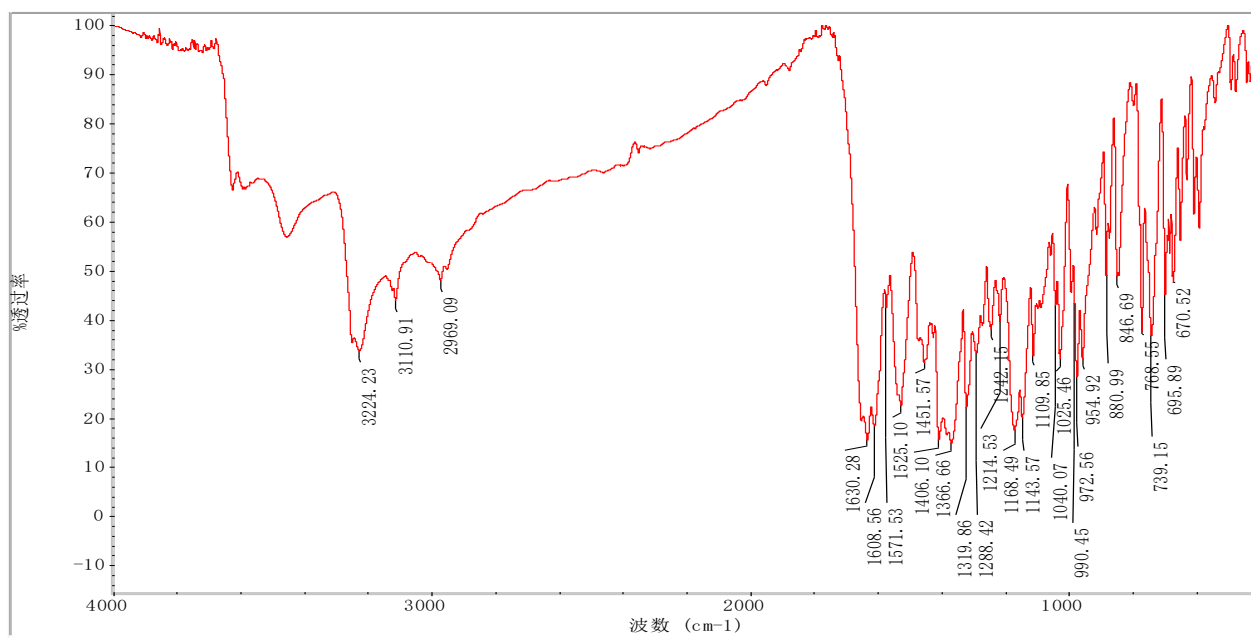
¹H-NMR Spectrum of Compound 5m



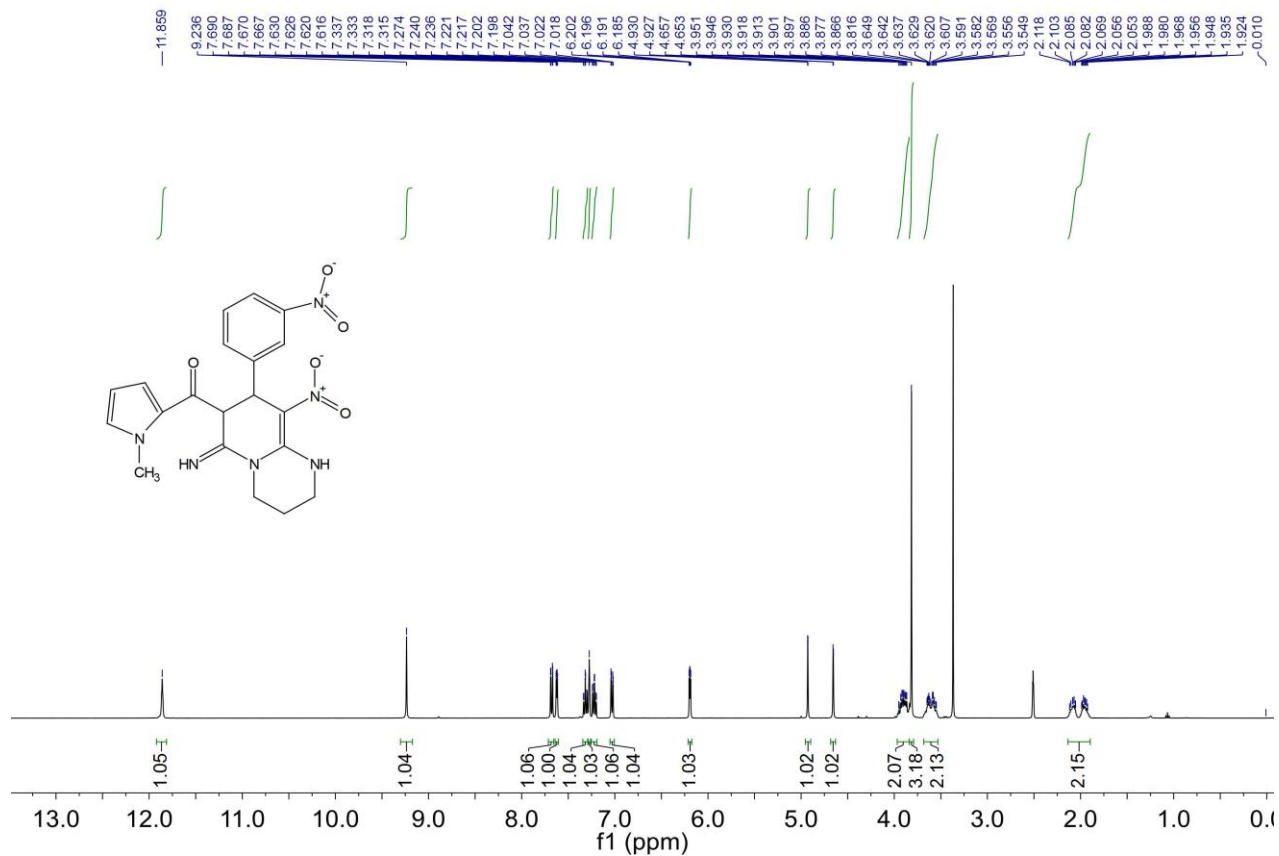
¹³C-NMR Spectrum of Compound 5m



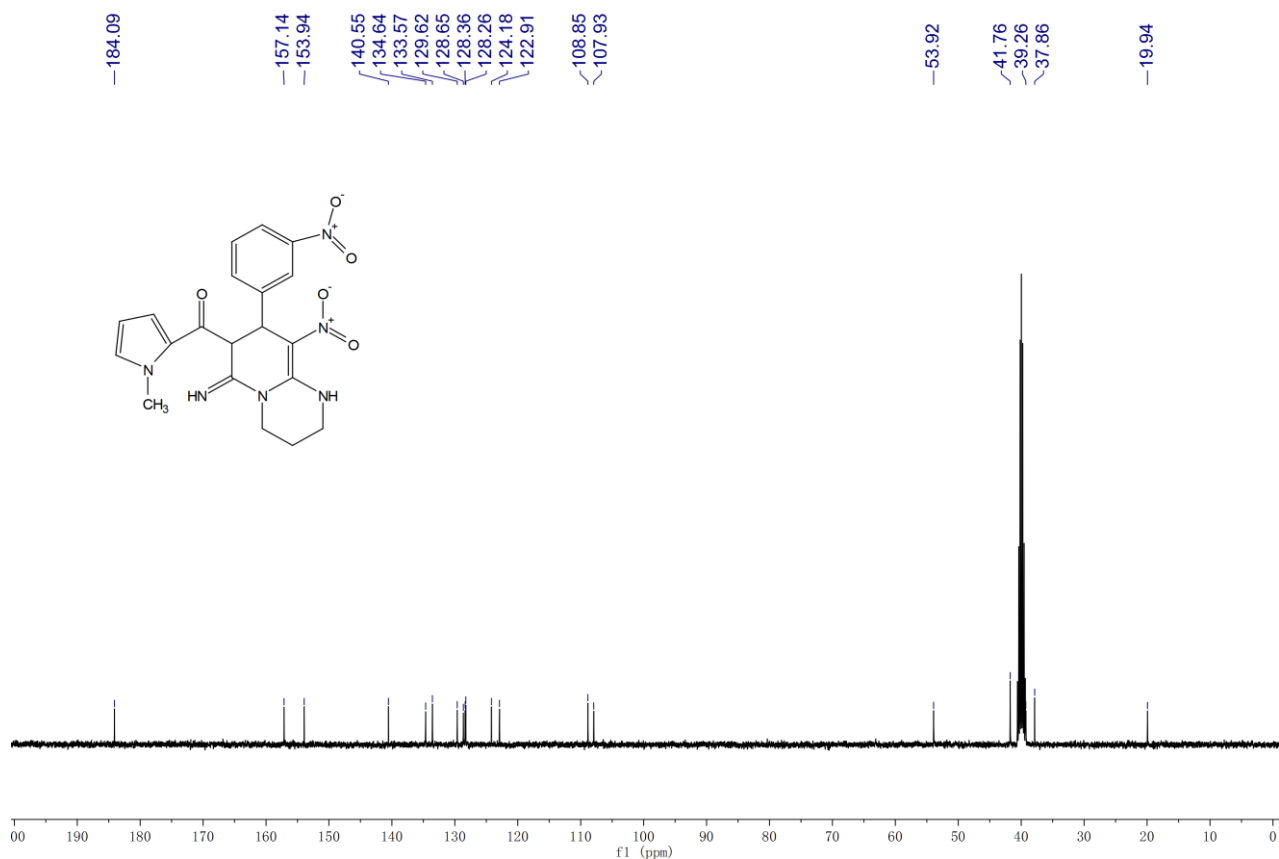
HRMS Spectrum of Compound 5m



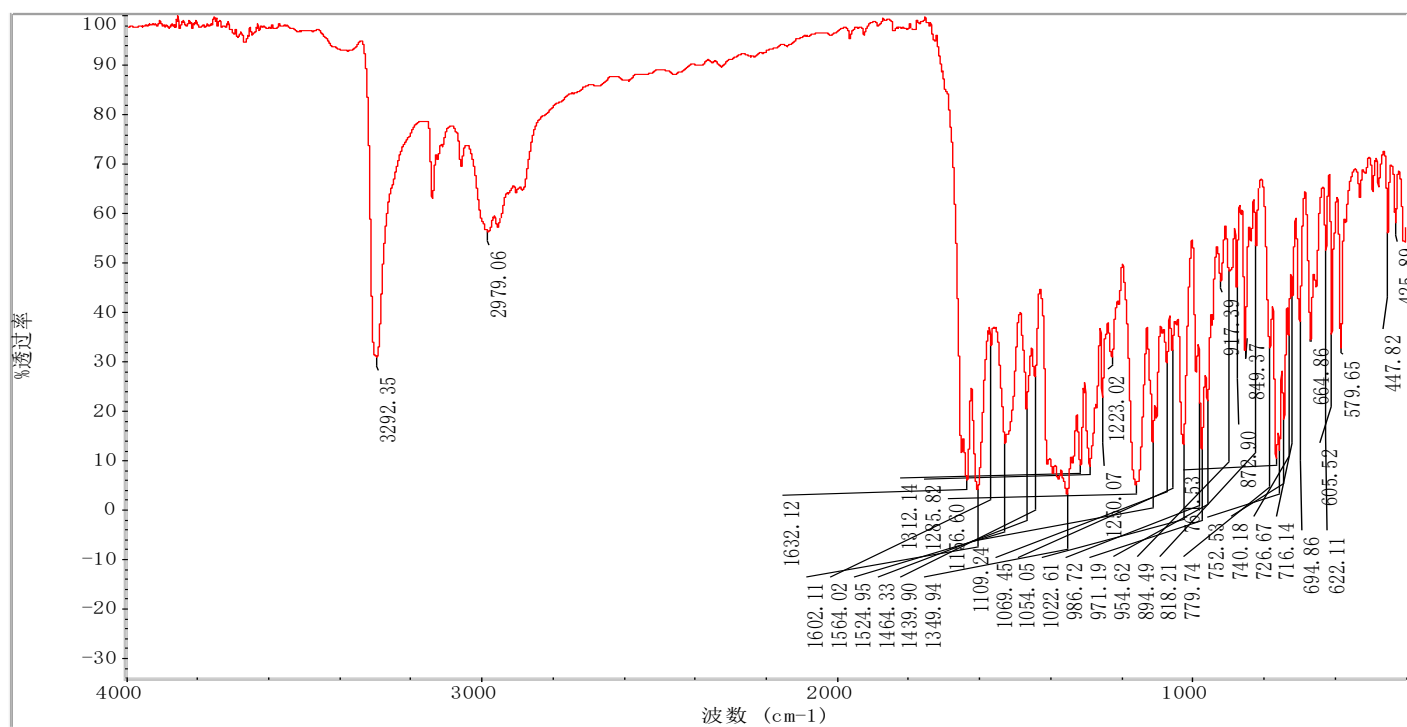
IR Spectrum of Compound 5m



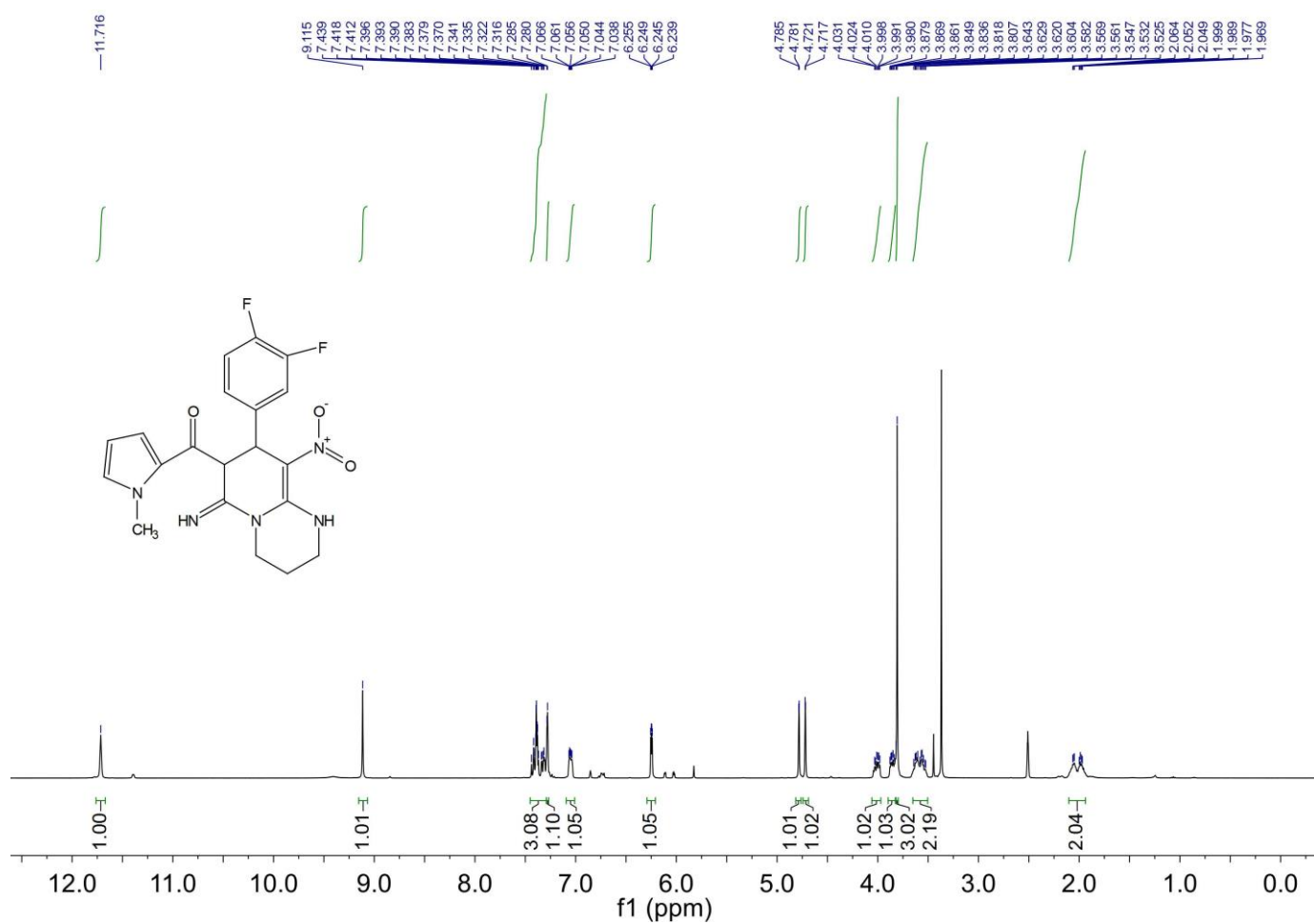
¹H-NMR Spectrum of Compound 5n



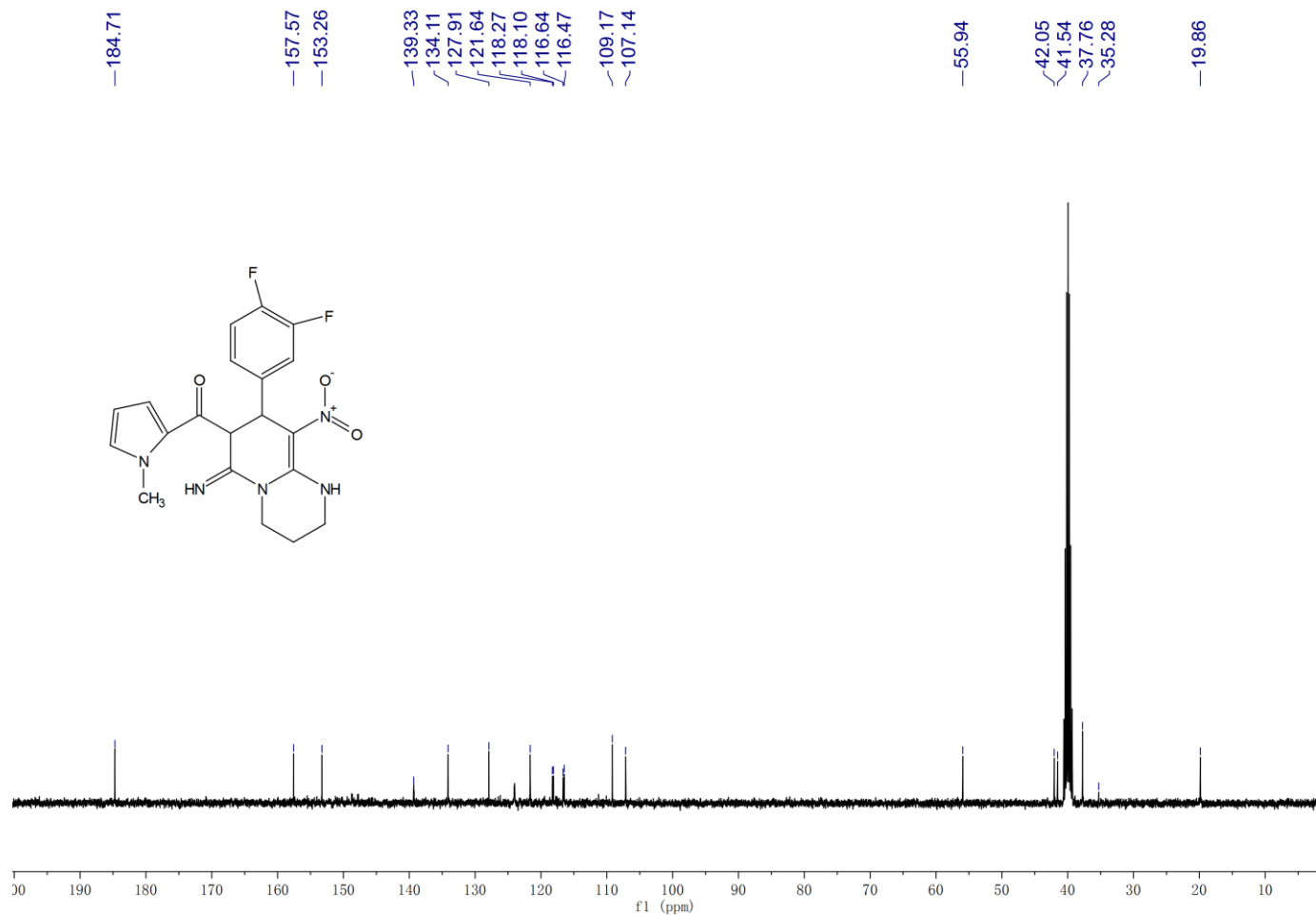
¹³C-NMR Spectrum of Compound 5n



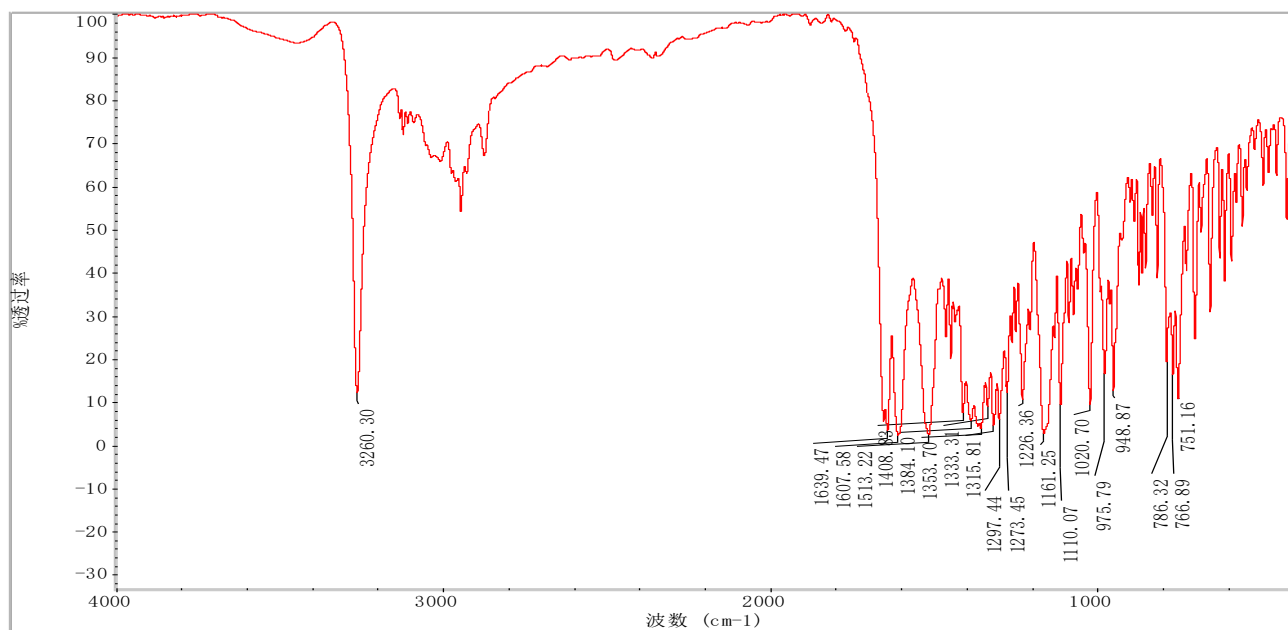
IR Spectrum of Compound 5n



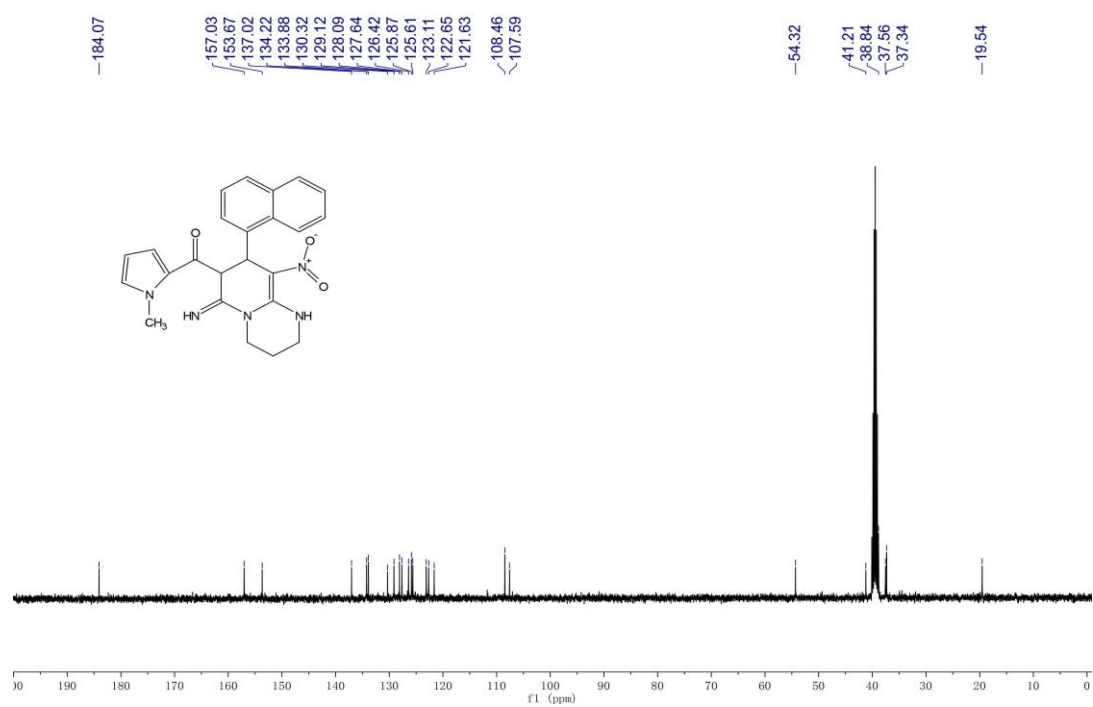
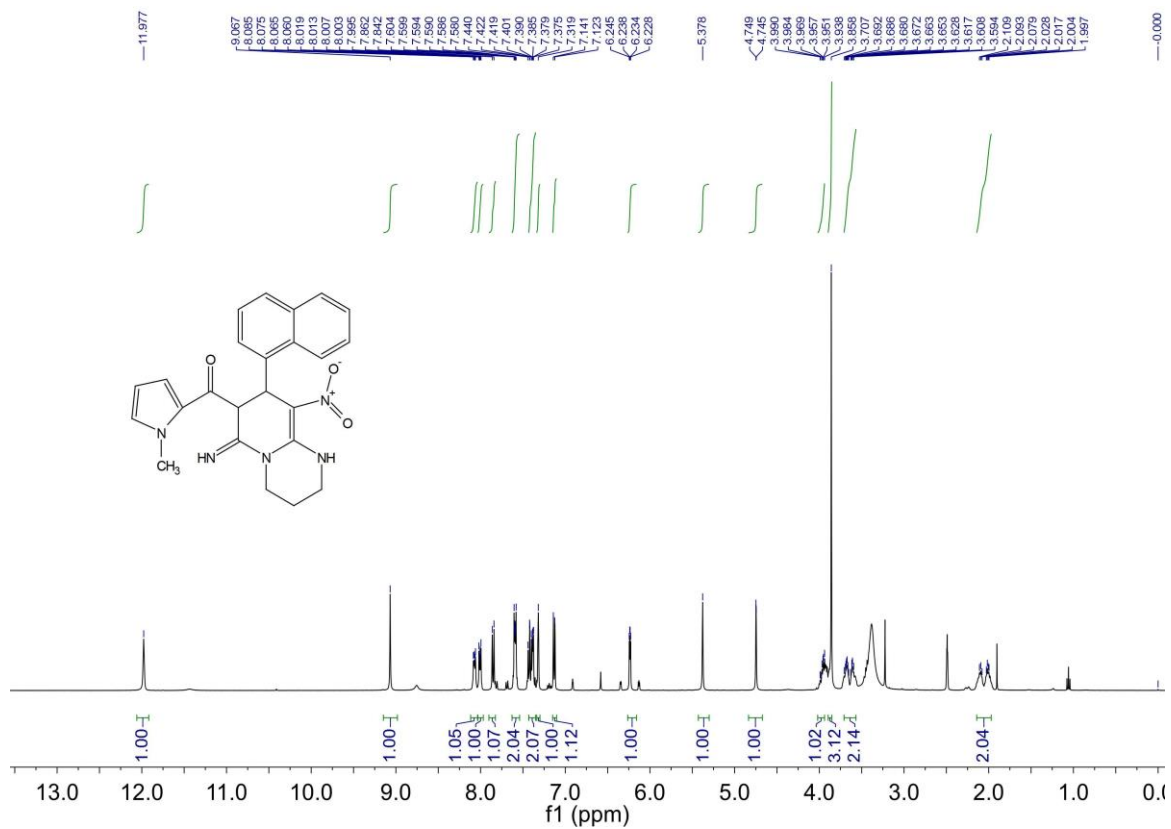
¹H-NMR Spectrum of Compound 5o

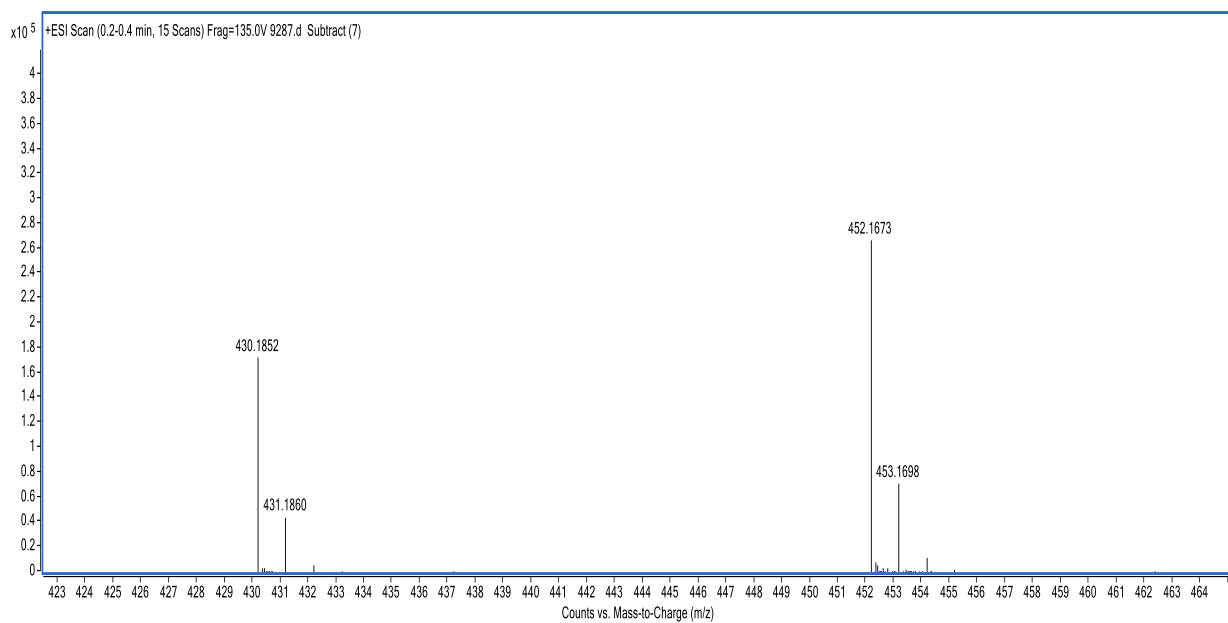


¹³C-NMR Spectrum of Compound 5o

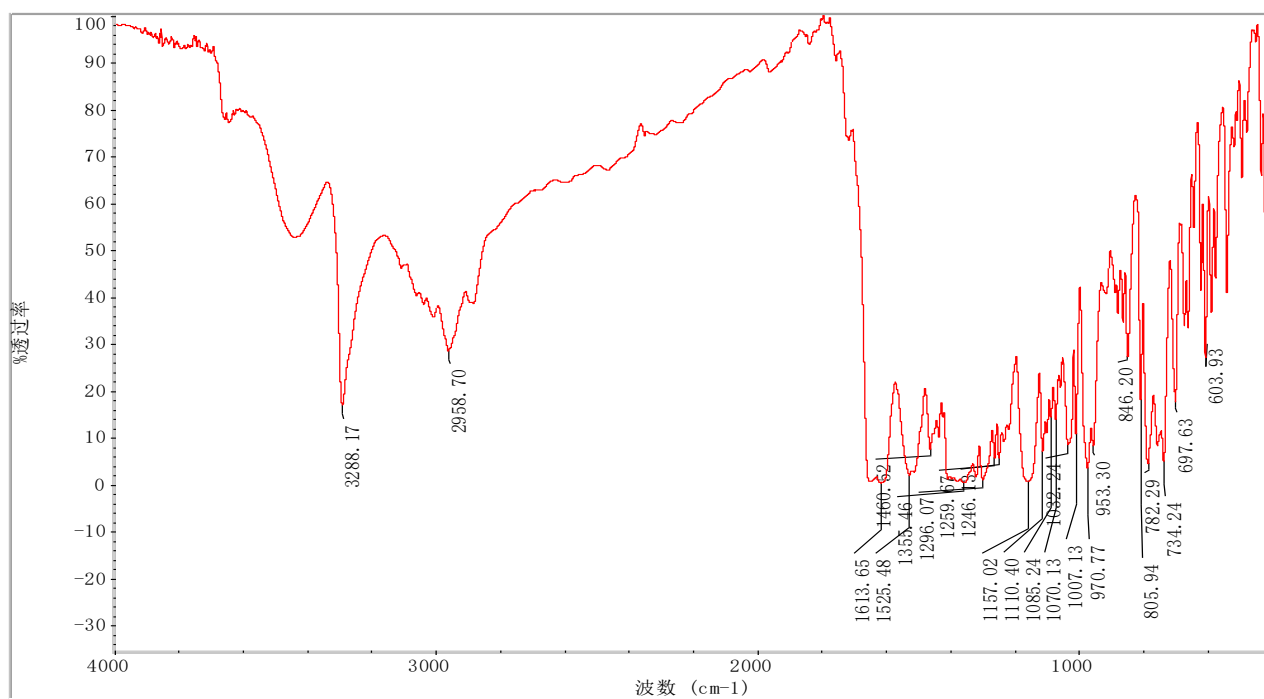


IR Spectrum of Compound 5n





HRMS Spectrum of Compound 5p



IR Spectrum of Compound 5p