

Supporting information

Phosphorous Acid Functionalized Polyacrylonitrile Fiber with Polarity
Tunable Surface Micro-environment for One-Pot C-C and C-N Bond
Formation Reactions

*Gang Xu^a, Lu Wang^a, Mengmeng Li^a, Minli Tao^a and Wenqin Zhang^{*a,b}*

^aDepartment of Chemistry, School of Sciences, Tianjin University, Tianjin, 300072, P. R. China.

^bCollaborative Innovation Center of Chemical Science and Engineering (Tianjin), Tianjin 300072, P. R. China.

* Corresponding author. E-mail: zhangwenqin@tju.edu.cn

1. Reagents and instruments

Commercially available polyacrylonitrile fiber (PANF, the number average molecular weight of the spinning solution is 53000 to 106000, 93.0% acrylonitrile, 6.5% methyl acrylate and 0.4-0.5% sodium styrene sulfonate) was cut into lengths of about 10 cm before use (from the Fushun Petrochemical Corporation of China). The aldehydes, 2-aminobenzamide, o-substituted aniline, ethanolamine, phosphorus oxychloride (POCl_3), n-butyl alcohol, benzylamine, 3-phenyl-1-propanol, and the other reagents *etc.* were analytical grade and used without further purification. The water was deionized.

The mechanical properties of different fiber samples were tested with an electronic single fiber strength tester YG(B)001A (Wenzhou Darong Textile Instrument Corporation, China) with clamping distance of 10 mm and an elongation speed of 20 mm min⁻¹ at room temperature. The FTIR spectra were obtained by an AVATAR360 FTIR spectrometer (Thermo Nicolet), all the samples were pulverized by cutting and made into pellets with KBr. The Elemental analysis (EA) were performed on a vario micro cube (Elementar). Scanning electron microscopy (SEM) (Hitachi, model S-4800) was used to characterize the surface of the modified fibers. X-ray powder diffraction spectra (XRD) were recorded by a D/MAX-2500 X-ray diffractometer (Rigaku Corporation). Thermodynamic characteristics fiber were investigated using a STA409PC TGA/ DSC simultaneous thermal analyzer (Netzsch company, Germany). ¹H NMR (400 MHz) and ¹³C NMR (101 MHz) spectra were recorded on BRUKER-AVANCE III instruments using tetramethylsilane as the internal standard. Nitrogen adsorption and desorption isotherms were measured by a Auto-sorb-iQA3200-4 surface area and pore size analyzer (Connor Instruments, USA). The samples were outgassed

at 200 °C overnight before measurements were made. HPLC analyses were performed on Waters Model 510.

2. Synthesis of the hydroxyethylation fiber

2.1 Synthesis of the PAN_{EA}F

Dried PANF (4.0 g), ethanolamine (40 mL) and water (60 mL) were placed in a three-necked flask. After the mixture was refluxed for 4 h, the fiber was filtered out and washed with water (60-70 °C) until no ethanolamine was attached to the fiber surface by non-covalent bonds, and then dried at 60 °C overnight to collect the hydroxyethylation fiber (PAN_{EA}F). The weight gain of PAN_{EA}F based on PANF was 15.1%.

2.2 Synthesis of the Bn-PAN_{EA}F

Dried PAN_{EA}F (3.0 g), Benzylamine (10 mL) and H₂O (30 mL) were placed in a three-necked flask, and the mixture was heated at 100 °C for 1.5 h. After cooling the mixture to room temperature, the fiber was filtered out and washed with water (60-70 °C) and ethanol until no benzylamine was attached to the fiber surface by non-covalent bonds. Then the fiber was dried at 60 °C overnight to get the Bn-PAN_{EA}F. The weight gain of Bn-PAN_{EA}F based on PAN_{EA}F was 8.3%.

2.3. Synthesis of the Bu-PAN_{EA}F

Dried PAN_{EA}F (1.0 g), *n*-butyl amine (10 mL), Na₂CO₃ (0.1 g) and H₂O (10 mL) were placed in a three-necked flask, and the mixture was heated at 100 °C for 1.5 h. After cooling the mixture to room temperature, the fiber was filtered out. The fiber was washed with water

(60-70 °C) and ethanol until no *n*-butyl amine was attached to the fiber surface by non-covalent bonds, and then dried at 60 °C overnight to get the Bu-PAN_{EA}F. The weight gain of Bu-PAN_{EA}F based on PAN_{EA}F was 6.6%.

3. Acid exchange capacity

Dried fibers (0.100 g) were immersed into 50 mL of 0.010 mol L⁻¹ NaOH for 4 h. The neutralized fibers were then filtered and the concentration of the remaining solution were determined by titration with 0.010 mol L⁻¹ HCl. The total acid contents were calculated based on the consumption of the base.

4. The WCA photographs of the fibers

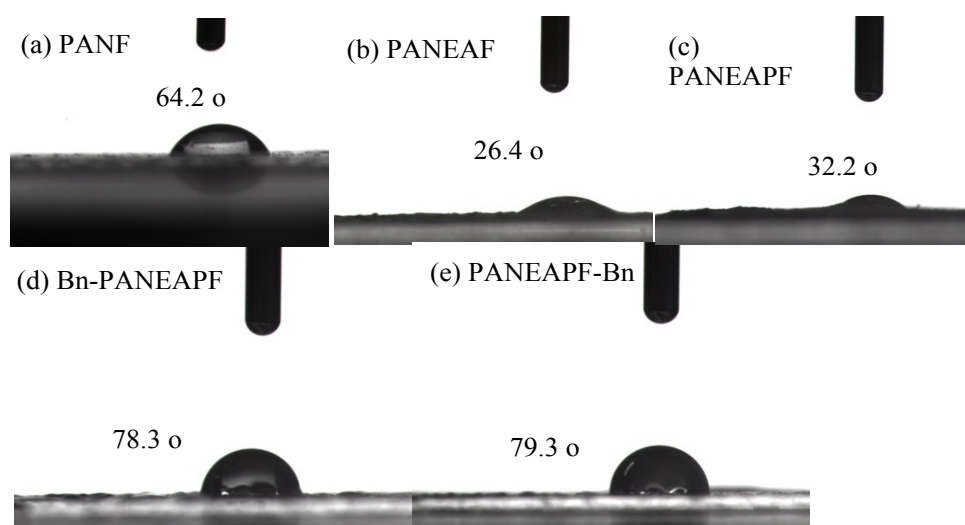


Fig. S1. The WCA photographs of (a) PANF, (b) PAN_{EA}F, (c) PAN_{EAP}F, (d) Bn-PAN_{EAP}F and (e) PAN_{EAP}F-Bn.

5. The FTIR spectra of PAN_{EAP}F, PAN_{EAP}F-Bn, Bu-PAN_{EAP}F, PAN_{EAP}F-Bu

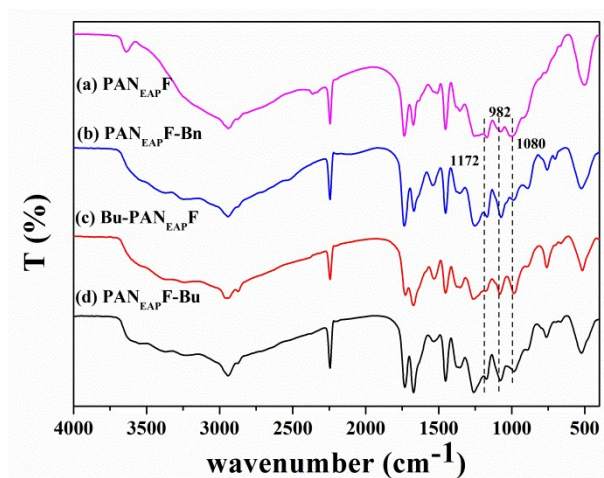


Fig S2. The FTIR spectra of (a) PAN_{EAP}F, (b) PAN_{EAP}F-Bn, (c) Bu-PAN_{EAP}F, (d) PAN_{EAP}F-Bu.

6. Specific surface area analysis

Table S1. Specific surface area analysis

Entry	Fiber	S _{BET} (m ² g ⁻¹)	V _{Tot} (cm ³ g ⁻¹)	Average pore diameter (nm)
1	PANF	41.1	0.034	3.27
2	PAN _{EAP} F	26.0	0.021	3.27
3	PAN _{EAP} F	28.0	0.119	3.44
4	Bn-PAN _{EAP} F	29.9	0.129	3.34
5	Bu-PAN _{EAP} F	38.9	0.177	3.38
6	PAN _{EAP} F-Bn	31.5	0.135	3.19
7	PAN _{EAP} F-Bu	37.0	0.158	3.51

7. The SEM images of PAN_{EAP}F, Bu-PAN_{EAP}F, BPAN_{EAP}F-Bn, PAN_{EAP}F-Bu

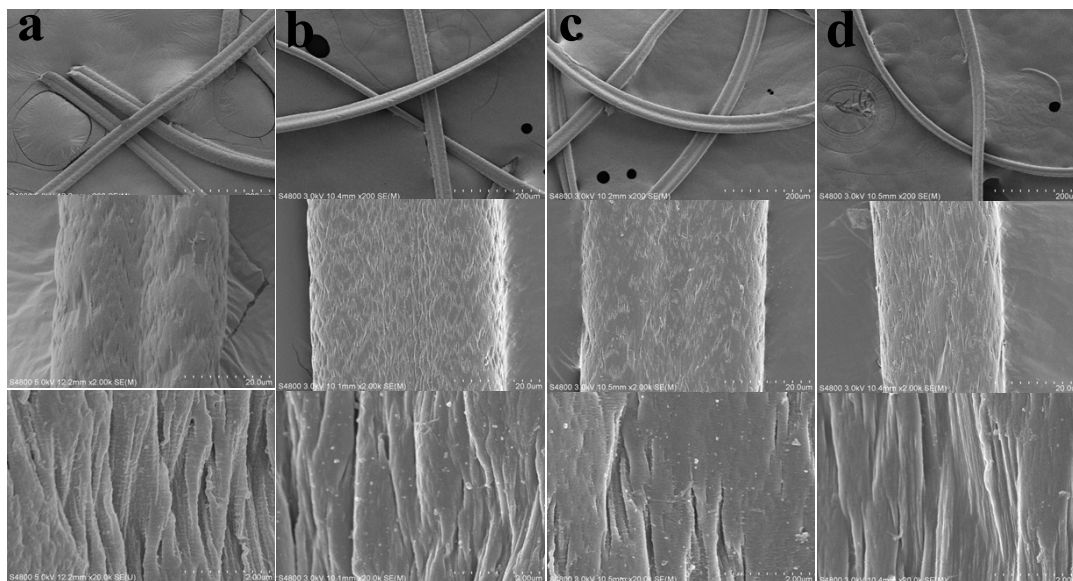


Fig. S3. The SEM images of (a) PAN_{EAP}F, (b) Bu-PAN_{EAP}F, (c) BPAN_{EAP}F-Bn, (d) PAN_{EAP}F-Bu.

8. Thermal stability of fibers

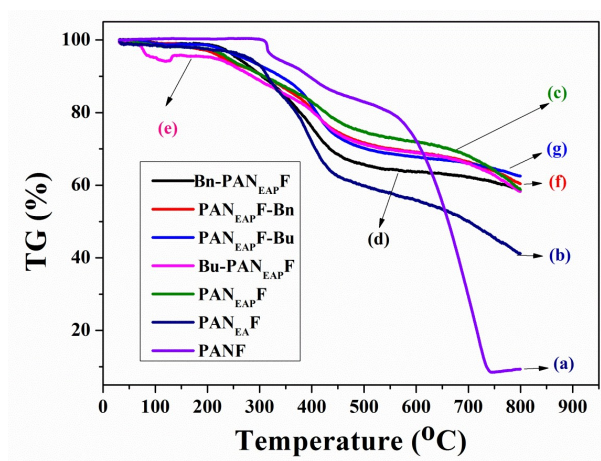


Fig. S4. TGA spectra of (a) PANF, (b) PAN_{EAP}F, (c) PAN_{EAP}F, (d) Bn-PAN_{EAP}F, (e) Bu-PAN_{EAP}F, (f) PAN_{EAP}F-Bn and (g) PAN_{EAP}F-Bu.

9. Catalytic test

9.1. General experimental procedure of the fiber catalyzed cyclocondensation reactions of 2-aminobenzamides with β -ketoesters for the synthesis of *N*-heterocycle derivatives:

The 2-aminobenzamides (1 mmol), β -ketoesters (1.5 mmol), fiber catalyst (5 mol%) and water (5 mL) were added in a screw-capped pressure bottle equipped with a magnetic stirrer. The reaction mixture was stirred at 100 °C for 15 h and monitored by TLC or HPLC. After completion of the reaction, the system was cooled to room temperature. The fiber catalyst was taken out with tweezers and washed with ethyl acetate (5 mL $\times$ 3) and was directly used for next run. The ethyl acetate solution was mixed with the reaction mixture. After extraction and washing with water, the organic phase was dried using Na₂SO₄. After evaporation of the solvent, the residue was purified by a silica gel column (eluent: ethyl acetate/petroleum ether=1/20-1/5) to get the corresponding pure product.

9.2. General experimental procedure of the fiber catalyzed Knoevenagel condensation of aromatic aldehydes with active methylene compounds:

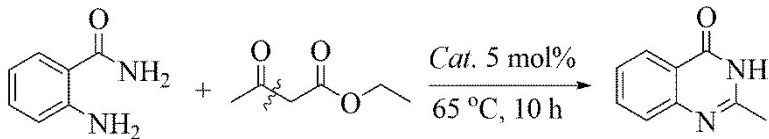
The aldehyde (1 mmol), malononitrile (1.1 mmol) and Bn-PAN_{EAP}F (5 mol%) were added to 10 mL of H₂O in a single-necked flask and the mixture was stirred at room temperature for 2 h. The progress of the reaction was monitored by TLC or HPLC. After the process was completed, the fiber catalyst was taken out with tweezers and washed with ethyl acetate (5 mL $\times$ 3) and then was directly used for next run. The ethyl acetate solution was mixed with the reaction mixture. After extraction and washing with water, the organic phase was dried using Na₂SO₄. After evaporation of the solvent, the residue was purified by a silica gel column (eluent: ethyl acetate/petroleum ether=1/50-1/10) to get the corresponding pure product.

9.3. General experimental procedure of the fiber catalyzed Biginelli reactions using Bn-PAN_{EAPF}

A mixture of aldehyde (1 mmol), methylene compound (1.2 mmol), urea (1.2 mmol), ethanol (5 mL) and Bn-PAN_{EAPF} (5 mol%) were added in a screw-capped pressure bottle equipped with a magnetic stirrer. The reaction mixture was stirred at 80 °C for 12 h and monitored by TLC or HPLC. After completion of the reaction, the system was cooled to room temperature. The fiber catalyst was taken out with tweezers and washed with ethanol (5 mL×3) and was directly used for next run. The ethanol solution was mixed with the reaction mixture, and the solvent of the mixture was evaporated under reduced pressure. The residue was washed with water (5 mL) and extracted with ethyl acetate (10 mL), the organic phase was dried using Na₂SO₄. After evaporation of the solvent, the product is almost pure. Further purification of the product was treated simply through a silica gel column (eluent: ethyl acetate/petroleum ether=1/20-1/1) .

9.4. The effect of the solvent polarity on the reaction

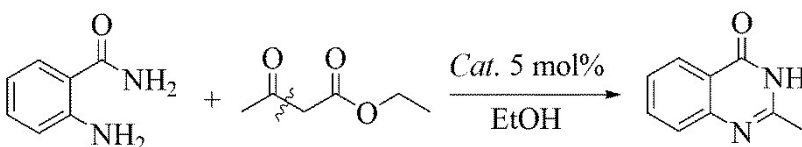
Table S2. The effect of the solvent polarity on the reaction^a

				
Entry	Catalyst	dosage	Solvent	Yield (%) ^b
1	PAN _{EAPF}	5 mol%	EtOH:H ₂ O (10:0)	59
2	PAN _{EAPF}	5 mol%	EtOH:H ₂ O (9:1)	51
3	PAN _{EAPF}	5 mol%	EtOH:H ₂ O (7:3)	36
4	PAN _{EAPF}	5 mol%	EtOH:H ₂ O (5:5)	30
5	PAN _{EAPF}	5 mol%	EtOH:H ₂ O (3:7)	31
6	PAN _{EAPF}	5 mol%	EtOH:H ₂ O (1:9)	29
7	PAN _{EAPF}	5 mol%	EtOH:H ₂ O (0:10)	44

^aReaction conditions: 2-aminobenzamide (1 mmol), ethyl acetoacetate (1.5 mmol), solvent (5 mL) and catalyst dosage based on acid content, the solvents with different polarities were obtained by adjusting the ratio of water to ethanol. ^bThe yields were obtained by HPLC using *m*-binitrobenzene as an internal standard.

9.5. Comparison of catalytic activity of fiber catalysts in ethanol

Table S3. Comparison of catalytic activity of fiber catalysts in ethanol^a

				
Entry	Catalyst	Temp (°C)	Time (h)	Yield (%) ^b
1	PAN _{EAP} F	65	10	59
2	Bn-PAN _{EAP} F	65	10	95
3	Bu-PAN _{EAP} F	65	10	93
4	PAN _{EAP} F-Bn	65	10	27
5	PAN _{EAP} F-Bu	65	10	40
6	PAN _{EAP} F	80	10	84
7	PAN _{EAP} F	80	15	96

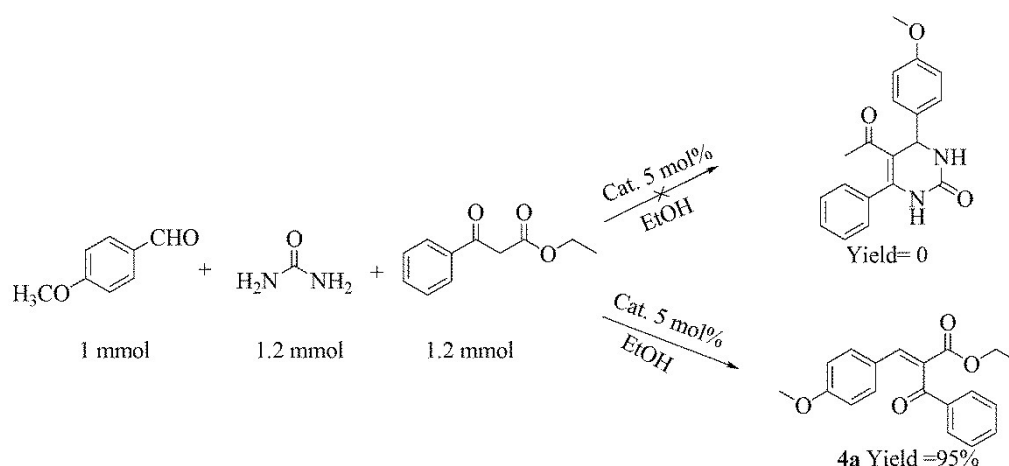
^aReaction conditions: 2-aminobenzamide (1 mmol), ethyl acetoacetate (1.5 mmol), solvent (5 mL) and catalyst dosage based on acid content. ^bThe yields were obtained by HPLC using *m*-binitrobenzene as an internal standard.

9.6. Comparison of the Knoevenagel condensation using different supported catalysts

Table S4. Comparison of the Knoevenagel condensation using different supported catalysts.

Entry	Catalyst	Catalyst dosage	solvent	Temperature /Time	Yield (%)	Ref.
1	Pd/Cz-MOF-253-800	5 mg	toluene	80 °C/5 h	99 (Conversion)	[1]
2	UiO-66-NH-RNH ₂	1 mol%	toluene	Rt/2 h	97	[2]
3	Fe ₃ O ₄ @SiO ₂ -3N	5 mg	THF	75 °C/15 min	100 (Conversion)	[3]
4	Microporous carbon nitride	100 mg	CH ₃ CN	90 °C /4 h	87	[4]
5	polystyrene-supported DABCO	10 mol%	CH ₃ OH	Rt/15 min	99	[5]
6	Fe ₃ O ₄ -cysteamine hydrochloride	50 mg	Ethanol–water (1/1)	50 °C/20 min	93	[6]
7	C/Co@DMAN	2 mol %	H ₂ O	Rt/7.5 h	97	[7]
8	Bn-PAN _{EAP} F	5 mol% (33 mg)	H ₂ O	Rt/2 h	94	This study

9.7. The reaction of p-methoxybenzaldehyde with urea and ethyl benzoylacetate



Scheme S1. The reaction of p-methoxybenzaldehyde with urea and ethyl benzoylacetate.

9.8. Comparison of the Biginelli reaction using different heterogeneous catalysts

Table S5. Comparison of the Biginelli reaction using different heterogeneous catalyst.

Entry	Catalyst	Catalyst dosage	Yield	Ref.
1	Yb(III)-resin	300 mg	72	[8]
2	Fe ₃ O ₄ @SBA-15	50 mg	82	[9]
3	PS-PEG-OSO ₃ H	300 mg	79	[10]
4	PPF-SO ₃ H	75 mg	89	[11]
5	Fe ₃ O ₄ @SiO ₂ -imid-PMA	30 mg	91	[12]
6	PSBIL	50 mg	92	[13]
7	Cobalt-based metal coordination polymers	20 mol %	94	[14]
8	Nano-Fe ₃ O ₄ @silica sulfuric acid	6 mg	89	[15]
9	PPA-SiO ₂	60 mg	94	[16]
10	Bn-PAN _{EAPF}	5 mol% (33 mg)	94	This study

10. NMR data of products 1a-1v, 2a-2p and 3a-3m.

Methylquinazolin-4(3H)-one (1a) ¹H NMR (400 MHz, CDCl₃) δ 12.36 (s, 1H), 8.29 (d, *J* = 7.9 Hz, 1H), 7.78 (t, *J* = 7.6 Hz, 1H), 7.69 (d, *J* = 8.1 Hz, 1H), 7.48 (t, *J* = 7.5 Hz, 1H), 2.62 (s, 3H); ¹³C NMR (101 MHz, DMSO) δ 162.16, 154.93, 149.10, 134.77, 126.79, 126.37, 126.17, 121.05, 21.83.

2-Ethylquinazolin-4(3H)-one (1b) ¹H NMR (400 MHz, DMSO) δ 12.18 (s, 1H), 8.09 (s, 1H), 7.77 (s, 1H), 7.61 (s, 1H), 7.46 (s, 1H), 2.63 (s, 2H), 1.25 (s, 3H); ¹³C NMR (101 MHz, DMSO) δ 162.27, 158.79, 149.42, 134.72, 127.27, 126.38, 126.15, 121.29, 28.30, 11.76.

2-Propylquinazolin-4(3H)-one (1c) ¹H NMR (400 MHz, CDCl₃) δ 12.03 (s, 1H), 8.23 (d, *J* = 7.9 Hz, 1H), 7.71 (t, *J* = 7.5 Hz, 1H), 7.64 (d, *J* = 8.1 Hz, 1H), 7.41 (t, *J* = 7.4 Hz, 1H), 2.73 (t, *J* = 7.6 Hz, 2H), 1.96 – 1.77 (m, 2H), 1.02 (t, *J* = 7.3 Hz, 3H); ¹³C NMR (101 MHz, CDCl₃) δ 163.38, 155.76, 148.49, 133.76, 126.20, 125.31, 125.20, 119.47, 36.74, 19.99, 12.73.

2-Cyclopropylquinazolin-4(3H)-one (1d) ^1H NMR (400 MHz, DMSO) δ 12.45 (s, 1H), 8.05 (d, $J = 7.9$ Hz, 1H), 7.72 (t, $J = 7.6$ Hz, 1H), 7.48 (d, $J = 8.2$ Hz, 1H), 7.39 (t, $J = 7.5$ Hz, 1H), 1.96 (m, 1H), 1.09 (m, 2H), 1.03 (m, 2H); ^{13}C NMR (101 MHz, DMSO) δ 162.08, 159.48, 149.58, 126.94, 126.21, 125.77, 121.09, 13.89, 9.97.

2-Chloromethylquinazolin-4(3H)-one (1e) ^1H NMR (400 MHz, CDCl_3) δ 9.492 (s, 1H), 8.30 (d, $J = 7.9$ Hz, 1H), 7.81 (m, $J = 7.6$ Hz, 1H), 7.68 (d, $J = 8.1$ Hz, 1H), 7.54 (m, $J = 7.5$ Hz, 1H), 4.56 (s, 2H); ^{13}C NMR (101 MHz, DMSO) δ 162.06, 152.71, 148.59, 135.10, 127.76, 127.64, 126.36, 121.73, 43.61.

2-Methoxymethylquinazolin-4(3H)-one (1f) ^1H NMR (400 MHz, CDCl_3) δ 9.67 (s, 1H), 8.22 (d, $J = 7.9$ Hz, 1H), 7.70 (t, $J = 7.6$ Hz, 1H), 7.59 (d, $J = 8.2$ Hz, 1H), 7.42 (t, $J = 7.5$ Hz, 1H), 4.43 (s, 2H), 3.48 (s, 3H); ^{13}C NMR (101 MHz, CDCl_3) δ 161.62, 152.68, 148.62, 134.82, 127.08, 126.86, 126.66, 121.62, 71.03, 59.42.

2,3-Dimethylquinazolin-4(3H)-one (1g) ^1H NMR (400 MHz, CDCl_3) δ 8.26 (d, $J = 7.9$ Hz, 1H), 7.72 (t, $J = 7.6$ Hz, 1H), 7.61 (d, $J = 8.1$ Hz, 1H), 7.44 (t, $J = 7.5$ Hz, 1H), 3.64 (s, 3H), 2.63 (s, 3H); ^{13}C NMR (101 MHz, CDCl_3) δ 162.32, 154.44, 147.27, 134.19, 126.80, 126.62, 126.40, 120.24, 31.05, 23.69.

6-Chloro-2,3-Dimethylquinazolin-4(3H)-one (1h) ^1H NMR (400 MHz, DMSO) δ 12.39 (s, 1H), 7.99 (s, 1H), 7.78 (d, $J = 8.6$ Hz, 1H), 7.58 (d, $J = 8.6$ Hz, 1H), 2.35 (s, 3H); ^{13}C NMR (101 MHz, DMSO) δ 161.14, 155.32, 148.15, 134.83, 130.52, 129.33, 125.12, 122.35, 21.93.

6-methoxy-2-methylquinazolin-4(3H)-one (1i) ^1H NMR (400 MHz, DMSO) δ 12.16 (s, 1H), 7.52 (d, $J = 8.8$ Hz, 1H), 7.46 (s, 1H), 7.37 (d, $J = 8.8$ Hz, 1H), 3.86 (s, 3H), 2.32 (s, 3H); ^{13}C NMR (101 MHz, DMSO) δ 162.01, 157.58, 152.25, 143.93, 128.71, 124.20, 121.79, 106.12, 55.98, 21.68.

2-Phenylquinazolin-4(3H)-one (1j) ^1H NMR (400 MHz, CDCl_3) δ 11.30 (s, 1H), 8.27 (d, $J = 7.9$ Hz, 1H), 8.17 (d, $J = 3.5$ Hz, 2H), 7.77 (t, $J = 8.7$ Hz, 2H), 7.53 (s, 3H), 7.45 (t, $J = 7.2$ Hz, 1H); ^{13}C NMR (101 MHz, CDCl_3) δ 163.61, 151.70, 149.50, 134.93, 132.84, 131.71, 129.13, 128.05, 127.31, 126.86, 126.42, 120.91.

2-(4-Methoxyphenyl)quinazolin-4(3H)-one (1k) ^1H NMR (400 MHz, DMSO) δ 12.44 (s, 1H), 8.20 (d, $J = 8.0$ Hz, 2H), 8.14 (d, $J = 7.7$ Hz, 1H), 7.81 (t, $J = 7.3$ Hz, 1H), 7.71 (d, $J = 8.0$ Hz, 1H), 7.49 (t, $J = 7.2$ Hz, 1H), 7.10 (d, $J = 8.0$ Hz, 2H), 3.86 (s, 3H); ^{13}C NMR (101 MHz, DMSO) δ 162.78, 162.33, 152.33, 149.41, 135.02, 129.93, 127.76, 126.60, 126.30, 125.26, 121.16, 114.47, 55.93.

6-Chloro-2-(4-methoxyphenyl)quinazolin-4(3H)-one (1l) ^1H NMR (400 MHz, DMSO) δ 12.59 (s, 1H), 8.19 (d, $J = 8.0$ Hz, 2H), 8.06 (s, 1H), 7.83 (d, $J = 8.5$ Hz, 1H), 7.72 (d, $J = 8.5$ Hz, 1H), 7.09 (d, $J = 8.0$ Hz, 2H), 3.86 (s, 3H); ^{13}C NMR (101 MHz, DMSO) δ 162.31, 161.95, 152.99, 148.16, 134.94, 130.55, 129.99, 125.25, 124.92, 122.20, 114.41, 55.72.

6-methoxy-2-(4-methoxyphenyl)quinazolin-4(3H)-one (1m) ^1H NMR (400 MHz, DMSO) δ 12.40 (s, 1H), 8.16 (d, $J = 8.3$ Hz, 2H), 7.66 (d, $J = 8.8$ Hz, 1H), 7.53 (s, 1H), 7.43 (d, $J = 8.7$ Hz, 1H), 7.08 (d, $J = 8.3$ Hz, 2H), 3.89 (s, 3H), 3.85 (s, 3H); ^{13}C NMR (101 MHz, DMSO) δ 162.58, 162.05, 157.90, 150.21, 143.89, 129.60, 129.47, 125.41, 124.57, 121.90, 114.44,

106.29, 56.08, 55.90.

2-(4-Fluorophenyl)quinazolin-4(3H)-one (1n) ¹H NMR (400 MHz, DMSO) δ 12.61 (s, 1H), 8.30 – 8.22 (m, 2H), 8.16 (d, J = 7.8 Hz, 1H), 7.85 (t, J = 7.5 Hz, 1H), 7.74 (d, J = 8.1 Hz, 1H), 7.53 (t, J = 7.4 Hz, 1H), 7.41 (t, J = 8.5 Hz, 2H). ¹³C NMR (101 MHz, DMSO) δ 165.67, 163.23, 162.69, 151.82, 149.12, 135.12, 130.89, 130.81, 129.69, 127.93, 127.09, 126.33, 121.35, 116.22, 116.00.

2-(4-Nitrophenyl)quinazolin-4(3H)-one (1o) ¹H NMR (400 MHz, DMSO) δ 12.86 (s, 1H), 8.44-8.38 (m, 4H), 8.19 (d, J = 7.5 Hz, 1H), 7.89 (t, J = 7.5 Hz, 1H), 7.81 (d, J = 8.0 Hz, 1H), 7.59 (t, J = 7.3 Hz, 1H).

Ethyl-2-(2-(4-nitrophenyl)-4-oxo-1,2,3,4-tetrahydroquinazolin-2-yl)acetate (1o') ¹H NMR (400 MHz, DMSO) δ 8.83 (s, 1H), 8.20 (d, J = 8.3 Hz, 2H), 7.82 (d, J = 8.3 Hz, 2H), 7.75 (s, 1H), 7.50 (d, J = 7.7 Hz, 1H), 7.26 (t, J = 7.6 Hz, 1H), 6.93 (d, J = 8.1 Hz, 1H), 6.65 (t, J = 7.3 Hz, 1H), 4.02 (q, J = 6.8 Hz, 2H), 3.04 (dd, J = 35.3, 14.9 Hz, 2H), 1.09 (t, J = 7.0 Hz, 3H).

2-Methyl-1H-benzoimidazole (1p) ¹H NMR (400 MHz, CDCl₃) δ 7.56 (dd, J = 2.5 Hz, 2H), 7.24 (dd, J = 9.1, 6.1 Hz, 2H), 2.66 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ 151.30, 138.38, 122.03, 114.37, 15.01.

2-(Methoxymethyl)-1H-benzoimidazole (1q) ¹H NMR (400 MHz, CDCl₃) δ 10.21 (s, 1H), 7.60 (s, 2H), 7.31 – 7.21 (m, 2H), 4.78 (s, 2H), 3.49 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ 151.57, 122.62, 116.69, 68.42, 59.10.

2-cyclopropyl-1H-benzo[d]imidazole (1r) ¹H NMR (400 MHz, CDCl₃) δ 7.55-7.450 (m, 2H), 7.23–7.13 (m, 2H), 2.13 (m, 1H), 1.29–1.20 (m, 2H), 1.09 (m, 2H).

2-chloromethyl-1H-benzimidazole (1s) ¹H NMR (400 MHz, CDCl₃) δ 7.54 (s, 2H), 7.27–7.19 (m, 2H), 4.80 (s, 2H).

2-(4-Methoxyphenyl)-1H-benzo[d]imidazole (1t) ¹H NMR (400 MHz, DMSO) δ 12.75 (s, 1H), 8.13 (d, J = 5.1 Hz, 2H), 7.57 (d, J = 42.9 Hz, 2H), 7.27–7.06 (m, 4H), 3.87 (s, 3H).

2-(4-Nitrophenyl)-1H-benzo[d]imidazole (1u) ¹H NMR (400 MHz, DMSO) δ 13.30 (s, 1H), 8.42 (s, 4H), 7.79–7.53 (m, 2H), 7.27 (s, 2H).

2-(pyridin-4-yl)-1H-benzo[d]imidazole (1v) ¹H NMR (400 MHz, DMSO) δ 13.27 (s, 1H), 8.77 (d, J = 4.3 Hz, 2H), 8.11 (d, J = 4.3 Hz, 2H), 7.66 (s, 2H), 7.28 (s, 2H).

2-(Phenylmethylene)malononitrile (2a) ¹H NMR (400 MHz, CDCl₃) δ 7.85 (d, J = 7.8 Hz, 2H), 7.72 (s, 1H), 7.57 (t, J = 7.4 Hz, 1H), 7.48 (t, J = 7.6 Hz, 2H).

2-(2-nitrobenzylidene)malononitrile (2b) ¹H NMR (400 MHz, CDCl₃) δ 8.46 (s, 1H), 8.37 (d, J = 8.6 Hz, 1H), 7.93–7.83 (m, 3H).

2-(3-nitrobenzylidene)malononitrile (2c) ¹H NMR (400 MHz, CDCl₃) δ 8.67 (s, 1H), 8.49 (d, J = 8.2 Hz, 1H), 8.34 (d, J = 7.9 Hz, 1H), 7.91 (s, 1H), 7.81 (t, J = 8.1 Hz, 1H).

2-(4-nitrobenzylidene)malononitrile (2d) ¹H NMR (400 MHz, CDCl₃) δ 8.40 (d, J = 8.5 Hz, 2H), 8.09 (d, J = 8.5 Hz, 2H), 7.90 (s, 1H).

2-(3-Methoxyphenylmethylene)malononitrile (**2e**) ^1H NMR (400 MHz, CDCl_3) δ 7.76 (s, 1H), 7.50 (s, 1H), 7.48–7.39 (m, 2H), 7.18 (d, $J = 7.3$ Hz, 1H), 3.87 (s, 3H).

2-(4-Methoxyphenylmethylene)malononitrile (**2f**) ^1H NMR (400 MHz, CDCl_3) δ 7.92 (d, $J = 8.7$ Hz, 2H), 7.66 (s, 1H), 7.02 (d, $J = 8.7$ Hz, 2H), 3.92 (s, 3H).

4-(*N*-dimethylamino benzylidene) malononitrile (**2g**) ^1H NMR (400 MHz, CDCl_3) δ 7.75 (d, $J = 8.7$ Hz, 2H), 7.40 (s, 1H), 6.62 (d, $J = 8.7$ Hz, 2H), 3.08 (s, 6H).

2-(4-hydroxyphenylmethylene)malononitrile (**2h**) ^1H NMR (400 MHz, DMSO) δ 8.38 (s, 1H), 7.96 (d, $J = 8.4$ Hz, 2H), 7.04 (d, $J = 8.4$ Hz, 2H); ^{13}C NMR (101 MHz, DMSO) δ 165.63 – 163.19 (m), 162.52 – 158.74 (m), 135.71 – 132.05 (m), 124.10 – 121.81 (m), 117.91 – 116.27 (m), 115.87 – 115.08 (m), 115.32 – 113.86 (m), 76.23 – 74.62 (m).

4-chlorobenzylidenemalonodinitrile (**2i**) ^1H NMR (400 MHz, CDCl_3) δ 7.87 (d, $J = 8.3$ Hz, 2H), 7.74 (s, 1H), 7.53 (d, $J = 8.3$ Hz, 2H).

2-(3-bromobenzylidene)malononitrile (**2j**) ^1H NMR (400 MHz, CDCl_3) δ 7.90 (s, 1H), 7.84 (d, $J = 7.9$ Hz, 1H), 7.73–7.62 (m, 2H), 7.37 (t, $J = 8.0$ Hz, 1H).

2-(2-trifluoromethylbenzylidene)malononitrile (**2k**) ^1H NMR (400 MHz, CDCl_3) δ 8.23 (s, 1H), 8.08 (d, $J = 7.3$ Hz, 1H), 7.85 (d, $J = 7.3$ Hz, 1H), 7.75 (p, $J = 7.4$ Hz, 2H).

[(2-chloro-3-quinolyl)methylidene]methane-1,1-dicarbonitrile (**2l**) ^1H NMR (400 MHz, CDCl_3) δ 9.02 (s, 1H), 8.37 (s, 1H), 8.07 (d, $J = 8.5$ Hz, 1H), 8.00 (d, $J = 8.2$ Hz, 1H), 7.94 (t, $J = 7.7$ Hz, 1H), 7.71 (t, $J = 7.6$ Hz, 1H).

2-(2-Furylmethylene)malonitrile (**2m**) ^1H NMR (400 MHz, CDCl_3) δ 7.90 (d, $J = 4.2$ Hz, 2H), 7.81 (d, $J = 3.7$ Hz, 1H), 7.31 – 7.25 (m, 1H), 1.59 (s, 1H).

Cinnamalmalononitrile (**2n**) ^1H NMR (400 MHz, CDCl_3) δ 7.66–7.57 (m, 3H), 7.53 – 7.41 (m, 3H), 7.28 (d, $J = 10.7$ Hz, 2H), 1.59 (d, $J = 3.2$ Hz, 1H).

Ethyl benzylidenecyanoacetate (**2o**) ^1H NMR (400 MHz, CDCl_3) δ 8.27 (s, 1H), 8.00 (d, $J = 7.7$ Hz, 2H), 7.54 (dq, $J = 14.8, 7.4$ Hz, 3H), 4.40 (q, $J = 7.1$ Hz, 2H), 1.41 (t, $J = 7.1$ Hz, 3H).

2-cyano-3-(4-nitrophenyl)-2-propenoic acid ethyl ester (**2p**) ^1H NMR (400 MHz, CDCl_3) δ 8.36 (d, $J = 8.4$ Hz, 2H), 8.31 (s, 1H), 8.14 (d, $J = 8.5$ Hz, 2H), 4.43 (q, $J = 7.1$ Hz, 2H), 1.43 (t, $J = 7.1$ Hz, 3H).

5-Ethoxycarbonyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (**3a**) ^1H NMR (400 MHz, DMSO) δ 9.25 (s, 1H), 7.79 (s, 1H), 7.38 (t, $J = 7.3$ Hz, 2H), 7.29 (d, $J = 7.0$ Hz, 3H), 5.20 (s, 1H), 4.04 (q, $J = 6.9$ Hz, 2H), 2.30 (s, 3H), 1.15 (t, $J = 7.0$ Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.81, 152.61, 148.83, 145.34, 128.86, 127.73, 126.72, 99.73, 59.65, 54.43, 18.25, 14.54.

5-Ethoxycarbonyl-4-(4-methylphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**3b**) ^1H NMR (400 MHz, DMSO) δ 9.14 (s, 1H), 7.68 (s, 1H), 7.12 (s, 4H), 5.11 (s, 1H), 3.99 (q, $J = 6.9$ Hz, 2H), 2.26 (d, $J = 8.2$ Hz, 6H), 1.11 (t, $J = 7.0$ Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.83, 152.66, 148.60, 142.43, 136.83, 129.35, 126.62, 99.91, 59.62, 54.11, 21.10, 18.22, 14.56.

5-Ethoxycarbonyl-4-(4-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**3c**) ^1H

NMR (400 MHz, DMSO) δ 9.14 (s, 1H), 7.66 (s, 1H), 7.15 (d, J = 8.3 Hz, 2H), 6.88 (d, J = 8.4 Hz, 2H), 5.10 (s, 1H), 3.99 (dd, J = 14.0, 6.9 Hz, 2H), 3.73 (s, 3H), 2.25 (s, 3H), 1.11 (t, J = 7.0 Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.85, 158.92, 152.64, 148.46, 137.53, 127.87, 114.17, 100.07, 59.61, 55.52, 53.82, 18.22, 14.56.

5-Ethoxycarbonyl-4-(3-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3d) ^1H NMR (400 MHz, DMSO) δ 9.11 (s, 1H), 7.24 (d, J = 14.7 Hz, 2H), 7.06 (d, J = 7.3 Hz, 1H), 6.99 (d, J = 8.0 Hz, 1H), 6.88 (t, J = 7.1 Hz, 1H), 5.50 (s, 1H), 3.93 (t, J = 6.9 Hz, 2H), 3.80 (s, 3H), 2.29 (s, 3H), 1.03 (t, J = 6.4 Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.82, 157.01, 152.67, 149.29, 132.10, 129.13, 127.55, 120.62, 111.61, 98.09, 59.44, 55.84, 49.38, 18.17, 14.48.

5-Ethoxycarbonyl-4-(2-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3e) ^1H NMR (400 MHz, DMSO) δ 9.18 (s, 1H), 7.72 (s, 1H), 7.25 (t, J = 7.8 Hz, 1H), 6.88 – 6.75 (m, 3H), 5.13 (s, 1H), 4.01 (q, J = 6.9 Hz, 2H), 3.73 (s, 3H), 2.25 (s, 3H), 1.12 (t, J = 7.0 Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.82, 159.69, 152.69, 148.89, 146.81, 130.02, 118.71, 112.87, 112.61, 99.64, 59.68, 55.43, 54.22, 18.23, 14.58.

5-Ethoxycarbonyl-4-(3-trifluoromethylphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3f) ^1H NMR (400 MHz, DMSO) δ 9.31 (s, 1H), 7.84 (s, 1H), 7.68–7.51 (m, 4H), 5.26 (s, 1H), 4.11–3.90 (m, 2H), 2.27 (s, 3H), 1.08 (t, J = 7.0 Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.59, 152.33, 149.59, 146.72, 130.78, 130.26, 129.50, 125.99, 124.55, 123.42, 99.03, 59.76, 54.23, 18.27, 14.38.

5-Ethoxycarbonyl-6-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (3g) ^1H NMR (400 MHz, DMSO) δ 9.36 (s, 1H), 8.22 (d, J = 8.3 Hz, 2H), 7.90 (s, 1H), 7.51 (d, J = 8.3 Hz, 2H), 5.28 (s, 1H), 3.99 (q, J = 6.9 Hz, 2H), 2.27 (s, 3H), 1.10 (t, J = 7.0 Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.53, 152.47, 152.24, 149.86, 147.18, 128.13, 124.29, 98.65, 59.86, 54.16, 18.34, 14.50.

4-(4-Chlorophenyl)-5-ethoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3h) ^1H NMR (400 MHz, DMSO) δ 9.23 (s, 1H), 7.76 (s, 1H), 7.40 (d, J = 8.0 Hz, 2H), 7.26 (d, J = 8.1 Hz, 2H), 5.15 (s, 1H), 3.99 (q, J = 6.8 Hz, 2H), 2.26 (s, 3H), 1.10 (t, J = 7.0 Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.68, 152.42, 149.18, 144.27, 132.26, 128.85, 128.66, 99.33, 59.72, 53.91, 18.27, 14.53.

4-(3-Bromophenyl)-5-ethoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3i) ^1H NMR (400 MHz, DMSO) δ 9.27 (s, 1H), 7.79 (s, 1H), 7.46 (d, J = 7.7 Hz, 1H), 7.40 (s, 1H), 7.32 (t, J = 7.7 Hz, 1H), 7.25 (d, J = 7.6 Hz, 1H), 5.15 (s, 1H), 4.08–3.93 (m, 2H), 2.26 (s, 3H), 1.11 (t, J = 7.0 Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.63, 152.39, 149.43, 147.96, 131.28, 130.62, 129.65, 125.74, 122.00, 99.08, 59.78, 54.07, 18.29, 14.51.

5-Ethoxycarbonyl-6-methyl-4-(2-thienyl)-3,4-dihydropyrimidin-2(1H)-one (3j) ^1H NMR (400 MHz, DMSO) δ 9.31 (s, 1H), 7.90 (s, 1H), 7.36 (d, J = 4.6 Hz, 1H), 6.98–6.87 (m, 2H), 5.43 (s, 1H), 4.07 (q, J = 7.0 Hz, 2H), 2.23 (s, 3H), 1.18 (t, J = 7.1 Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.50, 152.73, 149.27, 149.11, 127.12, 125.08, 123.98, 100.28, 59.82, 49.84, 18.15, 14.62.

5-(ethoxycarbonyl)-6-methyl-4-styryl-3,4-dihydropyrimidin-2(1H)-one (3k) ^1H NMR (400 MHz, DMSO) δ 9.16 (s, 1H), 7.56 (s, 1H), 7.41 (d, J = 6.7 Hz, 2H), 7.35–7.29 (m, 2H), 7.25 (m,

m 1H), 6.37 (d, $J = 15.7$ Hz, 1H), 6.20 (dd, $J = 15.4, 4.7$ Hz, 1H), 4.73 (s, 1H), 4.09 (dd, $J = 11.2, 6.5$ Hz, 2H), 2.21 (s, 3H), 1.21 (t, $J = 6.1$ Hz, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.68, 153.07, 149.02, 136.71, 130.44, 129.14, 128.58, 128.06, 126.81, 98.26, 59.69, 52.36, 18.19, 14.73.

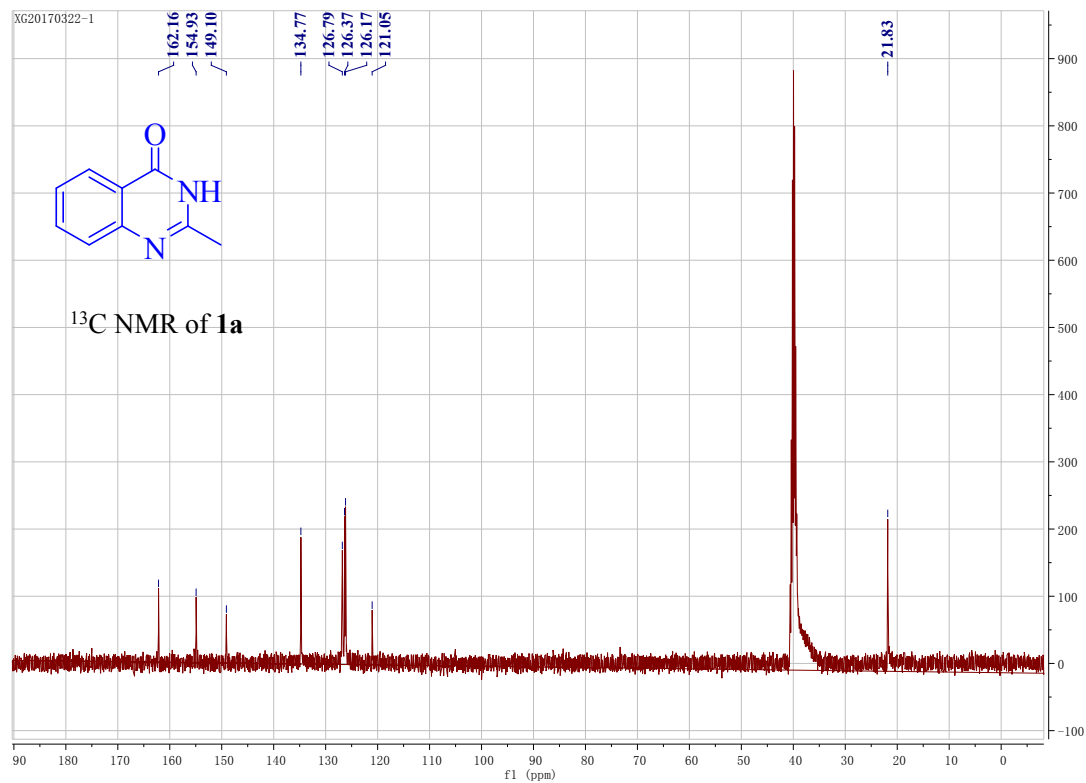
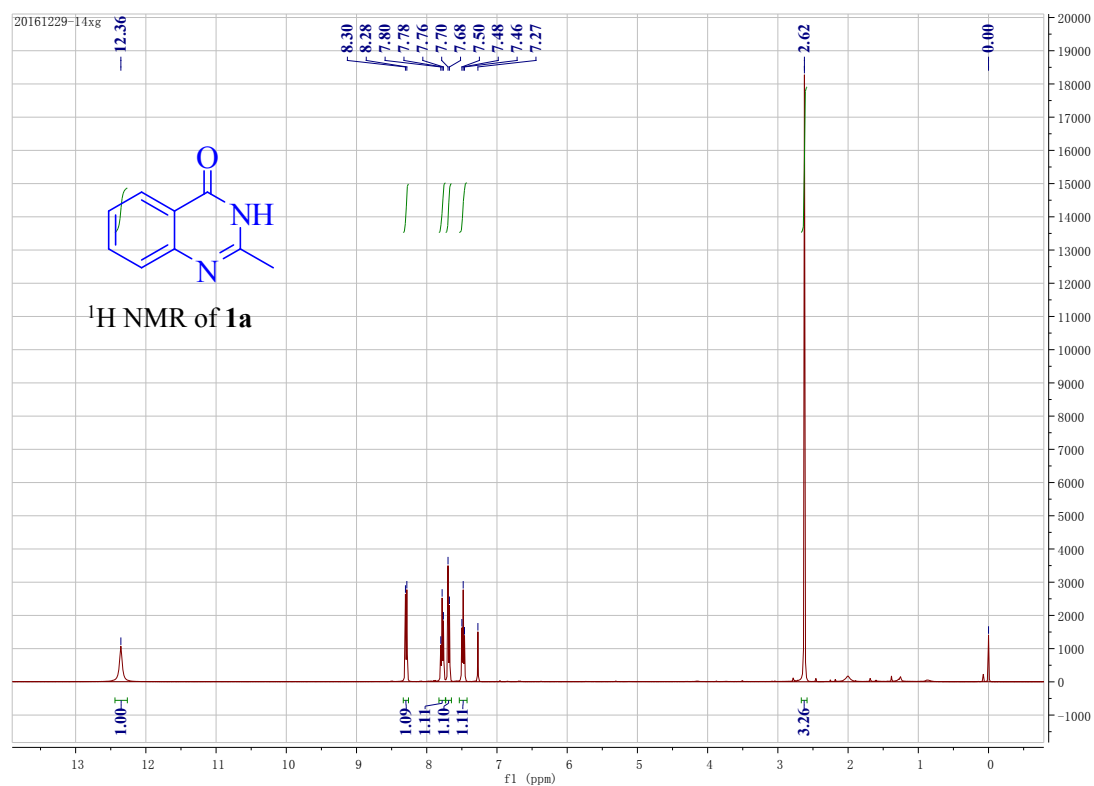
5-Acetyl-4-(4-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3l) ^1H NMR (400 MHz, DMSO) δ 9.24 (s, 1H), 7.85 (s, 1H), 7.24 (d, $J = 8.0$ Hz, 2H), 6.97 (d, $J = 8.0$ Hz, 2H), 5.29 (s, 1H), 3.80 (s, 3H), 2.36 (s, 3H), 2.16 (s, 3H); ^{13}C NMR (101 MHz, DMSO) δ 194.85, 158.99, 152.59, 148.27, 136.84, 128.12, 114.32, 110.08, 55.52, 53.80, 30.63, 19.31.

Benzyl 4-(4-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (3m) ^1H NMR (400 MHz, DMSO) δ 9.24 (s, 1H), 7.70 (s, 1H), 7.29 (s, 3H), 7.14 (dd, $J = 13.0, 6.8$ Hz, 4H), 6.87 (t, $J = 8.0$ Hz, 2H), 5.14 (s, 1H), 5.04 (q, $J = 12.8$ Hz, 2H), 3.73 (s, 3H), 2.27 (s, 3H); ^{13}C NMR (101 MHz, DMSO) δ 165.58, 158.99, 152.49, 149.41, 137.35, 137.03, 128.74, 128.17, 127.98, 127.96, 114.21, 99.52, 65.22, 55.55, 53.81, 18.32.

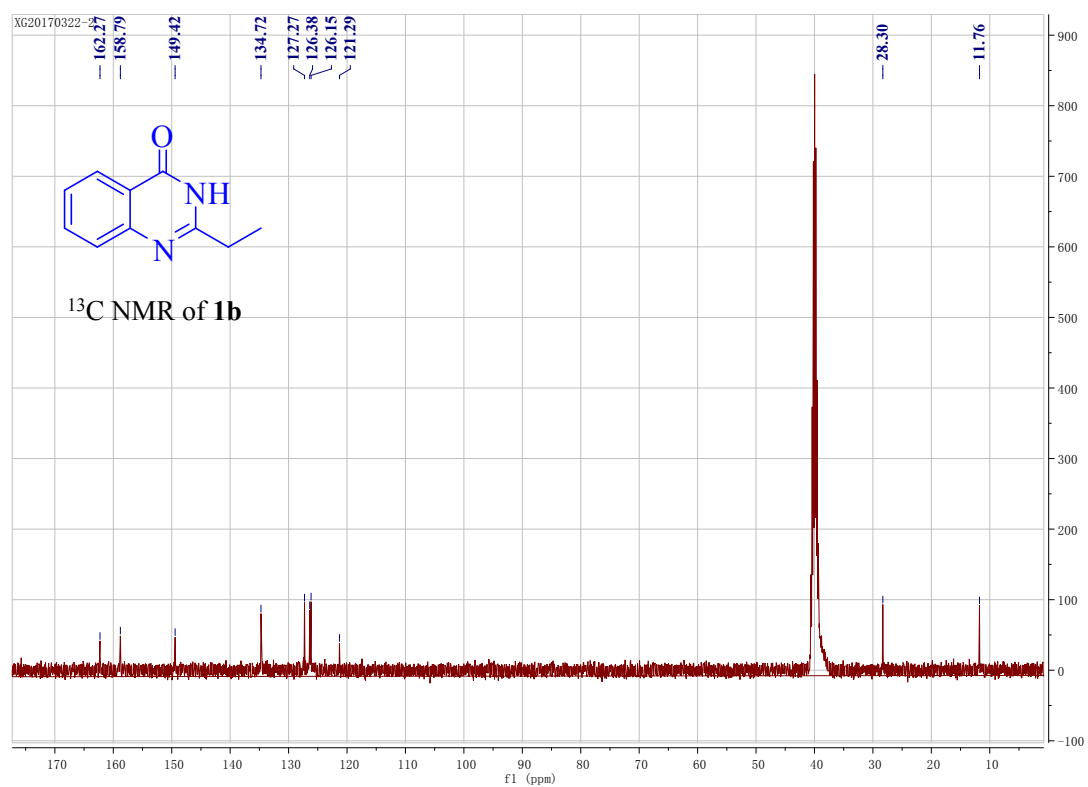
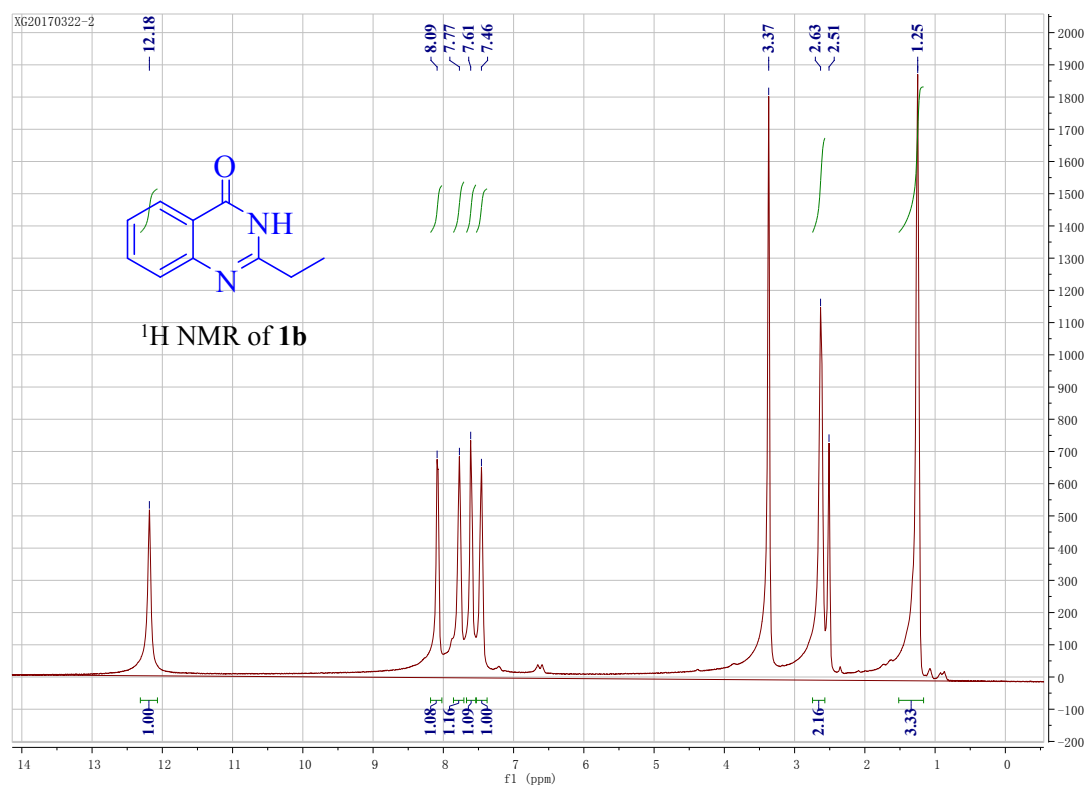
Ethyl 2-benzoyl-3-(4-methoxyphenyl)acrylate (3n) ^1H NMR (400 MHz, DMSO) δ 7.90 (d, $J = 8.9$ Hz, 3H), 7.67 (t, $J = 6.9$ Hz, 1H), 7.54 (t, $J = 7.5$ Hz, 2H), 7.35 (d, $J = 8.1$ Hz, 2H), 6.89 (d, $J = 8.1$ Hz, 2H), 4.16 (q, $J = 6.8$ Hz, 2H), 3.72 (s, 3H), 1.10 (dd, $J = 16.1, 9.3$ Hz, 3H).

11. NMR spectra of products 1a-1v, 2a-2p and 3a-3m.

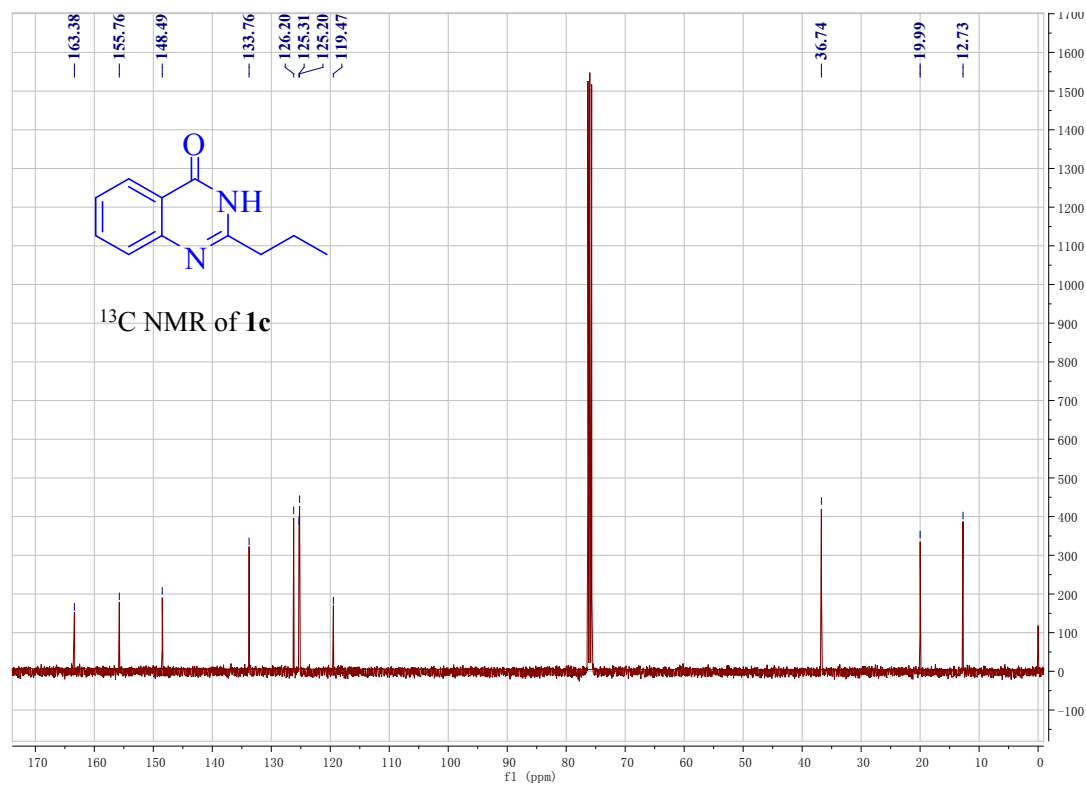
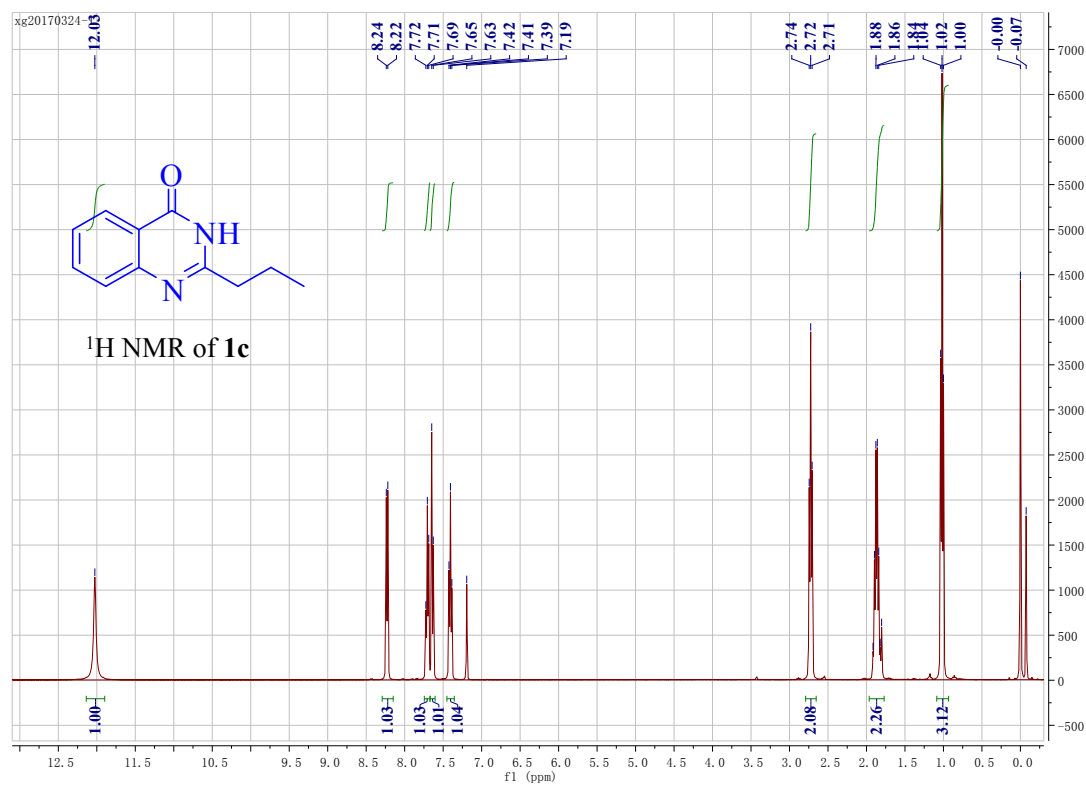
Methylquinazolin-4(3H)-one (1a)



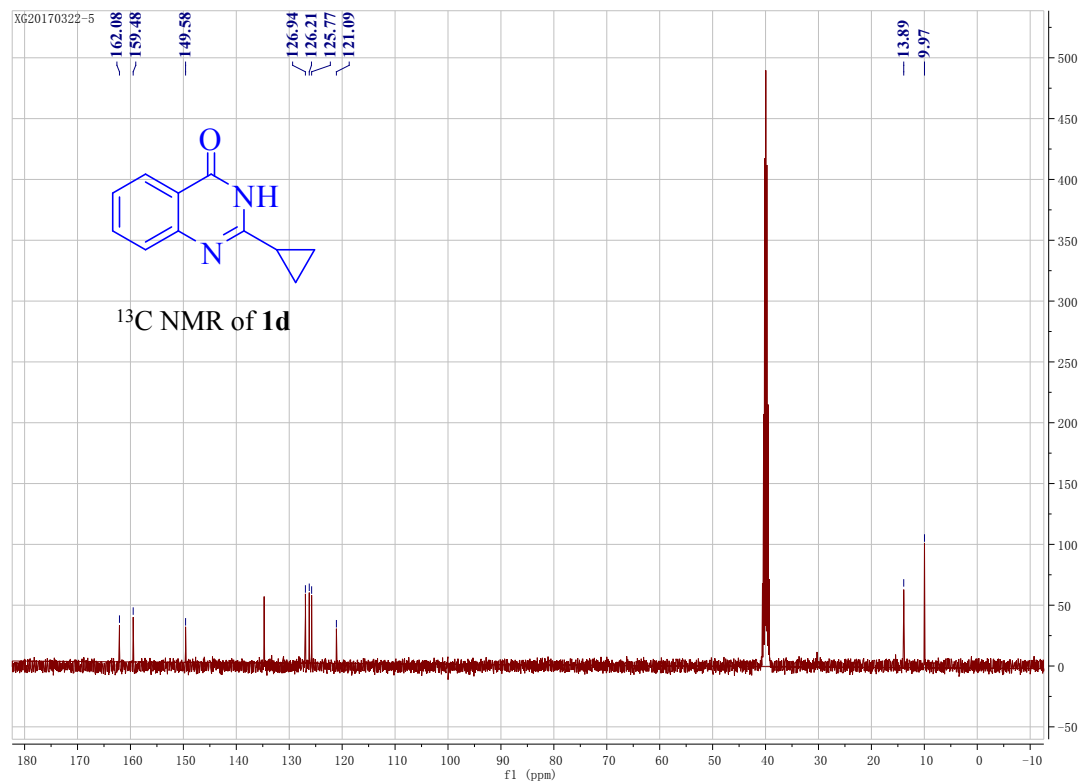
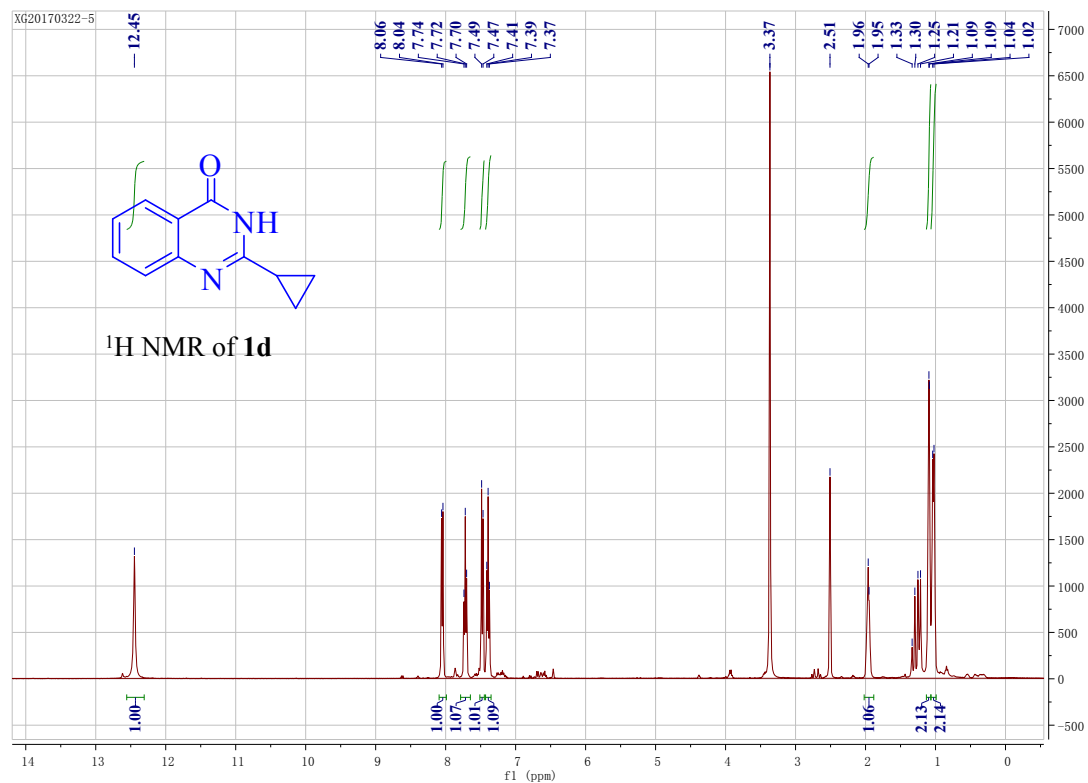
2-Ethylquinazolin-4(3H)-one (**1b**)



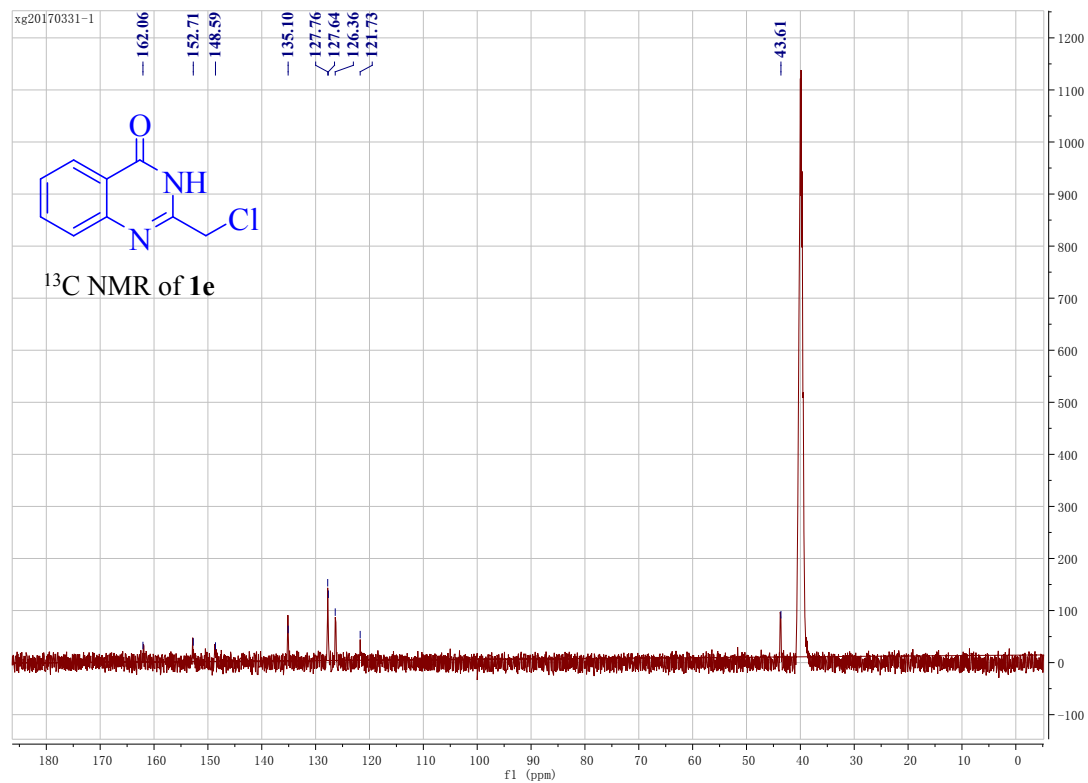
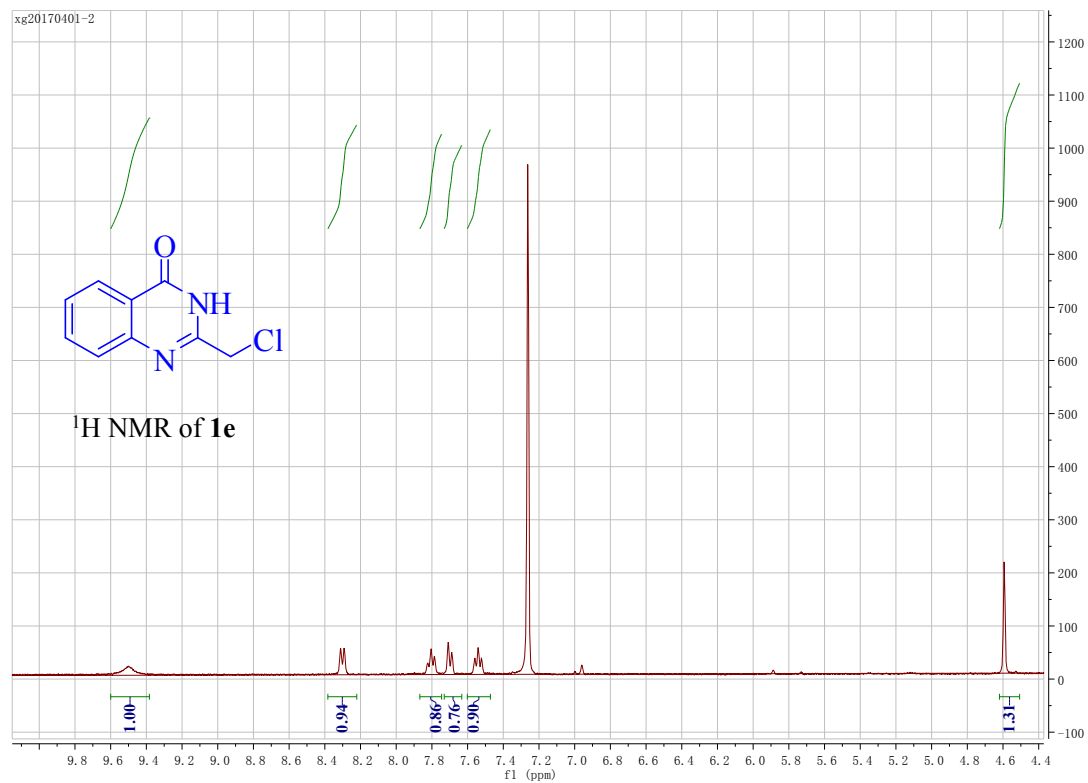
2-Propylquinazolin-4(3H)-one (**1c**)



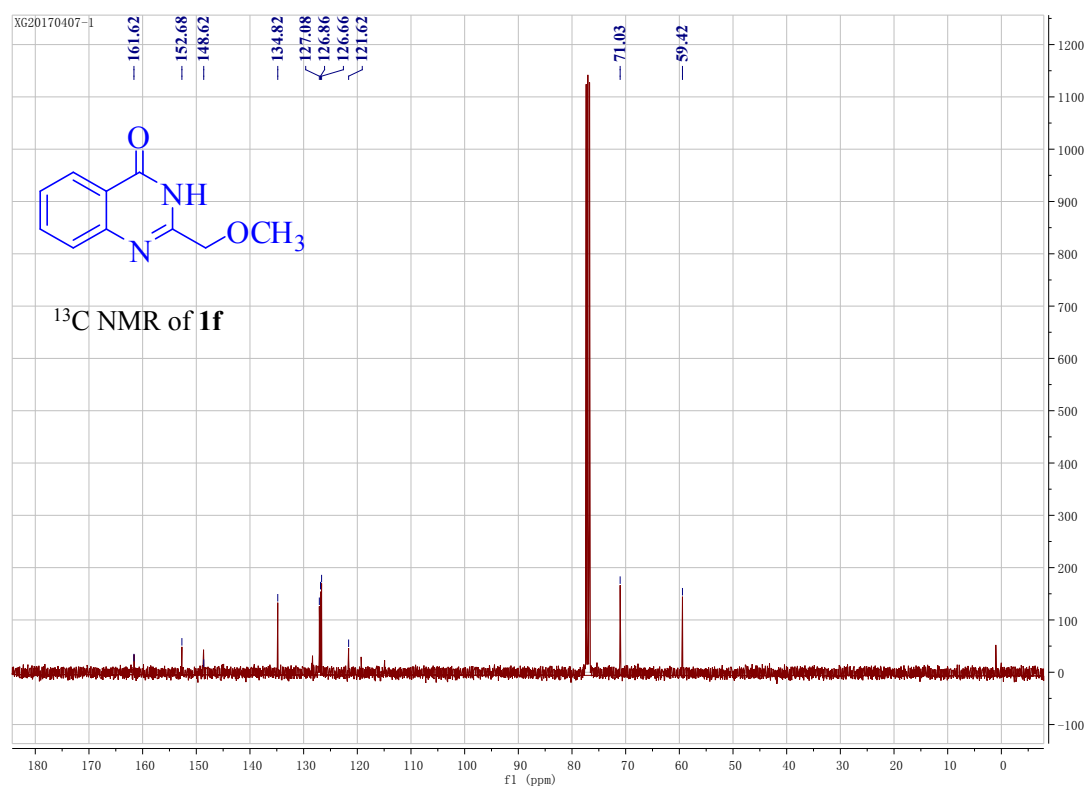
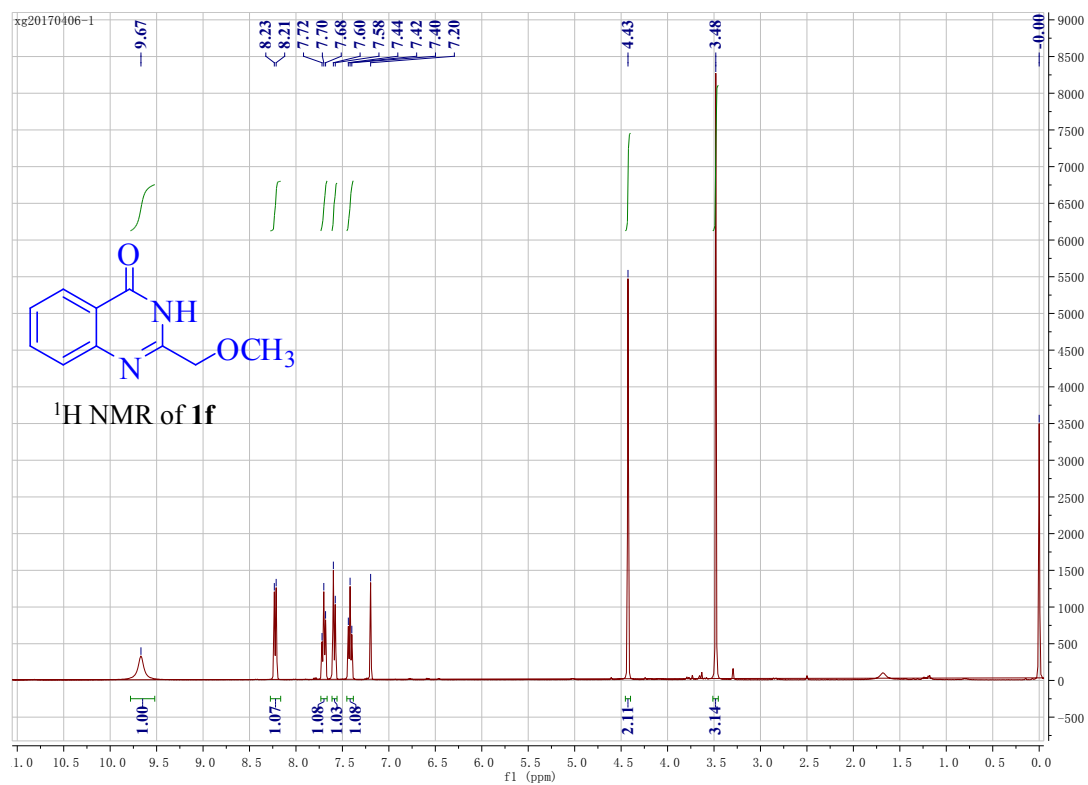
2-Cyclopropylquinazolin-4(3H)-one (**1d**)



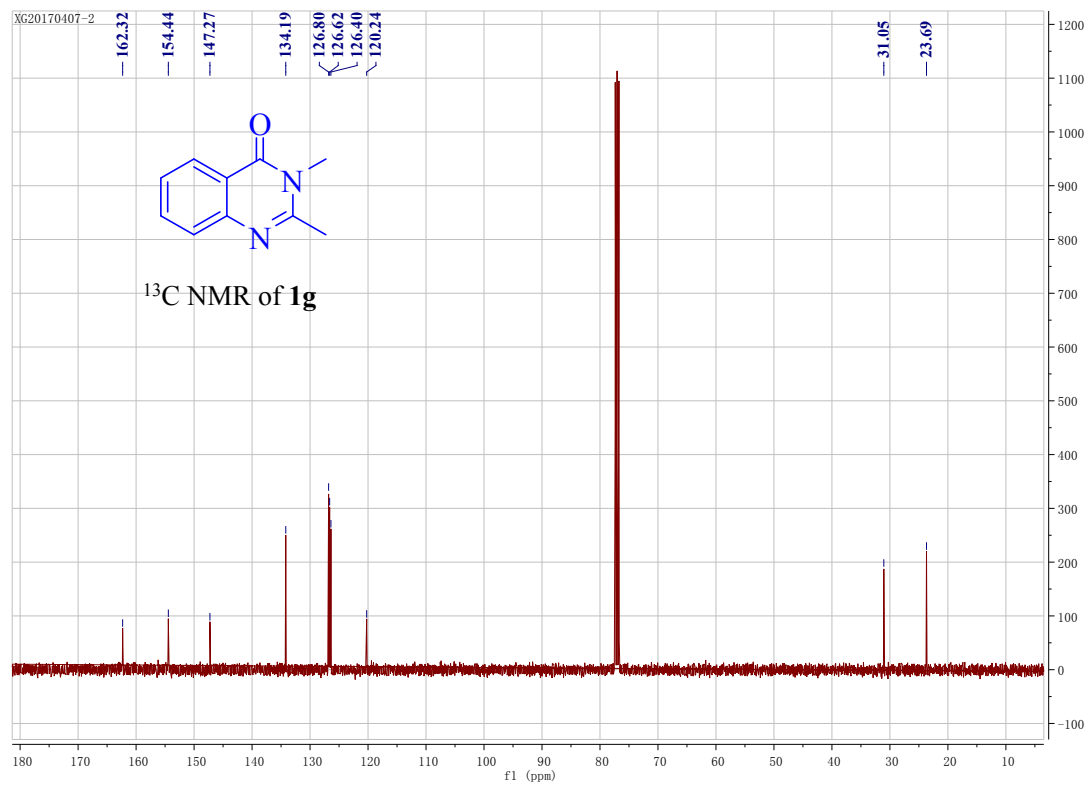
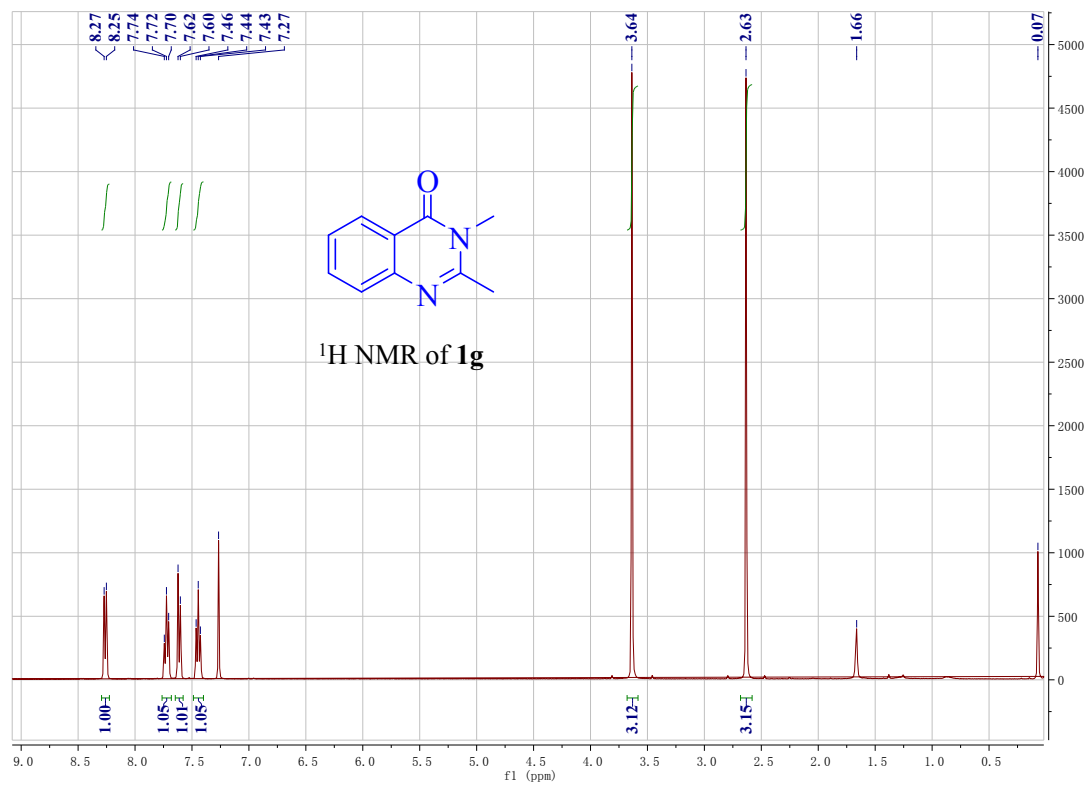
2-Chloromethylquinazolin-4(3H)-one (**1e**)



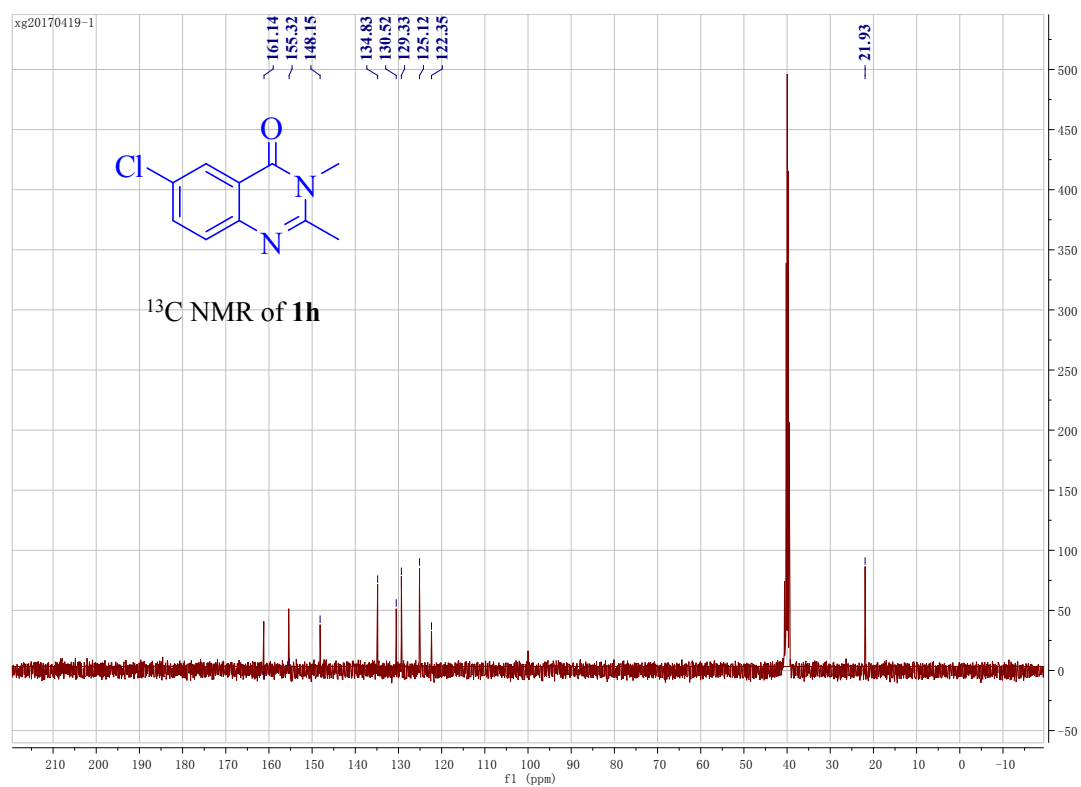
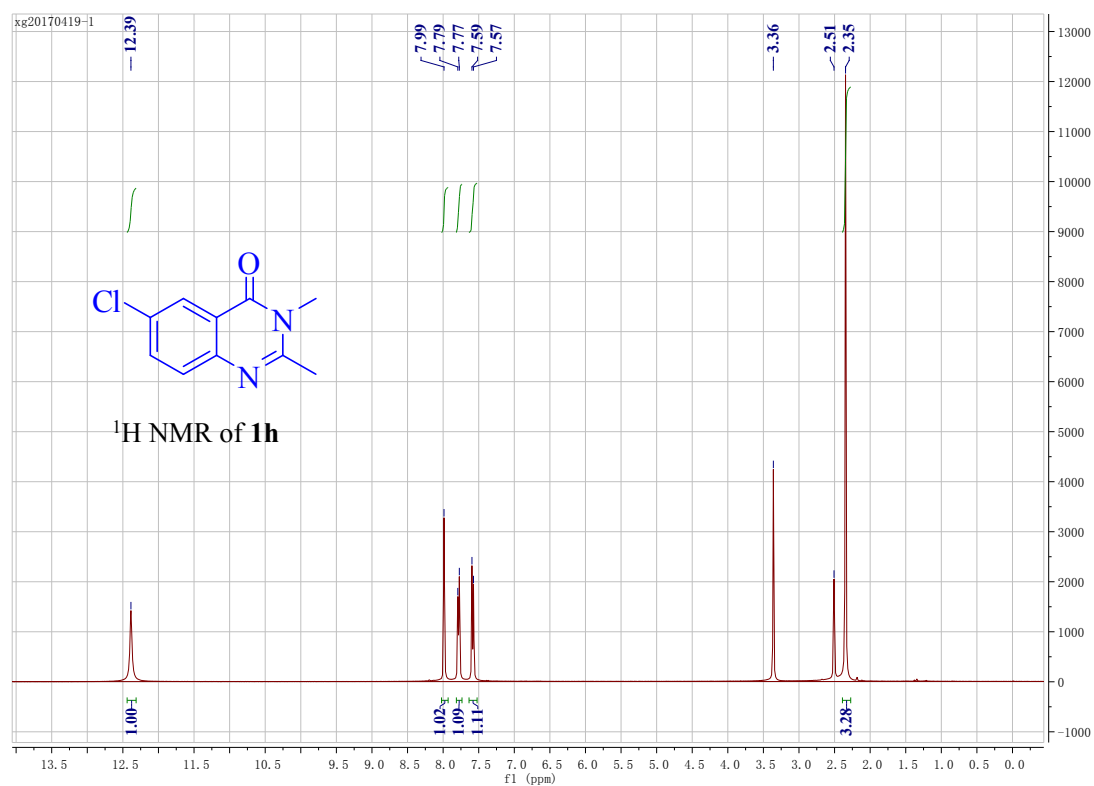
2-Methoxymethylquinazolin-4(3H)-one (1f)



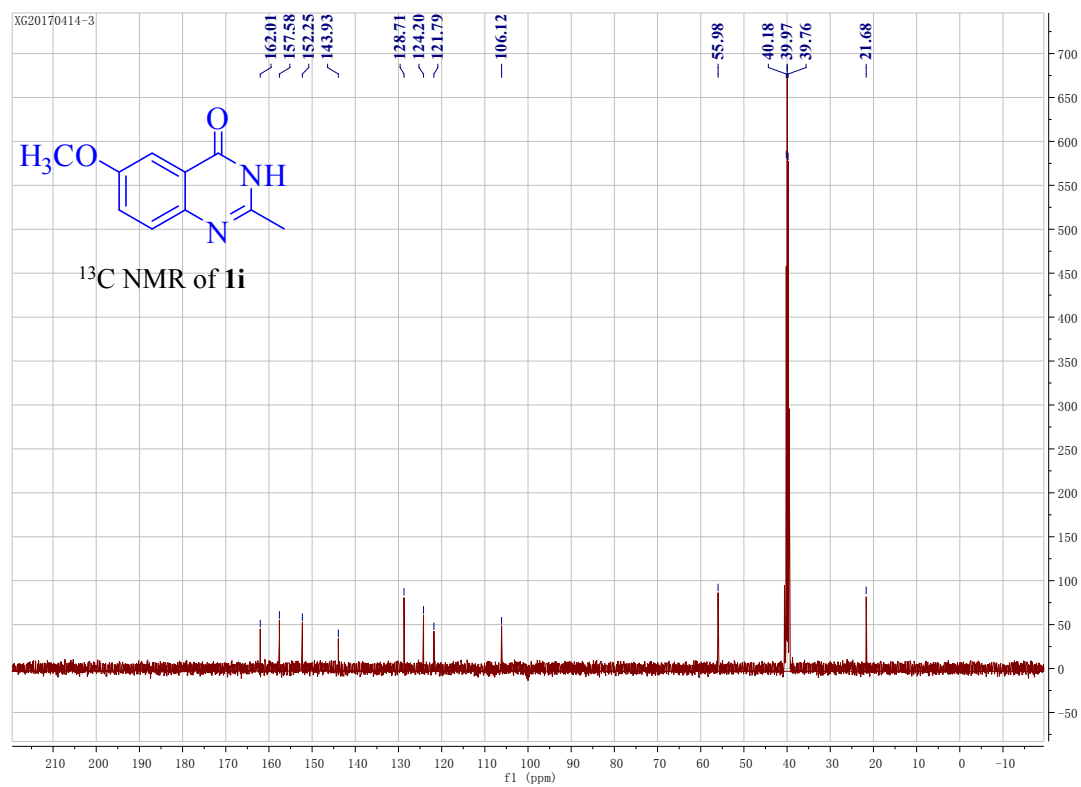
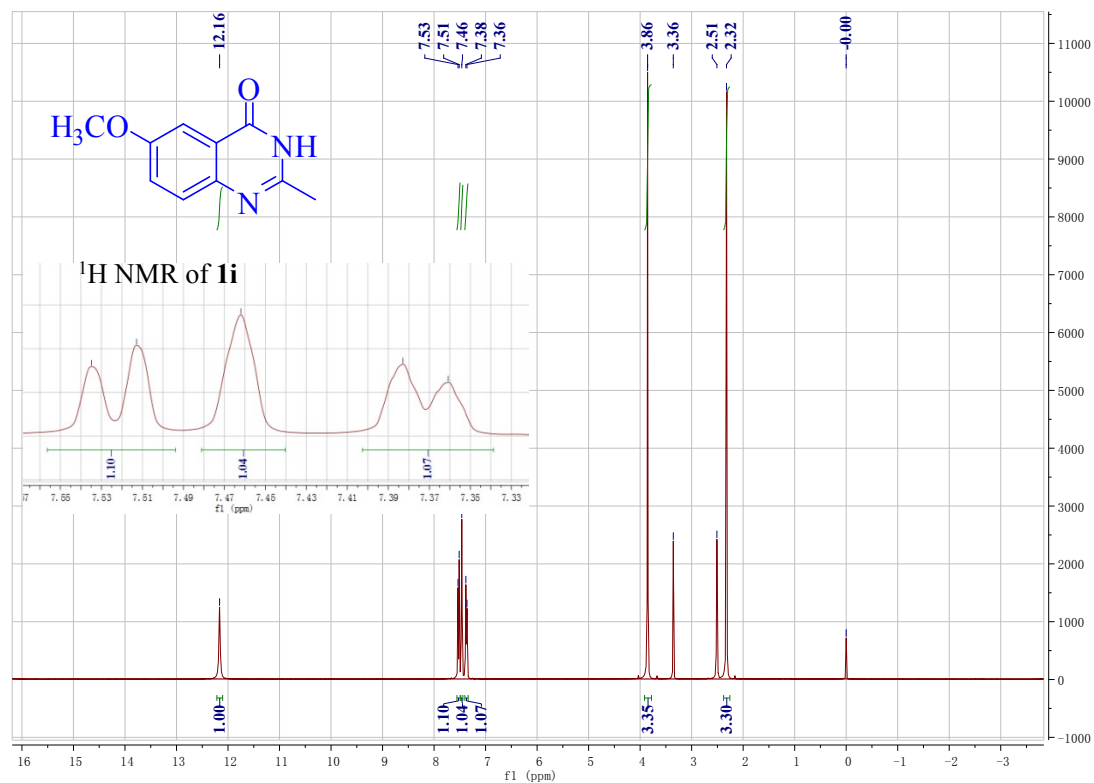
2,3-Dimethylquinazolin-4(3H)-one (**1g**)



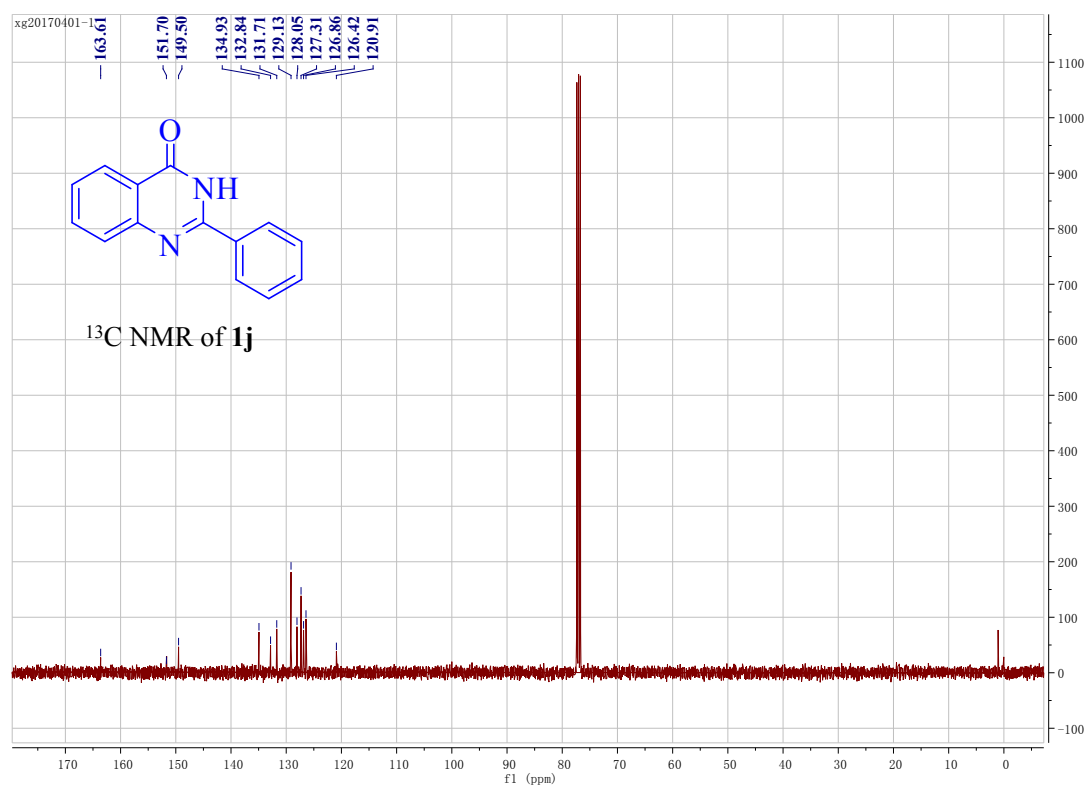
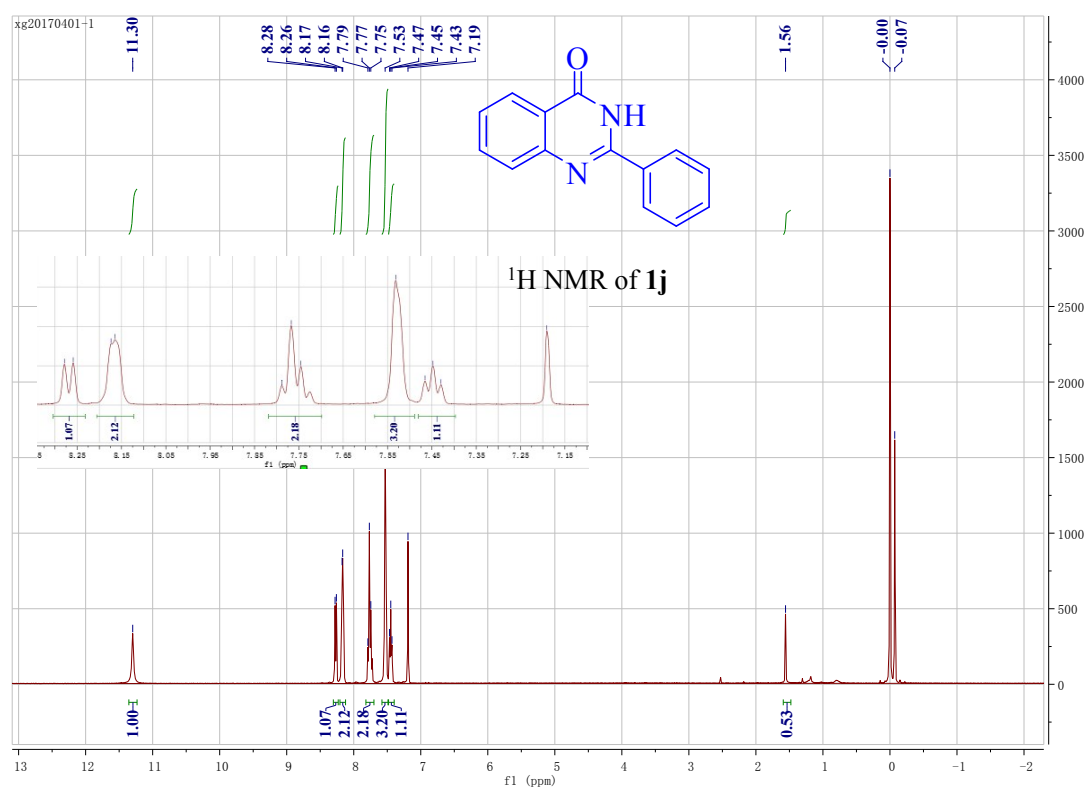
6-Chloro-2,3-Dimethylquinazolin-4(3H)-one (**1h**)



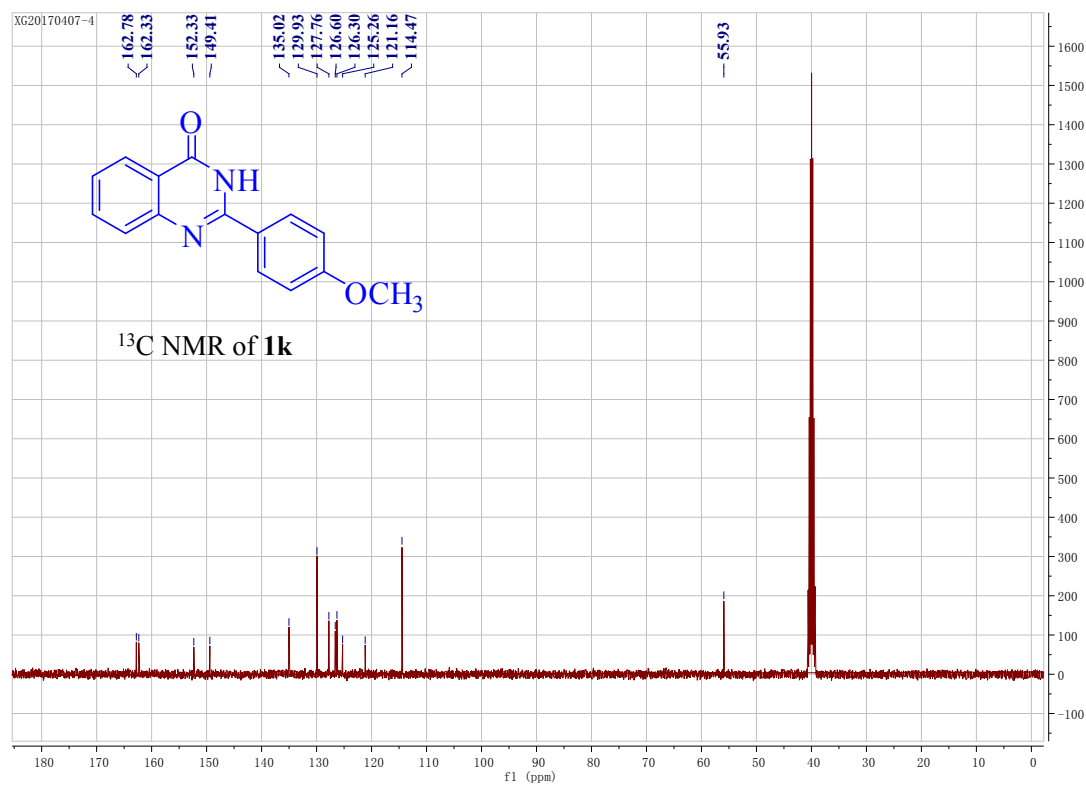
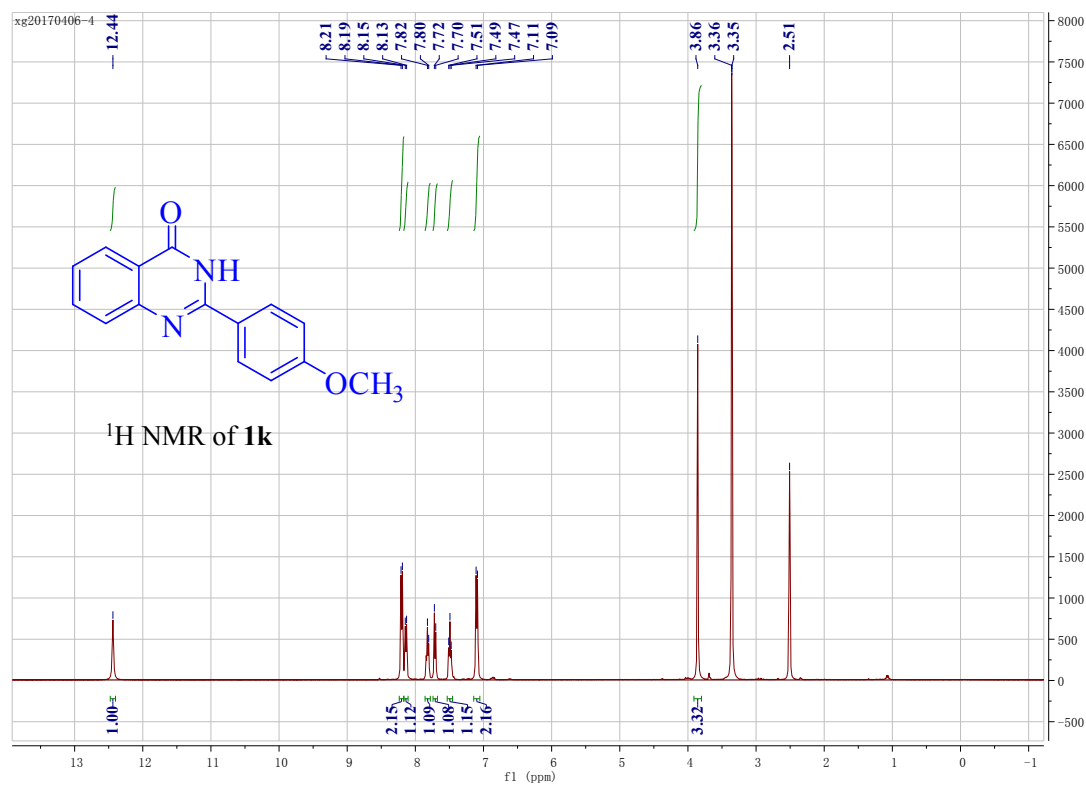
6-methoxy-2-methylquinazolin-4(3H)-one (**1i**)



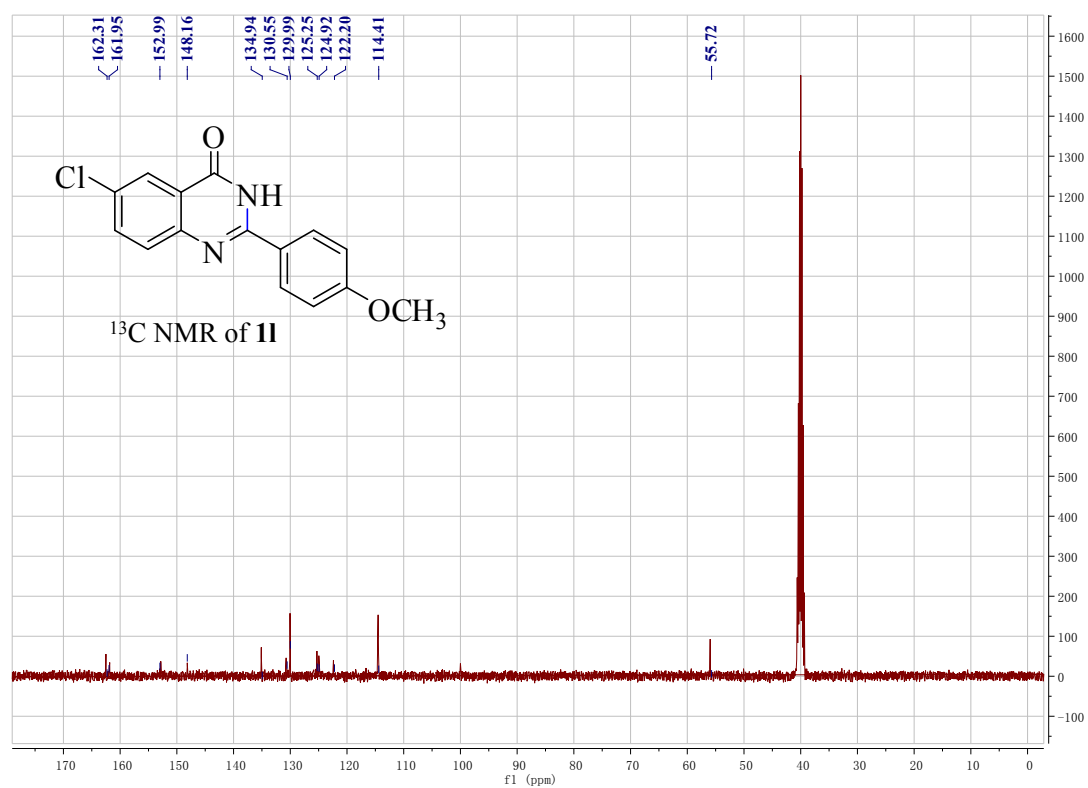
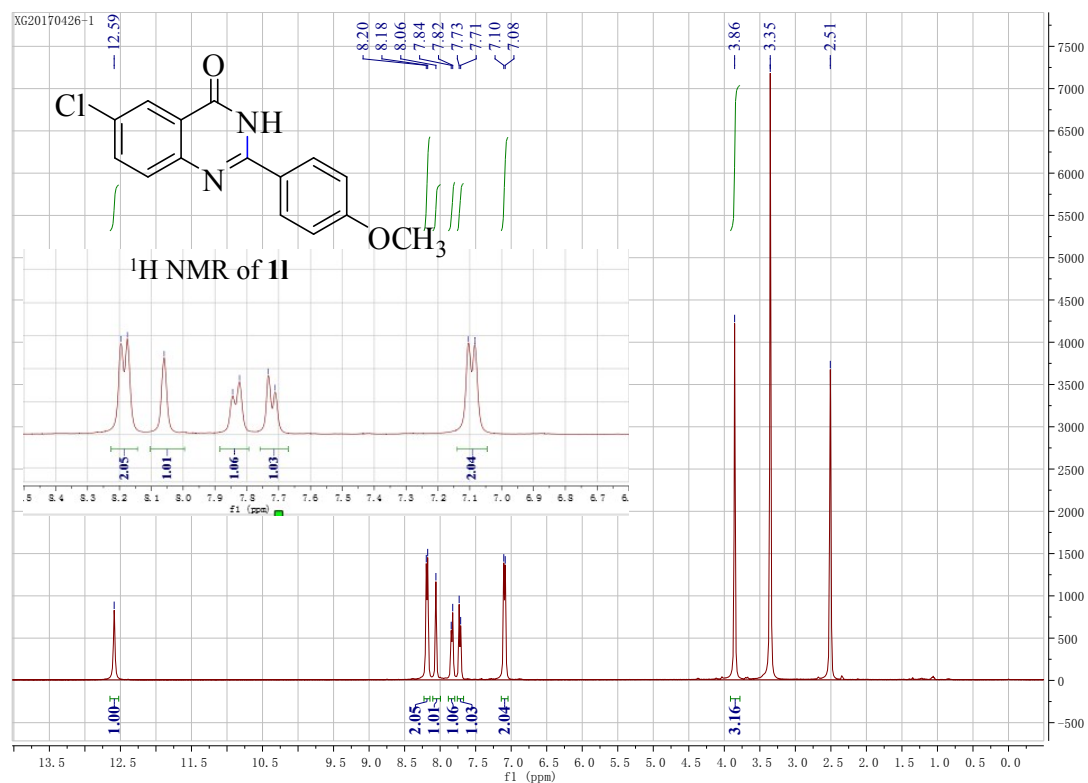
2-Phenylquinazolin-4(3H)-one (**1j**)



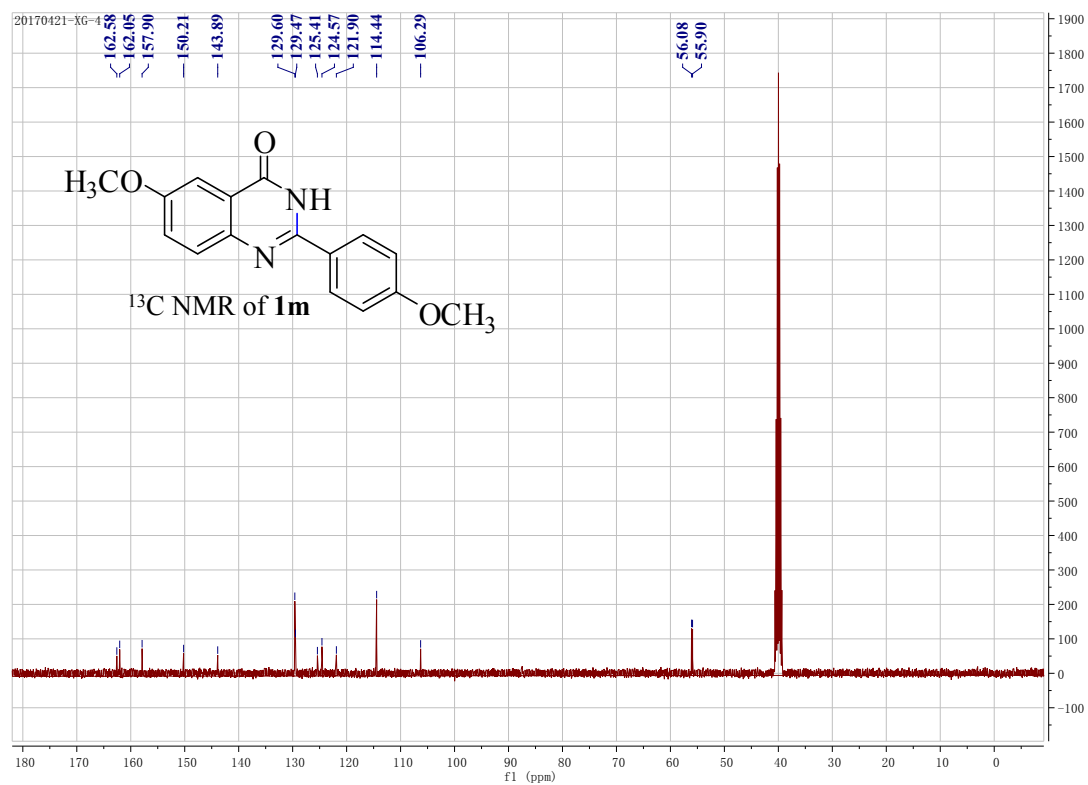
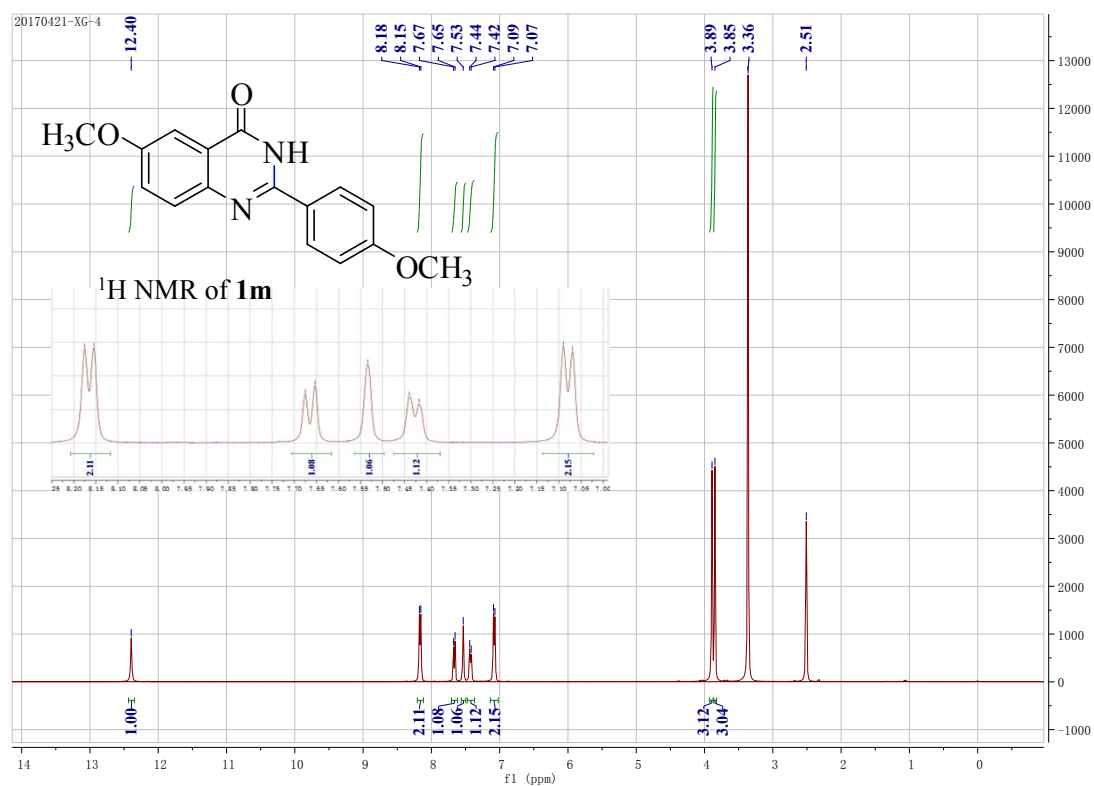
2-(4-Methoxyphenyl)quinazolin-4(3H)-one (**1k**)



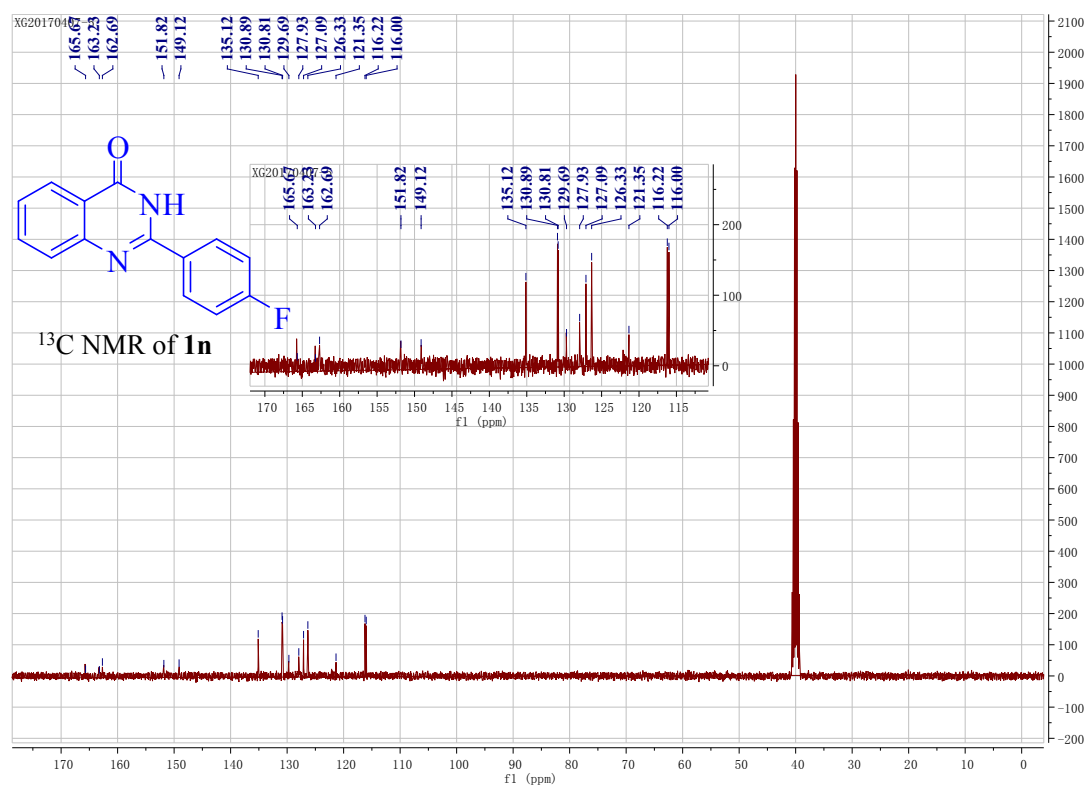
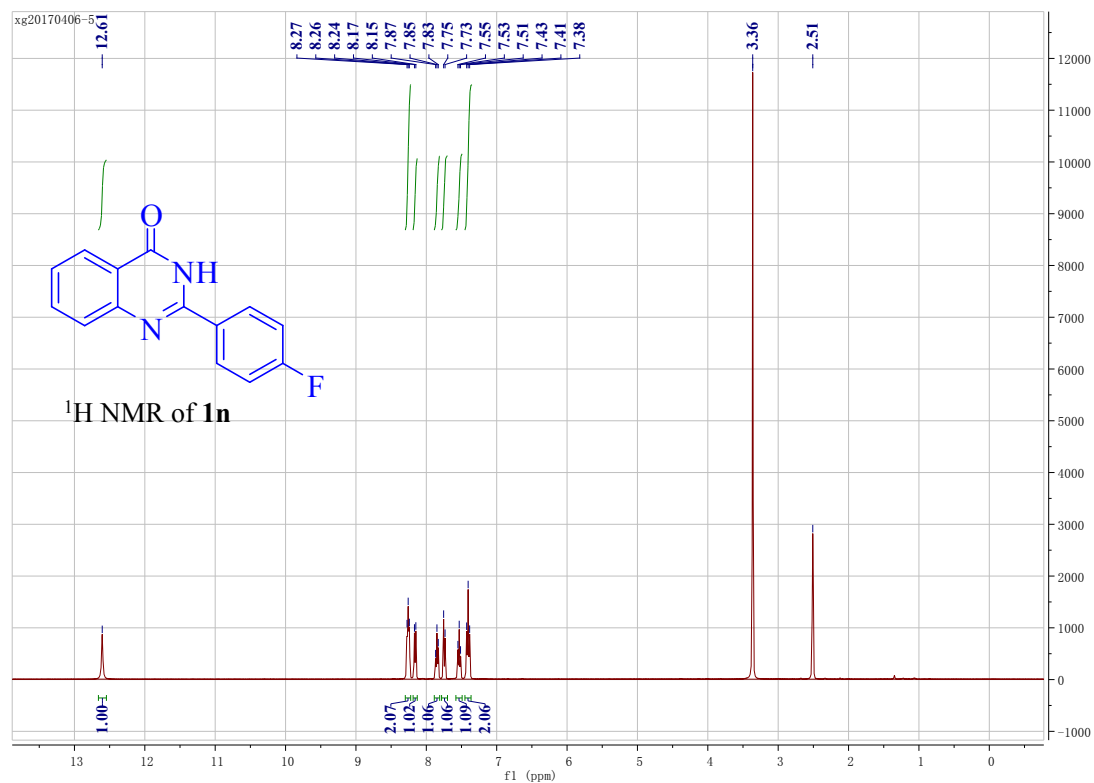
6-Chloro-2-(4-methoxyphenyl)quinazolin-4(3H)-one (**11**)



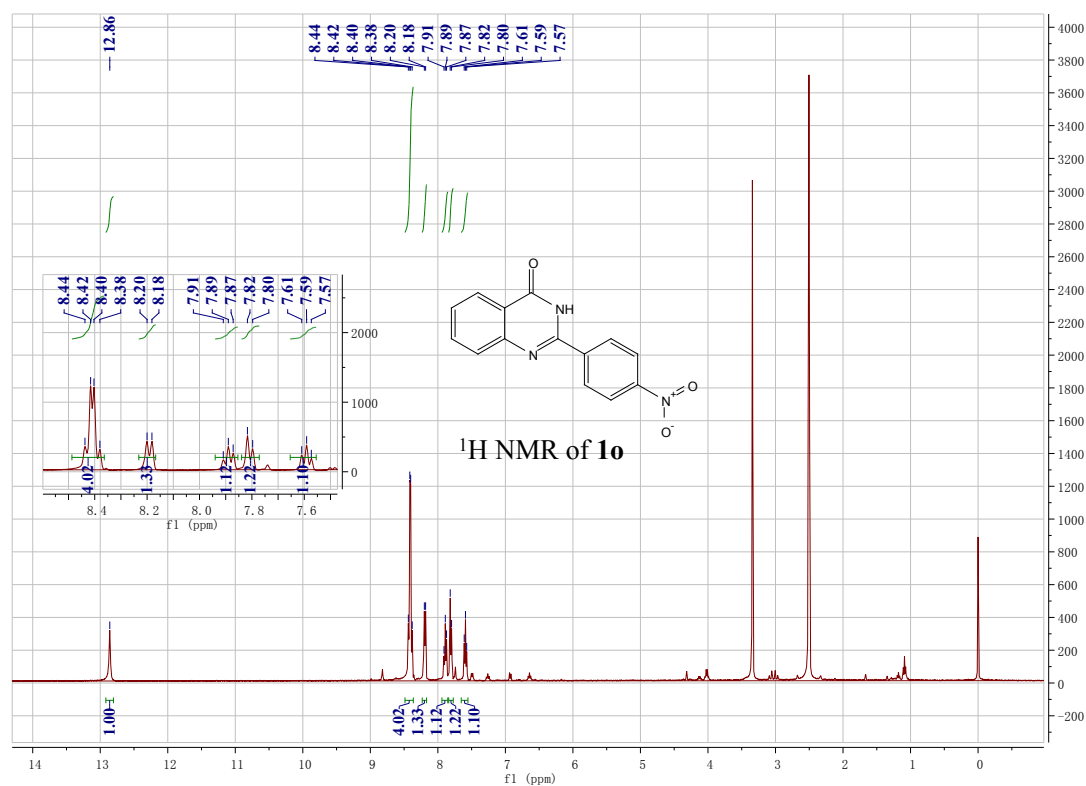
6-methoxy-2-(4-methoxyphenyl)quinazolin-4(3H)-one (**1m**)



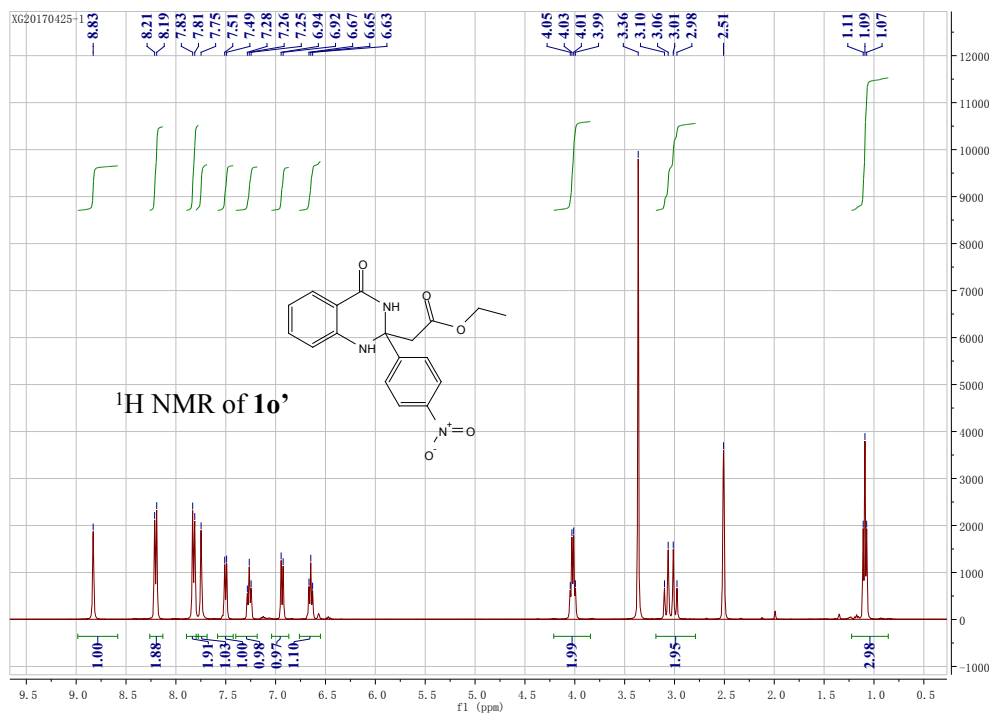
2-(4-Fluorophenyl)quinazolin-4(3H)-one (**1n**)



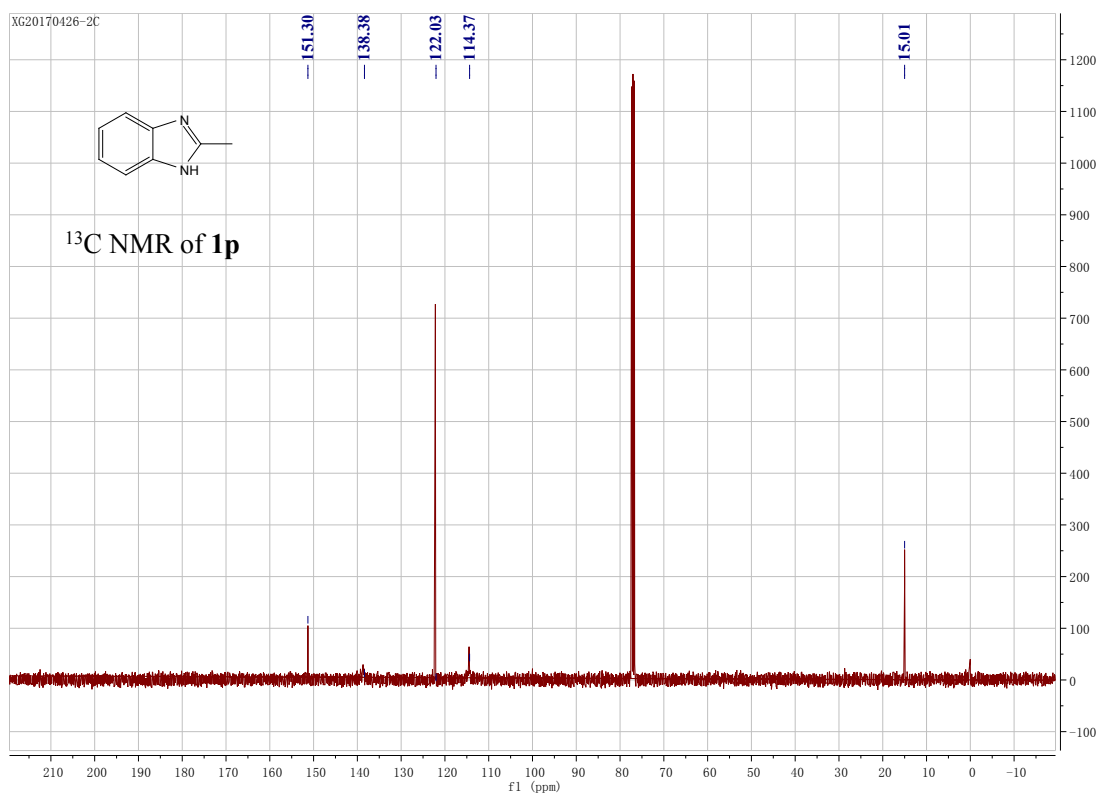
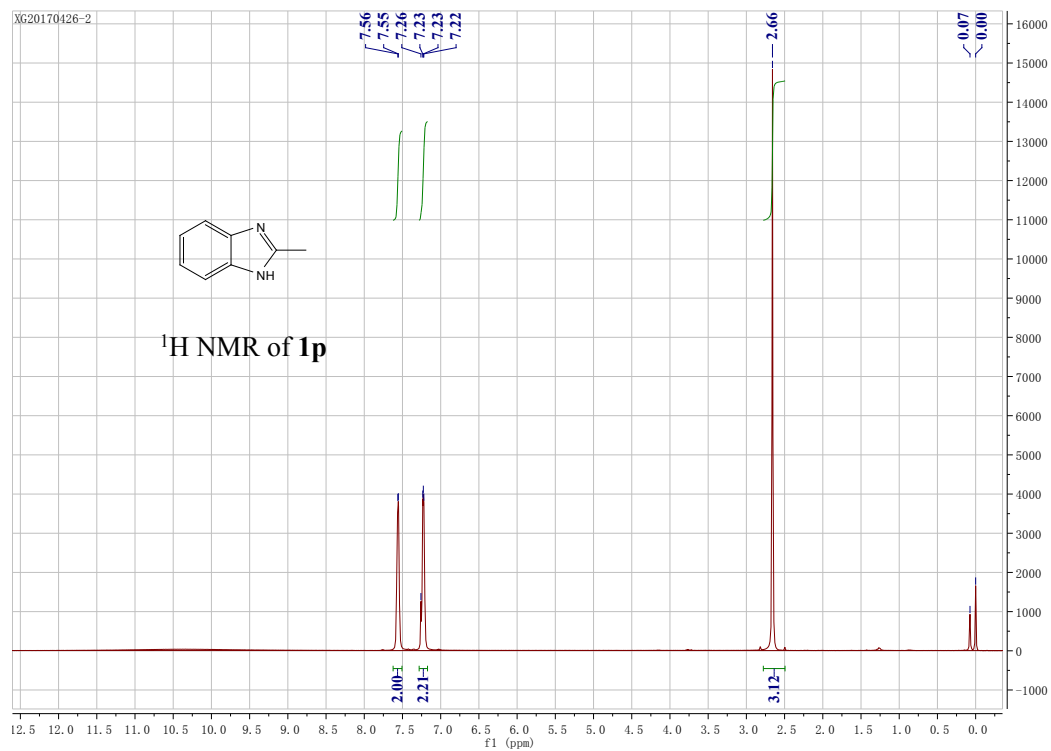
2-(4-Nitrophenyl)quinazolin-4(3H)-one (1o)



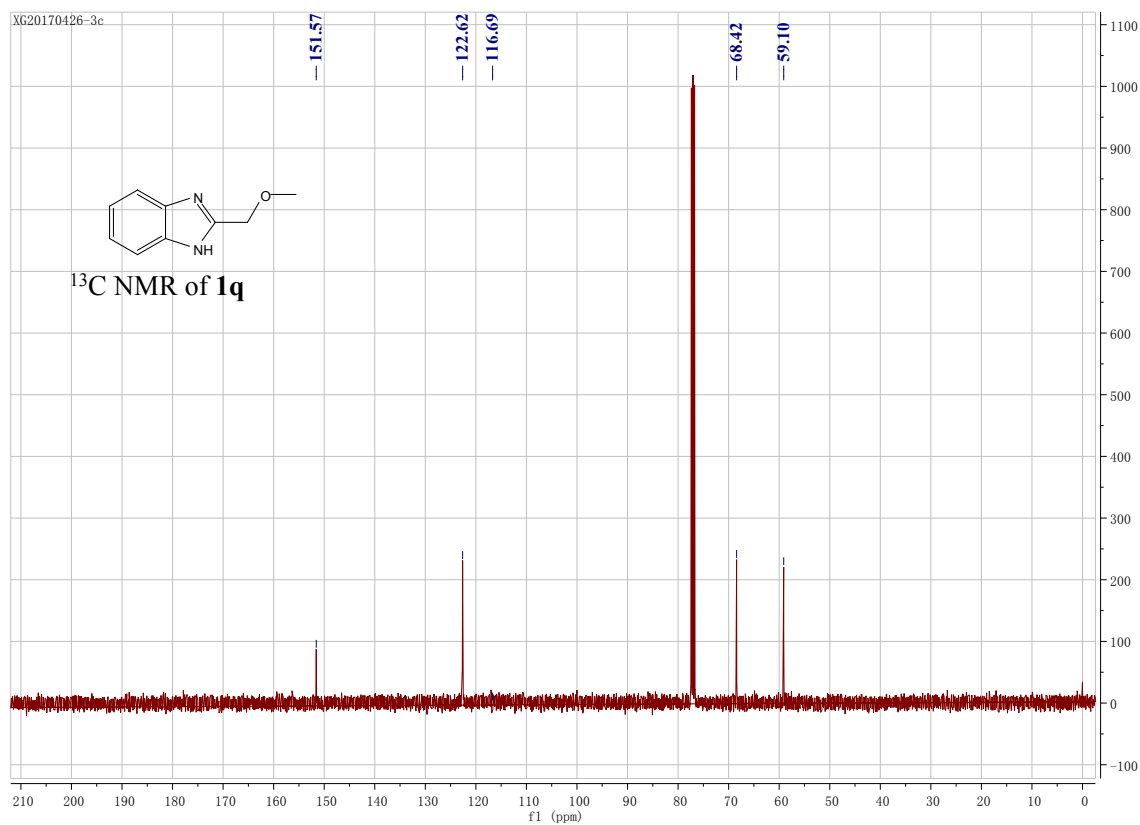
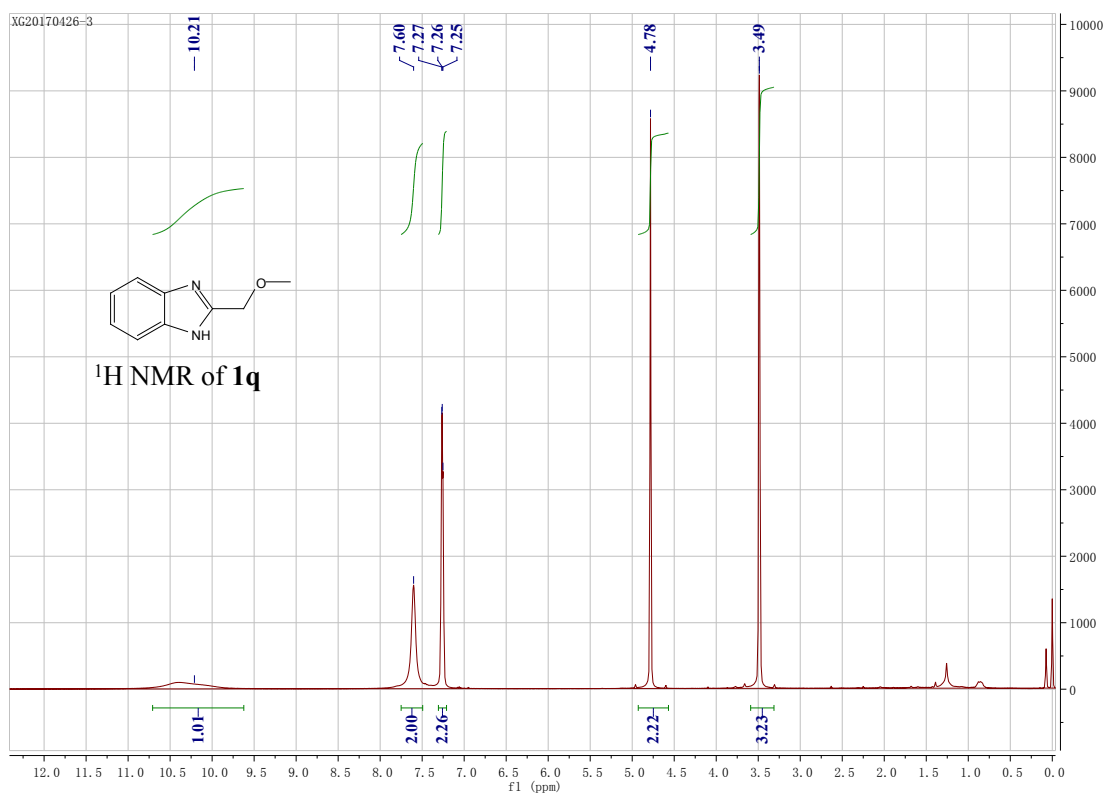
Ethyl-2-(2-(4-nitrophenyl)-4-oxo-1,2,3,4-tetrahydroquinazolin-2-yl)acetate (1o')



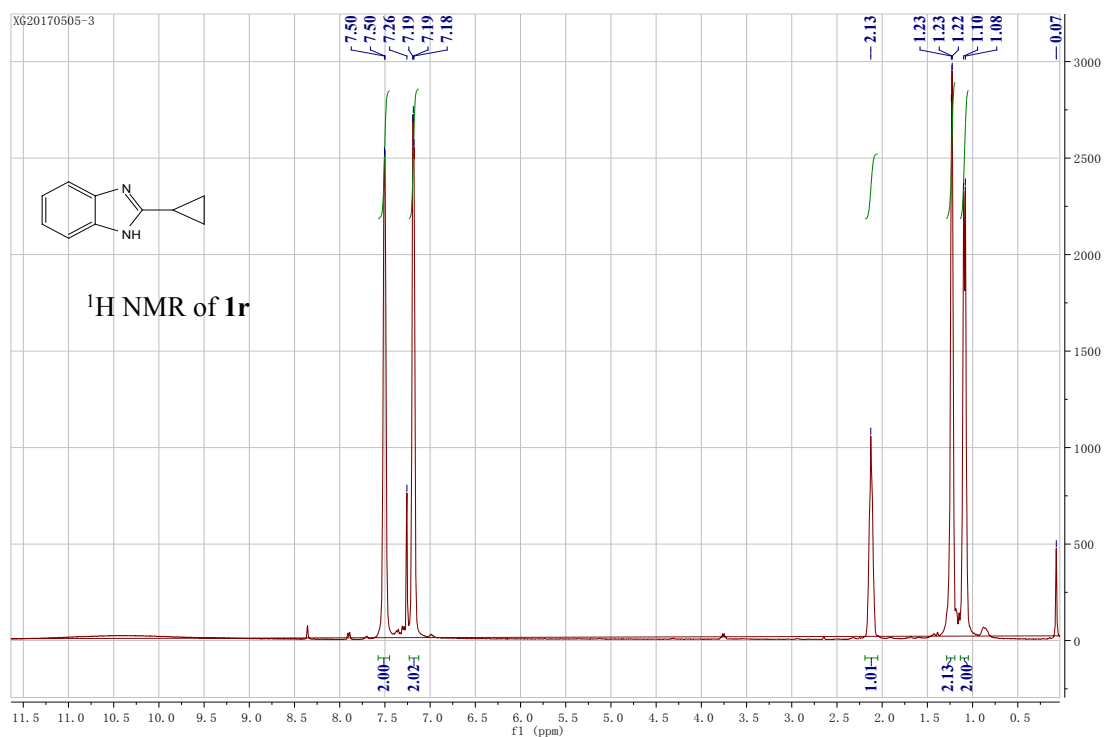
2-Methyl-1H-benzoimidazole (1p)



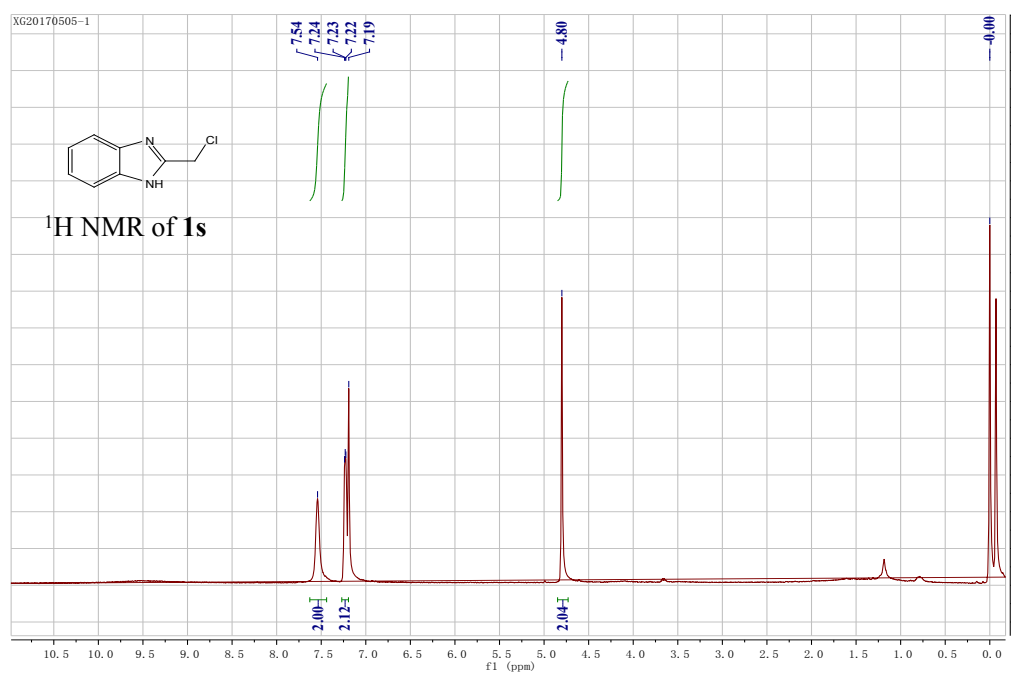
2-(Methoxymethyl)-1H-benzimidazole (1q)



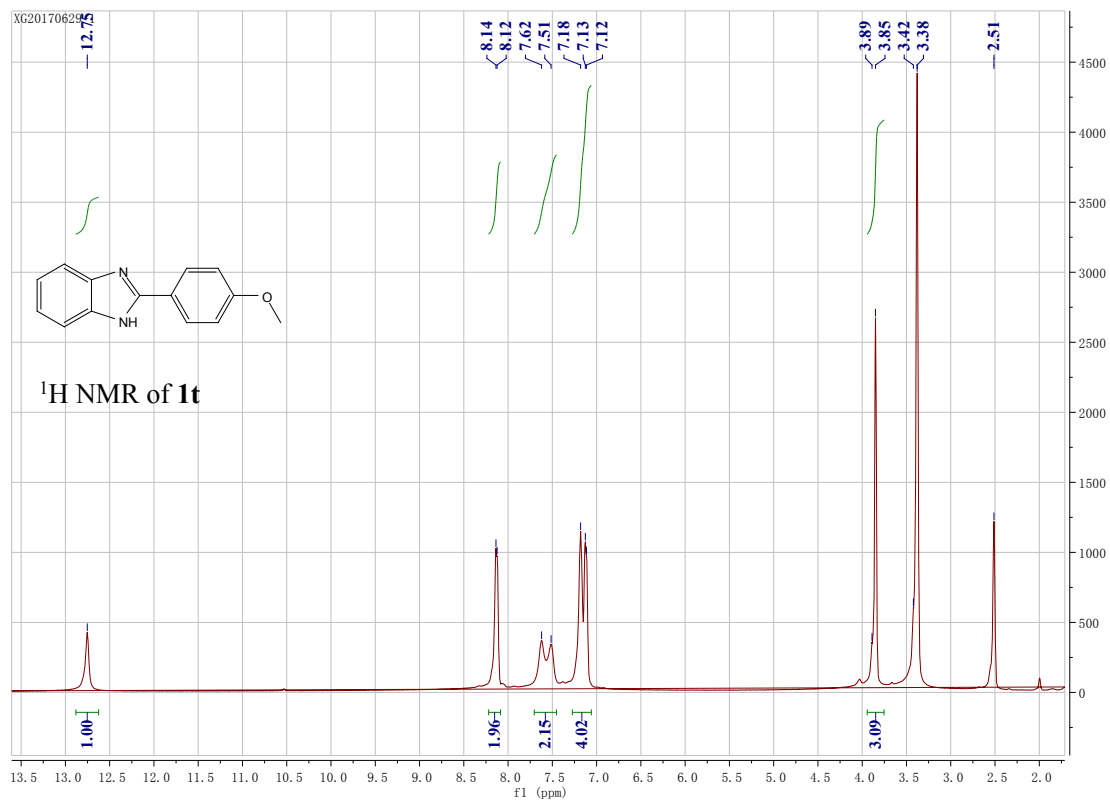
2-cyclopropyl-1H-benzo[d]imidazole (1r)



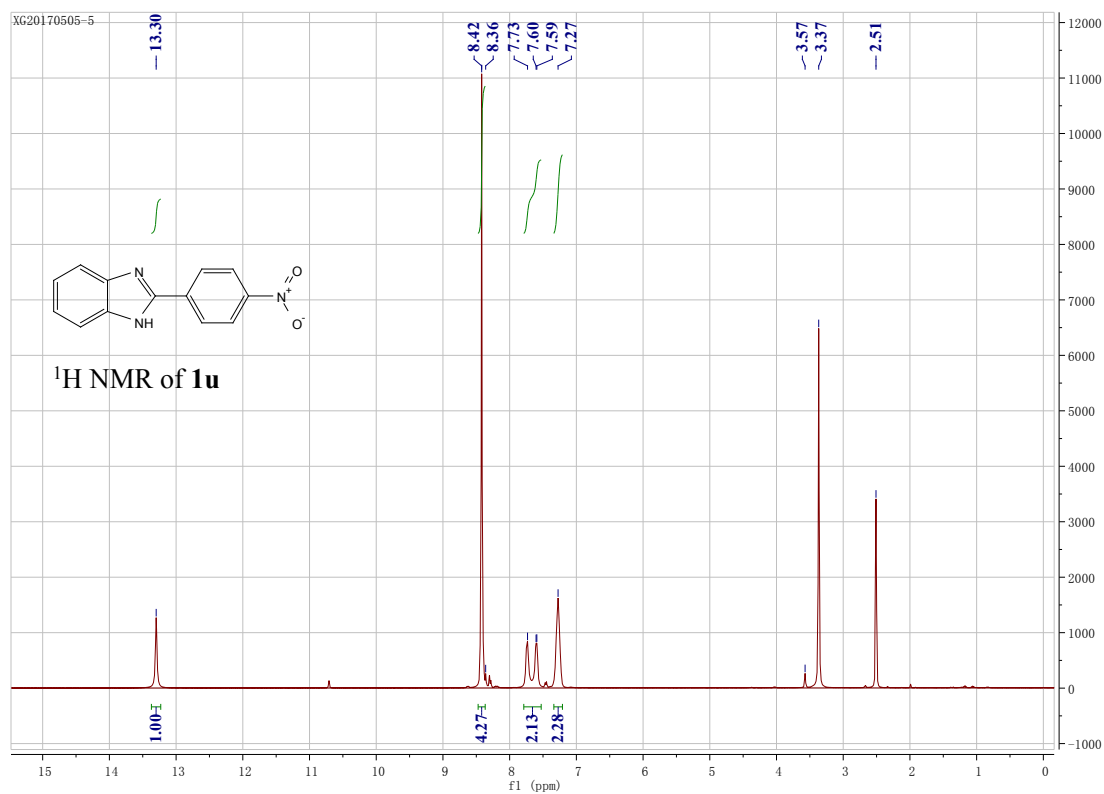
2-chloromethyl-1H-benzimidazole (1s)



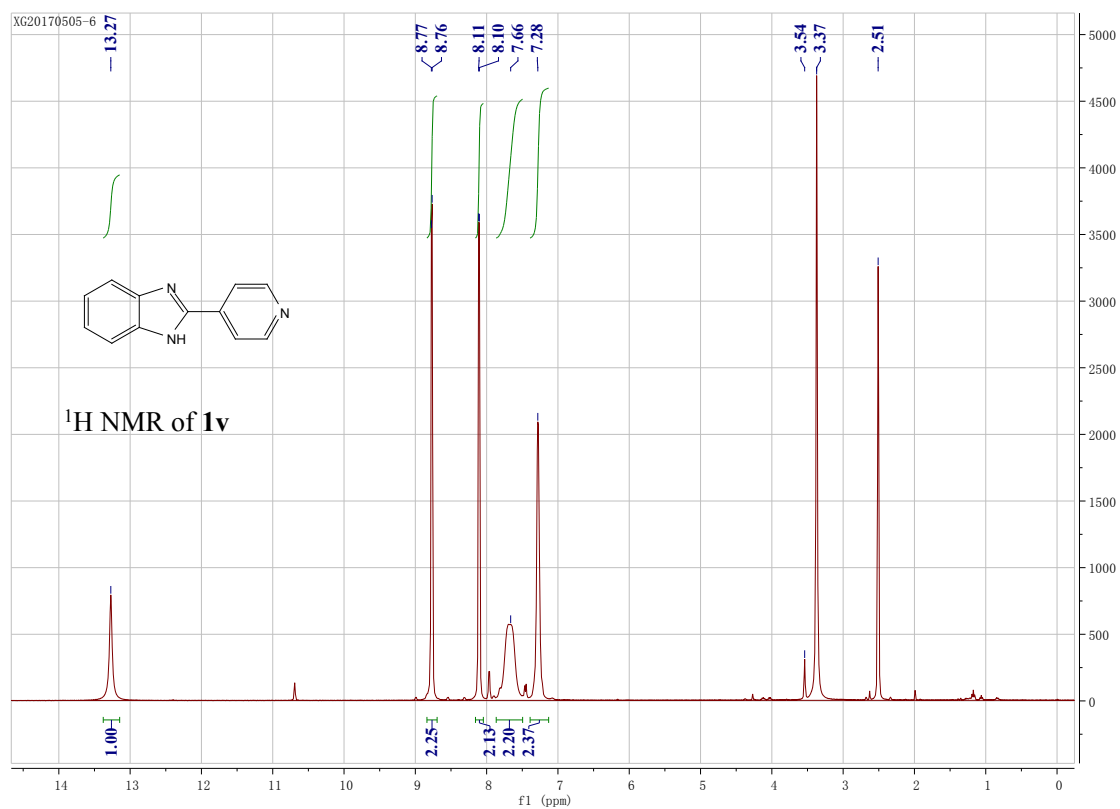
2-(4-Methoxyphenyl)-1H-benzo[d]imidazole (1t)



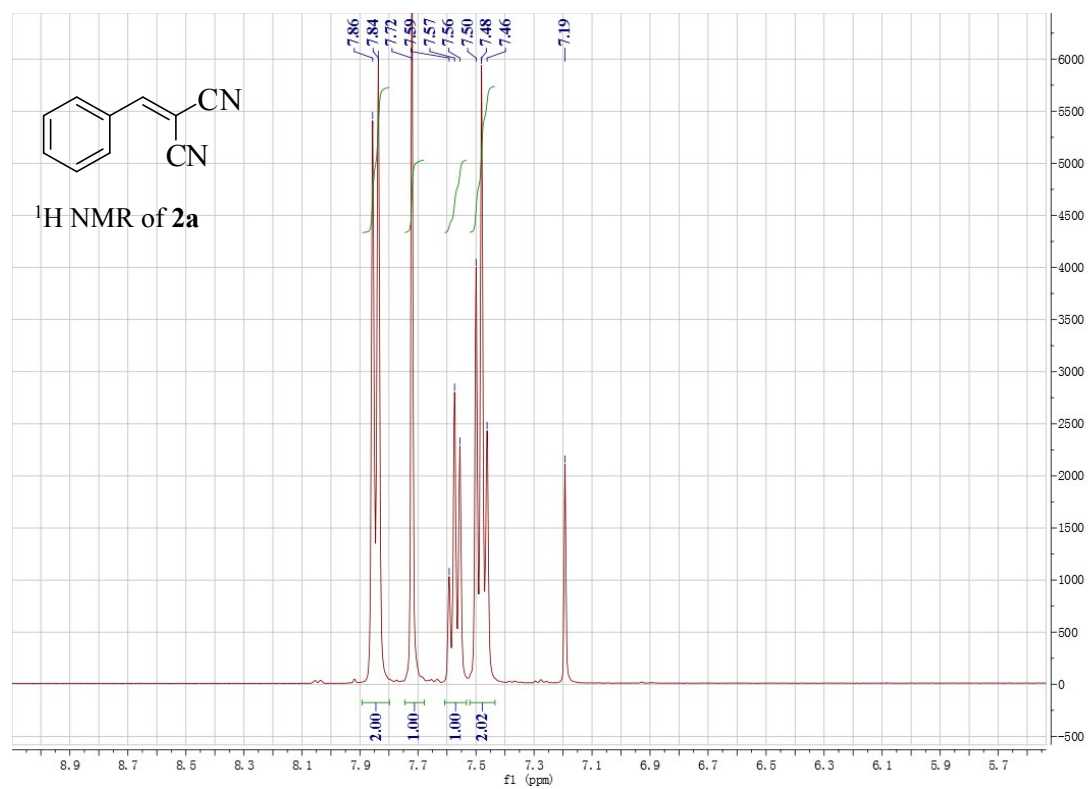
2-(4-Nitrophenyl)-1H-benzo[d]imidazole (1u)



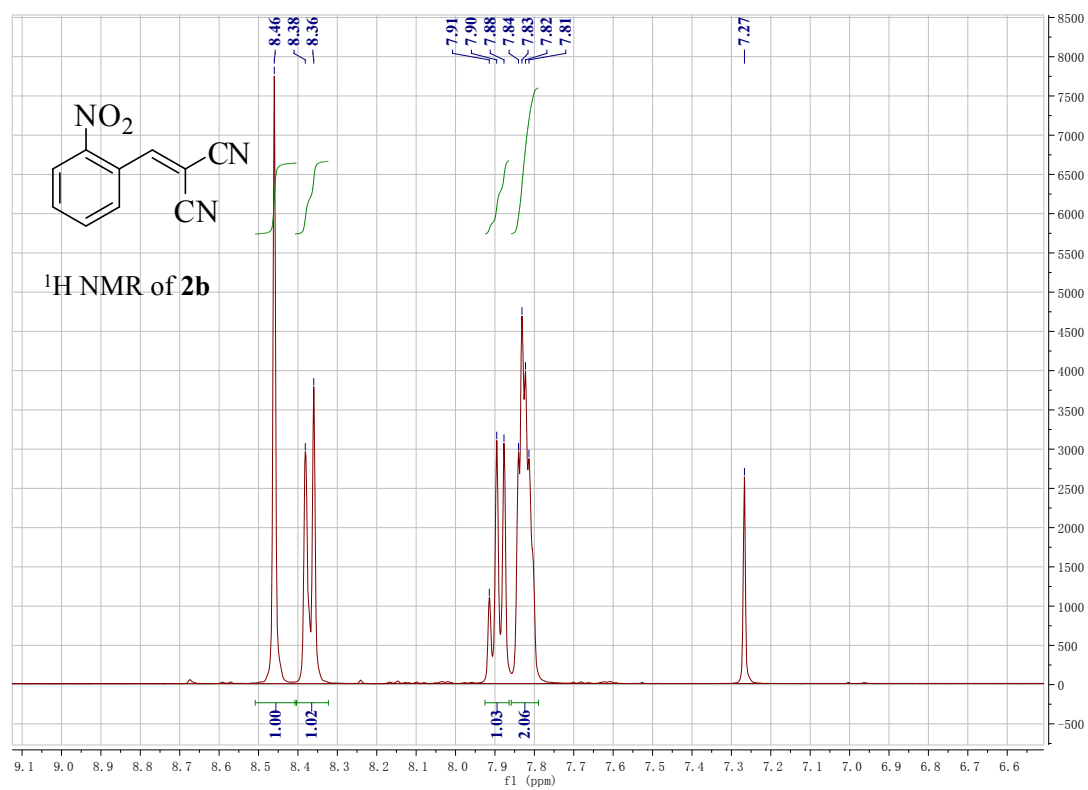
2-(pyridin-4-yl)-1H-benzo[d]imidazole (**1v**)



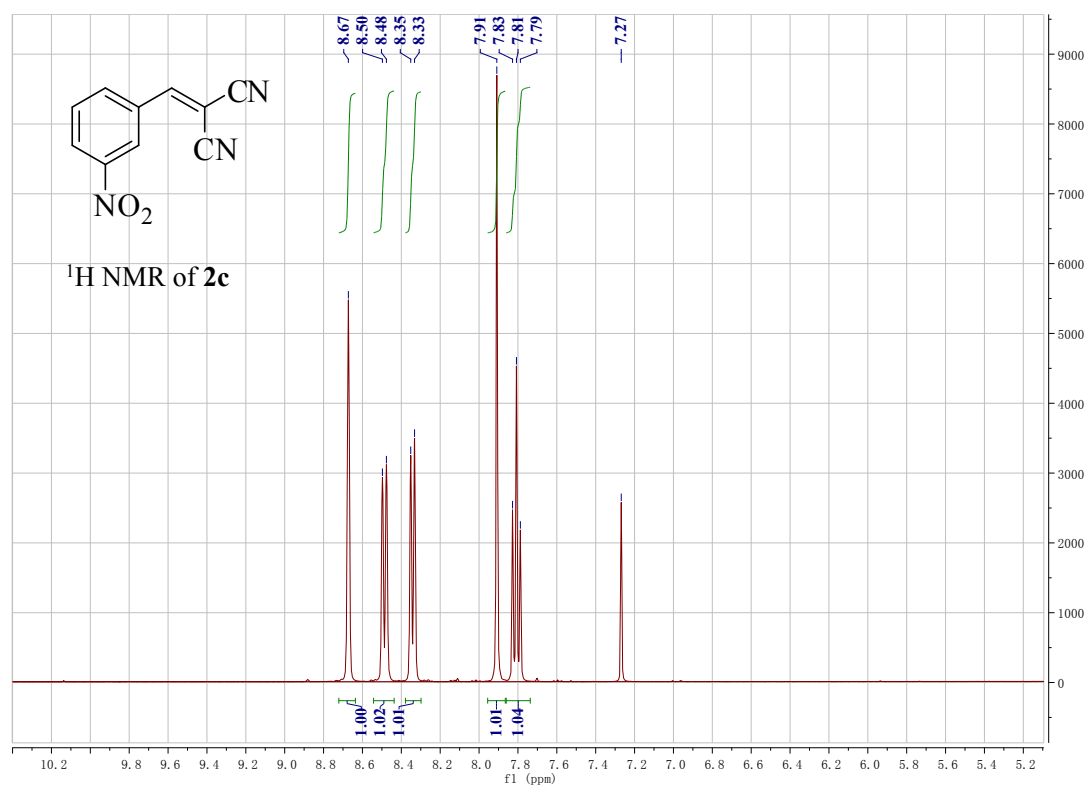
2-(Phenylmethylene)malononitrile (**2a**)



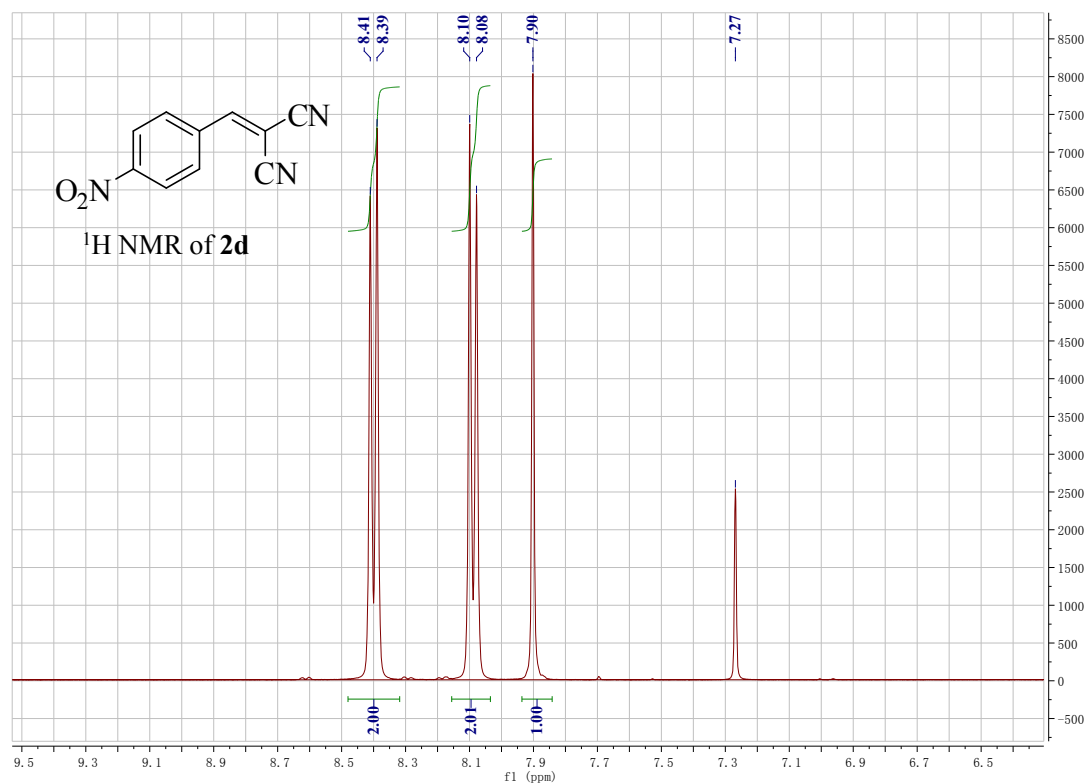
2-(2-nitrobenzylidene)malononitrile (**2b**)



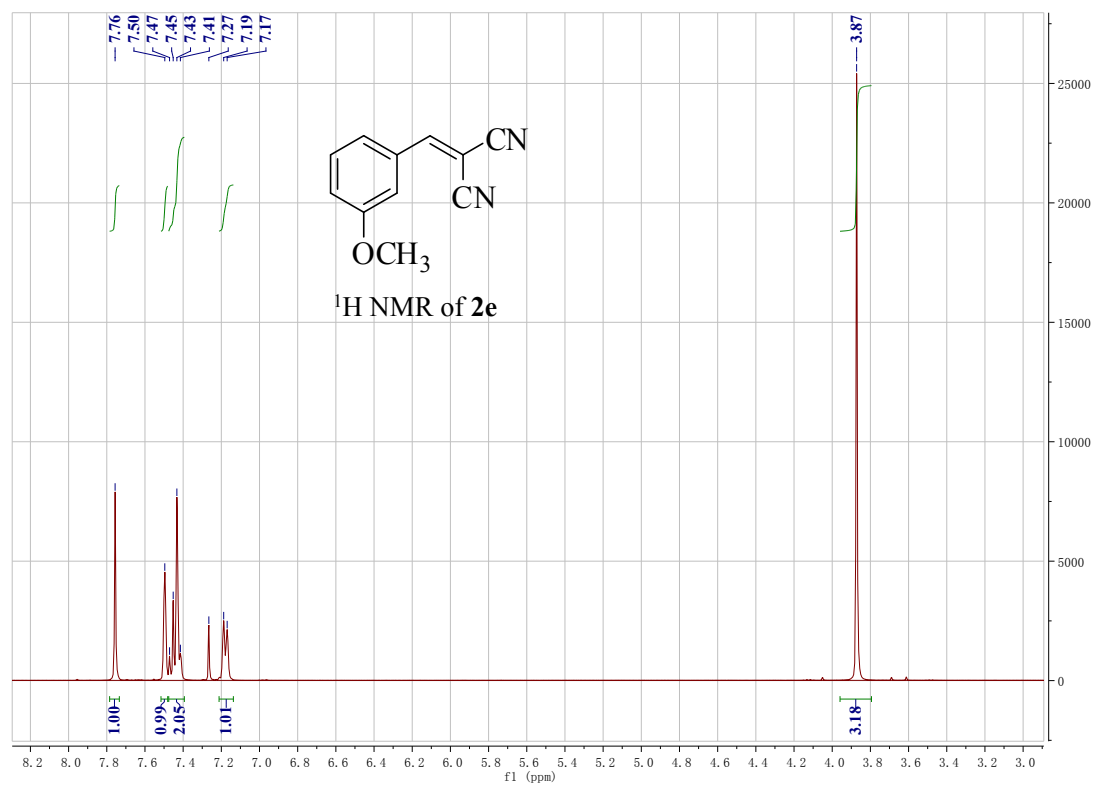
2-(3-nitrobenzylidene)malononitrile (**2c**)



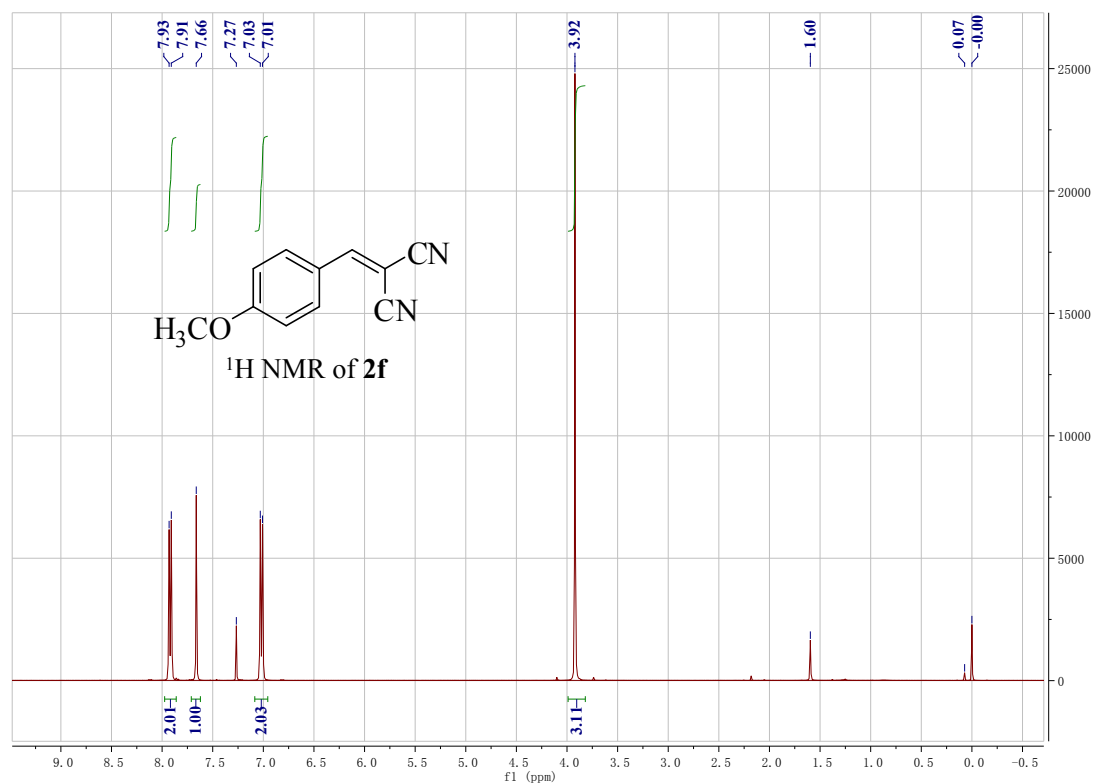
2-(4-nitrobenzylidene)malononitrile (**2d**)



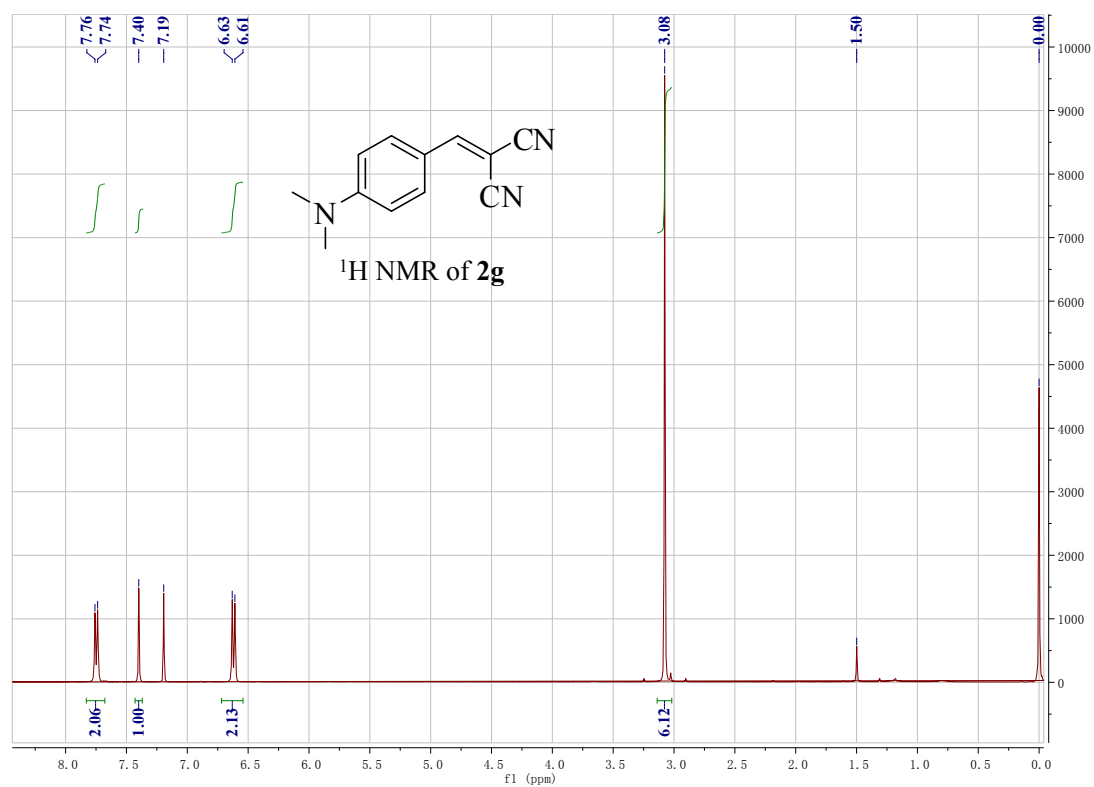
2-(3-Methoxyphenylmethylene)malononitrile (**2e**)



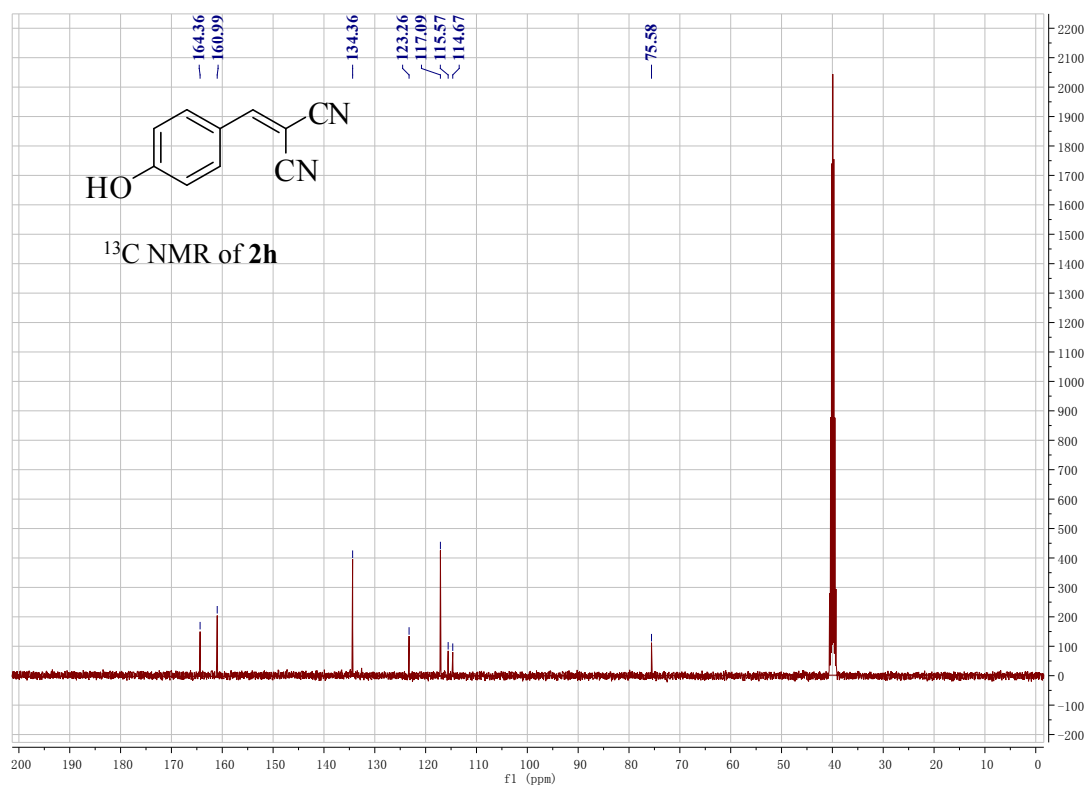
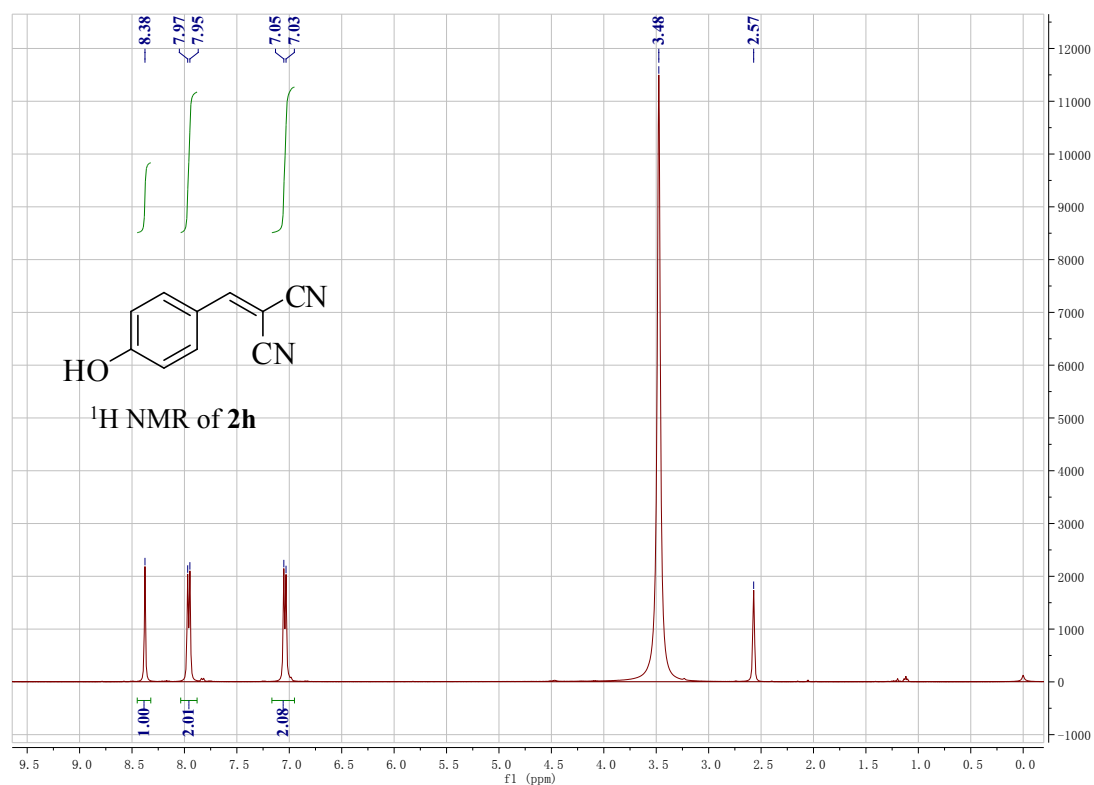
2-(4-Methoxyphenylmethylene)malononitrile (**2f**)



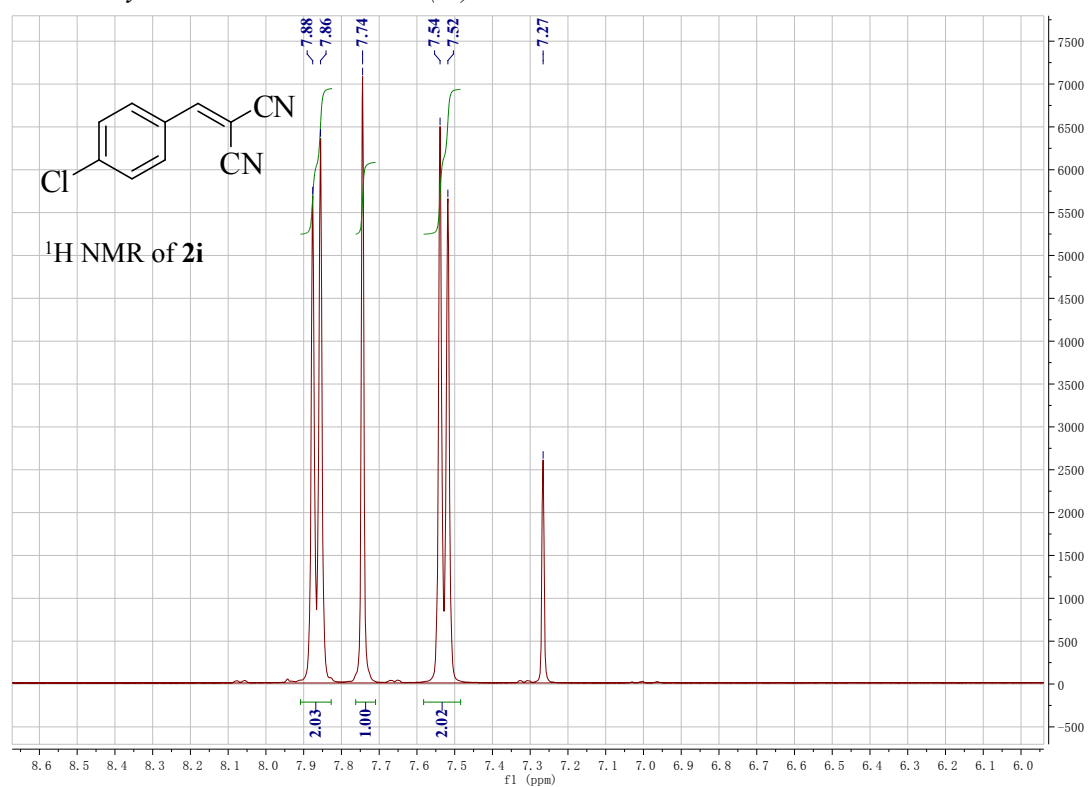
4-(N-dimethylamino benzylidene) malononitrile (**2g**)



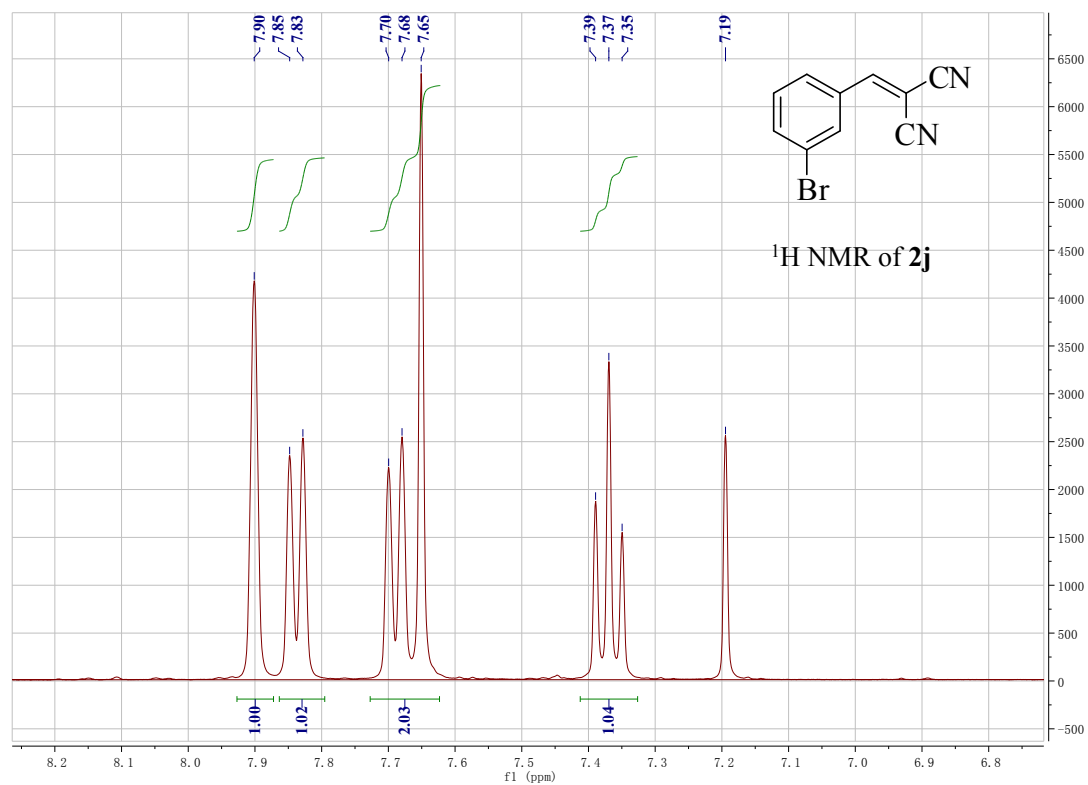
2-(4-hydroxyphenyl)methylene)malononitrile (**2h**)



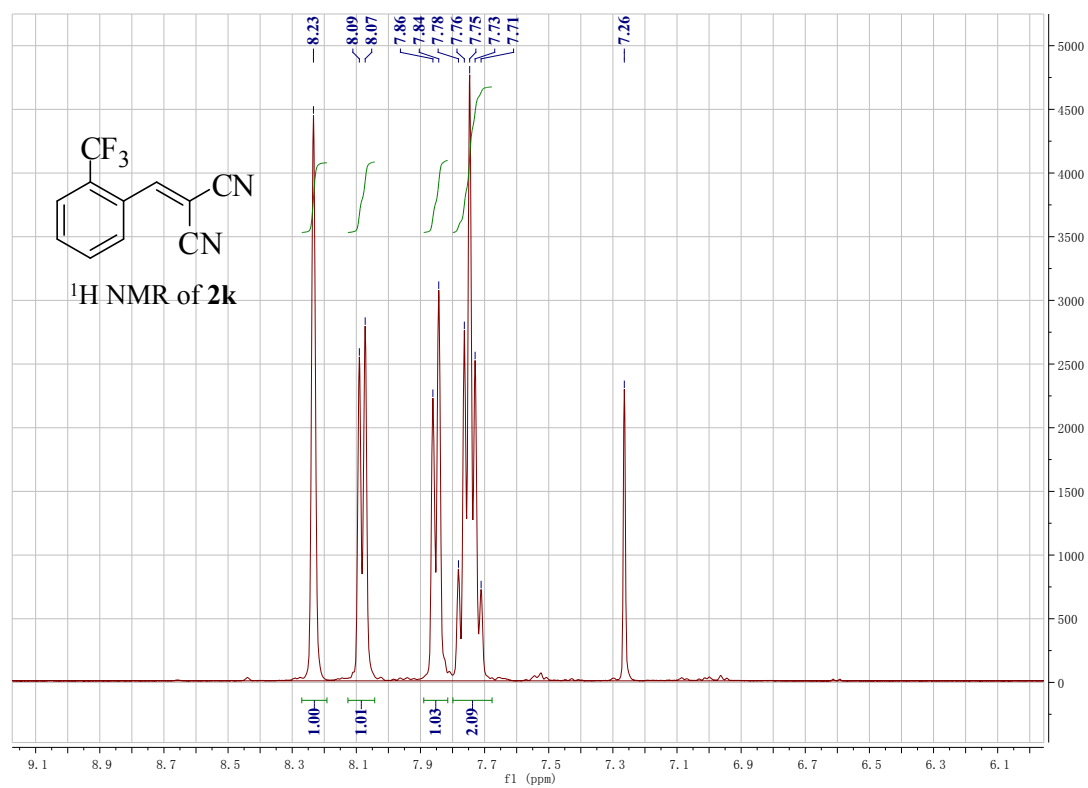
4-chlorobenzylidenemalononitrile (**2i**)



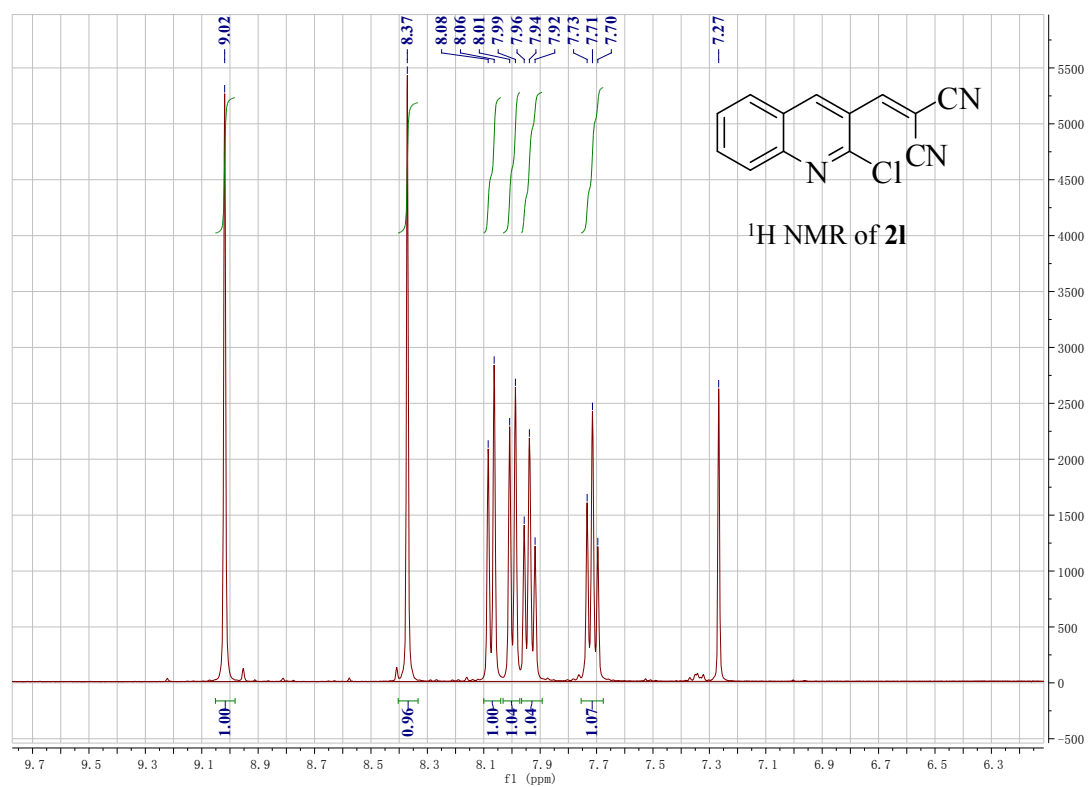
2-(3-bromobenzylidene)malononitrile (**2j**)



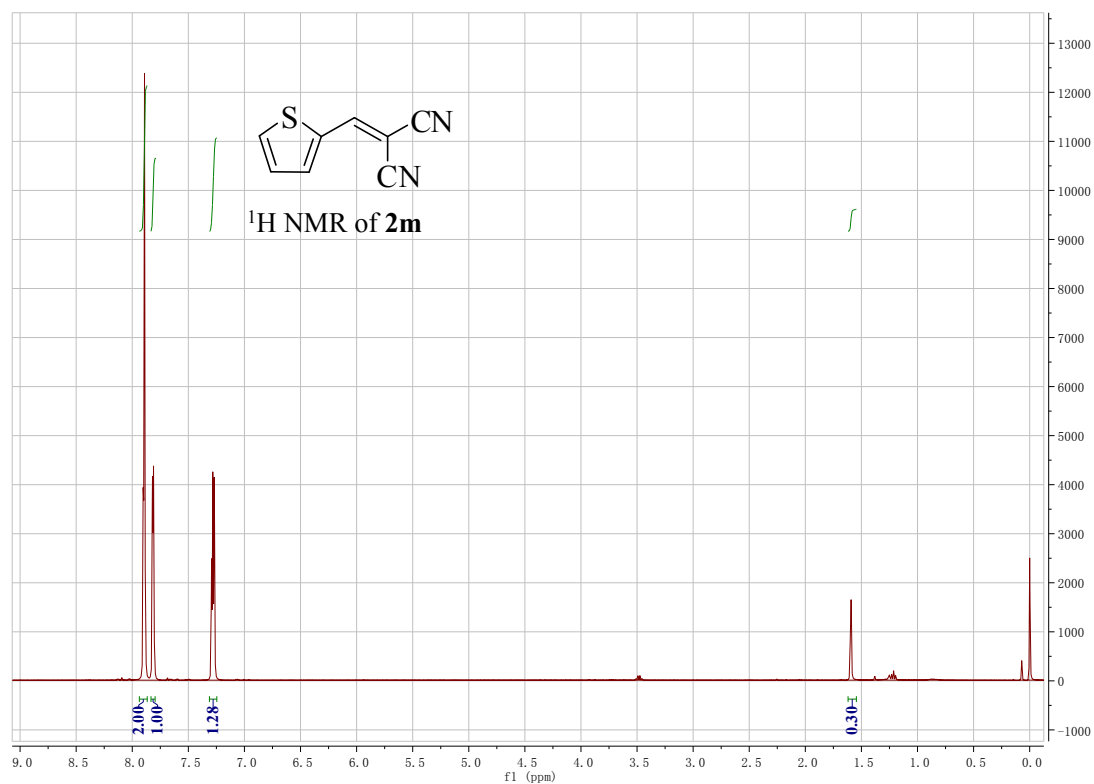
2-(2-trifluoromethylbenzylidene)malononitrile (2k)



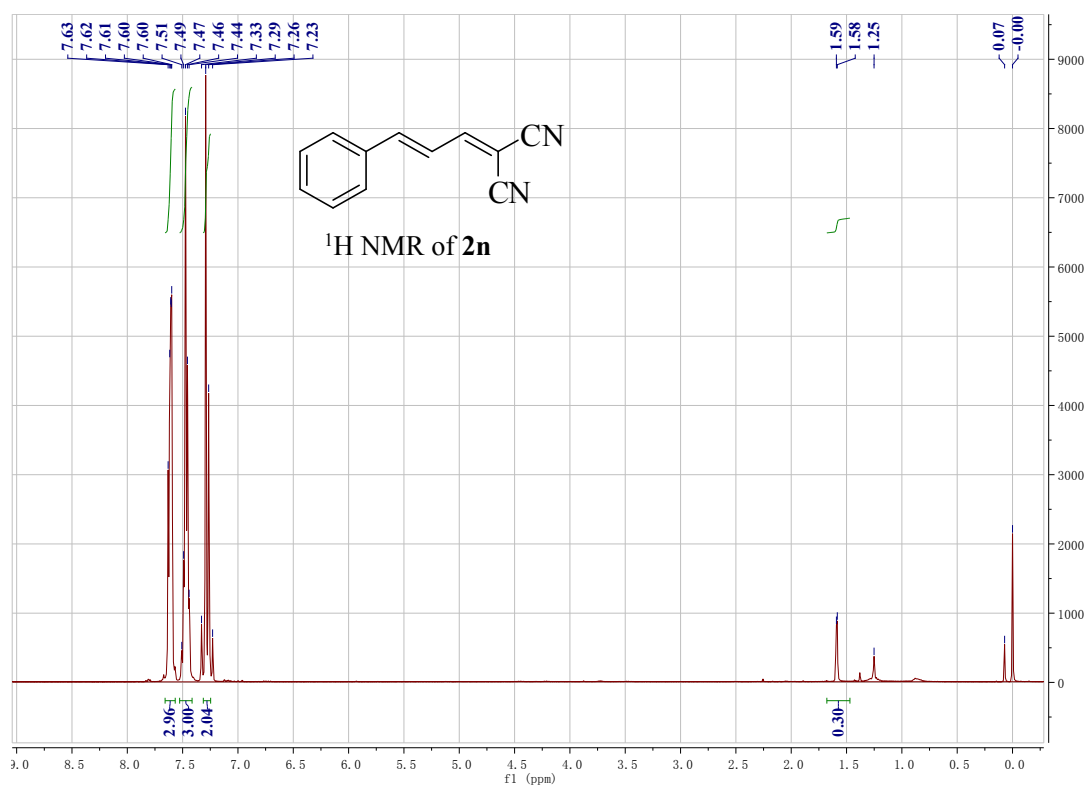
[(2-chloro-3-quinolyl)methylidene]methane-1,1-dicarbonitrile (2l)



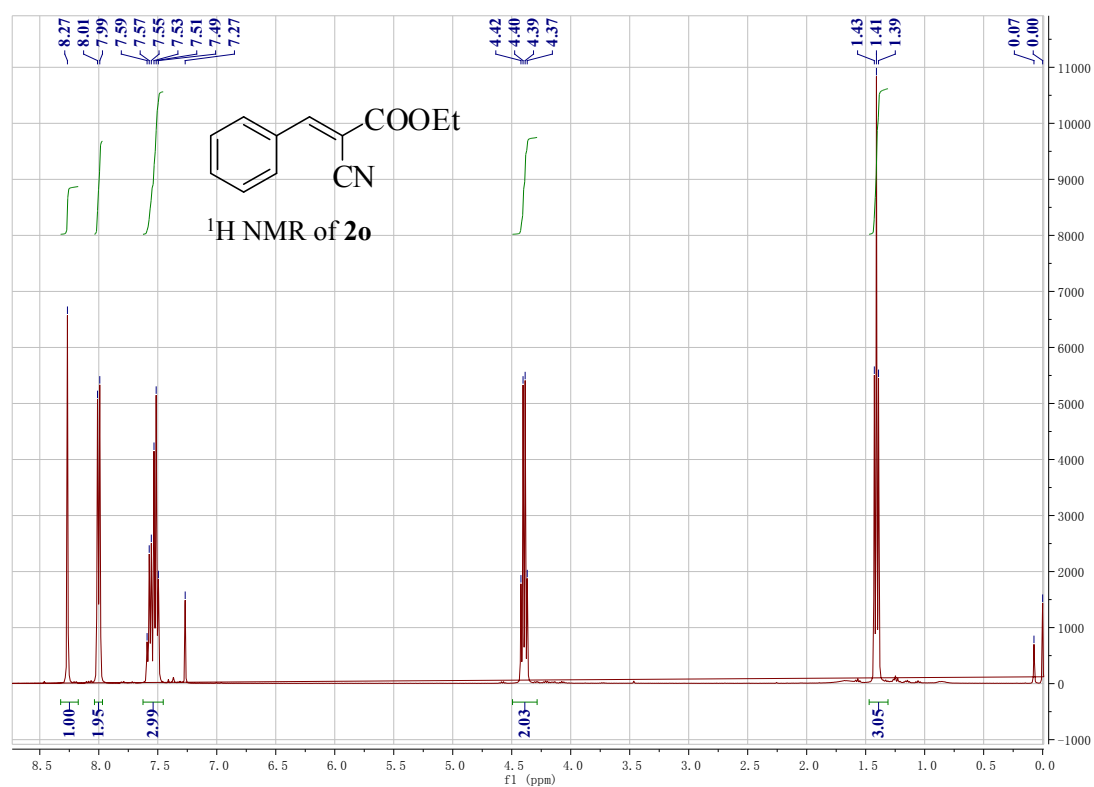
2-(2-Furylmethylene)malonitrile (**2m**)



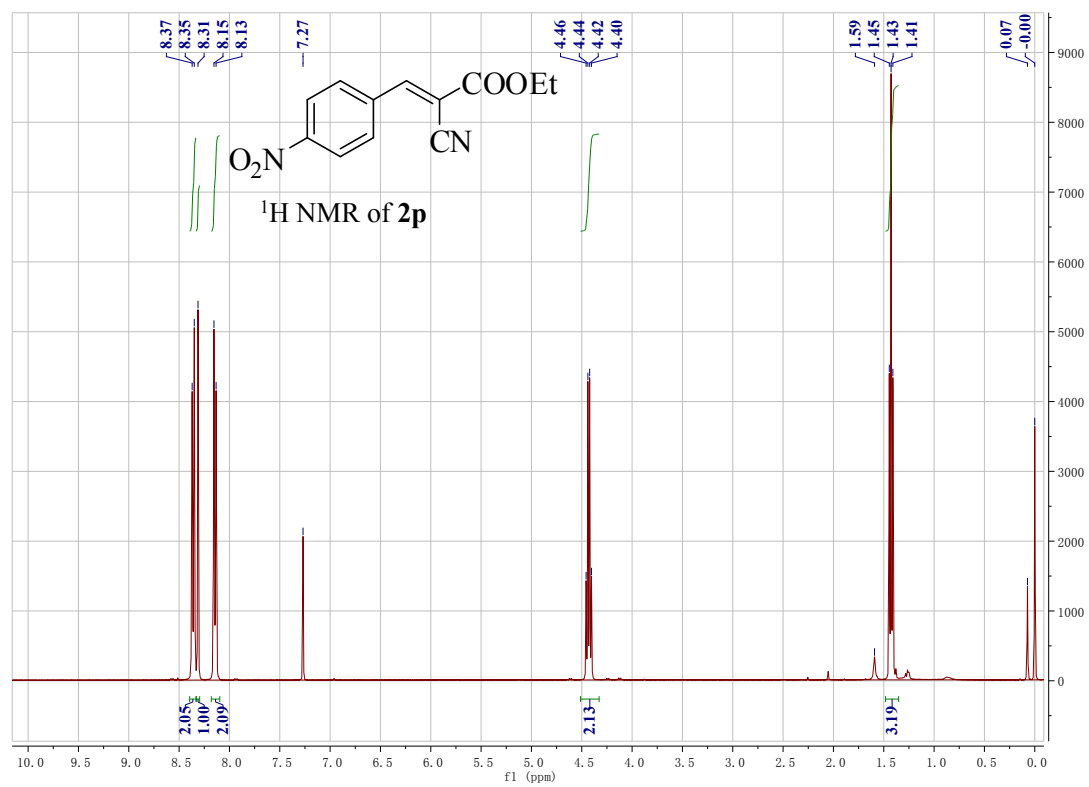
Cinnamalmalononitrile (**2n**)



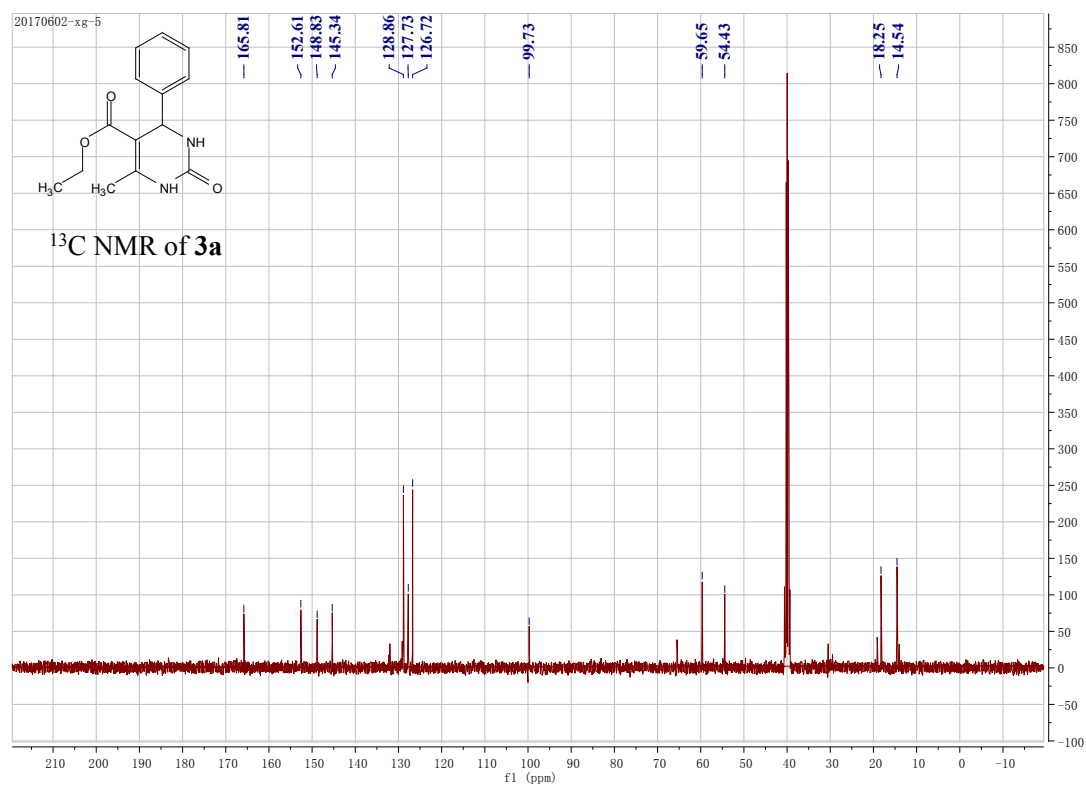
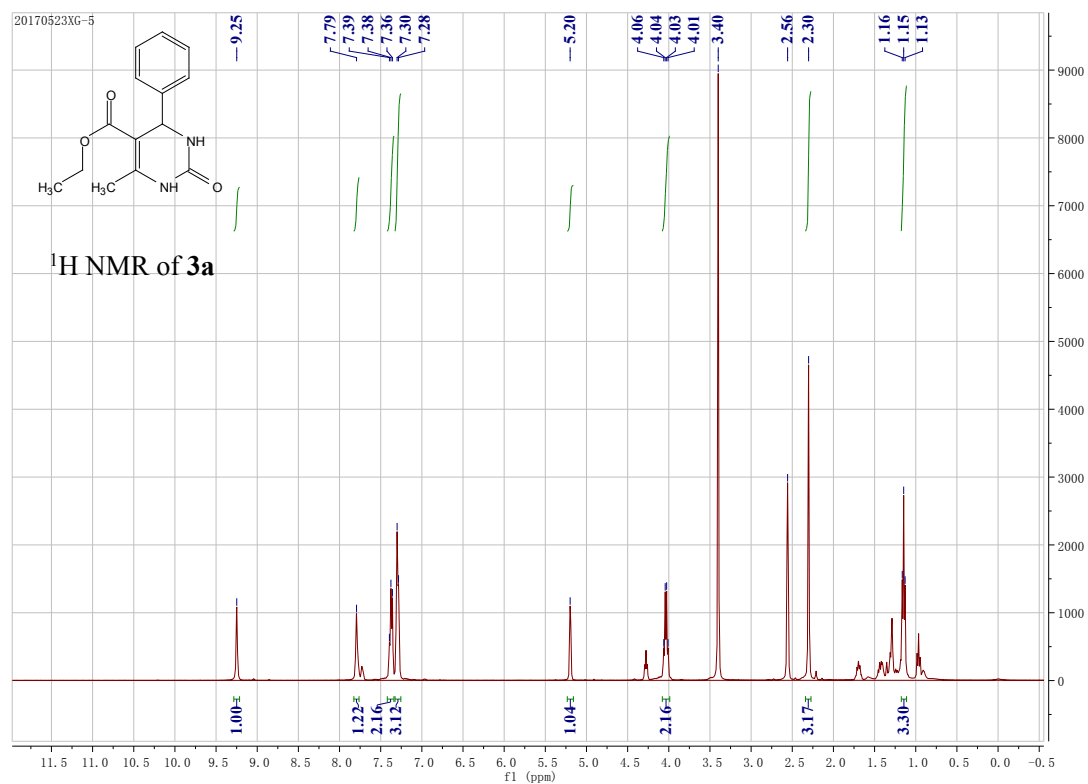
Ethyl benzylidenecyanoacetate (2o)



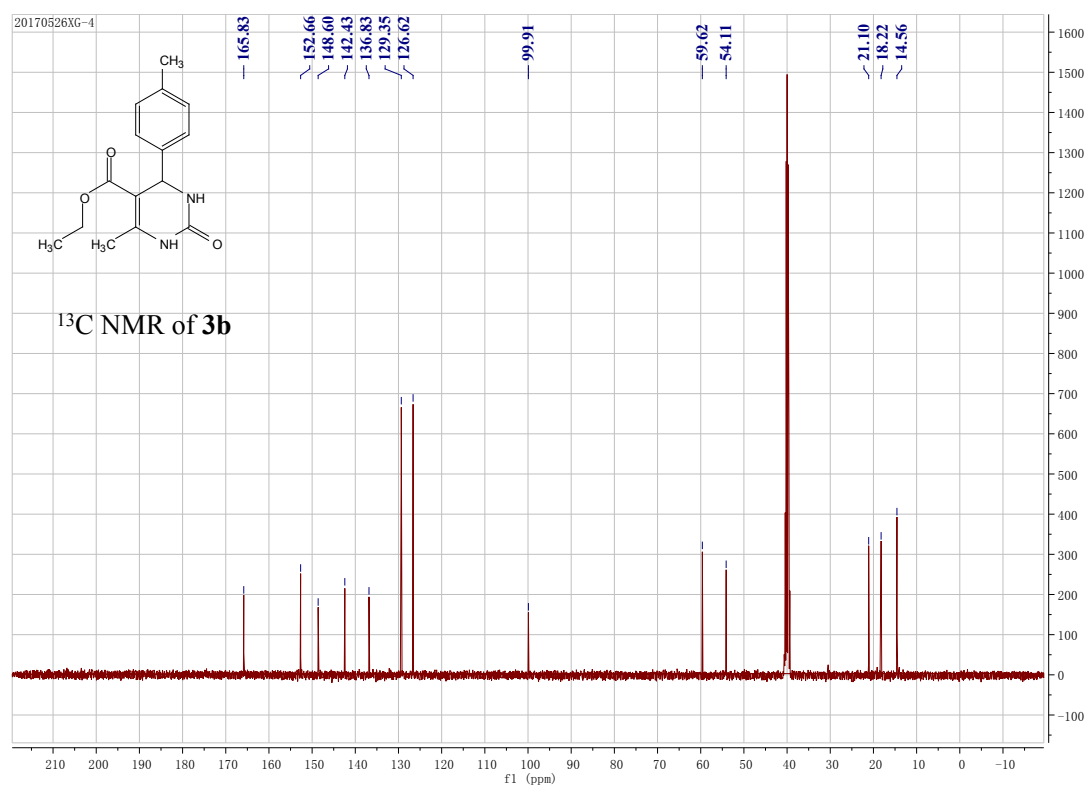
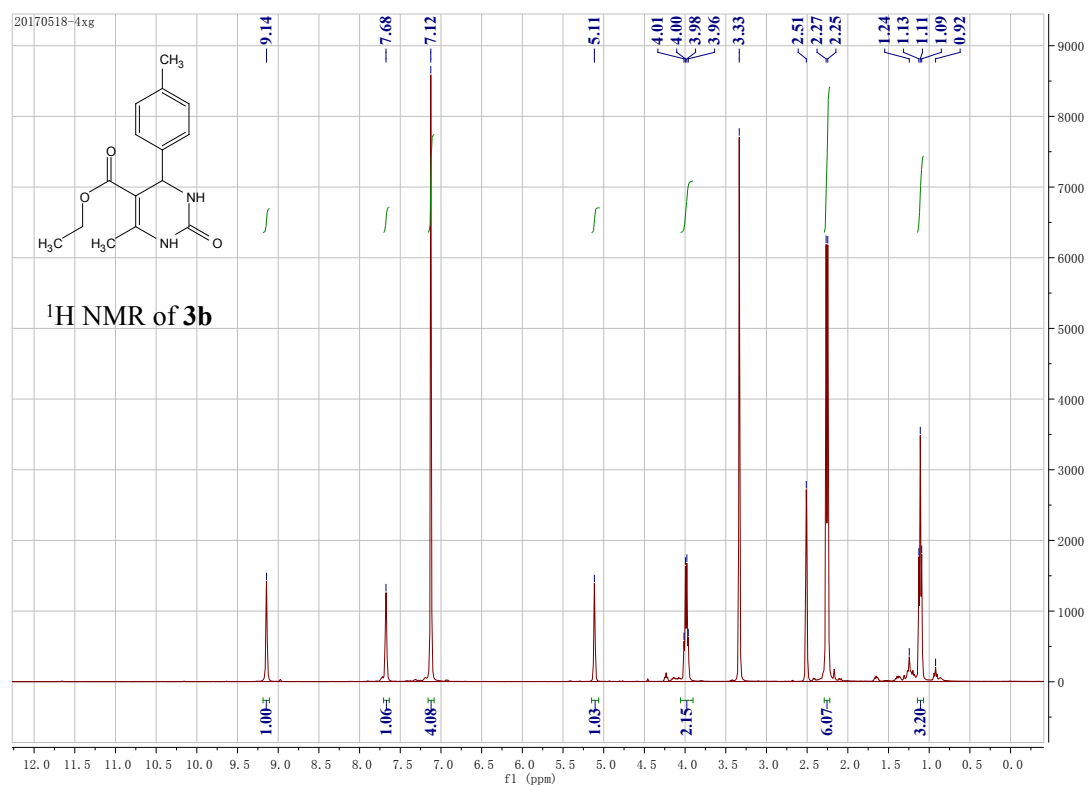
2-cyano-3-(4-nitrophenyl)-2-propenoic acid ethyl ester (2p)



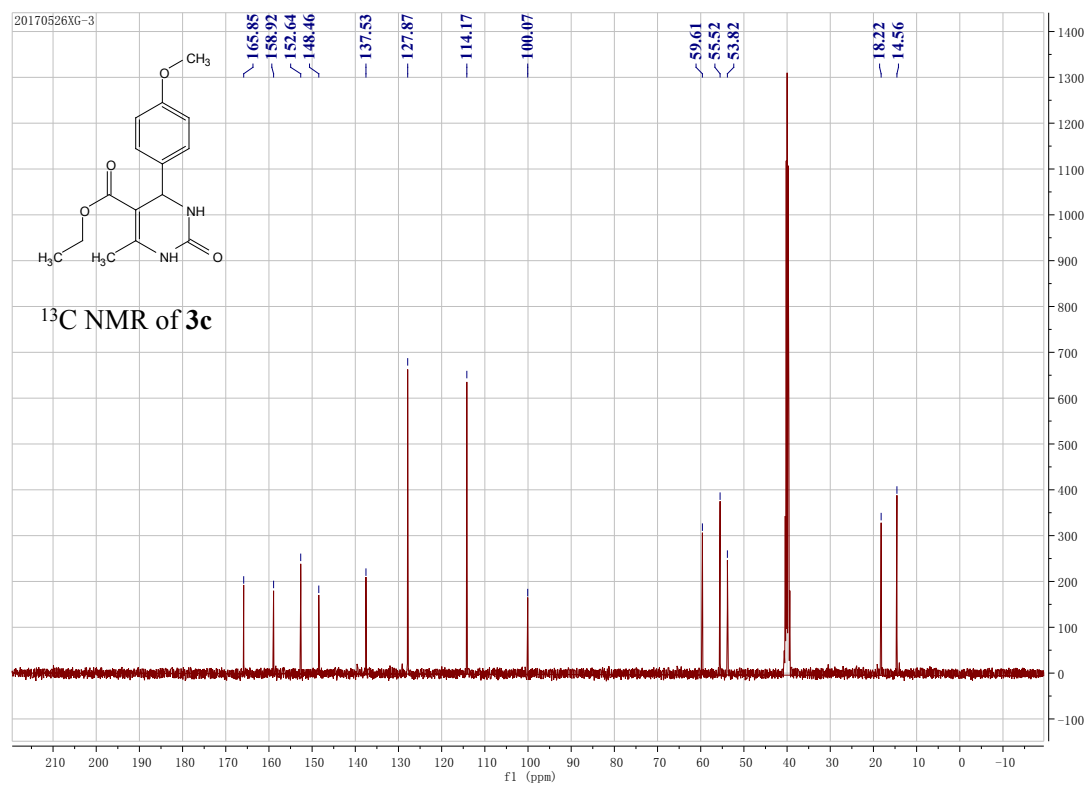
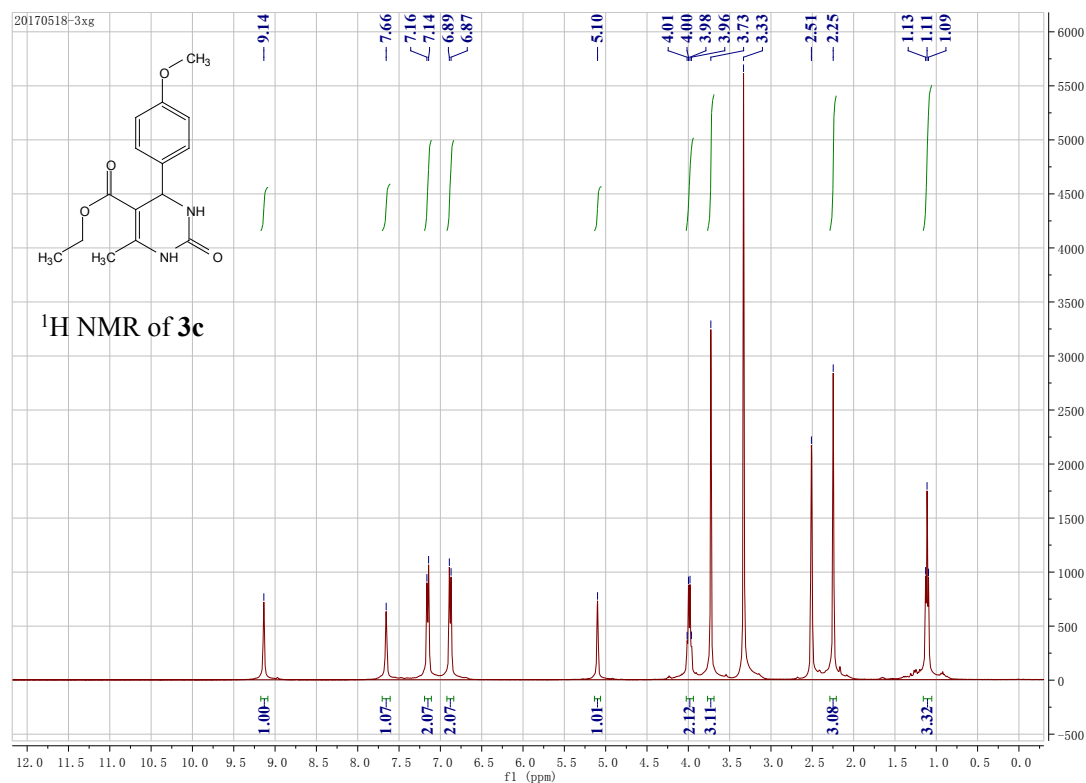
5-Ethoxycarbonyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (3a)



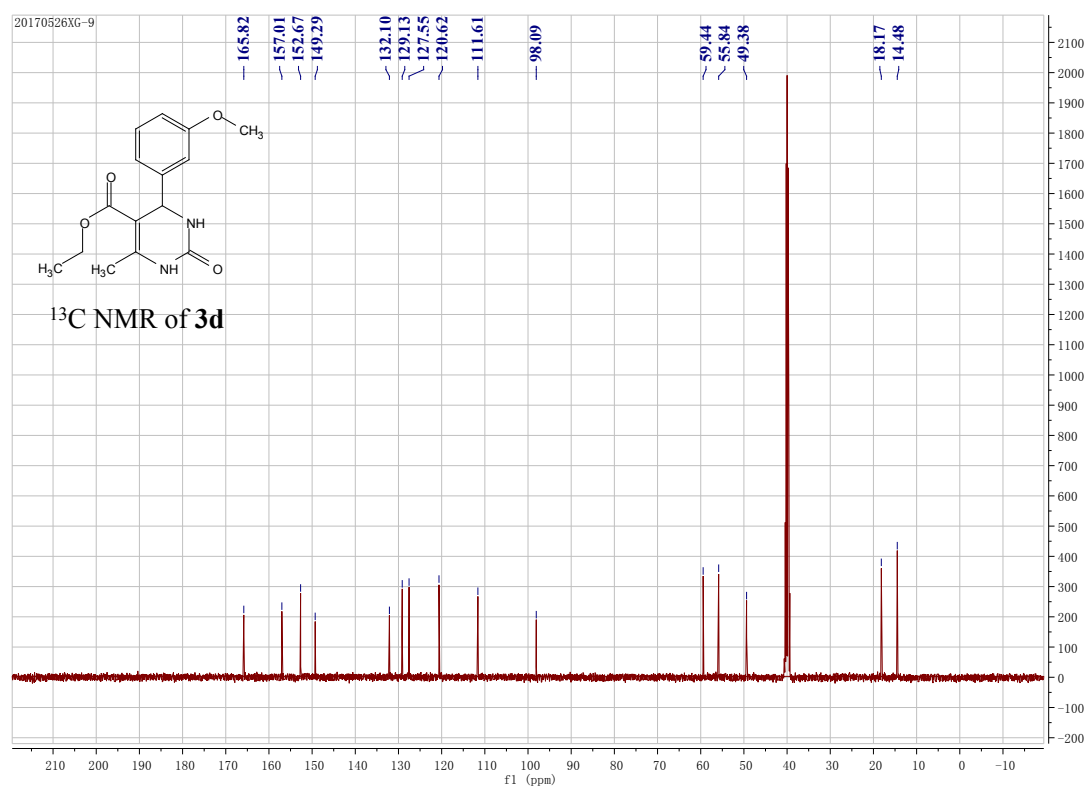
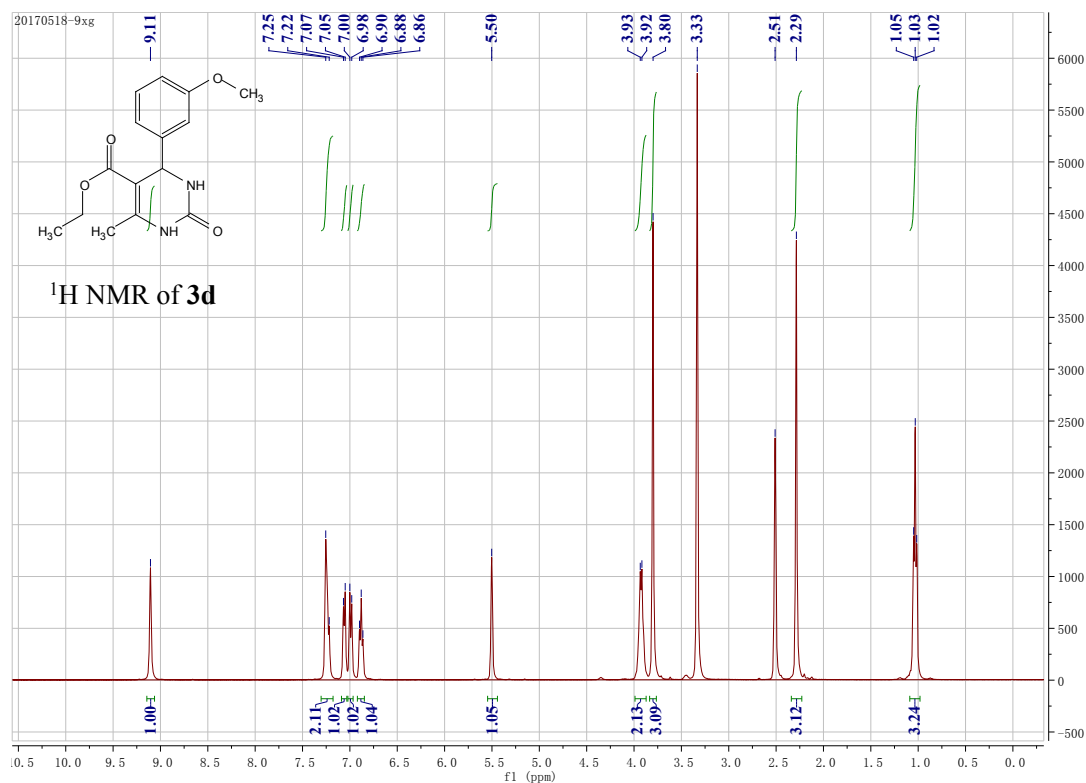
5-Ethoxycarbonyl-4-(4-methylphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3b)



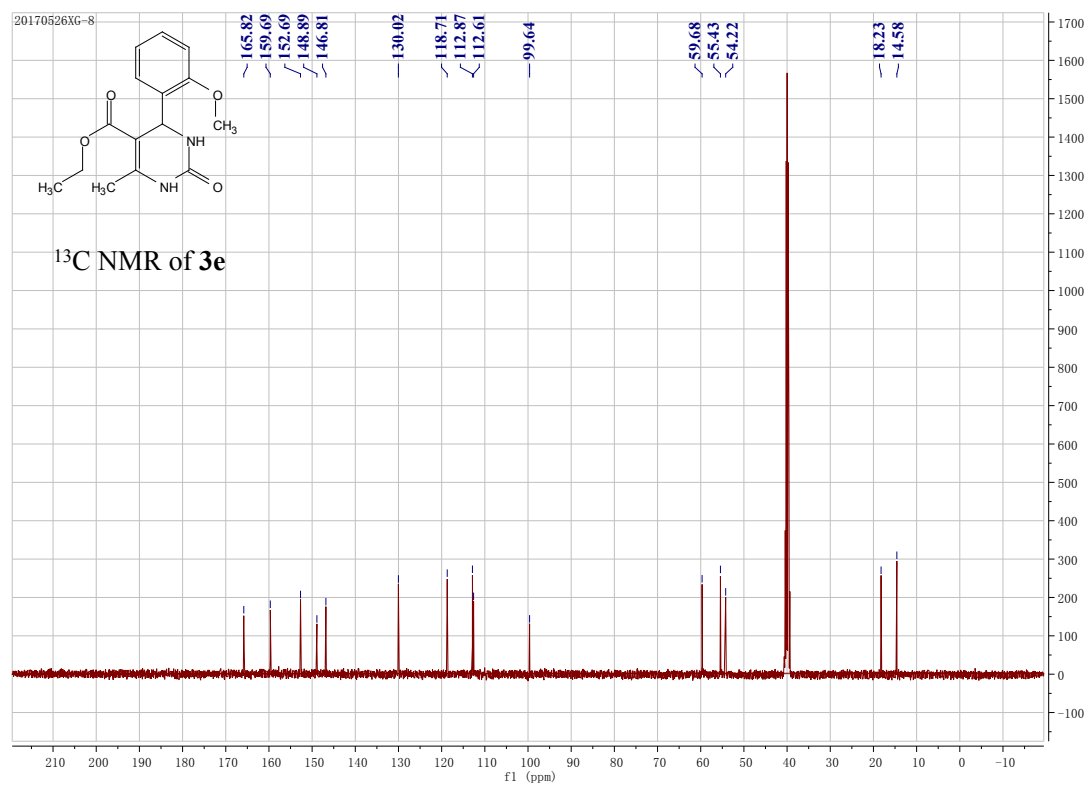
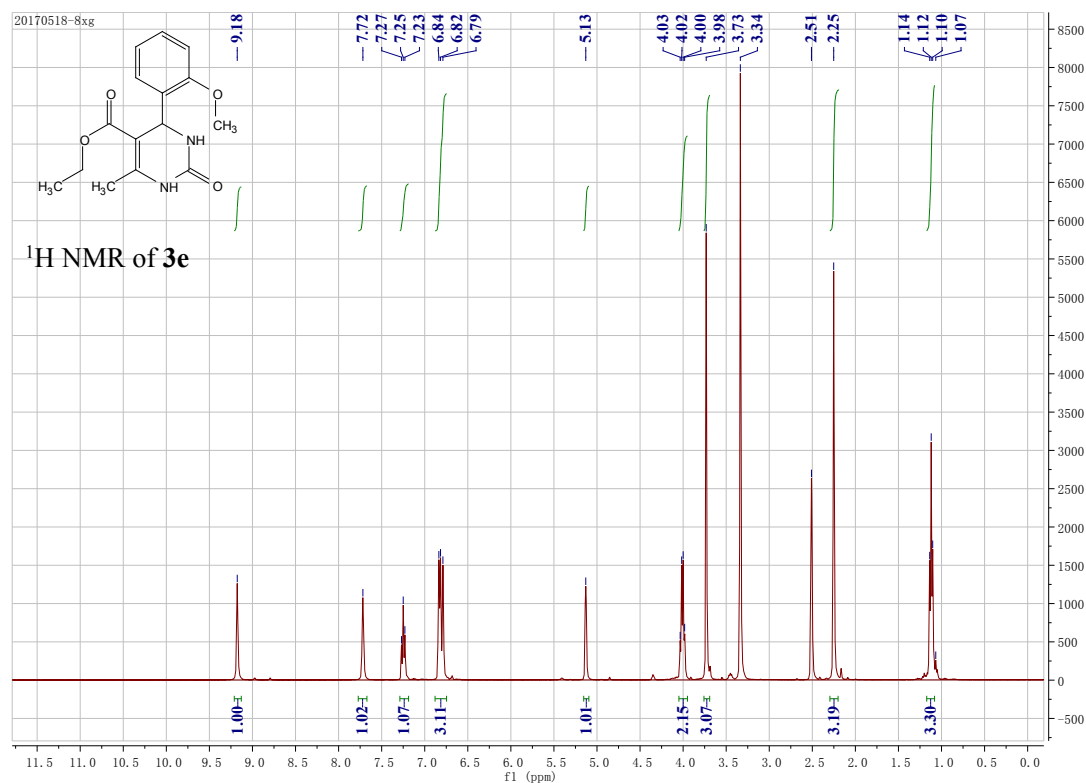
5-Ethoxycarbonyl-4-(4-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3c)



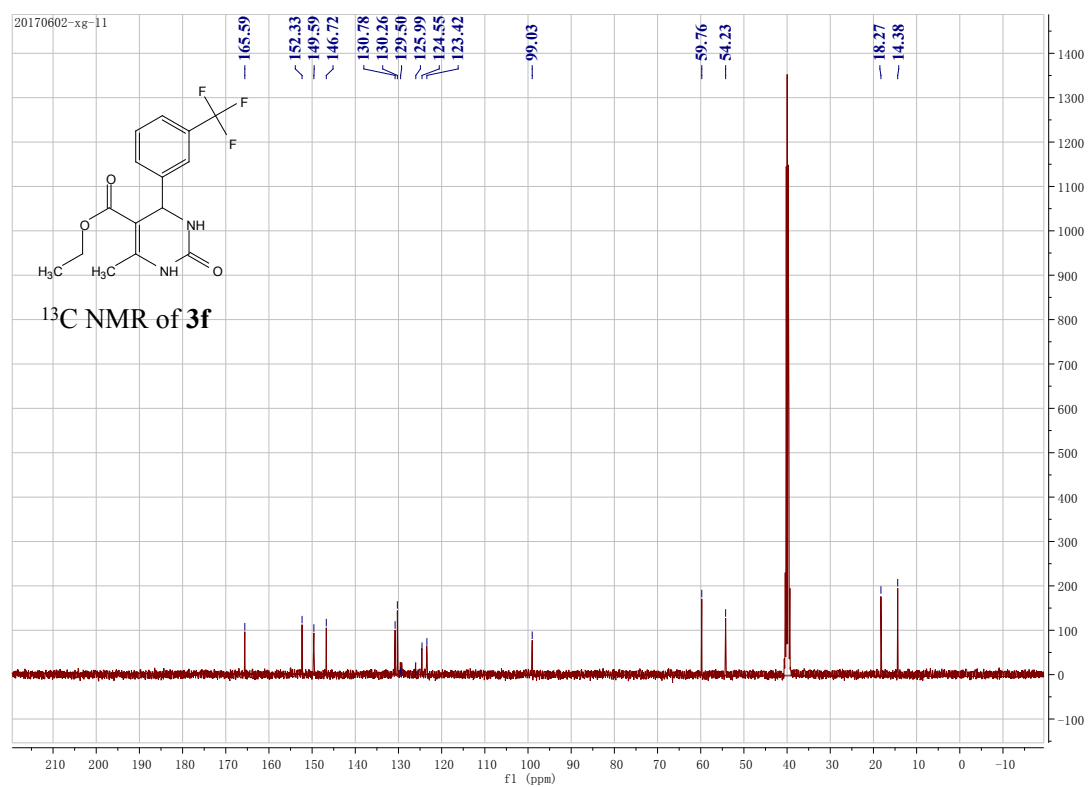
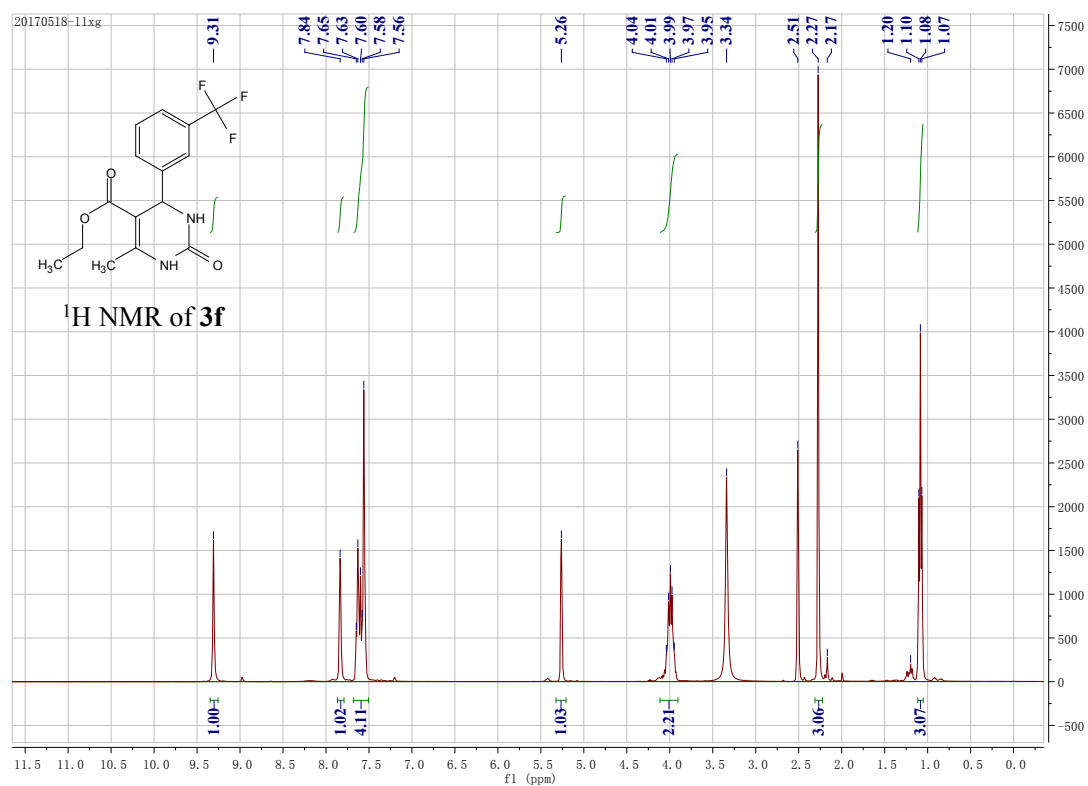
5-Ethoxycarbonyl-4-(3-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**3d**)



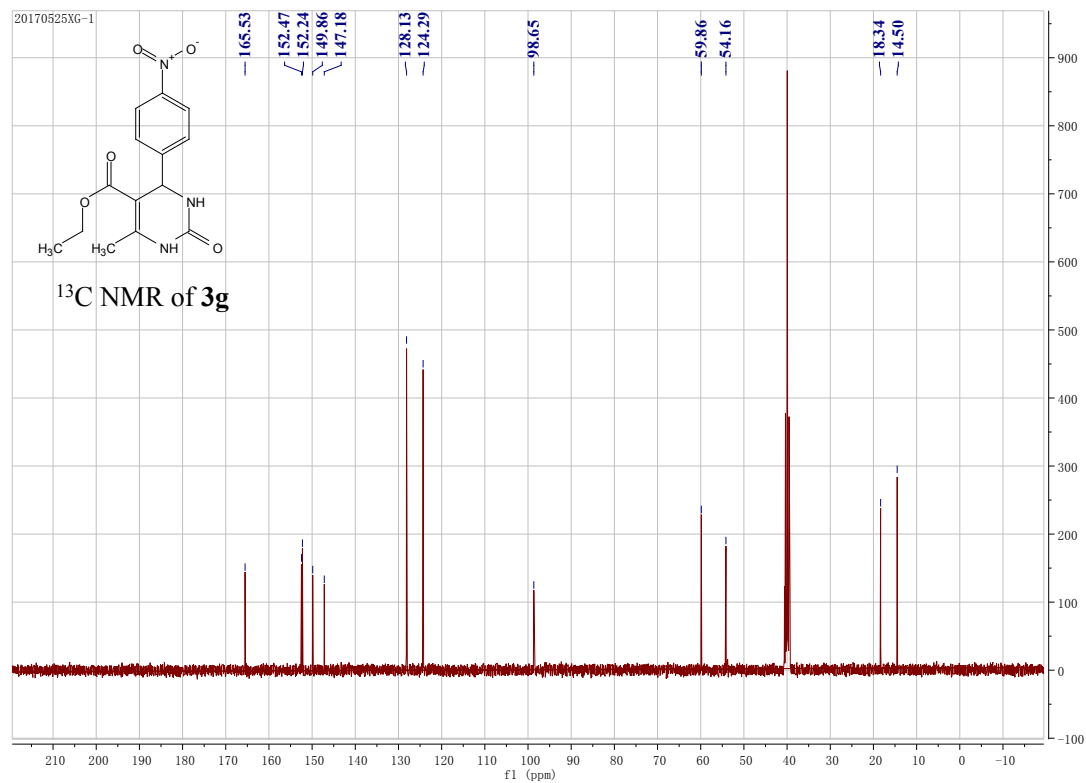
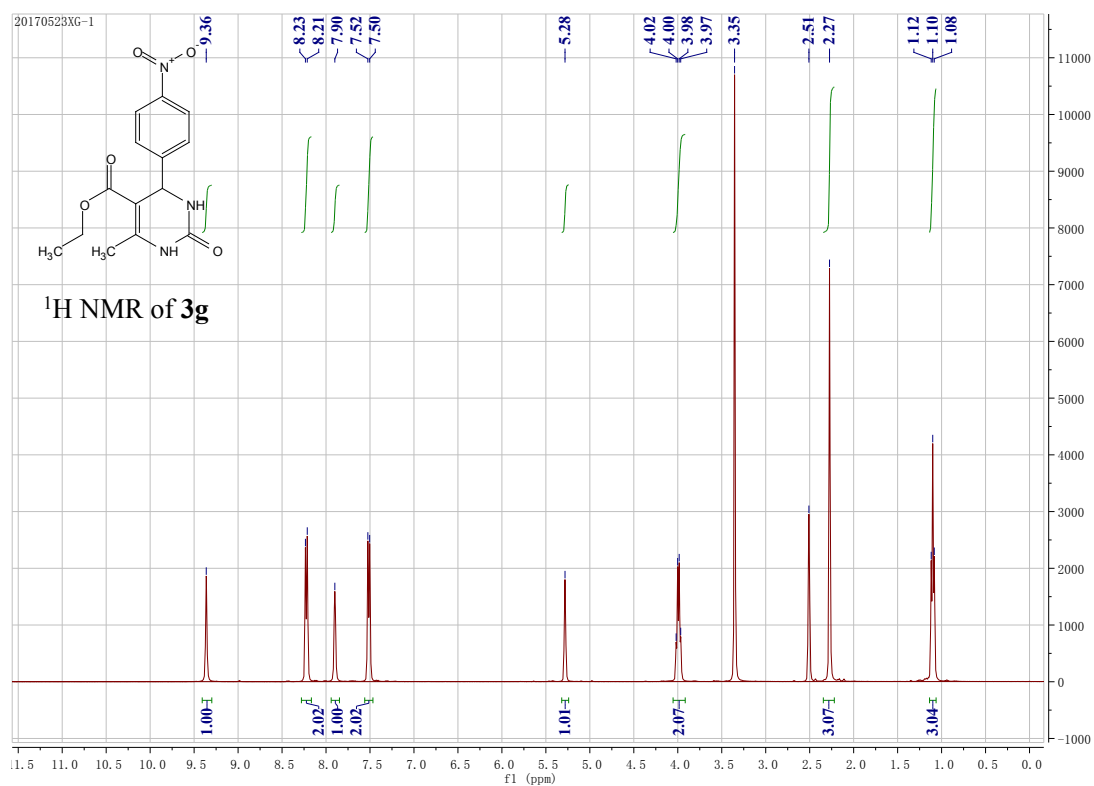
5-Ethoxycarbonyl-4-(2-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3e)



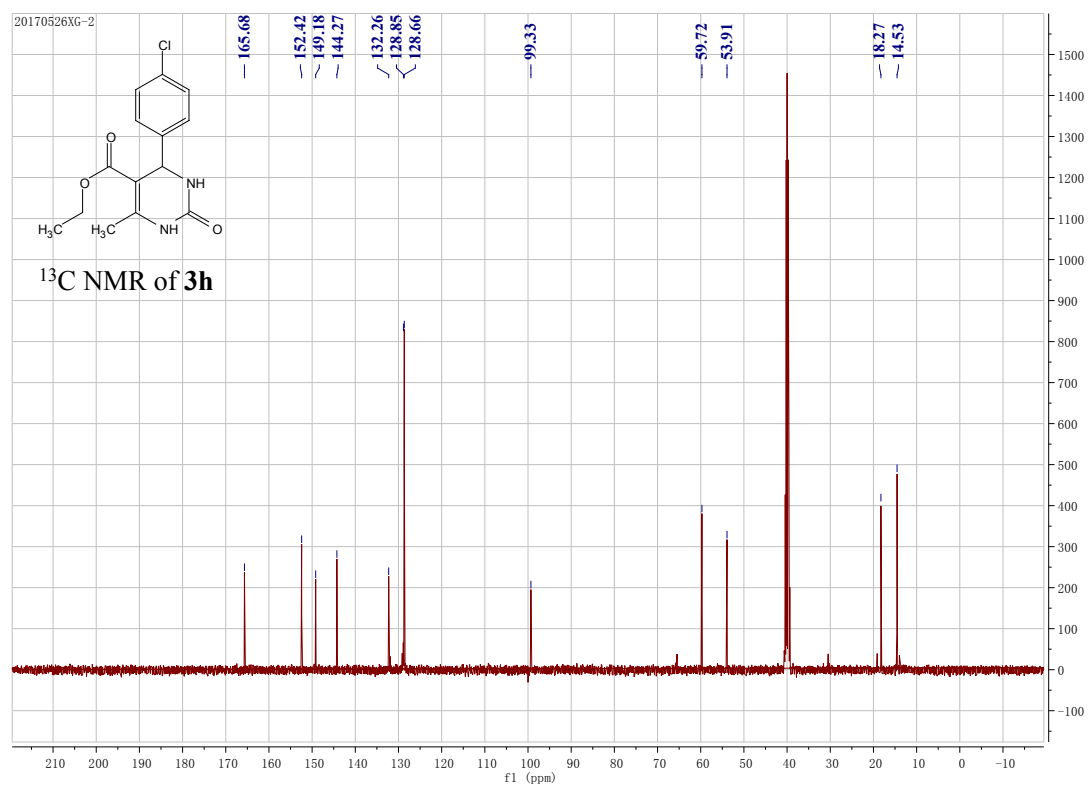
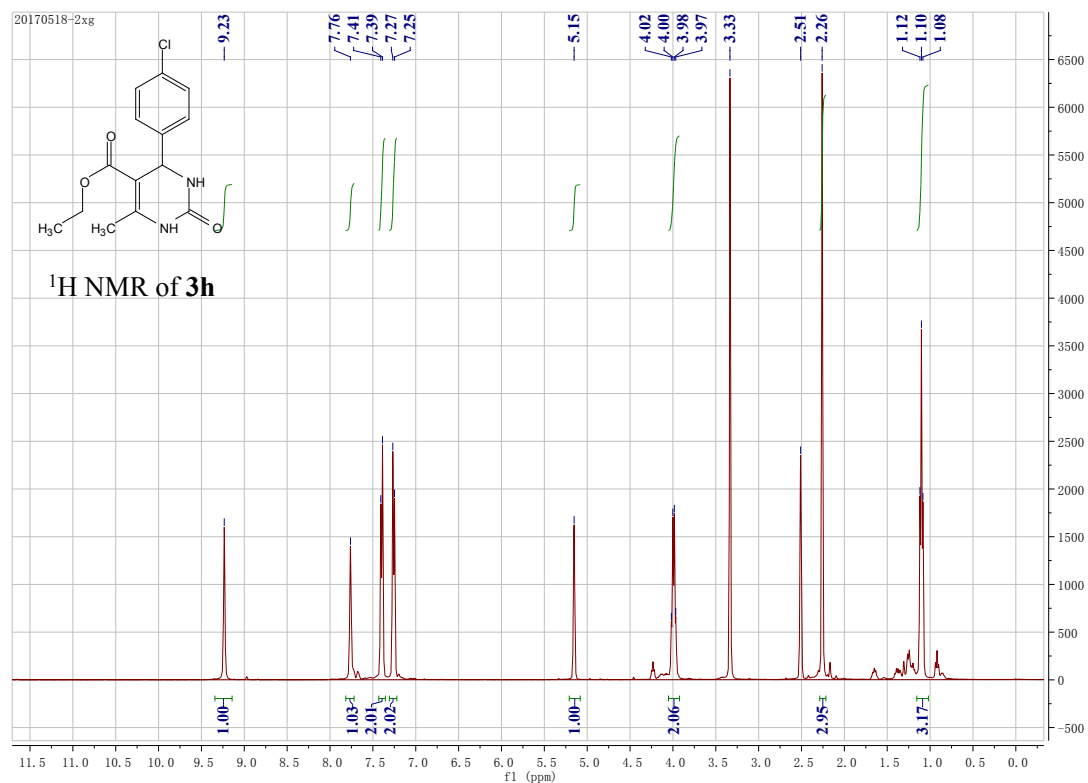
5-Ethoxycarbonyl-4-(3-trifluoromethylphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (3f)



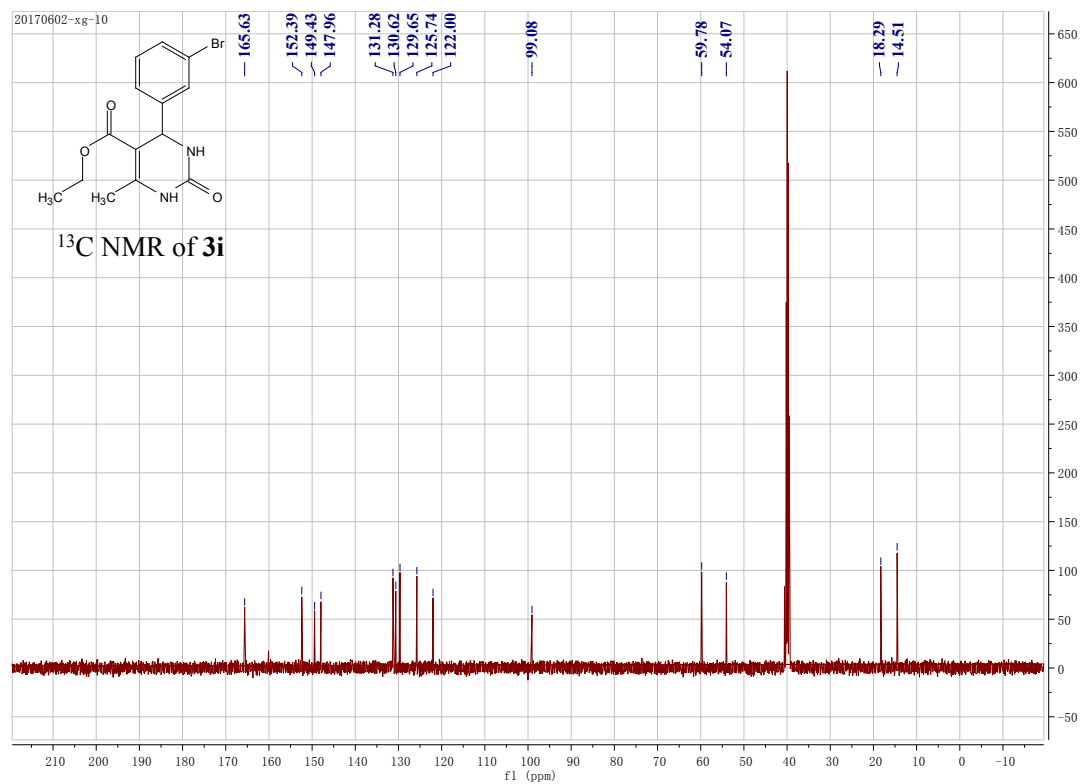
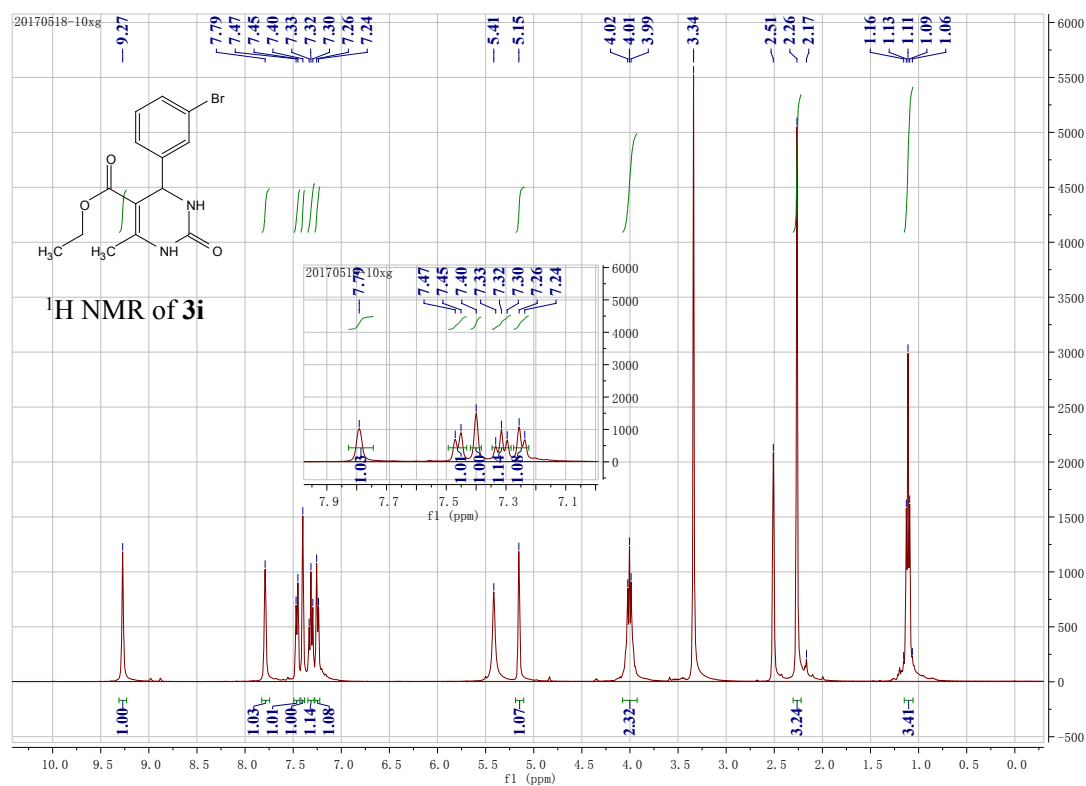
5-Ethoxycarbonyl-6-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (3g)



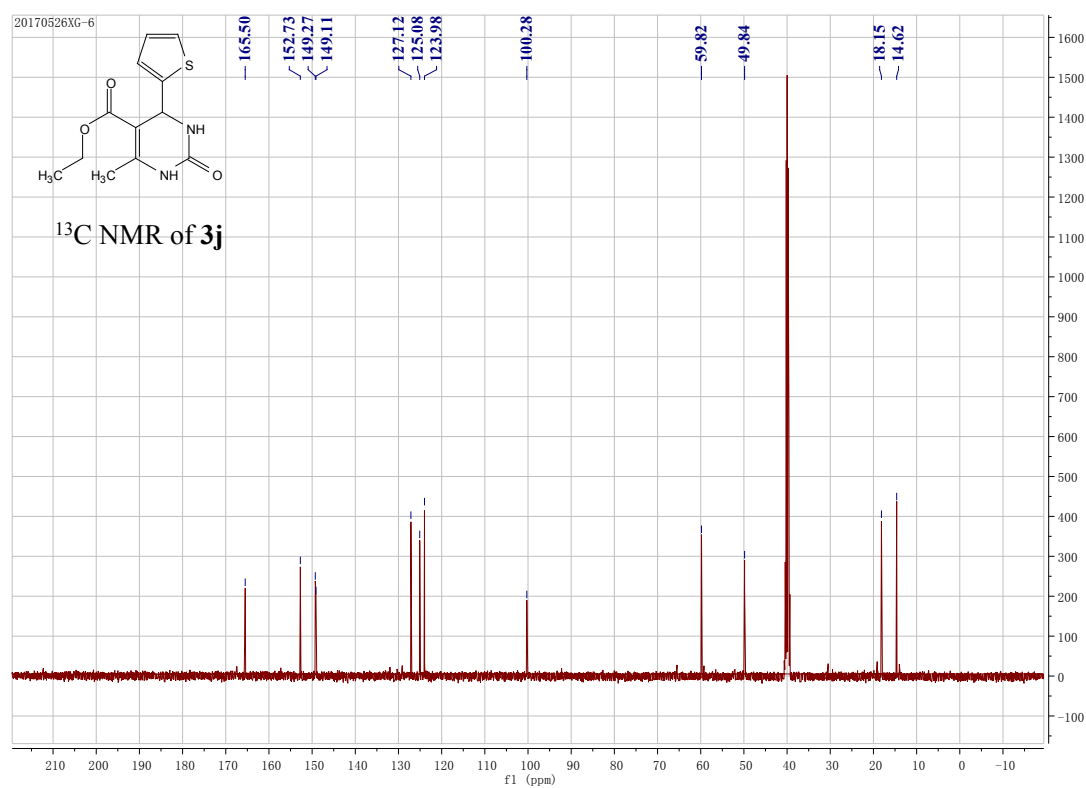
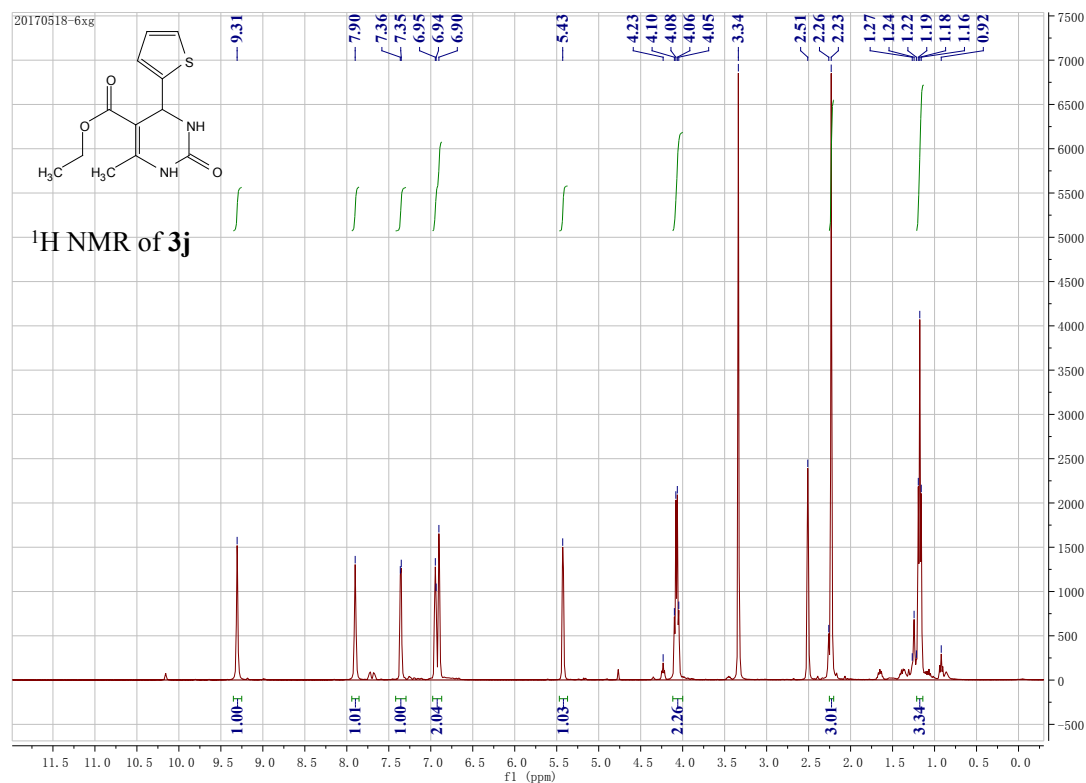
4-(4-Chlorophenyl)-5-ethoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**3h**)



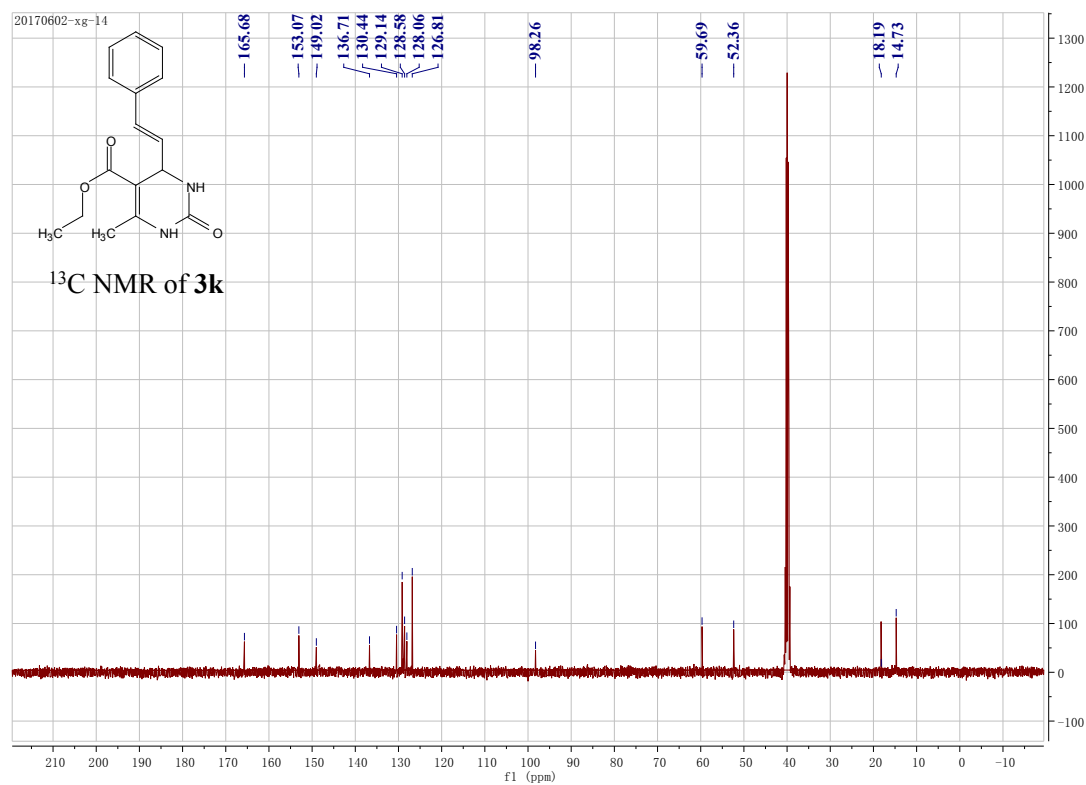
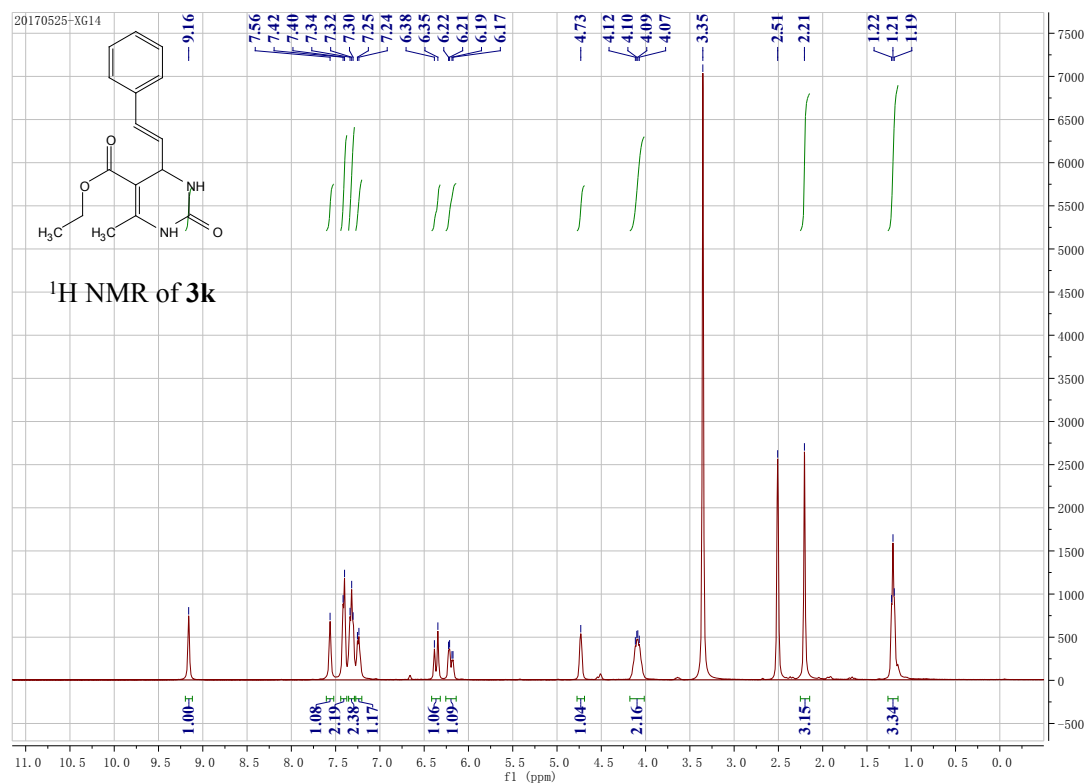
4-(3-Bromophenyl)-5-ethoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**3i**)



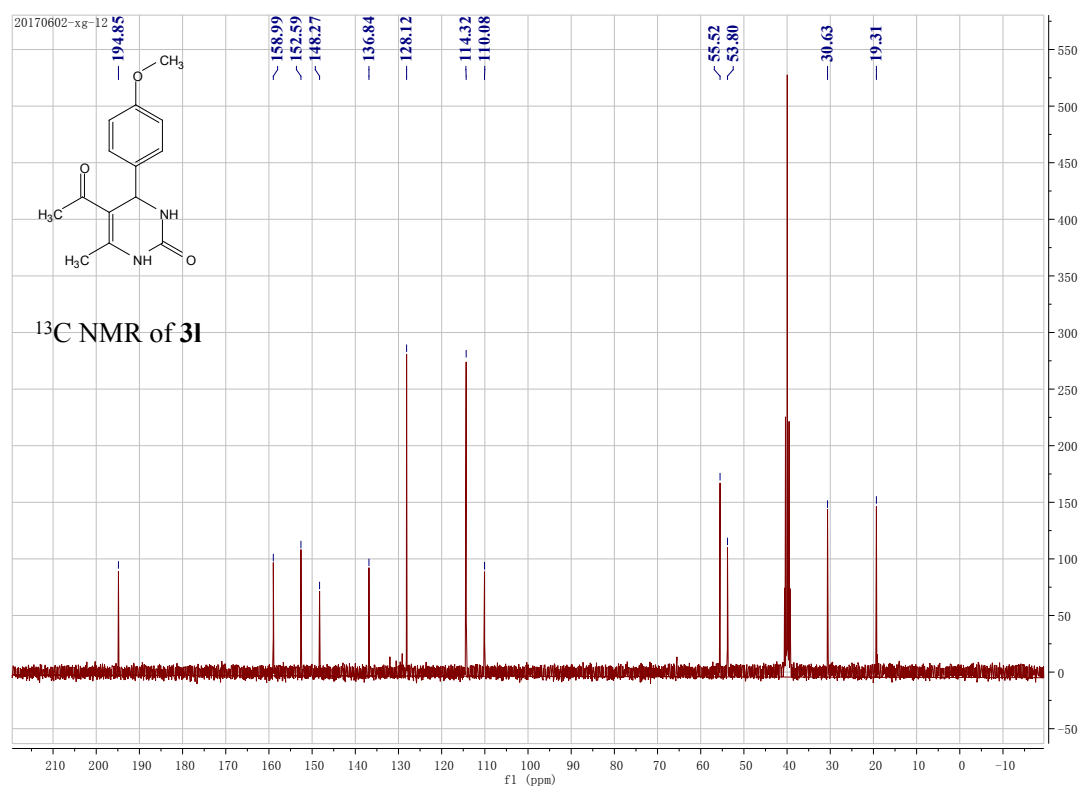
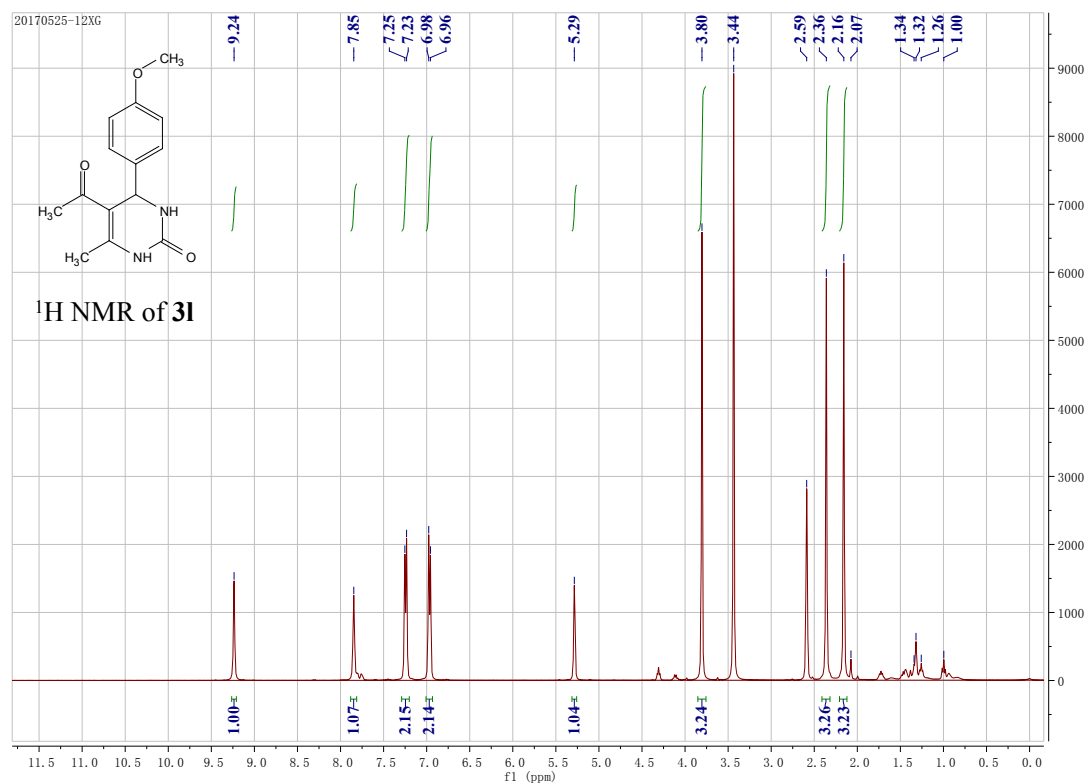
5-Ethoxycarbonyl-6-methyl-4-(2-thienyl)-3,4-dihydropyrimidin-2(1H)-one (3j)



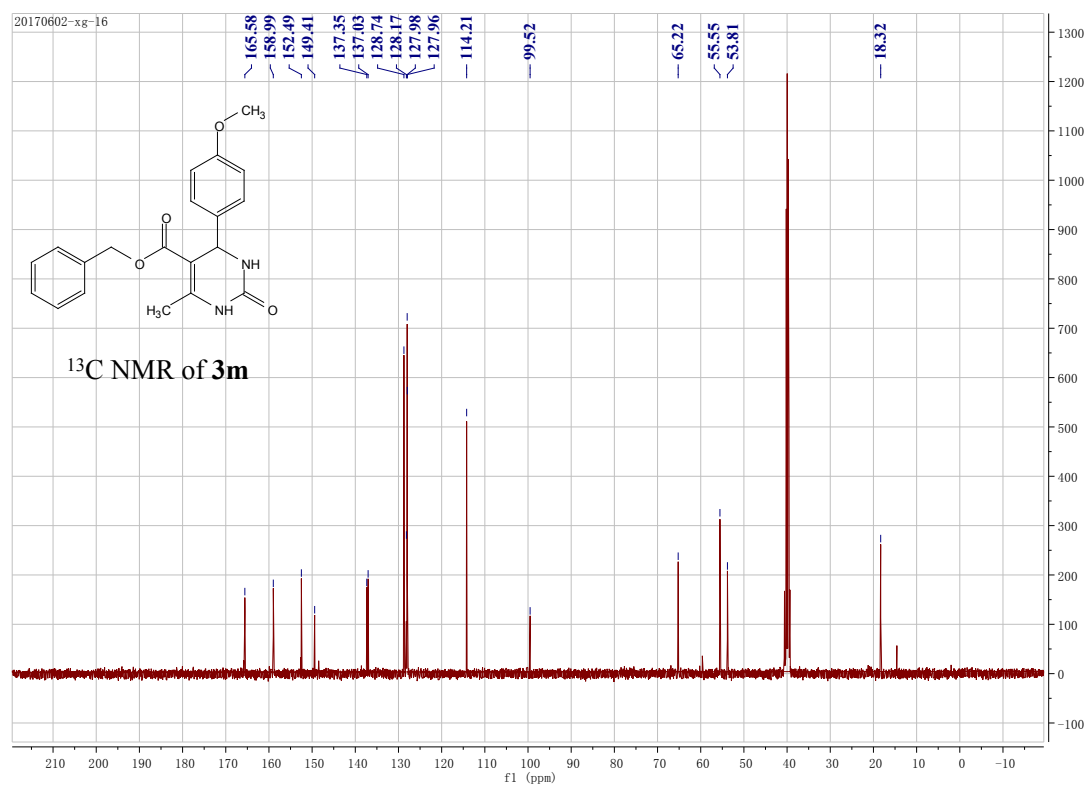
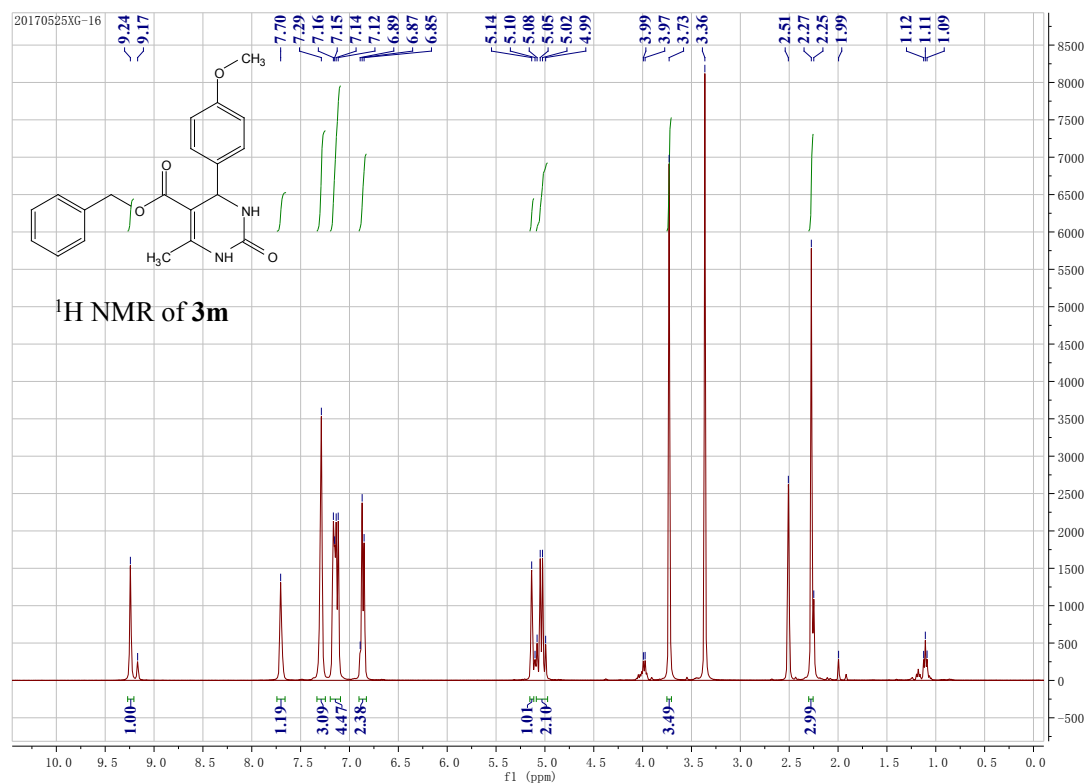
5-(ethoxycarbonyl)-6-methyl-4-styryl-3,4-dihydropyrimidin-2(1H)-one (**3k**)



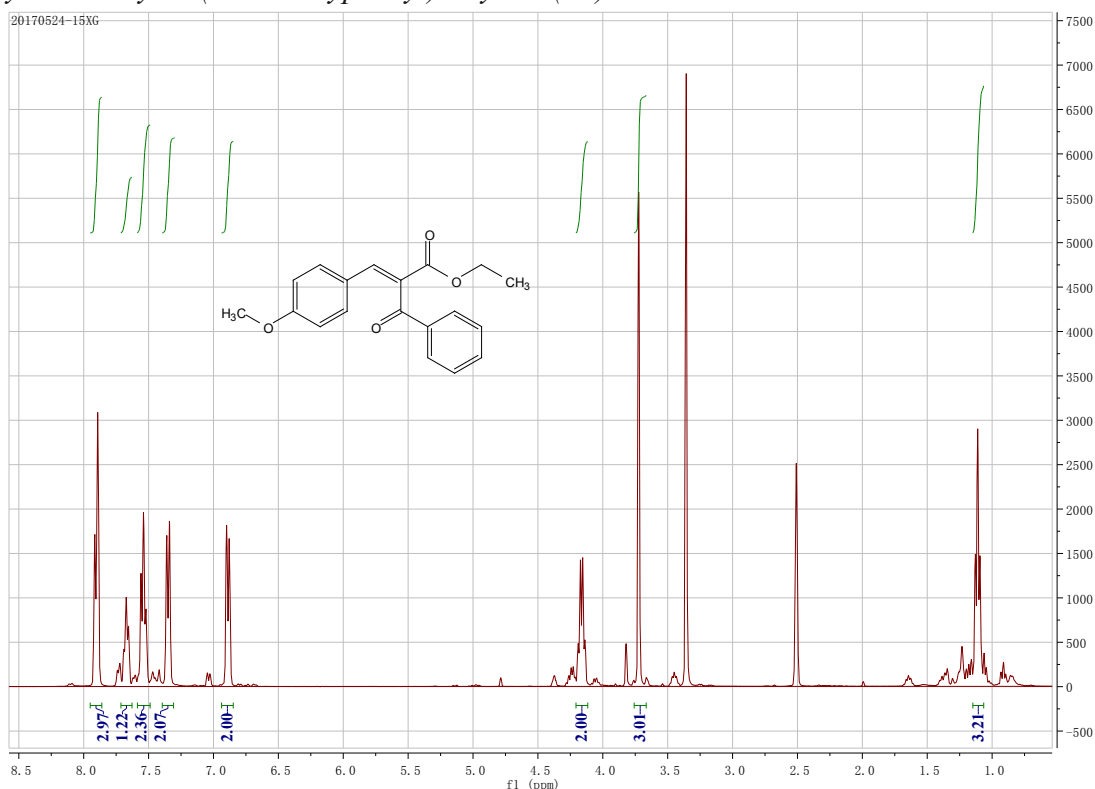
5-Acetyl-4-(4-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**3I**)



Benzyl 4-(4-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (3m)



Ethyl 2-benzoyl-3-(4-methoxyphenyl)acrylate (**3n**)



Notes and references

- [1] X. Li, B. Zhang, Y. Fang, W. Sun, Z. Qi, Y. Pei, S. Qi, P. Yuan, X. Luan, T. W. Goh, W. Huang, *Chem. Eur. J.* **2017**, *23*, 4266-4270.
- [2] Y. Luan, Y. Qi, H. Gao, R. S. Andriamantsoa, N. Zheng, G. Wang, *J. Mater. Chem. A* **2015**, *3*, 17320-17331.
- [3] J. B. M. de Resende Filho, G. P. Pires, J. M. G. de Oliveira Ferreira, E. E. S. Teotonio, J. A. Vale, *Catal. Lett.* **2017**, *147*, 167-180.
- [4] J. Xu, K. Shen, B. Xue, Y.-X. Li, *J. Mol. Catal. A: Chem.* **2013**, *372*, 105-113.
- [5] D.-Z. Xu, S. Shi, Y. Wang, *RSC Adv.* **2013**, *3*, 23075-23079.
- [6] R. Maleki, E. Kolvari, M. Salehi, N. Koukabi, *Appl. Organomet. Chem.* **2017**, 10.1002/aoc.3795.
- [7] E. M. Schneider, R. A. Raso, C. J. Hofer, M. Zeltner, R. D. Stettler, S. C. Hess, R. N. Grass, W. J. Stark, *J. Org. Chem.* **2014**, *79*, 10908-10915.
- [8] A. Dondoni, A. Massi, *Tetrahedron Lett.* **2001**, *42*, 7975-7978.
- [9] J. Mondal, T. Sen, A. Bhaumik, *Dalton Trans.* **2012**, *41*, 6173-6181.
- [10] Z.-J. Quan, Y.-X. Da, Z. Zhang, X.-C. Wang, *Catal. Commun.* **2009**, *10*, 1146-1148.
- [11] X.-L. Shi, H. Yang, M. Tao, W. Zhang, *RSC Adv.* **2013**, *3*, 3939-3945.
- [12] J. Javidi, M. Esmailpour, F. N. Dodeji, *RSC Adv.* **2015**, *5*, 308-315.
- [13] A. G. Khiratkar, P. N. Muskawar, P. R. Bhagat, *RSC Adv.* **2016**, *6*, 105087-105093.
- [14] J.-H. Wang, G.-M. Tang, S.-C. Yan, Y.-T. Wang, S.-J. Zhan, E. Zhang, Y. Sun, Y. Jiang, Y.-Z. Cui, *Appl. Organomet. Chem.* **2016**, *30*, 1009-1021.
- [15] B. Dam, A. K. Pal, A. Gupta, *Synth. Commun.* **2016**, *46*, 275-286.
- [16] M. Zeinali-Dastmalbaf, A. Davoodnia, M. M. Heravi, N. Tavakoli-Hoseini, A. Khojastehnezhad, H. A. Zamani, *Bull. Korean Chem. Soc.* **2011**, *32*, 656-658.