Supplementary Information for

ortho-Nb_2O_5 as a recyclable heterogeneous catalyst for the synthesis of 2-substituted benzimidazoles and quinazolinones in an aqueous medium *via* oxidative dehydrogenative coupling

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1. General information

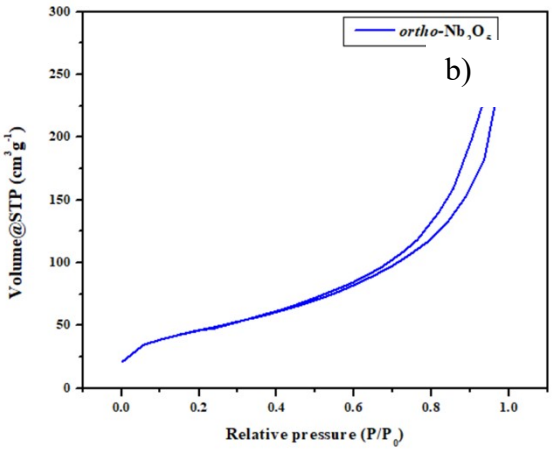
Unless otherwise mentioned, the purchased chemicals were used as such without further purification. All the solvents were purified and dried according to the standard procedures.[1] The catalysts were characterized by scanning electron microscopy (SEM) and powder X-ray diffraction (PXRD) analyses. PXRD analysis was done using a PAN Analytical B. V. Empyrean instrument equipped with Cu Kα radiation (40 kV, 40 mA). The measurements were done over a range of 5-80° at a scan rate of 0.5° per minute. SEM analysis was done using a Carl Zeiss EVO 18 instrument. The surface area was determined by BET analysis. BET analysis was done using a Quantachrome Nova 2200E BET surface area analyzer. UV-Vis spectra were recorded using a Shimadzu UV-2600 instrument. ^{1}H and ^{13}C NMR spectra were recorded using a Bruker 500 MHz instrument in DMSO-d_6 or $CDCl_3$ solvent, with tetramethylsilane (TMS) as an internal standard.

2. Procedure for the preparation of deformed orthorhombic Nb_2O_5 (*ortho*-Nb_2O_5)[2]

ortho-Nb_2O_5 was prepared by a conventional hydrothermal treatment of an aqueous ammonium niobium oxalate (ANO) solution. ANO (6 mmol) was completely dissolved in 40 mL of deionized (DI) water. The solution was transferred to a Teflon-linked stainless-steel autoclave (60 mL) and then heated at 175 °C for 3 days. The resulting precipitate was isolated by filtration, washed repeatedly with DI water, and dried at 80 °C overnight. After calcination of the dry powder at 400 °C for 4 h, *ortho*-Nb_2O_5 was obtained with a yield of 95%. The nature of the catalyst was studied by PXRD, SEM, and BET isotherm analyses.

3. BET isotherms of the catalysts

a)



b)

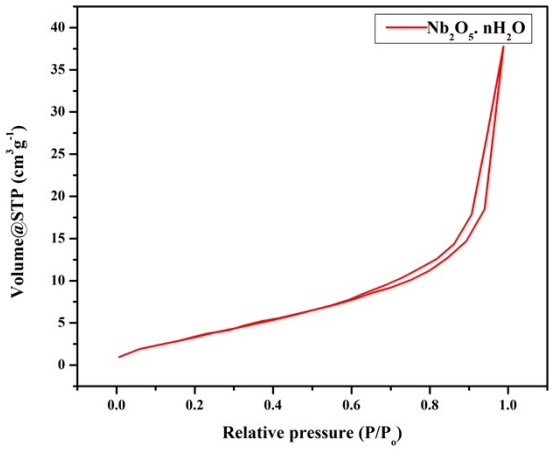


Fig. S1. BET isotherms of a) *ortho*-Nb_2O_5, and b) commercially sourced $Nb_2O_5 \cdot nH_2O$.

BJH desorption summary of *ortho*-Nb_2O_5

Surface area = 183.624 m^2/g

Pore volume = 0.430 cm^3/g

Pore diameter Dv(d) = 3.513 nm

BJH desorption summary of commercially sourced $Nb_2O_5 \cdot nH_2O$

Surface area = 23.622 m^2/g

Pore volume = 0.026 cm^3/g

Pore diameter Dv(d) = 2.368 nm

4. Procedure for the synthesis of 2-substituted benzimidazole derivatives

The Ace pressure tube was charged with 0.25 mmol of aryl diamine, 0.5 mmol of benzyl alcohol, 0.375 mmol of KOH, 15 mol% of catalyst, and 0.5 mL of water, and the mixture was allowed to stir at 120 °C. After 24 h, the organic components were extracted with ethyl acetate (3 × 5 mL), dried with anhydrous sodium sulfate, and filtered. The crude mixture was reduced under *vacuum* and purified by silica gel column chromatography using 15-50% ethyl acetate-hexane mixture as eluent. The formation of products was confirmed using ^1H and ^{13}C NMR spectroscopic techniques.

5. Procedure for the synthesis of 2-substituted quinazolinone derivatives

The above-mentioned procedure for the synthesis of 2-substituted benzimidazole derivatives was extended to the synthesis of quinazolinone derivatives by using 2-amino benzamides instead of aryl diamines.

6. Recyclability test

After the completion of the reaction, methanol was added to the reaction medium, and the mixture was centrifuged to separate the catalyst. Further, the catalyst was washed multiple times with methanol and water. Then, it was dried in a *vacuum* overnight and used for further cycles.

7. Detection of H_2O_2 during oxidative dehydrogenative coupling by UV-Vis spectroscopy[3]

The formation of H_2O_2 during oxidative dehydrogenative coupling was detected spectrophotometrically following the development of a characteristic absorption band for I_3^- at 354 nm (**Fig. S55**).[3] The reaction between *o*-phenylenediamine (1 mmol) and benzyl alcohol (2 mmol) was carried out under an aerial atmosphere, according to the procedure mentioned above. After 20 h of the reaction, the whole solution was extracted with dichloromethane (3 × 5 mL). The aqueous layer was then acidified with H_2SO_4 to pH 2 to stop further oxidation, and

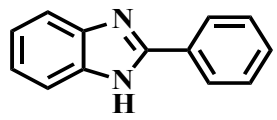
in the acidic medium, the superoxide radicals would undergo a disproportionation reaction to obtain hydrogen peroxide. After that, 1.0 mL of a 10% solution of KI and a few drops of 3% solution of ammonium molybdate were added. Hydrogen peroxide oxidizes I^- to I_2, which then reacts with excess I^- to form I_3^- according to the following chemical reactions.

$H_2O_2 + 2I^- + 2H^+ \rightarrow 2H_2O + I_2$;

$I_{2(aq)} + I^- \rightarrow I_3^-$

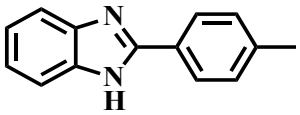
8. ^{1}H and ^{13}C NMR spectral data of 2-substituted benzimidazoles (3a-3n)

2-phenyl-1*H*-benzo[*d*]imidazole (3a)[4]



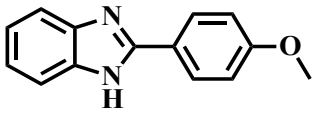
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.95 (s, 1H), 8.21 (d, J = 7.2 Hz, 2H), 7.62 (s, 1H), 7.56 (t, J = 7.5 Hz, 3H), 7.52–7.47 (m, 1H), 7.22 (dd, J = 6.0, 3.1 Hz, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 151.25, 143.78, 134.98, 130.15, 129.87, 128.97, 126.45, 122.58, 121.72, 118.88, 111.33.

2-(*p*-tolyl)-1*H*-benzo[*d*]imidazole (3b)[5]



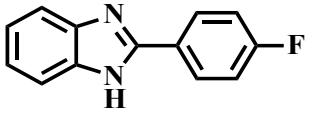
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.83 (s, 1H), 8.08 (d, J = 8.1 Hz, 2H), 7.65 (d, J = 7.3 Hz, 1H), 7.52 (d, J = 7.2 Hz, 1H), 7.37 (d, J = 8.1 Hz, 2H), 7.20 (t, J = 8.0 Hz, 2H), 2.39 (s, 3H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 151.85, 144.30, 140.03, 135.42, 129.98, 127.93, 126.86, 122.80, 122.03, 119.18, 111.66, 21.44.

2-(4-methoxyphenyl)-1*H*-benzo[*d*]imidazole (3c)[5]



^{1}H NMR (500 MHz, DMSO-d_6): δ 12.78 (s, 1H), 8.16 (d, J = 8.9 Hz, 2H), 7.59 (s, 2H), 7.21–7.16 (m, 2H), 7.12 (d, J = 8.9 Hz, 2H), 3.84 (s, 3H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 161.09, 151.87, 128.51, 123.21, 122.24, 114.83, 55.76.

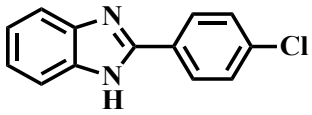
2-(4-fluorophenyl)-1*H*-benzo[*d*]imidazole (3d)[6]



^{1}H NMR (500 MHz, DMSO-d_6): δ 12.93 (s, 1H), 8.34–8.15 (m, 2H), 7.66 (d, J = 7.5 Hz, 1H), 7.53 (d, J = 7.5 Hz, 1H), 7.38–7.4 (m, 2H), 7.24–7.16 (m, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 163.63 ($^1J_{C–F}$ = 245.3 Hz), 150.88, 144.33, 135.58, 129.21 ($^3J_{C–F}$ = 9.1 Hz), 127.33 ($^4J_{C–F}$ = 4.2 Hz),

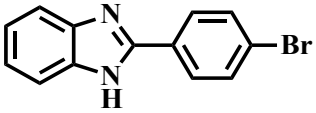
123.10, 122.26, 119.41, 116.55 ($^2J_{C-F}$ = 22.7 Hz), 111.85.

2-(4-chlorophenyl)-1*H*-benzo[*d*]imidazole (3e)[6]



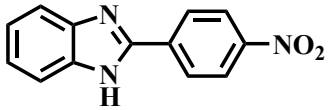
^{1}H NMR (500 MHz, DMSO-d_6): δ 13.00 (s, 1H), 8.22–8.16 (m, 2H), 7.68–7.63 (m, 2H), 7.62 (m, 2H), 7.26–7.20 (m, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 150.63, 144.23, 135.50, 134.96, 129.53, 128.61, 123.25, 122.31, 119.44, 111.88.

2-(4-bromophenyl)-1*H*-benzo[*d*]imidazole (3f)[7]



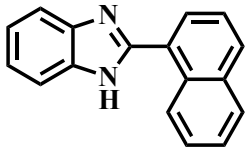
^{1}H NMR (500 MHz, DMSO-d_6): δ 13.00 (s, 1H), 8.13 (d, J = 8.5 Hz, 2H), 7.77 (d, J = 8.5 Hz, 2H), 7.68 (d, J = 7.5 Hz, 1H), 7.55 (d, J = 7.3 Hz, 1H), 7.27–7.17 (m, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 150.70, 144.22, 135.50, 132.45, 129.87, 128.83, 123.72, 123.27, 122.33, 119.45, 111.90.

2-(4-nitrophenyl)-1*H*-benzo[*d*]imidazole (3g)[8]



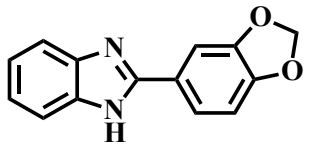
^{1}H NMR (500 MHz, DMSO-d_6): δ 13.30 (s, 1H), 8.46–8.39 (m, 4H), 7.74 (d, J = 7.9 Hz, 1H), 7.60 (d, J = 7.9 Hz, 1H), 7.28 (dt, J = 15.1, 7.0 Hz, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 149.48, 148.29, 144.30, 136.51, 135.71, 127.87, 127.49, 124.79, 124.08, 123.76, 122.81, 119.93, 112.29.

2-(naphthalen-1-yl)-1*H*-benzo[*d*]imidazole (3h)[5]



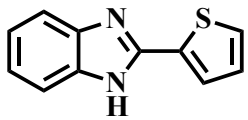
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.94 (s, 1H), 9.13 (d, J = 8.2 Hz, 1H), 8.12 (d, J = 7.9 Hz, 1H), 8.05 (dd, J = 11.7, 7.6 Hz, 2H), 7.80–7.59 (m, 5H), 7.28 (s, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 151.83, 134.10, 130.99, 130.62, 128.87, 128.33, 128.01, 127.54, 126.82, 125.75, 123.10, 122.05, 119.54, 111.75.

2-(benzo[*d*][1,3]dioxol-5-yl)-1*H*-benzo[*d*]imidazole (3i)[9]



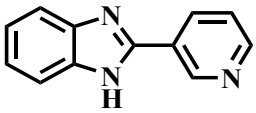
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.76 (s, 1H), 7.76–7.69 (m, 2H), 7.57 (s, 2H), 7.21–7.16 (m, 2H), 7.10 (d, J = 8.1 Hz, 1H), 6.13 (s, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 151.64, 149.21, 148.43, 124.85, 122.44, 121.45, 109.28, 107.06, 102.13.

2-(thiophen-2-yl)-1*H*-benzo[*d*]imidazole (3j)[5]



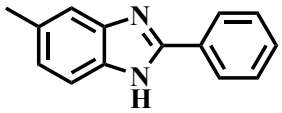
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.96 (s, 1H), 7.84 (dd, J = 3.7, 1.1 Hz, 1H), 7.73 (dd, J = 5.0, 1.1 Hz, 1H), 7.62 (d, J = 7.8 Hz, 1H), 7.51 (d, J = 7.6 Hz, 1H), 7.17–7.25 (m, 3H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 147.50, 144.08, 135.16, 134.19, 129.21, 128.75, 127.14, 123.09, 122.21, 119.01, 111.57.

2-(pyridin-3-yl)-1H-benzo[*d*]imidazole (3k)[10]



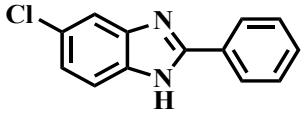
^{1}H NMR (500 MHz, DMSO-d_6): δ 13.11 (s, 1H), 9.36 (d, J = 1.5 Hz, 1H), 8.69 (dd, J = 4.8, 1.6 Hz, 1H), 8.52–8.49 (m, 1H), 7.72 (d, J = 7.5 Hz, 1H), 7.58-7.61 (m, 2H), 7.26 (t, J = 9.5 Hz, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 150.50, 148.85, 147.51, 143.67, 135.00, 133.75, 126.16, 124.01, 122.95, 121.99, 119.12, 111.53.

5-methyl-2-phenyl-1*H*-benzo[*d*]imidazole (3m)[11]



^{1}H NMR (500 MHz, DMSO-d_6): δ 12.76 (s, 1H), 8.17 (d, J = 7.2 Hz, 2H), 7.57–7.47 (m, 4H), 7.38 (s, 1H), 7.03 (d, J = 8.1 Hz, 1H), 2.44 (s, 3H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 151.38, 130.80, 130.11, 129.37, 126.78, 123.99, 21.81.

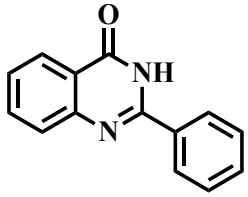
5-chloro-2-phenyl-1H-benzo[d]imidazole (3n)[5]



^{1}H NMR (500 MHz, DMSO-d_6): δ 13.08 (s, 1H), 8.21–8.10 (m, 2H), 7.81–7.40 (m, 5H), 7.25–7.13 (m, 1H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 153.15, 130.71, 130.21, 129.52, 127.10, 122.85.

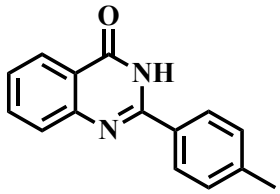
9. ^{1}H and ^{13}C NMR spectral data of 2-substituted quinazolinone derivatives (5a-5n)

2-phenylquinazolin-4(3*H*)-one (5a)[12]



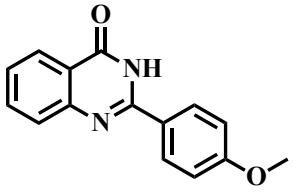
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.55 (s, 1H), 8.22–8.15 (m, 3H), 7.85 (t, J = 6.9 Hz, 1H), 7.75 (d, J = 7.9 Hz, 1H), 7.63–7.50 (m, 4H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.71, 152.78, 149.22, 135.08, 133.20, 131.87, 129.08, 128.24, 127.99, 127.07, 126.33, 121.46.

2-(*p*-tolyl)quinazolin-4(3*H*)-one (5b)[12]



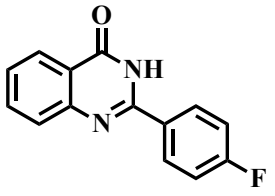
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.47 (s, 1H), 8.16 (d, J = 6.9 Hz, 1H), 8.11 (d, J = 8.2 Hz, 2H), 7.86–7.81 (m, 1H), 7.74 (d, J = 7.6 Hz, 1H), 7.52 (t, J = 7.0 Hz, 1H), 7.37 (d, J = 7.9 Hz, 2H), 2.40 (s, 3H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.72, 152.70, 149.31, 141.93, 135.06, 130.37, 129.67, 128.16, 127.90, 126.88, 126.31, 121.38, 21.47.

2-(4-methoxyphenyl)quinazolin-4(3*H*)-one (5c)[12]



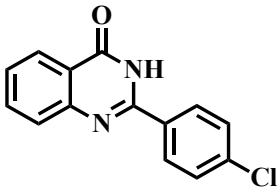
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.42 (s, 1H), 8.20 (d, J = 8.7 Hz, 2H), 8.14 (d, J = 7.3 Hz, 1H), 7.82 (t, J = 7.0 Hz, 1H), 7.71 (d, J = 8.1 Hz, 1H), 7.49 (t, J = 7.3 Hz, 1H), 7.09 (d, J = 8.9 Hz, 2H), 3.85 (s, 3H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.78, 162.34, 152.33, 149.41, 135.01, 129.93, 127.76, 126.59, 126.30, 125.27, 121.17, 114.46, 55.93.

2-(4-fluorophenyl)quinazolin-4(3*H*)-one (5d)[12]



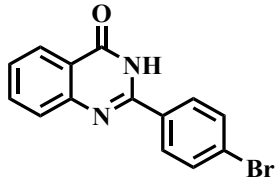
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.58 (s, 1H), 8.30–8.22 (m, 2H), 8.19–8.12 (m, 1H), 7.87–7.81 (m, 1H), 7.74 (d, J = 8.1 Hz, 1H), 7.53 (t, J = 7.0 Hz, 1H), 7.39 (t, J = 8.9 Hz, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 164.51 (d, $^1J_{C–F}$ = 249.5 Hz), 162.68, 151.83, 149.13, 135.10, 130.88, 130.84 (d, $^3J_{C–F}$ = 8.8 Hz), 129.69 (d, $^4J_{C–F}$ = 3.7 Hz), 127.93, 127.07, 126.32, 121.35, 116.12 (d, $^2J_{C–F}$ = 21.38 Hz).

2-(4-chlorophenyl)quinazolin-4(3*H*)-one (5e)[13]



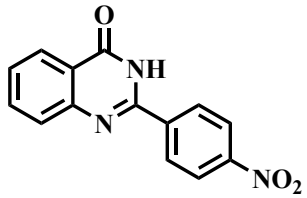
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.60 (s, 1H), 8.21 (d, J = 8.7 Hz, 2H), 8.17 (d, J = 7.8 Hz, 1H), 7.88–7.82 (m, 1H), 7.75 (d, J = 8.1 Hz, 1H), 7.63 (d, J = 8.7 Hz, 2H), 7.54 (t, J = 7.1 Hz, 1H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.10, 151.29, 148.53, 136.25, 134.62, 131.51, 129.57, 128.64, 127.48, 126.73, 125.82, 120.96.

2-(4-bromophenyl)quinazolin-4(3*H*)-one (5f)[14]



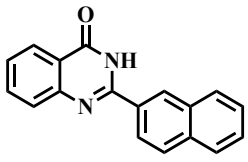
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.62 (s, 1H), 8.23–8.09 (m, 3H), 7.89–7.83 (m, 1H), 7.81–7.72 (m, 3H), 7.57–7.52 (m, 1H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.63, 151.96, 149.06, 135.17, 132.40, 132.10, 130.29, 128.01, 127.28, 126.36, 125.72, 121.49.

2-(4-nitrophenyl)quinazolin-4(3*H*)-one (5g)[15]



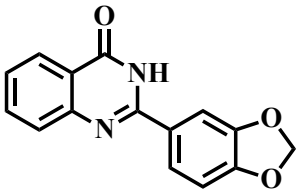
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.85 (s, 1H), 8.41 (dt, J = 6.7, 5.6 Hz, 4H), 8.20 (dd, J = 7.9, 1.2 Hz, 1H), 7.92–7.85 (m, 1H), 7.81 (d, J = 7.6 Hz, 1H), 7.63–7.56 (m, 1H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 163.13, 151.76, 149.37, 148.94, 139.49, 135.06, 129.76, 128.11, 127.61, 126.40, 124.07, 121.73.

2-(naphthalen-2-yl)quinazolin-4(3*H*)-one (5h)[12]



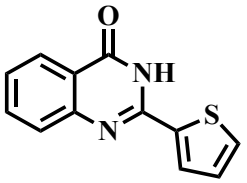
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.68 (s, 1H), 8.23 (d, J = 6.6 Hz, 1H), 8.18 (d, J = 7.5 Hz, 1H), 8.13 (d, J = 8.2 Hz, 1H), 8.06 (d, J = 7.2 Hz, 1H), 7.88 (t, J = 6.9 Hz, 1H), 7.81 (d, J = 7.0 Hz, 1H), 7.75 (d, J = 8.1 Hz, 1H), 7.68–7.64 (m, 1H), 7.62–7.57 (m, 3H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.38, 154.15, 149.21, 135.01, 133.61, 132.20, 130.85, 128.82, 128.16, 127.95, 127.55, 127.27, 126.85, 126.33, 125.69, 121.72.

2-(benzo[*d*][1,3]dioxol-5-yl)quinazolin-4(3*H*)-one (5i)[16]



^{1}H NMR (500 MHz, DMSO-d_6): δ 12.39 (s, 1H), 8.16–8.12 (m, 1H), 7.85–7.80 (m, 2H), 7.76 (d, J = 1.7 Hz, 1H), 7.71 (d, J = 7.9 Hz, 1H), 7.50 (t, J = 7.5 Hz, 1H), 7.09 (d, J = 8.2 Hz, 1H), 6.15 (s, 2H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.71, 152.07, 150.54, 149.23, 148.17, 135.07, 127.83, 126.97, 126.77, 126.30, 123.31, 121.22, 108.76, 108.03, 102.36.

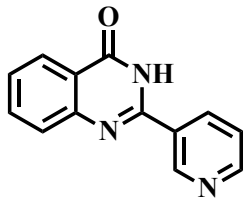
2-(thiophen-2-yl)quinazolin-4(3*H*)-one (5j)[17]



1H NMR (500 MHz, DMSO-d_6): δ 12.65 (s, 1H), 8.23 (d, J = 1.1 Hz, 1H), 8.12 (d, J = 7.8 Hz, 1H), 7.87 (d, J = 4.9 Hz, 1H), 7.80 (t, J = 7.6 Hz, 1H), 7.65 (d, J = 8.1 Hz, 1H), 7.48 (t, J = 7.5 Hz, 1H), 7.27–7.19 (m, 1H). ^{13}C NMR (126 MHz, DMSO-

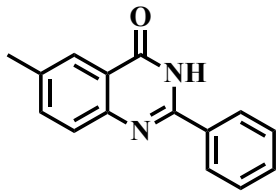
d_6): δ 162.26, 149.11, 148.31, 137.84, 135.17, 132.65, 129.88, 128.99, 127.42, 126.81, 126.47, 121.35.

2-(pyridin-3-yl)quinazolin-4(3*H*)-one (5k)[18]



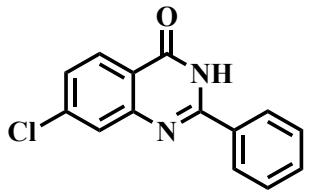
^{1}H NMR (500 MHz, DMSO-d_6): δ 12.75 (s, 1H), 9.30 (d, J = 1.8 Hz, 1H), 8.77 (dd, J = 4.7, 1.5 Hz, 1H), 8.54–8.46 (m, 1H), 8.18 (dd, J = 7.9, 1.1 Hz, 1H), 7.90–7.85 (m, 1H), 7.78 (d, J = 7.8 Hz, 1H), 7.62–7.54 (m, 2H).^{13}C NMR (126 MHz, DMSO-d_6): δ 162.57, 152.30, 151.23, 149.23, 149.01, 135.87, 135.20, 129.21, 128.07, 127.45, 126.36, 124.01, 121.62.

6-methyl-2-phenylquinazolin-4(3*H*)-one (5m)[13]



^{1}H NMR (500 MHz, DMSO-d_6): δ 12.46 (s, 1H), 8.18 (d, J = 7.0 Hz, 2H), 7.96 (s, 1H), 7.66 (s, 2H), 7.61–7.51 (m, 3H), 2.46 (s, 3H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.62, 151.93, 147.22, 136.78, 136.34, 133.26, 131.69, 129.05, 128.11, 127.86, 125.71, 121.20, 21.32.

7-chloro-2-phenylquinazolin-4(3*H*)-one (5n)[19]



^{1}H NMR (500 MHz, DMSO-d_6): δ 12.68 (s, 1H), 8.16 (dd, J = 14.3, 8.3 Hz, 3H), 7.80 (s, 1H), 7.64–7.53 (m, 4H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 162.13, 154.27, 150.36, 139.65, 132.86, 132.19, 129.11, 128.42, 128.39, 127.28, 127.06, 120.30.

10. ^{1}H and ^{13}C NMR spectra of 2-substituted benzimidazoles (3a-3n)

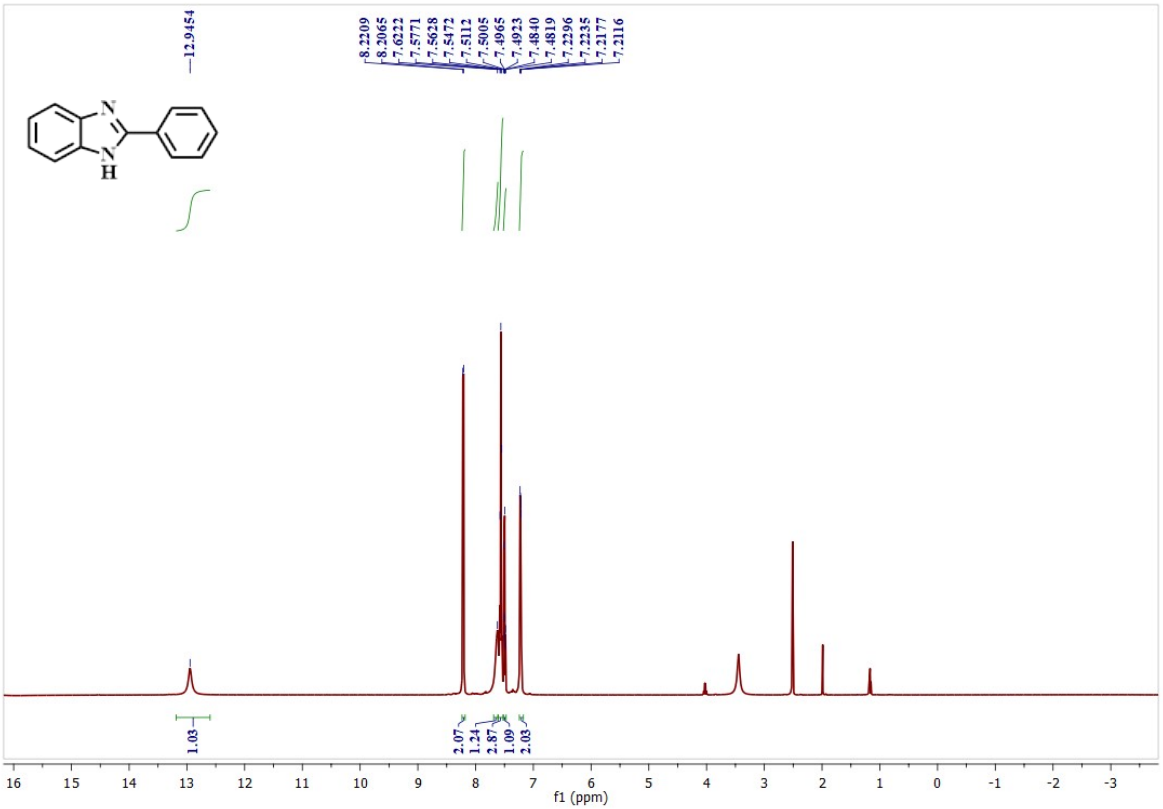


Fig. S2. ^{1}H NMR spectrum of **3a**.

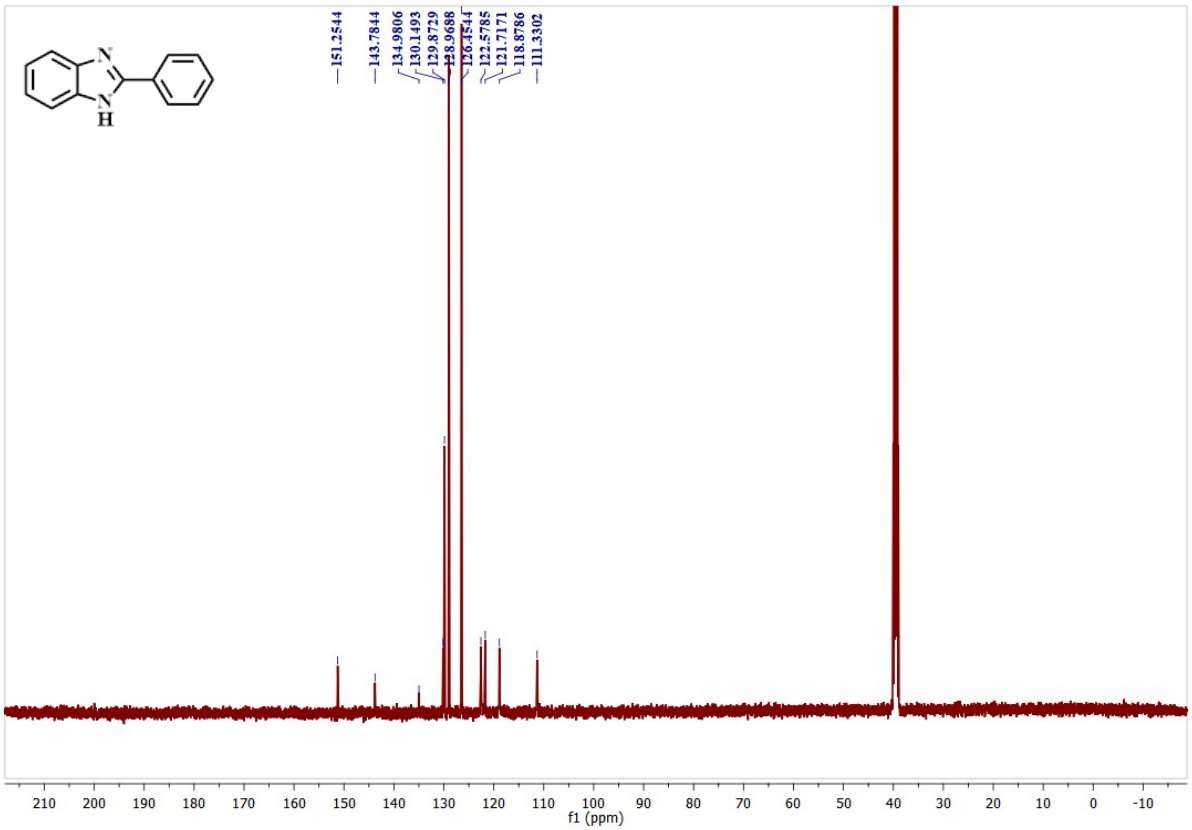


Fig. S3. ^{13}C NMR spectrum of **3a**.

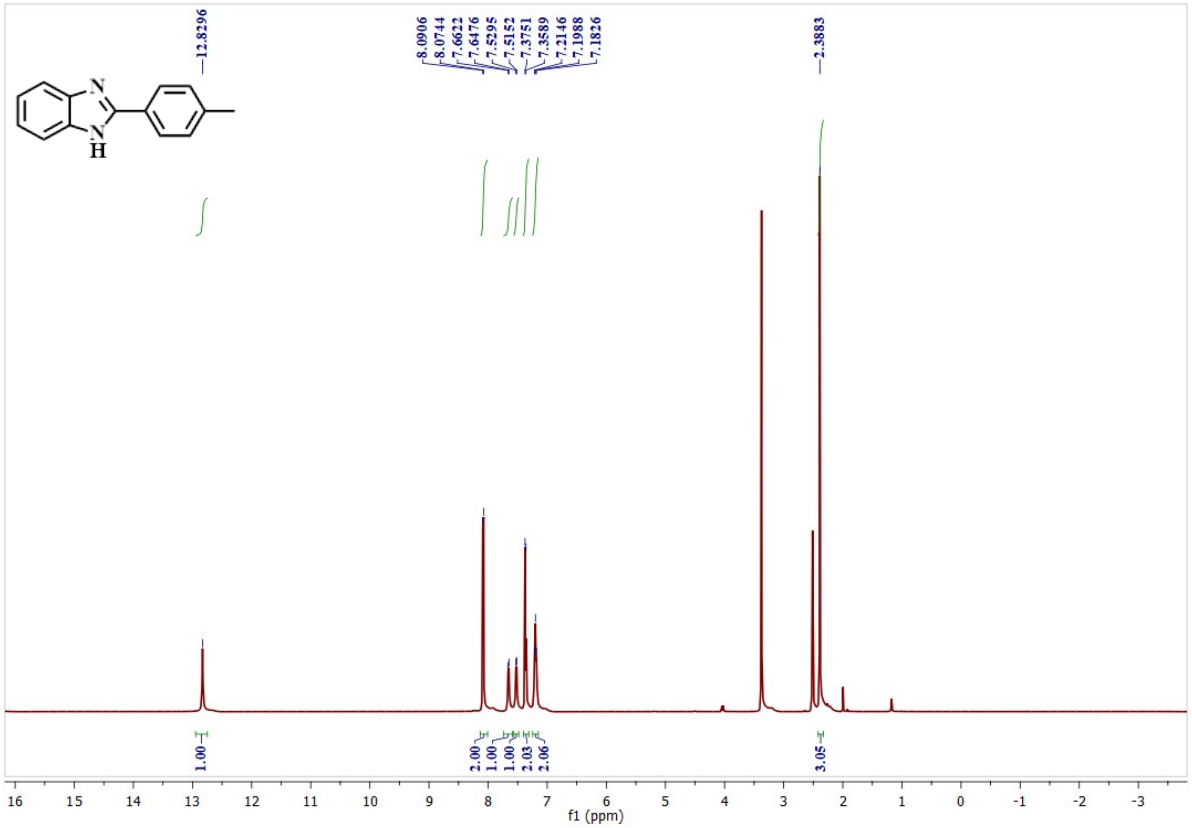


Fig. S4. ^{1}H NMR spectrum of **3b**.

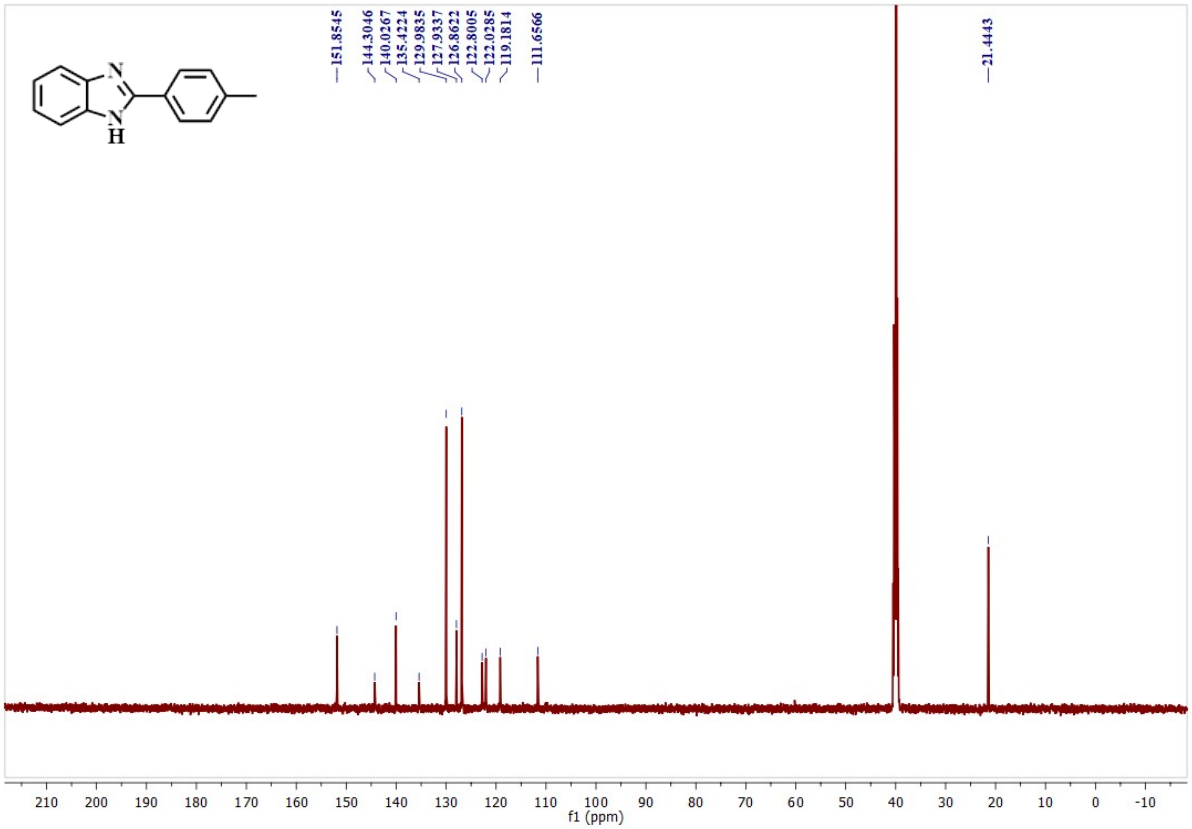


Fig. S5. ^{13}C NMR spectrum of **3b**.

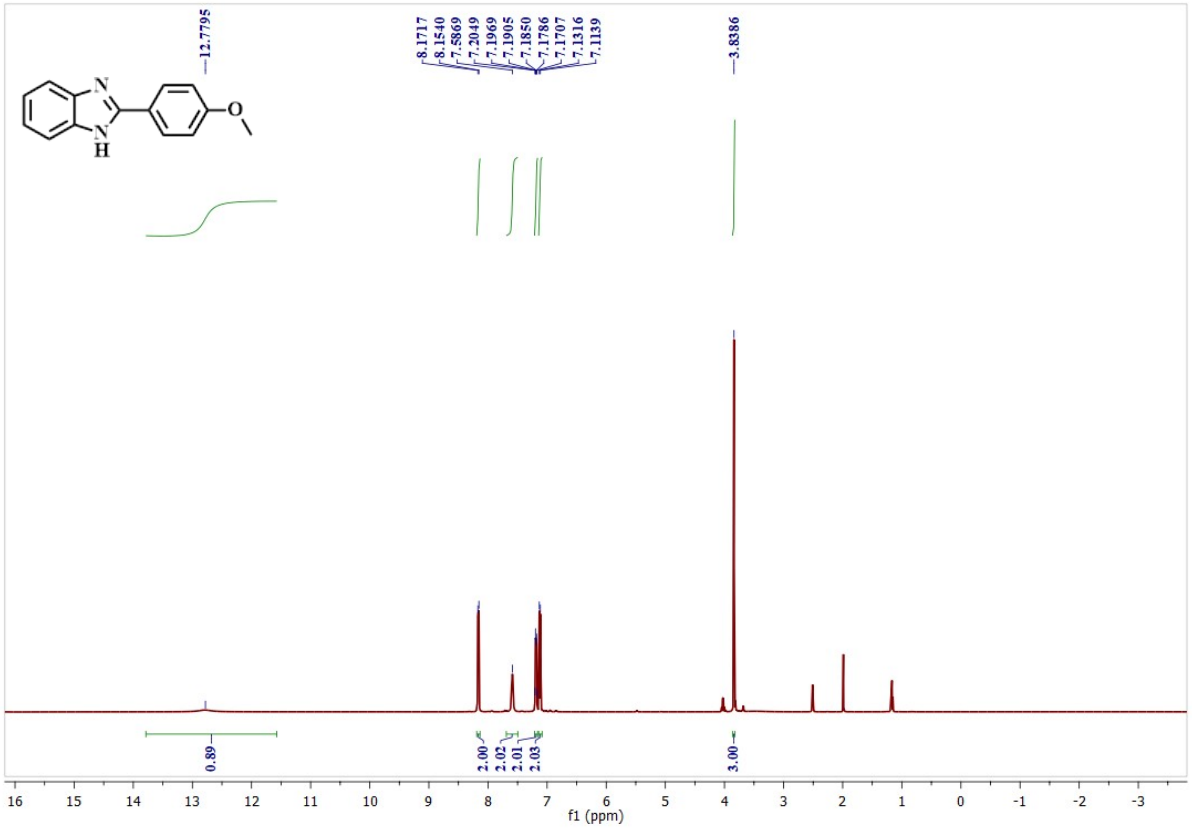


Fig. S6. ^{1}H NMR spectrum of **3c**.

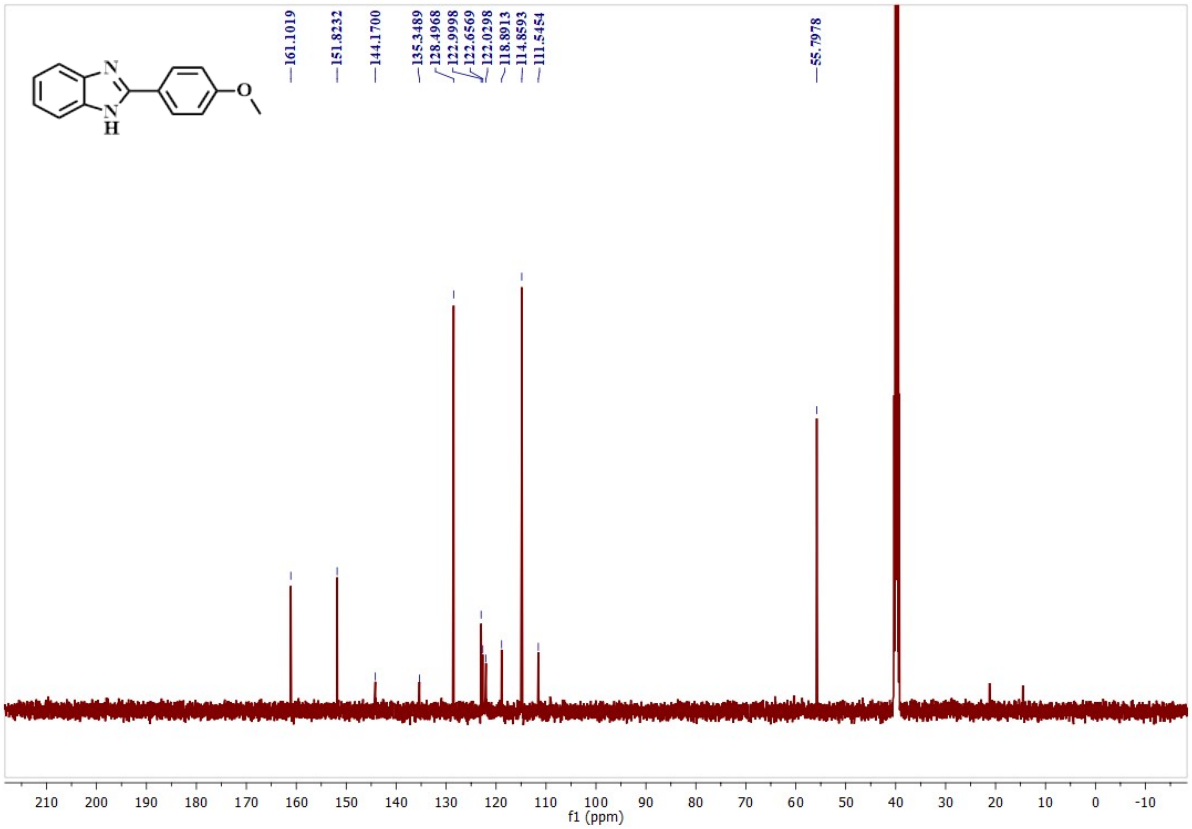


Fig. S7. ^{13}C NMR spectrum of **3c**.

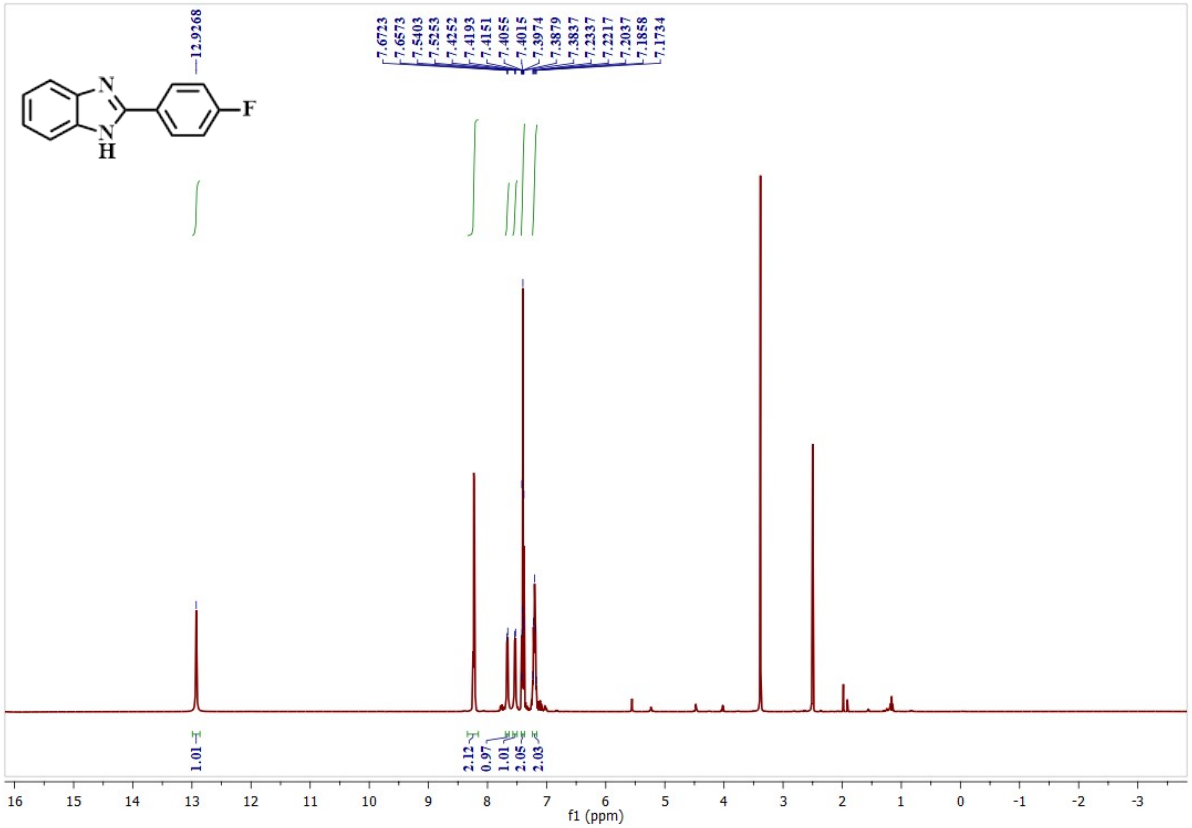


Fig. S8. ^{1}H NMR spectrum of **3d**.

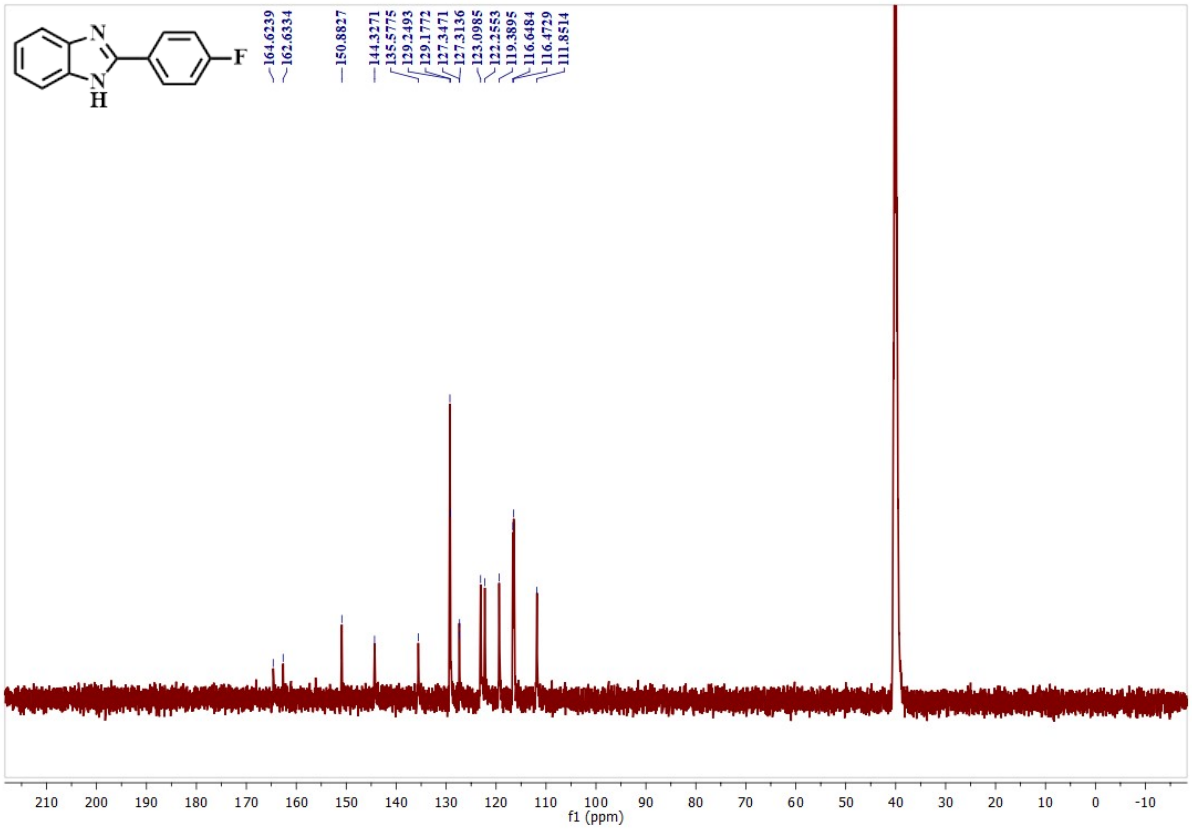


Fig. S9. ^{13}C NMR spectrum of **3d**.

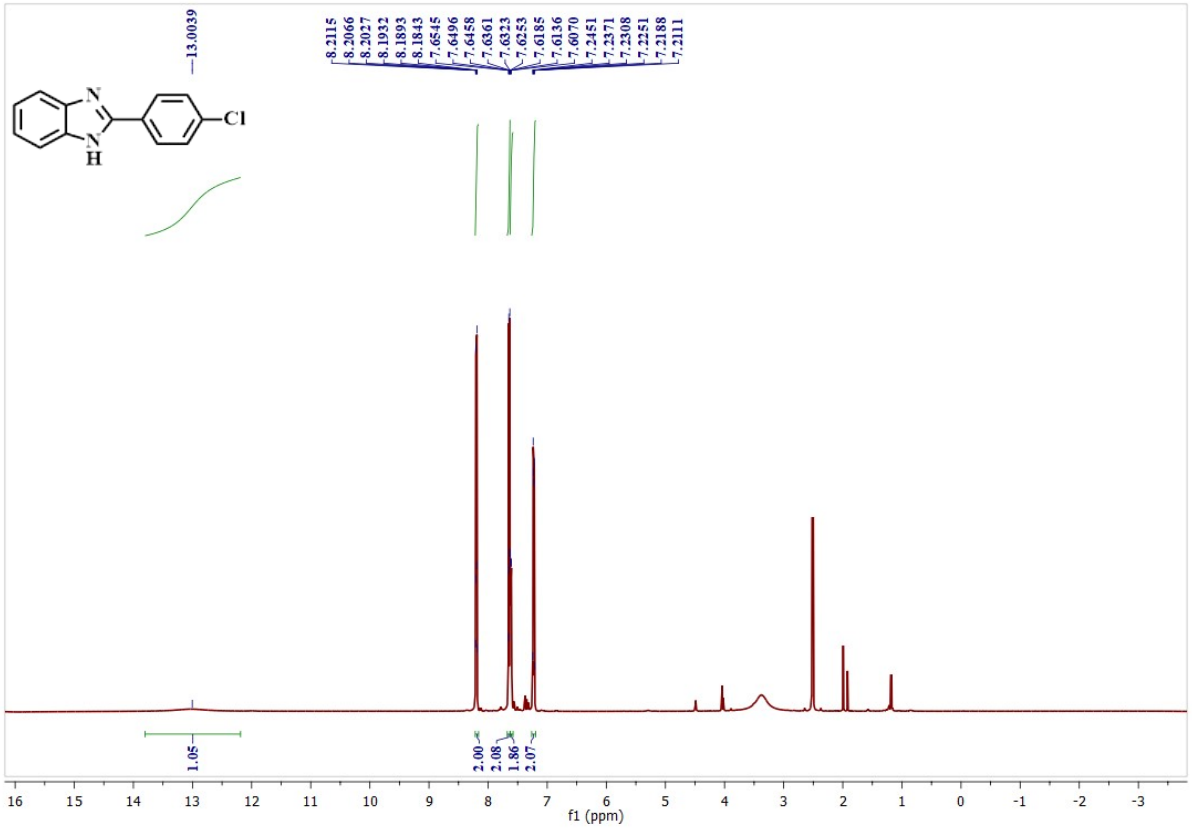


Fig. S10. ^{1}H NMR spectrum of **3e**.

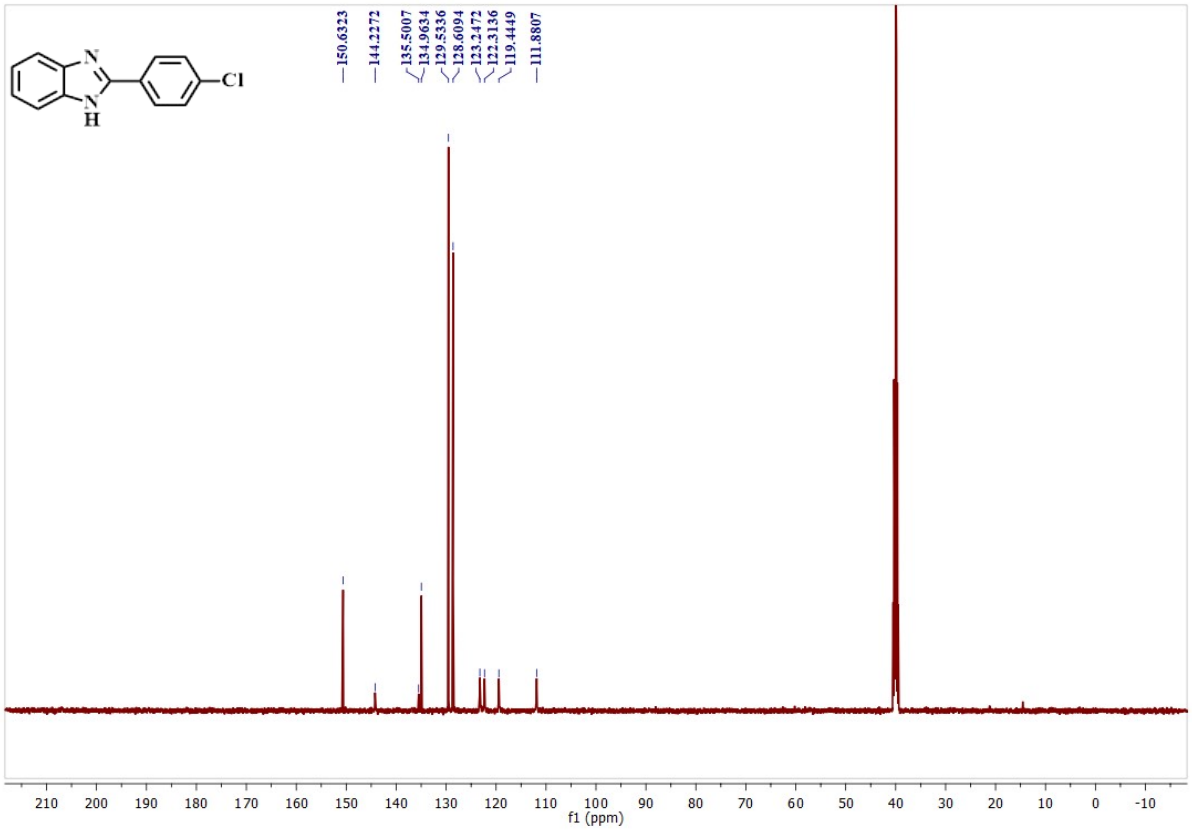


Fig. S11. ^{13}C NMR spectrum of **3e**.

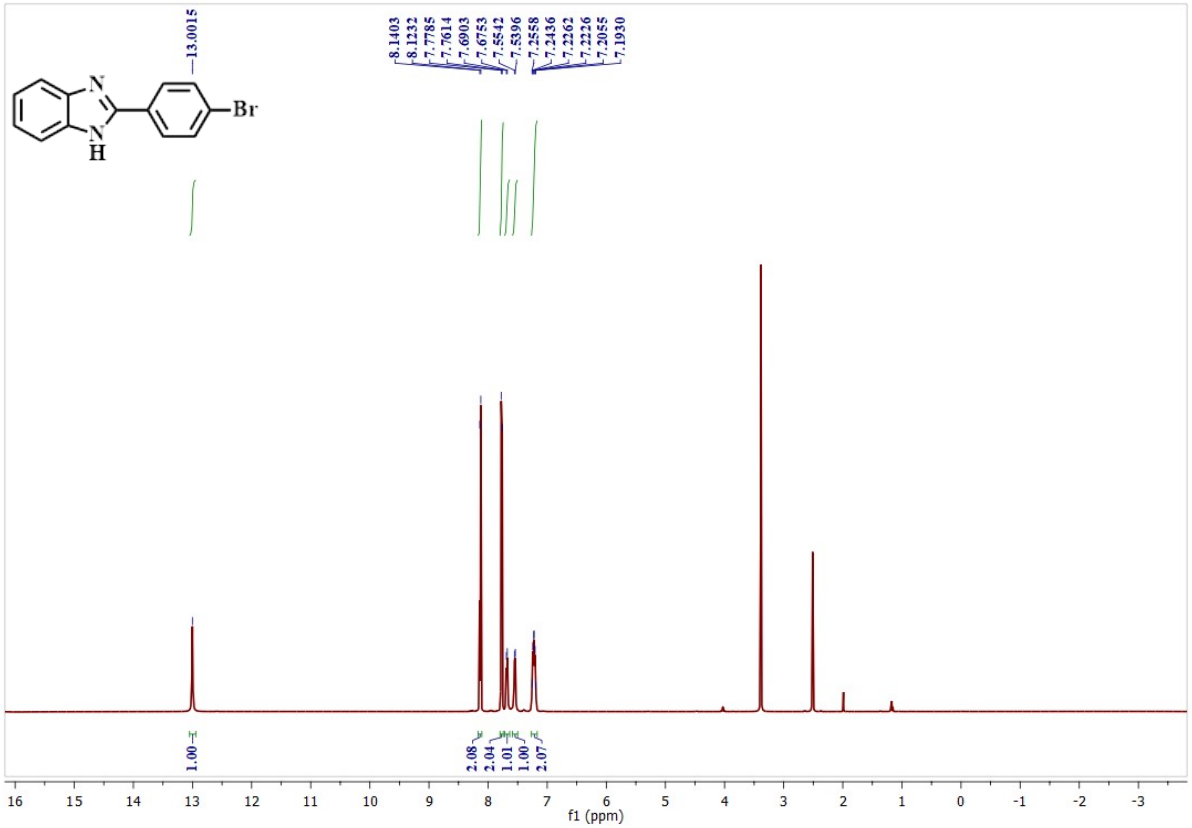


Fig. S12. ^{1}H NMR spectrum of **3f**.

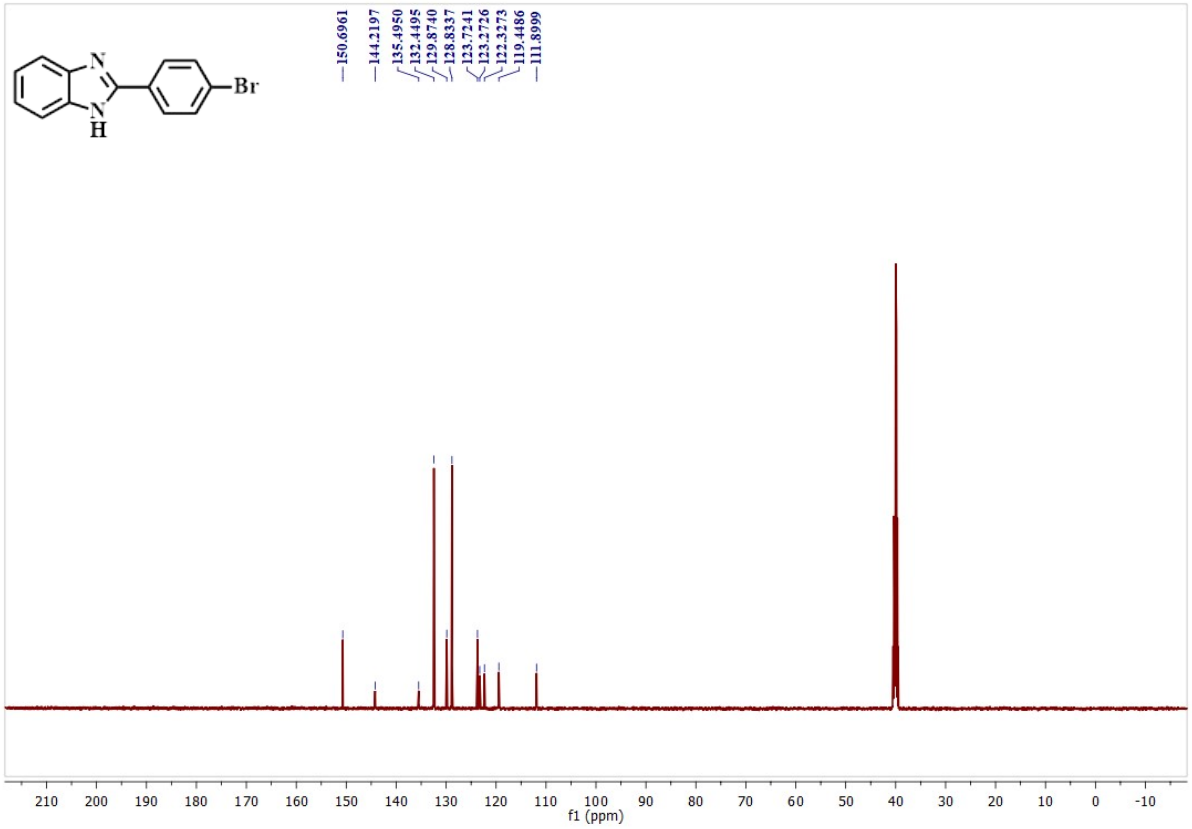


Fig. S13. ^{13}C NMR spectrum of **3f**.

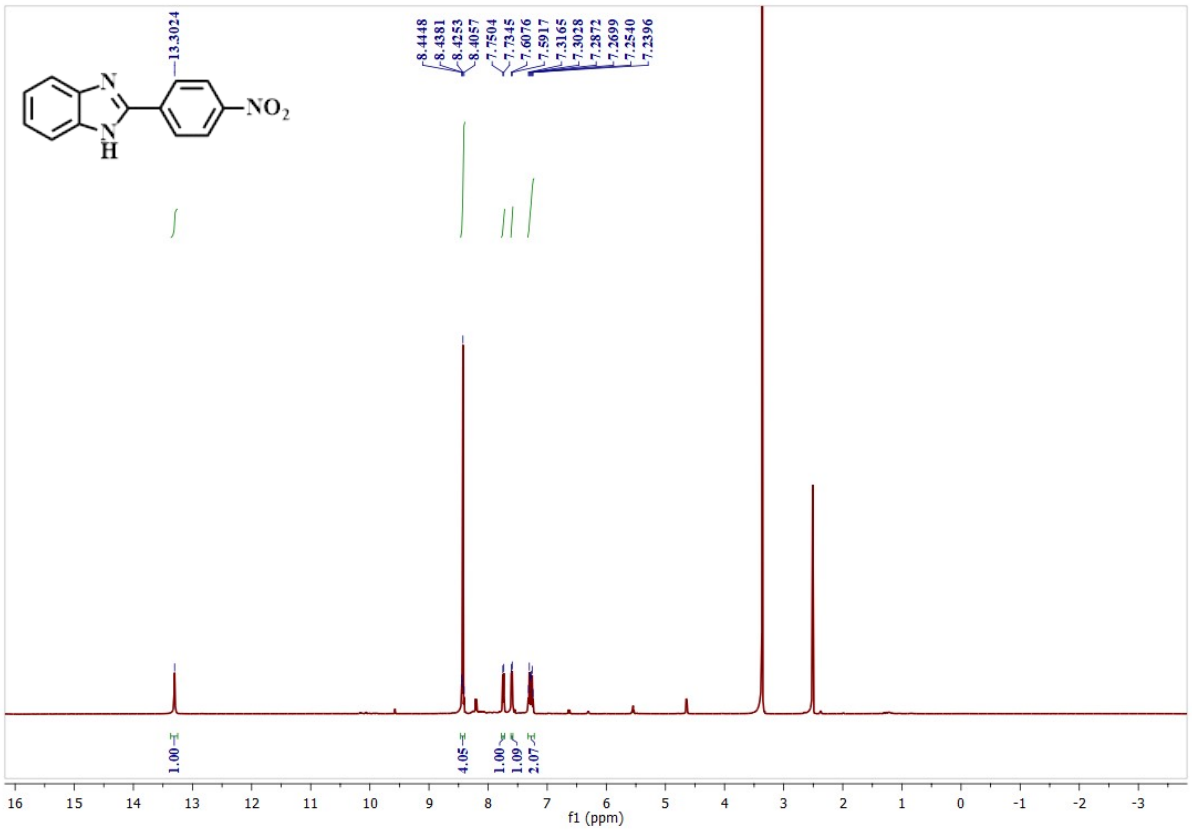


Fig. S14. ^{1}H NMR spectrum of **3g**.

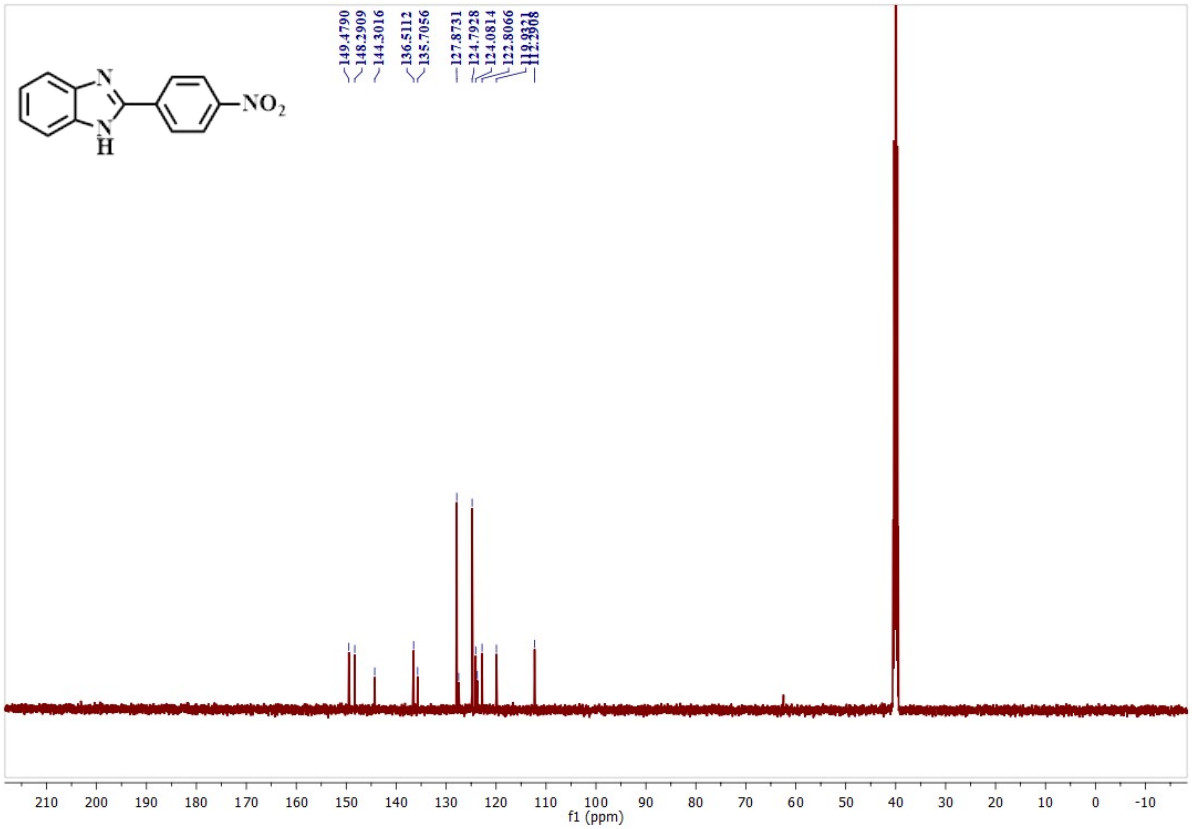


Fig. S15. ^{13}C NMR spectrum of **3g**.

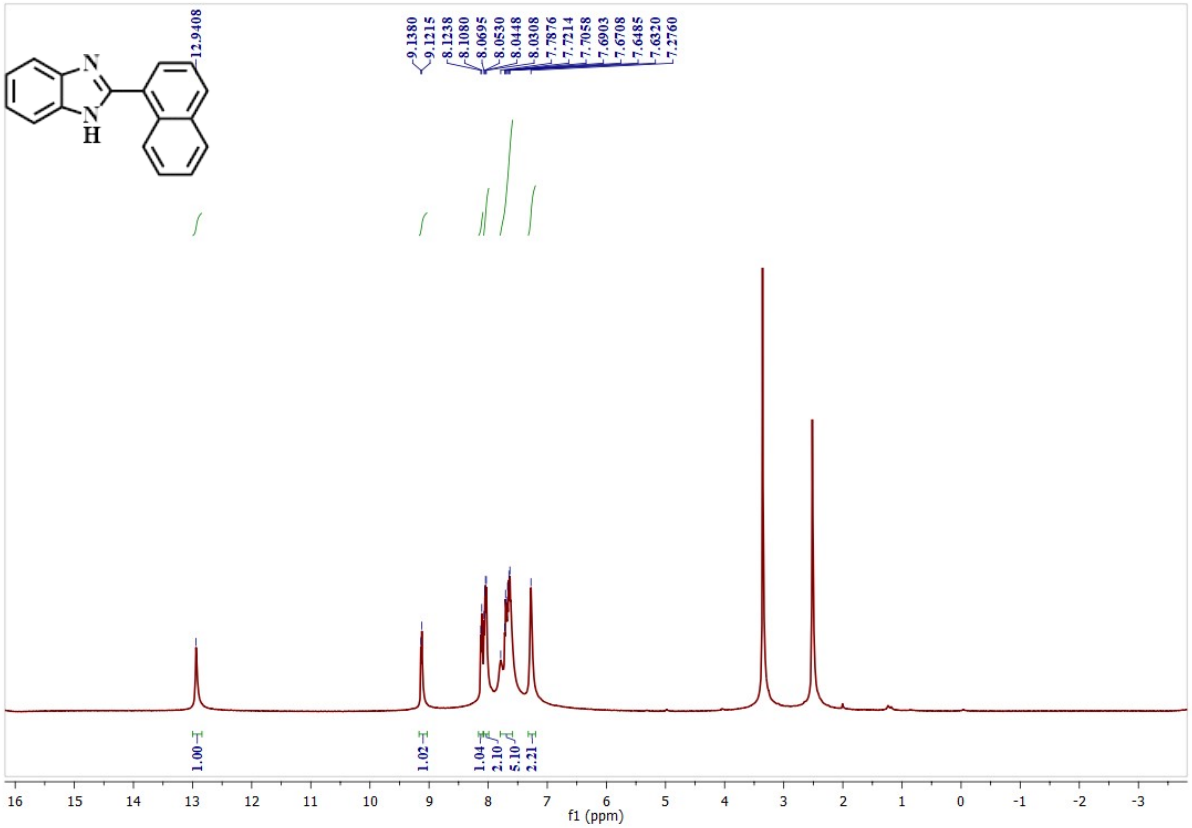


Fig. S16. ^{1}H NMR spectrum of **3h**.

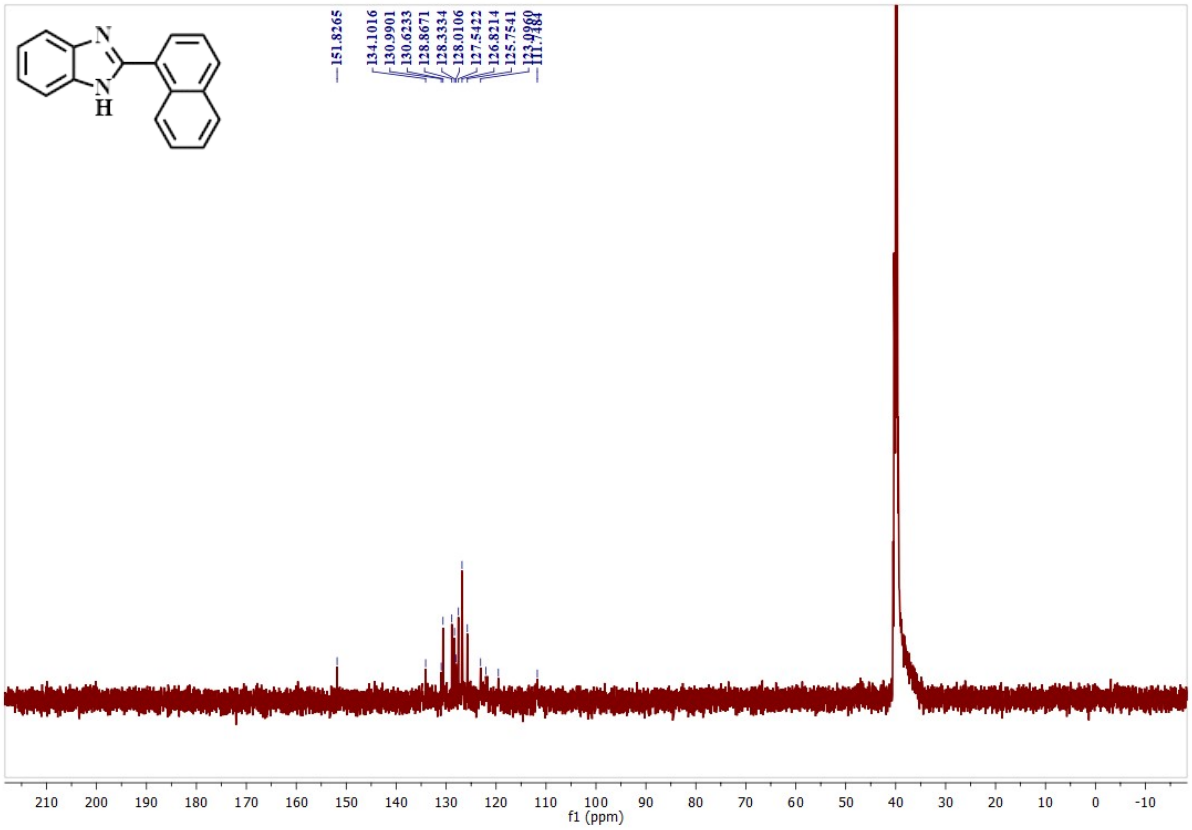


Fig. S17. ^{13}C NMR spectrum of **3h**.

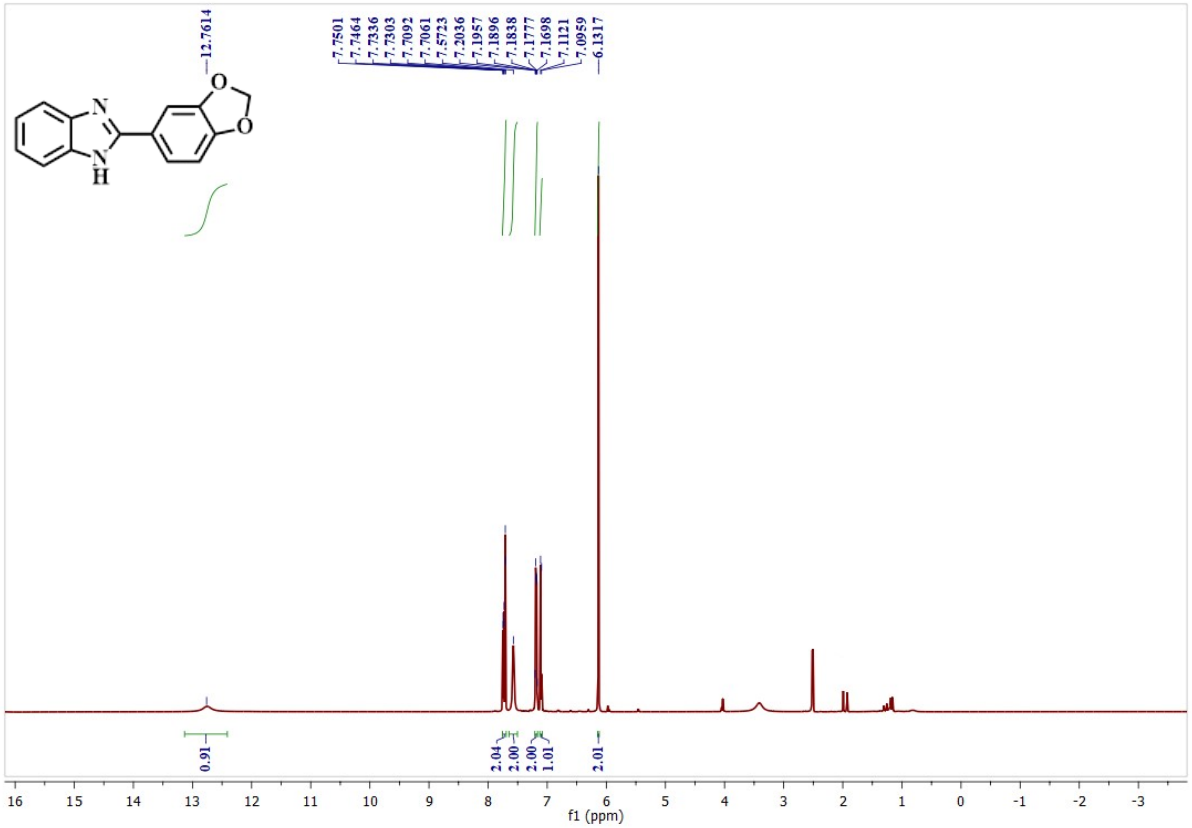


Fig. S18. ^{1}H NMR spectrum of **3i**.

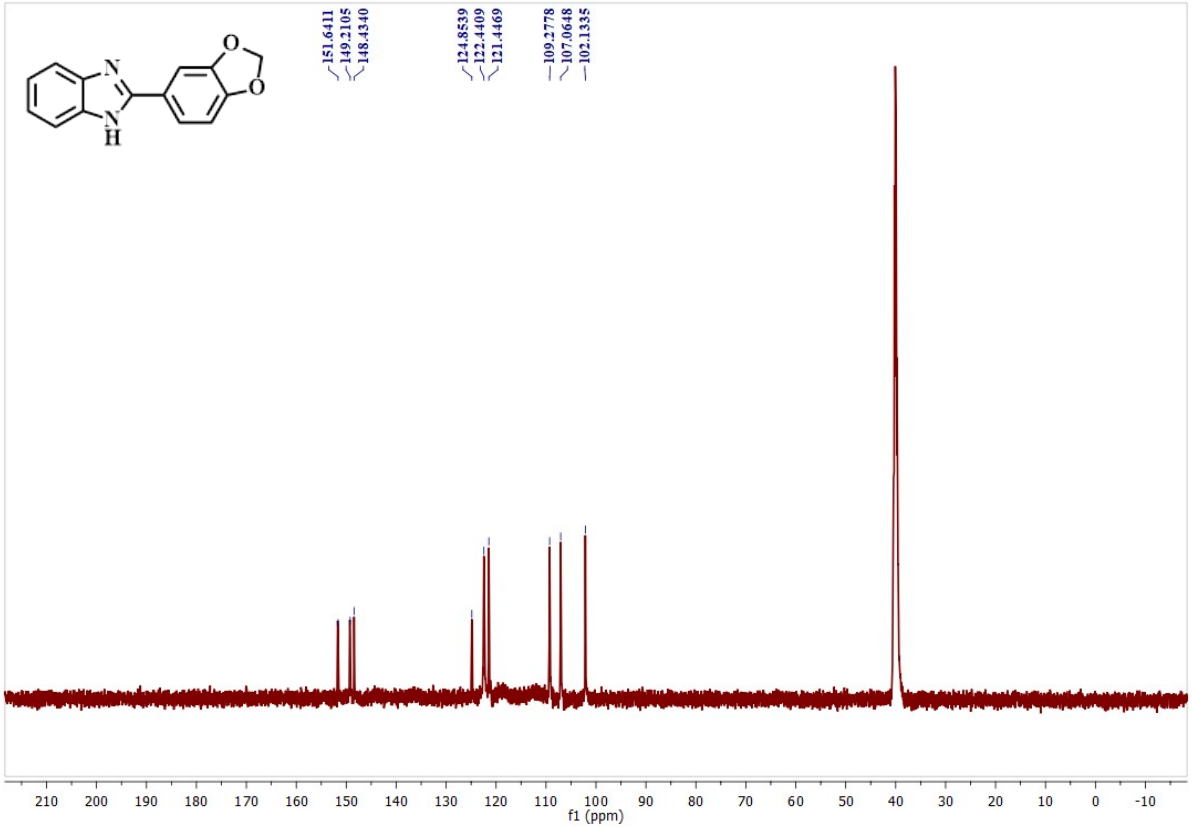


Fig. S19. ^{13}C NMR spectrum of **3i**.

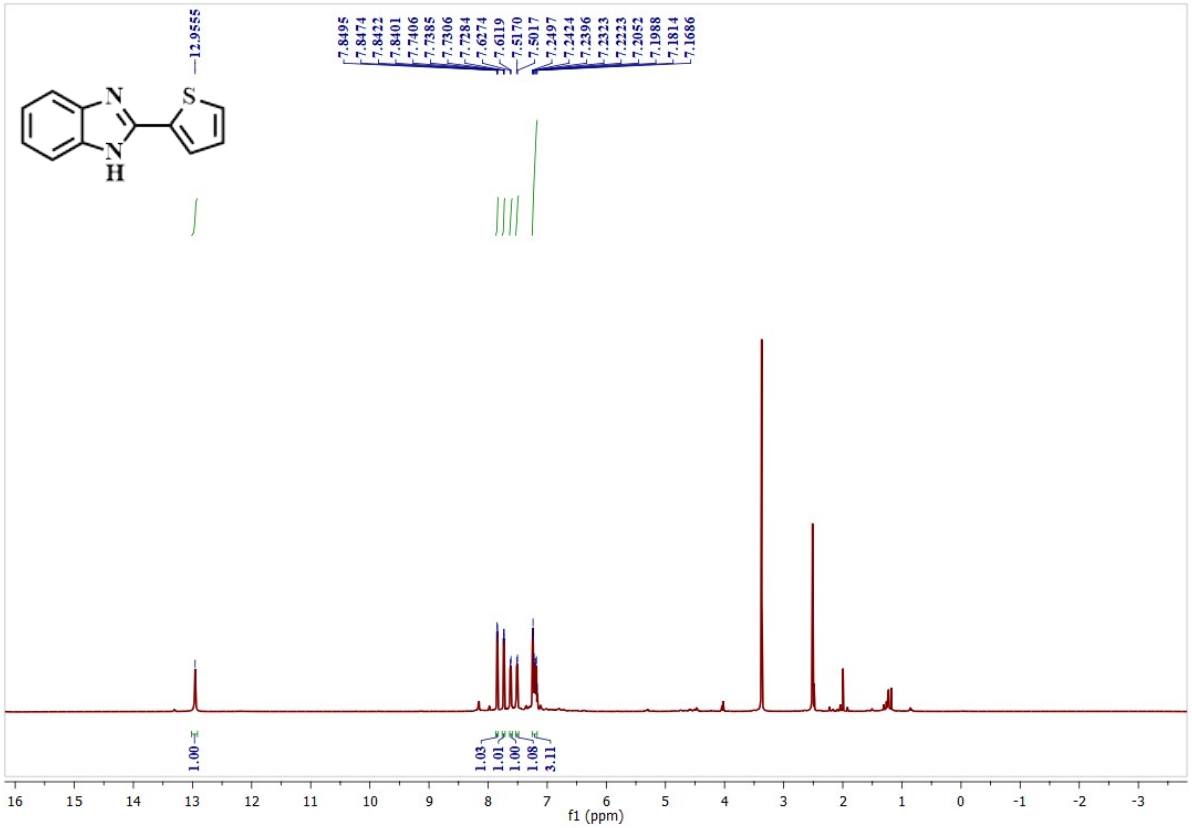


Fig. S20. ^{1}H NMR spectrum of **3j**.

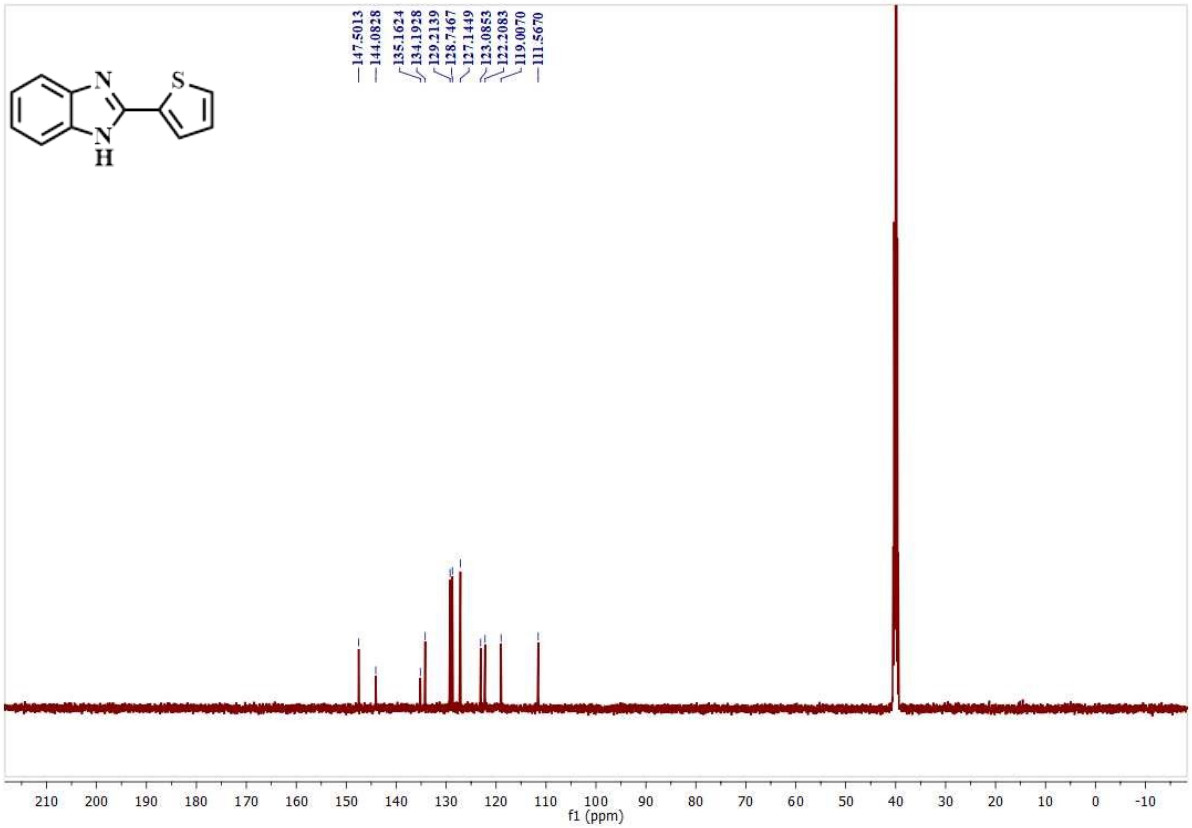


Fig. S21. ^{13}C NMR spectrum of **3j**.

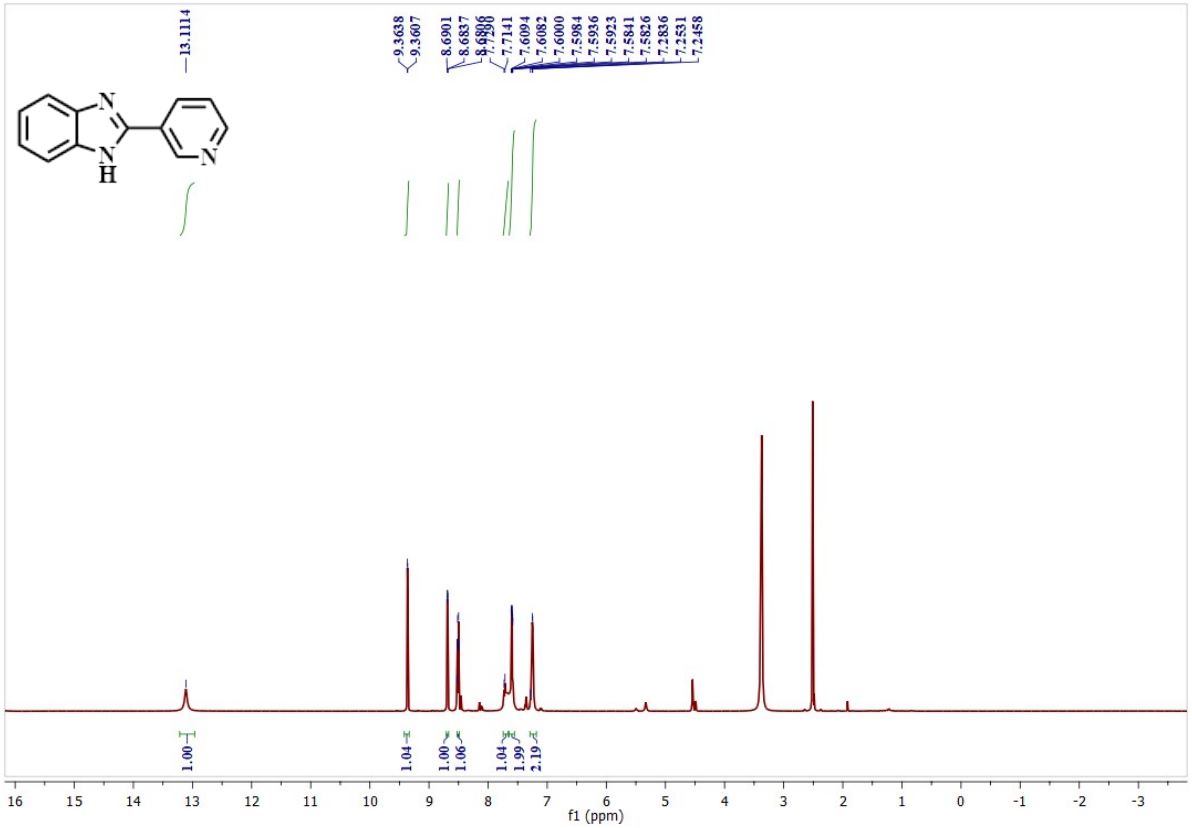


Fig. S22. ^{1}H NMR spectrum of **3k**.

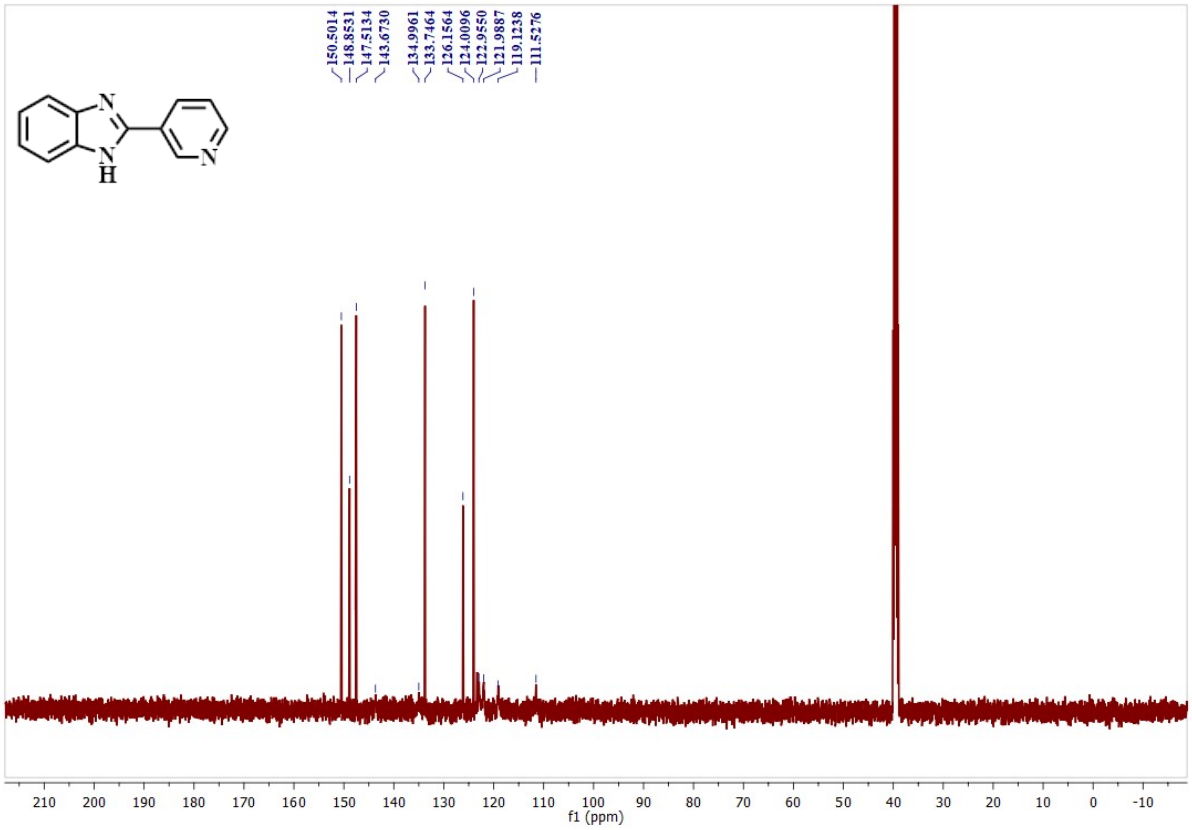


Fig. S23. ^{13}C NMR spectrum of **3k**.

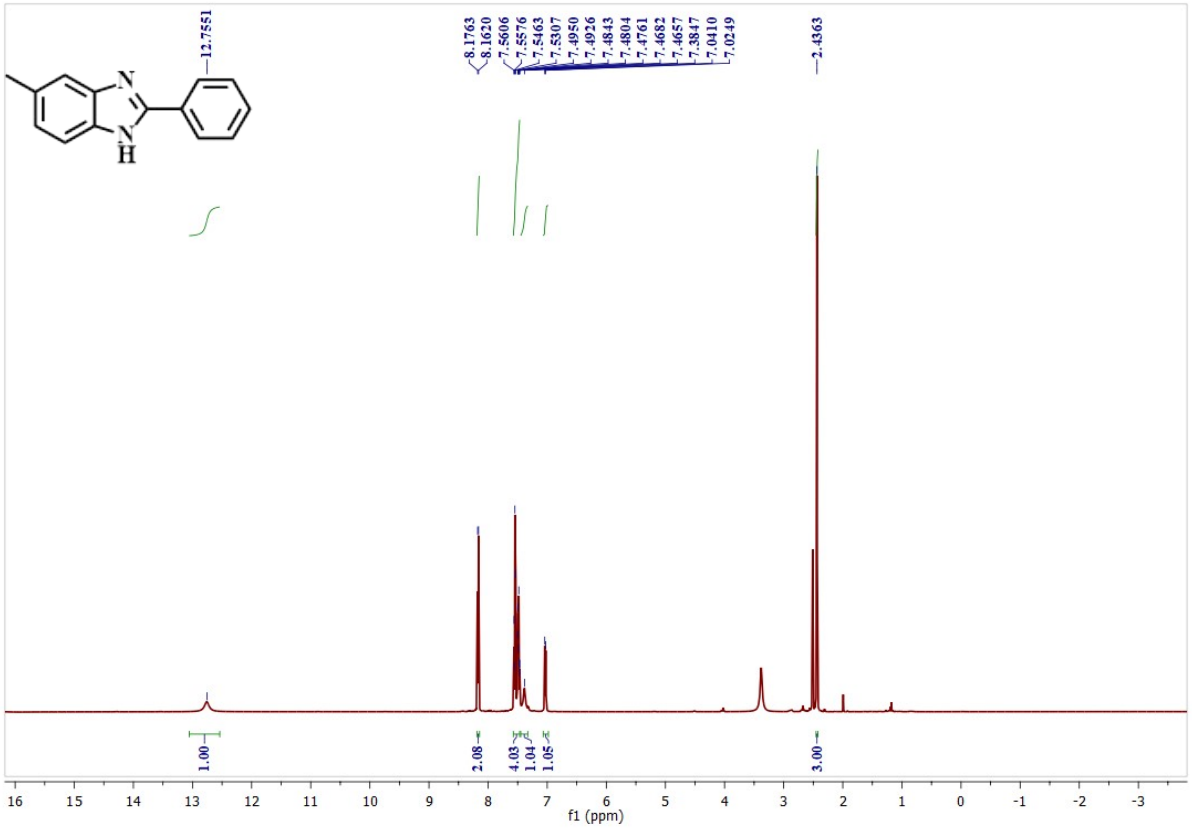


Fig. S24. ^{1}H NMR spectrum of **3m**.

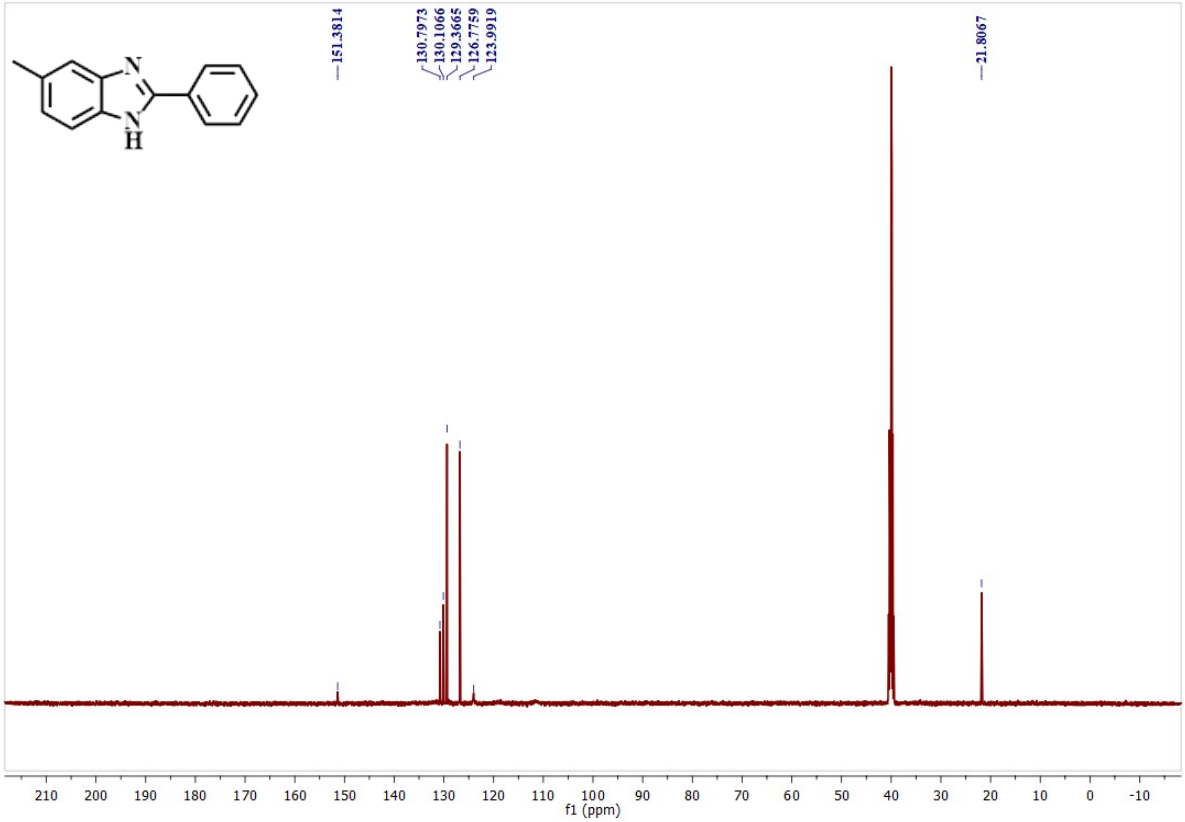


Fig. S25. ^{13}C NMR spectrum of **3m**.

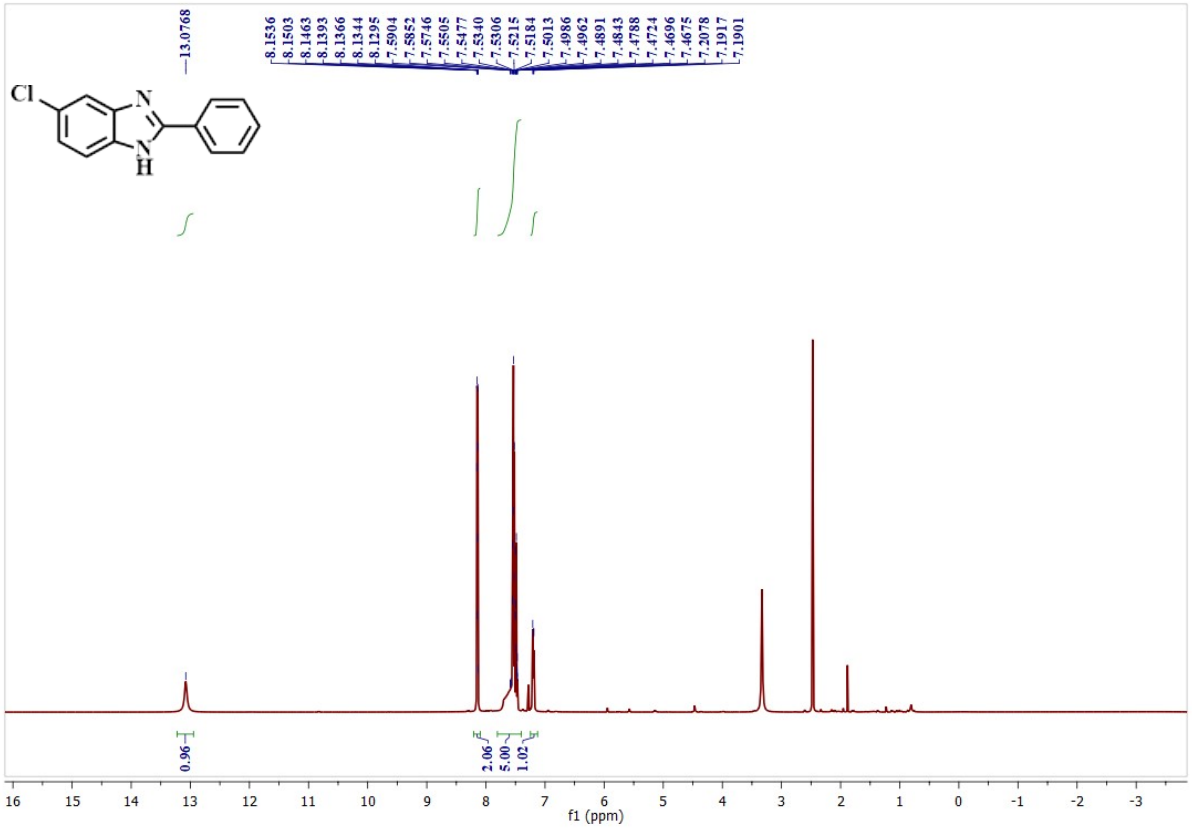


Fig. S26. ^{1}H NMR spectrum of **3n**.

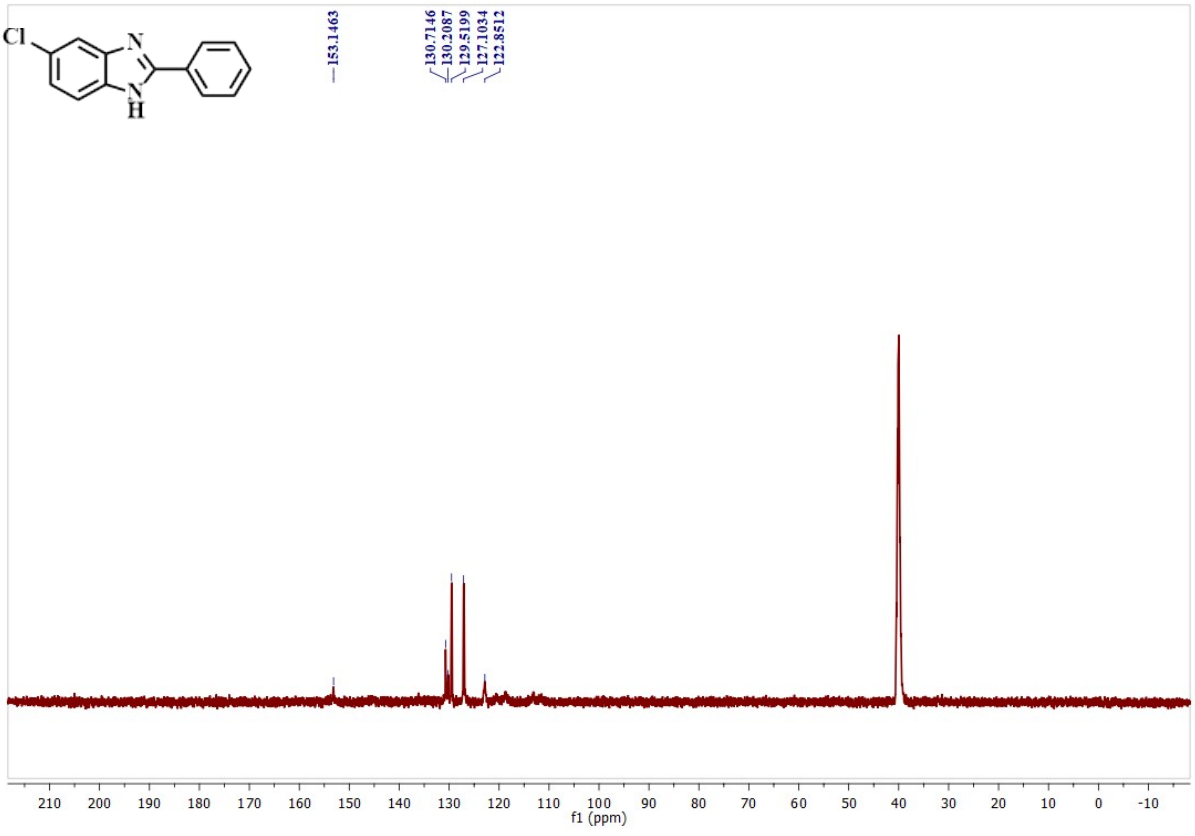


Fig. S27. ^{13}C NMR spectrum of **3n**.

11. ^{1}H and ^{13}C NMR spectra of 2-substituted quinazolinone derivatives (5a-5n)

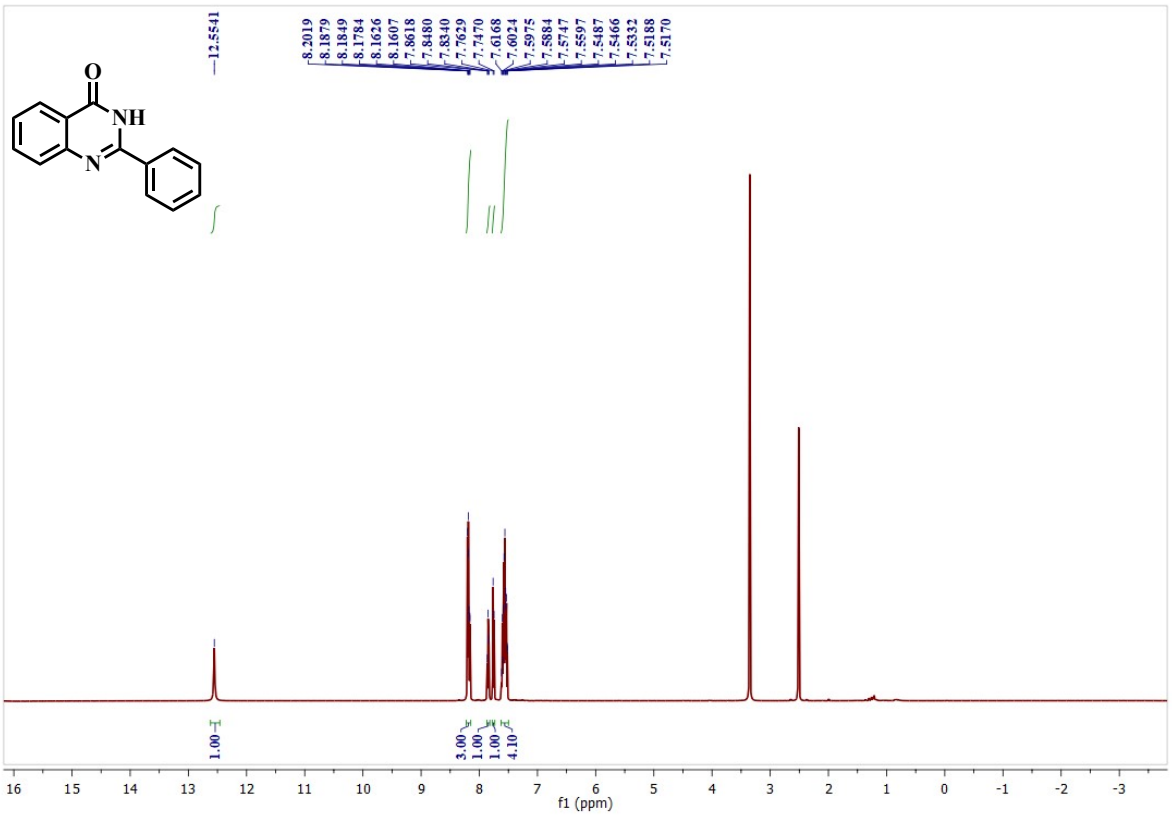


Fig. S28. ^{1}H NMR spectrum of **5a**.

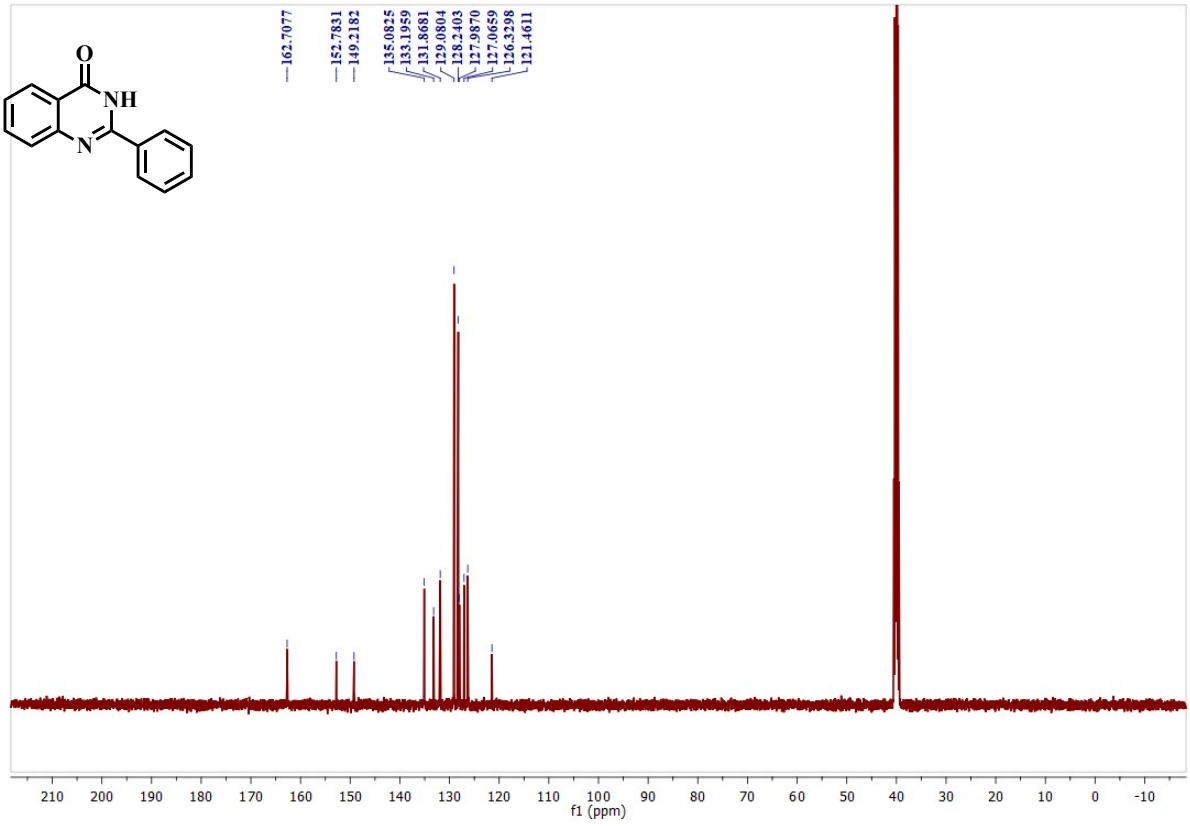


Fig. S29. ^{13}C NMR spectrum of **5a**.

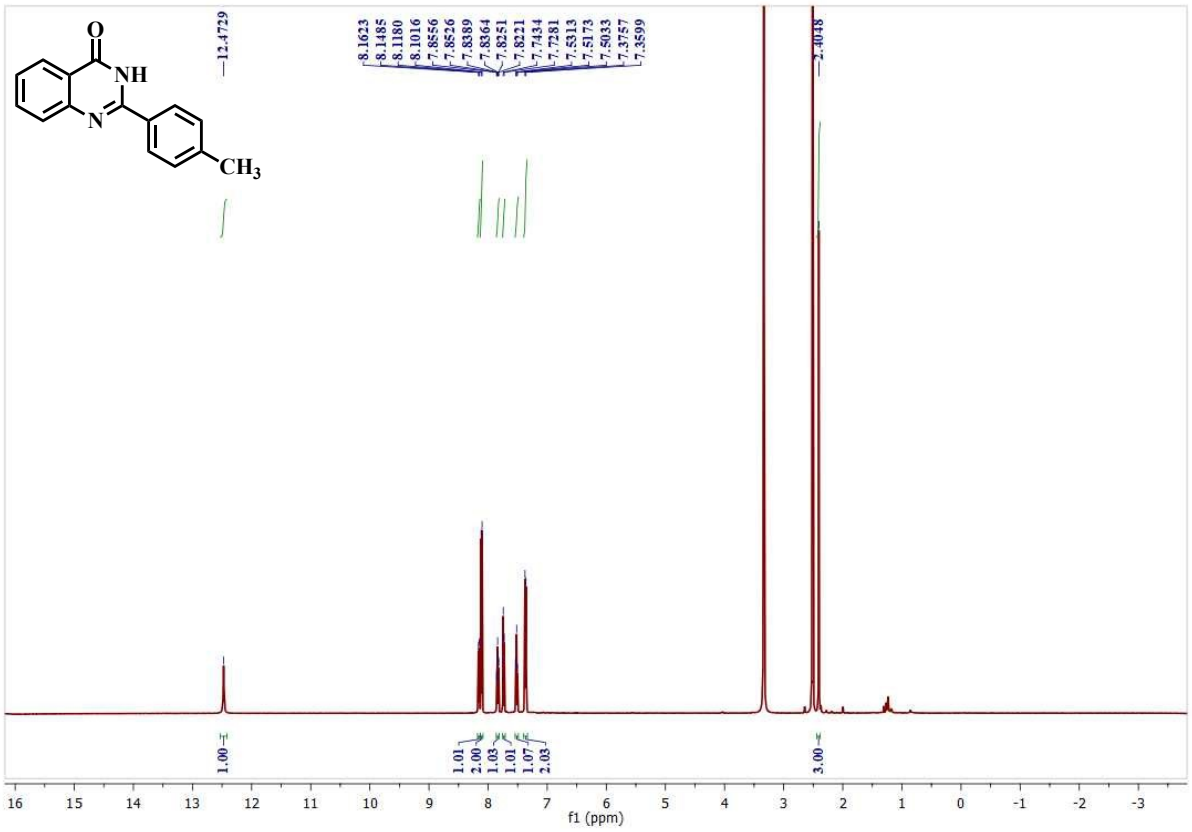


Fig. S30. ^{1}H NMR spectrum of **5b**.

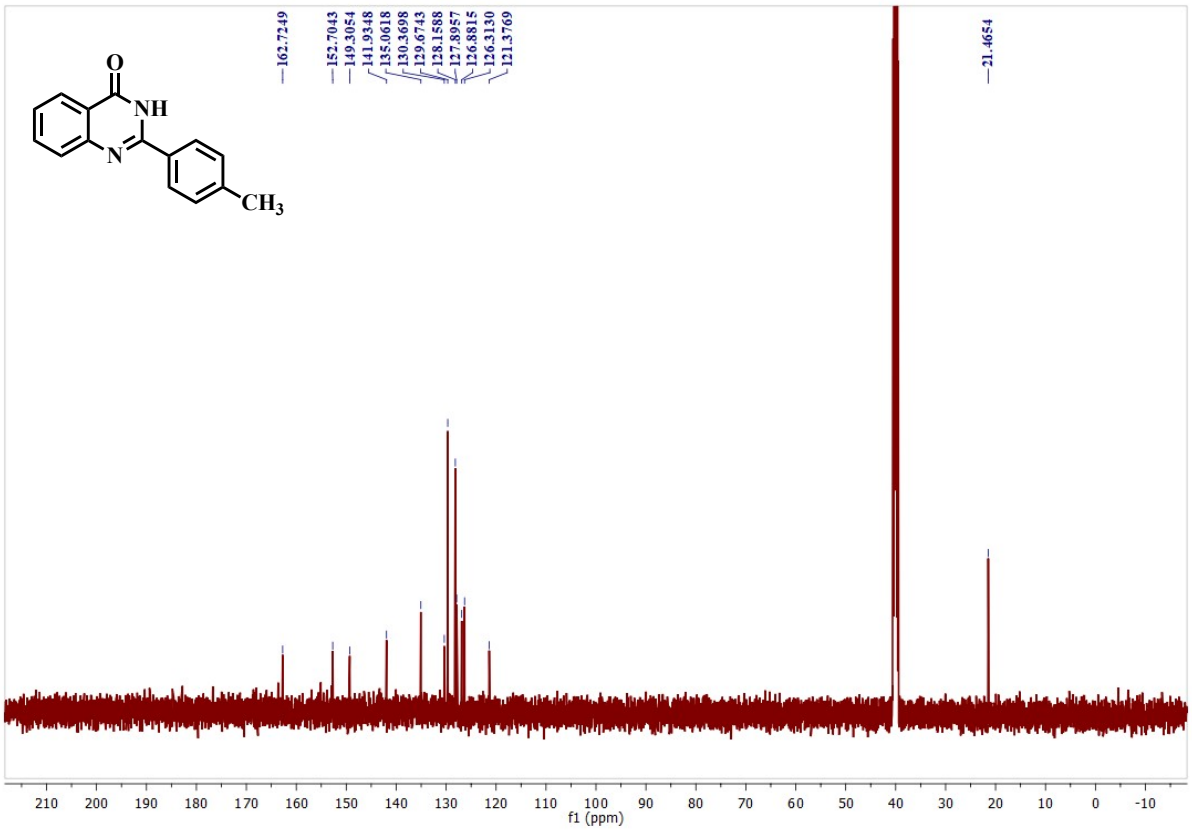


Fig. S31. ^{13}C NMR spectrum of **5b**.

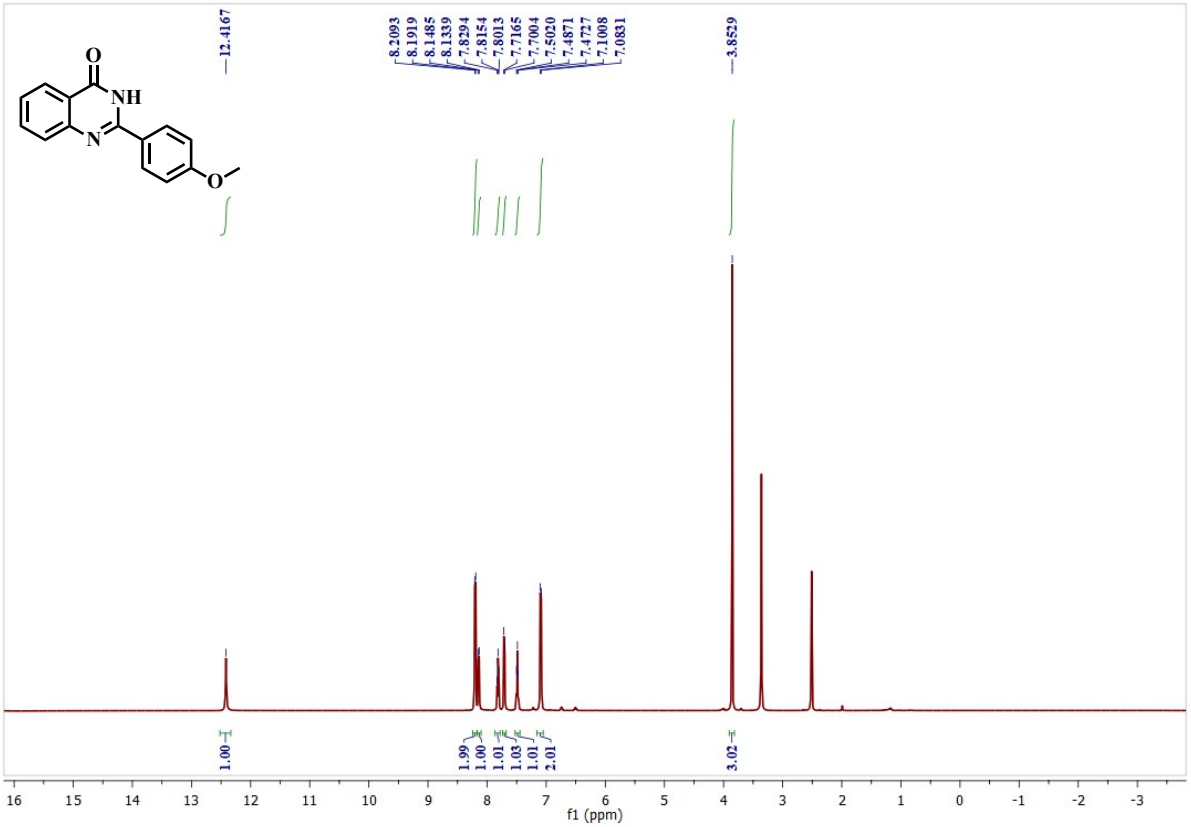


Fig. S32. ^{1}H NMR spectrum of **5c**.

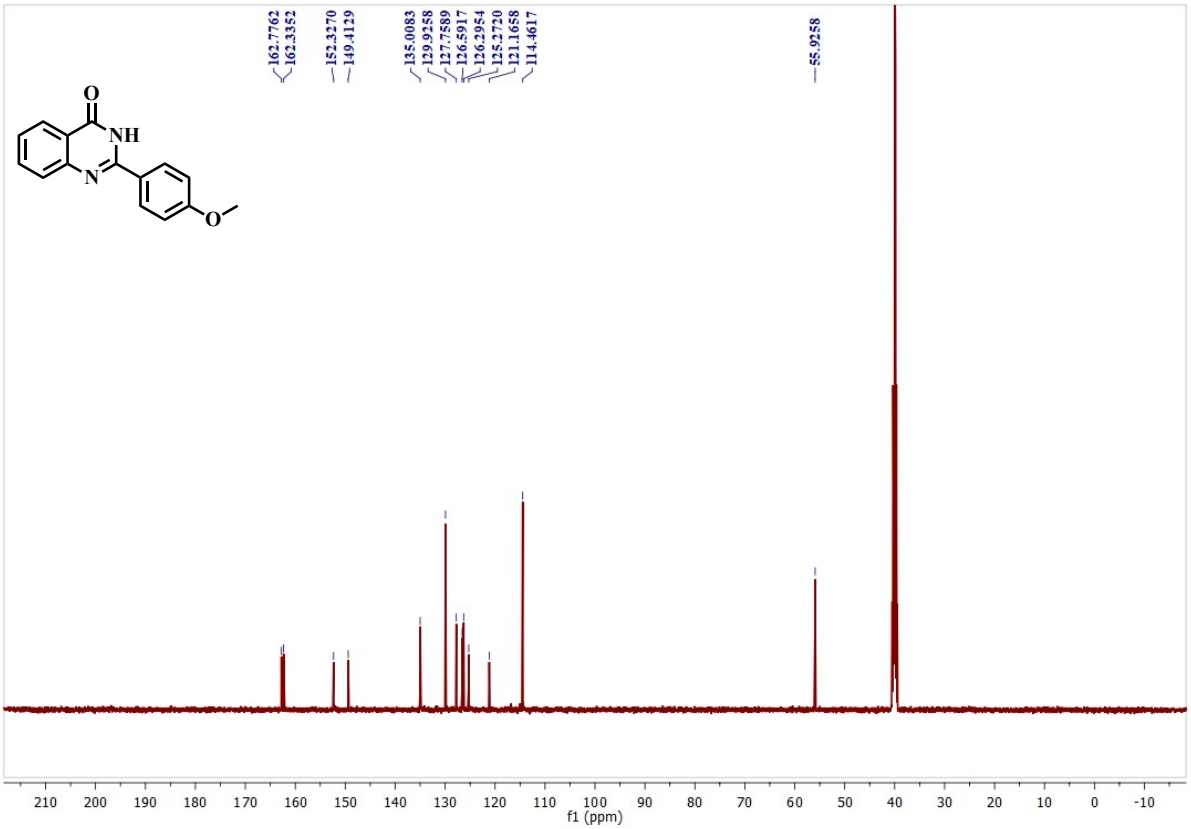


Fig. S33. ^{13}C NMR spectrum of **5c**.

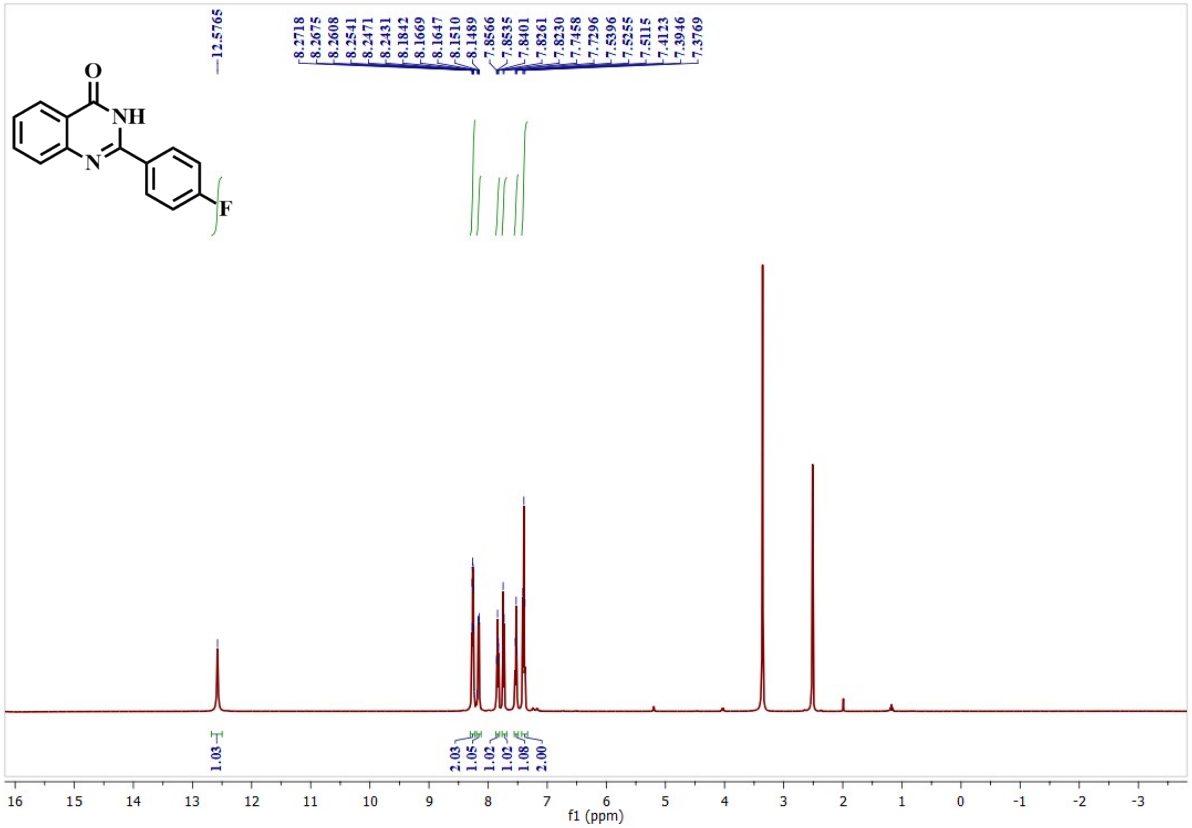


Fig. S34. ^{1}H NMR spectrum of **5d**.

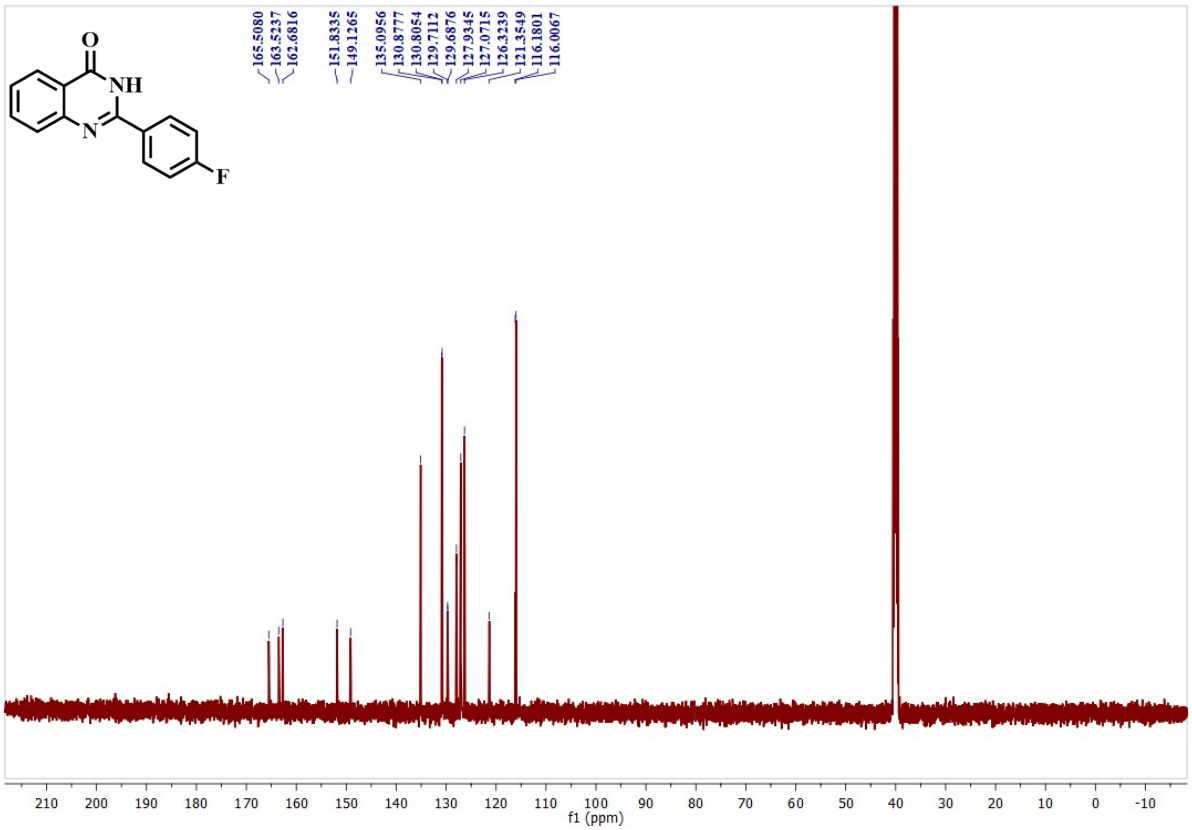


Fig. S35. ^{13}C NMR spectrum of **5d**.

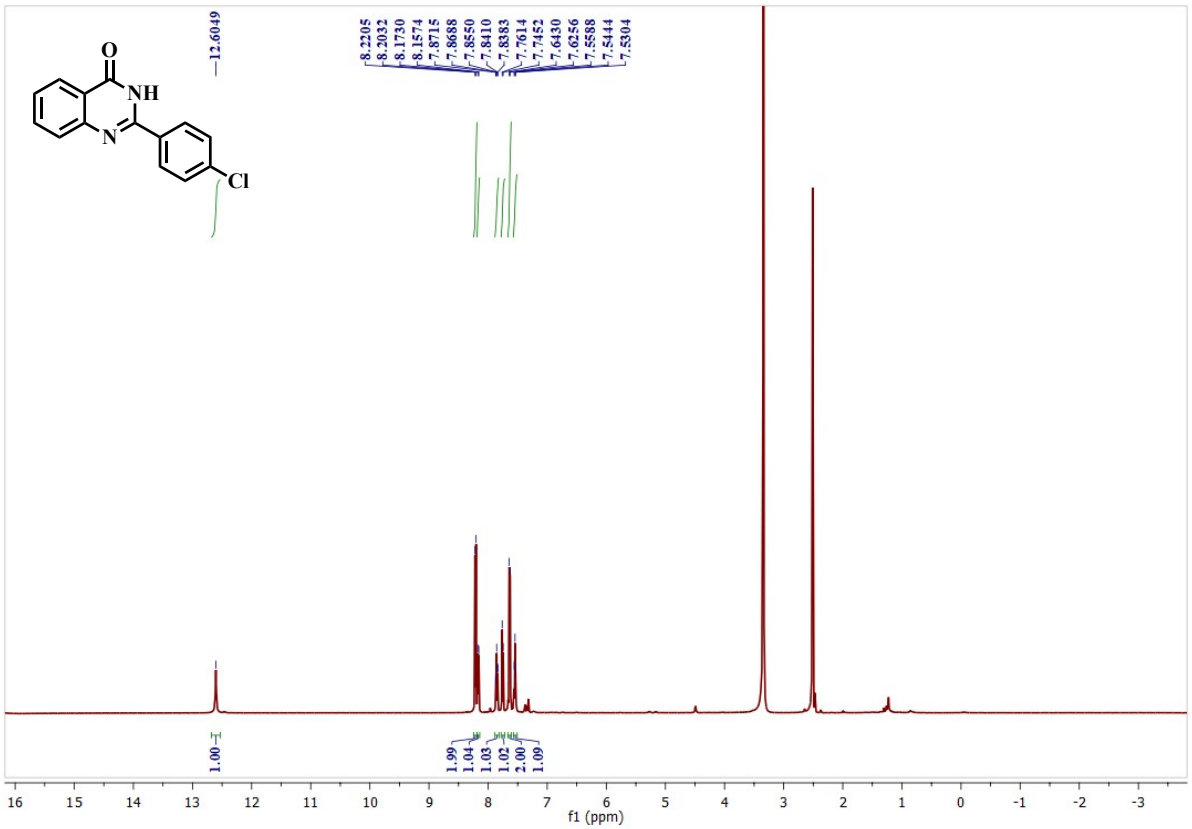


Fig. S36. ^{1}H NMR spectrum of **5e**.

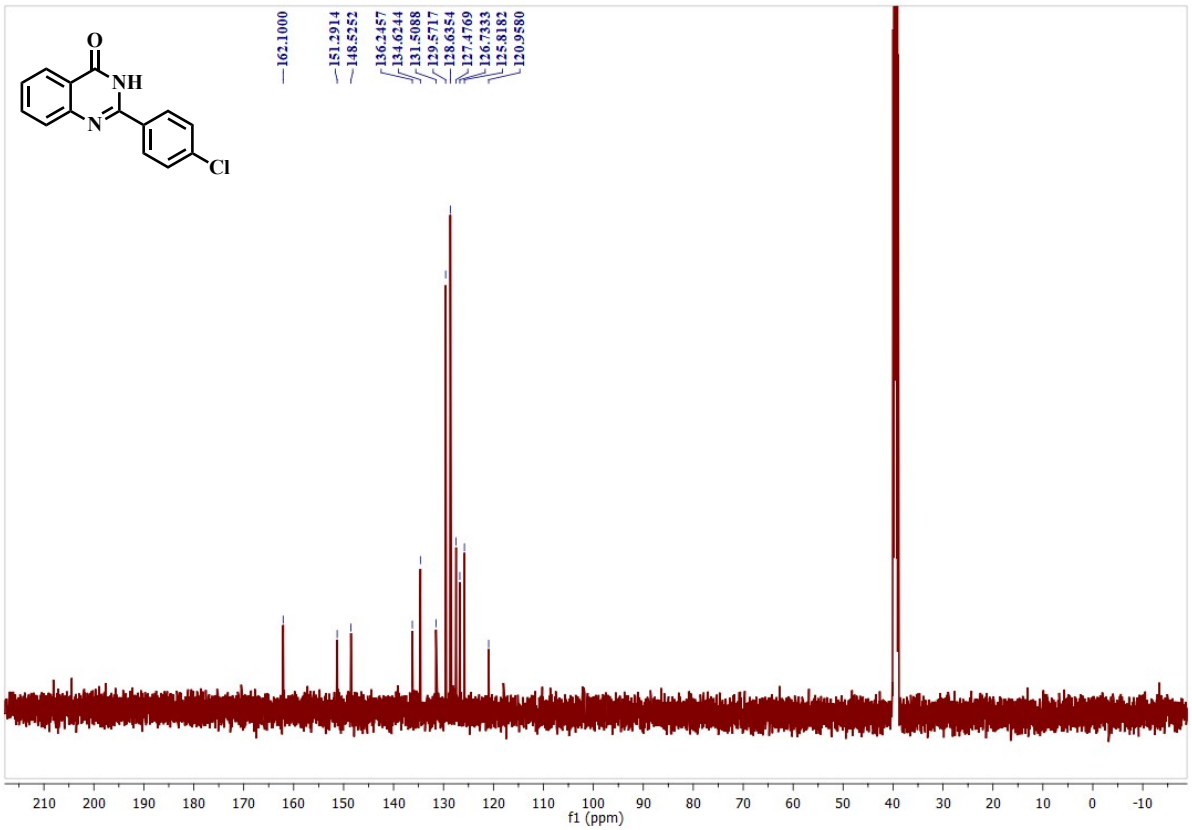


Fig. S37. ^{13}C NMR spectrum of **5e**.

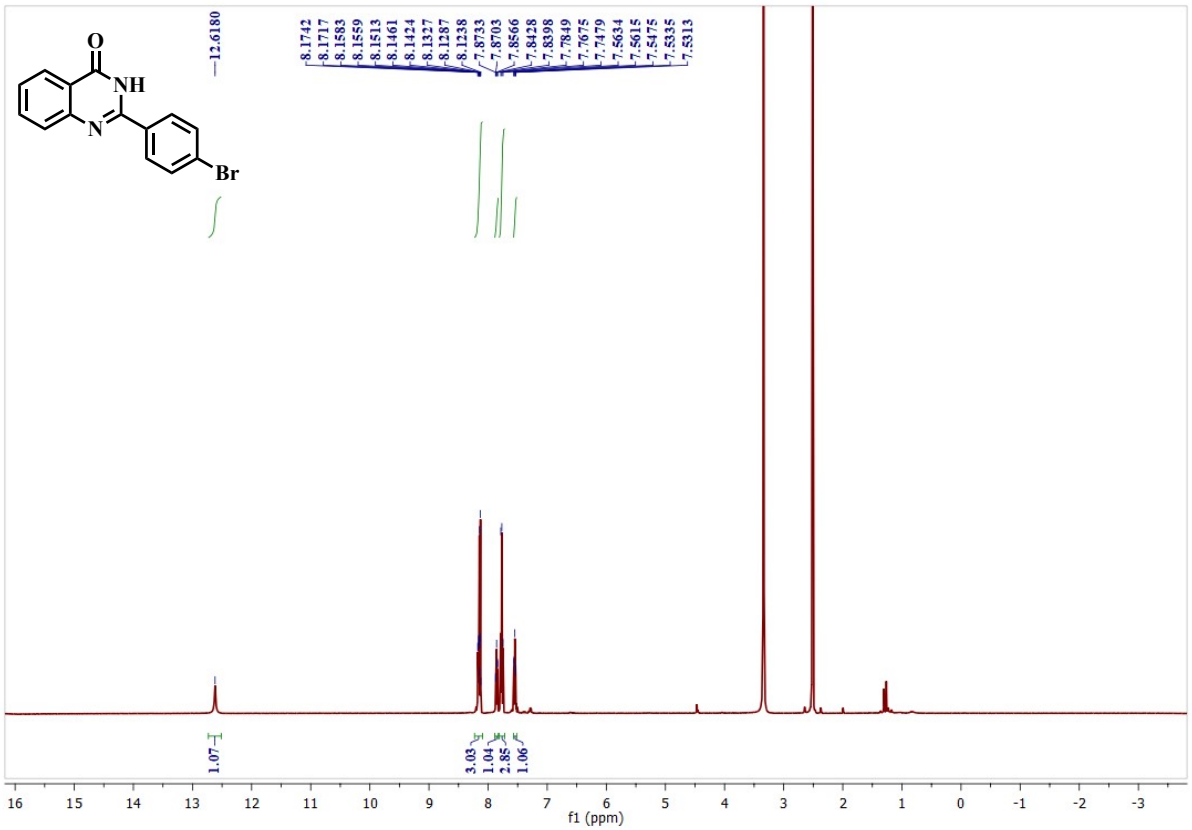


Fig. S38. ^{1}H NMR spectrum of **5f**.

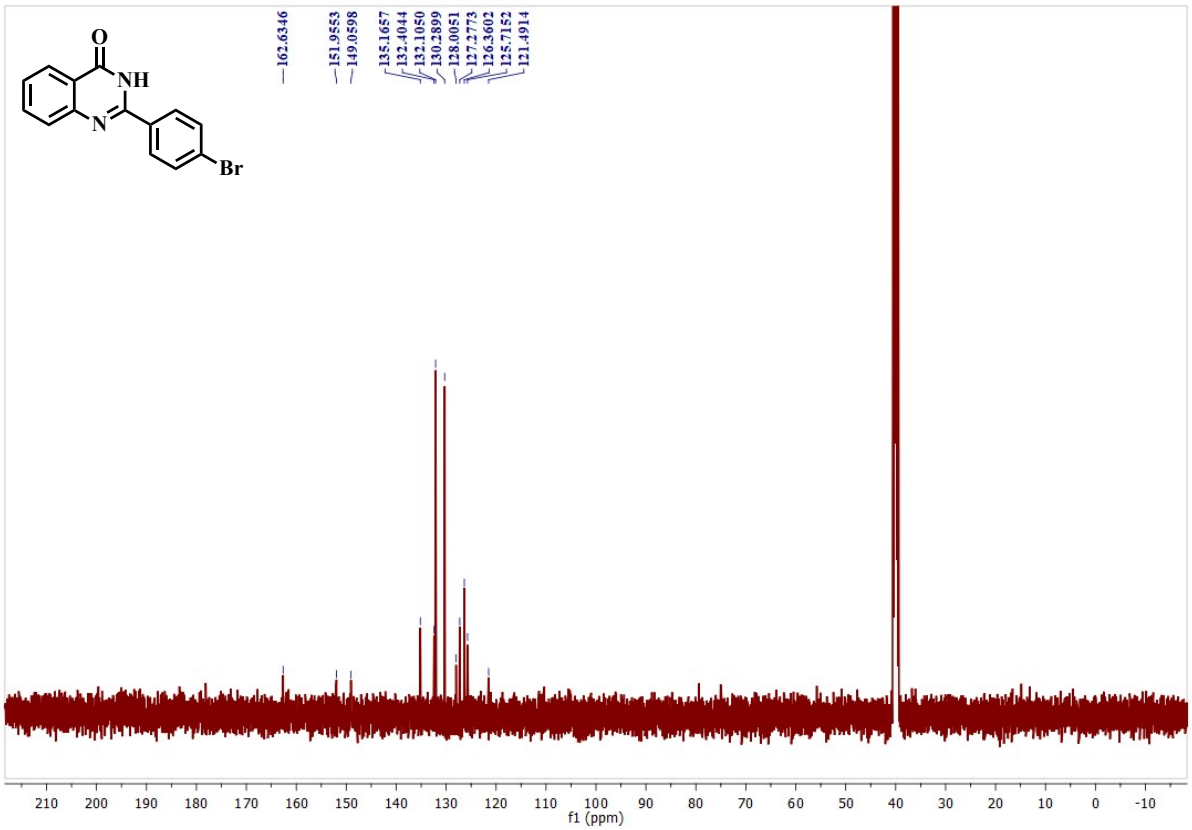


Fig. S39. ^{13}C NMR spectrum of **5f**.

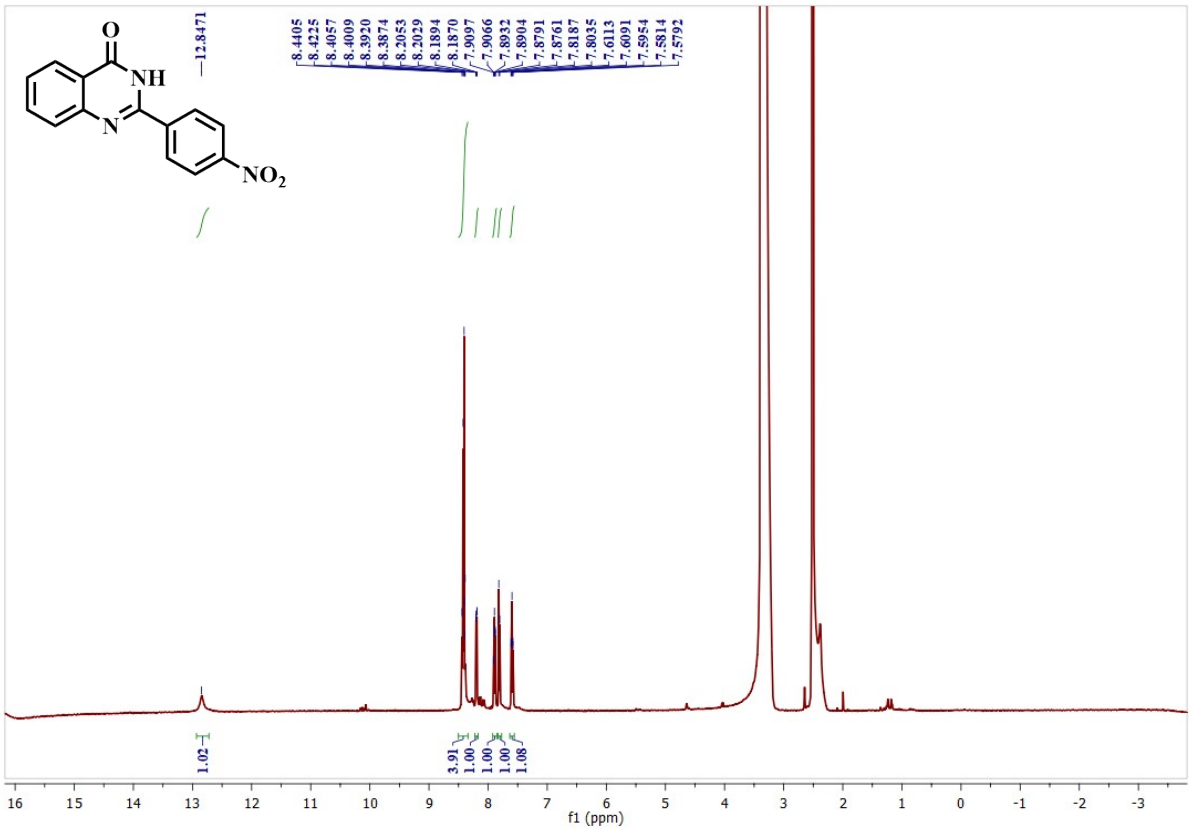


Fig. S40. ^{1}H NMR spectrum of **5g**.

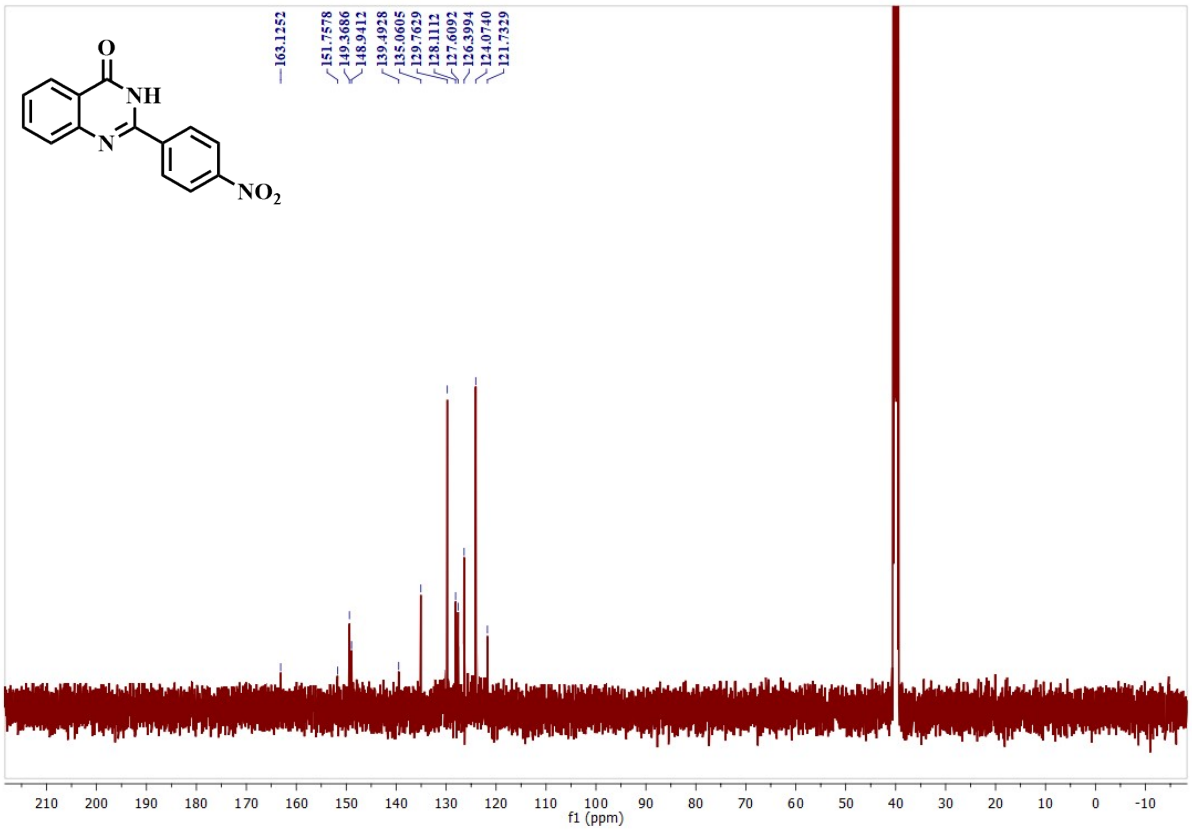


Fig. S41. ^{13}C NMR spectrum of **5g**.

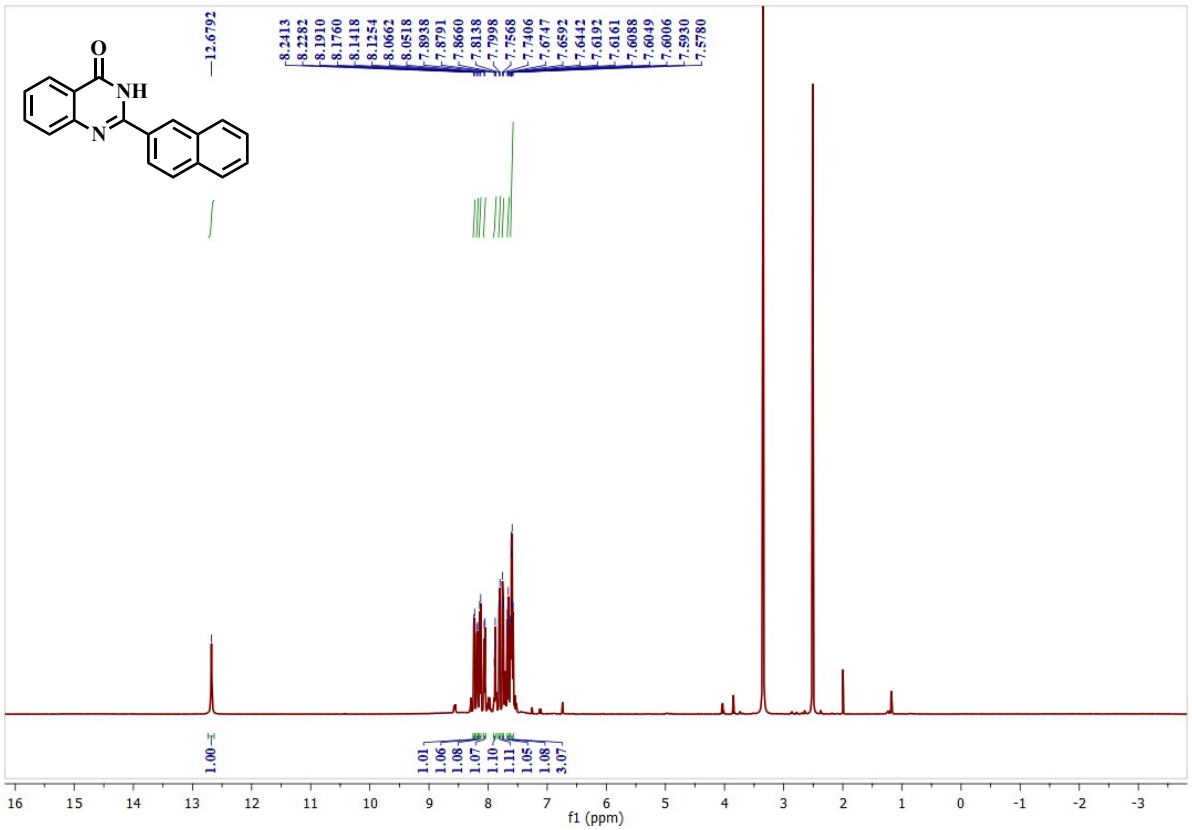


Fig. S42. ^{1}H NMR spectrum of **5h**.

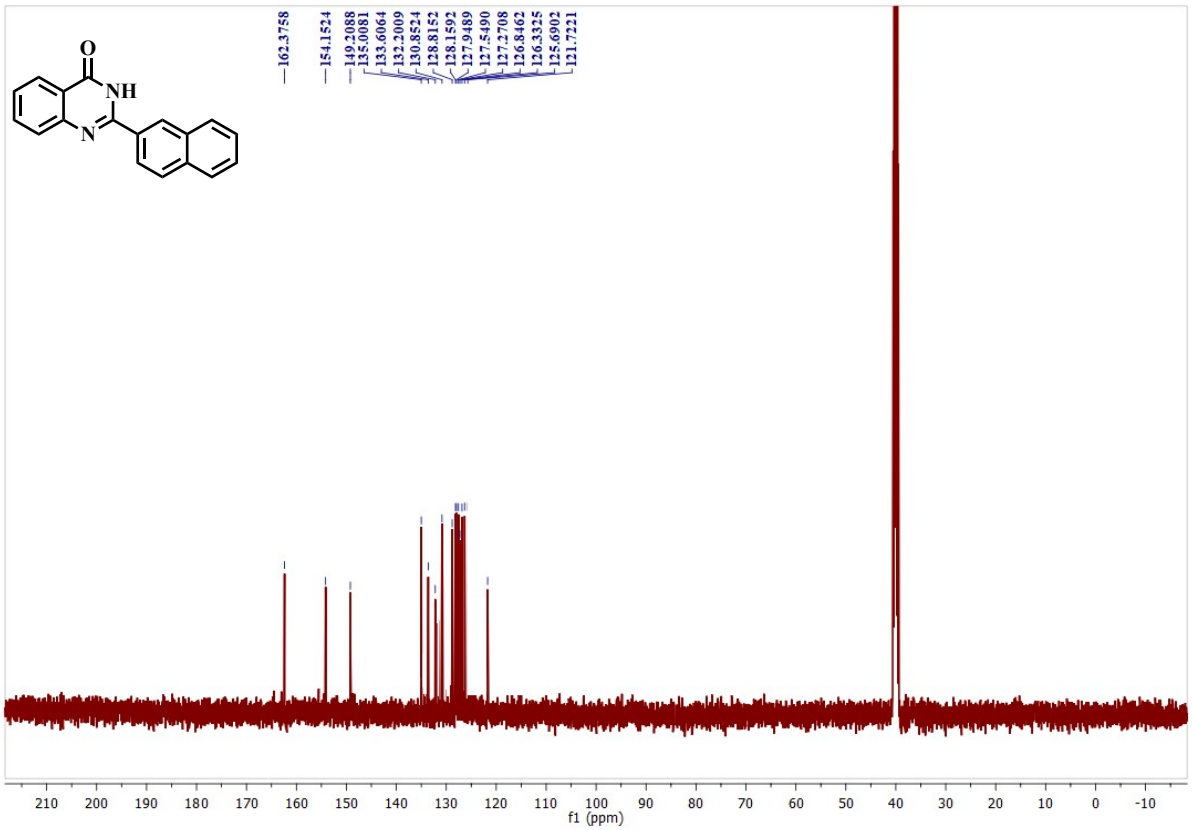


Fig. S43. ^{13}C NMR spectrum of **5h**.

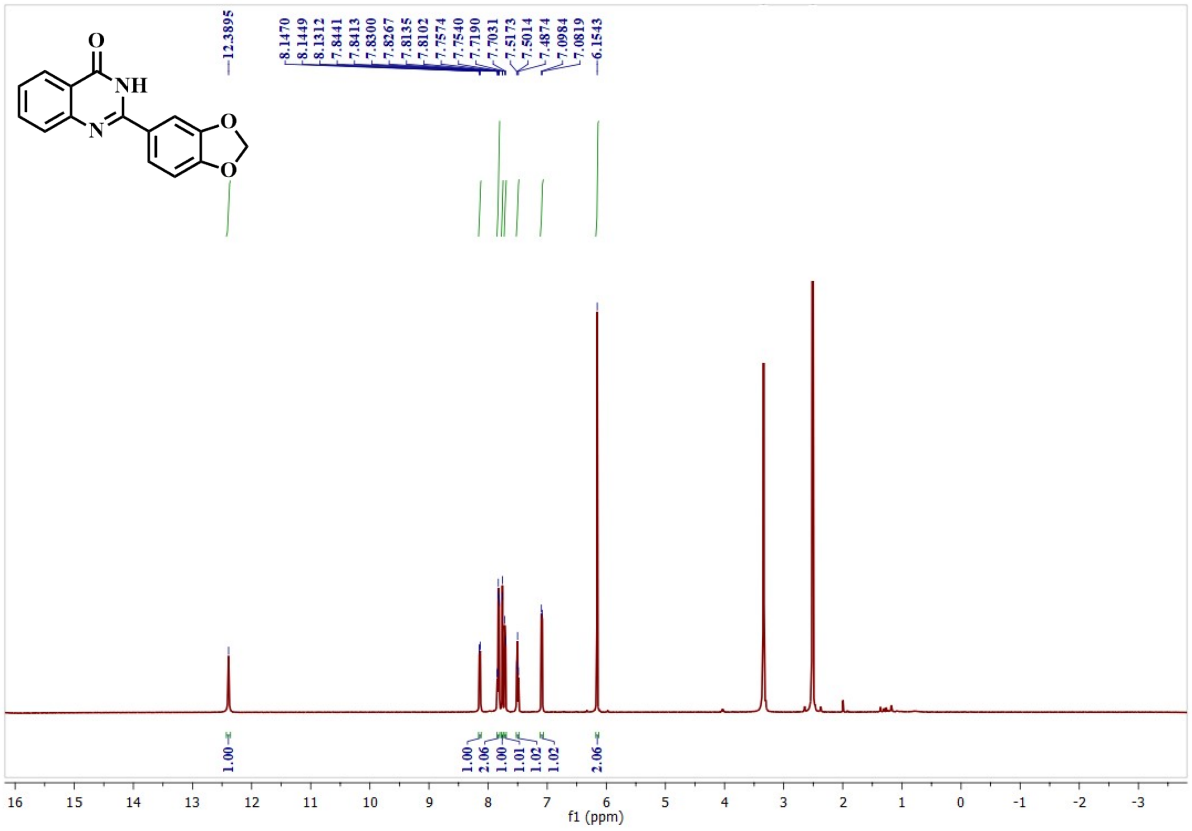


Fig. S44. ^{1}H NMR spectrum of **5i**.

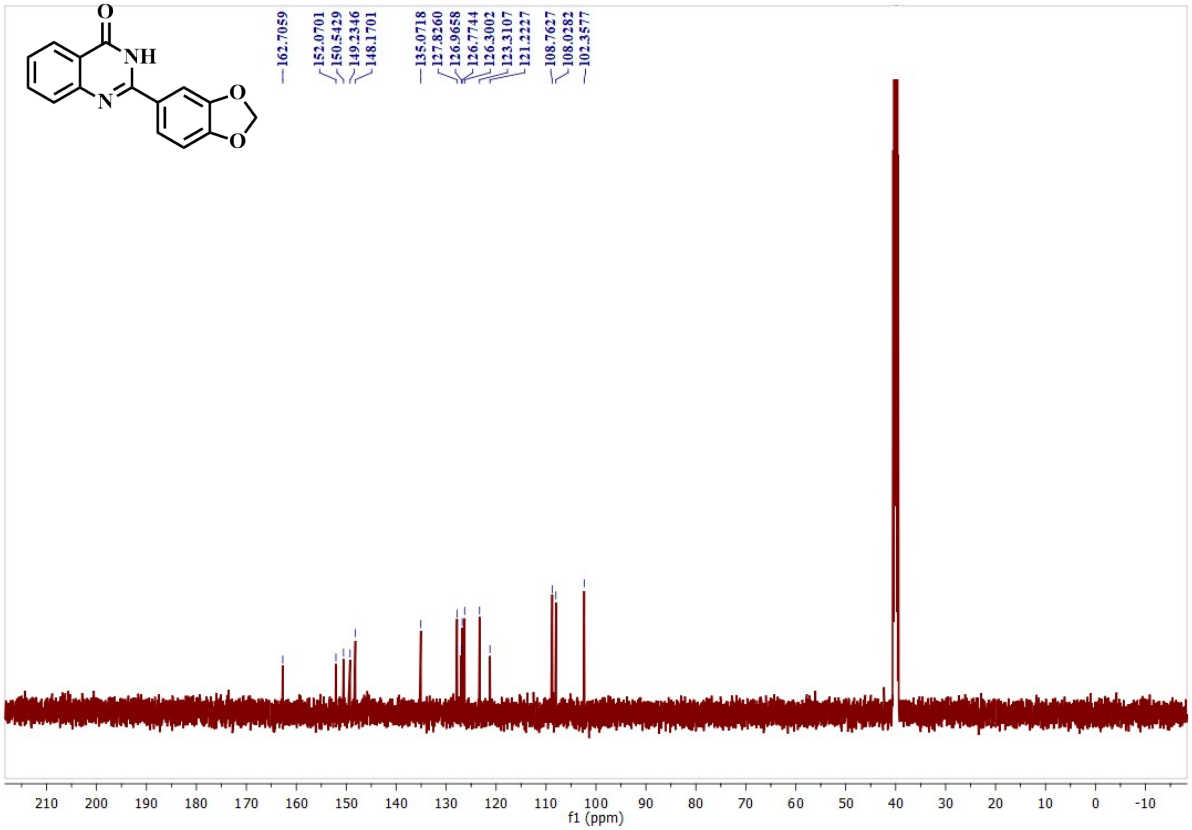


Fig. S45. ^{13}C NMR spectrum of **5i**.

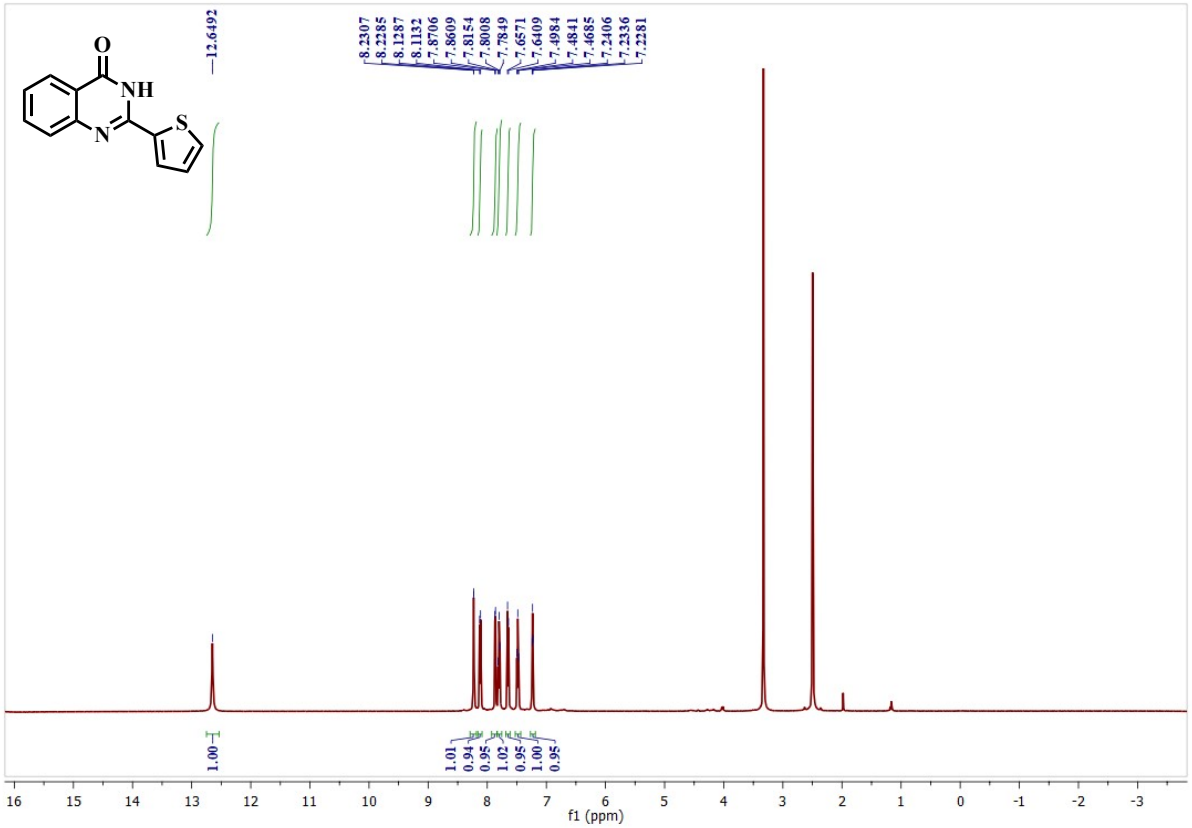


Fig. S46. ^{1}H NMR spectrum of **5j**.

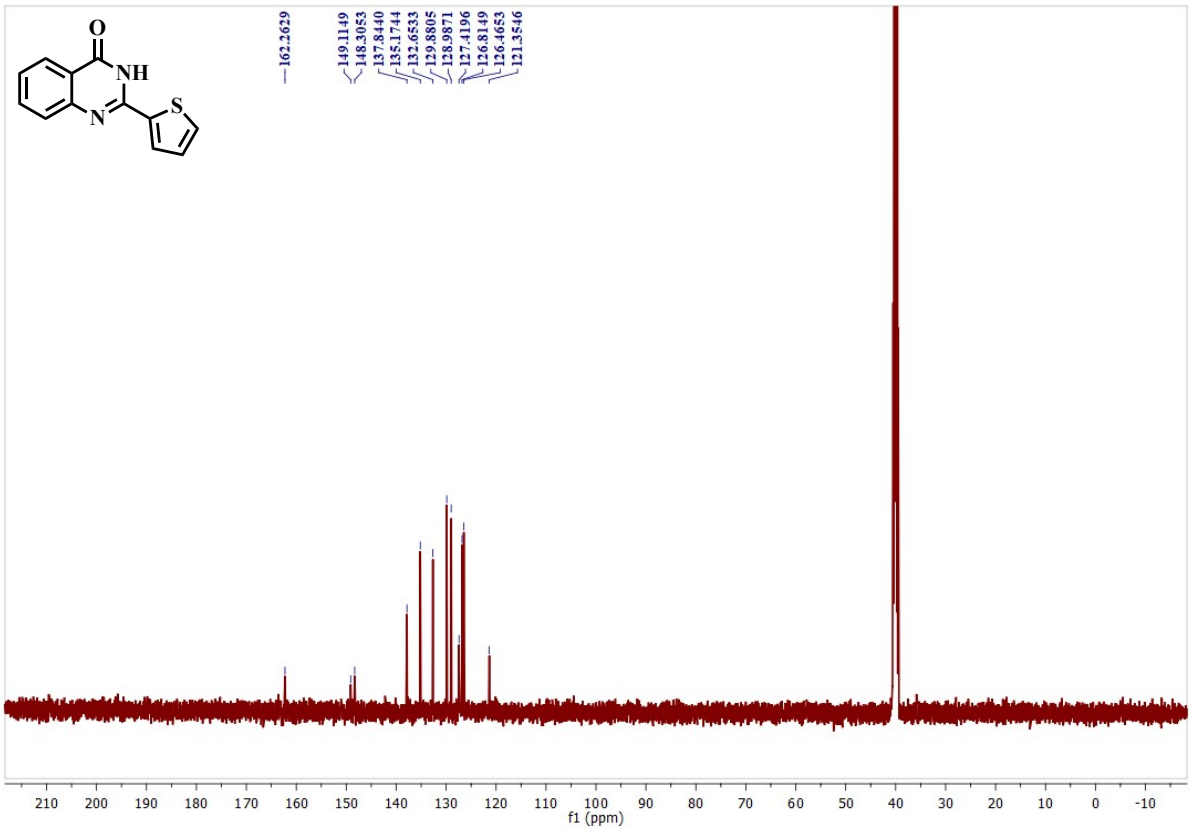


Fig. S47. ^{13}C NMR spectrum of **5j**.

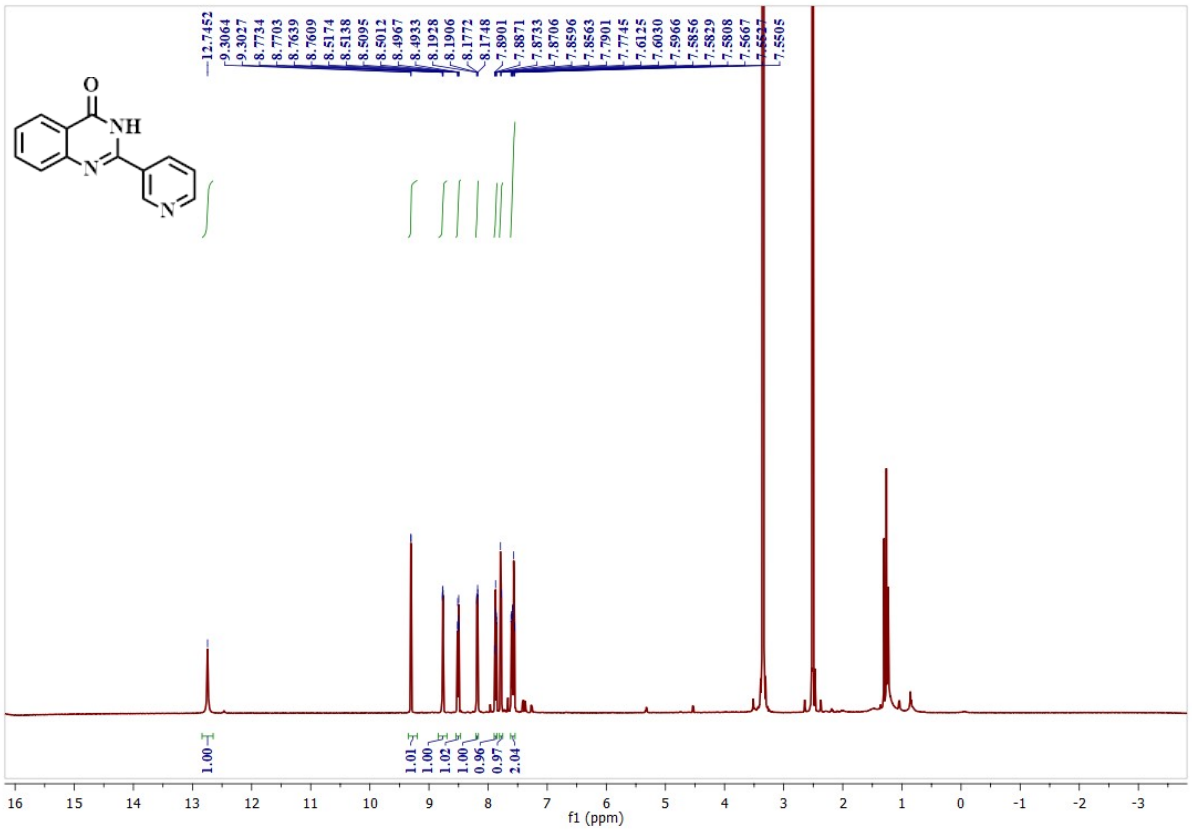


Fig. S48. ^{1}H NMR spectrum of **5k**.

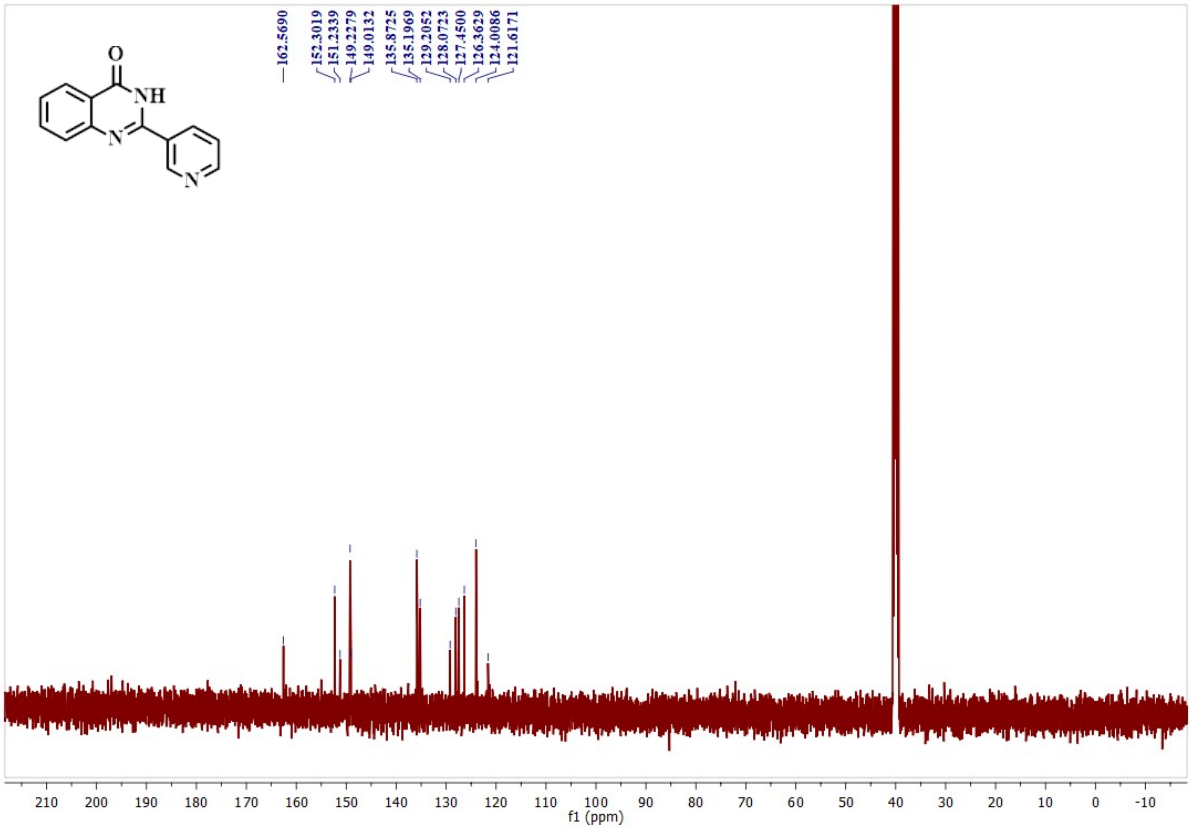


Fig. S49. ^{13}C NMR spectrum of **5k**.

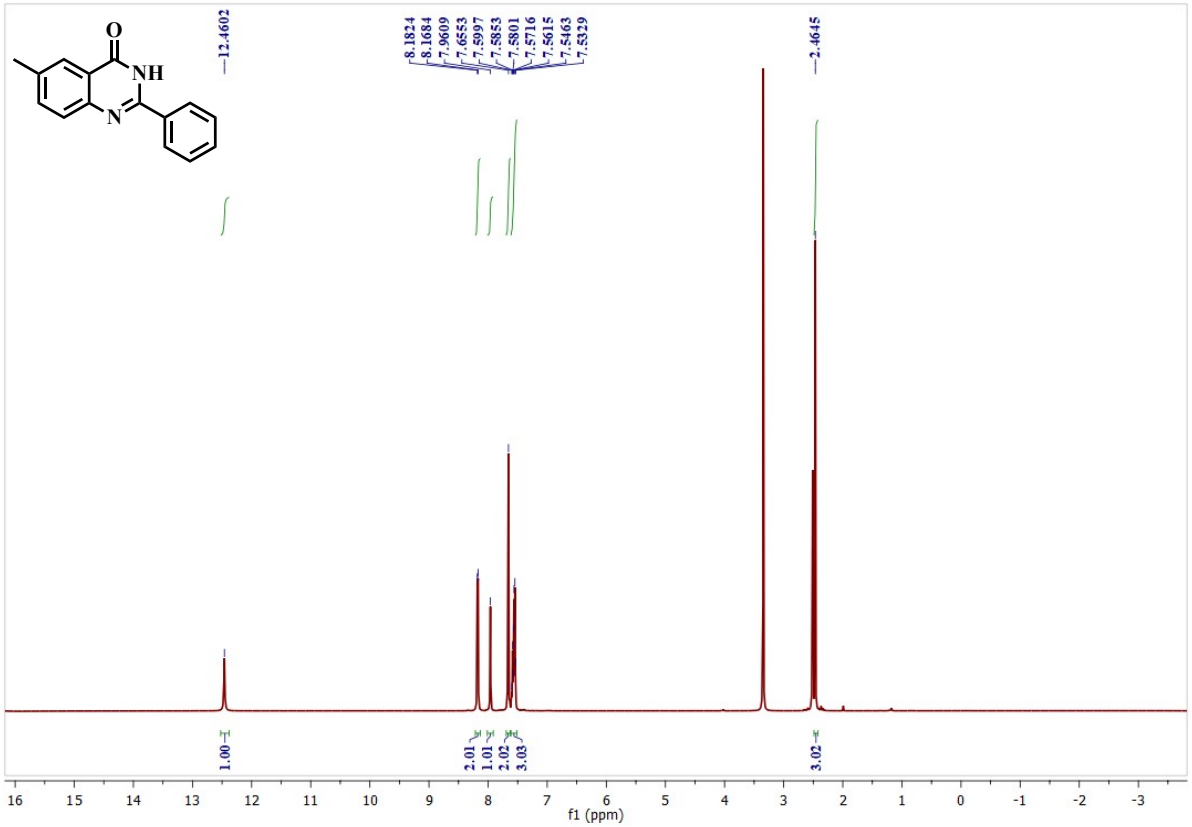


Fig. S50. ^{1}H NMR spectrum of **5m**.

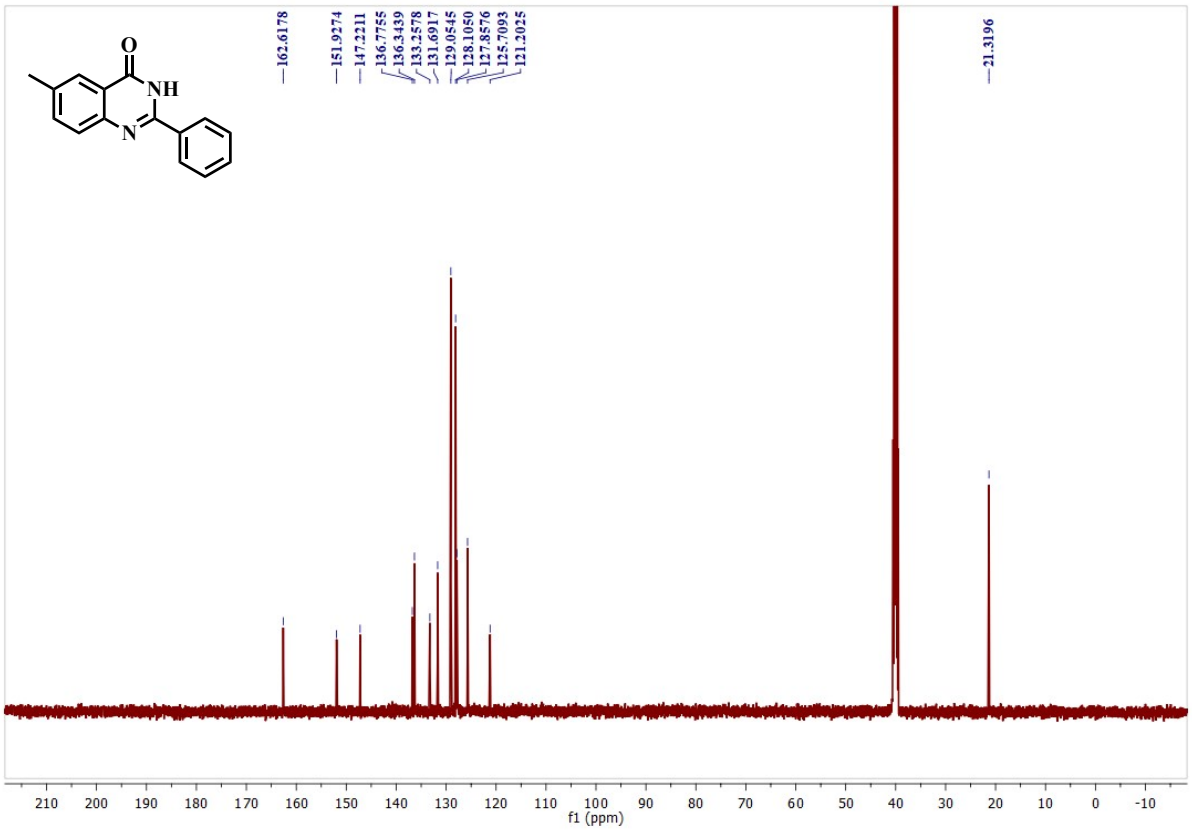


Fig. S51. ^{13}C NMR spectrum of **5m**.

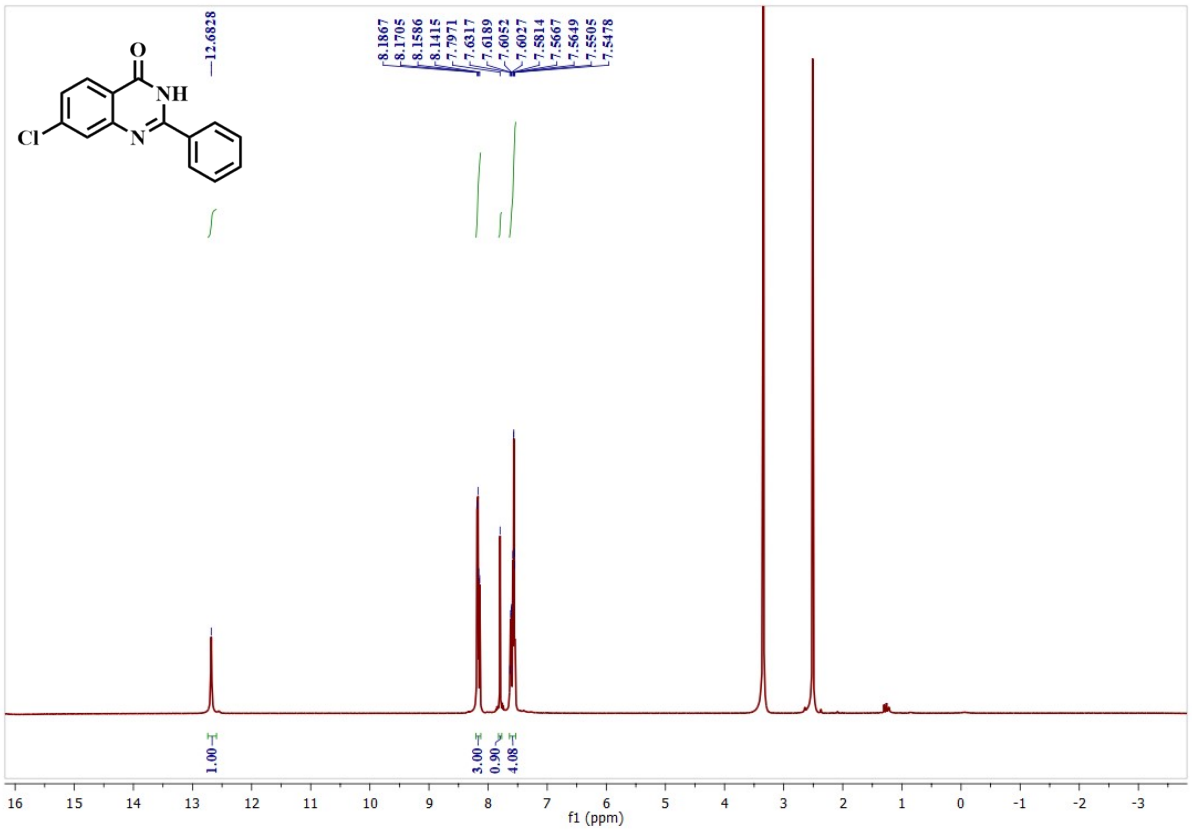


Fig. S52. ^{1}H NMR spectrum of **5n**.

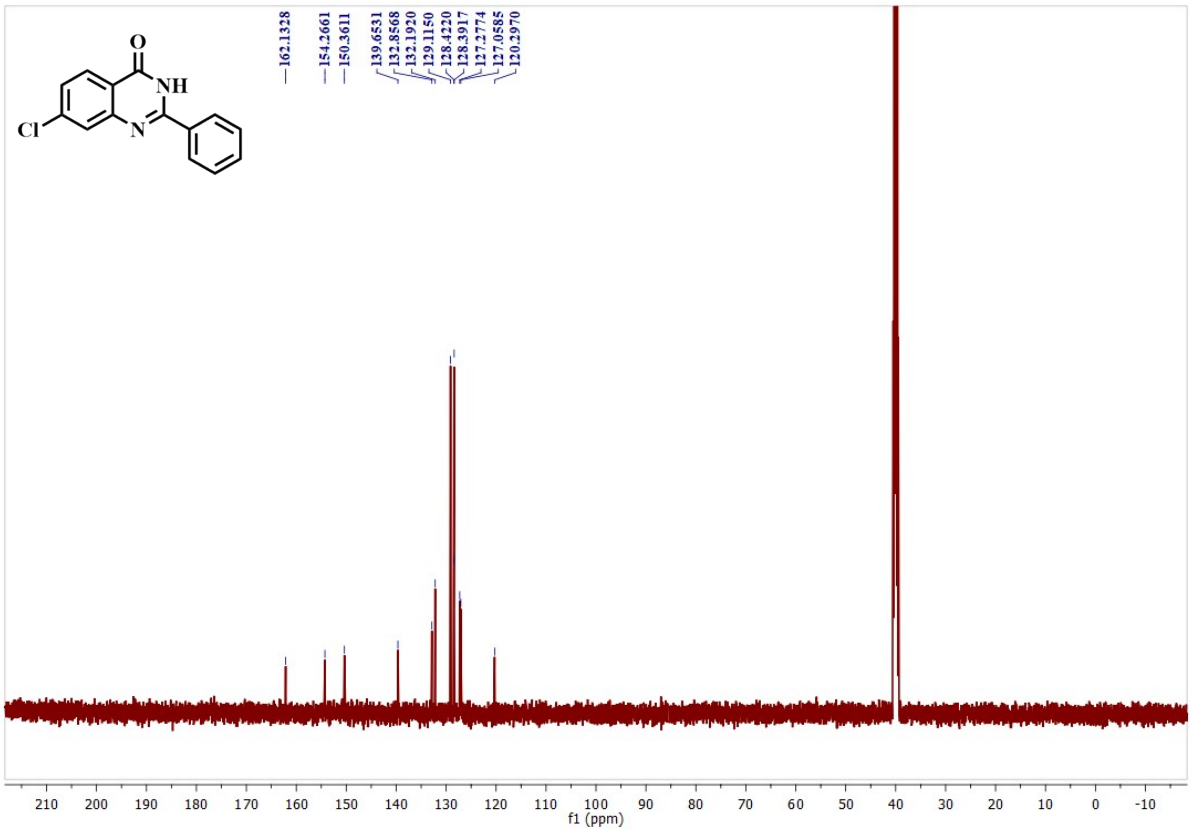


Fig. S53. ^{13}C NMR spectrum of **5n**.

12. PXRD and SAED patterns of the recycled catalysts

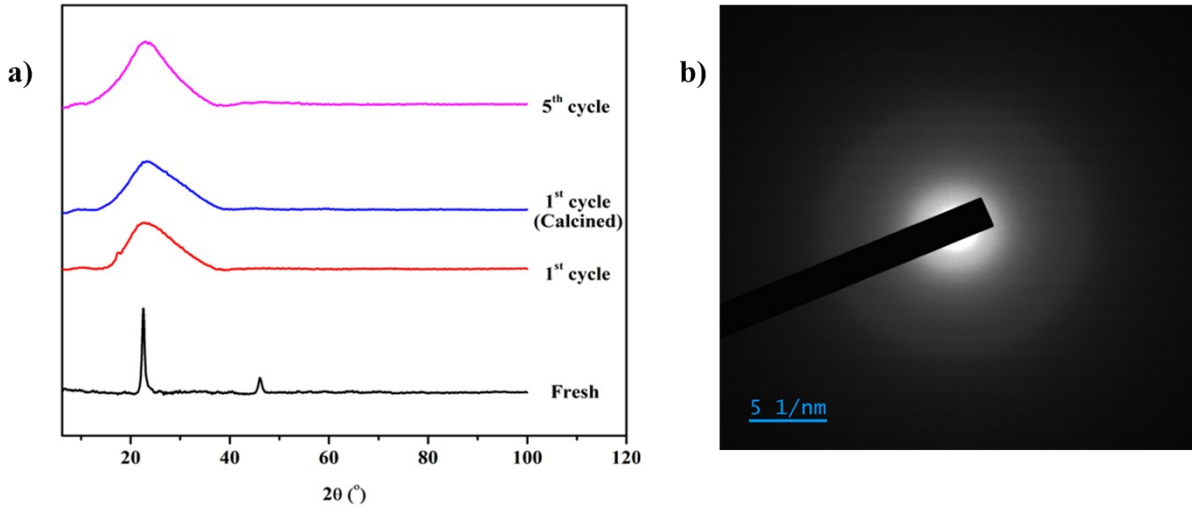


Fig. S54. a) PXRD patterns of the recycled catalysts, and b) SAED pattern of the recycled catalyst (after 1st cycle).

13. UV-Vis spectrum showing absorption changes during the formation of I_3^- in the presence of H_2O_2

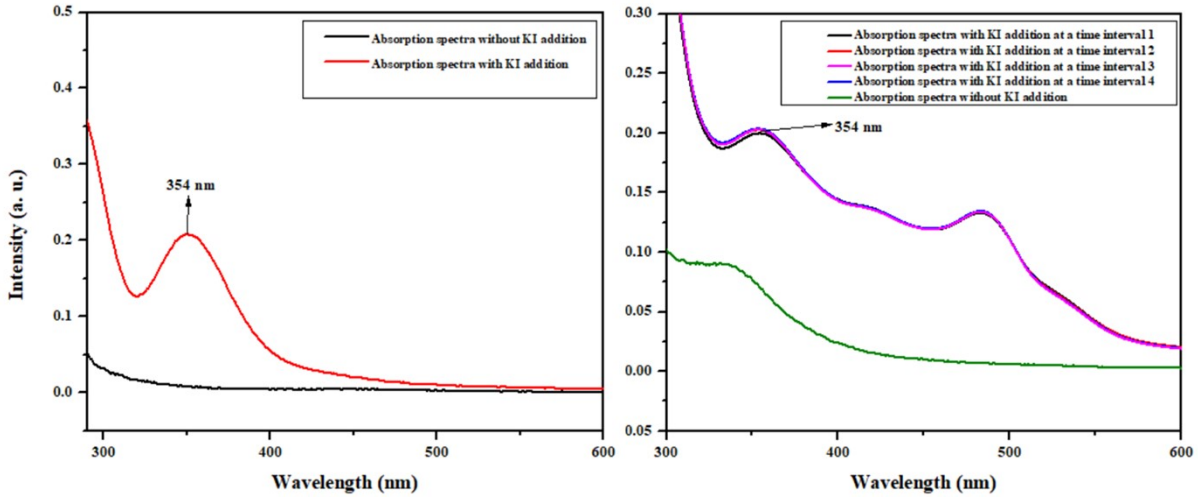


Fig. S55. Detection of H_2O_2. a) Absorption spectral changes during the formation of I_3^- due to hydrogen peroxide when aqueous medium obtained from the conversion of benzyl alcohol to benzaldehyde was analyzed b) Absorption spectral changes during the formation of I_3^- due to hydrogen peroxide when aqueous medium obtained from the synthesis of 2-substituted benzimidazole was analyzed.

14. ^{1}H NMR spectra of the crude mixtures

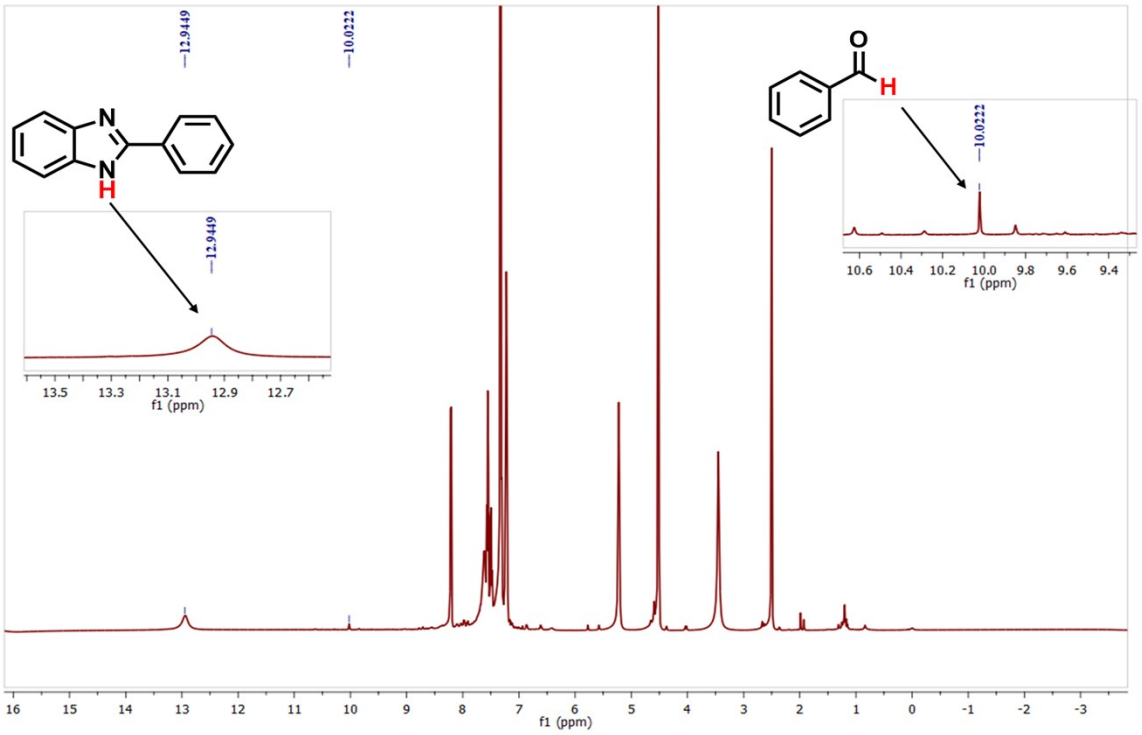


Fig. S56. ^{1}H NMR spectrum of the crude mixture obtained after 8 h of the benzimidazole synthesis reaction.

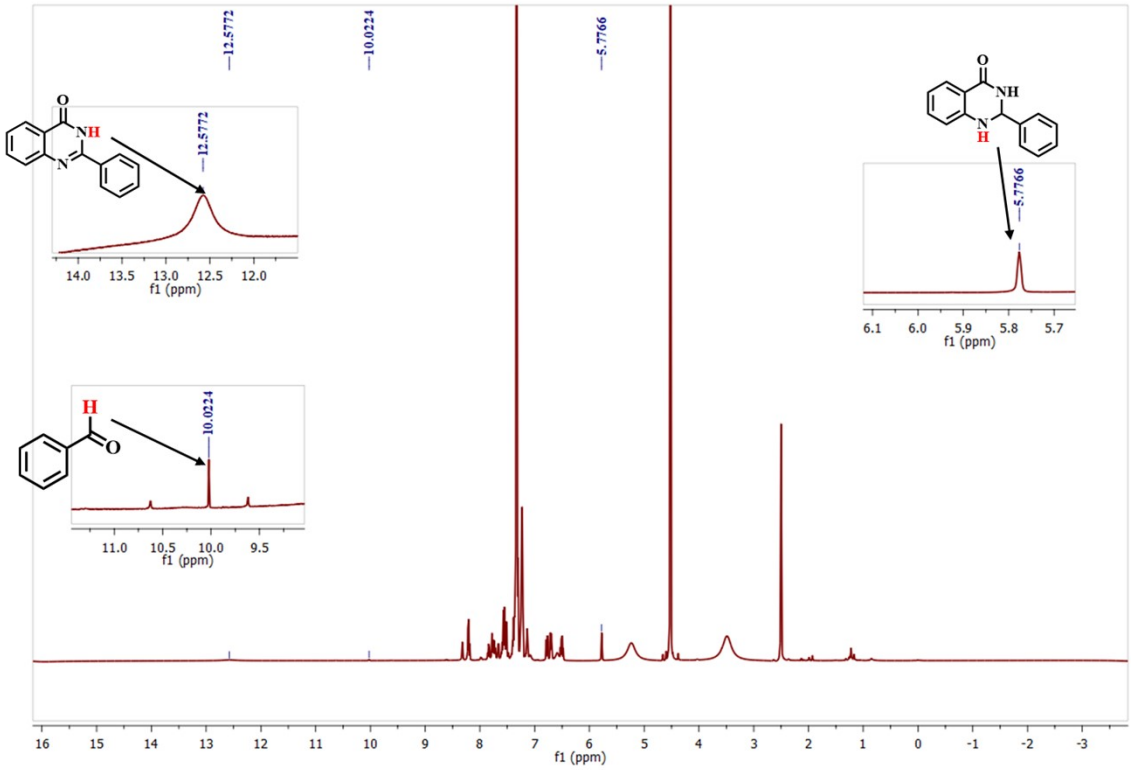


Fig. S57. ^{1}H NMR spectrum of the crude mixture obtained after 8 h of the quinazolinone synthesis reaction.

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