

Supporting Information (SI)

Silver(I)-Catalyzed Tandem Reaction of Enynones and 4-Alkynyl Isoxazoles: Regioselective Synthesis of Highly Functionalized 4*H*-furan[3,4-*c*]pyrroles

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Content

Experimental section	S2
The X-Ray Crystal Structure of 3b	S14
¹H and ¹³C NMR spectra of new compounds	S16

Experimental section

General Methods.

All reactions were carried out in a round flask with magnetic stirring. Unless otherwise noted, all reagents were purchased from Aladdin for direct use, or prepared as described in literature. DCM, DCE was dried over CaH before use. For chromatographic purification, 200-300 mesh silica gel (Qingdao, China) was employed. For thin layer chromatography (TLC) analysis, Merck 25 TLC aluminium sheets (silica gel 60 GF254, 0.25 mm) were used. ¹H NMR and ¹³C NMR were recorded on Bruker ARX 500 MHz spectrometer in CDCl₃ solution. The chemical shift was reported in parts per million (δ) relative to internal standard TMS (0 ppm), and coupling constants are reported as Hertz (Hz). Splitting patterns are designated as singlet (s), broad singlet (bs), doublet (d), triplet (t). Splitting patterns that could not be interpreted or easily visualized are designated as multiple (m). HRMS were performed on Thermo Scientific LTQ Qorbitrap XL mass spectrometer (ESI). Enynones **1** were known compounds and prepared according to literature procedures.

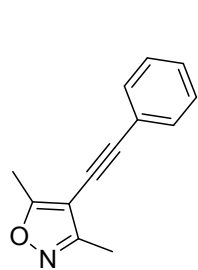
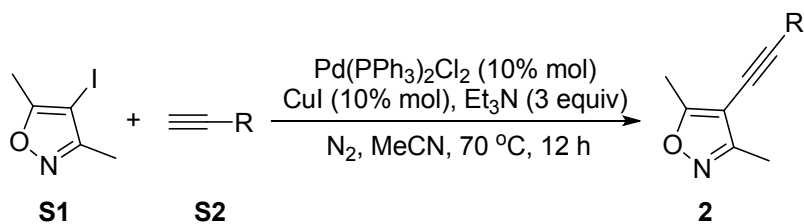
General procedure for the Ag(I)-catalyzed tandem reaction.

Synthesis of 3a. 3-(3-(4-chlorophenyl)prop-2-yn-1-ylidene)pentane-2,4-dione **1a** (0.2 mmol, 49 mg) was dissolved in DCE (2 mL). The solution was syringed dropwise into a vigorous stirring mixture of AgOTf (0.02 mmol, 5 mg) and 3,5-dimethyl-4-(phenylethynyl)isoxazole **2a** (0.24 mmol, 47 mg) in DCE (1 mL) at -20 °C under nitrogen atmosphere. After stirring for 1 hour, the reaction mixture was heated at 60 °C for 12 hours. Finally, the reaction mixture was concentrated in vacuum, and the residue was purified by flash chromatography (eluted with petroleum ether/ethyl acetate = 3:1), providing product **3a**.

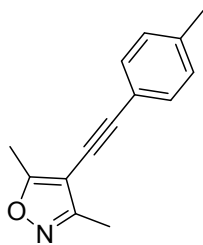
1 mmol scale for one representative example: Synthesis of 3a. The solution of 3-(3-(4-chlorophenyl)prop-2-yn-1-ylidene)pentane-2,4-dione **1a** (1 mmol, 246 mg) in DCE (5 mL) was syringed dropwise into a vigorous stirring mixture of AgOTf (0.1 mmol, 25 mg) and 3,5-dimethyl-4-(phenylethynyl)isoxazole **2a** (1.2 mmol, 237 mg) in DCE (3 mL) at -20 °C under nitrogen atmosphere. The mixture was stirred for two hours at -20 °C, and then at 60 °C for another 24 hours. Following this, the reaction mixture was concentrated in vacuum. The residue was purified by flash chromatography (eluted with petroleum ether/ethyl acetate = 3:1), affording product **3a** (306 mg, 70%).

Preparation of the substrates 2.

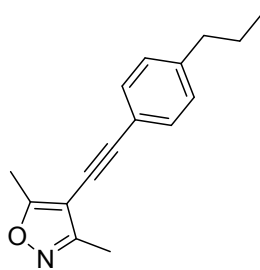
A mixture of 4-iodo-3,5-dimethylisoxazole **S1** (5 mmol, 1.11 g), acetylene **S2** (6 mmol), Pd(PPh₃)₂Cl₂ (0.5 mmol, 350 mg), CuI (0.5 mmol, 95 mg) and Et₃N (15 mmol, 1.5 g) in MeCN (10 ml) was stirred at 70 °C for 12 hours. The concentration of the reaction mixture in vacuum gave a residue. The purification of the residue by using flash chromatography (eluted with petroleum ether/ethyl acetate = 20:1 to 10 :1) afforded products **2**.



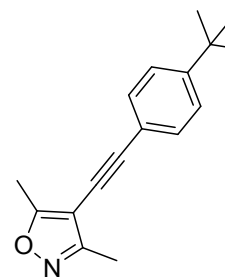
2a, 80%



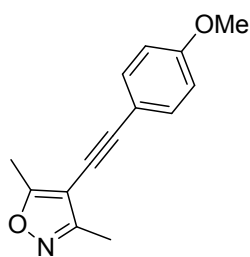
2b, 82%



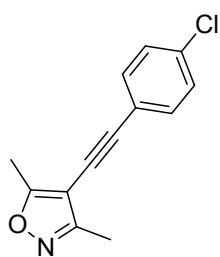
2c, 83%



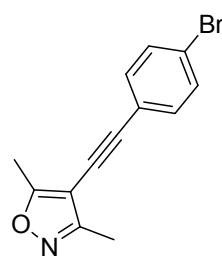
2d, 76%



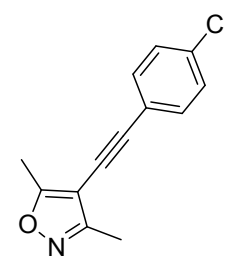
2e, 81%



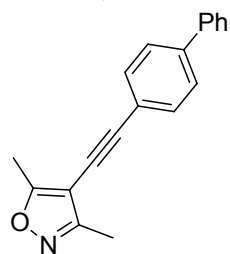
2f, 83%



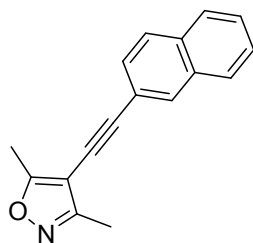
2g, 78%



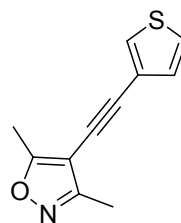
2h, 68%



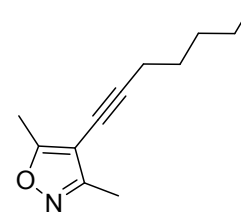
2i, 70%



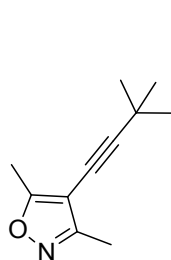
2j, 85%



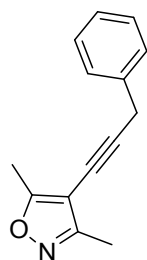
2k, 80%



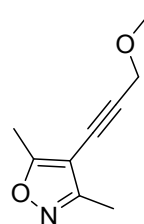
2l, 86%



2m, 84%

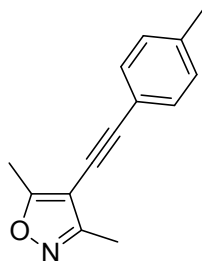


2n, 88%



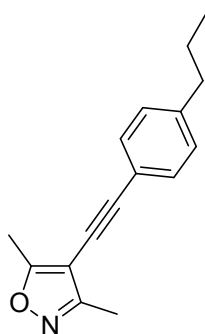
2o, 59%

3,5-dimethyl-4-(p-tolylethynyl)isoxazole (2b). Brown solid (846 mg, 82%), $R_f = 0.46$



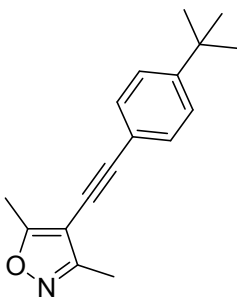
(PE : EA = 10 : 1), mp 78-79 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.41 (d, $J = 8.0$ Hz, 2H), 7.15 (d, $J = 7.5$ Hz, 2H), 2.47 (s, 3H), 2.36 (s, 3H), 2.34 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 170.8, 160.5, 138.7, 131.4, 129.2, 119.8, 101.3, 94.5, 76.6, 21.4, 11.9, 10.5. HRMS (ESI) [$\text{C}_{14}\text{H}_{13}\text{NO} + \text{H}$] $^+$ m/z calcd for 212.1070, found 212.1076.

3,5-dimethyl-4-((4-propylphenyl)ethynyl)isoxazole (2c). Brown oil (996 mg, 83%),



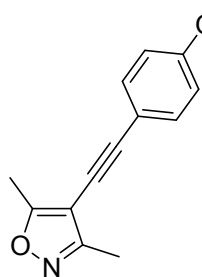
$R_f = 0.50$ (PE : EA = 10 : 1). $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.43 (d, $J = 7.5$ Hz, 2H), 7.16 (d, $J = 8.0$ Hz, 2H), 2.60 (t, $J = 7.5$ Hz, 2H), 2.48 (s, 3H), 2.34 (s, 3H), 1.69-1.61 (m, 2H), 0.96 (t, $J = 7.5$ Hz, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 170.8, 160.5, 143.5, 131.4, 128.6, 120.0, 101.3, 94.5, 76.6, 37.9, 24.3, 13.7, 11.9, 10.5. HRMS (ESI) [$\text{C}_{16}\text{H}_{17}\text{NO} + \text{H}$] $^+$ m/z calcd for 240.1383, found 240.1394.

4-((4-(tert-butyl)phenyl)ethynyl)-3,5-dimethylisoxazole (2d). Yellow solid (966 mg,



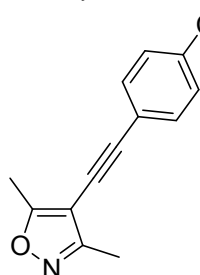
76%), $R_f = 0.48$ (PE : EA = 10 : 1), mp 98-100 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.48 (d, $J = 8.0$ Hz, 2H), 7.41 (d, $J = 8.5$ Hz, 2H), 2.51 (s, 3H), 2.37 (s, 3H), 1.36 (s, 9H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 170.9, 160.6, 151.9, 131.3, 125.5, 119.8, 101.4, 94.4, 76.6, 34.8, 31.2, 12.0, 10.6. HRMS (ESI) [$\text{C}_{17}\text{H}_{19}\text{NO} + \text{H}$] $^+$ m/z calcd for 254.1539, found 254.1547.

4-((4-methoxyphenyl)ethynyl)-3,5-dimethylisoxazole (2e). Yellow solid (923 mg,



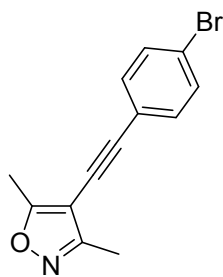
81%), $R_f = 0.39$ (PE : EA = 10 : 1), mp 74-76 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.45 (d, $J = 9.0$ Hz, 2H), 6.88 (d, $J = 9.0$ Hz, 2H), 3.82 (s, 3H), 2.48 (s, 3H), 2.34 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 170.7, 160.6, 159.9, 133.0, 114.8, 114.1, 101.4, 94.2, 75.8, 55.3, 11.9, 10.5. HRMS (ESI) [$\text{C}_{14}\text{H}_{13}\text{NO}_2 + \text{H}$] $^+$ m/z calcd for 228.1019, found 228.1025.

4-((4-chlorophenyl)ethynyl)-3,5-dimethylisoxazole (2f). Yellow solid (962 mg, 83%), $R_f = 0.44$ (PE : EA = 10 : 1), mp 85-87 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ



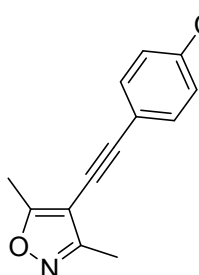
7.45-7.42 (m, 2H), 7.35-7.32 (m, 2H), 2.50 (s, 3H), 2.35 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 171.3, 160.5, 134.6, 132.7, 128.8, 121.3, 100.9, 93.2, 78.3, 12.0, 10.5. HRMS (ESI) [$\text{C}_{13}\text{H}_{10}\text{ClNO} + \text{H}$] $^+$ m/z calcd for 232.0524, found 232.0532.

4-((4-bromophenyl)ethynyl)-3,5-dimethylisoxazole (2g). Brown solid (1.07 g, 78%),



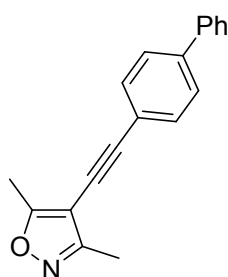
$R_f = 0.42$ (PE : EA = 10 : 1), mp 110-112 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.51-7.49 (m, 2H), 7.38-7.36 (m, 2H), 2.51 (s, 3H), 2.35 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 171.4, 160.5, 132.9, 131.7, 122.8, 121.7, 100.9, 93.3, 78.5, 12.1, 10.6. HRMS (ESI) [$\text{C}_{13}\text{H}_{10}\text{BrNO} + \text{H}$] $^+$ m/z calcd for 276.0019, found 276.0024.

4-((3,5-dimethylisoxazol-4-yl)ethynyl)benzotrile (2h). Yellow solid (759 mg,



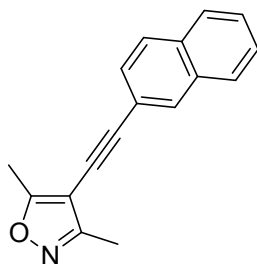
68%), $R_f = 0.31$ (PE : EA = 8 : 1), mp 118-120 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.60 (d, $J = 8.0$ Hz, 2H), 7.54 (d, $J = 8.5$ Hz, 2H), 2.48 (s, 3H), 2.31 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 172.1, 160.4, 132.1, 131.9, 127.6, 118.4, 111.7, 100.5, 92.8, 81.9, 12.1, 10.5. HRMS (ESI) [$\text{C}_{14}\text{H}_{10}\text{N}_2\text{O} + \text{H}$] $^+$ m/z calcd for 223.0886, found 223.0875.

4-([1,1'-biphenyl]-4-ylethynyl)-3,5-dimethylisoxazole (2i). Yellow solid (956 mg,

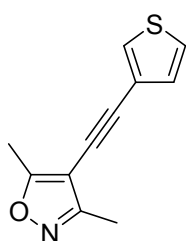


70%), $R_f = 0.44$ (PE : EA = 8 : 1), mp 138-140 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.66-7.61 (m, 6H), 7.52-7.48 (m, 2H), 7.43-7.40 (m, 2H), 2.55 (s, 3H), 2.41 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 171.2, 160.6, 141.3, 140.2, 132.0, 129.0, 127.8, 127.1, 127.1, 121.7, 101.3, 94.3, 78.0, 12.1, 10.6. HRMS (ESI) [$\text{C}_{19}\text{H}_{15}\text{NO} + \text{H}$] $^+$ m/z calcd for 274.1226, found 274.1237.

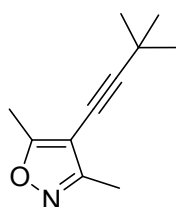
3,5-dimethyl-4-(naphthalen-2-ylethynyl)isoxazole (2j). Yellow solid (1.05 g, 85%), $R_f = 0.41$ (PE : EA = 10 : 1), mp 92-94 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 8.07 (s, 1H), 7.87-7.84 (m, 3H), 7.60-7.54 (m, 3H), 2.56 (s, 3H), 2.43 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 171.2, 160.7, 133.0, 132.9, 131.4, 128.2, 127.9, 127.8, 126.9, 126.8, 120.1, 101.3, 94.8, 77.7, 12.1, 10.7. HRMS (ESI) $[\text{C}_{17}\text{H}_{13}\text{NO} + \text{H}]^+$ m/z calcd for 248.1070, found 248.1078.



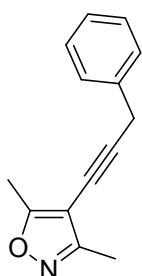
3,5-dimethyl-4-(thiophen-3-ylethynyl)isoxazole (2k). Brown solid (816 mg, 80%), $R_f = 0.44$ (PE : EA = 10 : 1), mp 85-86 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.54-7.53 (m, 1H), 7.34-7.32 (m, 1H), 7.20 (d, $J = 6.0$ Hz, 3H), 2.50 (s, 3H), 2.35 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 171.1, 160.6, 129.8, 128.9, 125.6, 121.8, 101.1, 89.3, 76.8, 12.0, 10.6. HRMS (ESI) $[\text{C}_{11}\text{H}_9\text{NOS} + \text{H}]^+$ m/z calcd for 204.0478, found 204.0483.



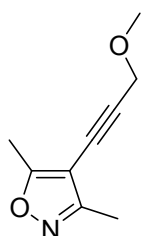
4-(3,3-dimethylbut-1-yn-1-yl)-3,5-dimethylisoxazole (2m). Yellow solid (747 mg, 84%), $R_f = 0.51$ (PE : EA = 15 : 1), mp 46-48 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 2.38 (s, 3H), 2.23 (s, 3H), 1.30 (s, 9H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 169.9, 160.6, 103.8, 101.4, 66.8, 31.1, 28.1, 11.7, 10.3. HRMS (ESI) $[\text{C}_{11}\text{H}_{15}\text{NO} + \text{Na}]^+$ m/z calcd for 178.1226, found 178.1227.



3,5-dimethyl-4-(3-phenylprop-1-yn-1-yl)isoxazole (2n). Brown oil (932 mg, 88%), $R_f = 0.44$ (PE : EA = 10 : 1). $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.45-7.38 (m, 4H), 7.33-7.29 (m, 1H), 3.89 (s, 2H), 2.47 (s, 3H), 2.34 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 170.8, 160.8, 136.5, 128.7, 127.9, 126.8, 101.3, 92.8, 70.7, 25.8, 11.9, 10.6. HRMS (ESI) $[\text{C}_{14}\text{H}_{13}\text{NO} + \text{H}]^+$ m/z calcd for 212.1070, found 212.1074.

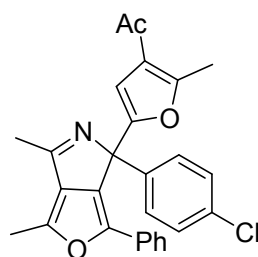


4-(3-methoxyprop-1-yn-1-yl)-3,5-dimethylisoxazole (2o). Brown oil (489 mg, 59%), $R_f = 0.38$ (PE : EA = 10 : 1). $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 4.29 (s,



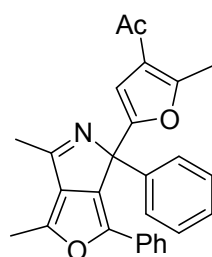
2H), 3.41 (s, 3H), 2.42 (s, 3H), 2.26 (s, 3H). ¹³C NMR (125.8 MHz, CDCl₃, ppm) δ 171.7, 160.7, 100.5, 90.2, 74.6, 60.3, 57.6, 11.9, 10.4. HRMS (ESI) [C₉H₁₁NO₂ + H]⁺ m/z calcd for 166.0863, found 166.0864.

1-(5-(4-(4-chlorophenyl)-1,6-dimethyl-3-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-



methylfuran-3-yl)ethanone (3a). Brown solid (69 mg, 78%), R_f = 0.38 (PE : EA = 2 : 1), mp 218-219 °C. ¹H NMR (500 MHz, CDCl₃, ppm) δ 7.58 (d, *J* = 7.0 Hz, 2H), 7.42 (d, *J* = 8.0 Hz, 2H), 7.32-7.29 (m, 4H), 7.24 (t, *J* = 7.5 Hz, 1H), 6.49 (s, 1H), 2.57 (s, 3H), 2.48 (s, 3H), 2.46 (s, 3H), 2.36 (s, 3H). ¹³C NMR (125.8 MHz, CDCl₃, ppm) δ 194.0, 163.3, 158.9, 151.5, 143.8, 141.9, 136.7, 134.0, 133.26, 133.04, 129.53, 129.36, 128.69, 128.46, 127.57, 124.96, 121.8, 108.9, 76.3, 29.2, 17.7, 14.5, 12.9. HRMS (ESI) [C₂₇H₂₂ClNO₃ + H]⁺ m/z calcd for 444.1361, found 444.1371.

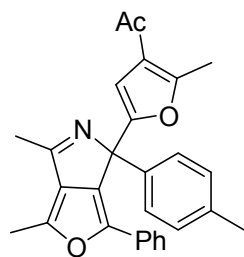
1-(5-(1,6-dimethyl-3,4-diphenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-



yl)ethanone (3b). Brown solid (57 mg, 70%), R_f = 0.41 (PE : EA = 2 : 1), mp 189-192 °C. ¹H NMR (500 MHz, CDCl₃, ppm) δ 7.61-7.59 (m, 2H), 7.49-7.47 (m, 2H), 7.35-7.28 (m, 5H), 7.24-7.21 (m, 1H), 6.51 (s, 1H), 2.57 (s, 3H), 2.48 (s, 3H), 2.46 (s, 3H), 2.33 (s, 3H). ¹³C NMR (125.8 MHz, CDCl₃, ppm) δ 194.2, 163.0, 158.8, 152.0, 143.7, 141.6, 138.1, 133.5, 133.4, 129.7, 128.5, 128.4, 128.1, 127.9, 127.4, 125.0, 121.7, 108.8, 76.9, 29.2, 17.7, 14.5, 12.9.

HRMS (ESI) [C₂₇H₂₃NO₃ + H]⁺ m/z calcd for 410.1751, found 410.1758.

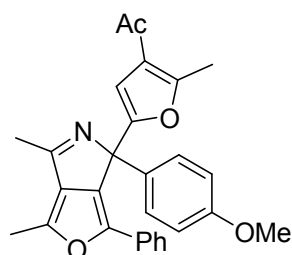
1-(5-(1,6-dimethyl-3-phenyl-4-(p-tolyl)-4H-furo[3,4-c]pyrrol-4-yl)-2-



methylfuran-3-yl)ethanone (3c). Brown solid (58 mg, 69%), R_f = 0.42 (PE : EA = 2 : 1), mp 178-180 °C. ¹H NMR (500 MHz, CDCl₃, ppm) δ 7.68 (d, *J* = 7.5 Hz, 2H), 7.38 (d, *J* = 8.0 Hz, 2H), 7.31-7.28 (m, 2H), 7.24-7.21 (m, 1H), 7.14 (d, *J* = 8.0 Hz, 2H), 6.51 (s, 1H), 2.57 (s, 3H), 2.47 (s, 3H), 2.46 (s, 3H), 2.35 (s, 3H), 2.33 (s, 3H). ¹³C NMR (125.8 MHz, CDCl₃, ppm) δ 194.2, 158.7, 143.7, 137.9, 129.7, 129.2, 128.4, 127.8, 127.7, 127.4, 127.4,

127.3, 125.0, 121.7, 108.8, 76.7, 29.2, 21.1, 17.6, 14.5, 12.9. HRMS (ESI) [C₂₈H₂₅NO₃ + H]⁺ m/z calcd for 424.1907, found 424.1921.

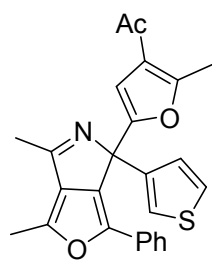
1-(5-(4-(4-methoxyphenyl)-1,6-dimethyl-3-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-



methylfuran-3-yl)ethanone (3d). Yellow solid (62 mg,

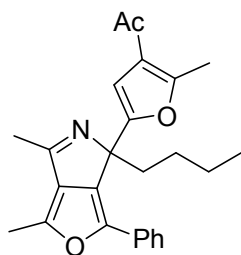
71%), $R_f = 0.27$ (PE : EA = 2 : 1), mp 170-172 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.62-7.60 (m, 2H), 7.43-7.42 (m, 2H), 7.31-7.28 (m, 2H), 7.24-7.21 (m, 1H), 6.88-6.85 (m, 2H), 6.49 (s, 1H), 3.80 (s, 3H), 2.56 (s, 3H), 2.47 (s, 3H), 2.44 (s, 3H), 2.33 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.2, 162.5, 159.3, 158.7, 152.3, 143.5, 141.4, 133.7, 133.4, 130.2, 129.8, 129.2, 128.4, 127.3, 125.0, 121.7, 113.8, 108.6, 76.5, 55.3, 29.2, 17.7, 14.6, 12.9. HRMS (ESI) $[\text{C}_{28}\text{H}_{25}\text{NO}_4 + \text{H}]^+$ m/z calcd for 440.1856, found 440.1868.

1-(5-(1,6-dimethyl-3-phenyl-4-(thiophen-3-yl)-4H-furo[3,4-c]pyrrol-4-yl)-2-



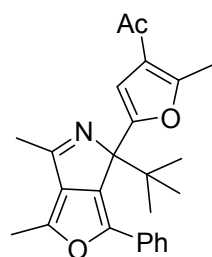
methylfuran-3-yl)ethanone (3e). Brown solid (50 mg, 60%), $R_f = 0.43$ (PE : EA = 2 : 1). $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.60-7.58 (m, 2H), 7.32-7.29 (m, 5H), 7.25-7.22 (m, 2H), 6.44 (s, 1H), 2.56 (s, 3H), 2.53 (s, 3H), 2.44 (s, 3H), 2.31 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.2, 162.8, 158.9, 151.4, 143.3, 141.6, 138.8, 133.8, 133.3, 129.7, 128.5, 127.8, 127.4, 126.1, 124.9, 123.4, 121.8, 108.4, 73.7, 29.2, 17.7, 14.6, 12.9. HRMS (ESI) $[\text{C}_{28}\text{H}_{25}\text{NO}_4 + \text{H}]^+$ m/z calcd for 416.1315, found 416.1329.

1-(5-(4-butyl-1,6-dimethyl-3-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-

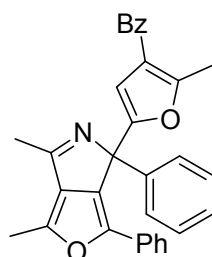


yl)ethanone (3f). Yellow solid (36 mg, 46%), $R_f = 0.33$ (PE : EA = 4 : 1), mp 159-160 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.61 (d, $J = 8.0$ Hz, 2H), 7.37 (t, $J = 8.0$ Hz, 2H), 7.25 (t, $J = 7.5$ Hz, 1H), 6.39 (s, 1H), 2.59-2.55 (m, 1H), 2.56 (s, 3H), 2.54 (s, 3H), 2.42 (s, 3H), 2.37-2.29 (m, 1H), 2.31 (s, 3H), 1.31-1.21 (m, 2H), 1.15-1.06 (m, 1H), 0.98-0.91 (m, 1H), 0.80 (t, $J = 7.5$ Hz, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.3, 163.0, 158.4, 152.5, 142.7, 141.0, 134.0, 133.2, 130.2, 128.6, 127.1, 124.5, 121.8, 106.7, 73.5, 34.7, 29.2, 25.5, 22.7, 17.6, 14.6, 13.9, 12.9. HRMS (ESI) $[\text{C}_{25}\text{H}_{27}\text{NO}_3 + \text{H}]^+$ m/z calcd for 390.2064, found 390.2073.

1-(5-(4-(tert-butyl)-1,6-dimethyl-3-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-



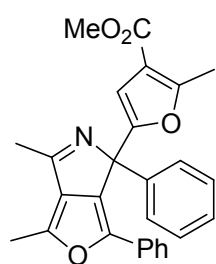
methylfuran-3-yl)ethanone (3g). Yellow solid (32 mg, 41%), $R_f = 0.40$ (PE : EA = 4 : 1), mp 168-169 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.82-7.80 (m, 2H), 7.37-7.34 (m, 2H), 7.29-7.26 (m, 1H), 6.52 (s, 1H), 2.63 (s, 3H), 2.46 (s, 3H), 2.39 (s, 3H), 2.34 (s, 3H), 0.96 (s, 9H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.6, 162.6, 157.5, 152.5, 144.0, 140.9, 133.8, 133.2, 131.3, 128.1, 127.8, 127.4, 121.7, 109.1, 81.3, 40.3, 29.3, 27.3, 17.5, 14.6, 12.7. HRMS (ESI) $[\text{C}_{25}\text{H}_{27}\text{NO}_3 + \text{H}]^+$ m/z calcd for 390.2064, found 390.2074.



(5-(1,6-dimethyl-3,4-diphenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-

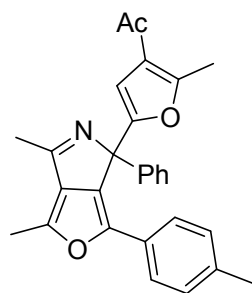
methylfuran-3-yl)(phenyl)methanone (3h). Yellow solid (31 mg, 33%), $R_f = 0.44$ (PE : EA = 2 : 1), mp 188-189 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.65 (d, $J = 7.0$ Hz, 2H), 7.61 (d, $J = 7.5$ Hz, 2H), 7.48-7.46 (m, 2H), 7.38-7.26 (m, 10H), 6.83 (s, 1H), 2.59 (s, 3H), 2.50 (s, 3H), 2.42 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.3, 163.8, 158.8, 152.4, 142.7, 141.3, 139.7, 137.1, 131.2, 128.5, 127.8, 126.8, 121.6, 108.0, 75.6, 65.2, 58.2, 29.1, 17.8, 14.5, 12.9. HRMS (ESI) $[\text{C}_{32}\text{H}_{25}\text{NO}_3 + \text{H}]^+$ m/z calcd for 472.1907, found 472.1921.

methyl 5-(1,6-dimethyl-3,4-diphenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-



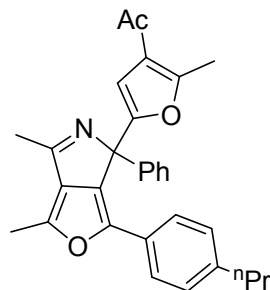
carboxylate (3j). Yellow solid (61 mg, 71%), $R_f = 0.35$ (PE : EA = 4 : 1), mp 201-203 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.59-7.57 (m, 2H), 7.51-7.49 (m, 2H), 7.34-7.27 (m, 5H), 7.23-7.20 (m, 1H), 6.51 (s, 1H), 3.78 (s, 3H), 2.55 (s, 3H), 2.48 (s, 3H), 2.42 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 164.5, 162.7, 159.7, 152.1, 143.6, 141.3, 138.1, 133.7, 133.5, 129.8, 128.4, 128.4, 128.0, 127.3, 124.9, 113.6, 109.2, 77.0, 51.3, 17.7, 13.9, 12.8. HRMS (ESI) $[\text{C}_{27}\text{H}_{23}\text{NO}_4 + \text{H}]^+$ m/z calcd for 426.1700, found 426.1708.

1-(5-(1,6-dimethyl-4-phenyl-3-(p-tolyl)-4H-furo[3,4-c]pyrrol-4-yl)-2-



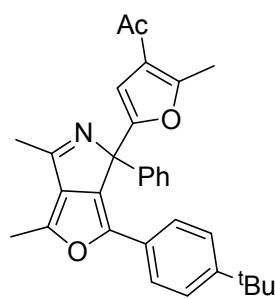
methylfuran-3-yl)ethanone (3k). Brown solid (59 mg, 69%), $R_f = 0.35$ (PE : EA = 2 : 1), mp 168-169 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.52-7.48 (m, 4H), 7.35-7.31 (m, 3H), 7.10 (d, $J = 8.0$ Hz, 2H), 6.49 (s, 1H), 2.55 (s, 3H), 2.49 (s, 3H), 2.45 (s, 3H), 2.33 (s, 6H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.1, 163.0, 158.7, 152.1, 144.0, 141.2, 138.3, 137.3, 133.2, 132.7, 129.0, 128.5, 128.0, 127.9, 127.0, 125.0, 121.7, 108.8, 76.9, 29.1, 21.2, 17.6, 14.5, 12.8. HRMS (ESI) $[\text{C}_{28}\text{H}_{25}\text{NO}_3 + \text{H}]^+$ m/z calcd for 424.1907, found 424.1918.

1-(5-(1,6-dimethyl-4-phenyl-3-(4-propylphenyl)-4H-furo[3,4-c]pyrrol-4-yl)-2-



methylfuran-3-yl)ethanone (3l). Brown solid (64 mg, 71%), $R_f = 0.42$ (PE : EA = 2 : 1), mp 158-159 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.52-7.49 (m, 4H), 7.36-7.31 (m, 3H), 7.15 (d, $J = 8.0$ Hz, 2H), 6.50 (s, 1H), 2.57 (t, $J = 7.5$ Hz, 2H), 2.55 (s, 3H), 2.49 (s, 3H), 2.46 (s, 3H), 2.33 (s, 3H), 1.68-1.60 (m, 2H), 0.95 (t, $J = 7.0$ Hz, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.3, 163.0, 158.8, 152.1, 144.0, 142.1, 141.3, 138.3, 133.3, 132.7, 128.5, 128.1, 127.9, 127.3, 125.0, 121.7, 108.8, 37.8, 29.2, 24.4, 17.7, 14.6, 13.8, 12.9. HRMS (ESI) $[\text{C}_{30}\text{H}_{29}\text{NO}_3 + \text{H}]^+$ m/z calcd for 452.2220, found 452.2231.

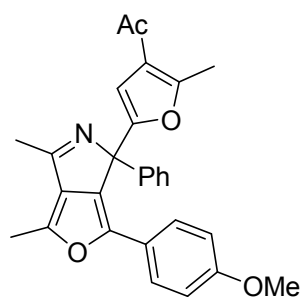
1-(5-(3-(4-(tert-butyl)phenyl)-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3m).



Yellow solid (61 mg, 65%), $R_f = 0.40$ (PE : EA = 2 : 1), mp 178-179 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.54-7.50 (m, 4H), 7.37-7.30 (m, 5H), 6.50 (s, 1H), 2.55 (s, 3H), 2.49 (s, 3H), 2.45 (s, 3H), 2.34 (s, 3H), 1.32 (s, 9H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.3, 163.0, 158.8, 152.2, 150.4, 143.9, 141.2, 138.3, 133.3, 132.8, 128.5, 128.0, 128.0, 127.0, 125.3, 124.8, 121.7, 108.8, 76.9, 34.6, 31.2, 29.2, 17.7, 14.6, 12.9. HRMS (ESI)

$[\text{C}_{31}\text{H}_{31}\text{NO}_3 + \text{H}]^+$ m/z calcd for 466.2377, found 466.2387.

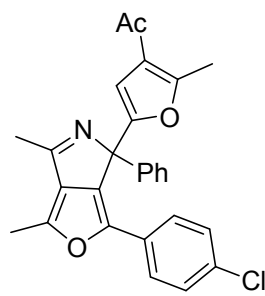
1-(5-(3-(4-methoxyphenyl)-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3n).



Yellow solid (64 mg, 73%), $R_f = 0.23$ (PE : EA = 2 : 1), mp 156-157 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.54-7.51 (m, 2H), 7.49-7.46 (m, 2H), 7.35-7.31 (m, 3H), 6.85-6.82 (m, 2H), 6.50 (s, 1H), 3.80 (s, 3H), 2.54 (s, 3H), 2.49 (s, 3H), 2.46 (s, 3H), 2.33 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.1, 163.2, 159.0, 158.7, 152.2, 143.8, 138.3, 133.1, 131.5, 128.5, 128.1, 127.9, 126.5, 122.7, 121.7, 113.8, 108.8, 76.8, 55.3, 29.1, 17.6, 14.5,

12.8. HRMS (ESI) $[\text{C}_{30}\text{H}_{29}\text{NO}_3 + \text{H}]^+$ m/z calcd for 440.1856, found 440.1864.

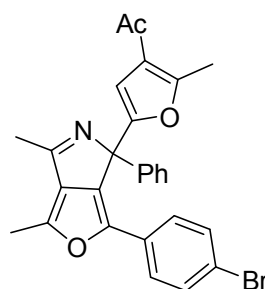
1-(5-(3-(4-chlorophenyl)-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3o).



Brown solid (54 mg, 60%), $R_f = 0.38$ (PE : EA = 2 : 1), mp 210-212 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.54-7.52 (m, 2H), 7.44-7.42 (m, 2H), 7.35-7.32 (m, 3H), 7.27-7.24 (m, 2H), 6.51 (s, 1H), 2.56 (s, 3H), 2.48 (s, 3H), 2.46 (s, 3H), 2.35 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.1, 162.9, 158.7, 151.9, 142.7, 137.9, 134.0, 133.5, 133.1, 128.6, 128.6, 128.2, 128.2, 127.7, 126.2, 121.8, 108.8, 76.9, 29.2, 17.7, 14.5, 12.9. HRMS (ESI) $[\text{C}_{27}\text{H}_{22}\text{ClNO}_3 + \text{H}]^+$

m/z calcd for 444.1361, found 444.1351.

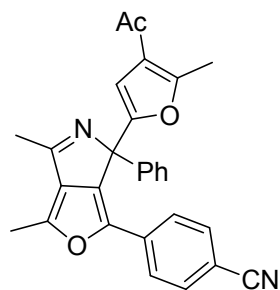
1-(5-(3-(4-bromophenyl)-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3p).



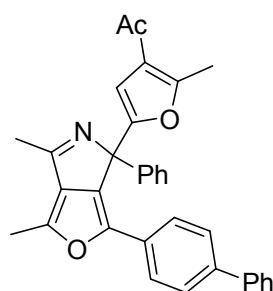
Brown solid (58 mg, 59%), $R_f = 0.35$ (PE : EA = 2 : 1), mp 238-239 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.48-7.41 (m, 6H), 7.35-7.32 (m, 3H), 7.35-7.32 (m, 3H), 7.27-7.24 (m, 2H), 6.53 (s, 1H), 2.56 (s, 3H), 2.48 (s, 3H), 2.47 (s, 3H), 2.35 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.0, 162.9, 158.6, 151.7, 142.6, 141.9, 137.7, 134.0, 133.4, 131.4, 128.5, 128.5, 128.1, 127.6, 126.3, 121.6, 121.2,

108.7, 76.8, 29.1, 17.5, 14.5, 12.8. HRMS (ESI) $[C_{27}H_{22}BrNO_3 + H]^+$ m/z calcd for 488.0856, found 488.0848.

4-(4-(4-acetyl-5-methylfuran-2-yl)-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-3-yl)benzonitrile (3q). Yellow solid (47 mg, 54%), $R_f = 0.20$ (PE : EA = 2 : 1), mp 258-259 °C. 1H NMR (500 MHz, $CDCl_3$, ppm) δ 7.69-7.67 (m, 2H), 7.57-7.55 (m, 2H), 7.39-7.37 (m, 2H), 7.36-7.34 (m, 3H), 6.55 (s, 1H), 2.59 (s, 3H), 2.46 (s, 3H), 2.45 (s, 3H), 2.36 (s, 3H). ^{13}C NMR (125.8 MHz, $CDCl_3$, ppm) δ 194.0, 162.8, 158.7, 151.5, 143.4, 141.8, 137.5, 137.2, 134.0, 133.5, 132.2, 128.8, 128.5, 127.6, 125.1, 121.8, 118.9, 110.2, 108.9, 77.2, 29.2, 17.7, 14.5, 13.0. HRMS (ESI) $[C_{28}H_{22}N_2O_3 + H]^+$ m/z calcd for 435.1703, found 435.1697.

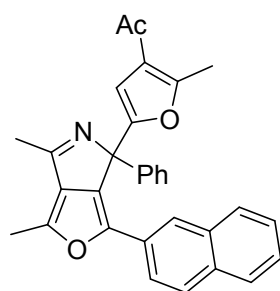


1-(5-(3-([1,1'-biphenyl]-4-yl)-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3r). Yellow solid (55 mg, 57%), $R_f = 0.20$ (PE : EA = 2 : 1), mp 273-274 °C. 1H NMR (500 MHz, $CDCl_3$, ppm) δ 7.66 (d, $J = 7.0$ Hz, 2H), 7.58 (d, $J = 8.0$ Hz, 2H), 7.53 (d, $J = 8.0$ Hz, 2H), 7.49 (d, $J = 6.5$ Hz, 2H), 7.42 (t, $J = 8.0$ Hz, 2H), 7.36-7.31 (m, 4H), 6.53 (s, 1H), 2.56 (s, 3H), 2.47 (s, 3H), 2.45 (s, 3H), 2.33 (s, 3H). ^{13}C NMR (125.8 MHz, $CDCl_3$, ppm) δ 194.2, 163.0, 158.8, 152.1, 143.5, 141.7, 140.4, 139.9, 138.1, 133.7, 133.5, 128.8, 128.7, 128.6, 128.2, 127.9,

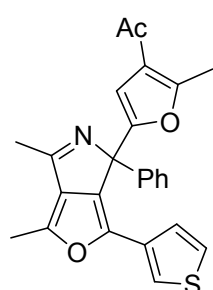


127.5, 127.0, 126.9, 125.4, 121.8, 108.8, 77.0, 29.2, 17.7, 14.6, 12.9. HRMS (ESI) $[C_{33}H_{27}NO_3 + H]^+$ m/z calcd for 486.2064, found 486.2055.

1-(5-(1,6-dimethyl-3-(naphthalen-2-yl)-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3s). Yellow solid (59 mg, 64%), $R_f = 0.37$ (PE : EA = 2 : 1), mp 228-229 °C. 1H NMR (500 MHz, $CDCl_3$, ppm) δ 8.10 (s, 1H), 7.80-7.77 (m, 1H), 7.75-7.70 (m, 3H), 7.54-7.51 (m, 2H), 7.48-7.44 (m, 2H), 7.37-7.33 (m, 3H), 6.57 (s, 1H), 2.61 (s, 3H), 2.49 (s, 3H), 2.48 (s, 3H), 2.34 (s, 3H). ^{13}C NMR (125.8 MHz, $CDCl_3$, ppm) δ 194.2, 163.0, 158.8, 152.2, 143.8, 141.8, 138.3, 134.2, 133.6, 133.3, 132.5, 128.6, 128.2, 128.0, 127.9, 127.7, 127.2, 126.5, 126.2,



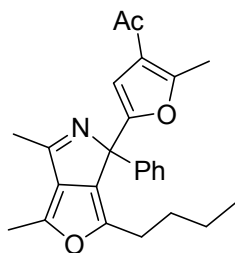
123.7, 123.2, 121.8, 108.9, 77.1, 29.2, 17.7, 14.6, 13.0. HRMS (ESI) $[C_{31}H_{25}NO_3 + H]^+$ m/z calcd for 460.1907, found 460.1898.



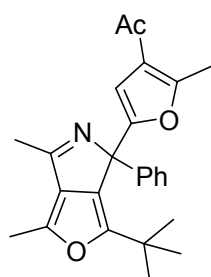
1-(5-(1,6-dimethyl-4-phenyl-3-(thiophen-3-yl)-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3t). Brown solid (51

mg, 61%), $R_f = 0.41$ (PE : EA = 3 : 1), mp 213-215 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.44-7.40 (m, 3H), 7.33-7.28 (m, 3H), 7.39-7.37 (m, 2H), 7.24-7.21 (m, 2H), 6.48 (s, 1H), 2.50 (s, 3H), 2.45 (s, 3H), 2.42 (s, 3H), 2.31 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.2, 163.2, 158.7, 152.4, 141.1, 140.9, 138.4, 132.8, 132.0, 131.3, 128.5, 128.1, 127.7, 125.7, 125.6, 121.7, 120.6, 108.3, 76.6, 29.2, 17.8, 14.5, 12.8. HRMS (ESI) $[\text{C}_{25}\text{H}_{21}\text{NO}_3\text{S} + \text{H}]^+$ m/z calcd for 416.1315, found 416.1307.

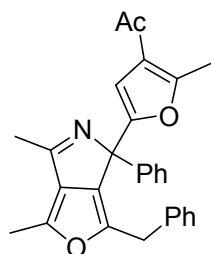
1-(5-(3-butyl-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3u). Yellow solid (51 mg, 65%), $R_f = 0.37$ (PE : EA = 4 : 1), mp 168-169 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.38-7.26 (m, 5H), 6.52 (s, 1H), 2.68 (t, $J = 7.5$ Hz, 3H), 2.54 (s, 3H), 2.42 (s, 3H), 2.42 (s, 3H), 2.38 (s, 3H), 1.58-1.49 (m, 2H), 1.29-1.25 (m, 2H), 0.86 (t, $J = 7.5$ Hz, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.3, 163.9, 158.4, 153.2, 146.0, 140.5, 140.2, 132.4, 131.0, 128.4, 127.7, 127.0, 121.7, 107.5, 75.3, 30.4, 29.1, 27.1, 22.3, 17.7, 14.5, 13.8, 12.7. HRMS (ESI) $[\text{C}_{25}\text{H}_{27}\text{NO}_3 + \text{H}]^+$ m/z calcd for 390.2064, found 390.2061.



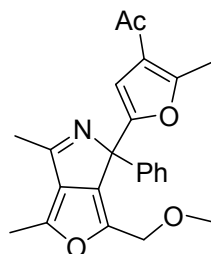
1-(5-(3-(tert-butyl)-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3v). Yellow solid (45 mg, 58%), $R_f = 0.41$ (PE : EA = 4 : 1), mp 172-174 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.33-7.26 (m, 5H), 6.52 (s, 1H), 2.59 (s, 3H), 2.44 (s, 3H), 2.39 (s, 3H), 2.37 (s, 3H), 1.13 (s, 9H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.3, 162.6, 158.3, 152.9, 152.6, 140.1, 139.7, 131.8, 130.8, 128.1, 127.7, 127.7, 121.8, 109.3, 76.4, 33.3, 29.2, 29.2, 17.6, 14.5, 12.7. HRMS (ESI) $[\text{C}_{25}\text{H}_{27}\text{NO}_3 + \text{H}]^+$ m/z calcd for 390.2064, found 390.2056.



1-(5-(3-benzyl-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3w). Brown solid (58 mg, 70%), $R_f = 0.41$ (PE : EA = 2 : 1), mp 180-181 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.40-7.38 (m, 2H), 7.36-7.21 (m, 3H), 7.26-7.20 (m, 3H), 7.05 (d, $J = 8.5$ Hz, 2H), 6.49 (s, 1H), 4.05 (s, 2H), 2.45 (s, 3H), 2.43 (s, 6H), 2.33 (s, 3H). $^{13}\text{C NMR}$ (125.8 MHz, CDCl_3 , ppm) δ 194.4, 164.0, 158.7, 152.9, 143.4, 141.5, 139.9, 137.8, 133.9, 131.1, 128.5, 128.5, 128.4, 127.8, 127.0, 126.5, 121.6, 107.7, 75.4, 33.3, 29.2, 17.8, 14.4, 12.9. HRMS (ESI) $[\text{C}_{28}\text{H}_{25}\text{NO}_3 + \text{H}]^+$ m/z calcd for 424.1907, found 424.1904.

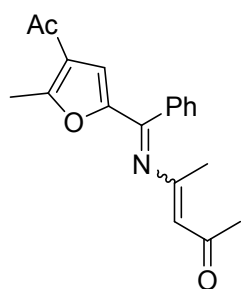


1-(5-(3-(methoxymethyl)-1,6-dimethyl-4-phenyl-4H-furo[3,4-c]pyrrol-4-yl)-2-methylfuran-3-yl)ethanone (3x). Yellow solid (45 mg, 59%), $R_f = 0.41$ (PE : EA = 2 : 1), mp 168-169 °C. $^1\text{H NMR}$ (500 MHz, CDCl_3 , ppm) δ 7.39 (d, $J = 8.0$ Hz, 2H), 7.35-7.27 (m, 3H), 6.63 (s, 1H),



4.43 (dd, $J = 12.5, 16.0$ Hz, 2H), 3.32 (s, 3H), 2.55 (s, 3H), 2.46 (s, 6H), 2.45 (s, 3H), 2.38 (s, 3H). ^{13}C NMR (125.8 MHz, CDCl_3 , ppm) δ 194.3, 163.7, 158.8, 152.4, 142.7, 141.3, 139.7, 137.1, 131.2, 128.5, 127.8, 126.8, 121.6, 108.0, 75.6, 65.2, 58.2, 29.1, 17.8, 14.5, 12.9. HRMS (ESI) $[\text{C}_{23}\text{H}_{23}\text{NO}_4 + \text{H}]^+$ m/z calcd for 378.1700, found 378.1711.

4-(((4-acetyl-5-methylfuran-2-yl)(phenyl)methylene)amino)pent-3-en-2-one (6b).



Yellow oil (55 mg, mixture of two geometric isomers (10:3), 88%), $R_f = 0.45$ (PE : EA = 4 : 1). ^1H NMR (500 MHz, CDCl_3 , ppm) δ 7.51-7.40 (m, 7H), 6.77 (s, 0.31H), 6.69 (s, 1H), 5.48 (s, 0.31H), 5.41 (s, 1H), 2.70 (s, 0.91H), 2.67 (s, 3H), 2.38 (s, 0.9H), 2.36 (s, 3H), 2.29 (s, 0.91H), 2.09 (s, 3H), 2.05 (s, 0.92H), 1.93 (s, 3H). ^{13}C NMR (125.8 MHz, CDCl_3 , ppm) δ 197.7, 193.6, 193.3, 162.0, 161.5, 130.4, 129.9, 128.5, 128.3, 128.3, 127.8, 123.3, 123.0, 118.6, 32.0, 29.1, 23.4, 19.7, 14.9, 14.8. HRMS (ESI) $[\text{C}_{19}\text{H}_{19}\text{NO}_3 + \text{H}]^+$ m/z calcd for 310.1438, found 310.1430.

The X-Ray Crystal Structure of 3b.

Important Crystal Data for **3b**. Crystallographic data have been deposited with the Cambridge Crystallographic Data Centre; deposition no. CCDC-2051532.

Bond precision: C-C = 0.0025 Å Wavelength = 1.54184

Cell: a = 9.1471(4) b = 9.2244(3) c = 14.1426(6)
 Alpha = 92.216(3) beta = 107.400(4) gamma = 108.470(3)

Temperature: 293 K

	Calculated	Reported
Volume	1068.25(8)	1068.25(8)
Space group	P -1	P -1
Hall group	-P 1	-P 1
Moiety formula	C ₂₇ H ₂₃ N O ₃	C ₂₇ H ₂₃ N O ₃
Sum formula	C ₂₇ H ₂₃ N O ₃	C ₂₇ H ₂₃ N O ₃
Mr	409.46	409.46
D _x , g cm ⁻³	1.273	1.273
Z	2	2
Mu (mm ⁻¹)	0.660	0.660
F ₀₀₀	432.0	432.0

F000' 433.28
h, k, lmax 10, 10, 16
Nref 3766
Tmin, Tmax 0.876, 0.936
Tmin' 0.876
Correction method = # Reported T Limits: Tmin = 0.771 Tmax = 1.000
AbsCorr = MULTI-SCAN

Data completeness = 0.995 Theta(max) = 66.585

R(reflections) = 0.0441(3441) wR2(reflections) = 0.1159(3749)

S = 1.027 Npar = 284

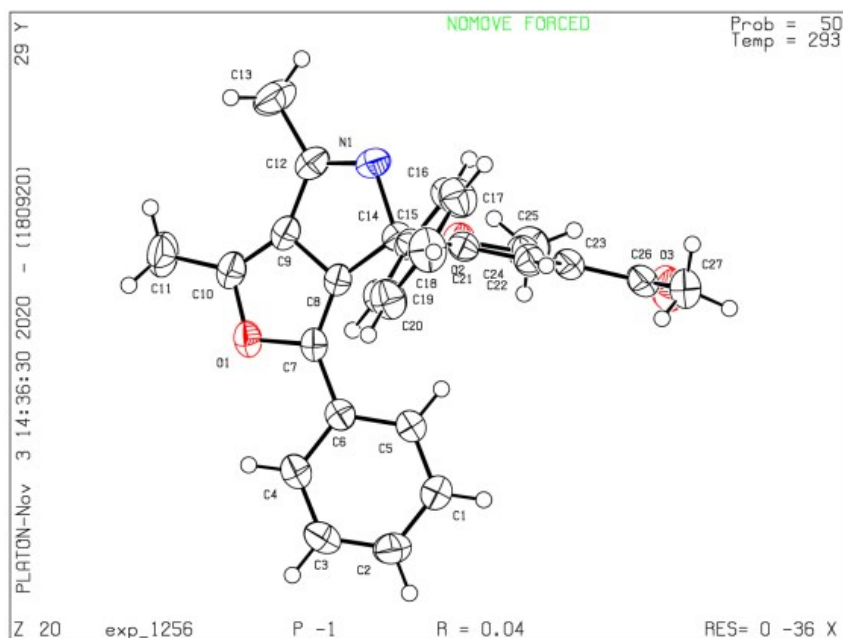
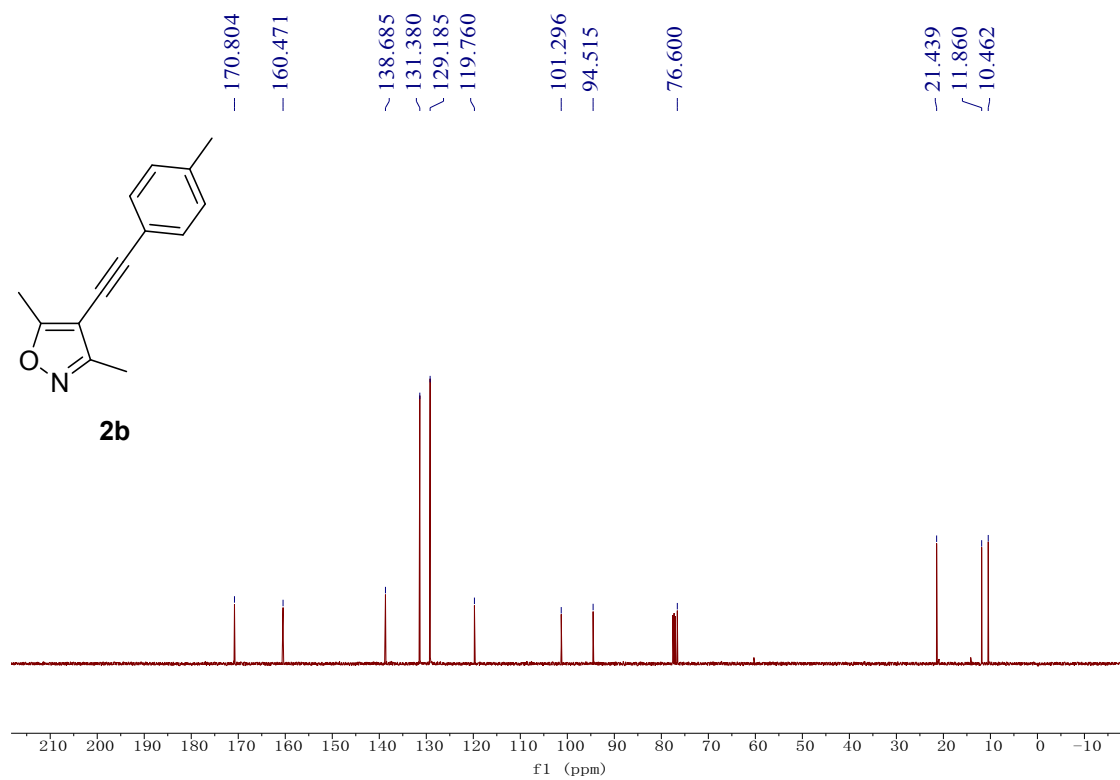
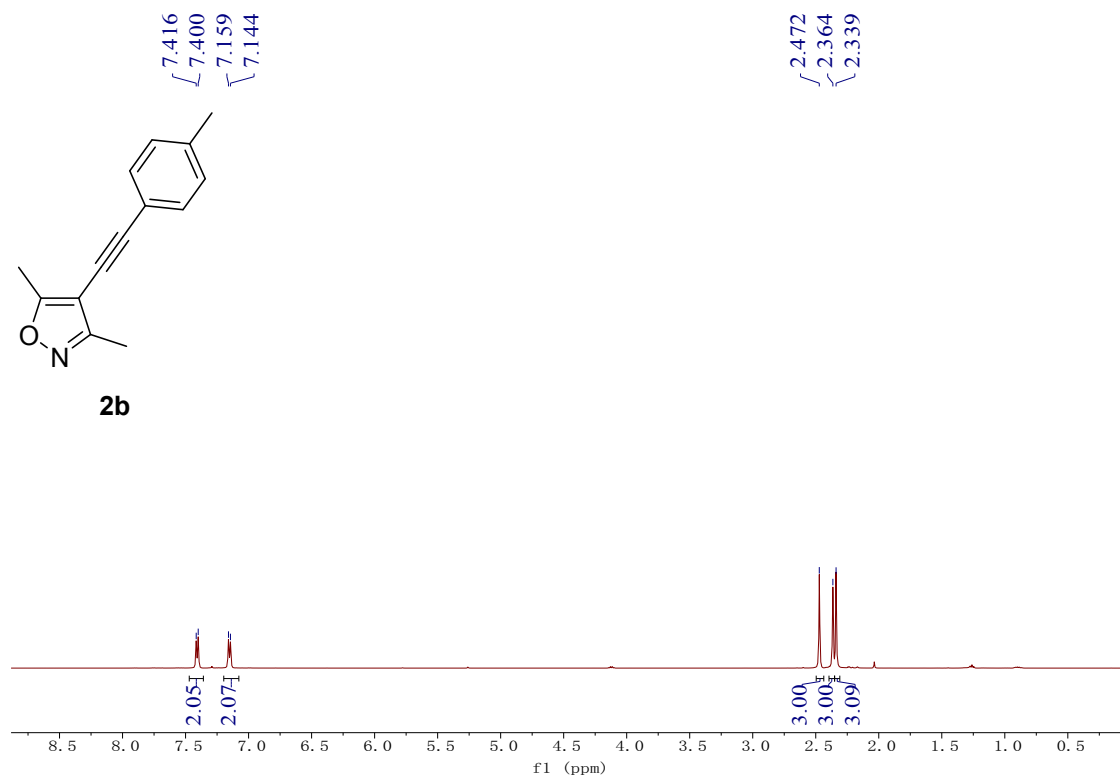
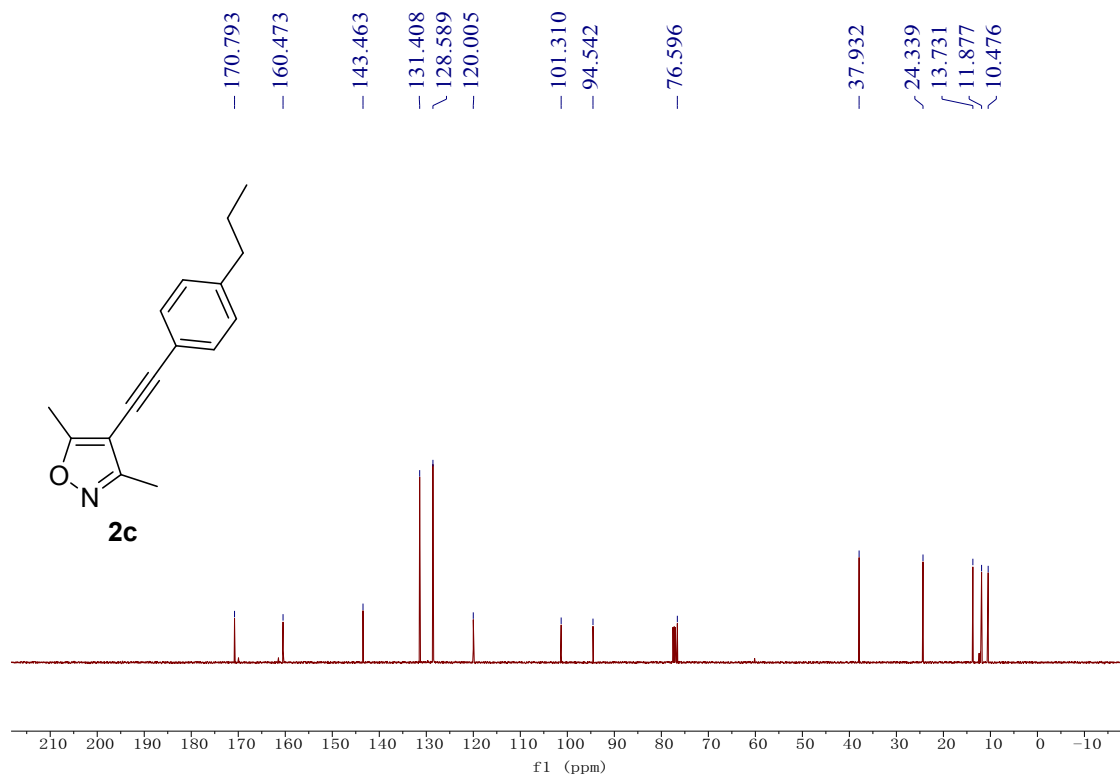
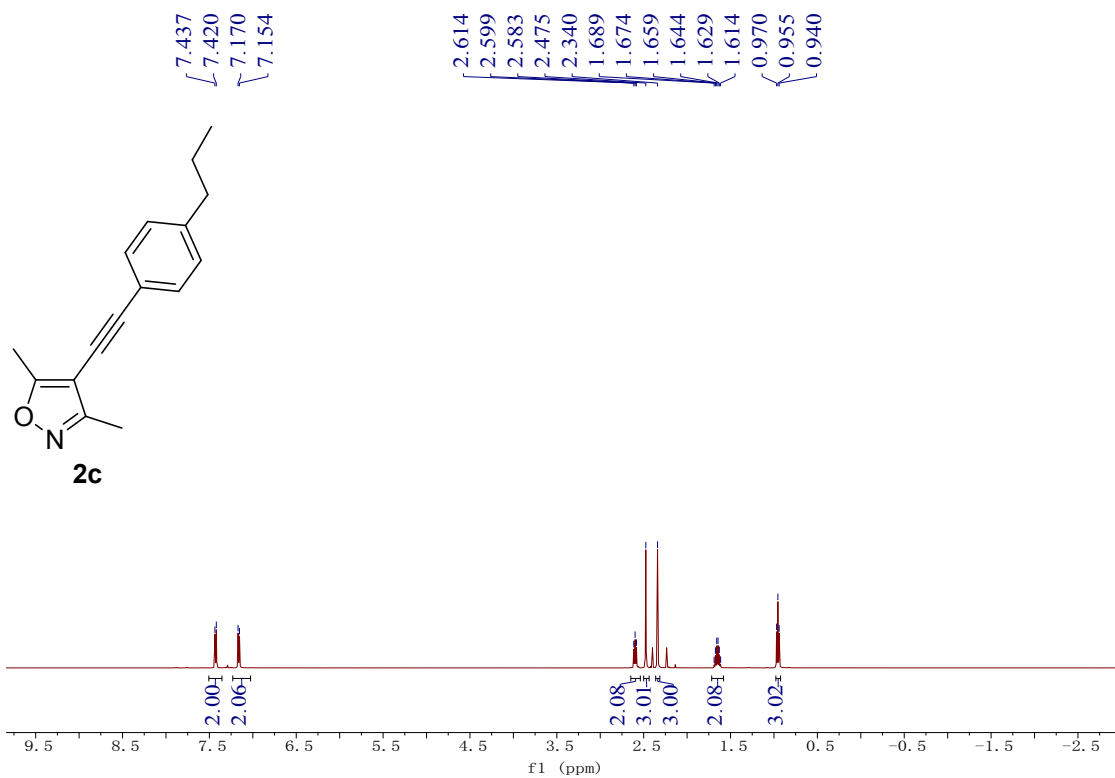


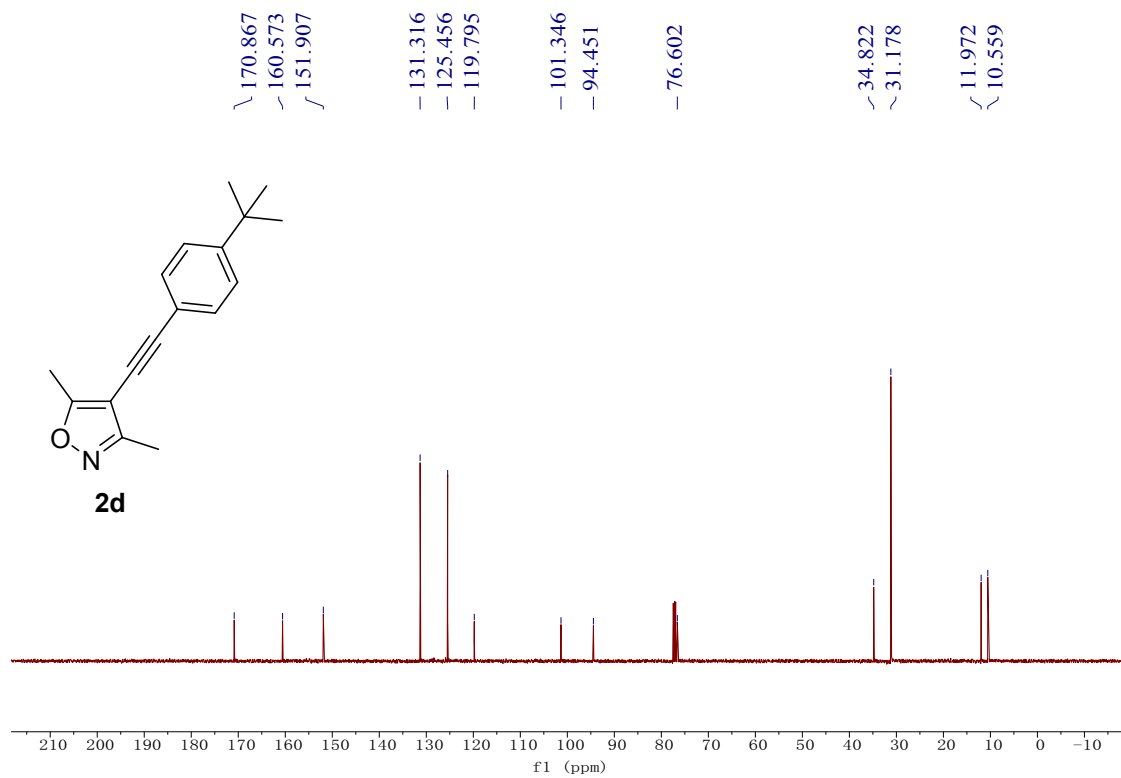
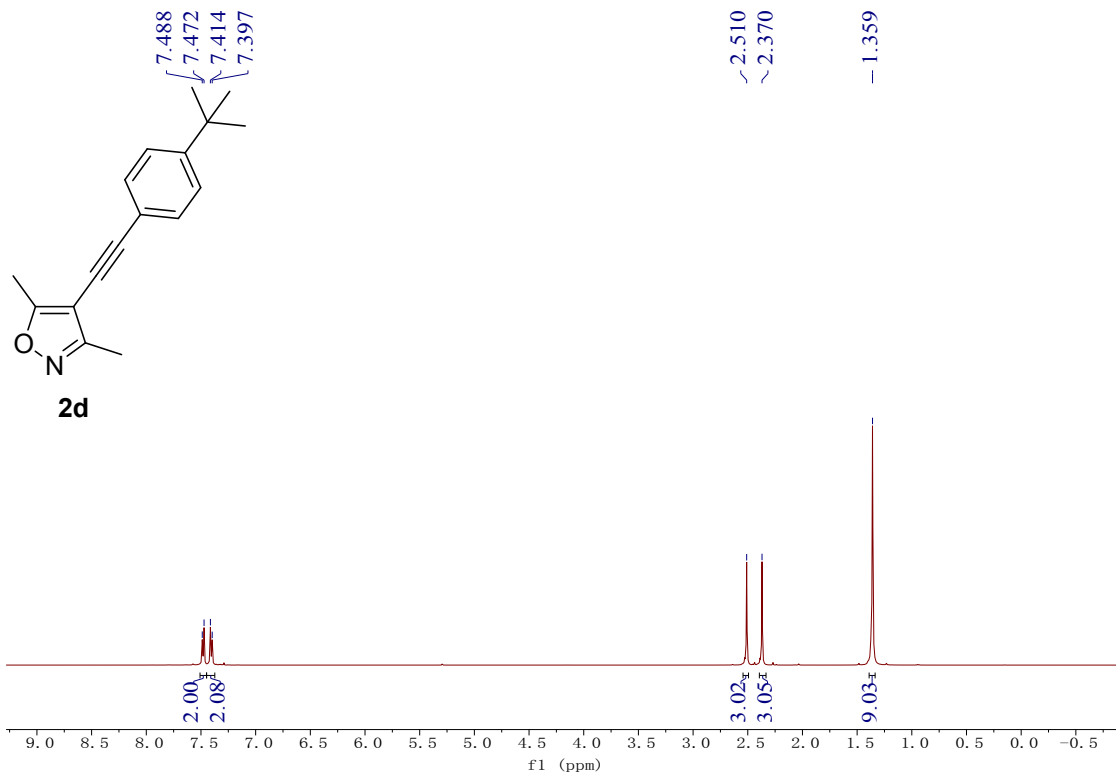
Figure S1 X-ray crystal structure of product **3b**.

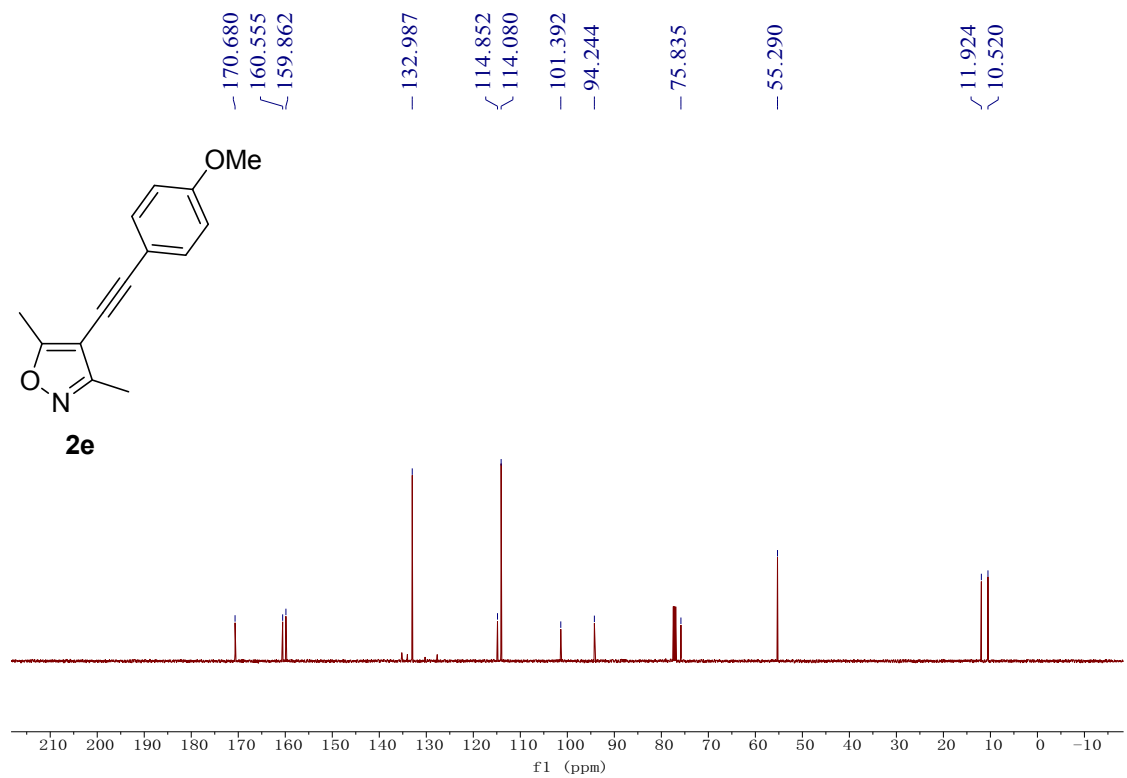
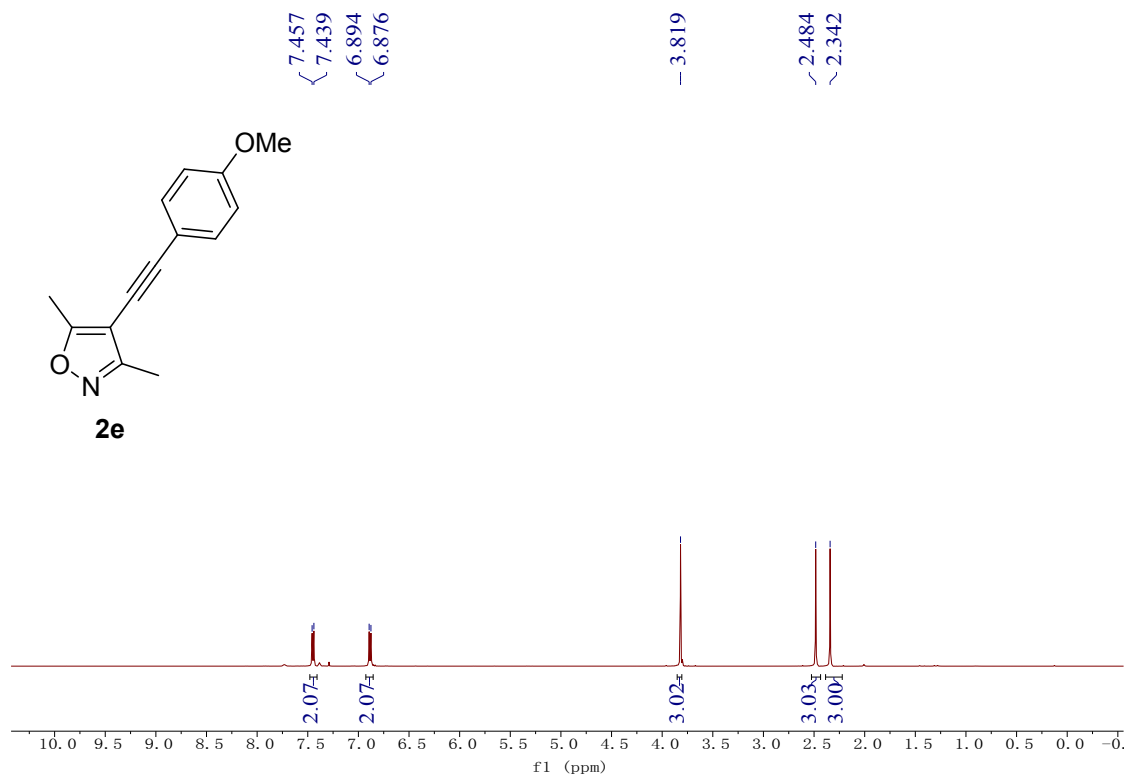
Procedure for the preparation of sample: Compound **3b** (60 mg) was dissolved in DCM (2 ml). Then *n*-hexane (5 ml) was added in above mixture. The mixture was stayed in dry and dark place. Solvents was slowly evaporated to form the crystal (CCDC-2051532).

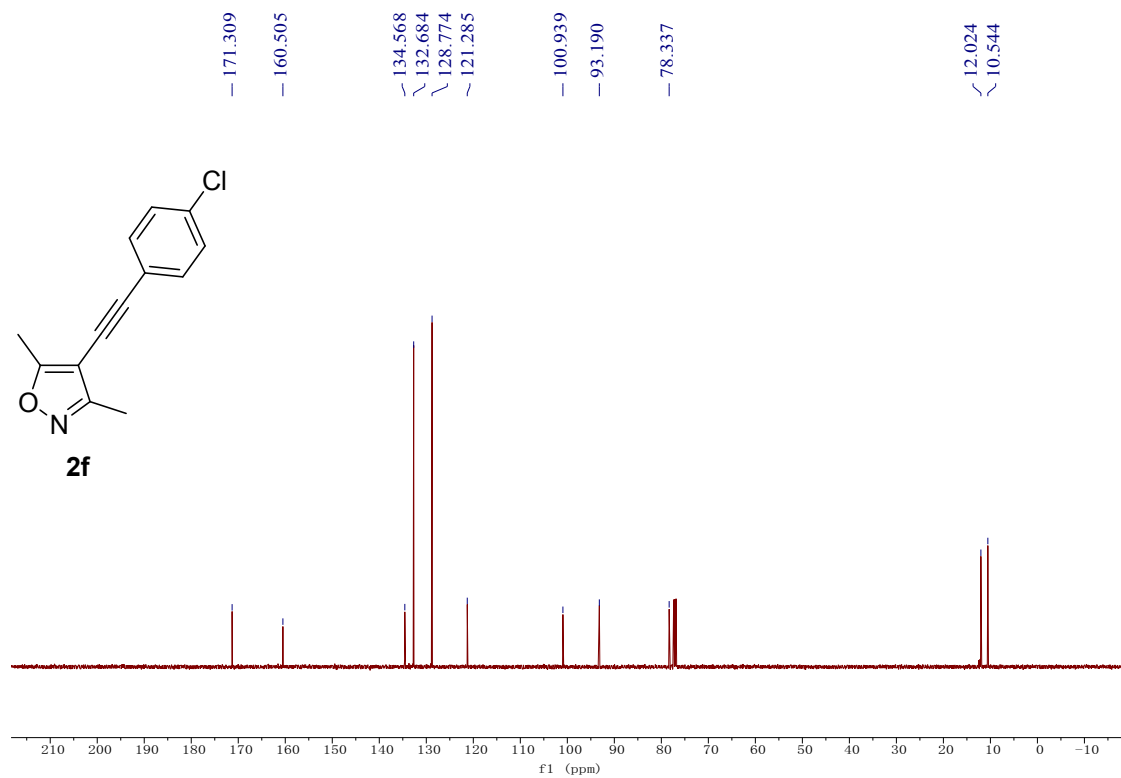
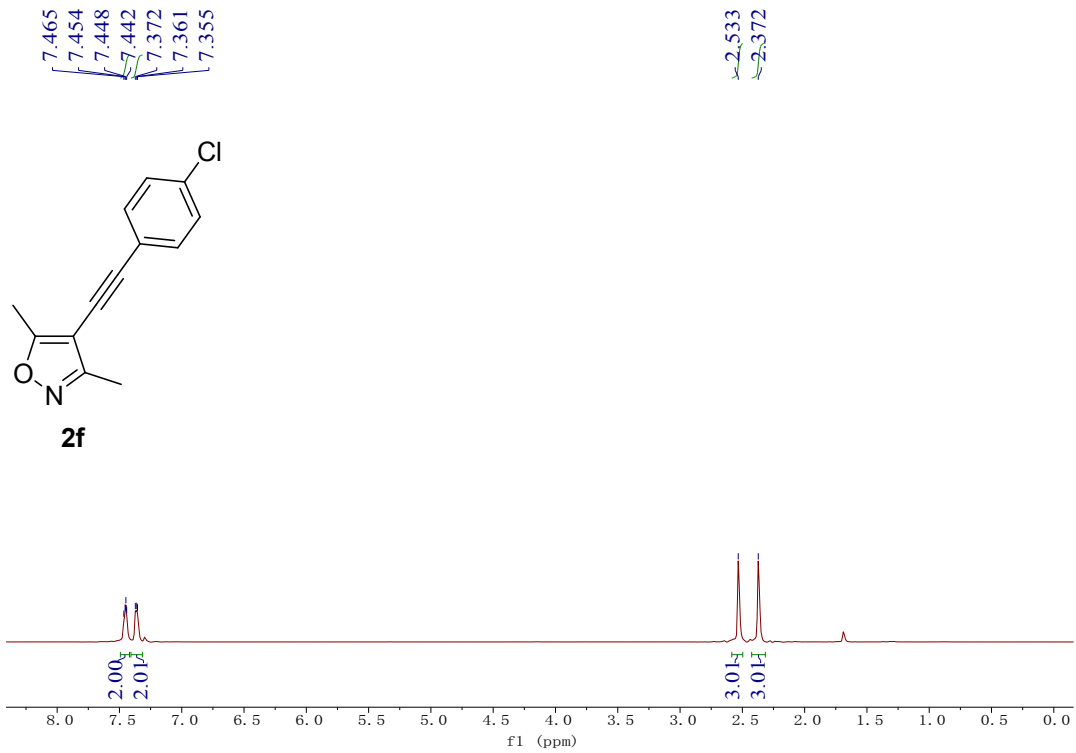
^1H and ^{13}C NMR spectra of new compounds

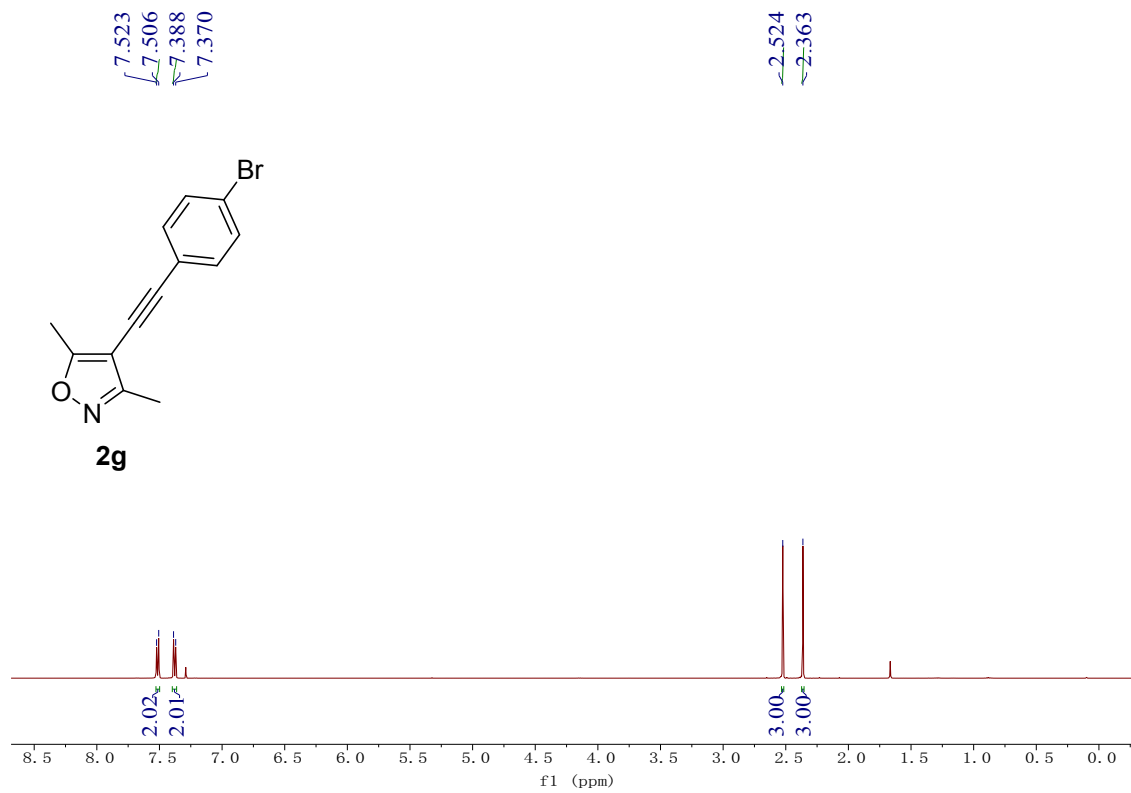


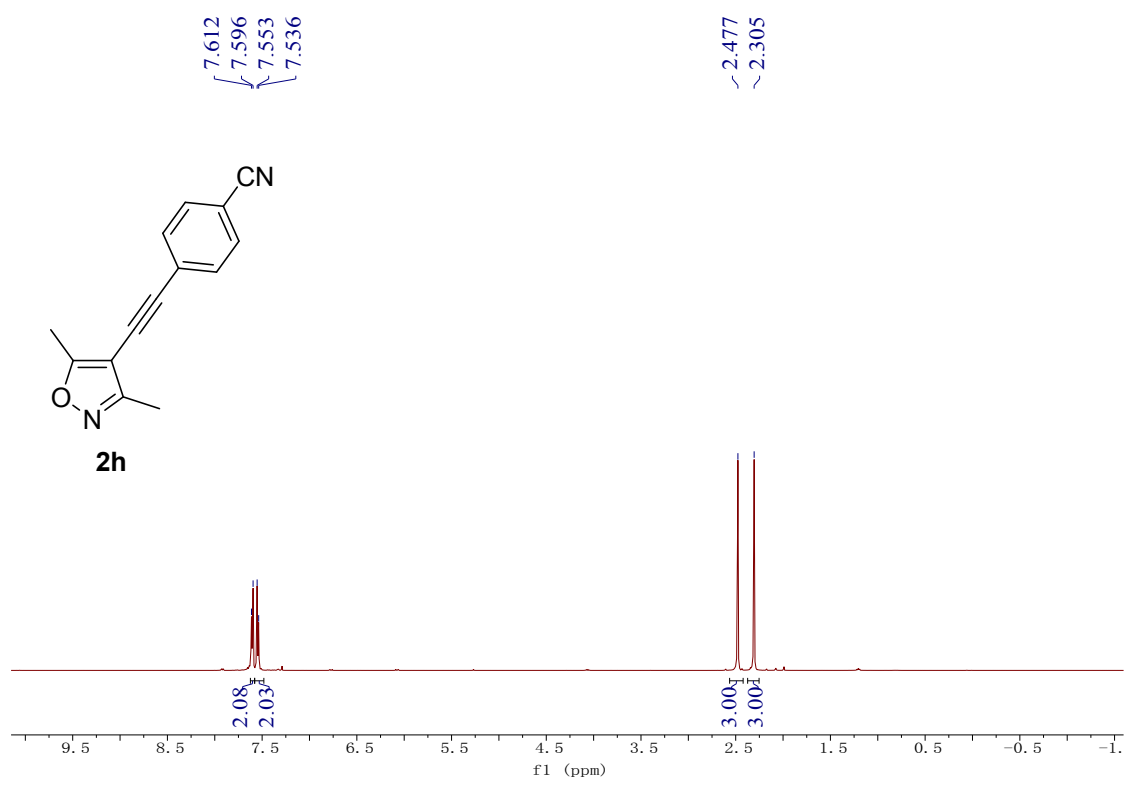
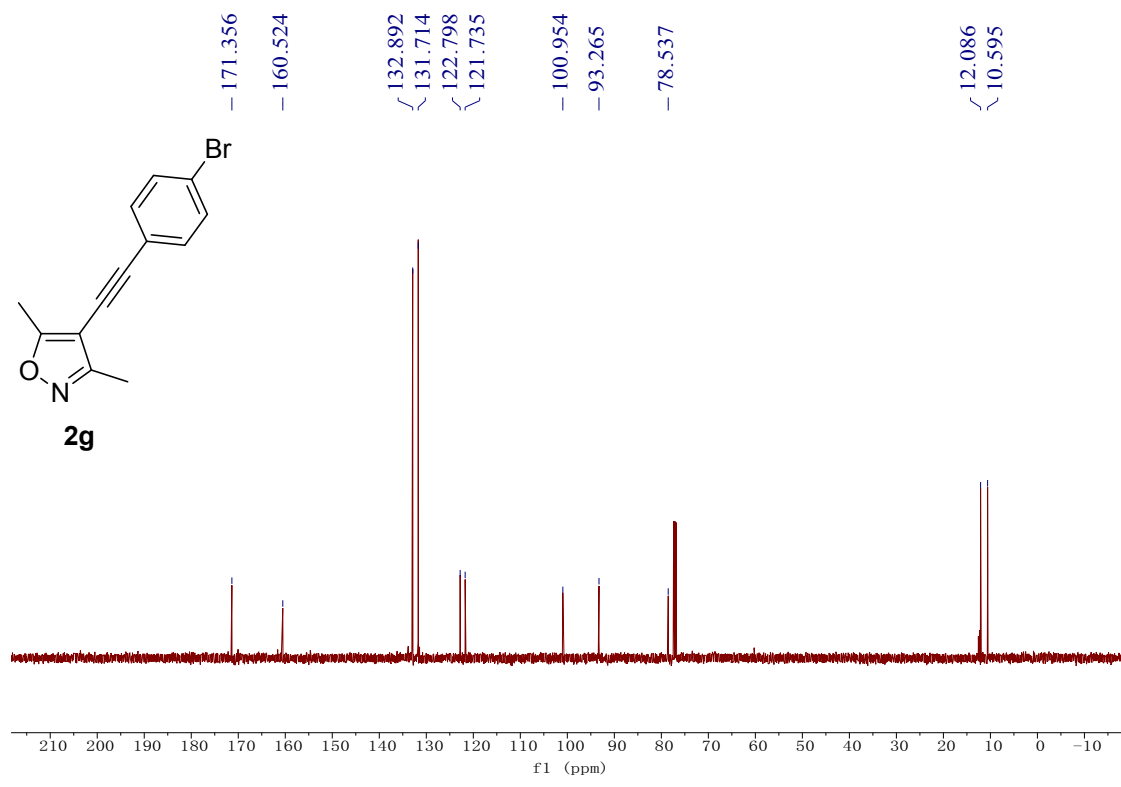


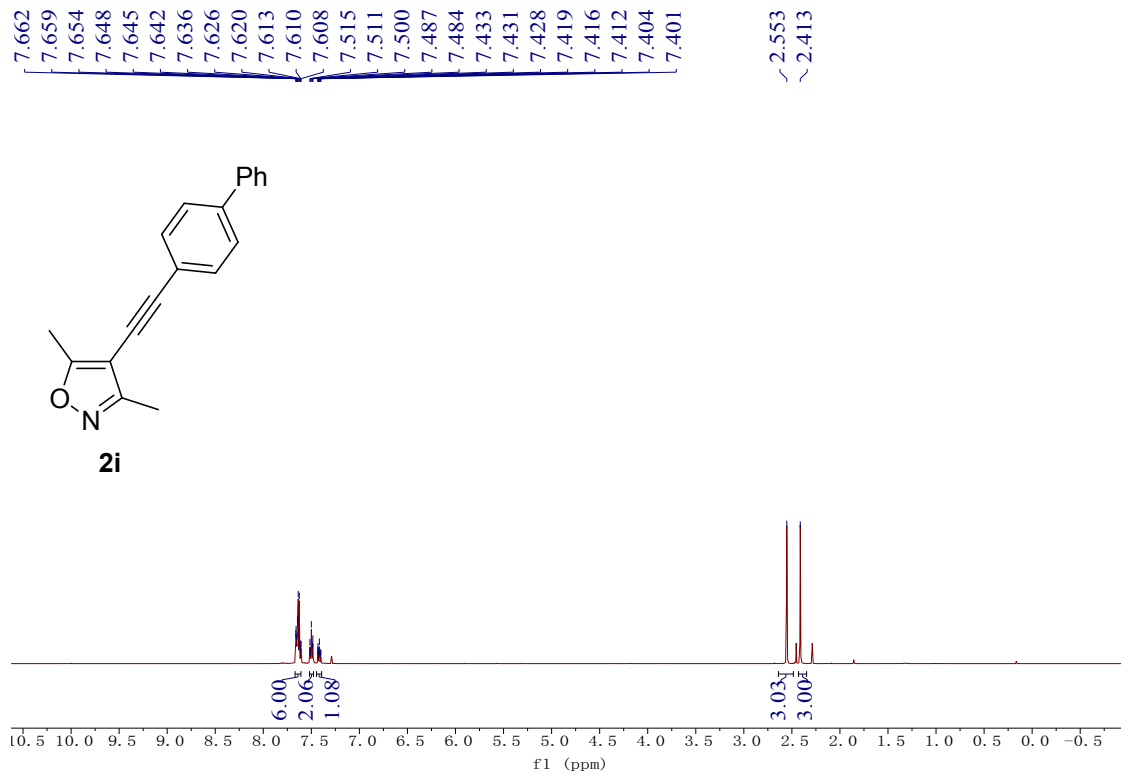
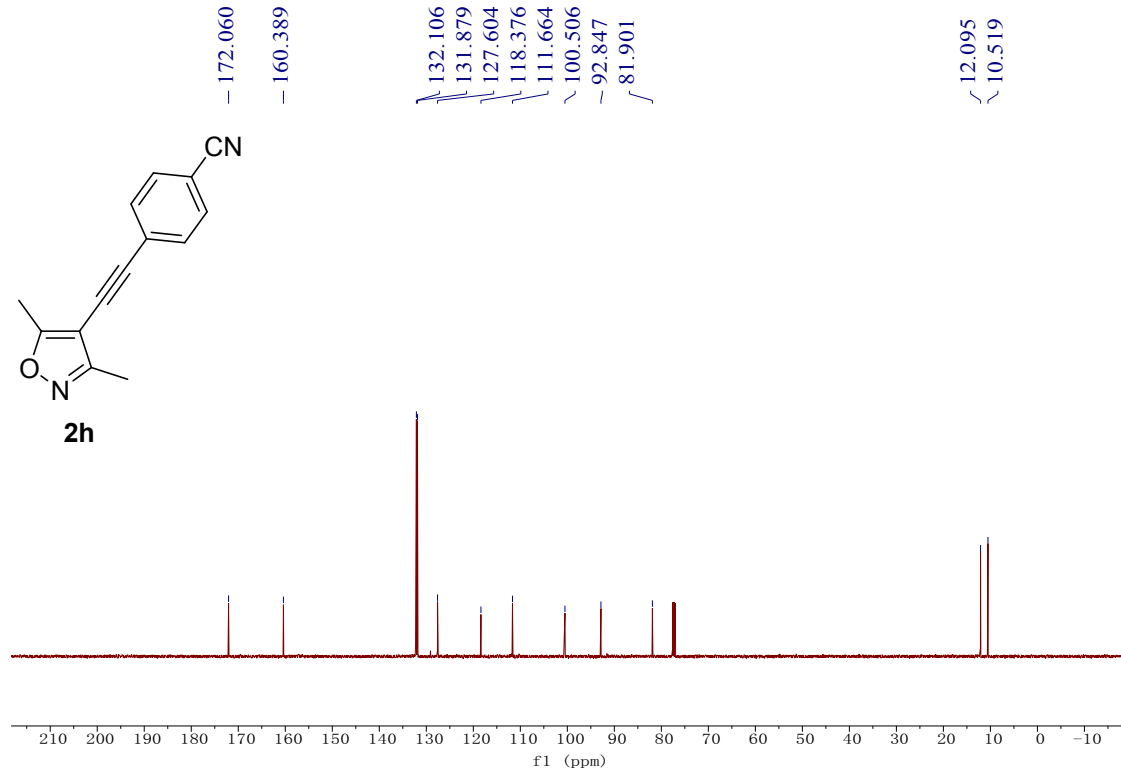


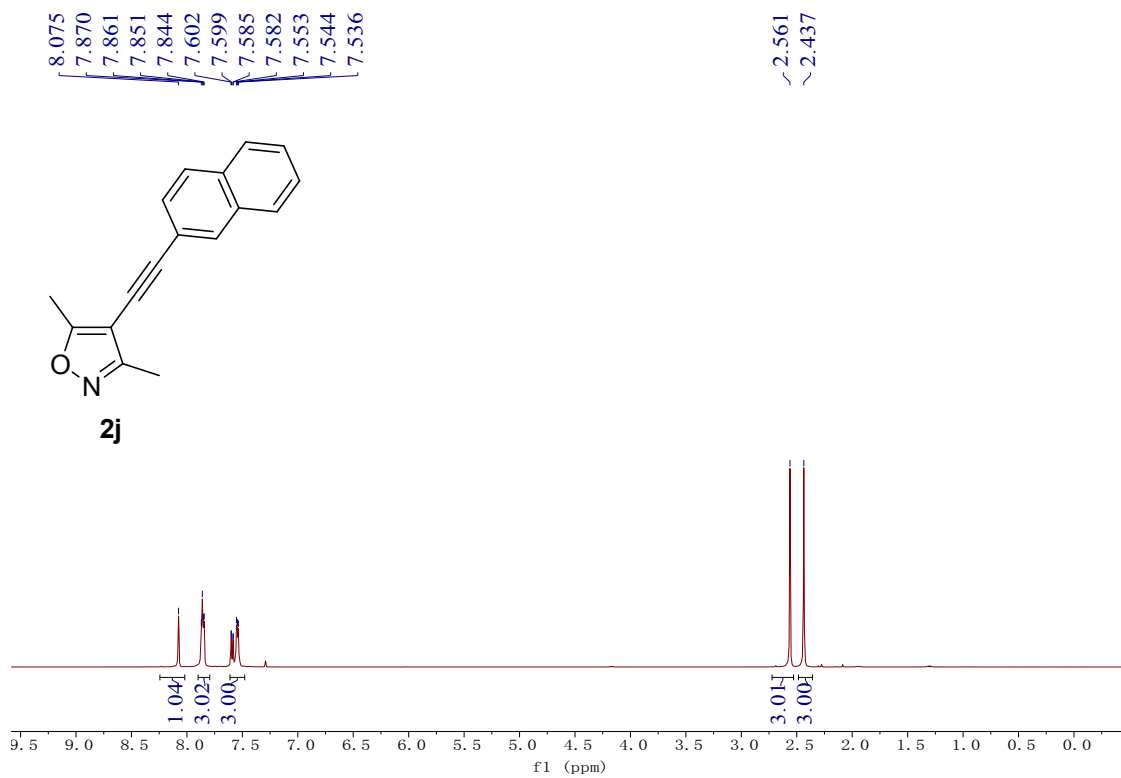
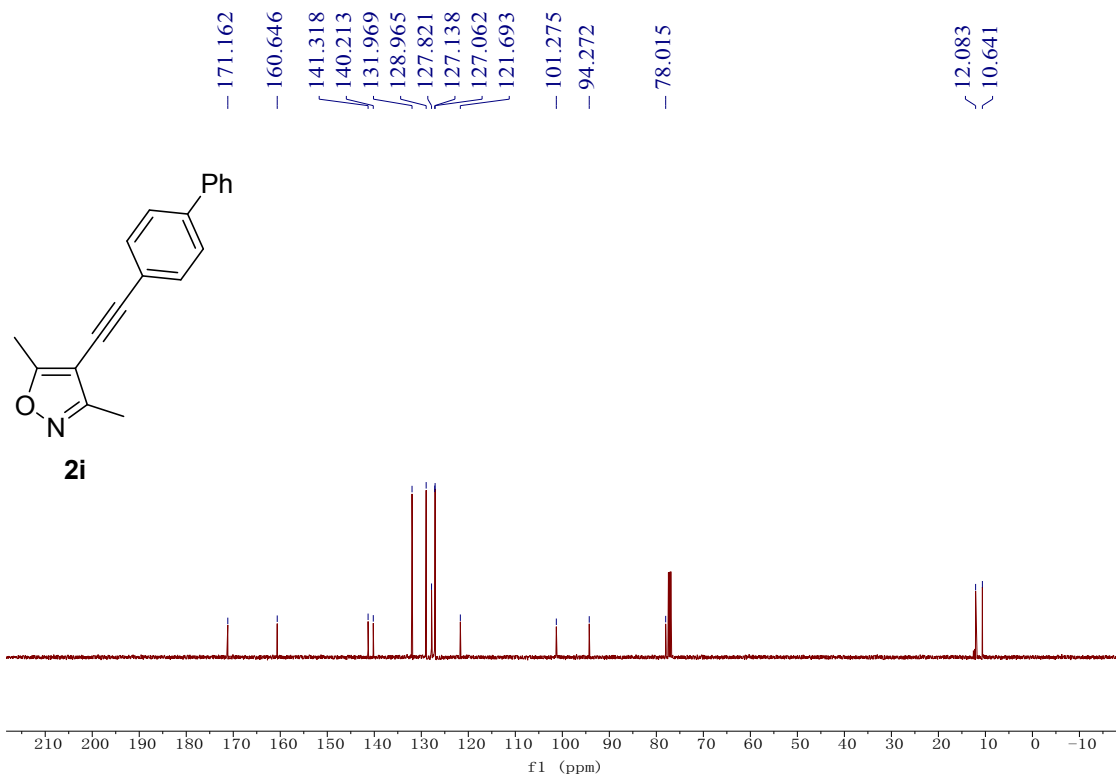


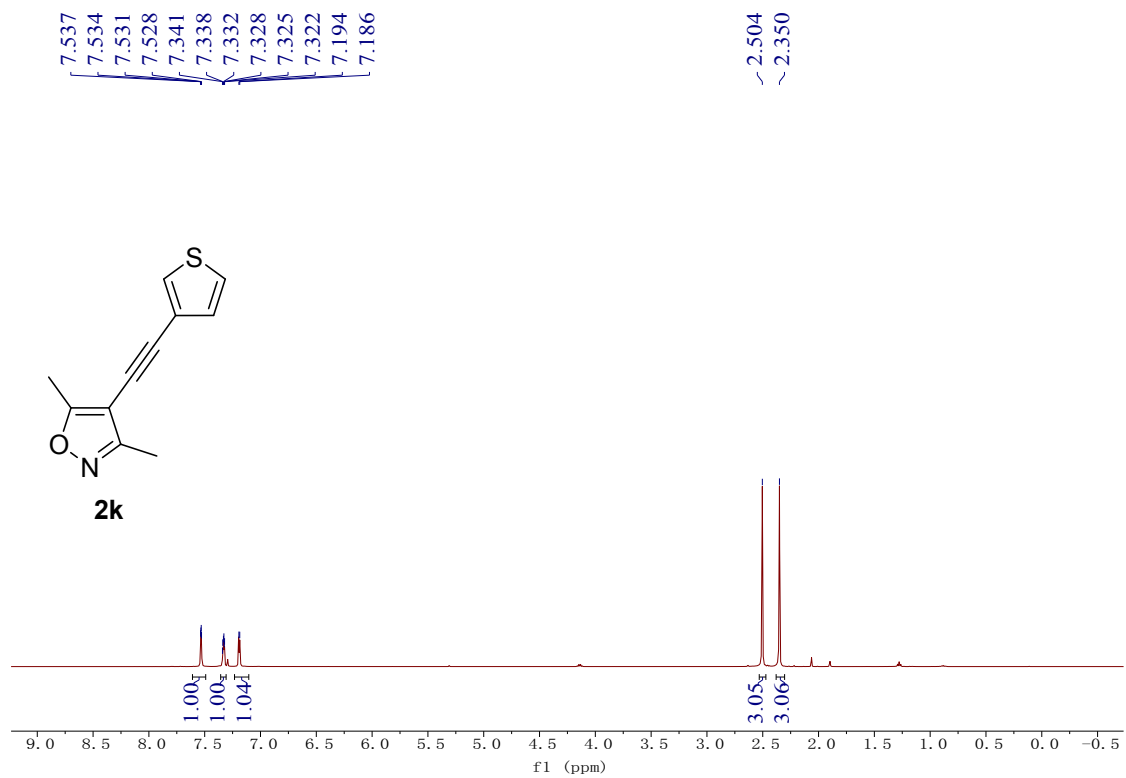
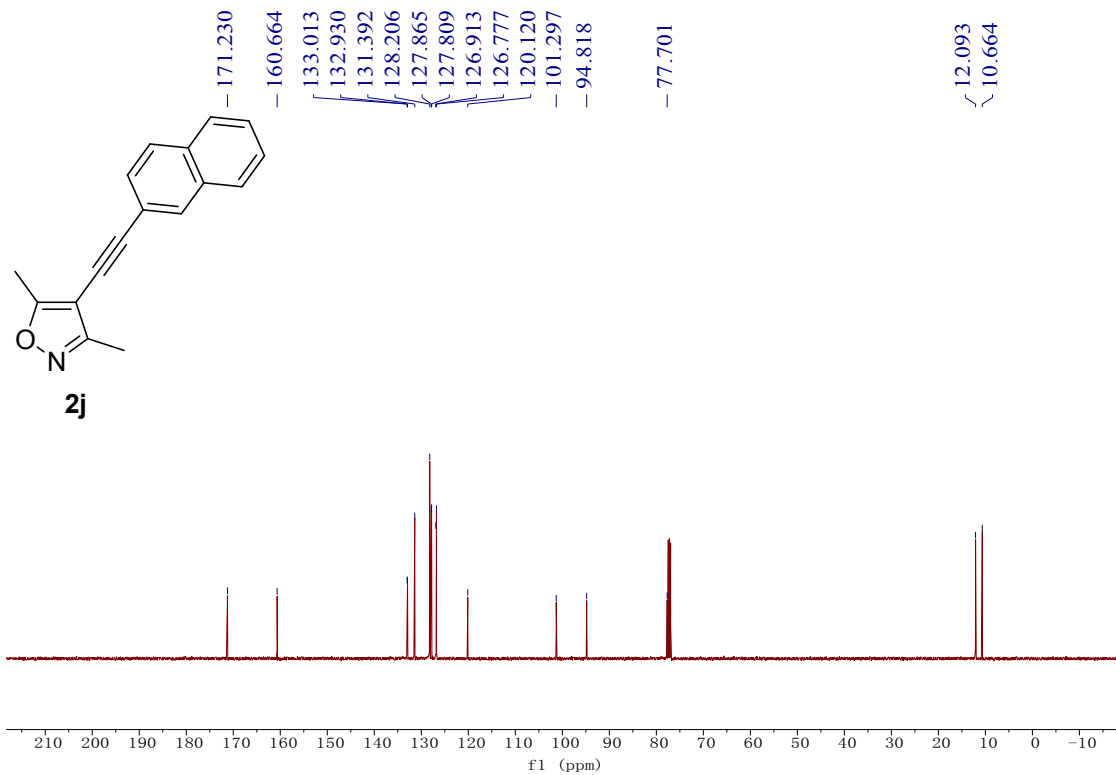


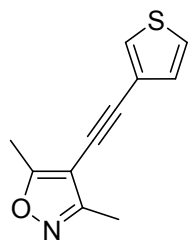






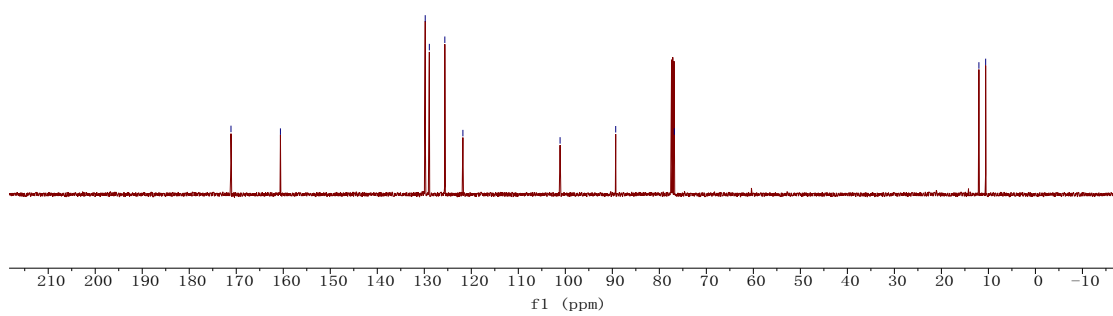


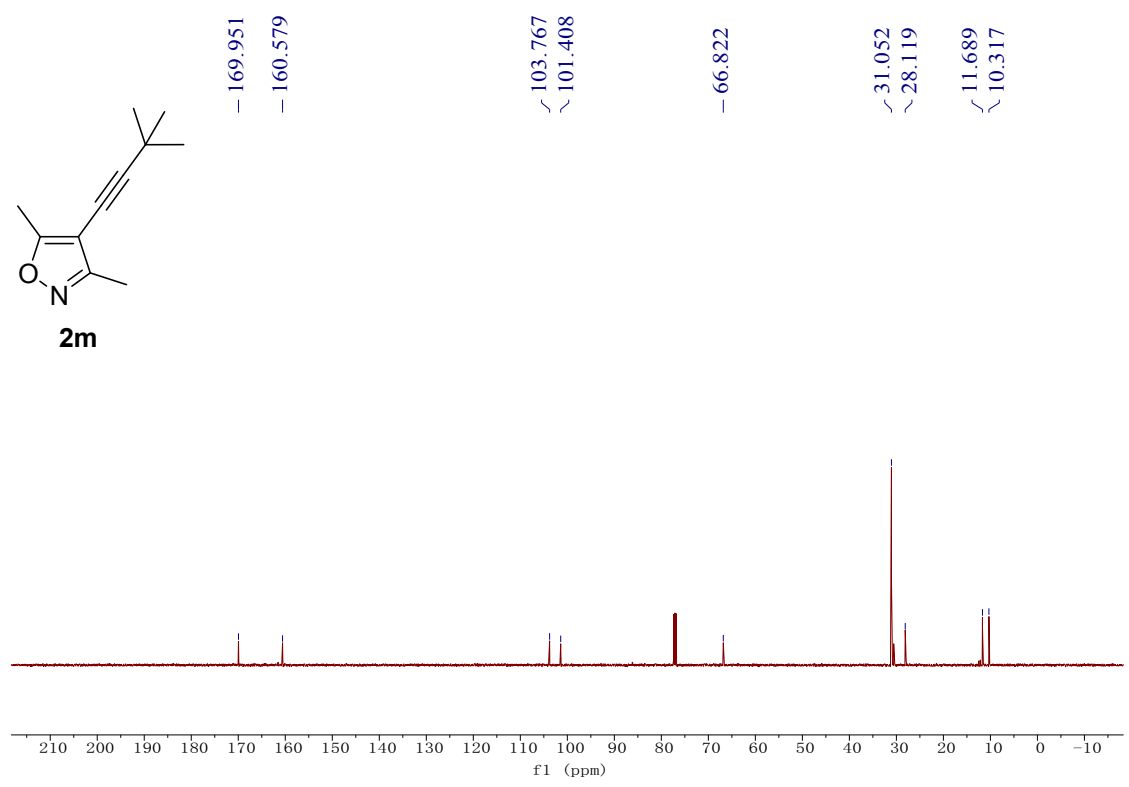
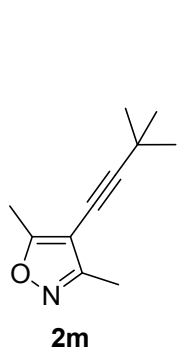
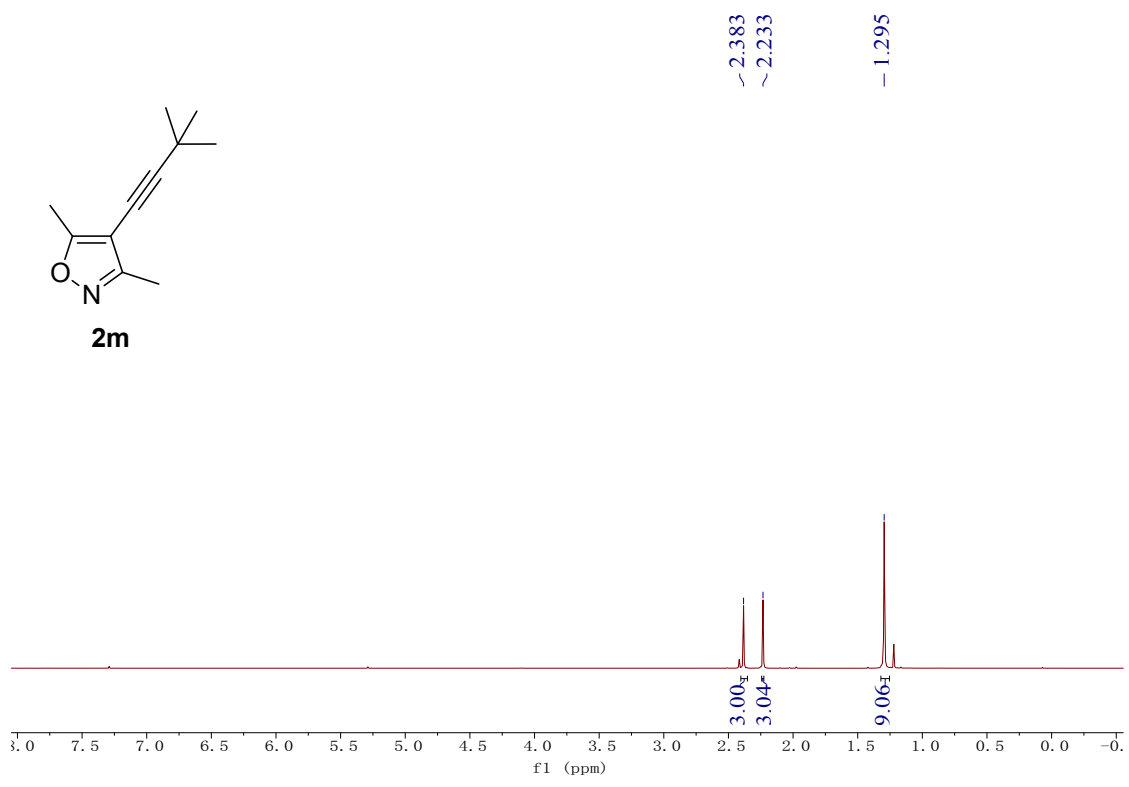
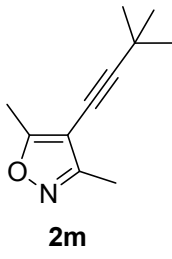


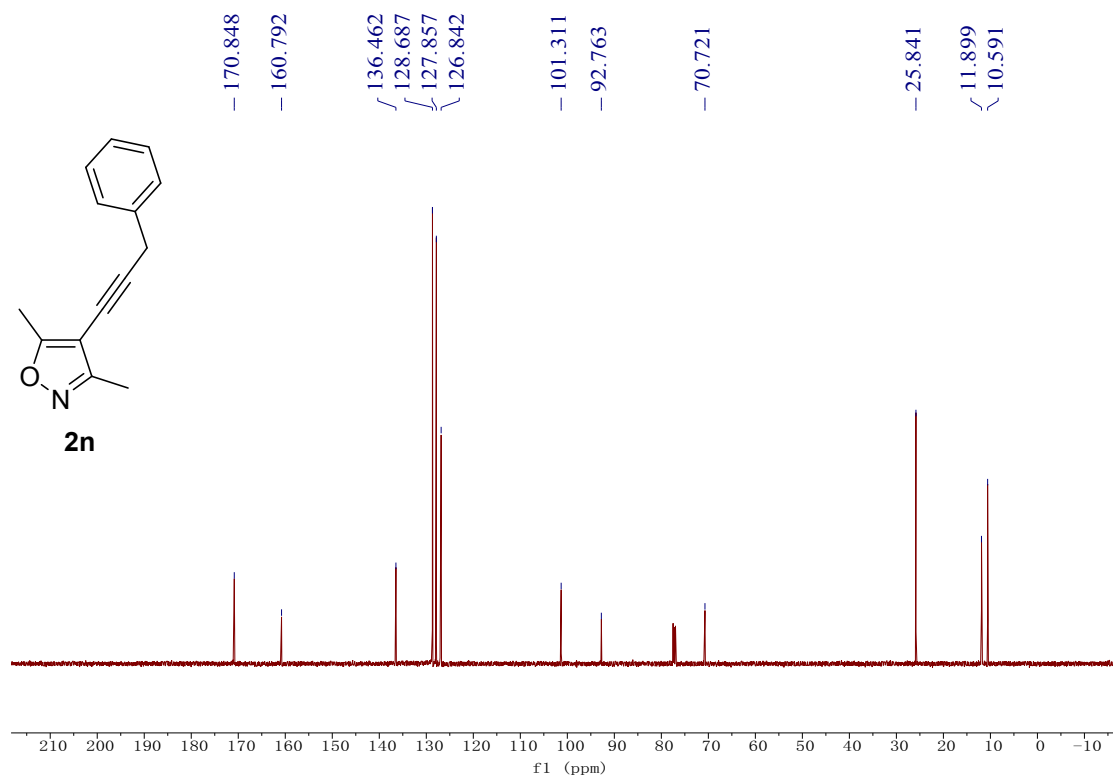
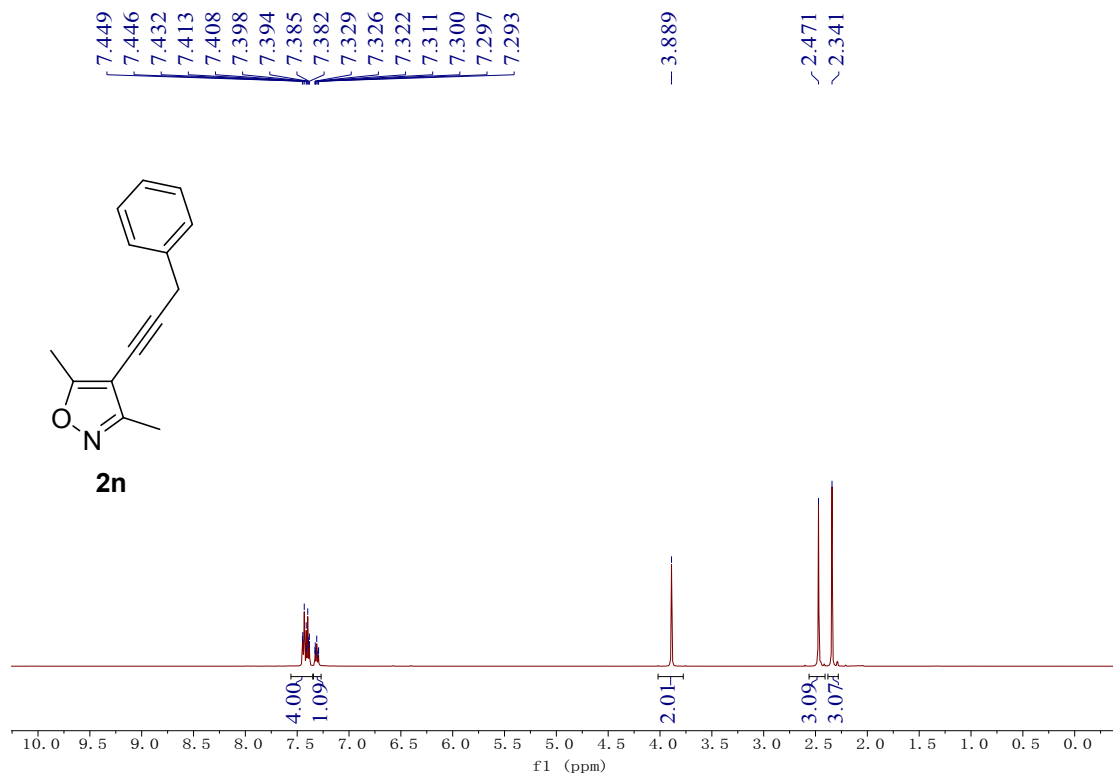


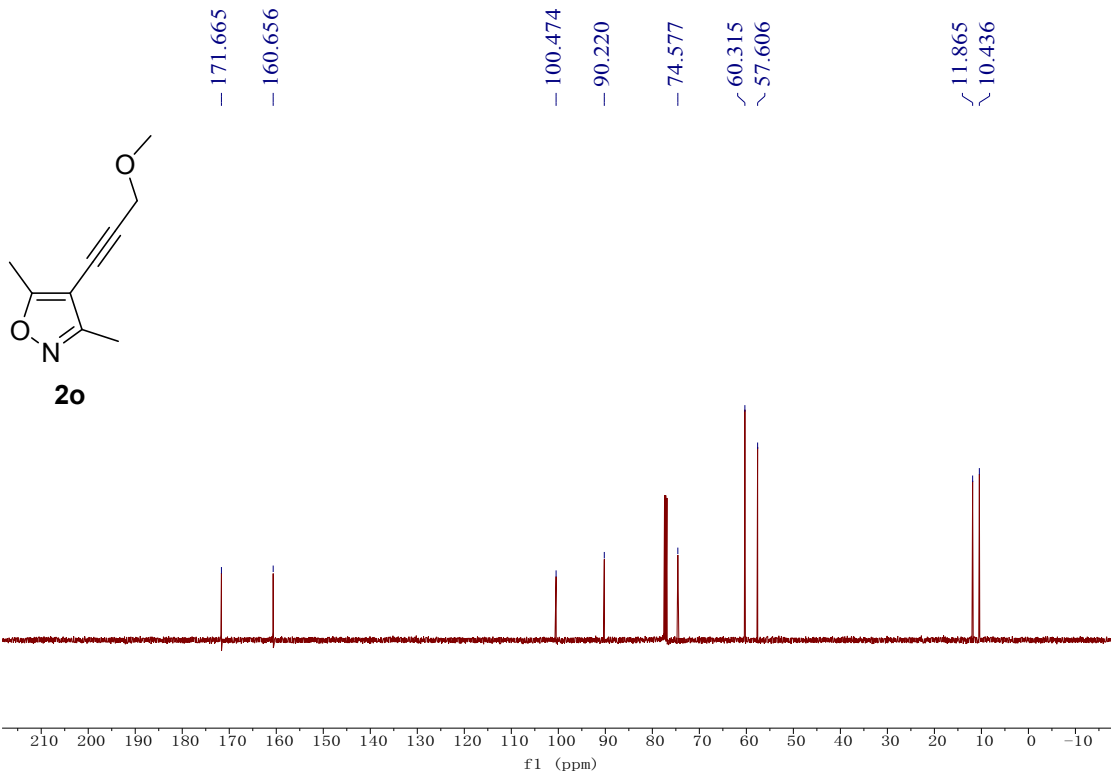
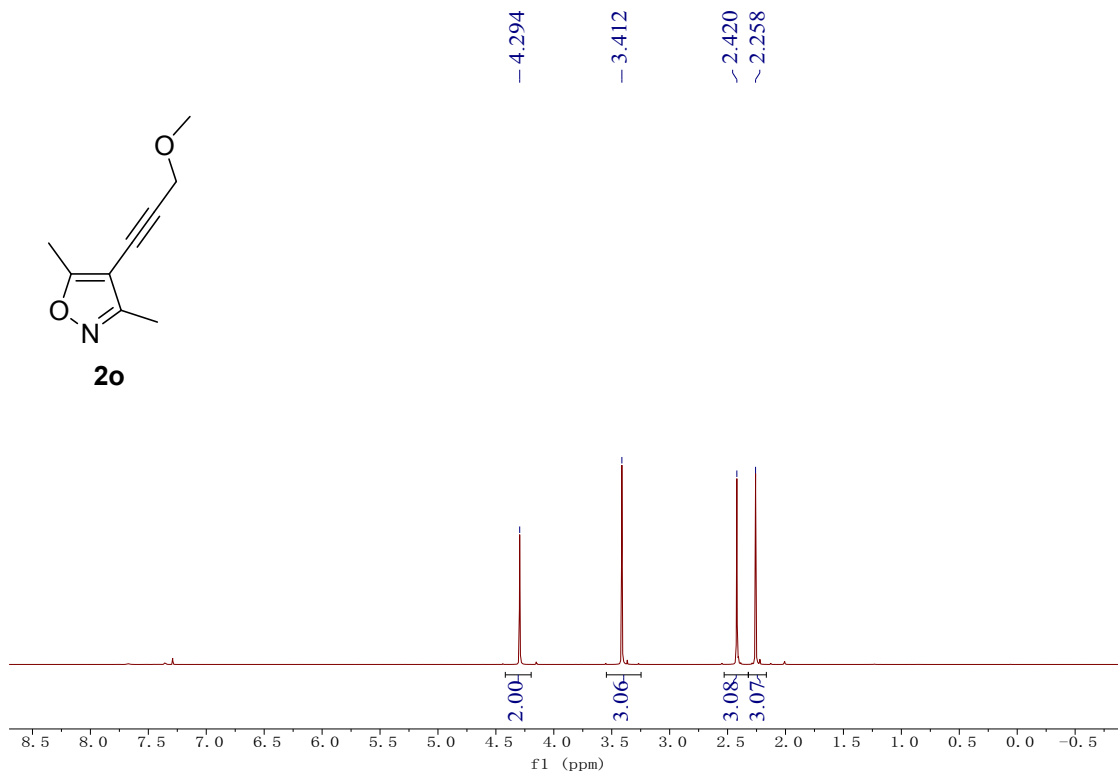
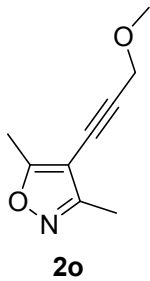
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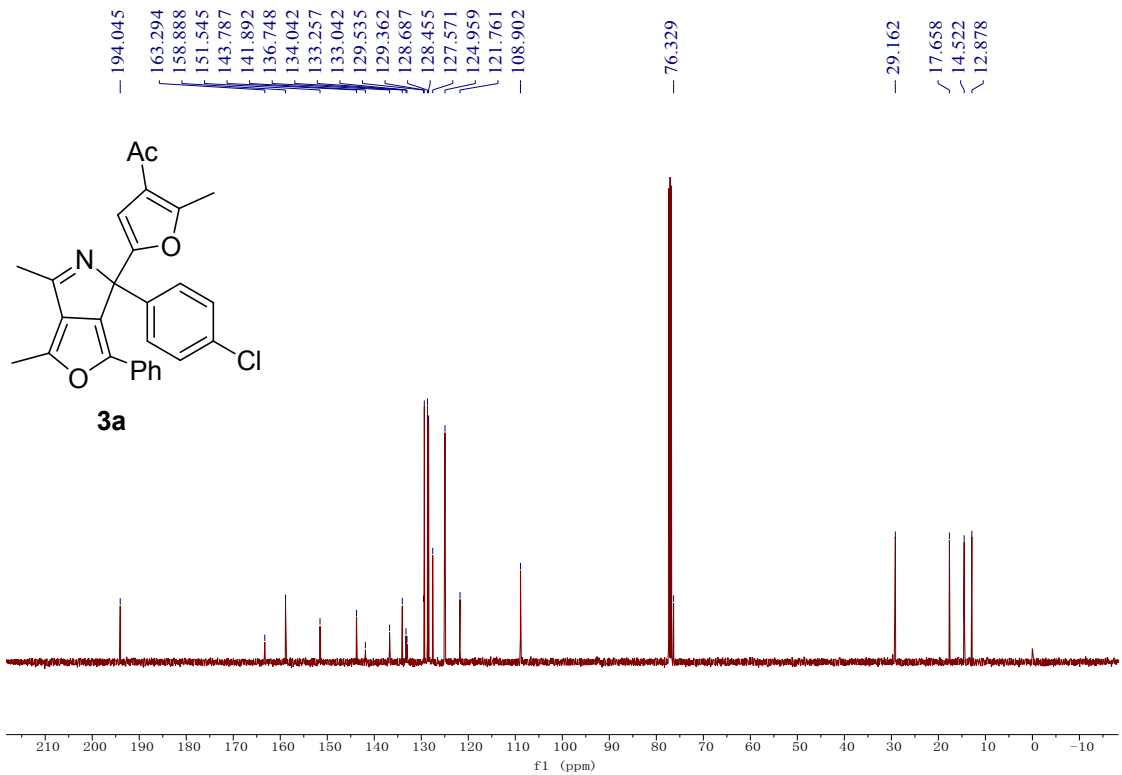
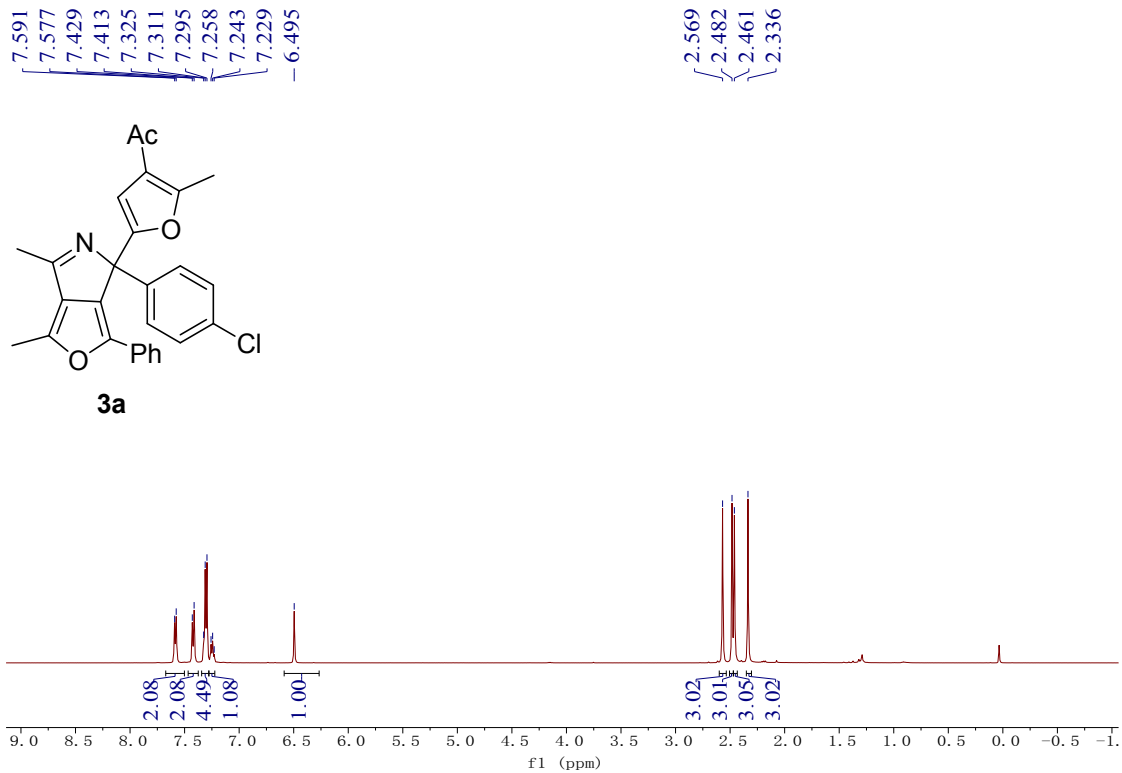
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/ 12.021
/ 10.584



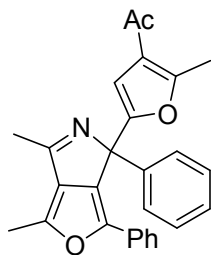




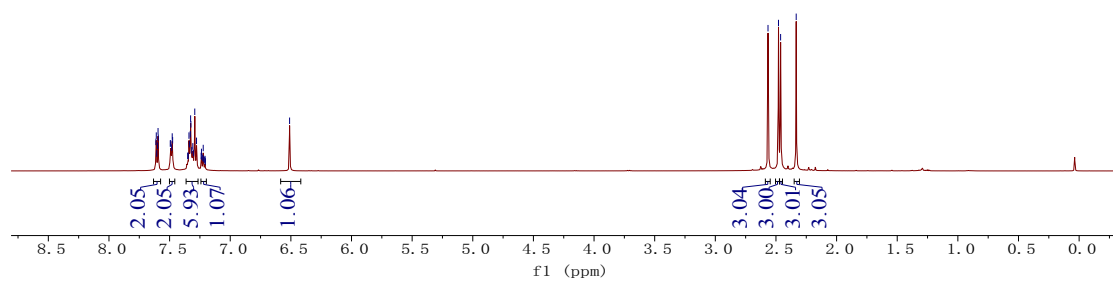




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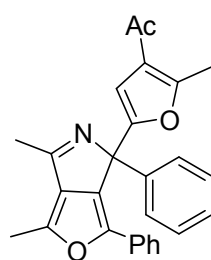
3b



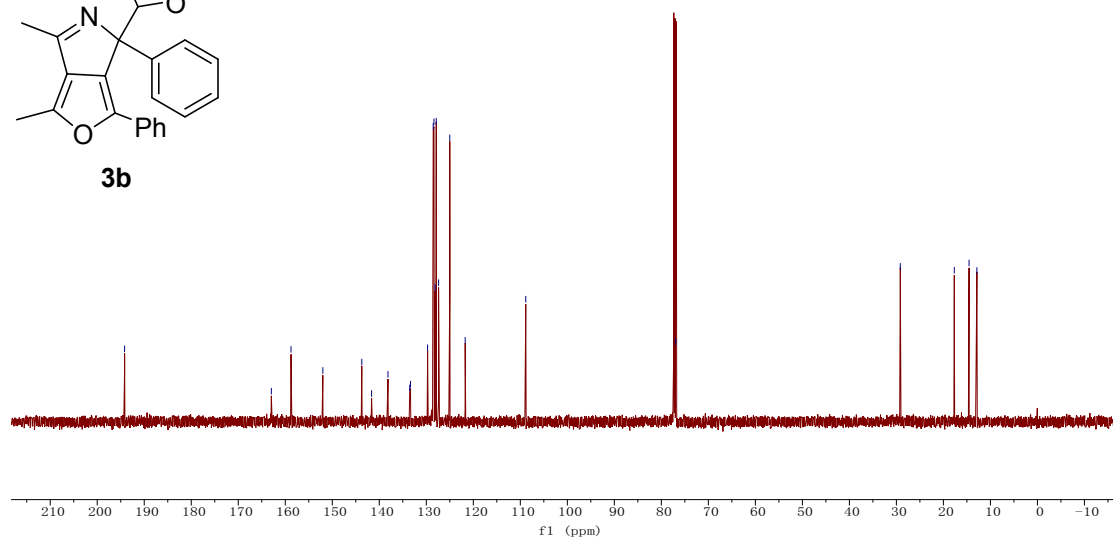
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76.950

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3b



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