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Convenient preparation of (*E*)-3-arylidene-4-diazopyrrolidine-2,5-diones in array format

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Experimental part

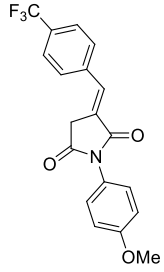
General considerations

All commercial reagents were used without purification. NMR spectra were recorded using Bruker Avance III spectrometer (^{1}H: 400.13 MHz; ^{13}C: 100.61 MHz; chemical shifts are reported as parts per million (δ, ppm); the residual solvent peaks were used as internal standards: 7.26 and 2.50 ppm for ^{1}H in $CDCl_3$ and DMSO-d_6 respectively, 39.52 and 77.16 ppm for ^{13}C in DMSO-d_6 and $CDCl_3$ respectively; multiplicities are abbreviated as follows: s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet, br = broad, dd = doublet of doublets, dt = doublet of triplets, ddd = doublet/doublets of doublets; coupling constants, *J*, are reported in Hz. Mass spectrum were recovered using Bruker microTOF spectrometer (ionization by electrospray, positive ions detection). Melting points were determined in open capillary tubes on Stuart SMP50 Automatic Melting Point Apparatus.

General procedure for the preparation of benzylidene succinimides 1 (GP1)

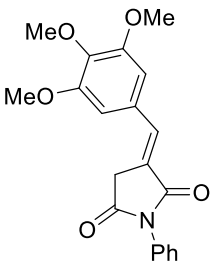
To a solution of corresponding maleimide (10 mmol) in methanol (50 ml) was added triphenylphosphine (11 mmol) and the mixture was stirred for 20 minutes followed by addition of the appropriate aldehyde (10.5 mmol). In several minutes the precipitated formed, the reaction mixture was stirred at ambient temperature for 4–16 hours. Upon cooling in ice bath the precipitate was filtered off, washed with methanol (20 ml) and dried in air to afford benzylidene succinimide **1** which was used in diazo transfer step without further purification.

(*E*)-1-(4-Methoxyphenyl)-3-(4-(trifluoromethyl)benzylidene)pyrrolidine-2,5-dione (1a): The

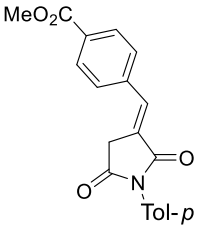


title compound was synthesized from 1-(4-methoxyphenyl)-1*H*-pyrrole-2,5-dione (812 mg, 4.0 mmol) and 4-(trifluoromethyl)benzaldehyde (766 mg, 4.4 mmol) according to GP1. Yield 1.07 g (74%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.78 – 7.75 (m, 3H, =CH and ArH), 7.66 (d, *J* = 8.2 Hz, 2H, ArH), 7.31 (d, *J* = 9.0 Hz, 2H, ArH), 7.04 (d, *J* = 9.0 Hz, 2H, ArH), 3.87 (s, 3H, OCH_3), 3.78 (d, *J* = 2.4 Hz, 2H, CH_2).

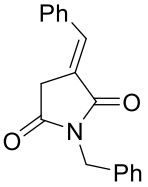
(*E*)-1-Phenyl-3-(3,4,5-trimethoxybenzylidene)pyrrolidine-2,5-dione [S1] **(1b):** The title compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione (692 mg, 4.0 mmol) and 3,4,5-trimethoxybenzaldehyde (864 mg 4.4 mmol) according to GP1. Yield 1.31 g (93%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.65 (t, *J* = 2.3 Hz, 1H, =CH), 7.51 (t, *J* = 7.5 Hz, 2H, 2ArH), 7.45 – 7.37 (m, 3H, 3ArH), 6.76 (s, 2H, 2ArH), 3.93 (s, 3H, OCH_3), 3.91 (s, 6H, $2OCH_3$), 3.77 (d, *J* = 2.3 Hz, 2H, CH_2).



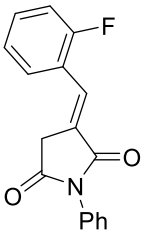
Methyl (*E*)-4-[(2,5-dioxo-1-(*p*-tolyl)pyrrolidin-3-ylidene)methyl]benzoate (1c): The title compound was synthesized from 1-(*p*-tolyl)-1*H*-pyrrole-2,5-dione (748 mg, 4.0 mmol) and methyl 4-formylbenzoate (722 mg, 4.4 mmol) according to GP1. Yield 1.19 g (89%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.16 (d, *J* = 8.3 Hz, 2H, 2ArH), 7.78 (t, *J* = 2.2 Hz, 1H, =CH), 7.62 (d, *J* = 8.4 Hz, 2H, 2ArH), 7.33 (d, *J* = 8.3 Hz, 2H, 2ArH), 7.27 (d, *J* = 7.8 Hz, 2H, 2ArH), 3.98 (s, 3H, $COOCH_3$), 3.79 (d, *J* = 2.4 Hz, 2H, CH_2), 2.43 (s, 3H, CH_3).



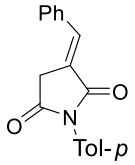
(*E*)-1-Benzyl-3-benzylidenepyrrolidine-2,5-dione [S2] **(1d):** The title compound was synthesized from 1-benzyl-1*H*-pyrrole-2,5-dione (748 mg, 4.0 mmol) and benzaldehyde (466 mg, 4.4 mmol) according to GP1. Yield 930 mg (84%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.65 (t, *J* = 2.4 Hz, 1H, =CH), 7.52 – 7.43 (m, 7H, ArH), 7.37 – 7.30 (m, 3H, ArH), 4.82 (s, 2H, NCH_2), 3.60 (d, *J* = 2.3 Hz, 2H, CH_2CO).



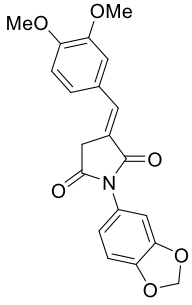
(*E*)-3-(2-Fluorobenzylidene)-1-phenylpyrrolidine-2,5-dione[1] **(1e):** The title compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione (692 mg, 4.0 mmol) and 2-fluorobenzaldehyde (544 mg, 4.4 mmol) according to GP1. Yield 1.04 g (93%), white solid. ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.79 (t, *J* = 7.6 Hz, 1H, =CH), 7.70 – 7.64 (m, 1H, ArH), 7.54 (t, *J* = 7.6 Hz, 3H, 3ArH), 7.48 – 7.42 (m, 1H, ArH), 7.40 – 7.35 (m, 4H, 4ArH), 3.86 (d, *J* = 2.2 Hz, 2H, CH_2).



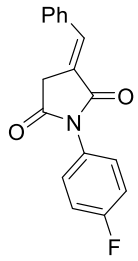
(*E*)-3-Benzylidene-1-(*p*-tolyl)pyrrolidine-2,5-dione [S3] **(1f):** The title compound was synthesized from 1-(*p*-tolyl)-1*H*-pyrrole-2,5-dione (748 mg, 4.0 mmol) and benzaldehyde (466 mg, 4.4 mmol) according to GP1. Yield 940 mg (85%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77 (s, 1H, =CH), 7.57 (d, *J* = 6.7 Hz, 2H, 2ArH), 7.54 – 7.45 (m, 3H, 3ArH), 7.36 – 7.27 (m, 4H, 4ArH), 3.78 (s, 2H, CH_2), 2.43 (s, 3H, CH_3).



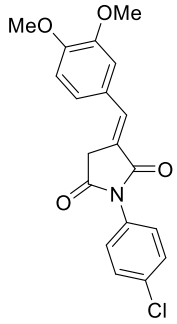
(*E*)-1-(Benzo[*d*][1,3]dioxol-5-yl)-3-(3,4-dimethoxybenzylidene)pyrrolidine-2,5-dione (1g): The title compound was synthesized from 1-(benzo[*d*][1,3]dioxol-5-yl)-1*H*-pyrrole-2,5-dione (868 mg, 4.0 mmol) and 3,4-dimethoxy-benzaldehyde (730 mg, 4.4 mmol) according to GP1. Yield 1.38 g (94%), white solid. 1H NMR (400 MHz, $CDCl_3$) δ 7.71 – 7.66 (m, 1H, =CH), 7.29 (d, *J* = 1.7 Hz, 1H, ArH), 7.18 (d, *J* = 8.3 Hz, 1H, ArH), 7.07 (s, 1H, ArH), 7.02 – 6.96 (m, 1H, ArH), 6.92 (dd, *J* = 8.7, 1.7 Hz, 1H, ArH), 6.86 (dd, *J* = 6.3, 1.8 Hz, 1H, ArH), 6.04 (d, *J* = 1.6 Hz, 2H, CH_2), 3.97 (s, 3H, OCH_3), 3.96 (s, 3H, OCH_3), 3.74 (s, 2H, CH_2).

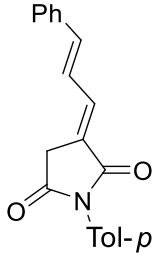


(*E*)-3-Benzylidene-1-(4-fluorophenyl)pyrrolidine-2,5-dione (1h): The title compound was synthesized from 1-(4-fluorophenyl)-1*H*-pyrrole-2,5-dione (764 mg, 4.0 mmol) and benzaldehyde (466 mg, 4.4 mmol) according to GP1. Yield 976 mg (87%), white solid. 1H NMR (400 MHz, $CDCl_3$) δ 7.77 (t, *J* = 2.3 Hz, 1H, =CH), 7.59 – 7.54 (m, 2H, 2ArH), 7.54 – 7.47 (m, 3H, 3ArH), 7.40 (dd, *J* = 9.0, 4.8 Hz, 2H, 2ArH), 7.21 (t, *J* = 8.6 Hz, 2H, 2ArH), 3.78 (d, *J* = 2.4 Hz, 2H, CH_2).



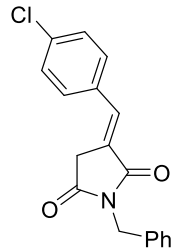
(*E*)-1-(4-Chlorophenyl)-3-(3,4-dimethoxybenzylidene)pyrrolidine-2,5-dione (1i): The title compound was synthesized from 1-(4-chlorophenyl)-1*H*-pyrrole-2,5-dione (828 mg, 4.0 mmol) and 3,4-dimethoxybenzaldehyde (730 mg, 4.4 mmol) according to GP1. Yield 1.23 g (86%), white solid. 1H NMR (400 MHz, $CDCl_3$) δ 7.54 – 7.49 (m, 1H, =CH), 7.40 (d, *J* = 8.7 Hz, 2H, 2ArH), 7.37 (d, *J* = 8.4 Hz, 2H, 2ArH), 7.13 – 710 (m, 2H, 2ArH), 7.00 – 6.97 (m, 1H, ArH), 3.91 (s, 3H, OCH_3), 3.89 (s, 3H, OCH_3), 3.67 (d, *J* = 2.2 Hz, 2H, CH_2).



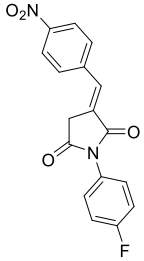


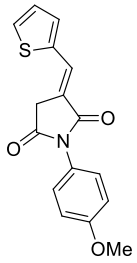
(*E*)-3-((*E*)-3-Phenylallylidene)-1-(*p*-tolyl)pyrrolidine-2,5-dione (1j): The title compound was synthesized from 1-(*p*-tolyl)-1*H*-pyrrole-2,5-dione (748 mg, 4.0 mmol) and cinnamaldehyde (580 mg, 4.4 mmol) according to GP1. Yield 872 mg (72%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.55 (d, *J* = 6.9 Hz, 2H, 2ArH), 7.50 – 7.45 (m, 1H, CH) 7.45 – 7.37 (m, 3H, 3ArH), 7.33 – 7.26 (m, 4H, 4ArH), 7.07 (d, *J* = 15.5 Hz, 1H, CH), 6.87 (dd, *J* = 15.4, 11.5 Hz, 1H, CH), 3.59 (d, *J* = 2.1 Hz, 2H, CH_2), 2.42 (s, 3H, CH_3).

(*E*)-1-Benzyl-3-(4-chlorobenzylidene)pyrrolidine-2,5-dione [S4] **(1k):** The title compound was synthesized from 1-benzyl-1*H*-pyrrole-2,5-dione (748 mg, 4.0 mmol) and 4-chlorobenzaldehyde (618 mg, 4.4 mmol) according to GP1. Yield 1.05 g (84%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.59 (t, *J* = 2.4 Hz, 1H, =CH), 7.48 – 7.40 (m, 6H, ArH), 7.37 – 7.28 (m, 3H, ArH), 4.81 (s, 2H, NCH_2), 3.56 (d, *J* = 2.4 Hz, 2H, CH_2CO).



(*E*)-1-(4-Fluorophenyl)-3-(4-nitrobenzylidene)pyrrolidine-2,5-dione (1l): The title compound was synthesized from 1-(4-fluorophenyl)-1*H*-pyrrole-2,5-dione (764 mg, 4.0 mmol) and 4-nitrobenzaldehyde (664 mg, 4.4 mmol) according to GP1. Yield 834 mg (64%), white solid. ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.30 (d, *J* = 8.8 Hz, 2H, 2ArH), 7.99 (d, *J* = 8.9 Hz, 2H, 2ArH), 7.70 (t, *J* = 2.3 Hz, 1H, =CH), 7.46 – 7.35 (m, 4H, 4ArH), 3.93 (d, *J* = 2.4 Hz, 2H, CH_2).





(*E*)-1-(4-Methoxyphenyl)-3-(thiophen-2-ylmethylene)pyrrolidine-2,5-dione (1m): The title compound was synthesized from 1-(4-methoxyphenyl)-1*H*-pyrrole-2,5-dione (812 mg, 4.0 mmol) and thiophene-2-carbaldehyde (493 mg, 4.4 mmol) according to GP1. Yield 825 mg (69%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.94 (t, *J* = 2.3 Hz, 1H, =CH), 7.65 (d, *J* = 5.1 Hz, 1H, ArH), 7.43 (d, *J* = 3.7 Hz, 1H, ArH), 7.31 (d, *J* = 9.0 Hz, 2H, ArH), 7.21 (dd, *J* = 5.1, 3.7 Hz, 1H, ArH), 7.03 (d, *J* = 9.0 Hz, 2H, ArH), 3.86 (s, 3H, OCH_3), 3.68 (d, *J* = 2.3 Hz, 2H, CH_2).

(*E*)-3-(4-(Dimethylamino)benzylidene)-1-phenylpyrrolidine-2,5-dione (1n): The title compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 4-(dimethylamino)-benzaldehyde according to GP1. Yield 2.56 g (84%), yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (t, *J* = 2.3 Hz, 1H, =CH), 7.58 – 7.45 (m, 4H), 7.44 – 7.39 (m, 3H), 6.78 (d, *J* = 8.9 Hz, 2H), 3.74 (d, *J* = 2.2 Hz, 2H, CH_2), 3.09 (s, 6H, $N(CH_3)_2$).

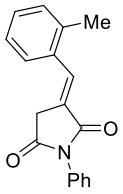
(*E*)-3-(4-Methoxybenzylidene)-1-phenylpyrrolidine-2,5-dione [S5] **(1o):** The title compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 4-methoxybenzaldehyde according to GP1. Yield 2.49 g (85%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (t, *J* = 2.4 Hz, 1H, =CH), 7.56 – 7.49 (m, 4H), 7.46 – 7.38 (m, 3H), 7.02 (d, *J* = 8.8 Hz, 2H), 3.90 (s, 3H, OCH_3), 3.75 (d, *J* = 2.3 Hz, 2H, CH_2).

(*E*)-3-(3-Methoxybenzylidene)-1-phenylpyrrolidine-2,5-dione [S2] **(1p):** The title compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 3-methoxybenzaldehyde according to GP1. Yield 2.40 g (82%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.74 (t, *J* = 2.5 Hz, 1H, =CH), 7.53 (t, *J* = 7.5 Hz, 2H), 7.47 – 7.39 (m, 4H), 7.16 (dt, *J* = 8.0, 1.2 Hz, 1H), 7.07 (t, *J* = 2.1 Hz, 1H), 7.02 (ddd, *J* = 8.2, 2.6, 0.9 Hz, 1H), 3.89 (s, 3H, OCH_3), 3.79 (d, *J* = 2.4 Hz, 2H, CH_2).

(*E*)-3-(2-Methoxybenzylidene)-1-phenylpyrrolidine-2,5-dione (1q): The title compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 2-methoxybenzaldehyde according to GP1. Yield 2.29 g (78%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.18 (t, *J* = 2.5 Hz, 1H, =CH), 7.58 – 7.38 (m, 7H), 7.06 (t, *J* = 7.6 Hz, 1H), 7.00 (d, *J* = 8.3 Hz, 1H), 3.94 (s, 3H, OCH_3), 3.73 (d, *J* = 2.5 Hz, 2H, CH_2).

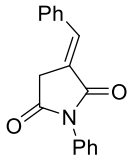
(*E*)-3-(4-Methylbenzylidene)-1-phenylpyrrolidine-2,5-dione [S2] **(1r):** The title compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 4-methylbenzaldehyde according to GP1. Yield 2.41 g (87%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.74 (t, *J* = 2.5 Hz, 1H, =CH), 7.55 – 7.50 (m, 2H), 7.48 – 7.39 (m, 5H), 7.31 (d, *J* = 8.1 Hz, 2H), 3.77 (d, *J* = 2.4 Hz, 2H, CH_2), 2.44 (s, 3H, CH_3).

(*E*)-3-(2-Methylbenzylidene)-1-phenylpyrrolidine-2,5-dione [S6] **(1s):** The title compound



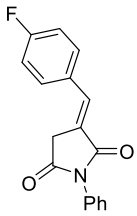
was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 2-methylbenzaldehyde according to GP1. Yield 2.23 g (80%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.01 (t, *J* = 2.5 Hz, 1H, =CH), 7.56 – 7.50 (m, 2H), 7.48 – 7.40 (m, 4H), 7.38 – 7.29 (m, 3H), 3.73 (d, *J* = 2.4 Hz, 2H, CH_2), 2.50 (s, 3H), CH_3).

(*E*)-3-Benzylidene-1-phenylpyrrolidine-2,5-dione [S5] **(1t):** The title compound was



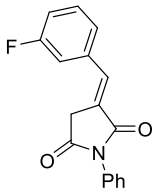
synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and benzaldehyde according to GP1. Yield 2.16 g (82%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77 (t, *J* = 2.5 Hz, 1H, =CH), 7.59 – 7.39 (m, 10H), 3.78 (d, *J* = 2.5 Hz, 2H, CH_2).

(*E*)-3-(4-Fluorobenzylidene)-1-phenylpyrrolidine-2,5-dione [S2] **(1u):** The title compound



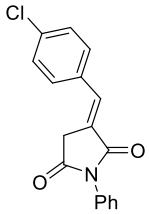
was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 4-fluorobenzaldehyde according to GP1. Yield 2.42 g (86%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.74 (t, *J* = 2.5 Hz, 1H, =CH), 7.59 – 7.50 (m, 4H), 7.47 – 7.38 (m, 3H), 7.20 (t, *J* = 8.6 Hz, 2H), 3.75 (d, *J* = 2.4 Hz, 2H, CH_2).

(*E*)-3-(3-Fluorobenzylidene)-1-phenylpyrrolidine-2,5-dione [S1] **(1v):** The title compound



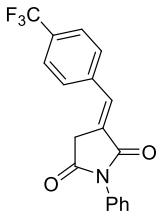
was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 3-fluorobenzaldehyde according to GP1. Yield 2.28 g (81%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 (t, *J* = 2.5 Hz, 1H, =CH), 7.56 – 7.39 (m, 6H), 7.35 (d, *J* = 7.9 Hz, 1H), 7.25 (dt, *J* = 9.8, 2.1 Hz, 1H), 7.18 (tdd, *J* = 8.4, 2.6, 0.9 Hz, 1H), 3.78 (d, *J* = 2.5 Hz, 2H, CH_2).

(*E*)-3-(4-Chlorobenzylidene)-1-phenylpyrrolidine-2,5-dione [S7] **(1w):** The title compound



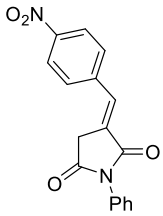
was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 4-chlorobenzaldehyde according to GP1. Yield 2.68 g (90%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (t, *J* = 2.5 Hz, 1H, =CH), 7.56 – 7.50 (m, 2H), 7.49 (s, 4H), 7.47 – 7.39 (m, 3H), 3.76 (d, *J* = 2.5 Hz, 2H, CH_2).

(*E*)-1-Phenyl-3-(4-(trifluoromethyl)benzylidene)pyrrolidine-2,5-dione [S2]**: (1x)** The title



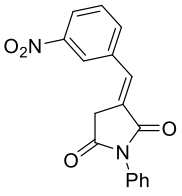
compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 4-(trifluoromethyl)-benzaldehyde according to GP1. Yield 2.69 g (81%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 – 7.65 (m, 3H), 7.61 (d, *J* = 8.2 Hz, 2H), 7.45 (t, *J* = 7.6 Hz, 2H), 7.40 – 7.28 (m, 3H), 3.74 (d, *J* = 2.5 Hz, 2H, CH_2).

(*E*)-3-(4-Nitrobenzylidene)-1-phenylpyrrolidine-2,5-dione [S1] **(1y):** The title compound was



synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 4-nitrobenzaldehyde according to GP1. Yield 2.68 g (90%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.25 (d, *J* = 8.8 Hz, 2H), 7.72 (d, *J* = 8.8 Hz, 2H), 7.69 (t, *J* = 2.5 Hz, 1H, =CH), 7.49 – 7.40 (m, 2H), 7.39 – 7.34 (m, 1H), 7.33 – 7.28 (m, 2H), 3.80 (d, *J* = 2.5 Hz, 2H, CH_2).

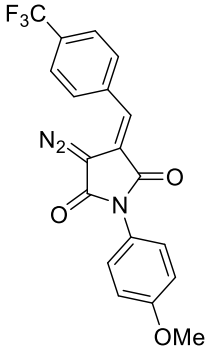
(*E*)-3-(3-Nitrobenzylidene)-1-phenylpyrrolidine-2,5-dione [8] **(1z):** The title compound was synthesized from 1-phenyl-1*H*-pyrrole-2,5-dione and 3-nitrobenzaldehyde according to GP1. Yield 2.87 g (93%), white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.34 (t, *J* = 2.0 Hz, 1H), 8.22 (ddd, *J* = 8.3, 2.3, 1.0 Hz, 1H), 7.91 – 7.86 (m, 1H), 7.69 (t, *J* = 2.5 Hz, 1H, =CH), 7.66 (t, *J* = 8.0 Hz, 1H), 7.49 – 7.41 (m, 2H), 7.39 – 7.34 (m, 1H), 7.34 – 7.30 (m, 2H), 3.82 (d, *J* = 2.5 Hz, 2H, CH_2).



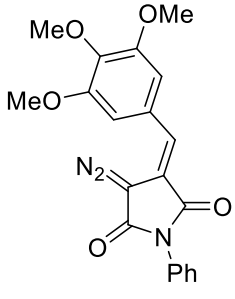
Preparation of diazo benzylidene succinimides 2 (GP2)

To a stirred solution/suspension of imide **1** (2 mmol) in CH_2Cl_2 (15 ml) were added 4-nitrophenylsulfonyl azide (479 mg, 2.1 mmol) and DBU (313 µl, 2.1 mmol), and the mixture was stirred at ambient temperature for 1–2 hours (controlled by TLC). The resulting mixture was subjected to flash column chromatography on silica gel eluting with CH_2Cl_2 to afford pure diazo compound **2**.

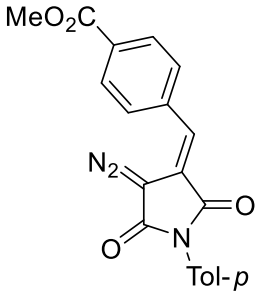
(*E*)-3-Diazo-1-(4-methoxyphenyl)-3-(4-trifluoromethylbenzylidene)pyrrolidine-2,5-dione (2a): The title compound was synthesized from imide **1a** (722 mg, 2.0 mmol) according to GP2. Yield: 623 mg (81%). Orange solid; m.p. 154.0–156.1 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.74 (s, 1H, =CH), 7.74 (d, *J* = 8.1 Hz, 2H, ArH), 7.49 (d, *J* = 8.1 Hz, 2H, ArH), 7.34 (d, *J* = 9.0 Hz, 2H, ArH), 7.03 (d, *J* = 9.0 Hz, 2H, ArH), 3.87 (s, 3H, OCH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.8, 163.9, 159.6, 136.9, 131.2 (q, *J* = 32.9 Hz), 129.1, 127.8, 125.7 (q, *J* = 3.7 Hz), 125.6, 124.2, 123.68 (q, *J* = 272.3 Hz), 118.8, 114.6, 60.1, 55.5. HRMS (ESI), *m/z* calcd for $C_{19}H_{12}F_3N_3NaO_3$ $[M+Na]^+$ 410.0723 found 410.0717.



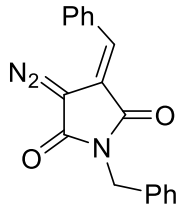
(*E*)-3-Diazo-1-phenyl-4-(3,4,5-trimethoxybenzylidene)pyrrolidine-2,5-dione (**2b**): The title compound was synthesized from imide **1b** (706 mg, 2.0 mmol) according to GP2. Yield: 697 mg (92%). Red-orange solid; m.p. 108.8–111.6 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.69 (s, 1H, =CH), 7.56 – 7.47 (m, 2H, 2ArH), 7.44 – 742 (m 3H, 3ArH), 6.59 (s, 2H, 2ArH), 3.93 (s, 6H, $2OCH_3$) 3.92 (s, 3H, OCH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.2, 164.0, 153.4, 139.5, 131.8, 129.2, 128.5, 126.4, 115.5, 106.4, 67.1, 61.0, 60.2, 56.1. HRMS (ESI), *m/z* calcd for $C_{20}H_{18}N_3O_5$ $[M+H]^+$ 380.1241 found 380.1263 calcd for $C_{20}H_{17}N_3NaO_5$ $[M+Na]^+$ 402.1060 found 402.1048.



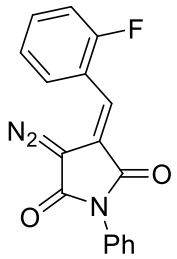
Methyl (*E*)-4-{[4-diazo-2,5-dioxo-1-(*p*-tolyl)pyrrolidin-3-ylidene]methyl}benzoate (**2c**): The title compound was synthesized from imide **1c** (642 mg, 2.0 mmol) according to GP2. Yield: 628 mg (87%). Yellow solid; m.p. 186.0–189.5 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 (d, *J* = 8.0 Hz, 2H, ArH), 7.74 (s, 1H, =CH), 7.43 (d, *J* = 8.0 Hz, 2H, ArH), 7.30 (d, *J* = 10.0 Hz, 4H, ArH), 3.97 (s, 3H, $COOCH_3$), 2.42 (s, 1H, CH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.2, 165.8, 163.9, 138.8, 137.7, 130.7, 129.8, 129.8, 129.0, 128.8, 126.2, 126.2, 118.5, 60.3, 52.4, 21.2. HRMS (ESI), *m/z* calcd for $C_{20}H_{17}N_3NaO_5$ $[M+Na]^+$ 402.1060 found 402.1048.



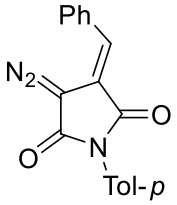
(*E*)-1-Benzyl-3-benzylidene-3-diazopyrrolidine-2,5-dione (2d): The title compound was synthesized from imide **1d** (554 mg, 2.0 mmol) according to GP2. Yield 503 mg (83%). Orange solid; m.p. 100.3–101.4 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (s, 1H, =CH), 7.47 – 7.42 (m, 4H, ArH), 7.40 – 7.31 (m, 6H, ArH), 4.84 (s, 2H, CH_2). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.8, 164.6, 135.8, 133.4, 129.4, 128.9, 128.8, 128.7, 128.7, 128.0, 127.7, 117.2, 59.7, 42.9. HRMS (ESI), *m/z* calcd for $C_{18}H_{13}N_3NaO_2$ $[M+Na]^+$ 326.0900 found 326.0891.



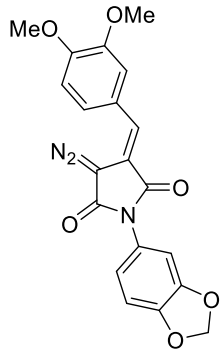
(*E*)-3-Diazo-4-(2-fluorobenzylidene)-1-phenylpyrrolidine-2,5-dione (**2e**): The title compound was synthesized from imide **1e** (562 mg, 2.0 mmol) according to GP2. Yield 602 mg (98%). Yellow solid; m.p. 145.9–146.4 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 (s, 1H, =CH), 7.56 – 7.49 (m, 2H, ArH), 7.47 – 7.39 (m, 4H, ArH), 7.33 (td, *J* = 7.5, 1.9 Hz, 1H, ArH), 7.26 (td, *J* = 7.5, 1.1 Hz, 1H, ArH), 7.22 – 7.16 (m, 1H, ArH). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.8, 163.6, 160.3 (d, *J* = 251.0 Hz), 132.1 (d, *J* = 9.9 Hz), 131.8, 131.5 (d, *J* = 8.5 Hz), 130.3 (d, *J* = 2.0 Hz), 129.2, 128.6, 128.4, 126.5, 124.3 (d, *J* = 3.5 Hz), 118.9, 116.2 (d, *J* = 21.4 Hz), 60.9. HRMS (ESI), *m/z* calcd for $C_{17}H_{11}FN_3O_2$ $[M+H]^+$ 308.0830 found 308.0822.



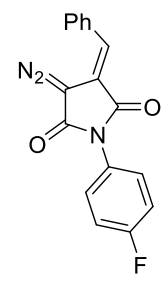
(*E*)-3-Benzylidene-4-diazo-1-(*p*-tolyl)pyrrolidine-2,5-dione (**2f**): The title compound was synthesized from imide **1f** (554 mg, 2.0 mmol) according to GP2. Yield 551 mg (91%). Yellow solid; m.p. 116.5–117.4 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77 (s, 1H, =CH), 7.50 – 7.45 (m, 2H, ArH), 7.44 – 7.36 (m, 3H, ArH), 7.32 (s, 4H, ArH), 2.43 (s, 3H, CH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.3, 164.1, 138.6, 133.4, 129.8, 129.5, 129.2, 129.0, 128.7, 128.0, 126.3, 116.7, 60.1, 21.2. HRMS (ESI), *m/z* calcd for $C_{18}H_{14}N_3O_2$ $[M+H]^+$ 304.1081 found 304.1086.



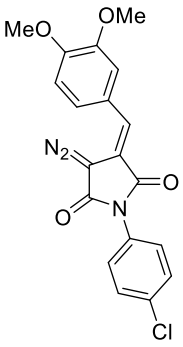
(*E*)-1-(Benzo[*d*][1,3]dioxol-5-yl)-3-diazo-4-(3,4-dimethoxybenzylidene)pyrrolidine-2,5-dione (**2g**): The title compound was synthesized from imide **1g** (678 mg, 2.0 mmol) according to GP2, but the product was purely soluble in DCM and it was filtered, washed with DCM (5 mL) and dried *in vacuo*. Yield 747 mg (95%). Orange solid; m.p. 174.5–176.3 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (s, 1H, =CH), 7.00 – 6.90 (m, 3H, ArH), 6.89 – 6.86 (m, 3H, ArH), 6.05 (s, 2H, CH_2), 3.96 (s, 3H, OCH_3), 3.95 (s, 3H, OCH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.6, 164.3, 150.5, 149.2, 148.1, 147.8, 128.8, 126.3, 125.3, 122.8, 120.6, 114.4, 111.8, 111.1, 108.4, 107.9, 101.8, 60.1, 56.1, 55.8. HRMS (ESI), *m/z* calcd for $C_{20}H_{15}N_3NaO_6$ $[M+Na]^+$ 416.0853 found 416.0864.



(*E*)-3-Benzylidene-4-diazo-1-(4-fluorophenyl)pyrrolidine-2,5-dione (**2h**): The title compound was synthesized from imide **1h** (562 mg, 2.0 mmol) according to GP2, yield 546 mg (89%); and according to GP3, yield 362 mg (59%). Light orange solid; m.p. 125.1–126.6 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77 (s, 1H, =CH), 7.55 – 7.36 (m, 7H, ArH), 7.19 (t, *J* = 8.6 Hz, 2H, ArH). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.1, 163.9, 162.1 (d, *J* = 248.7 Hz), 133.3, 129.0, 128.8, 128.4, 128.3 (d, *J* = 9.1 Hz), 127.7 (d, *J* = 2.8 Hz), 124.9, 116.5, 116.2 (d, *J* = 22.9 Hz), 60.2. HRMS (ESI), *m/z* calcd for $C_{17}H_{11}FN_3O_2$ $[M+H]^+$ 308.0830 found 308.0842.

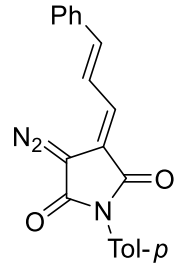


(*E*)-1-(4-Chlorophenyl)-3-diazo-4-(3,4-dimethoxybenzylidene)pyrrolidine-2,5-dione (**2i**):

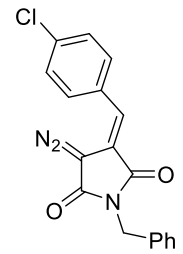


The title compound was synthesized from imide **1i** (714 mg, 2.0 mmol) according to GP2. Yield 684 mg (89%). Orange solid; m.p. 146.2–148.0 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (s, 1H, =CH), 7.48 (d, *J* = 8.8 Hz, 2H, 2ArH), 7.41 (d, *J* = 8.8 Hz, 2H, 2ArH), 6.98 (dd, *J* = 8.4, 1.8 Hz, 1H, ArH), 6.95 (d, *J* = 8.3 Hz, 1H, ArH), 6.87 (d, *J* = 1.7 Hz, 1H, ArH), 3.95 (s, 3H, OCH_3), 3.95 (s, 3H, OCH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.2, 163.8, 150.6, 149.2, 134.2, 130.4, 129.3, 129.1, 127.6, 126.1, 122.8, 114.1, 111.8, 111.1, 60.3, 56.0, 55.9. HRMS (ESI), *m/z* calcd for $C_{19}H_{15}ClN_3O_4$ $[M+H]^+$ 384.0746 found 384.0738.

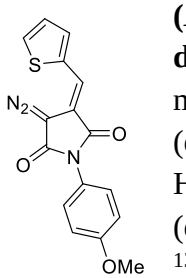
(*E*)-3-Diazo-4-(*E*-3-phenylallylidene)-1-(*p*-tolyl)pyrrolidine-2,5-dione (**2j**): The title compound was synthesized from imide **1j** (606 mg, 2.0 mmol) according to GP3. Yield 553 mg (84%). Light-orange solid; m.p. 178.0–180.1 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.49 – 7.35 (m, 6H, ArH, =CH), 7.33 – 7.27 (m, 4H, ArH), 7.03 (d, *J* = 15.2 Hz, 1H, =C<u>H</u>-Ph), 6.90 (dd, *J* = 15.1, 12.0 Hz, 1H, <u>H</u>C=CH-Ph), 2.42 (s, 3H, CH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.1, 163.9, 142.2, 138.6, 135.7, 129.8, 129.7, 129.2, 129.0, 127.9, 127.5, 126.3, 120.2, 114.4, 60.6, 21.2. HRMS (ESI), *m/z* calcd for $C_{20}H_{17}N_3NaO_5$ $[M+Na]^+$ 402.1060 found 402.1048.



(*E*)-1-Benzyl-3-(4-chlorobenzylidene)-4-diazopyrrolidine-2,5-dione (2k): The title compound was synthesized from imide **1k** (622 mg, 2.0 mmol) according to GP2. Yield 633 mg (94%). Orange solid; m.p. 134.5–135.1 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.59 (s, 1H, =CH), 7.48 – 7.39 (m, 4H, ArH), 7.39 – 7.29 (m, 3H, ArH), 7.24 (d, *J* = 8.5 Hz, 2H, ArH), 4.83 (s, 2H, CH_2). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.5, 164.4, 135.7, 135.5, 131.8, 130.1, 129.1, 128.8, 128.7, 128.1, 126.2, 117.7, 59.7, 42.9. HRMS (ESI), *m/z* calcd for $C_{18}H_{12}ClN_3NaO_2$ $[M+Na]^+$ 360.0510 found 360.0516.



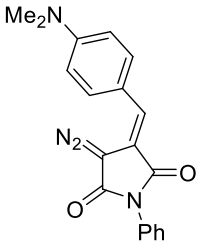
(*E*)-3-Diazo-1-(4-fluorophenyl)-4-(4-nitrobenzylidene)pyrrolidine-2,5-dione (**2l**): The title compound was synthesized from imide **1l** (652 mg, 2.0 mmol) according to GP2. Yield 380 mg (54%). Orange solid; m.p. 160.2–161.6 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.32 (d, *J* = 8.7 Hz, 2H, ArH), 7.80 (d, *J* = 8.7 Hz, 2H, ArH), 7.74 (s, 1H, =CH), 7.49 (dd, *J* = 9.0, 5.1 Hz, 2H, ArH), 7.39 (t, *J* = 8.8 Hz, 2H, ArH). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.2, 163.4, 162.3 (d, *J* = 249.3 Hz), 147.8, 139.6, 129.6, 128.3 (d, *J* = 8.8 Hz), 127.4 (d, *J* = 3.5 Hz), 124.7, 124.1, 119.7, 116.3 (d, *J* = 23.0 Hz), 60.5. HRMS (ESI), *m/z* calcd for $C_{17}H_9FN_4NaO_4$ $[M+Na]^+$ 375.0500 found 375.0518.



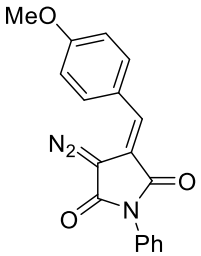
(*E*)-3-Diazo-1-(4-methoxyphenyl)-3-(thiophen-2-ylmethylene)pyrrolidine-2,5-dione (2m): The title compound was synthesized from imide **1m** (598 mg, 2.0 mmol) according to GP2. Yield 572 mg (88%). Orange solid; m.p. 138.0–140.5 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.85 (s, 1H, =CH), 7.57 (dd, *J* = 5.1, 1.0 Hz, 1H, ArH), 7.34 (d, *J* = 9.0 Hz, 2H, ArH), 7.26 (d, *J* = 3.7 Hz, 1H, ArH), 7.17 (dd, *J* = 5.1, 3.7 Hz, 1H, ArH), 7.02 (d, *J* = 9.0 Hz, 2H, ArH), 3.86 (s, 3H, OCH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.7, 164.5, 159.5, 137.2, 131.6, 130.4, 128.5,

127.8, 124.5, 120.8, 114.5, 113.8, 60.2, 55.5. HRMS (ESI), *m/z* calcd for $C_{16}H_{11}N_3NaO_3S$ $[M+Na]^+$ 348.0413 found 348.0415.

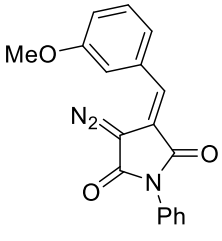
(*E*)-3-Diazo-4-(4-(dimethylamino)benzylidene)-1-phenylpyrrolidine-2,5-dione (2n): The title compound was synthesized from imide **1n** (613 mg, 2.0 mmol) according to GP2, yield 612 mg (92%). Dark orange solid; m.p. 118.2–120.0 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (s, 1H, =CH), 7.54 – 7.48 (m, 2H), 7.47 – 7.38 (m, 3H), 7.30 (d, *J* = 8.8 Hz, 2H), 6.74 (d, *J* = 8.9 Hz, 2H), 3.07 (s, 6H, $N(CH_3)_2$). ^{13}C NMR (101 MHz, $CDCl_3$) δ 167.2, 164.5, 151.1, 132.2, 131.2, 130.0, 129.1, 128.3, 126.6, 121.1, 111.8, 110.6, 60.2 (C=N_2), 40.1. HRMS (ESI), *m/z* calcd for $C_{19}H_{16}N_4NaO_2$ $[M+Na]^+$ 355.1165 found 355.1167.



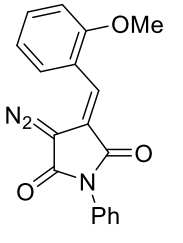
(*E*)-3-Diazo-4-(4-methoxybenzylidene)-1-phenylpyrrolidine-2,5-dione (2o): The title compound was synthesized from imide **1o** (587 mg, 2.0 mmol) according to GP2, yield 613 mg (96%). Orange solid; m.p. 113.5–114.4 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 (s, 1H, =CH), 7.54 – 7.49 (m, 2H), 7.46 – 7.40 (m, 3H), 7.34 (d, *J* = 8.7 Hz, 2H), 6.99 (d, *J* = 8.7 Hz, 2H), 3.88 (s, 3H, OCH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.5, 164.2, 160.8, 131.9, 130.8, 129.1, 128.5, 128.4, 126.5, 126.0, 114.40, 114.37, 60.1 (C=N_2), 55.5. HRMS (ESI), *m/z* calcd for $C_{18}H_{13}N_3NaO_3$ $[M+Na]^+$ 342.0849 found 342.0859.



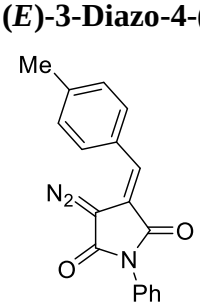
(*E*)-3-Diazo-4-(3-methoxybenzylidene)-1-phenylpyrrolidine-2,5-dione (2p): The title compound was synthesized from imide **1p** (587 mg, 2.0 mmol) according to GP2, yield 562 mg (88%). Orange solid; m.p. 120.0–120.8 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.75 (s, 1H, =CH), 7.56 – 7.48 (m, 2H), 7.47 – 7.41 (m, 3H), 7.38 (t, *J* = 7.9 Hz, 1H), 7.00 – 6.94 (m, 2H), 6.89 (t, *J* = 2.1 Hz, 1H), 3.88 (s, 3H, OCH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.2, 164.0, 159.8, 134.6, 131.8, 129.8, 129.2, 128.5, 128.1, 126.5, 121.5, 116.8, 115.5, 114.1, 60.3 (C=N_2), 55.3. HRMS (ESI), *m/z* calcd for $C_{18}H_{13}N_3NaO_3$ $[M+Na]^+$ 342.0849 found 342.0853.



(*E*)-3-Diazo-4-(2-methoxybenzylidene)-1-phenylpyrrolidine-2,5-dione (2q): The title compound was synthesized from imide **1q** (587 mg, 2.0 mmol) according to GP2, yield 536 mg (84%). Orange solid; m.p. 118.8–119.4 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.86 (s, 1H, =CH), 7.55 – 7.49 (m, 2H), 7.47 – 7.37 (m, 4H), 7.30 – 7.25 (m, 1H), 7.05 (td, *J* = 7.5, 1.0 Hz, 1H), 6.97 (dd, *J* = 8.4, 0.9 Hz, 1H), 3.91 (s, 3H, OCH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.3, 164.0, 157.4, 132.0, 131.3, 130.1, 129.1, 128.4, 126.5, 124.6, 122.2, 120.3, 116.8, 110.9, 61.0 (C=N_2), 55.4. HRMS (ESI), *m/z* calcd for $C_{18}H_{13}N_3NaO_3$ $[M+Na]^+$ 342.0849 found 342.0852.

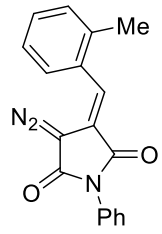


(*E*)-3-Diazo-4-(4-methylbenzylidene)-1-phenylpyrrolidine-2,5-dione (2r): The title compound was synthesized from imide **1r** (556 mg, 2.0 mmol) according to GP2, yield 485 mg (80%). Orange solid; m.p. 124.3–125.6 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.76 (s, 1H, =CH), 7.55 – 7.49 (m, 2H), 7.48 – 7.40 (m, 3H), 7.28 (s, 4H), 2.43 (s, 3H, CH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.4, 164.1, 140.1, 131.9, 130.6, 129.5, 129.2, 129.1, 128.52, 128.48, 126.5, 115.7, 60.2

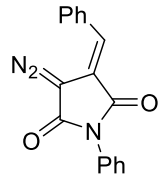


(C=N_2), 21.5. HRMS (ESI), *m/z* calcd for $C_{18}H_{13}N_3NaO_2$ $[M+Na]^+$ 326.0900 found 326.0886.

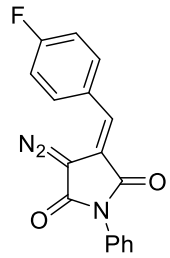
(*E*)-3-Diazo-4-(2-methylbenzylidene)-1-phenylpyrrolidine-2,5-dione (2s): The title compound was synthesized from imide **1s** (556 mg, 2.0 mmol) according to GP2, yield 528 mg (87%). Orange solid; m.p. 101.1–101.9 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.84 (s, 1H, =CH), 7.56 – 7.50 (m, 2H), 7.47 – 7.42 (m, 3H), 7.36 – 7.23 (m, 4H), 2.43 (s, 3H, CH_3). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.1, 163.8, 137.8, 132.4, 131.8, 130.7, 129.7, 129.2, 128.5, 128.3, 127.1, 126.5, 125.4, 117.5, 60.2 (C=N_2), 20.0. HRMS (ESI), *m/z* calcd for $C_{18}H_{13}N_3NaO_2$ $[M+Na]^+$ 326.0900 found 326.0892.



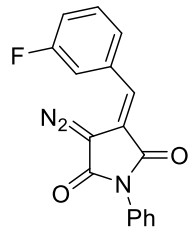
(*E*)-3-Benzylidene-4-diazo-1-phenylpyrrolidine-2,5-dione (2t): The title compound was synthesized from imide **1t** (527 mg, 2.0 mmol) according to GP2, yield 486 mg (84%). Orange solid; m.p. 125.8–126.4 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.79 (s, 1H, =CH), 7.55 – 7.37 (m, 10H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.2, 164.0, 133.4, 131.9, 129.6, 129.2, 129.0, 128.8, 128.5, 128.2, 126.5, 116.7, 60.2 (C=N_2), 20.0. HRMS (ESI), *m/z* calcd for $C_{17}H_{11}N_3NaO_2$ $[M+Na]^+$ 312.0743 found 312.0732.



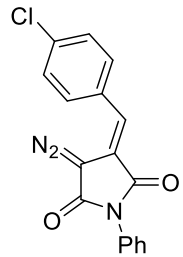
(*E*)-3-Diazo-4-(4-fluorobenzylidene)-1-phenylpyrrolidine-2,5-dione (2u): The title compound was synthesized from imide **1u** (563 mg, 2.0 mmol) according to GP2, yield 498 mg (81%). Orange solid; m.p. 133.2–133.8 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 (s, 1H, =CH), 7.55 – 7.49 (m, 2H), 7.47 – 7.41 (m, 3H), 7.37 (dd, *J* = 8.7, 5.3 Hz, 2H), 7.18 (t, *J* = 8.6 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 166.0, 163.9, 163.2 (d, *J* = 251.7 Hz), 131.8, 130.9 (d, *J* = 8.5 Hz), 129.6 (d, *J* = 3.7 Hz), 129.2, 128.6, 127.0, 126.5, 116.6, 116.1 (d, *J* = 22.0 Hz), 60.0 (C=N_2). HRMS (ESI), *m/z* calcd for $C_{17}H_{10}FN_3NaO_2$ $[M+Na]^+$ 330.0649 found 330.0644.



(*E*)-3-Diazo-4-(3-fluorobenzylidene)-1-phenylpyrrolidine-2,5-dione (2v): The title compound was synthesized from imide **1v** (563 mg, 2.0 mmol) according to GP2, yield 467 mg (76%). Orange solid; m.p. 136.8–137.3 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (s, 1H, =CH), 7.57 – 7.48 (m, 2H), 7.51 – 7.39 (m, 4H), 7.21 – 7.03 (m, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.8, 163.8, 162.7 (d, *J* = 248.5 Hz), 135.5 (d, *J* = 7.8 Hz), 131.7, 130.4 (d, *J* = 8.5 Hz), 129.2, 128.6, 126.5, 126.4, 124.6 (d, *J* = 3.1 Hz), 117.9, 116.6 (d, *J* = 21.4 Hz), 115.7 (d, *J* = 22.2 Hz), 60.3 (C=N_2). HRMS (ESI), *m/z* calcd for $C_{17}H_{10}FN_3NaO_2$ $[M+Na]^+$ 330.0649 found 330.0637.



(*E*)-3-(4-Chlorobenzylidene)-4-diazo-1-phenylpyrrolidine-2,5-dione (2w): The title compound was synthesized from imide **1w** (595 mg, 2.0 mmol) according to GP2, yield 498 mg (77%). Orange solid; m.p. 133.8–134.7 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.70 (s, 1H, =CH), 7.54 – 7.49 (m, 2H), 7.48 – 7.41 (m, 5H), 7.31 (d, *J* = 8.1 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.9, 163.8, 135.6, 131.8, 131.7, 130.2, 129.2, 129.1, 128.6, 126.6, 126.4, 117.2, 60.2 (C=N_2). HRMS (ESI), *m/z* calcd for $C_{17}H_{10}ClN_3NaO_2$ $[M+Na]^+$ 346.0354 found 346.0355.



(*E*)-3-Diazo-1-phenyl-4-(4-(trifluoromethyl)benzylidene)pyrrolidine-2,5-dione (2x): The title compound was synthesized from imide **1x** (663 mg, 2.0 mmol) according to GP2, yield 536 mg (75%). Orange solid; m.p. 145.4–145.7 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.75 (s, 1H, =CH), 7.74 (d, *J* = 8.3 Hz, 2H), 7.56 – 7.41 (m, 7H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.6, 163.7, 136.8, 131.6, 131.2 (q, *J* = 32.5 Hz), 129.2, 129.1, 128.7, 126.4, 125.8 (q, *J* = 3.9 Hz), 125.7, 123.7 (q, *J* = 272.3 Hz), 118.7, 60.3 (C=N_2). HRMS (ESI), *m/z* calcd for $C_{18}H_{10}F_3N_3NaO_2$ $[M+Na]^+$ 380.0617 found 380.0628.

(*E*)-3-Diazo-4-(4-nitrobenzylidene)-1-phenylpyrrolidine-2,5-dione (2y): The title compound was synthesized from imide **1y** (617 mg, 2.0 mmol) according to GP2, yield 221 mg (33%). Orange solid; m.p. 160.0–160.9 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.34 (d, *J* = 8.8 Hz, 2H), 7.75 (s, 1H, =CH), 7.58 – 7.49 (m, 4H), 7.49 – 7.41 (m, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.3, 163.5, 147.8, 139.7, 131.5, 129.6, 129.3, 128.8, 126.4, 124.5, 124.0, 119.7, 60.5 (C=N_2). HRMS (ESI), *m/z* calcd for $C_{17}H_{10}N_4NaO_4$ $[M+Na]^+$ 357.0594 found 357.0606.

(*E*)-3-Diazo-4-(3-nitrobenzylidene)-1-phenylpyrrolidine-2,5-dione (2z): The title compound was synthesized from imide **1z** (617 mg, 2.0 mmol) according to GP2, yield 201 mg (30%). Orange solid; m.p. 142.5–143.1 °C (decomp). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.27 (dt, *J* = 7.1, 2.3 Hz, 1H), 8.22 (s, 1H), 7.75 (s, 1H, =CH), 7.72 – 7.65 (m, 2H), 7.56 – 7.50 (m, 2H), 7.48 – 7.42 (m, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.4, 163.5, 148.4, 135.1, 134.5, 131.6, 130.0, 129.3, 128.7, 126.4, 124.4, 124.0, 123.2, 119.3, 60.3 (C=N_2). HRMS (ESI), *m/z* calcd for $C_{17}H_{10}N_4NaO_4$ $[M+Na]^+$ 357.0594 found 357.0596.

Crystallographic data for compounds 2f and 2j

X-ray Single Crystal analyses were performed on Agilent Technologies «Xcalibur» and «Supernova» diffractometers with monochromated MoKα or CuKα radiation, respectively. Crystals were measuring at the temperature of 100 K The structures has been solved by the Superflip [S9] and ShelXS [S10] structure solution programs using Charge Flipping and Direct Methods, respectively, and refined with the ShelXL [S11] refinement incorporated in the OLEX2 program package [S12]. CCDC # 2014276 (**2f**), CCDC # 1999007 (**2j**) contain the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via http://www.ccdc.cam.ac.uk.

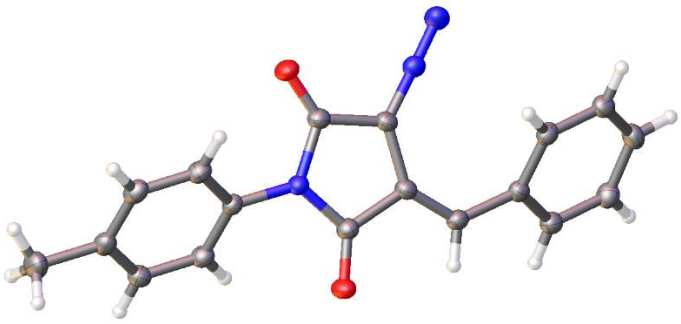


Figure S1 ORTEP representation of compound **2f**

Table S1 Crystal data and structure refinement for **2f**	
Empirical formula	$C_{18}H_{13}N_3O_2$
Formula weight	303.32
Temperature/K	100.00(10)
Crystal system	monoclinic
Space group	$P2_1$
a/Å	9.34450(10)
b/Å	13.38360(10)
c/Å	23.8971(2)
α/°	90
β/°	99.4670(10)
γ/°	90
Volume/Å^3	2947.94(5)
Z	2
ρ_{calc}g/cm^3	1.367
μ/mm^{-1}	0.747
F(000)	1264.0
Crystal size/mm^3	0.15 × 0.12 × 0
Radiation	CuKα (λ = 1.54184)
2Θ range for data collection/°	7.5 to 154.934
Index ranges	-11 ≤ h ≤ 11, -16 ≤ k ≤ 16, -30 ≤ l ≤ 28
Reflections collected	60289
Independent reflections	12289 [R_{int} = 0.0347, R_{sigma} = 0.0269]
Data/restraints/parameters	12289/1/833
Goodness-of-fit on F^2	1.013
Final R indexes [I>=2σ (I)]	R_1 = 0.0446, wR_2 = 0.1249
Final R indexes [all data]	R_1 = 0.0454, wR_2 = 0.1259
Largest diff. peak/hole / e Å^{-3}	0.48/-0.22
CCDC # 2014276	

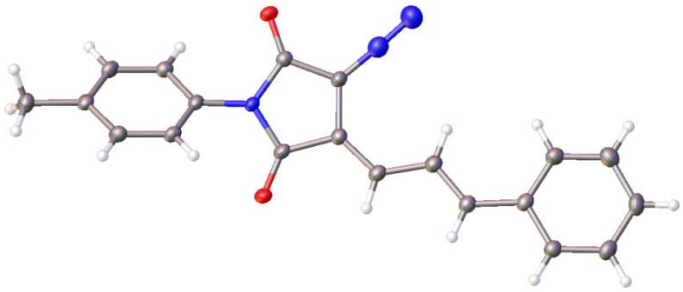


Figure S2 ORTEP representation of compound **2j**

Table S2 Crystal data and structure refinement for **2j**	
Empirical formula	$C_{20}H_{15}N_3O_2$
Formula weight	329.35
Temperature/K	100.00(11)
Crystal system	triclinic
Space group	P-1
a/Å	6.21290(10)
b/Å	9.2395(2)
c/Å	14.5466(3)
α/°	79.089(2)
β/°	85.627(2)
γ/°	76.324(2)
Volume/Å^3	796.26(3)
Z	2
ρ_{calc}g/cm^3	1.374
μ/mm^{-1}	0.737
F(000)	344.0
Crystal size/mm^3	0.16 × 0.1 × 0.06
Radiation	CuKα (λ = 1.54184)
2Θ range for data collection/°	6.192 to 140.918
Index ranges	-7 ≤ h ≤ 7, -11 ≤ k ≤ 7, -17 ≤ l ≤ 17
Reflections collected	8190
Independent reflections	3031 [R_{int} = 0.0298, R_{sigma} = 0.0309]
Data/restraints/parameters	3031/0/227
Goodness-of-fit on F^2	1.080
Final R indexes [I>=2σ (I)]	R_1 = 0.0525, wR_2 = 0.1544
Final R indexes [all data]	R_1 = 0.0563, wR_2 = 0.1583
Largest diff. peak/hole / e Å^{-3}	0.21/-0.57
CCDC # 1999007	

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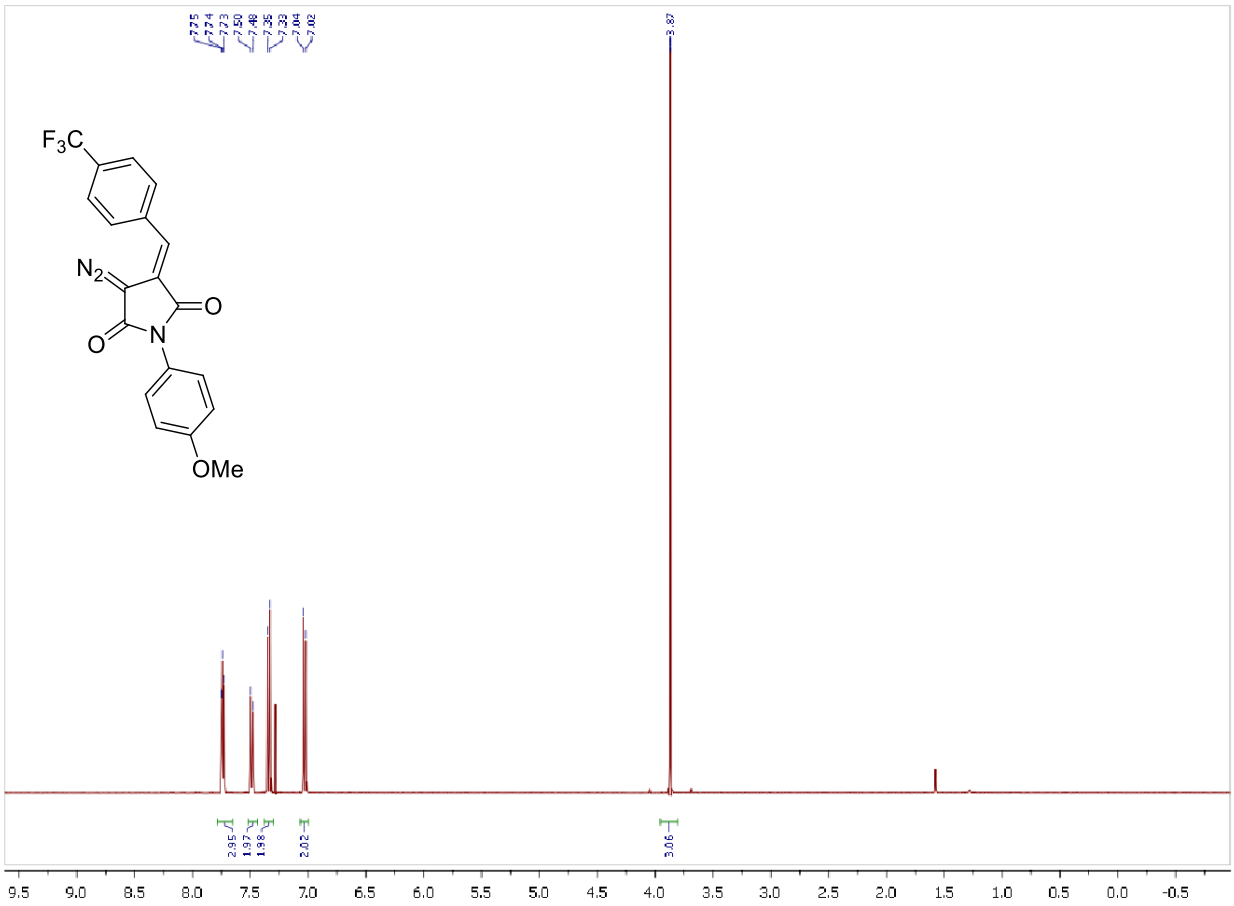

Figure S3 ^{1}H NMR spectrum of compound **2a**

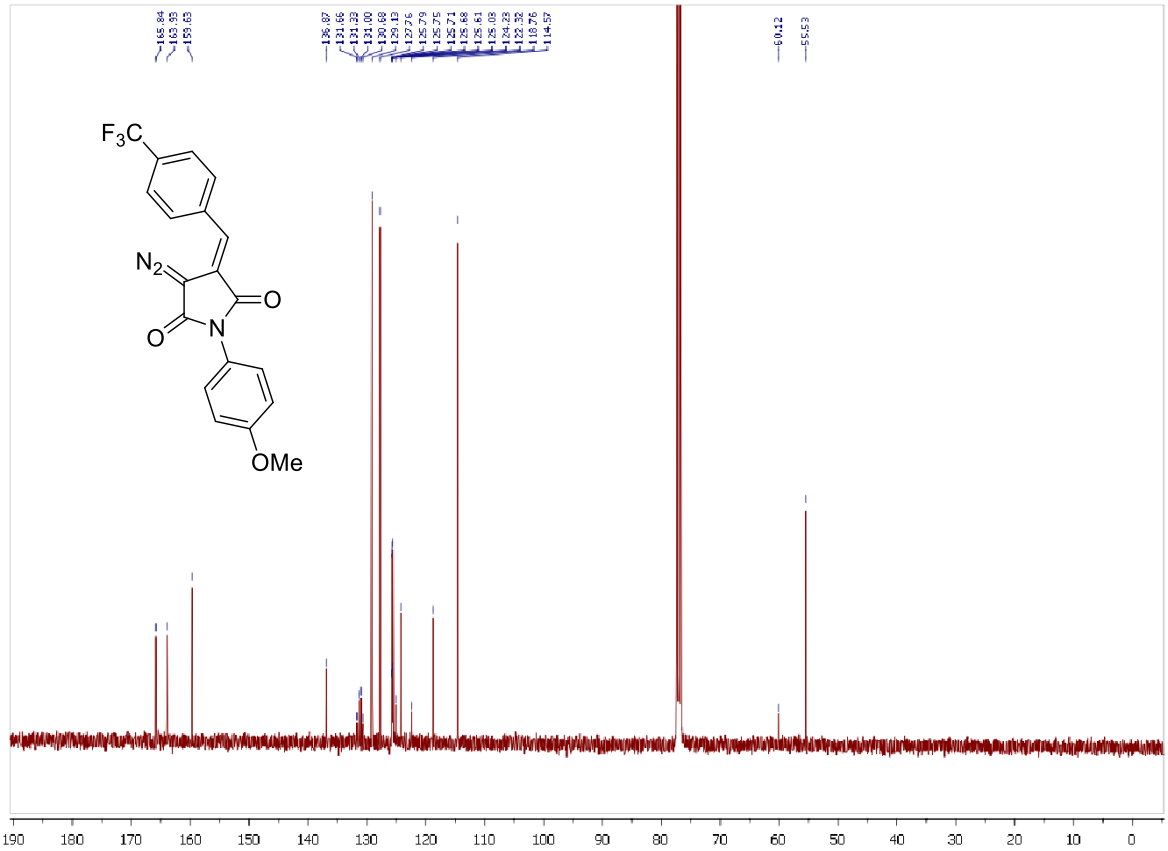

Figure S4 ^{13}C NMR spectrum of compound **2a**

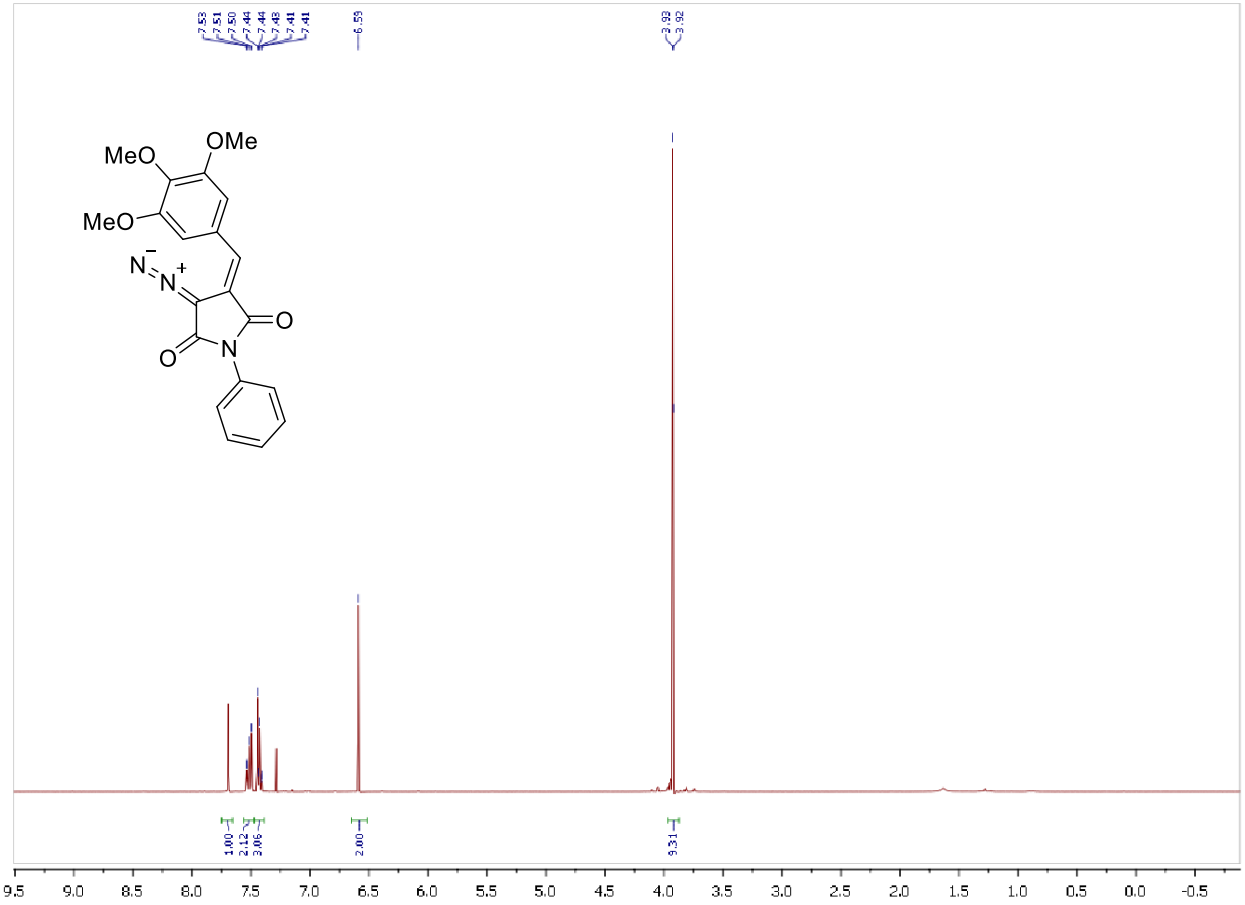

Figure S5 ^{1}H NMR spectrum of compound **2b**

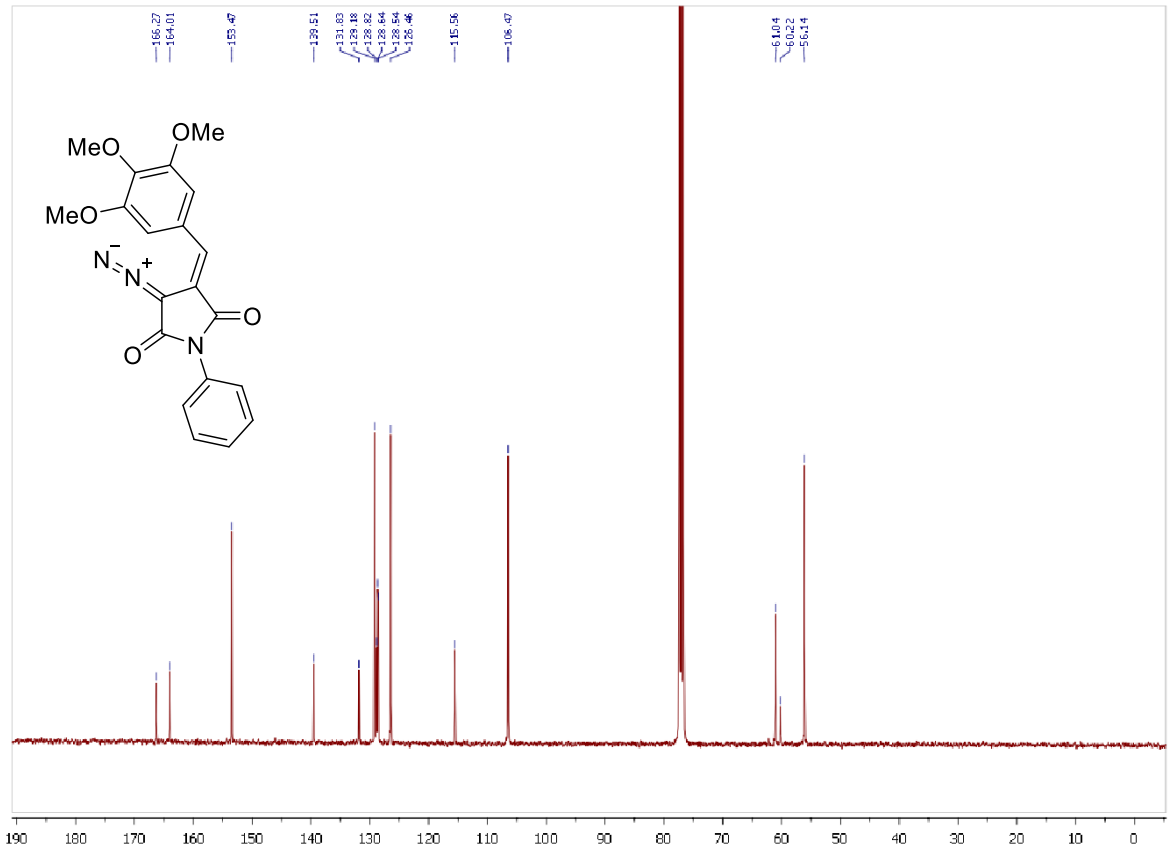

Figure S6 ^{13}C NMR spectrum of compound **2b**

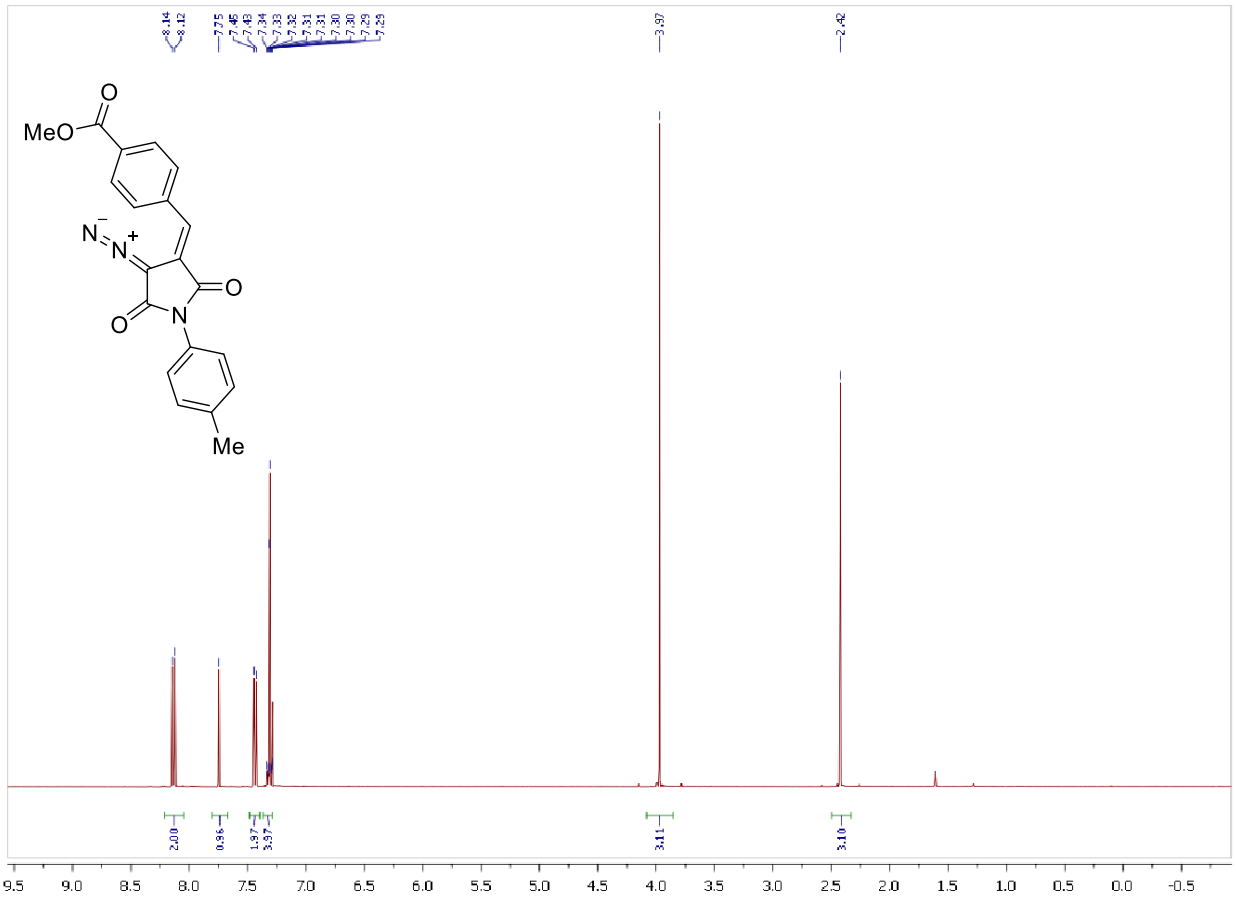

Figure S7 ^{1}H NMR spectrum of compound **2c**

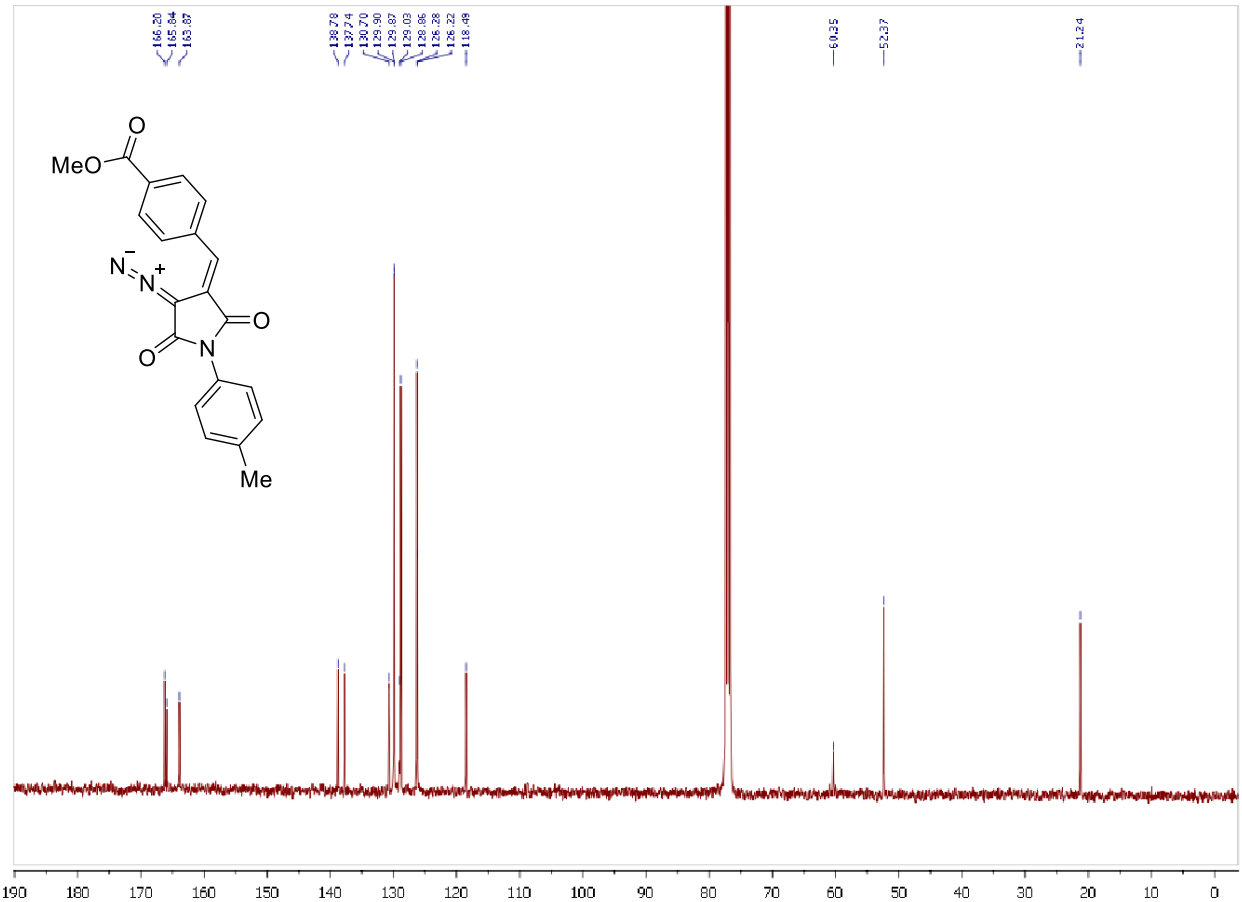

Figure S8 ^{13}C NMR spectrum of compound **2c**

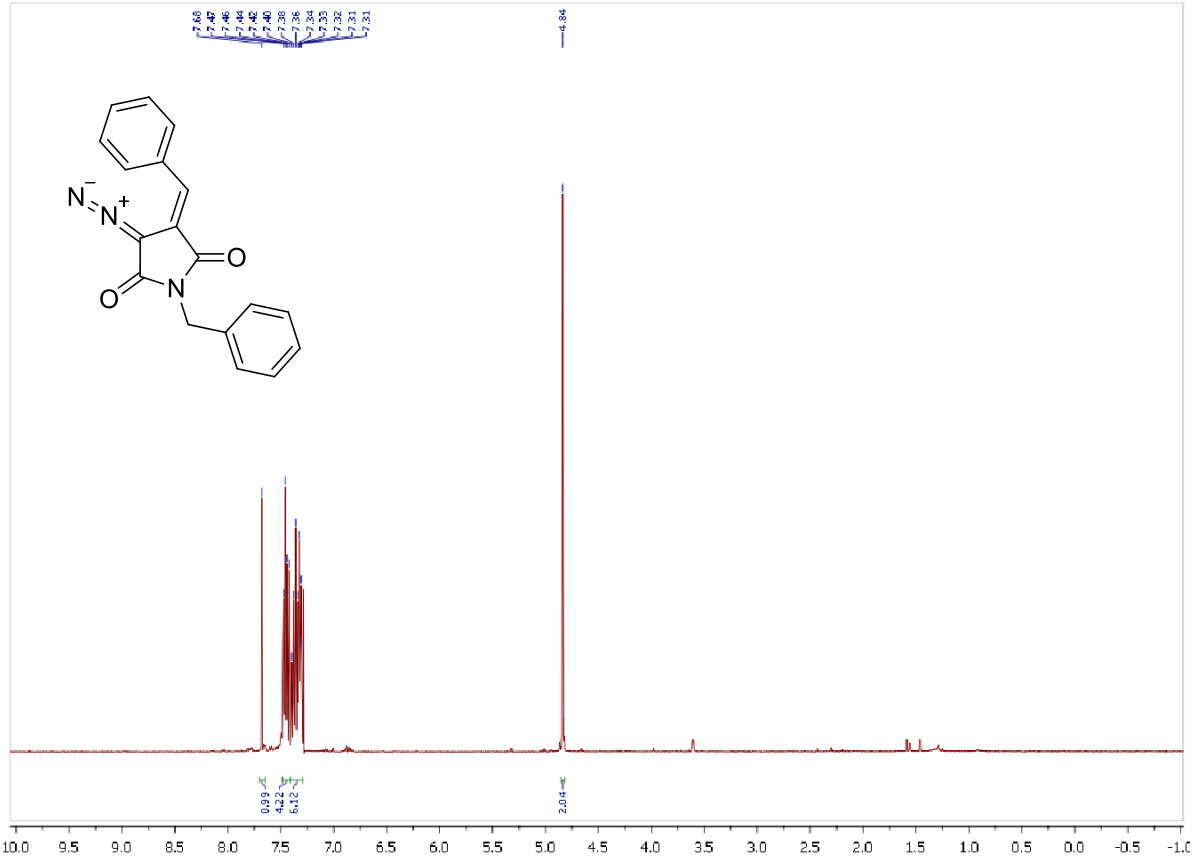

Figure S9 ^{1}H NMR spectrum of compound **2d**

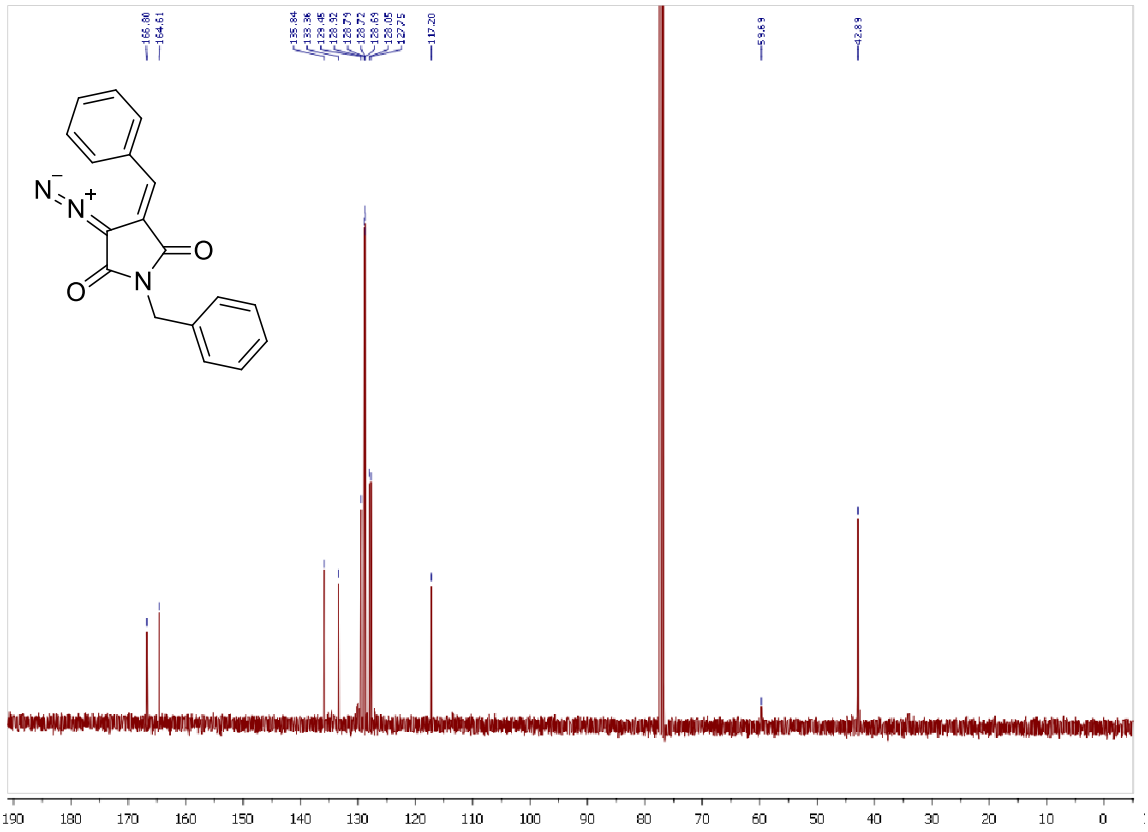

Figure S10 ^{13}C NMR spectrum of compound **2d**

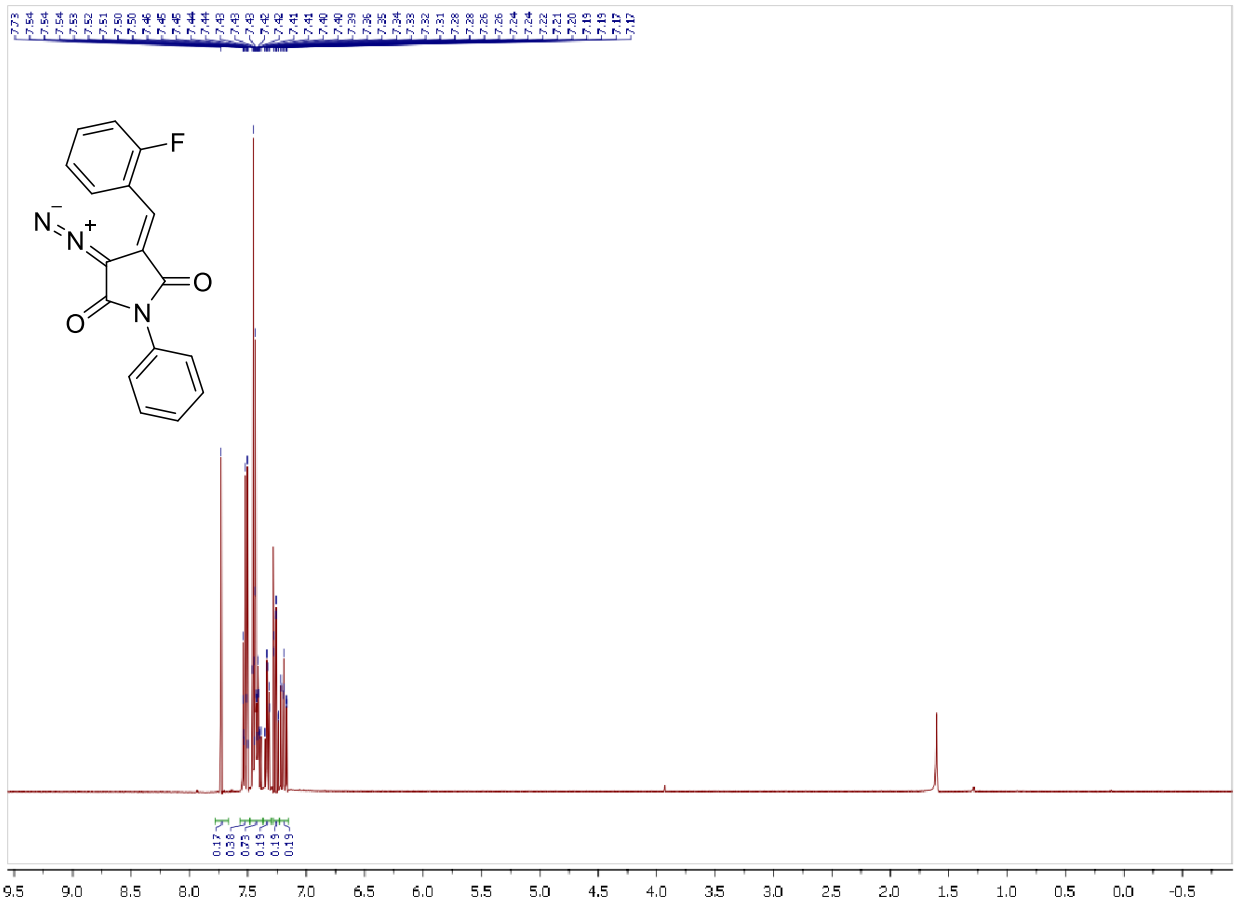

Figure S11 ^{1}H NMR spectrum of compound **2e**

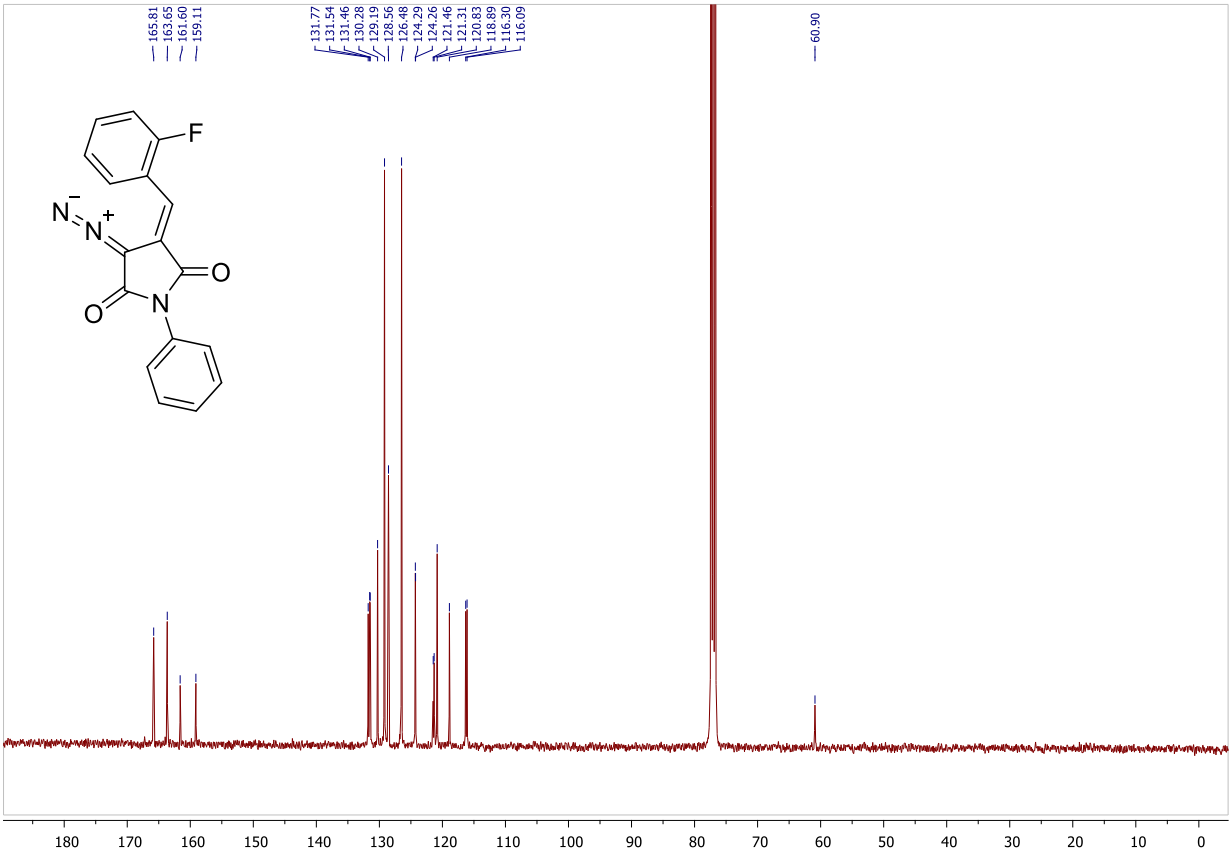

Figure S12 ^{13}C NMR spectrum of compound **2e**

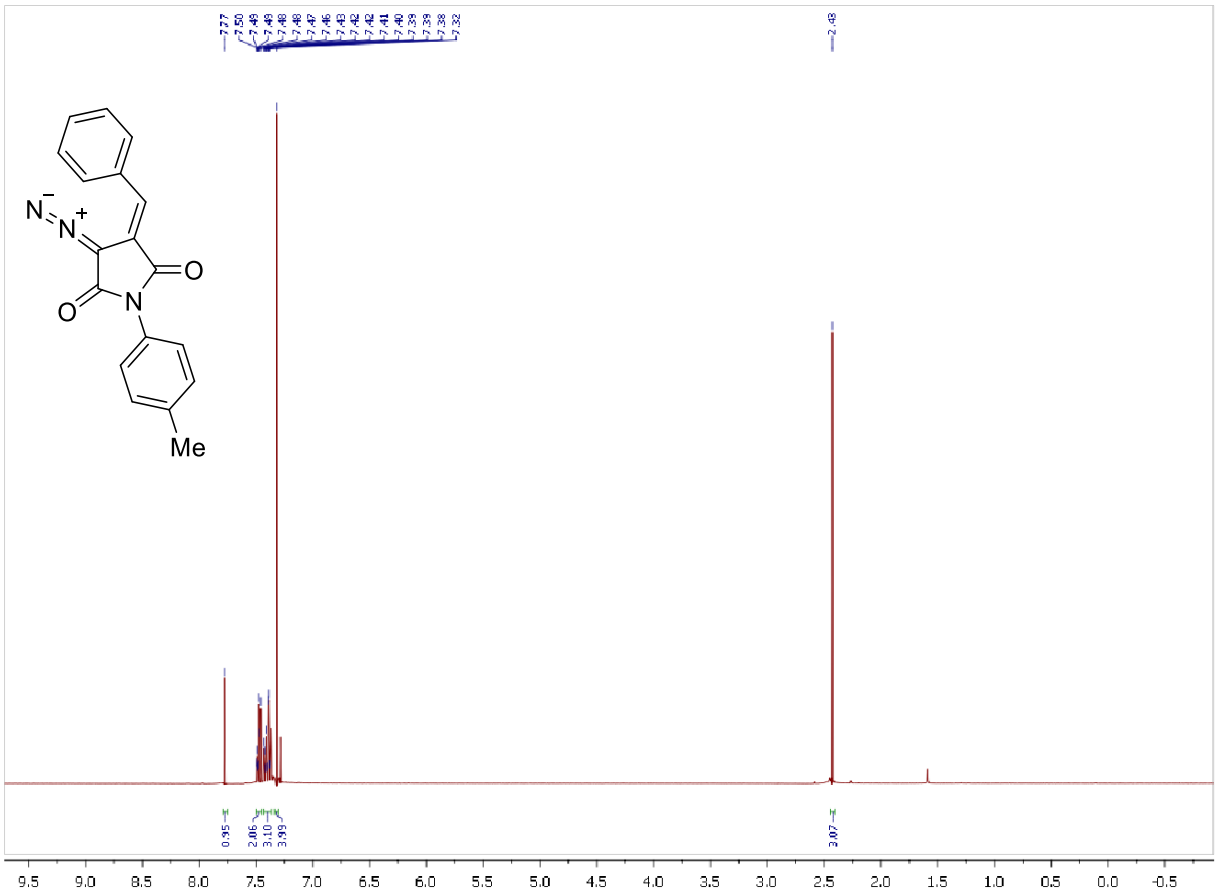

Figure S13 ^{1}H NMR spectrum of compound **2f**

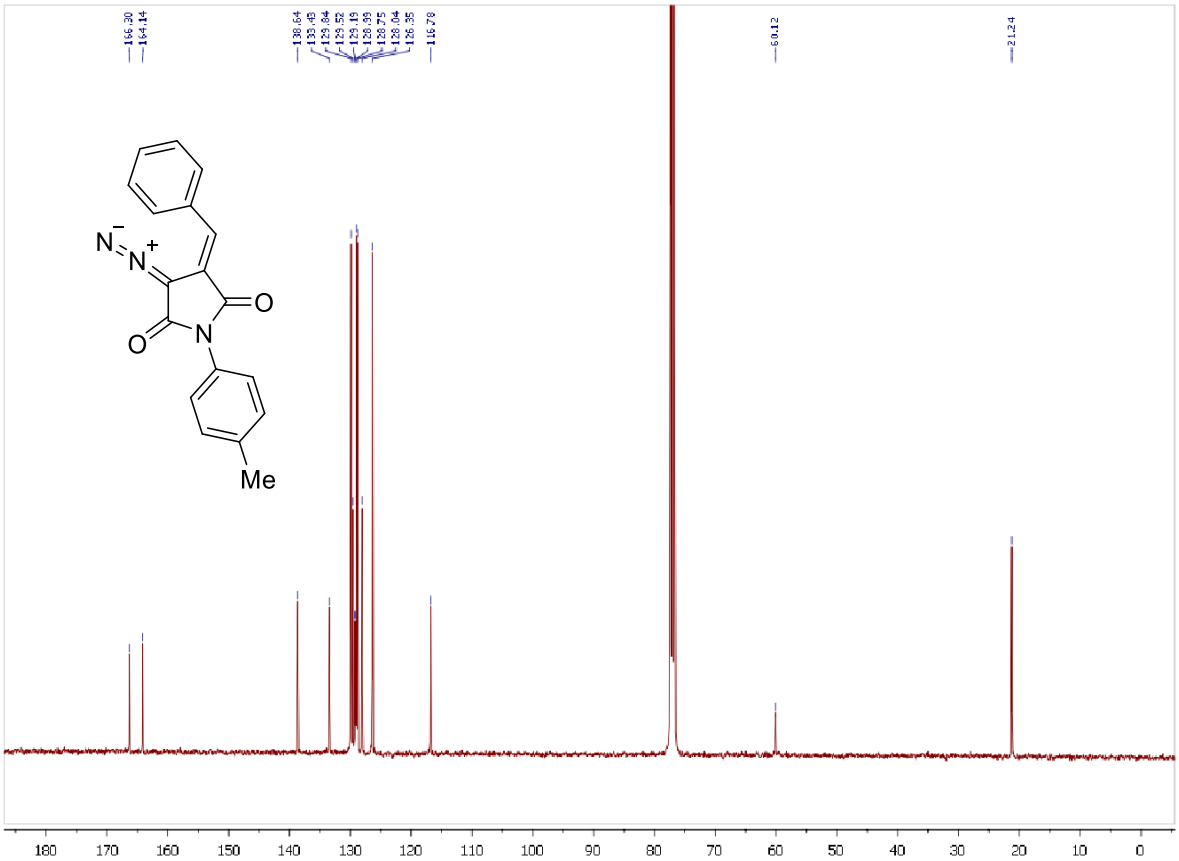

Figure S14 ^{13}C NMR spectrum of compound **2f**

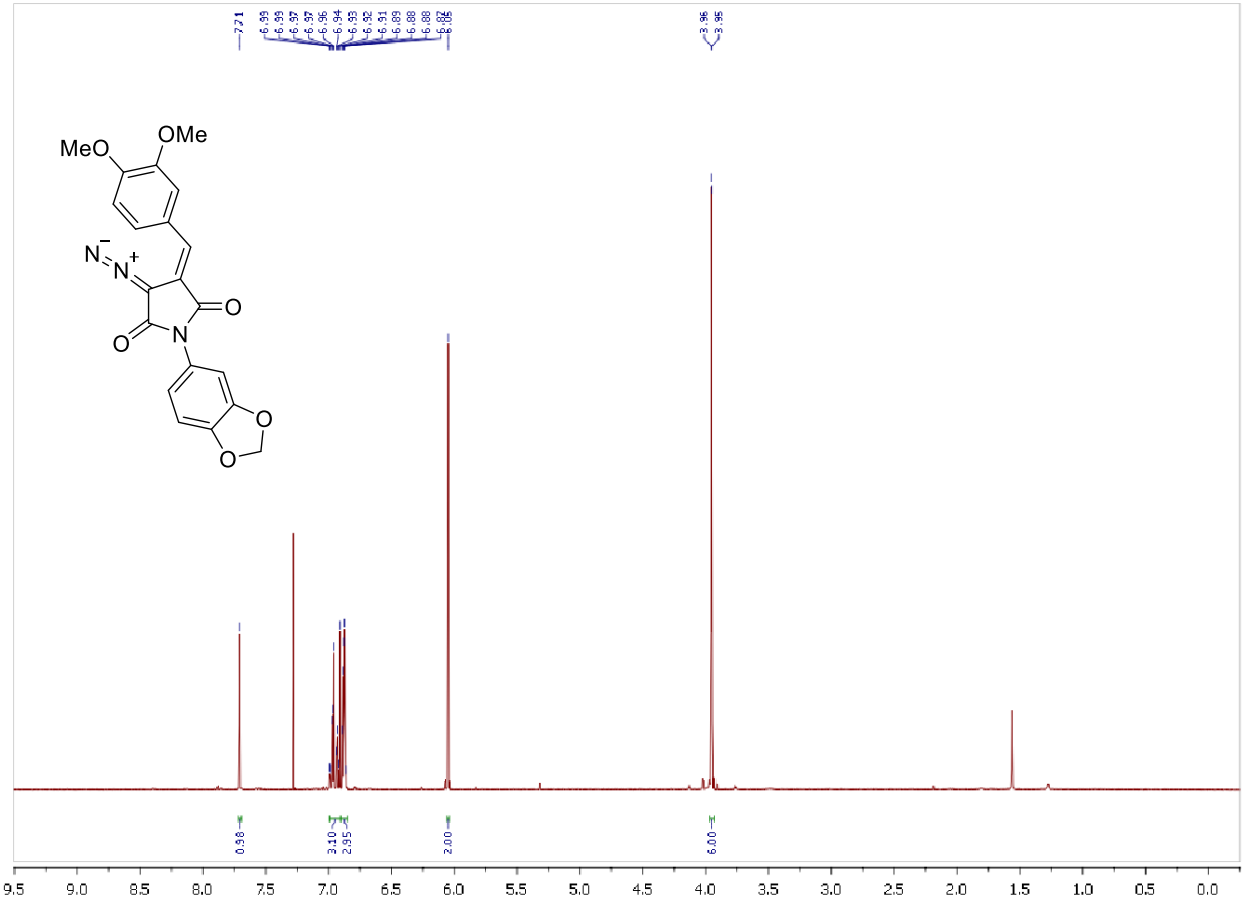

Figure S15 ^{1}H NMR spectrum of compound **2g**

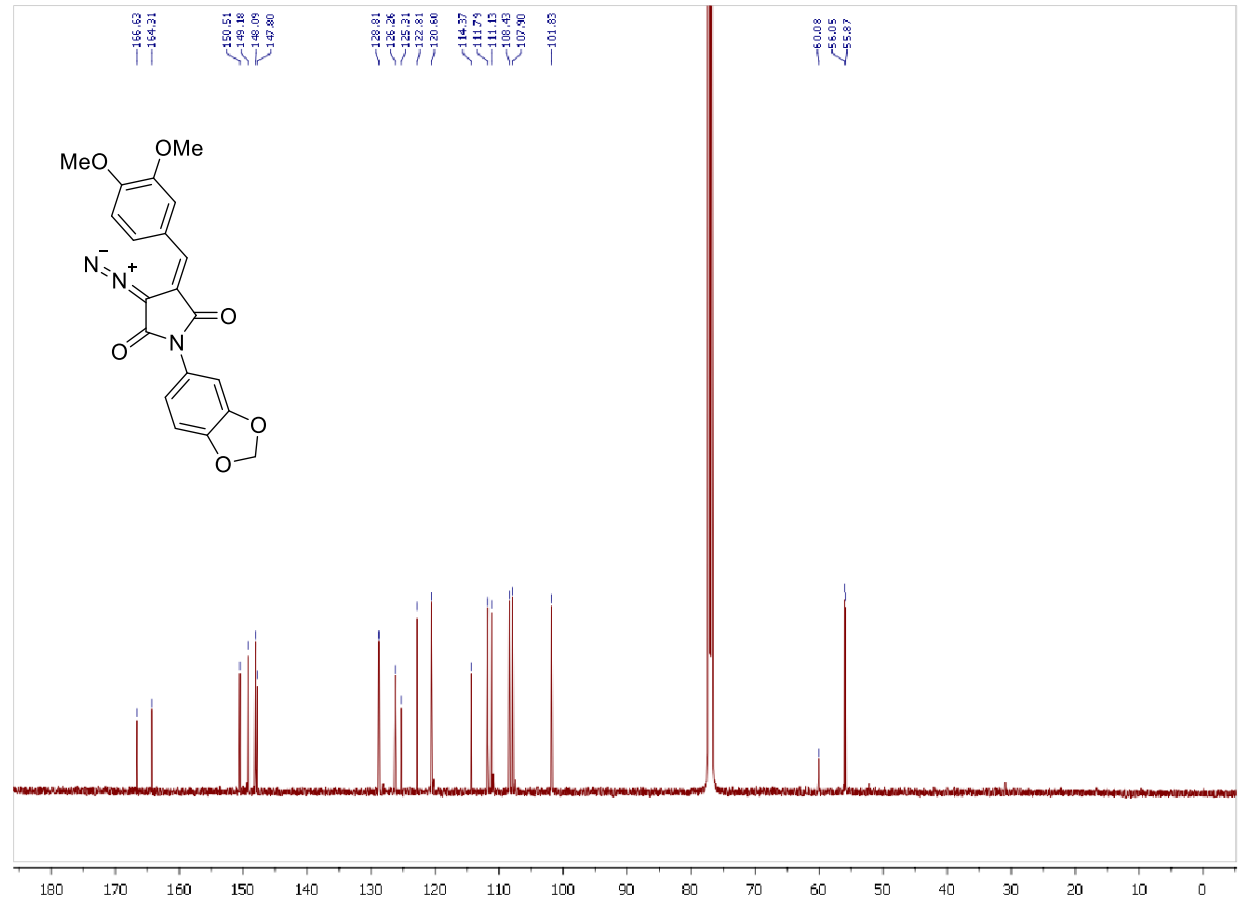

Figure S16 ^{13}C NMR spectrum of compound **2g**

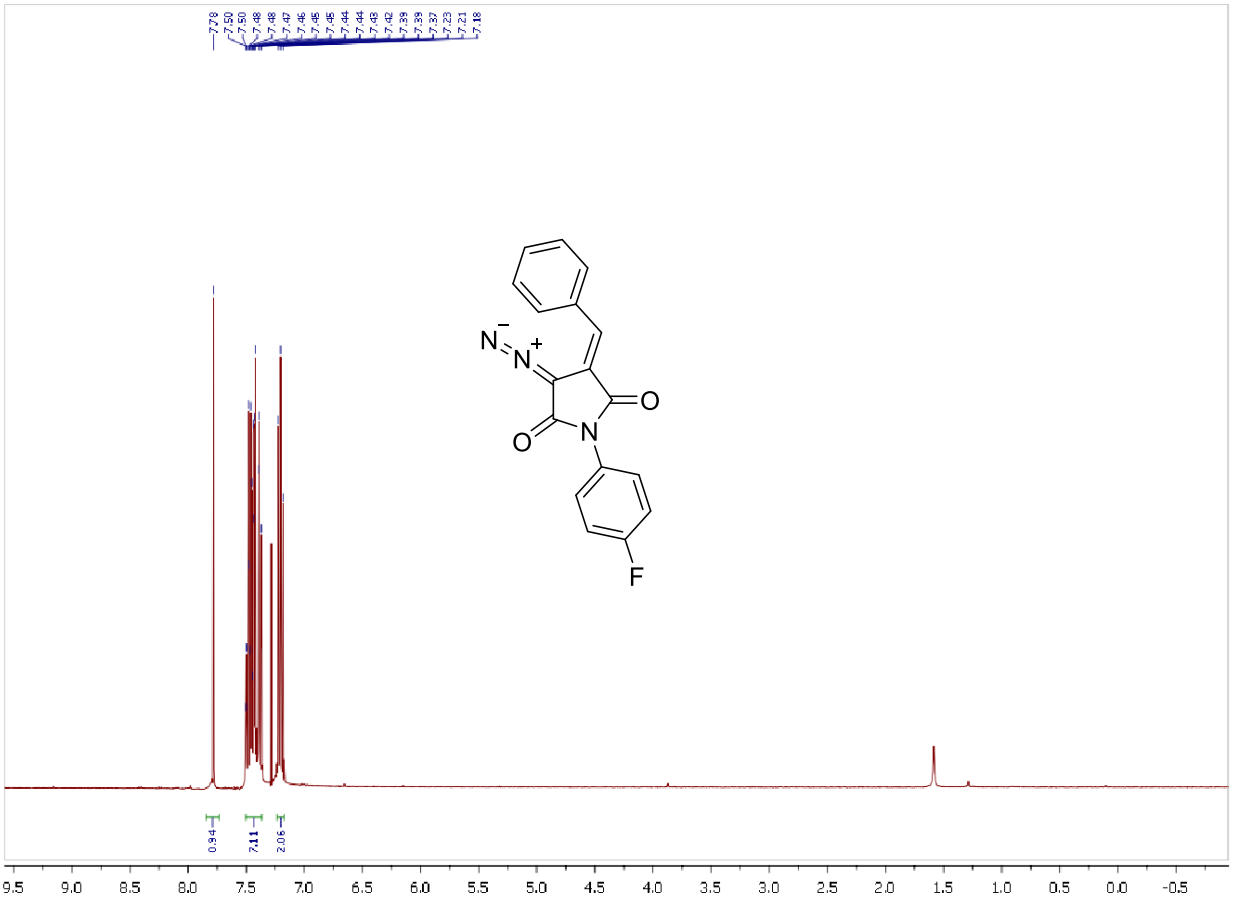

Figure S17 ^{1}H NMR spectrum of compound **2h**

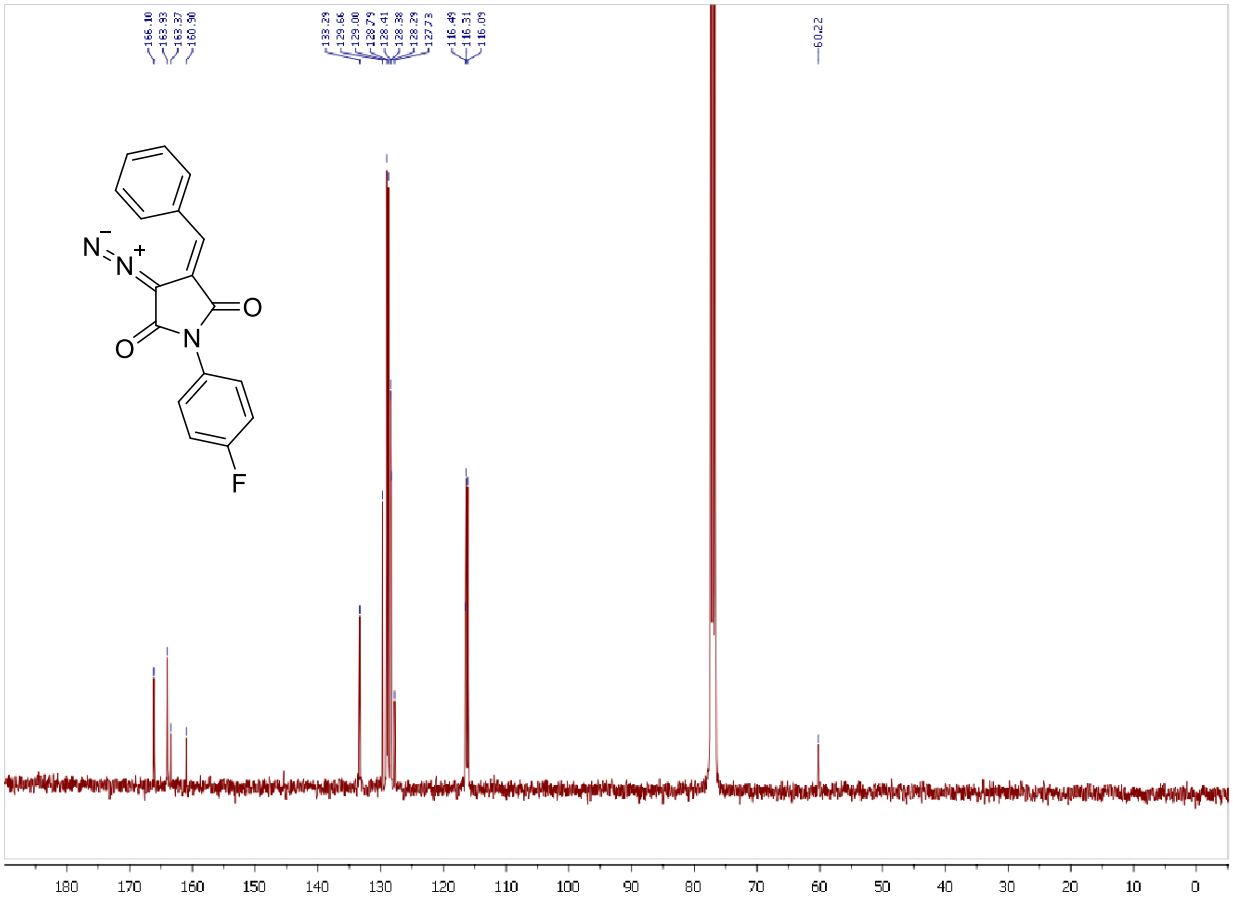

Figure S18 ^{13}C NMR spectrum of compound **2h**

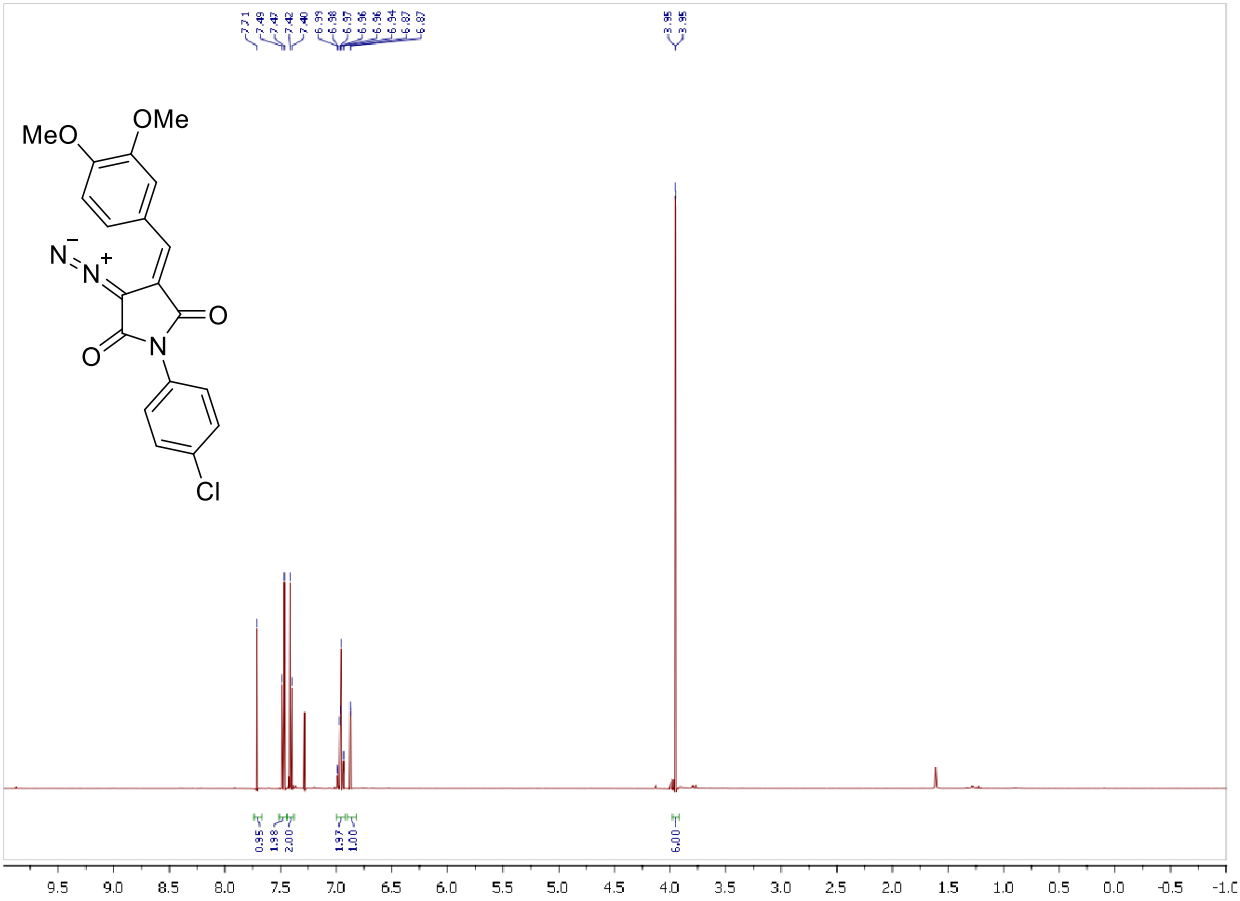

Figure S19 ^{1}H NMR spectrum of compound **2i**

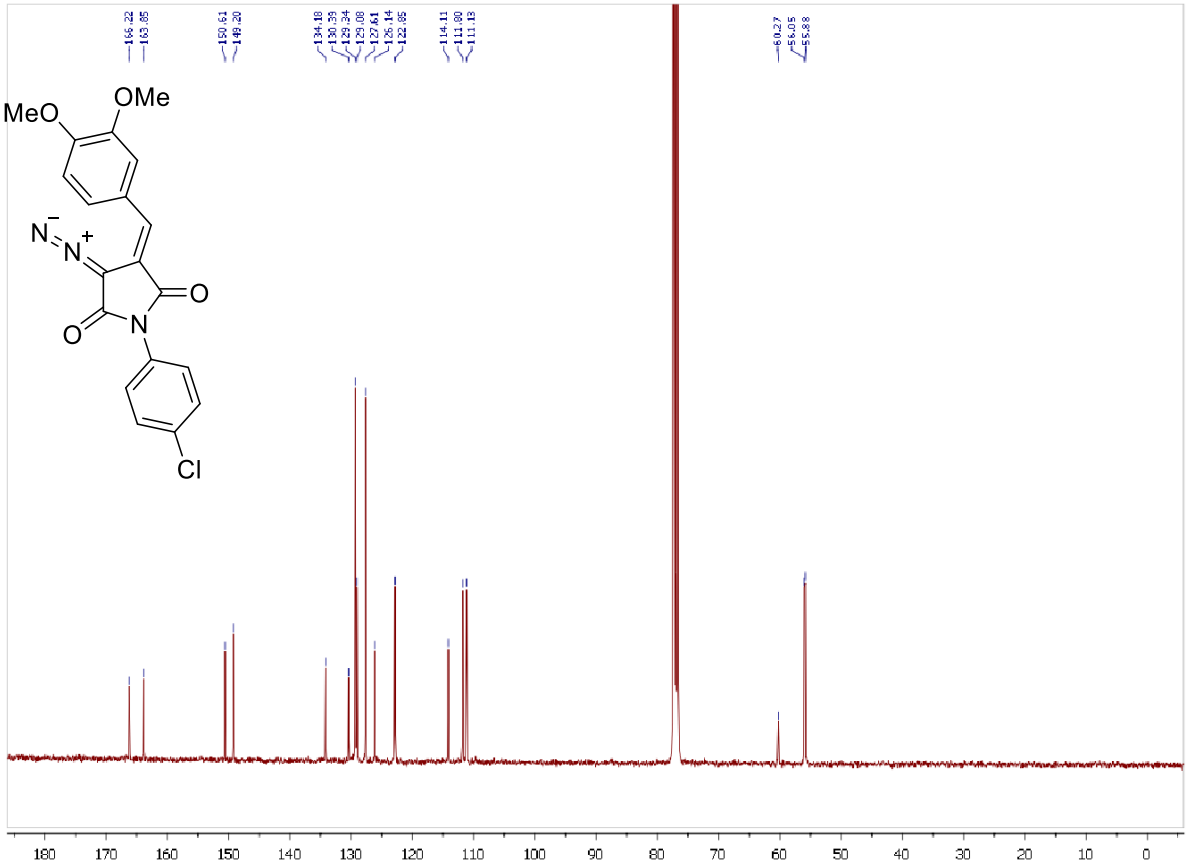

Figure S20 ^{13}C NMR spectrum of compound **2i**

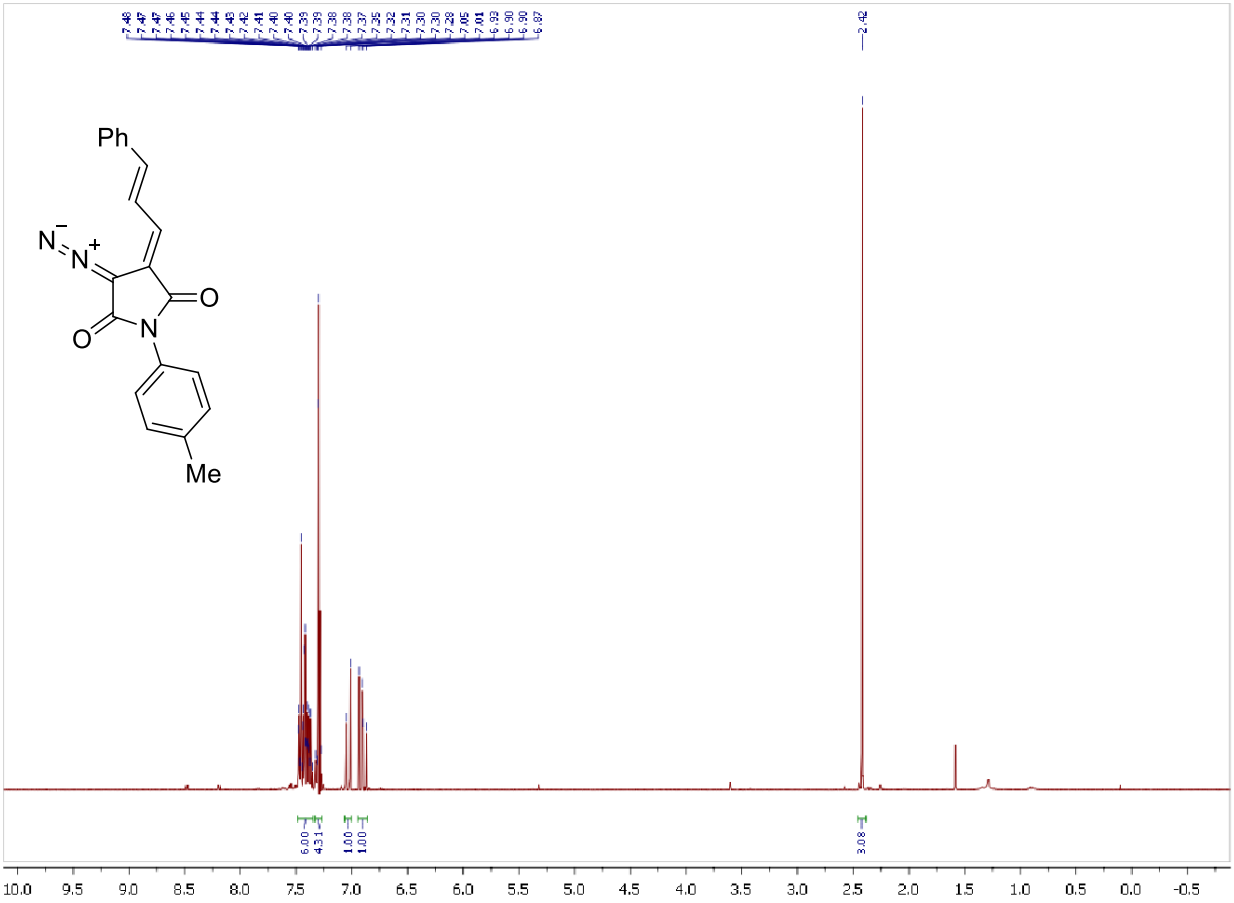

Figure S21 ^{1}H NMR spectrum of compound **2j**

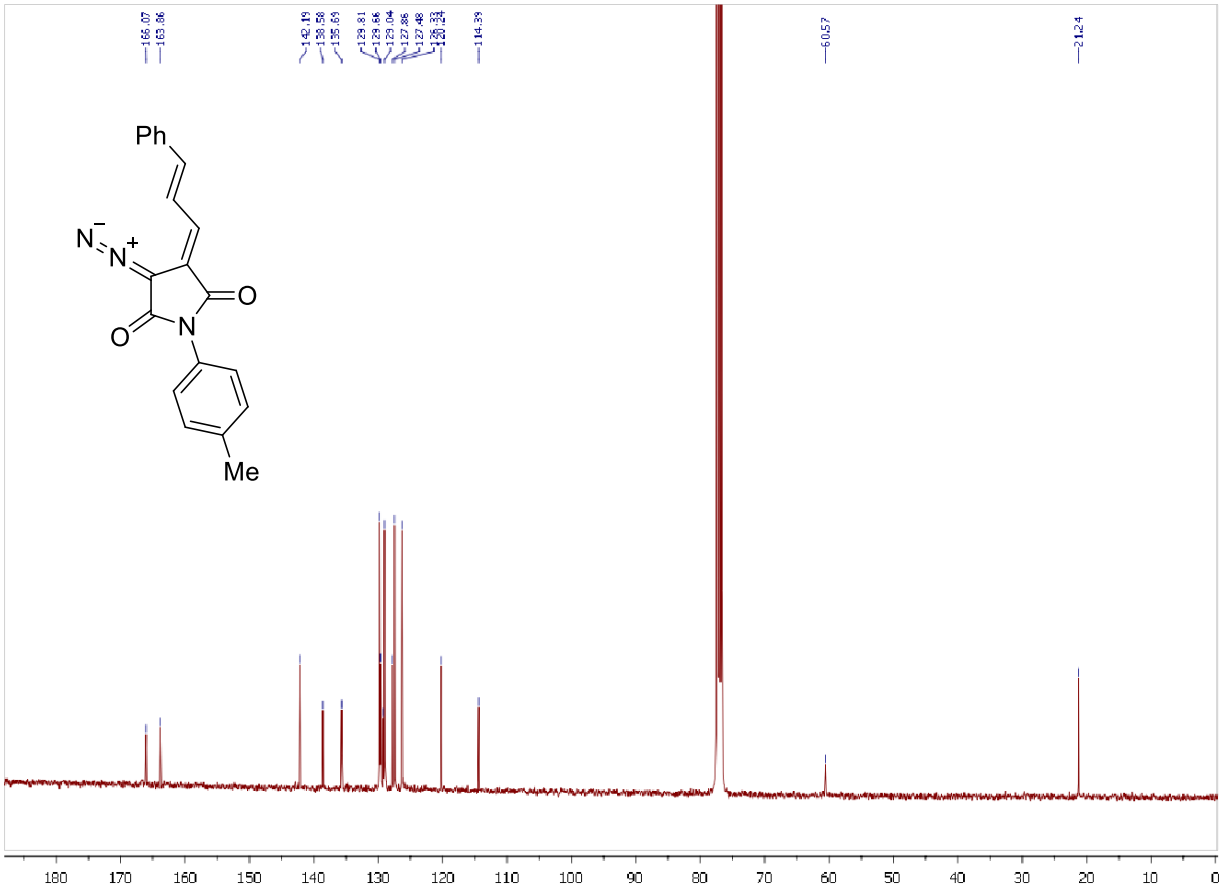

Figure S22 ^{13}C NMR spectrum of compound **2j**

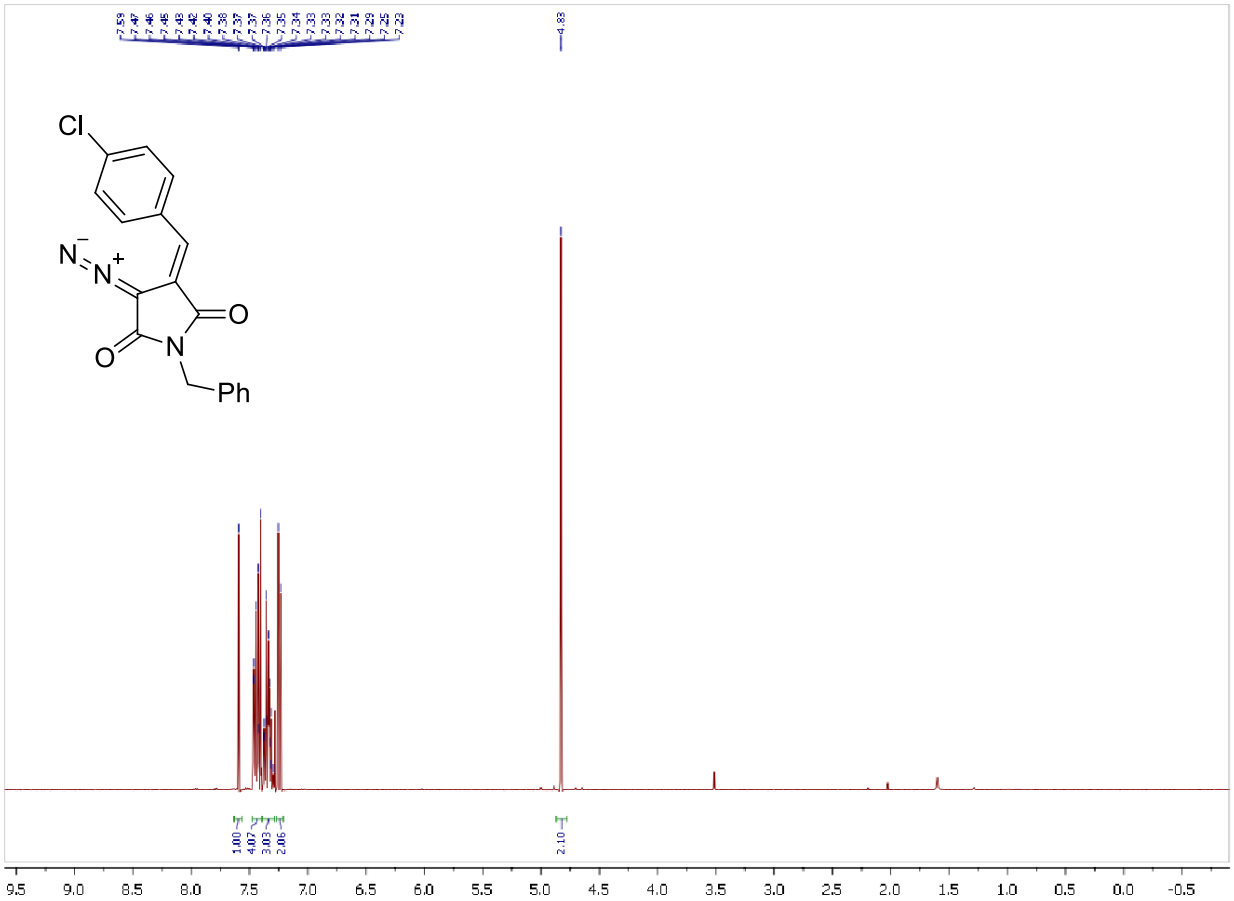

Figure S23 ^{1}H NMR spectrum of compound **2k**

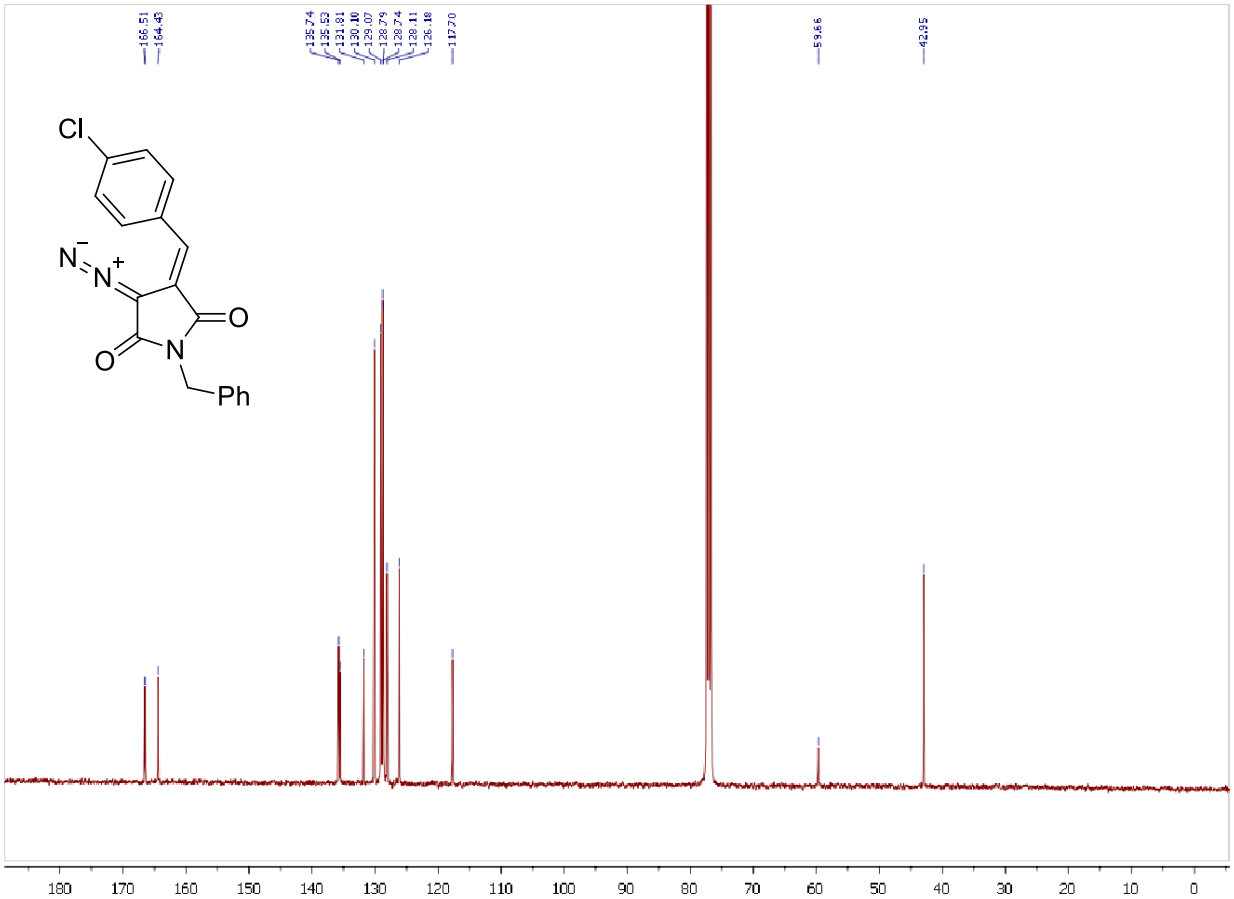

Figure S24 ^{13}C NMR spectrum of compound **2k**

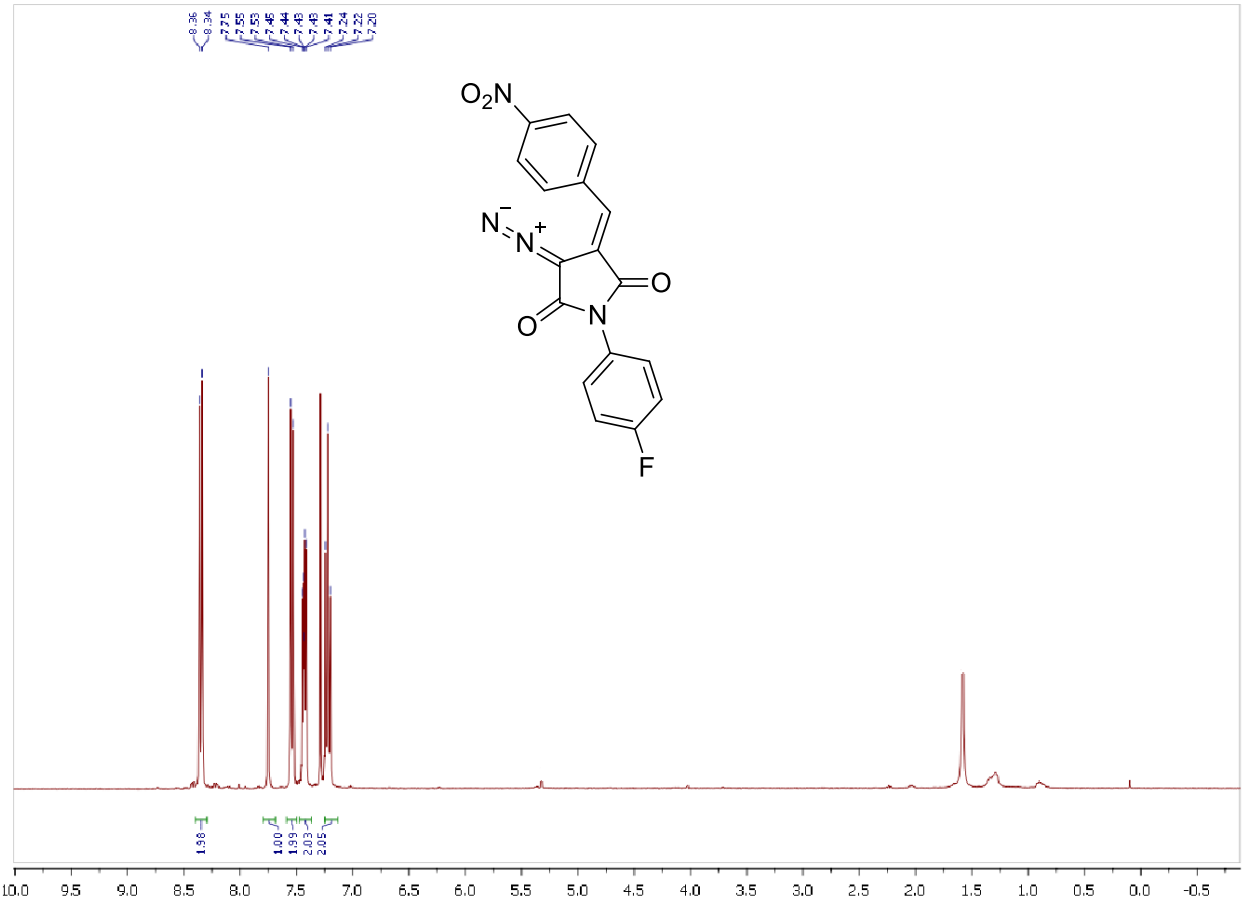

Figure S25 ^{1}H NMR spectrum of compound **2l**

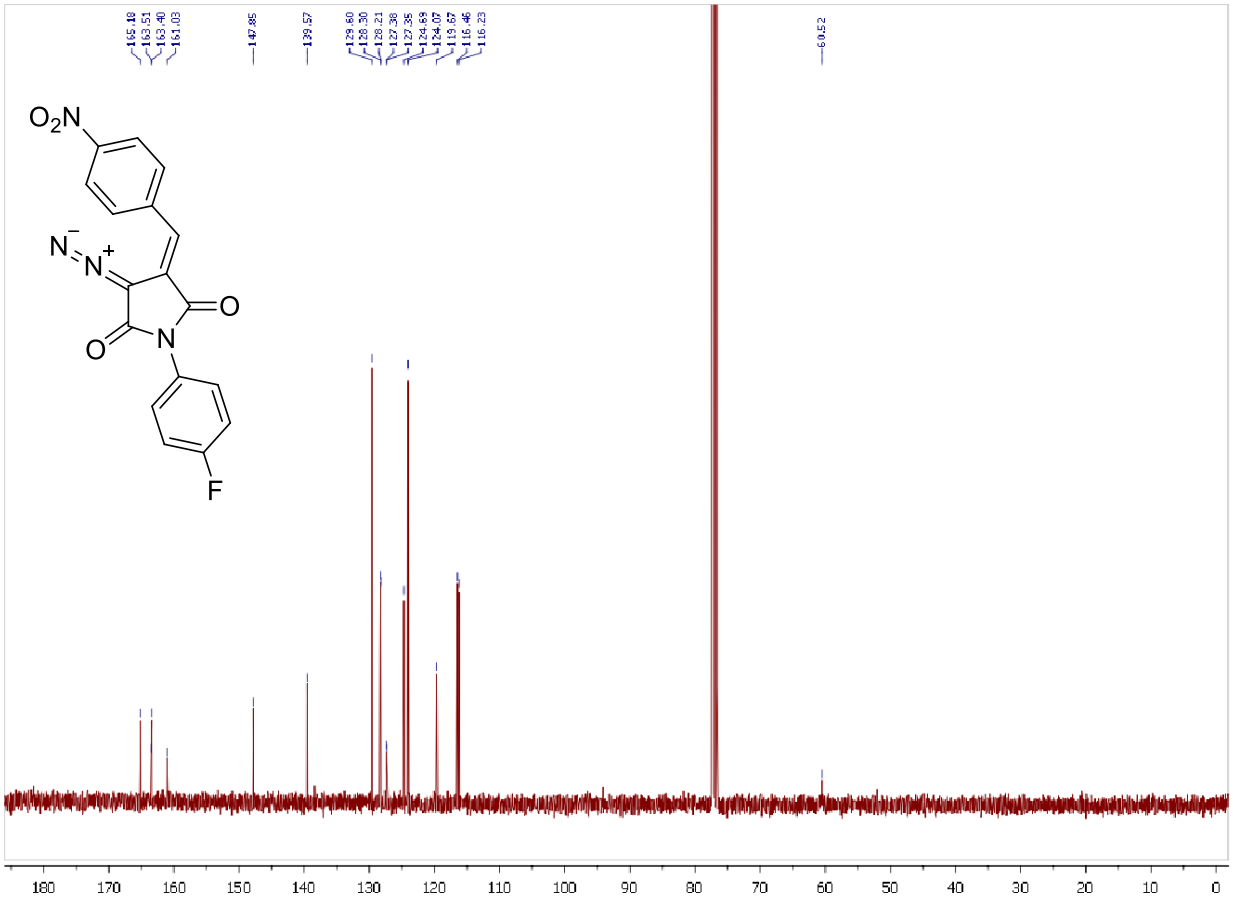

Figure S26 ^{13}C NMR spectrum of compound **2l**

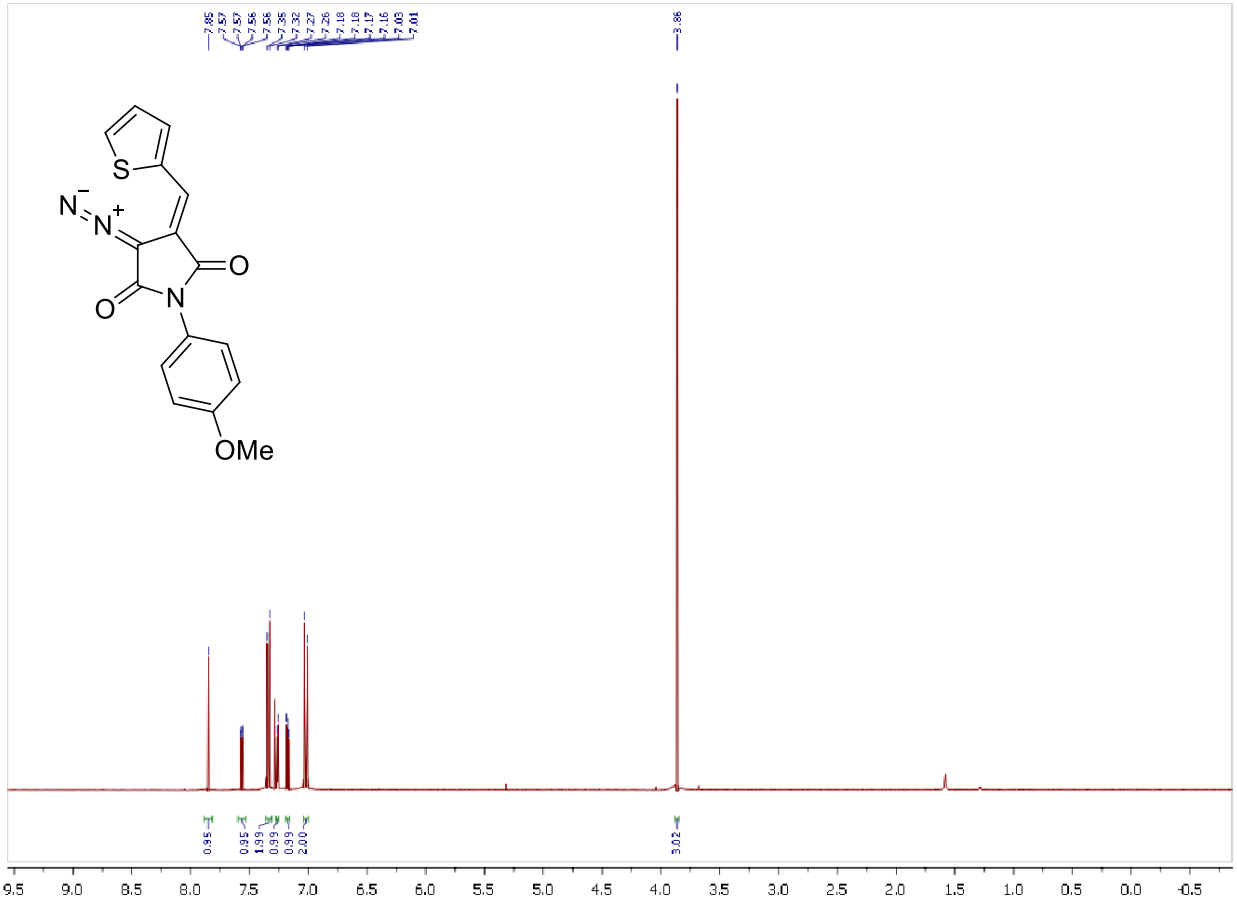

Figure S27 ^{1}H NMR spectrum of compound **2m**

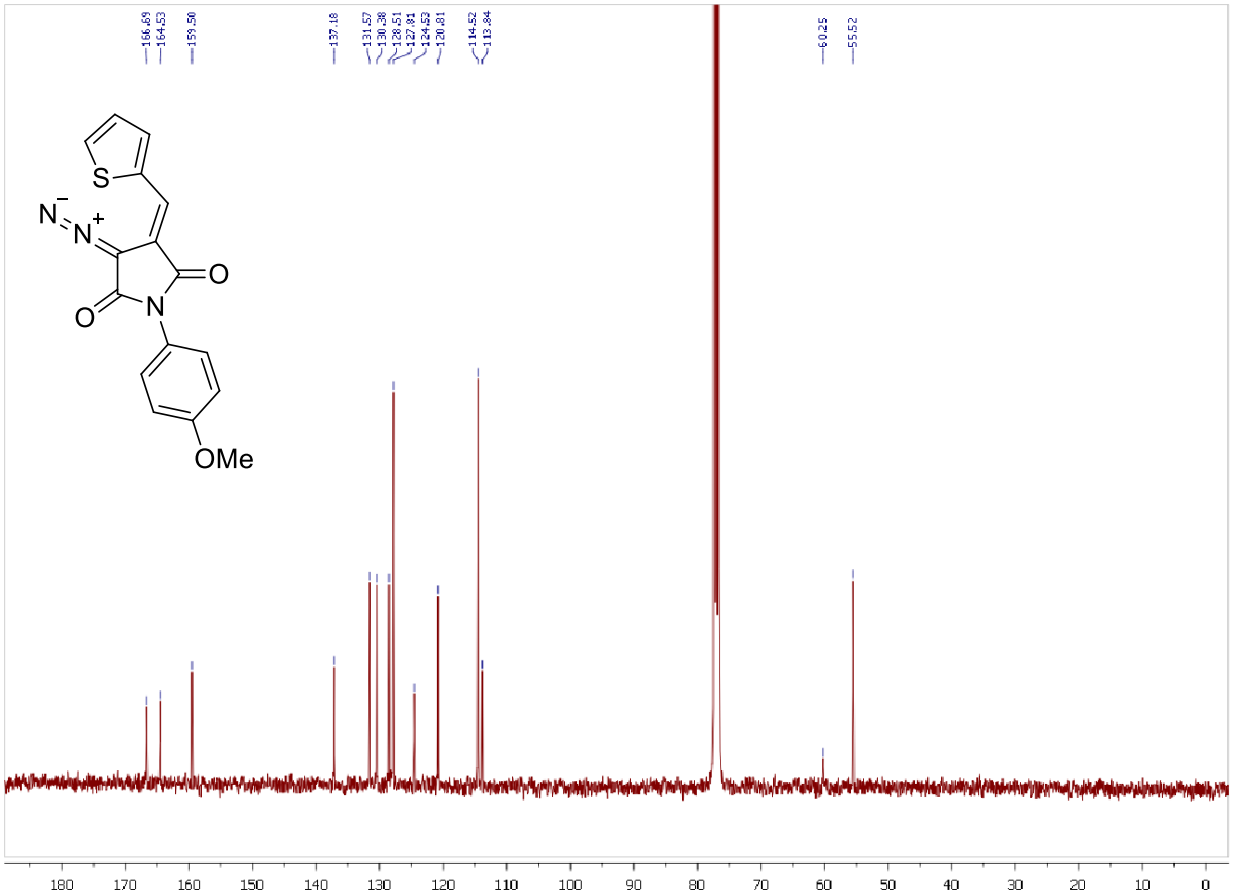

Figure S28 ^{13}C NMR spectrum of compound **2m**

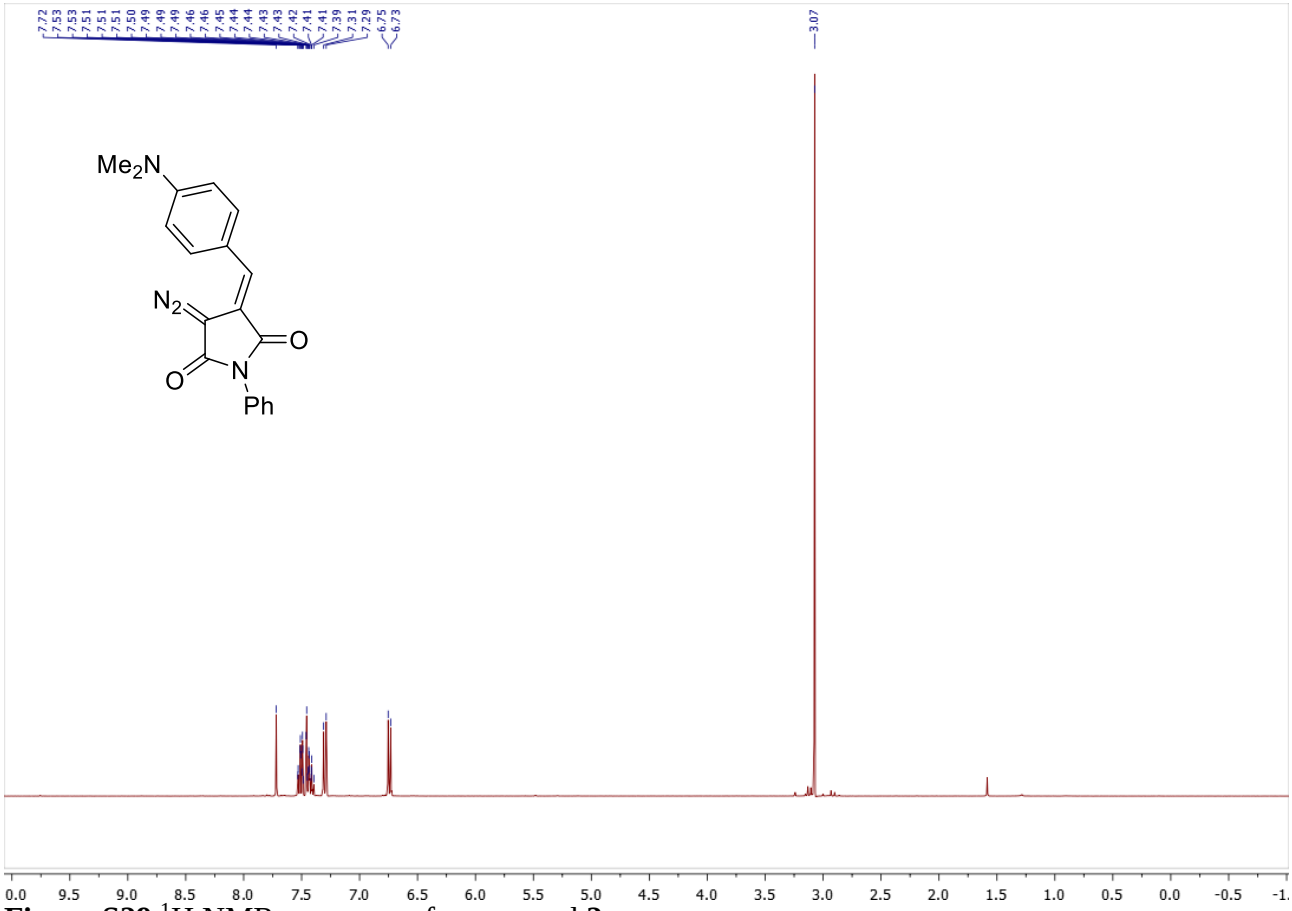

Figure S29 ^{1}H NMR spectrum of compound **2n**

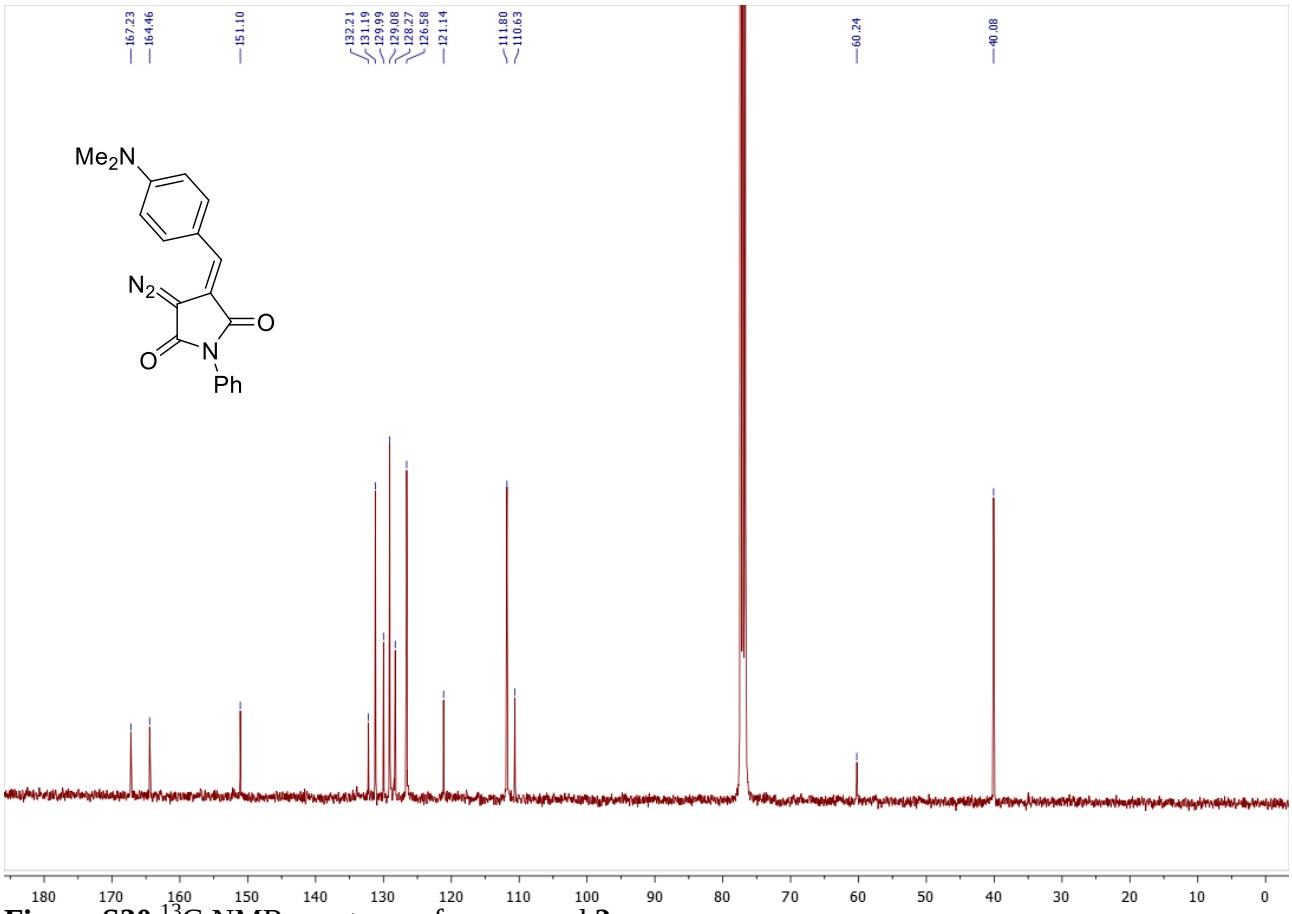

Figure S30 ^{13}C NMR spectrum of compound **2n**

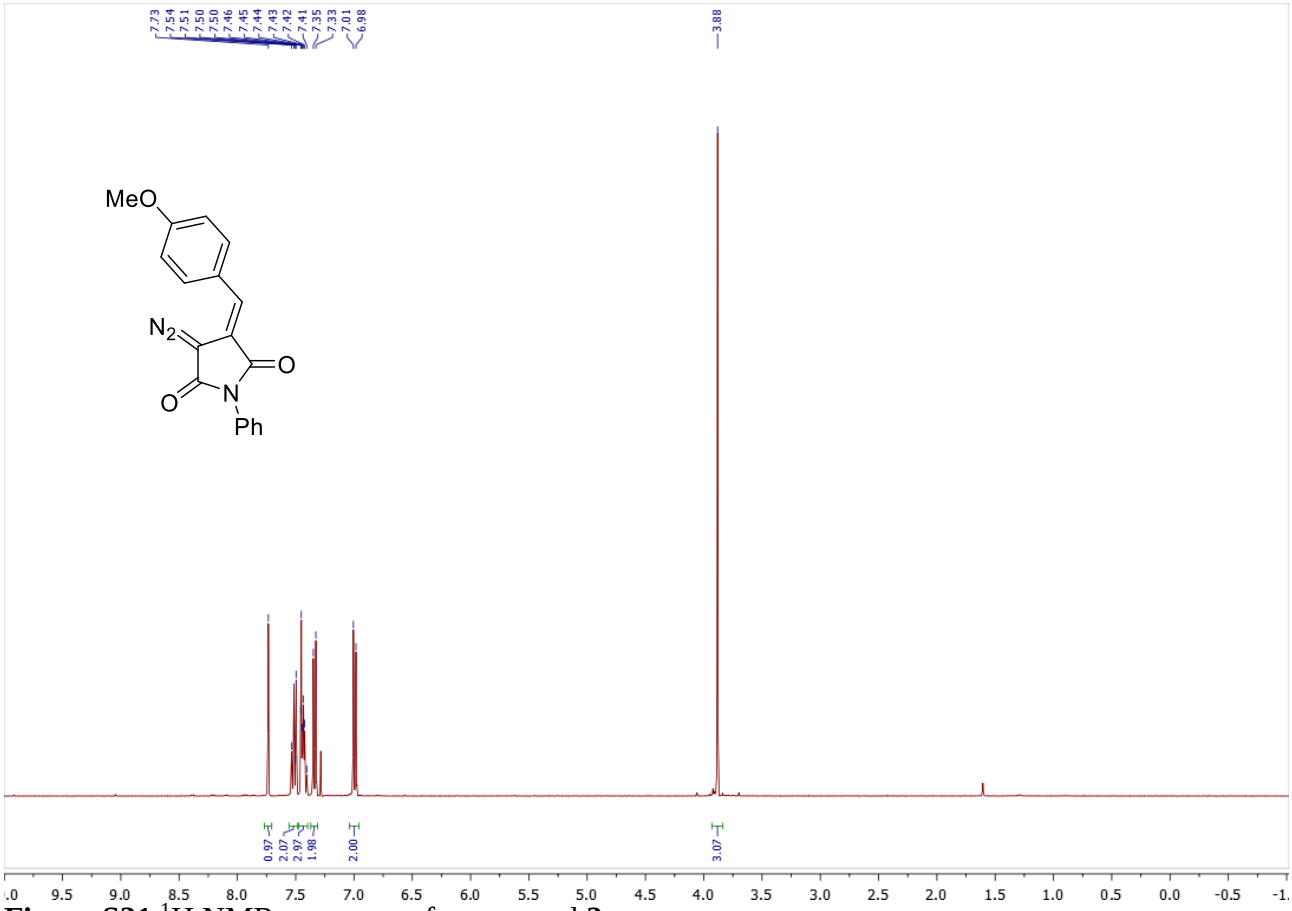

Figure S31 ^{1}H NMR spectrum of compound **2o**

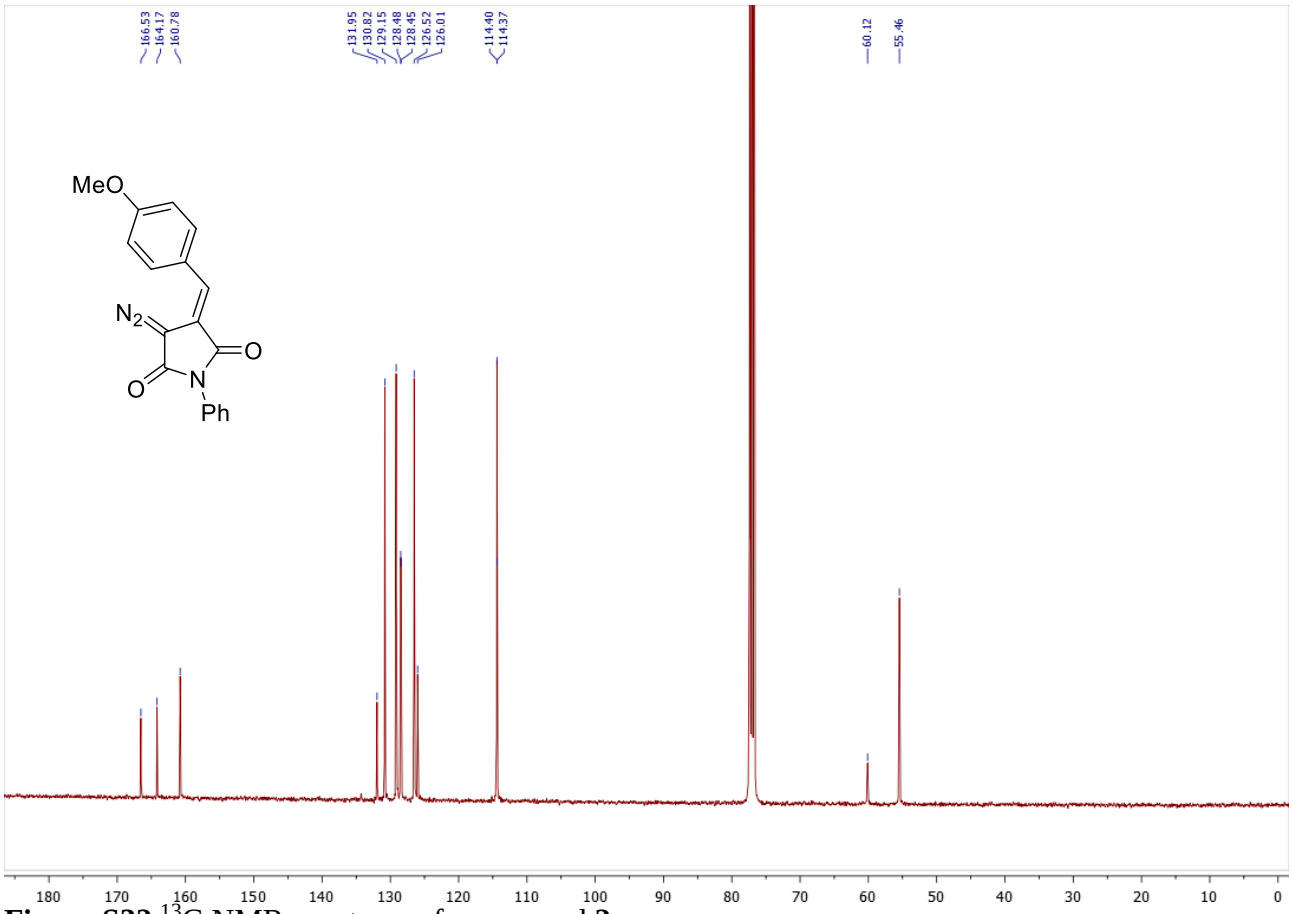

Figure S32 ^{13}C NMR spectrum of compound **2o**

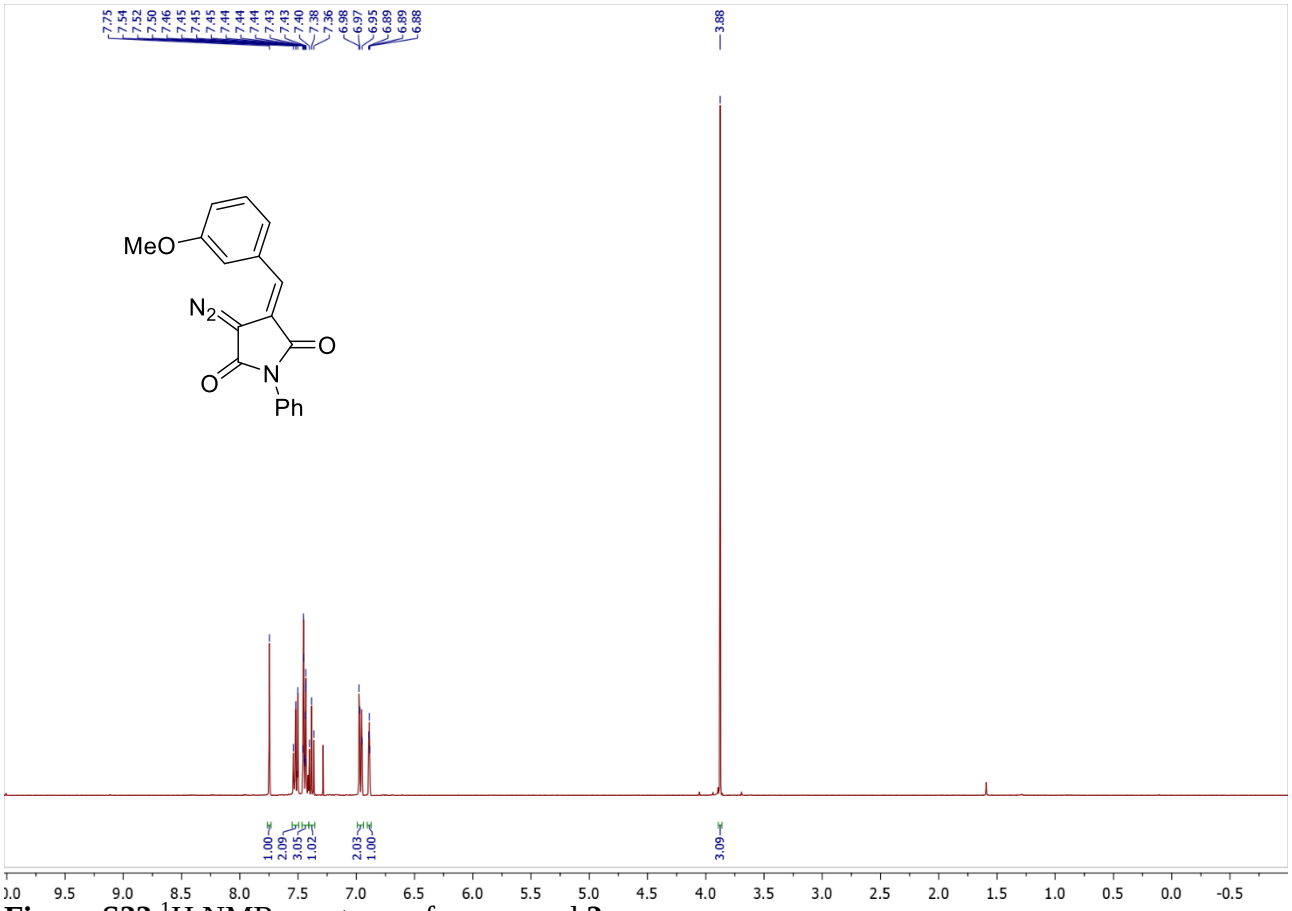

Figure S33 ^{1}H NMR spectrum of compound **2p**

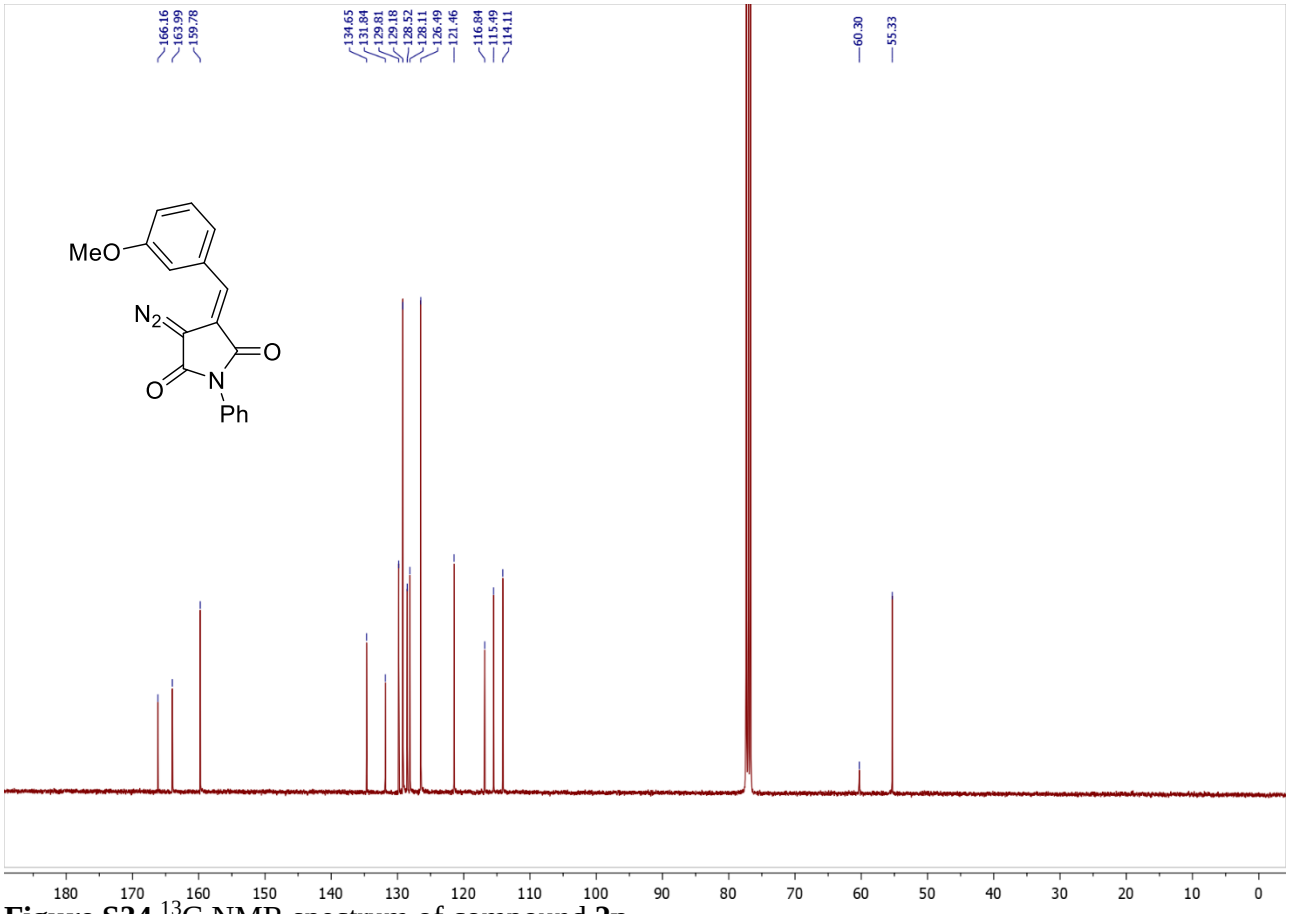

Figure S34 ^{13}C NMR spectrum of compound **2p**

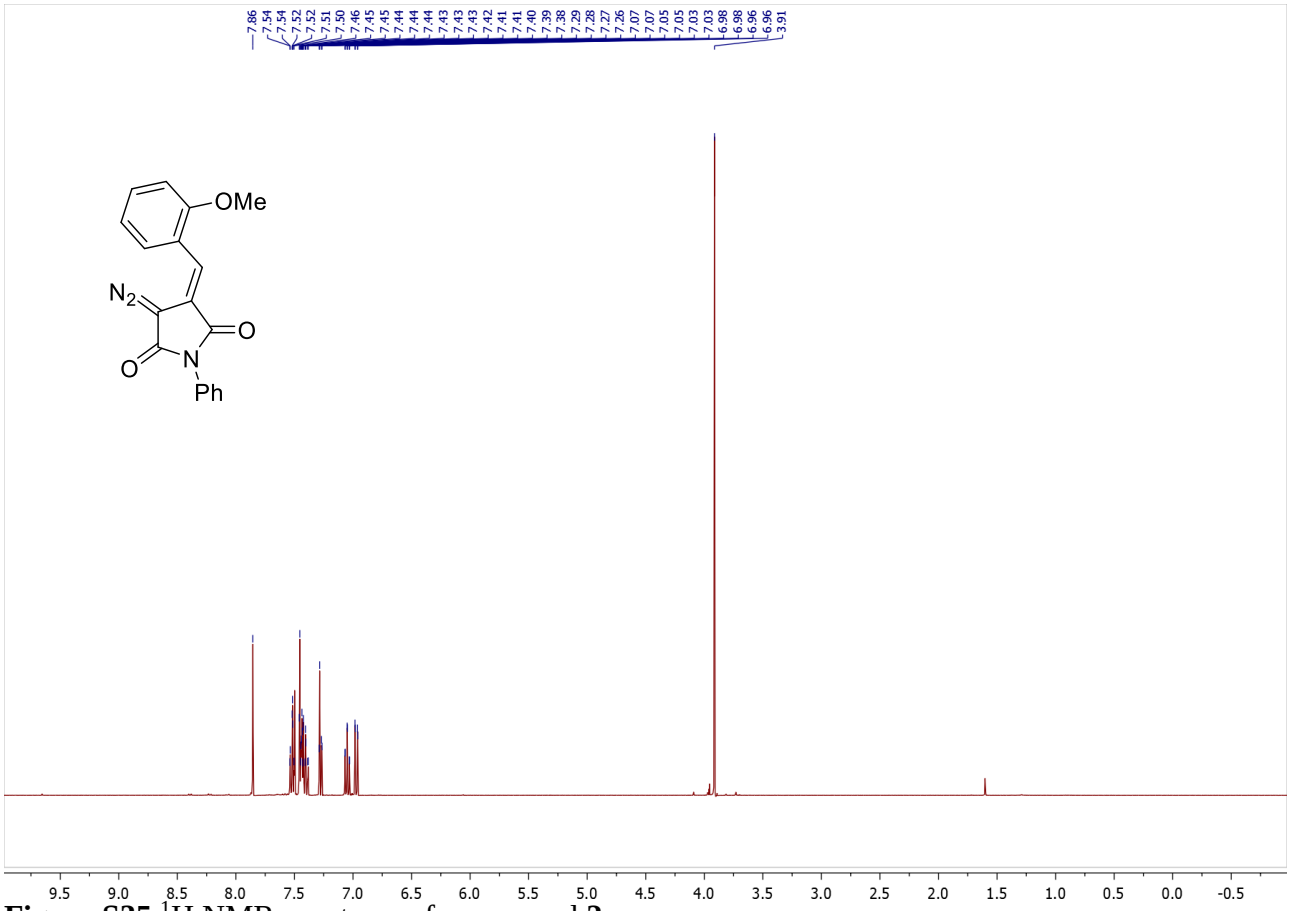

Figure S35 ^{1}H NMR spectrum of compound **2q**

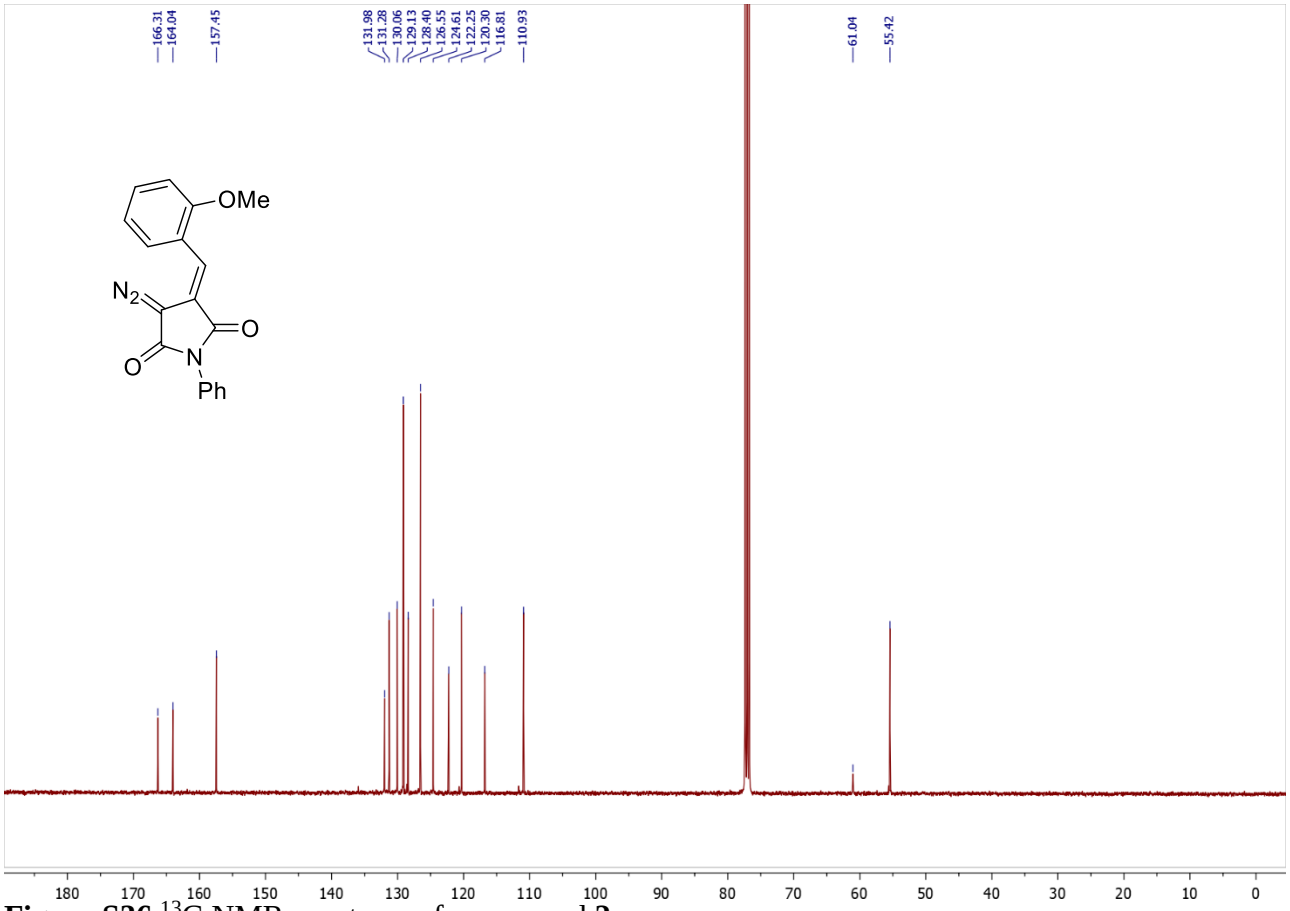

Figure S36 ^{13}C NMR spectrum of compound **2q**

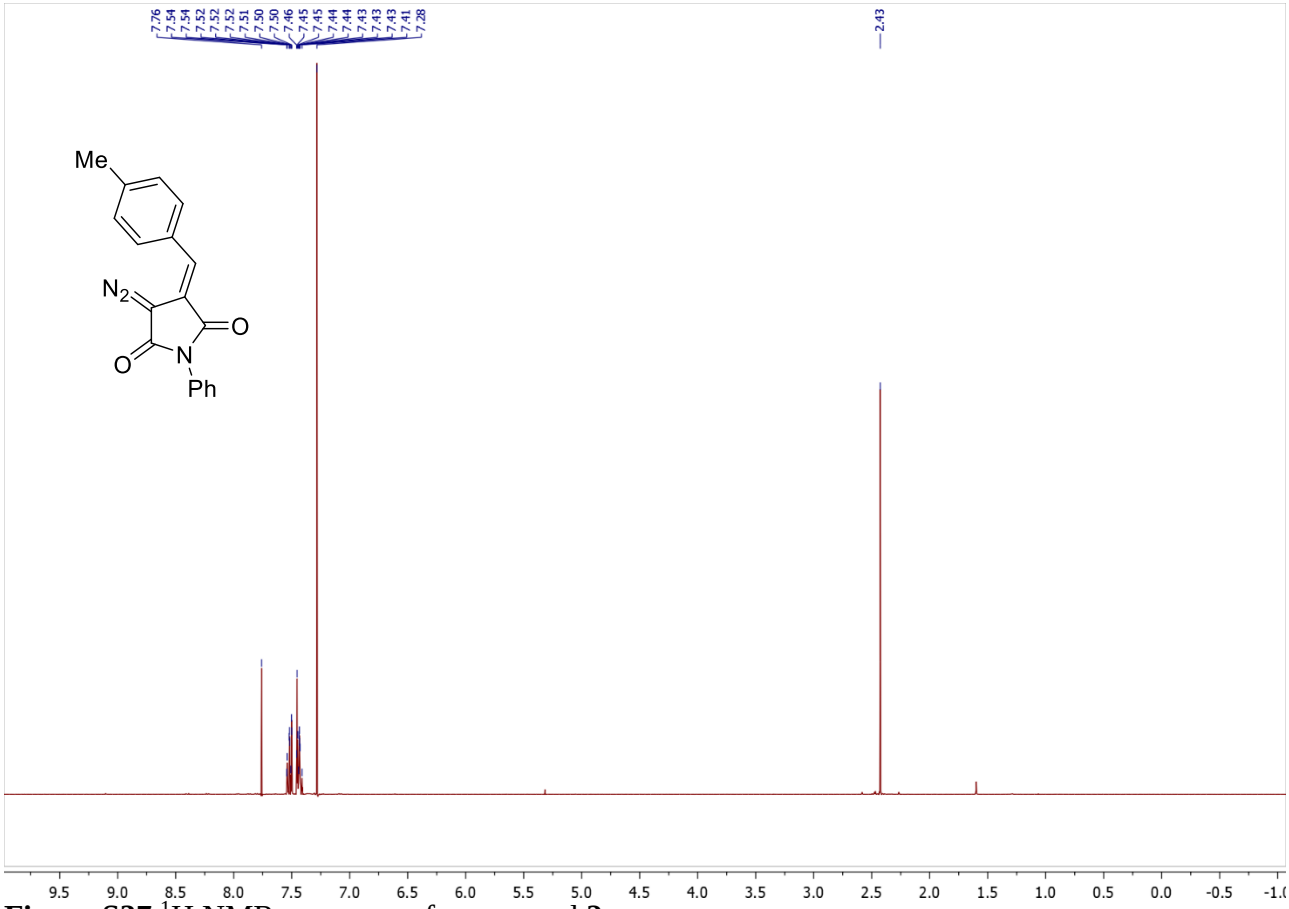

Figure S37 ^{1}H NMR spectrum of compound **2r**

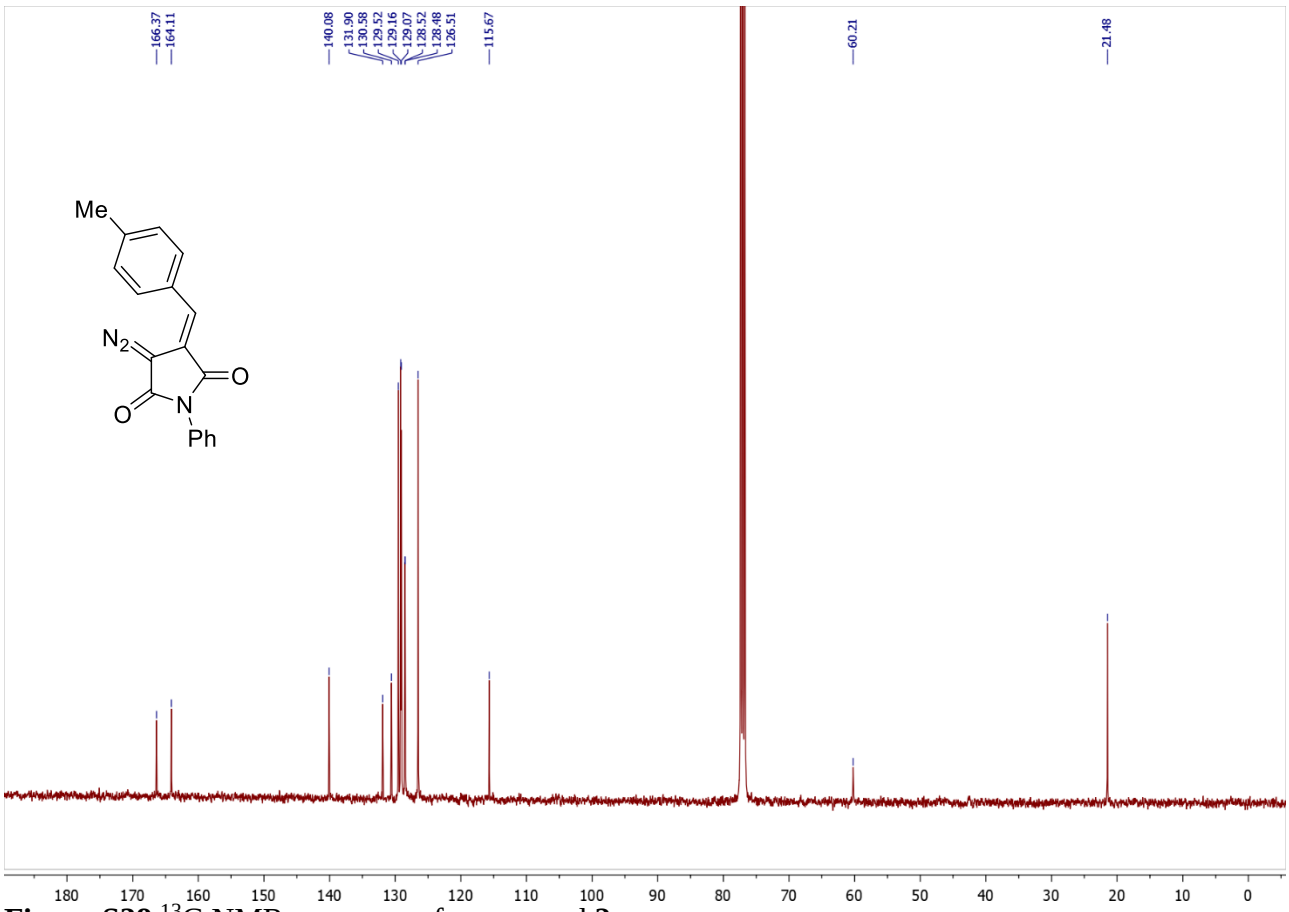

Figure S38 ^{13}C NMR spectrum of compound **2r**

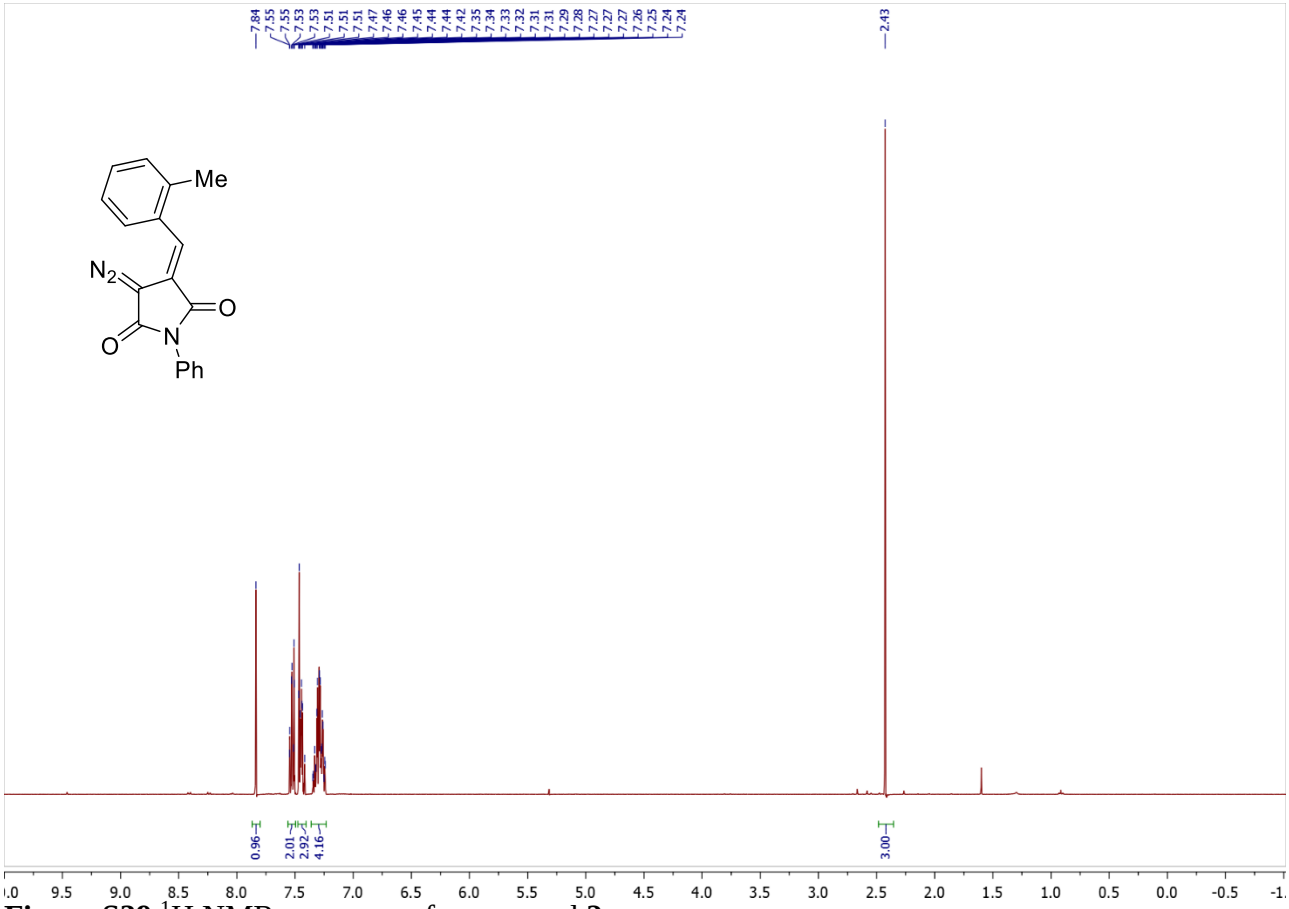

Figure S39 ^{1}H NMR spectrum of compound **2s**

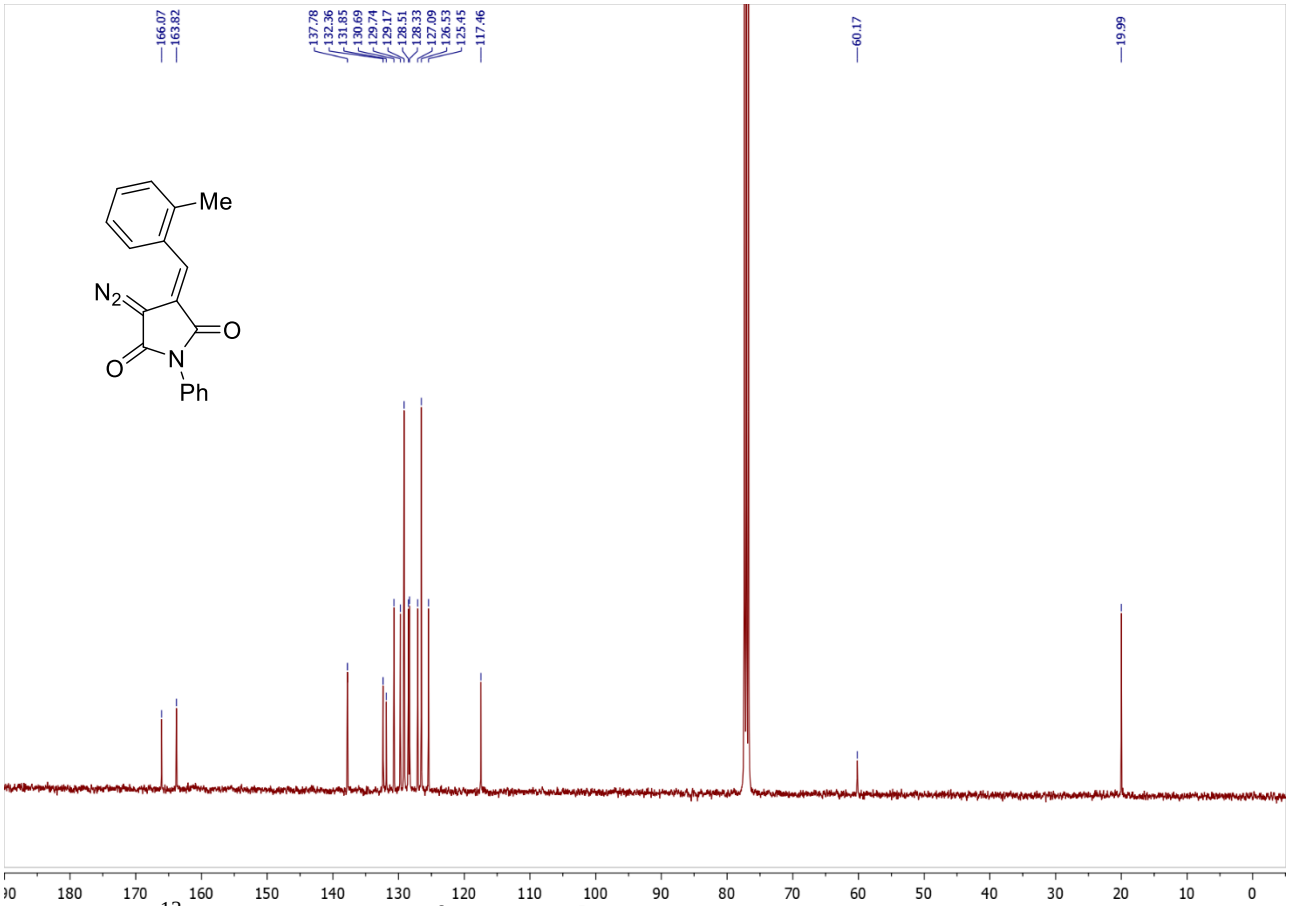

Figure S40 ^{13}C NMR spectrum of compound **2s**

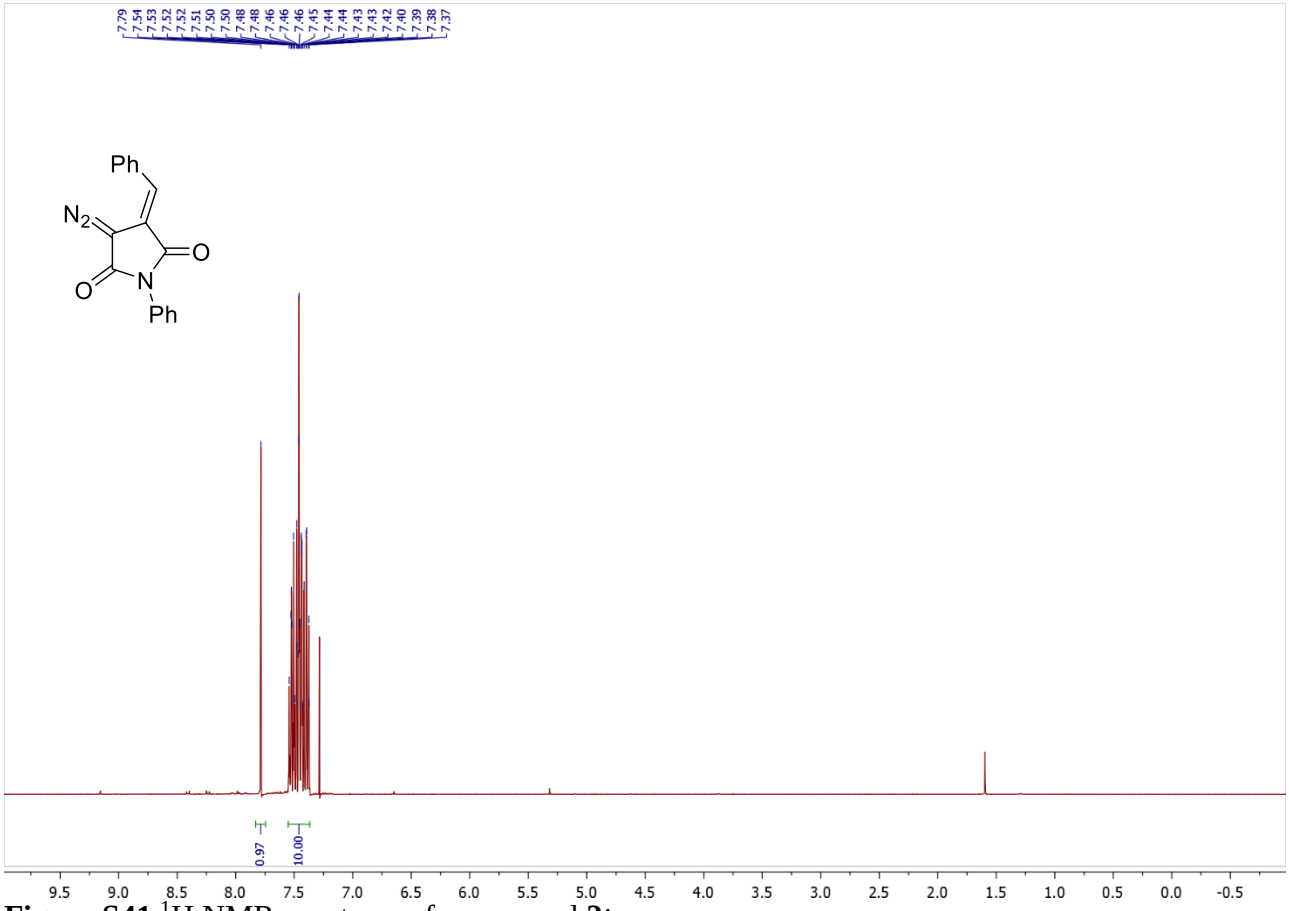

Figure S41 ^{1}H NMR spectrum of compound **2t**

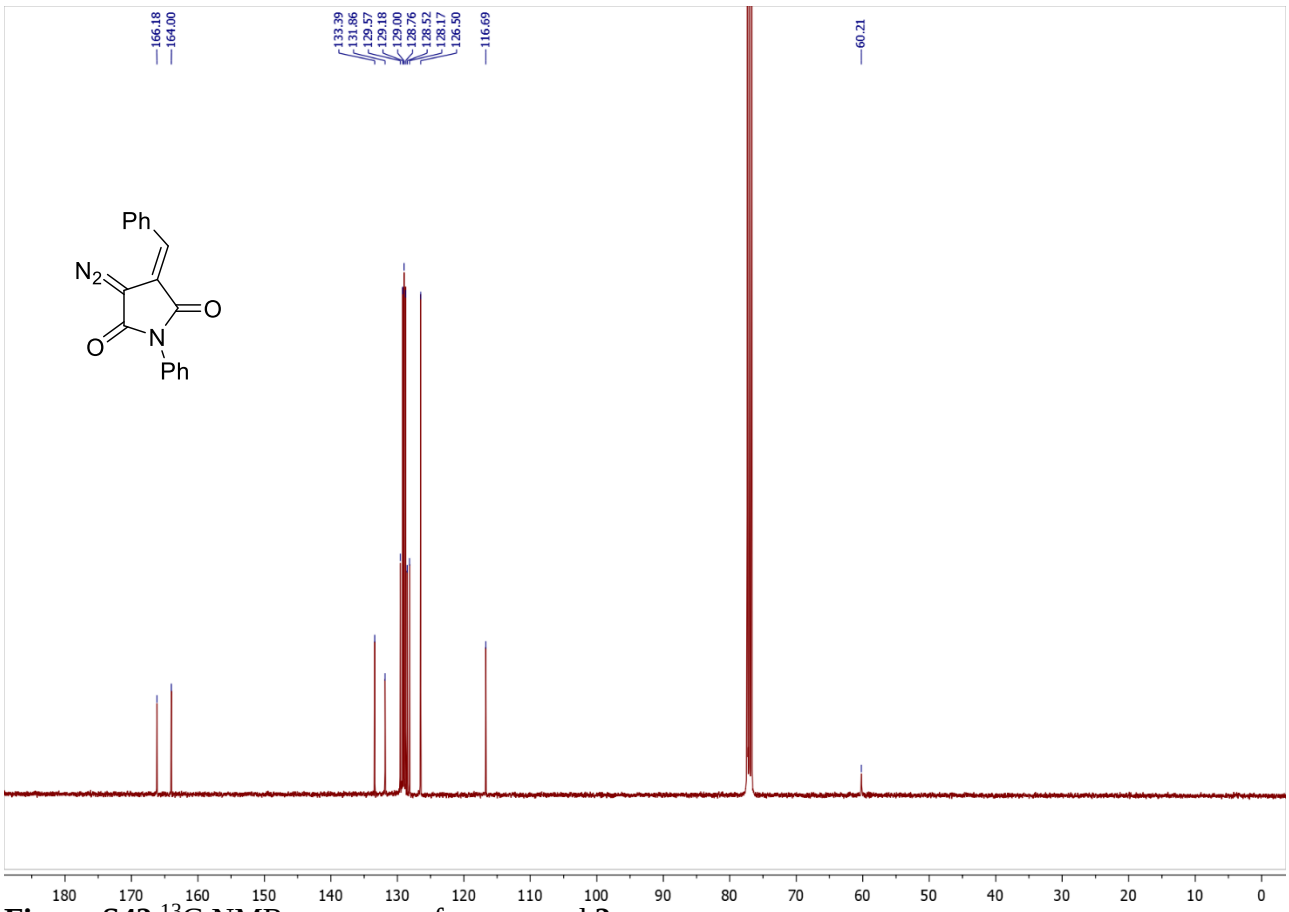

Figure S42 ^{13}C NMR spectrum of compound **2t**

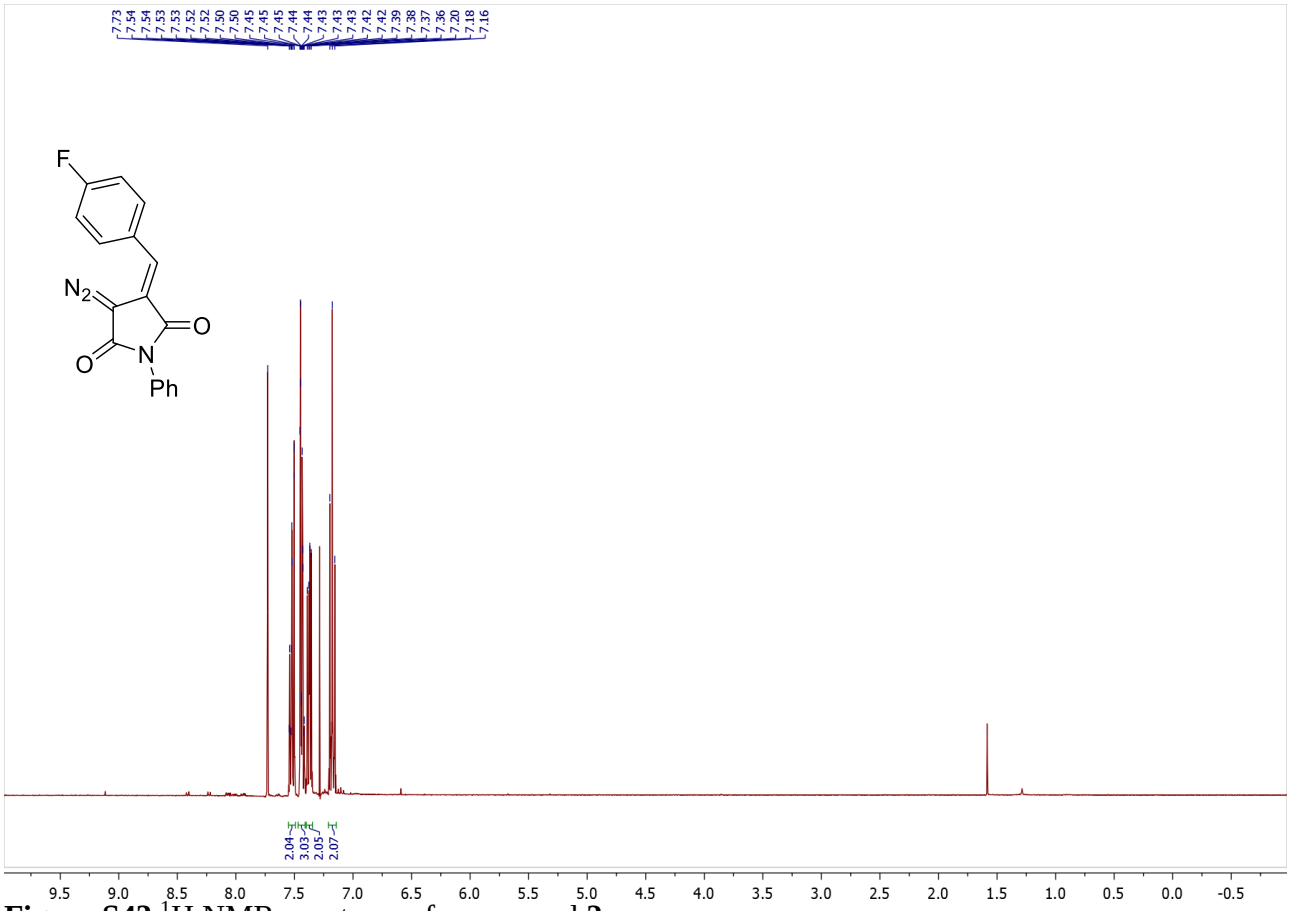

Figure S43 ^{1}H NMR spectrum of compound **2u**

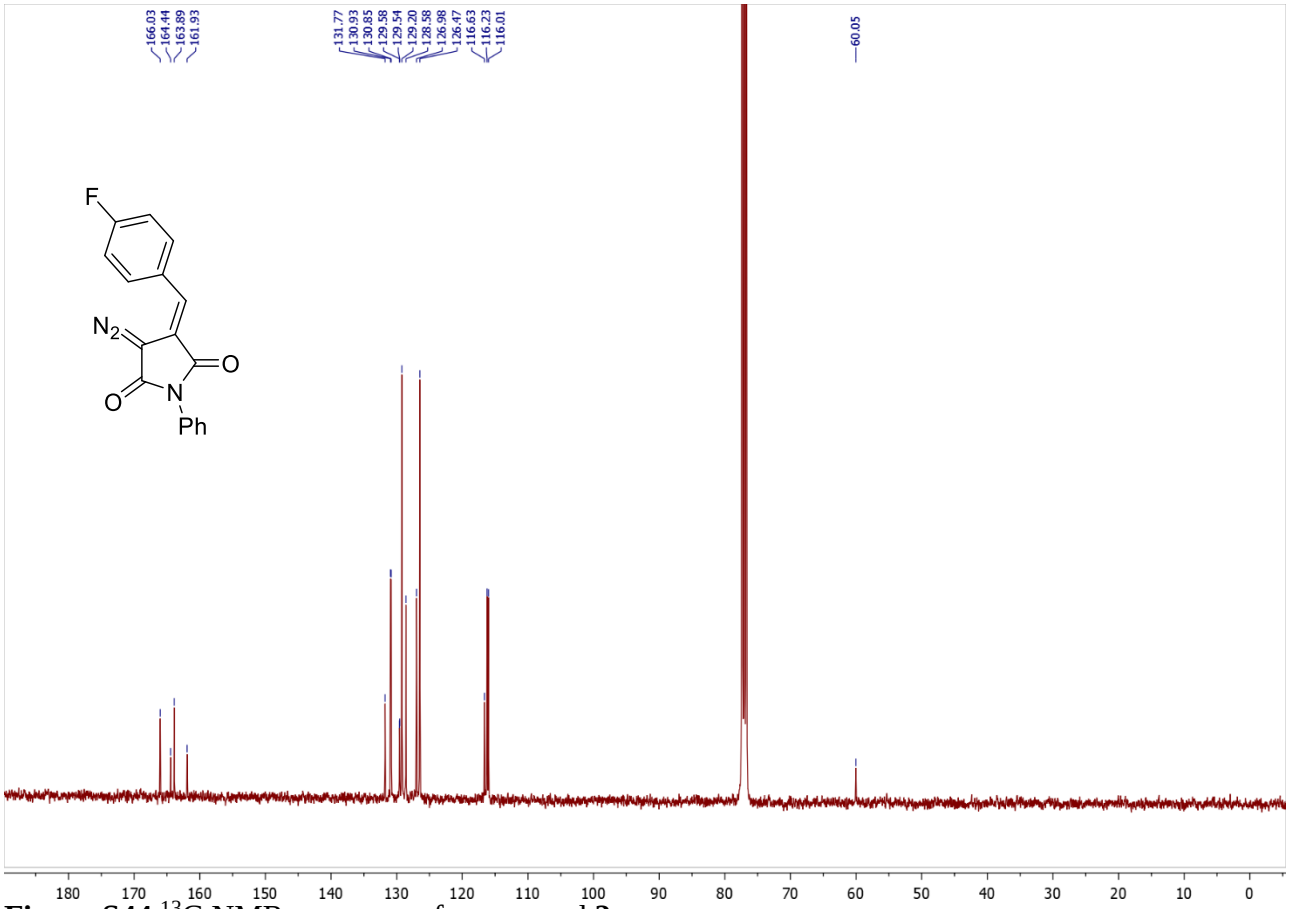

Figure S44 ^{13}C NMR spectrum of compound **2u**

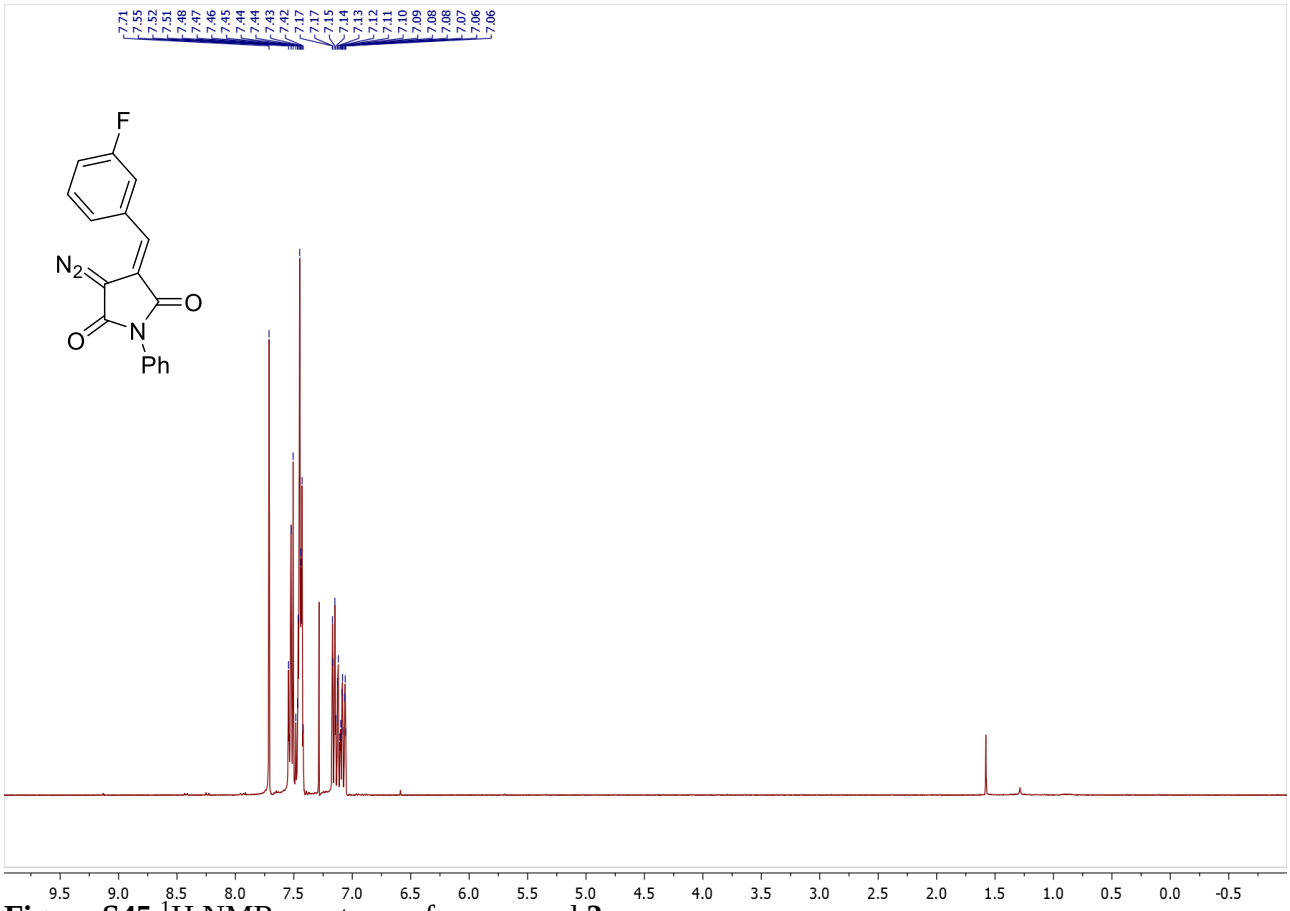

Figure S45 ^{1}H NMR spectrum of compound **2v**

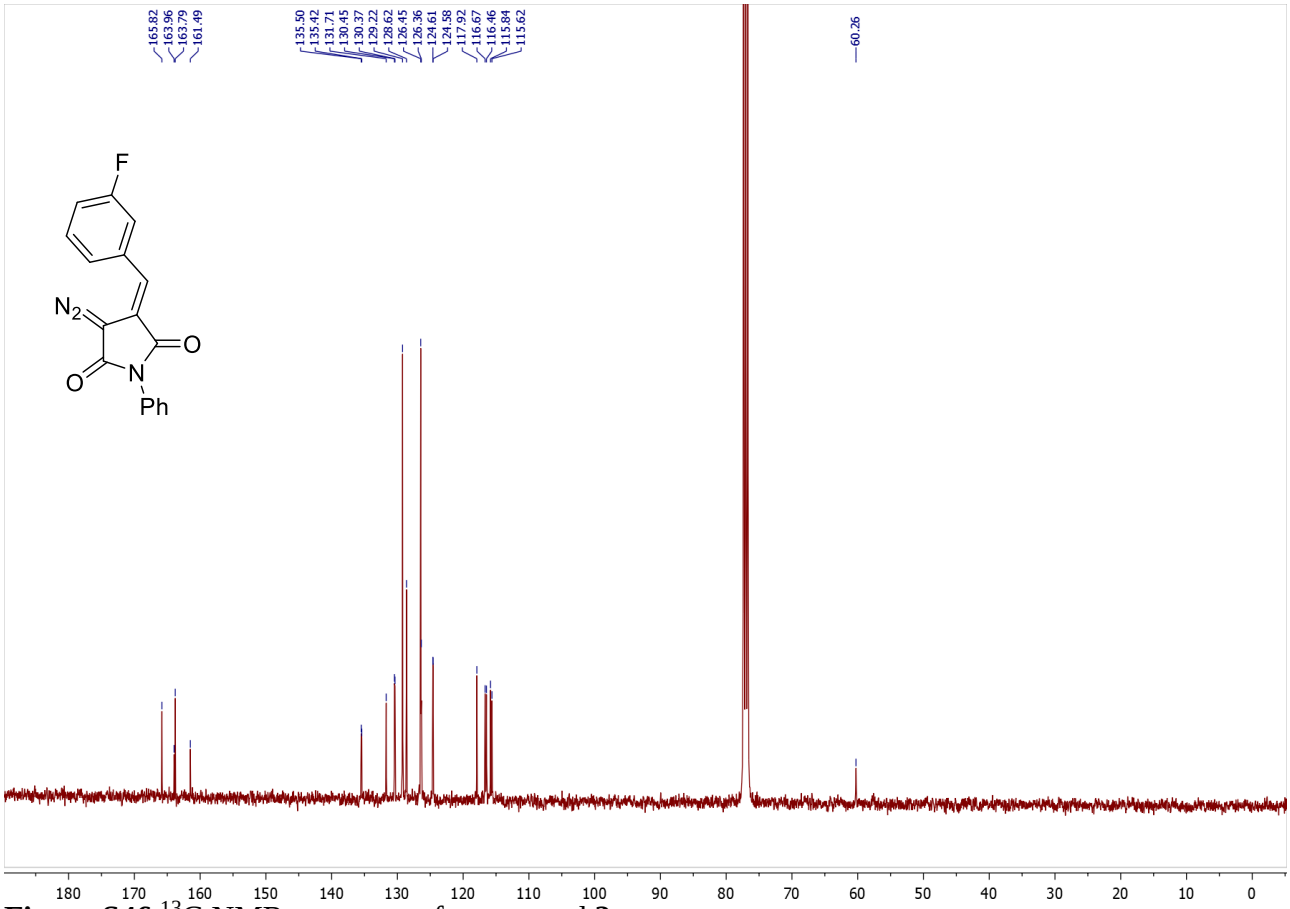

Figure S46 ^{13}C NMR spectrum of compound **2v**

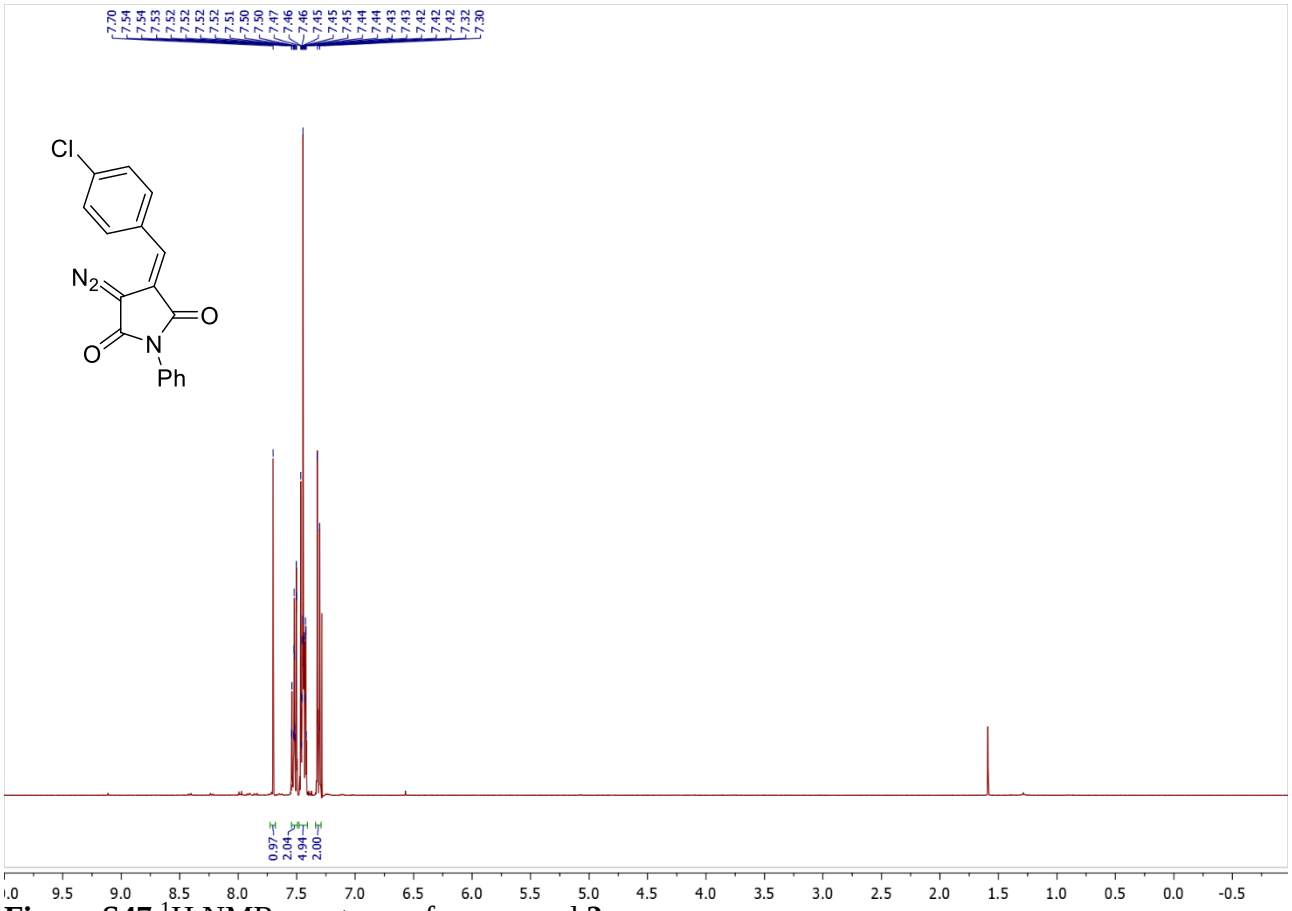

Figure S47 ^{1}H NMR spectrum of compound **2w**

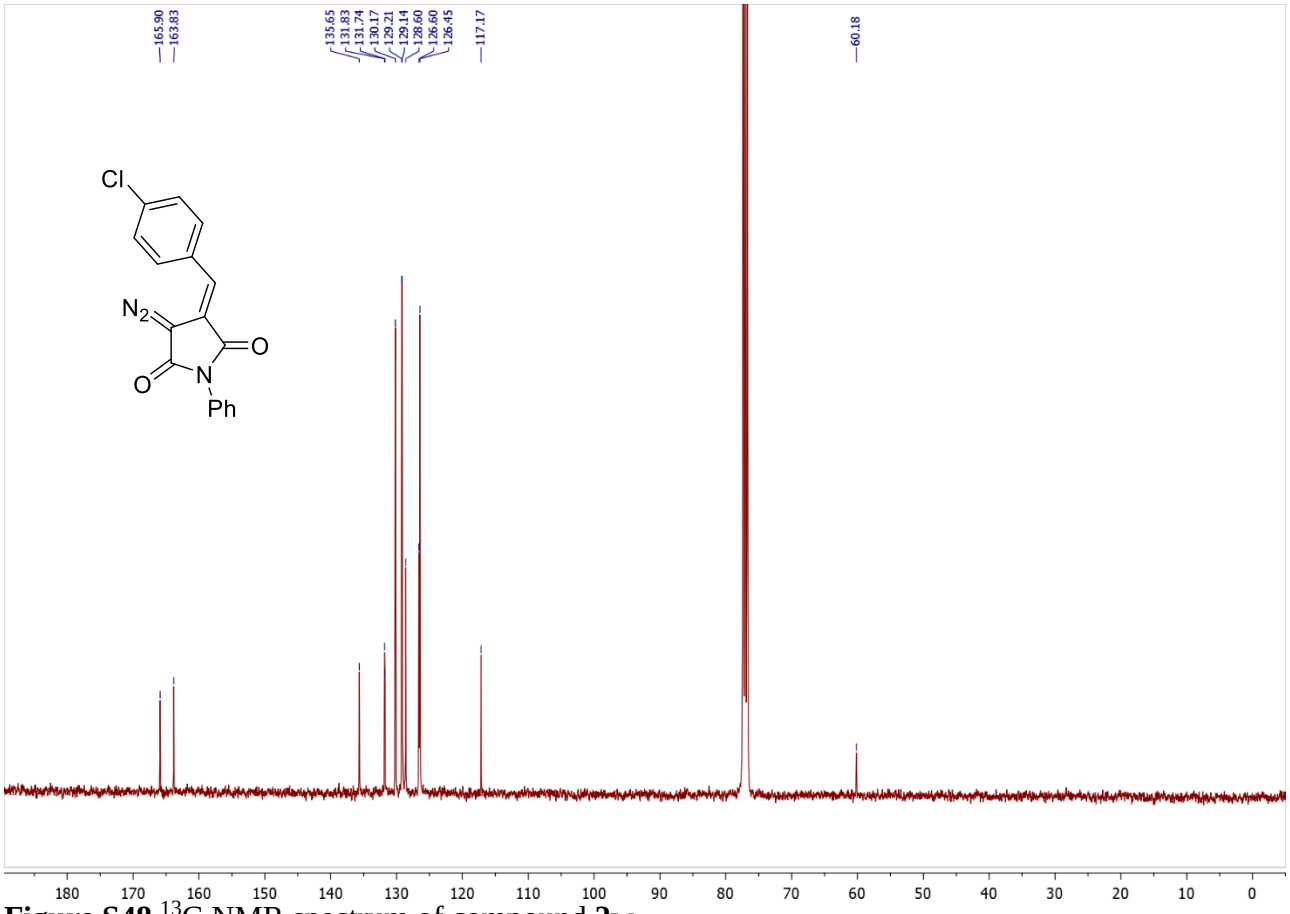

Figure S48 ^{13}C NMR spectrum of compound **2w**

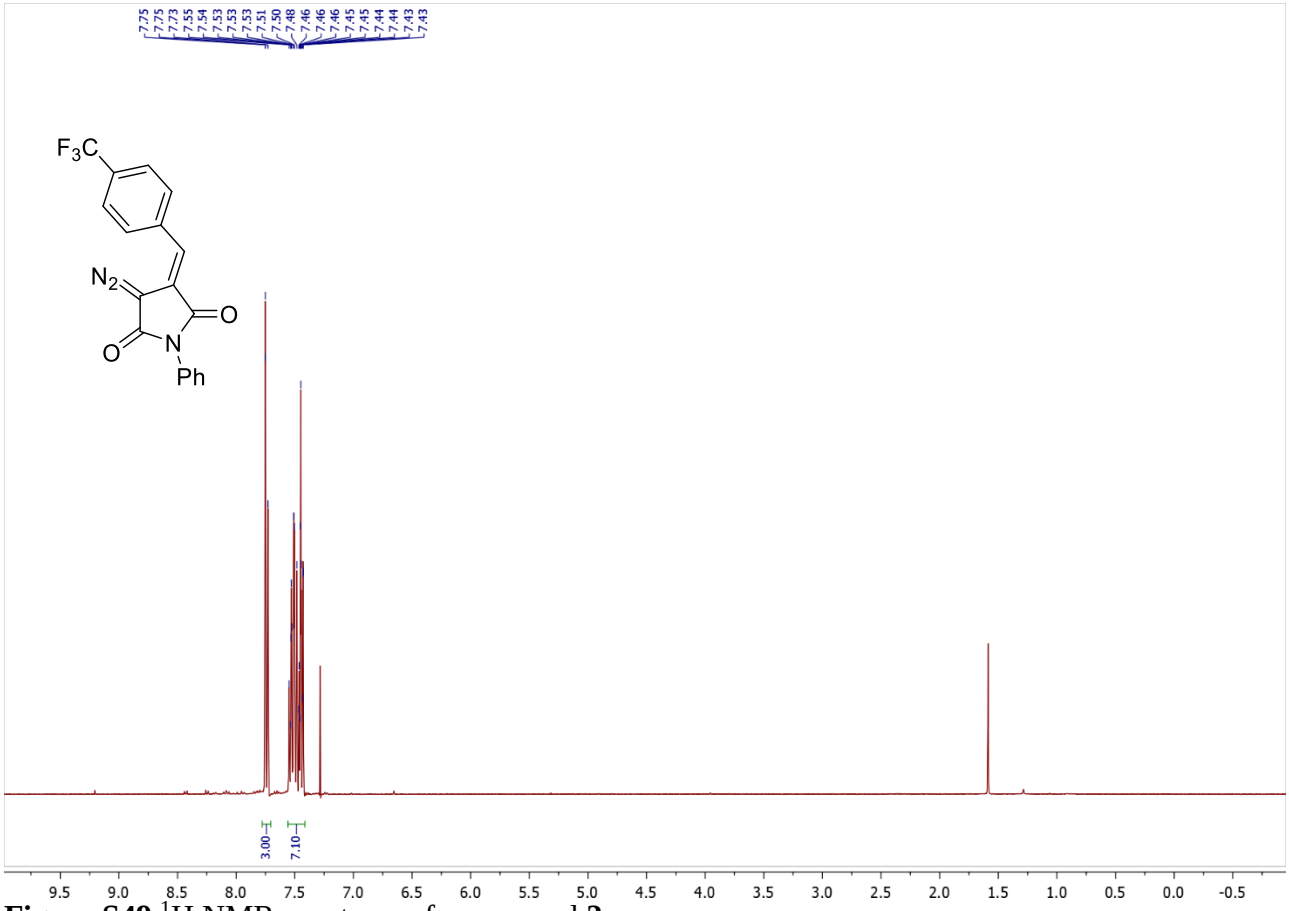

Figure S49 ^{1}H NMR spectrum of compound **2x**

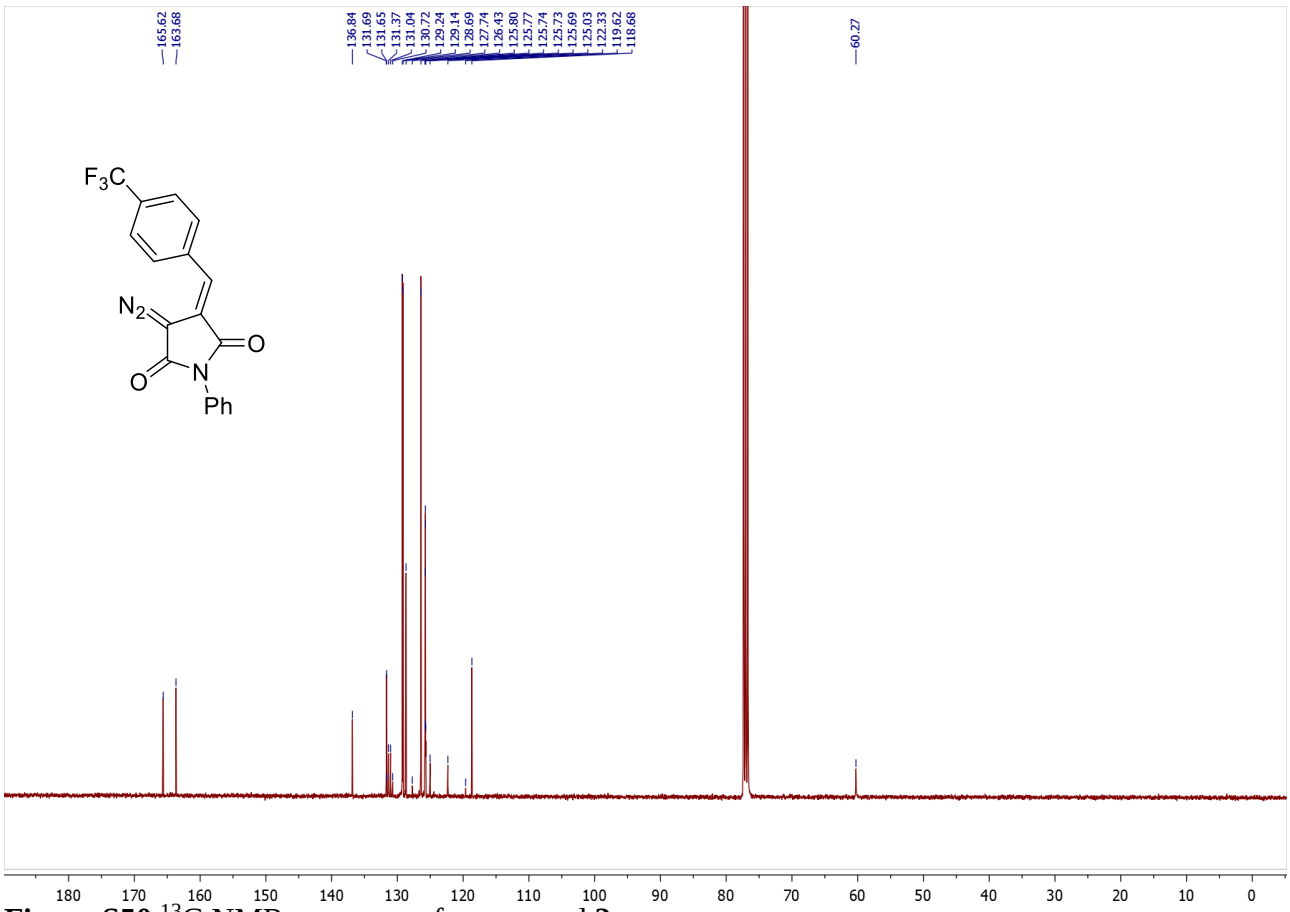

Figure S50 ^{13}C NMR spectrum of compound **2x**

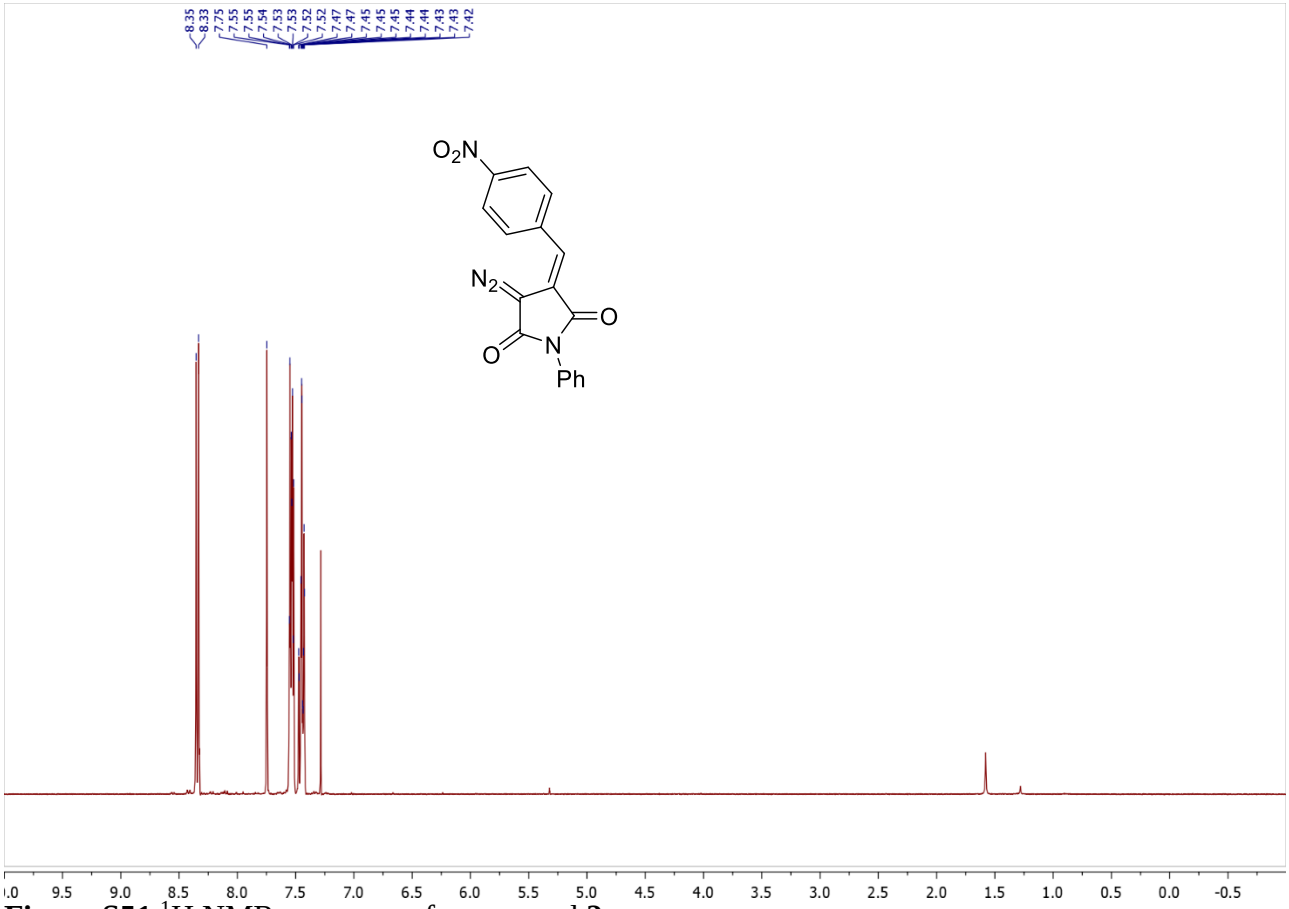

Figure S51 ^{1}H NMR spectrum of compound **2y**

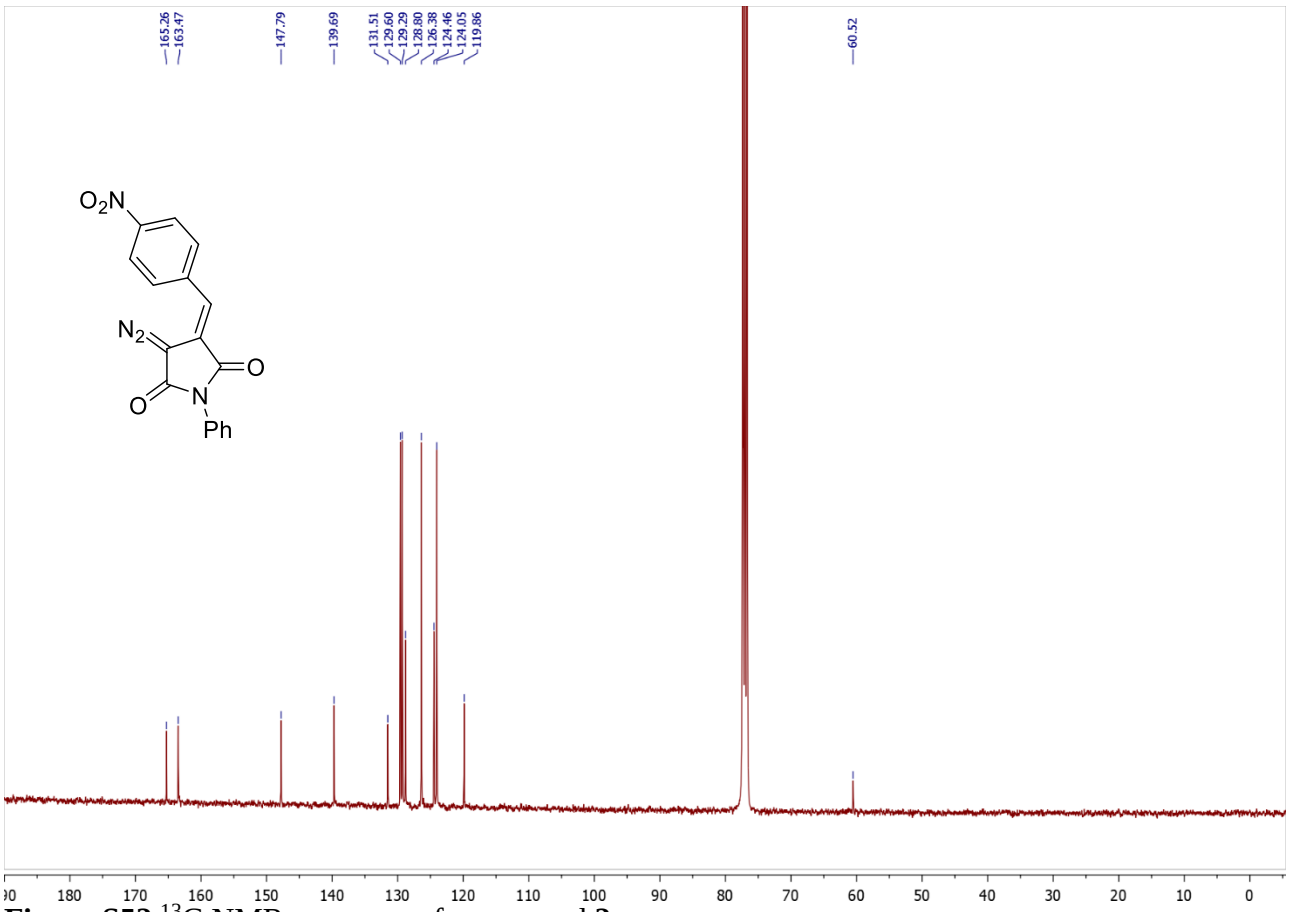

Figure S52 ^{13}C NMR spectrum of compound **2y**

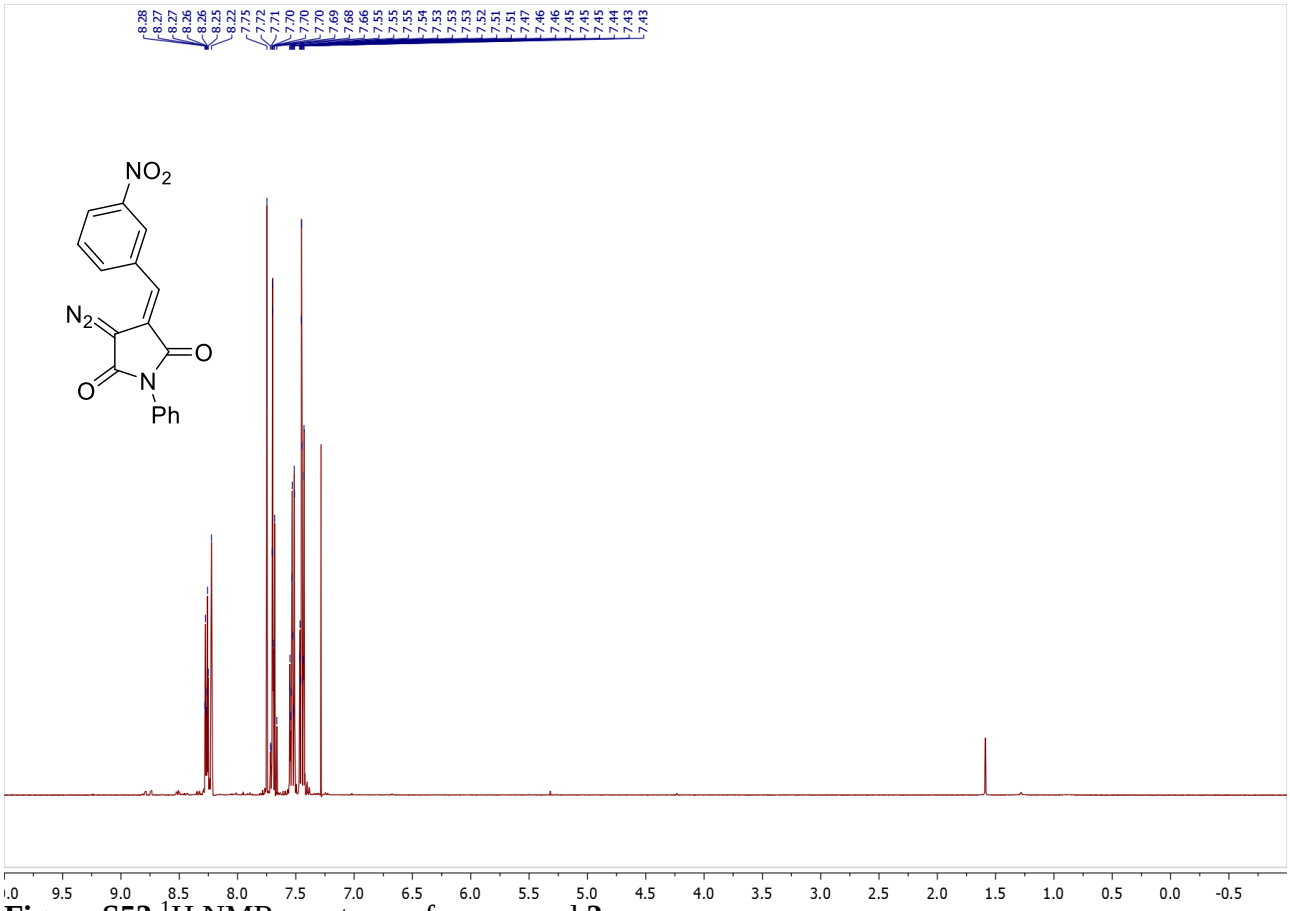

Figure S53 ^{1}H NMR spectrum of compound **2z**

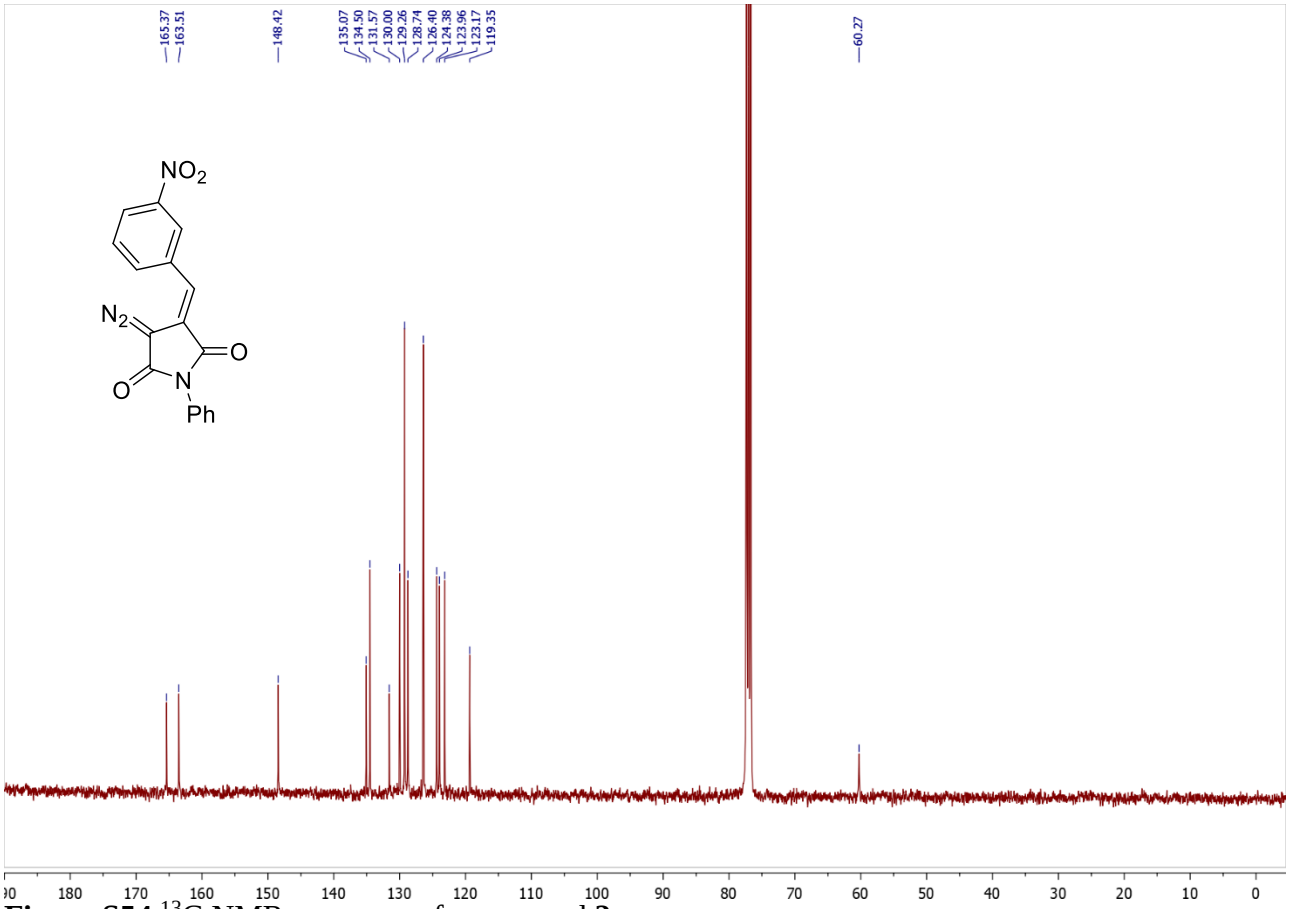

Figure S54 ^{13}C NMR spectrum of compound **2z**