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Supporting Information

Straightforward synthesis of diverse dipyrazolylmethane derivatives and their application for fluorescence sensing of Cu^{2+} ions

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Contents

I. **General Information**

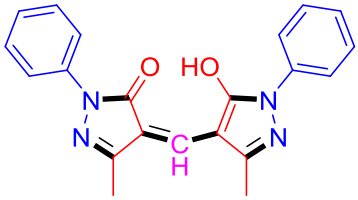
All experiments were carried out in a nitrogen atmosphere. Merck, pre-coated silica gel plates (Art. 5554) with a fluorescent indicator were used for analytical thin layer chromatography (TLC). Flash column chromatography was performed using silica gel 9385 (Merck). ^{1}H nuclear magnetic resonance (NMR) spectra were recorded on Varian 300 MHz or 600 MHz spectrometers and the chemical shifts were reported in parts per million (δ) relative to tetramethylsilane (TMS) (0 ppm) as the internal standard or relative to the resonance of the residual protonated solvent (^{1}H: $CDCl_3$, δ = 7.24 ppm; DMSO, δ = 2.50 ppm). The following abbreviations were used to describe the peak patterns where appropriate: singlet (s), doublet (d), triplet (t), quartet (q), and multiplet (m). The coupling constants, *J*, are reported in Hertz (Hz). The ^{13}C NMR spectra were obtained using 75 MHz or 150 MHz spectrometers and referenced to the internal solvent signals (^{13}C: $CDCl_3$, δ = 77.0 ppm; DMSO, δ = 39.51 ppm). All melting points were obtained on a Fisher-Johns melting point apparatus and were uncorrected. The Fourier transform infrared (FT-IR) spectra were recorded on a Jasco FTIR 5300 spectrophotometer. The high-resolution mass spectra **(**HRMS) were measured using a JEOL JMS-600 mass spectrometer (positive ion EI mode) at the Korean Basic Science Institute.

II. General Procedures for the preparation of Dipyrazolylmethanes (4a-4p)

p-Toluenesulfonic acid monohydrate (*p*-TsOH • H_2O, 1 equiv.) was added to a solution of β-keto esters (1.0 mmol) and phenyl hydrazine hydrochlorides (1.0 mmol) in dry DMF (5.0 ml). The reaction mixture was stirred at 140 ^oC for the requested time. After the reaction was complete, as indicated by TLC, the mixture was allowed to cool to room temperature, quenched with water, and extracted with ethyl acetate. The organic layer was then dried over anhydrous $MgSO_4$ and the solvent was removed *in vacuo*. The residue was finally purified by column chromatography on silica gel using hexane-ethyl acetate mixture as the eluent to give the corresponding products **4a–4p**.

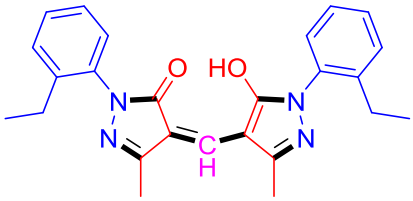
III. Spectroscopic Data for dipyrazolylmethanes

(*Z*)-4-((5-Hydroxy-3-methyl-1-phenyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2-phenyl-2,4-dihydro-3*H*-pyrazol-3-one (4a)[s1]



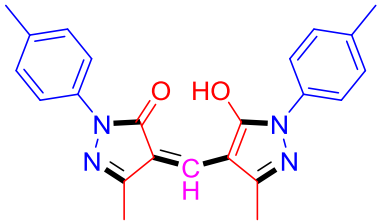
Yellow solid (134 mg, 75%): mp 127-129 ^oC; ^1H NMR (600 MHz, $CDCl_3$) δ 17.93 (s, 1H), 7.91 (d, J = 7.8 Hz, 4H), 7.44 (dd, J = 8.4, 7.8 Hz, 4H), 7.29-7.26 (m, 2H), 7.25 (s, 1H), 2.37 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 161.1, 152.6, 138.0, 137.6, 128.8, 126.3, 120.8, 109.4, 12.7; IR (KBr): 3471, 2924, 1623, 1382, 1103, 896, 754, 667 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{18}N_4O_2$: 358.1430. Found: 358.1430.

(*Z*)-2-(2-Ethylphenyl)-4-((1-(2-ethylphenyl)-5-hydroxy-3-methyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4b)



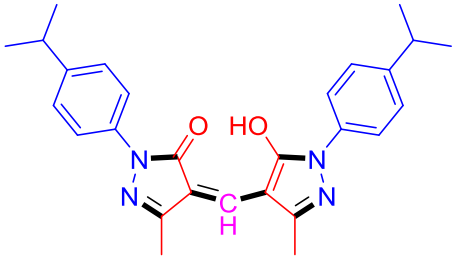
Yellow solid (141 mg, 68%): mp 174-176 ^oC; ^1H NMR (300 MHz, $CDCl_3$) δ 7.34-7.32 (m, 5H), 7.26-7.24 (m, 4H), 2.59 (q, J = 7.5 Hz, 4H), 2.35 (s, 6H), 1.14 (t, J = 7.5 Hz, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 161.9, 152.2, 141.2, 138.7, 135.0, 129.4, 129.1, 127.3, 126.4, 108.2, 24.3, 14.2, 12.8; IR (KBr): 3451, 2964, 1609, 1317, 1081, 763, 650 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{25}H_{26}N_4O_2$: 414.2056. Found: 414. 414.2057.

(*Z*)-4-((5-Hydroxy-3-methyl-1-(*p*-tolyl)-1*H*-pyrazol-4-yl)methylene)-5-methyl-2-(*p*-tolyl)-2,4-dihydro-3*H*-pyrazol-3-one (4c)



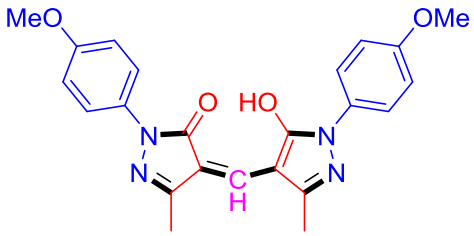
Yellow solid (141 mg, 73%): mp 229-231 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.69 (d, J = 8.1 Hz, 4H), 7.18-7.14 (m, 5H), 2.30 (s, 6H), 2.27 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 161.0, 152.5, 138.1, 136.3, 135.2, 129.4, 121.0, 109.4, 21.0, 12.9; IR (KBr): 3484, 2370, 1623, 1494, 1380, 1327, 1110, 1003, 889, 753, 663 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{23}H_{22}N_4O_2$: 386.1743. Found: 386.1745.

(*Z*)-4-((5-Hydroxy-1-(4-isopropylphenyl)-3-methyl-1*H*-pyrazol-4-yl)methylene)-2-(4-isopropylphenyl)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4d)



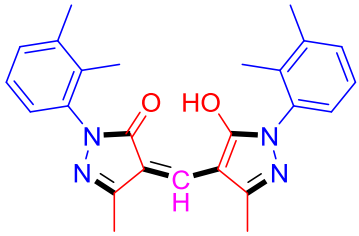
Yellow solid (153 mg, 69%): mp 236-238 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.83 (d, J = 8.1 Hz, 4H), 7.32 (d, J = 8.4 Hz, 4H), 7.24 (s, 1H), 3.02-2.91 (m, 2H), 2.36 (s, 6H), 1.30 (d, J = 6.6 Hz, 12H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 161.0, 152.5, 147.3, 138.0, 135.4, 126.8, 121.1, 109.3, 33.7, 23.9, 12.7; IR (KBr): 3458, 2963, 1606, 1315, 1079, 822, 592 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{27}H_{30}N_4O_2$: 442.2369. Found: 442.2371.

(*Z*)-4-((5-Hydroxy-1-(4-methoxyphenyl)-3-methyl-1*H*-pyrazol-4-yl)methylene)-2-(4-methoxyphenyl)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4e)[s2]



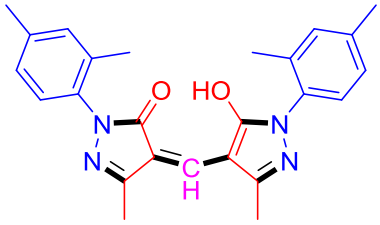
Yellow solid (151 mg, 72%): mp 211-213 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.79 (d, J = 8.7 Hz, 4H), 7.28 (s, 1H), 6.97 (d, J = 9.0 Hz, 4H), 3.85 (s, 6H), 2.37 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 160.7, 158.0, 152.5, 138.1, 130.9, 122.8, 114.0, 109.3, 55.5, 12.9; IR (KBr): 3477, 2941, 1616, 1511, 1323, 1167, 1009, 821, 596 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{23}H_{22}N_4O_4$: 418.1641. Found: 418.1642.

(*Z*)-2-(2,3-Dimethylphenyl)-4-((1-(2,3-dimethylphenyl)-5-hydroxy-3-methyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4f)[s3]



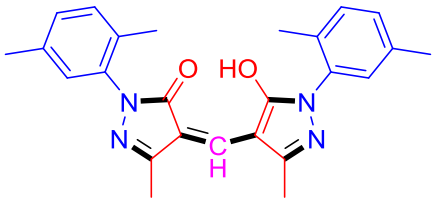
Orange solid (145 mg, 70%): mp 236-238 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.26 (s, 1H), 7.13-7.06 (m, 6H), 2.28 (s, 6H), 2.22 (s, 6H), 2.02 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 157.1, 147.7, 134.1, 133.5, 131.0, 129.3, 125.9, 121.3, 120.0, 103.6, 15.6, 9.9, 8.2; IR (KBr): 3467, 2929, 1608, 1318, 998, 775, 604 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{25}H_{26}N_4O_2$: 414.2056. Found: 414.2052.

(*Z*)-2-(2,4-Dimethylphenyl)-4-((1-(2,4-dimethylphenyl)-5-hydroxy-3-methyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4g)



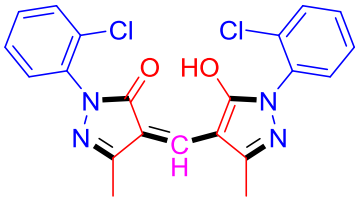
Yellow solid (155 mg, 75%): mp 156-158 ^{o}C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 17.59 (s, 1H), 7.31(s, 1H), 7.16 (d, J = 7.8 Hz, 2H), 7.08 (s, 2H), 7.05 (d, J = 8.4 Hz, 2H), 2.35 (s, 6H), 2.32 (s, 6H), 2.18 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 161.7, 152.3, 139.0, 138.6, 134.9, 133.1, 131.5, 127.2, 126.7, 108.3, 21.1, 17.9, 12.9; IR (KBr): 3490, 2957, 1613, 1320, 1081, 812 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{25}H_{26}N_4O_2$: 414.2056. Found: 414.2052.

(*Z*)-2-(2,5-Dimethylphenyl)-4-((1-(2,5-dimethylphenyl)-5-hydroxy-3-methyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4h)



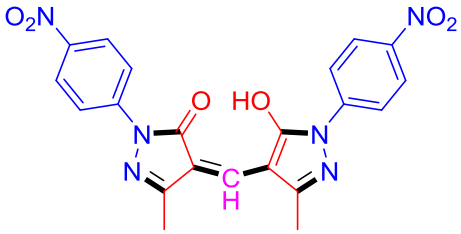
Yellow solid (162 mg, 78%): mp 212-214 ^{o}C; ^{1}H NMR (300 MHz, CDCl3) δ 7.25 (s, 1H), 7.10-7.02 (m, 6H), 2.29 (s, 6H), 2.24 (s, 6H), 2.12 (s, 6H); ^{13}C NMR (75 MHz, CDCl3) δ 161.7, 152.5, 138.8, 136.4, 135.5, 132.1, 130.8, 130.0, 127.4, 108.4, 20.7, 17.6, 13.0; IR (KBr): 3460, 2955, 1610, 1531, 1446, 1321, 1137, 1002, 805, 668 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{25}H_{26}N_4O_2$: 414.2056. Found: 414.2057.

(*Z*)-2-(2-Chlorophenyl)-4-((1-(2-chlorophenyl)-5-hydroxy-3-methyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4i)



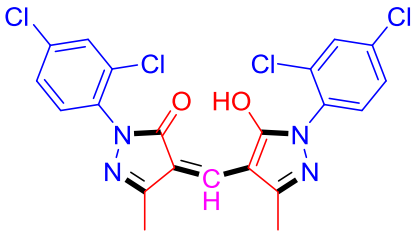
Yellow solid (160 mg, 75%): mp 208-210 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.51-7.48 (m, 2H), 7.43-7.40 (m, 2H), 7.36-7.34 (m, 5H), 2.37 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 162.2, 152.8, 139.0, 134.4, 131.9, 130.4, 129.1, 127.4, 108.3, 13.0; IR (KBr): 3452, 2925, 1626, 1384, 1326, 1100, 1002, 818, 726 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{16}Cl_2N_4O_2$: 426.0650. Found: 426.0652.

(*Z*)-4-((5-Hydroxy-3-methyl-1-(4-nitrophenyl)-1*H*-pyrazol-4-yl)methylene)-5-methyl-2-(4-nitrophenyl)-2,4-dihydro-3*H*-pyrazol-3-one (4j)[s4]



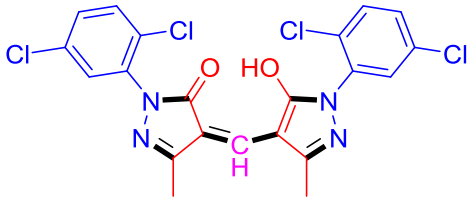
Yellow solid (134 mg, 60%): mp >300 °C; ^{1}H NMR (600 MHz, DMSO-d_6) δ 13.23 (s, 1H), 8.26 (d, *J* = 9.6 Hz, 2H), 8.23 (d, *J* = 9.0 Hz, 2H), 8.18 (d, *J* = 9.0 Hz, 2H), 7.94 (s, 1H), 7.78 (d, *J* = 9.0 Hz, 2H), 2.12 (s, 6H); IR (KBr): 3424, 2927, 2256, 1616, 1511, 1323, 1167, 1009, 821, 596 cm^{-1}.

(*Z*)-2-(2,4-Dichlorophenyl)-4-((1-(2,4-dichlorophenyl)-5-hydroxy-3-methyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4k)



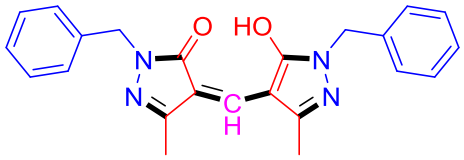
Yellow solid (180 mg, 73%): mp 196-198 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.51 (d, *J* = 1.5 Hz, 2H), 7.37-7.31 (m, 5H), 2.35 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 162.3, 153.1, 139.1, 135.7, 133.1, 132.9, 130.3, 129.8, 127.8, 108.4, 12.9; IR (KBr): 3457, 1621, 1374, 1089, 825 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{14}Cl_4N_4O_2$: 493.9871. Found: 493.9869.

(*Z*)-2-(2,5-Dichlorophenyl)-4-((1-(2,5-dichlorophenyl)-5-hydroxy-3-methyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4l)



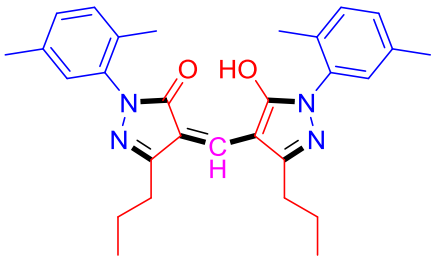
Yellow solid (173 mg, 70%): mp 182-184 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$+DMSO-d_6) δ 7.37-7.26 (m, 3H), 7.15-7.06 (m, 4H), 2.25 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$+DMSO-d_6) δ 161.7, 152.9, 138.7, 130.0, 129.9, 127.4, 124.1, 124.0, 116.4, 116.2, 108.0, 12.5; IR (KBr): 3426, 2256, 1632, 1416, 1005, 753, 644 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{14}Cl_4N_4O_2$: 493.9871. Found: 493.9868.

(*Z*)-2-Benzyl-4-((1-benzyl-5-hydroxy-3-methyl-1*H*-pyrazol-4-yl)methylene)-5-methyl-2,4-dihydro-3*H*-pyrazol-3-one (4m)



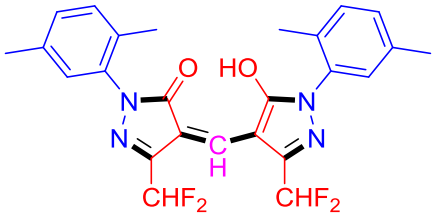
Yellow solid (149 mg, 77%): mp 154-156 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.43-7.36 (m, 10H), 7.35 (s, 1H), 5.11 (s, 4H), 2.33 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 161.3, 151.7, 138.2, 135.9, 128.5, 127.9, 127.6, 108.0, 49.2, 12.6; IR (KBr): 3456, 2935, 1617, 1528, 1445, 1323, 1113, 693 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{23}H_{22}N_4O_2$: 386.1743. Found: 386.1743.

(*Z*)-2-(2,5-Dimethylphenyl)-4-((1-(2,5-dimethylphenyl)-5-hydroxy-3-propyl-1*H*-pyrazol-4-yl)methylene)-5-propyl-2,4-dihydro-3*H*-pyrazol-3-one (4n)



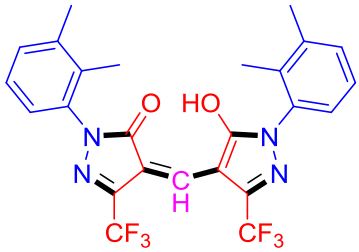
Yellow liquid (169 mg, 72%): ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.40 (s, 1H), 7.17-7.08 (m, 6H), 2.68 (t, *J* = 7.5 Hz, 4H), 2.31 (s, 6H), 2.18 (s, 6H), 1.77 (q, *J* = 7.5 Hz, 4H), 1.05 (t, *J* = 7.5 Hz, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 161.8, 155.9, 138.3, 136.4, 135.5, 132.1, 130.8, 130.0, 127.4, 107.6, 29.1, 22.0, 20.7, 17.6, 14.0; IR (neat): 3549, 2943, 1613, 1366, 1098, 819 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{29}H_{34}N_4O_2$: 470.2682. Found: 470.2682.

(*Z*)-5-(Difluoromethyl)-4-((3-(difluoromethyl)-1-(2,5-dimethylphenyl)-5-hydroxy-1*H*-pyrazol-4-yl)methylene)-2-(2,5-dimethylphenyl)-2,4-dihydro-3*H*-pyrazol-3-one (4o)



Yellow solid (177 mg, 73%): mp 168 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.29 (s, 1H), 7.21-7.14 (m, 4H), 7.10 (s, 2H), 6.59 (t, $J_{H\text{-}F}$ = 53.1 Hz, 2H), 2.33 (s, 6H), 2.17 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 162.3, 147.7 (t, $J_{C\text{-}F}$ = 29.1 Hz), 141.2, 136.7, 134.7, 132.1, 131.0, 130.7, 127.3, 115.3, 112.1, 109.0, 105.9, 20.7, 17.4; IR (KBr): 3597, 2932, 1623, 1524, 1383, 1042, 835, 739 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{25}H_{22}F_4N_4O_2$: 486.1679. Found: 486. 1681.

(*Z*)-2-(2,3-Dimethylphenyl)-4-((1-(2,3-dimethylphenyl)-5-hydroxy-3-(trifluoromethyl)-1*H*-pyrazol-4-yl)methylene)-5-(trifluoromethyl)-2,4-dihydro-3*H*-pyrazol-3-one (4p)



Yellow solid (191 mg, 73%): mp 191-193 °C; ^{1}H NMR (300 MHz, $CDCl_3$+DMSO-d_6) δ 7.30-7.19 (m, 6H), 5.84 (s, 2H), 2.37 (s, 6H), 2.04 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$+DMSO-d_6) δ 153.4, 140.6 (q, $J_{C\text{-}F}$ = 37.4 Hz), 137.5, 135.7, 133.8, 130.1, 125.1, 124.7, 122.5, 119.0, 83.9, 19.4, 13.4; IR (KBr): 3411, 2255, 1654, 1492, 1245, 1002, 859, 710 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{25}H_{20}F_6N_4O_2$: 522.1490. Found: 522.1488.

IV. Fluorescence measurements

Chloride salts of Al^{3+}, Ca^{2+}, Co^{2+}, Cr^{3+}, Cu^{2+}, Fe^{3+}, Hg^{2+}, Ni^{2+}, lead nitrate (Pb^{2+}), and cadmium acetate (Cd^{2+}) were purchased from Sigma-Aldrich and used as received. For fluorescence analysis, dipyrazolylmethane derivatives **4a, 4e, 4j, 4k**, and **4p** (1.50 mL, 1.0 x 10^{-4} M in MeCN) were transferred to cuvettes and the corresponding emission spectrum were measured at an excitation wavelength of 330 nm. The selectivity of compound **4j** was tested against ten different metal cations, such as Al^{3+}, Ca^{2+}, Cd^{2+}, Co^{2+}, Cr^{3+}, Cu^{2+}, Fe^{3+}, Hg^{2+}, Ni^{2+}, and Pb^{2+} ions (25 μM in MeCN). Furthermore, the sensitivity of compound **4j** towards different concentrations of Cu^{2+} ions was examined. The fluorescence measurements were carried out using a Hitachi Fluorescence Spectrophotometer F-7000 with a 5 nm slit width and a scan speed at 240 nm/min.

The quenching efficiency of Cu^{2+} ions was calculated using the Stern-Volmer relation (1) and the limit of detection (LOD) of dipyrazolylmethane **4j** was computed using the formula (2).

$$F_0/F = 1 + K_{sv}[Q] \qquad (1)$$

where F_0 and F are the fluorescence intensities of dipyrazolylmethane **4j** in the absence and presence of Cu^{2+} ions, respectively, K_{sv} is the Stern–Volmer quenching constant and [Q] is the concentration of Cu^{2+} ions.

$$\text{LOD} = \frac{3S}{b} \qquad (2)$$

where S is the standard deviation of F_0 (5 results) and *b* is the slope of the linear line (Fig. S1). [s5]

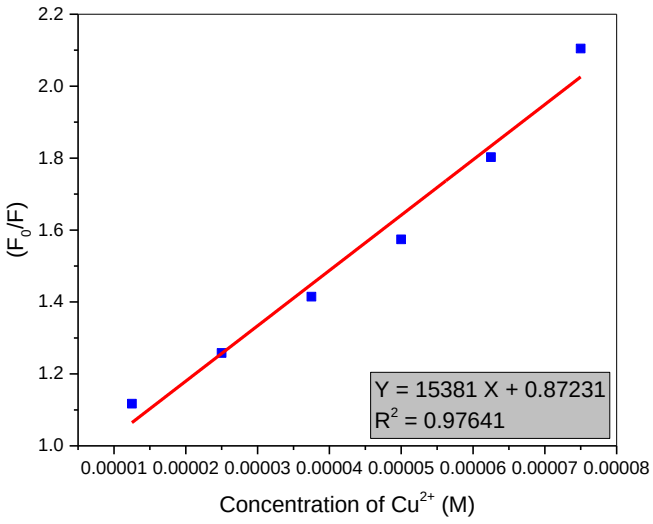


Fig. S1 Stern–Volmer plot of fluorescence intensity ratios (F_0/F) versus Cu^{2+} ions concentrations.

V. References

s1 F. F. Wong and Y.-Y. Huang, *Tetrahedron*, 2011, **67**, 3863-3867.

s2 Y. Ma, J. Wang and H. Ma, *Heterocycles*, 2014, **89**, 1645-1655.

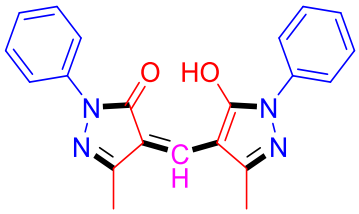
s3 CCDC 1474418 for **4f** contains the supplementary crystallographic data for this paper. The data can be obtained free of charge from The Cambridge Crystallographic Data Centre *via* www.ccdc.cam.ac.uk/data_request/cif.

s4 (a) Y. A. Azev, S. V. Shorshnev and D. Gabel, *Pharm. Chem. J.*, 2002, **36**, 146-150; (b) A. Kreutzberger and K. Kolter, *J. Heterocycl. Chem.*, 1986, **23**, 781-784; (c) W. Häensel, *Justus Liebigs Ann. Chem.*, 1976, 1680-1688.

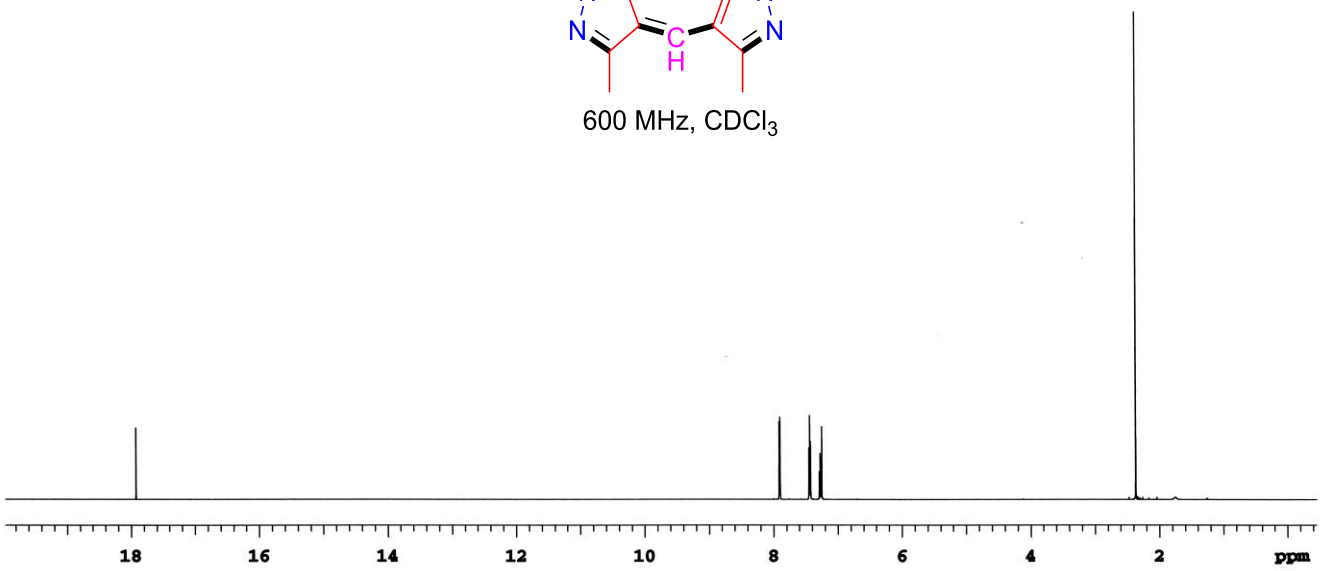
s5 T. N. J. I. Edison, R. Atchudan, J.-J. Shim, S. Kalimuthu, B.-C. Ahn, Y. R. Lee, J. Photochem. Photobiol. B, 2016, **158**, 235-242.

VI. NMR Spectra

^{1}H NMR of Compound **4a**



600 MHz, $CDCl_3$



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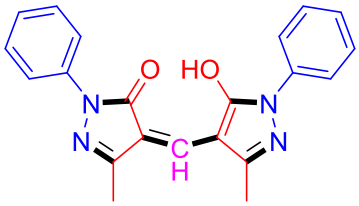
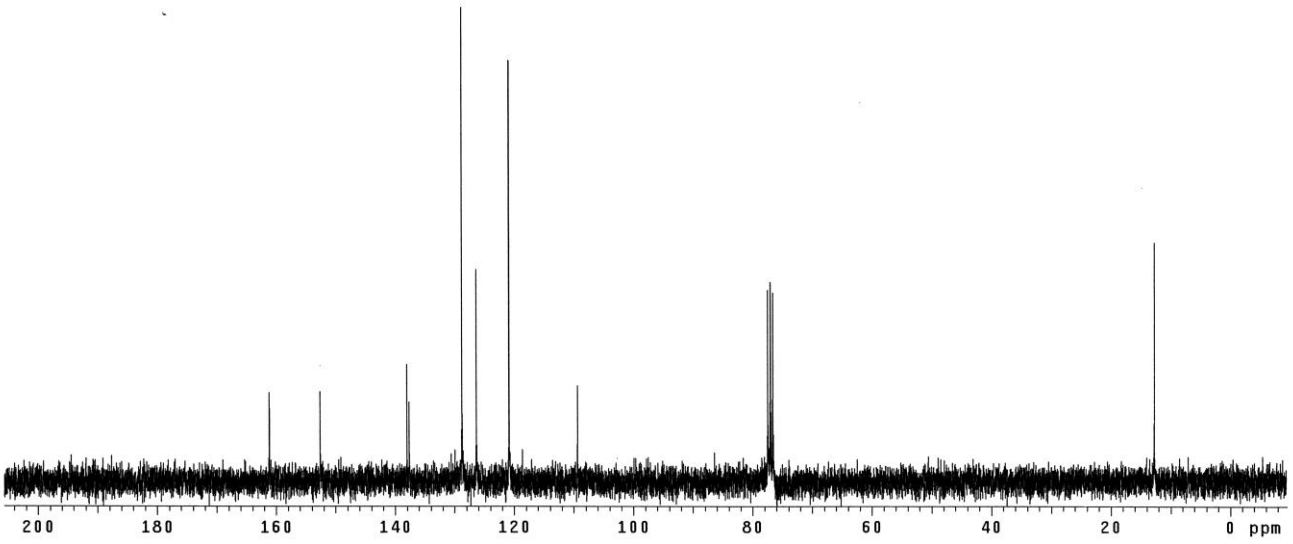
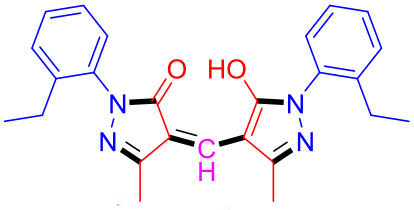


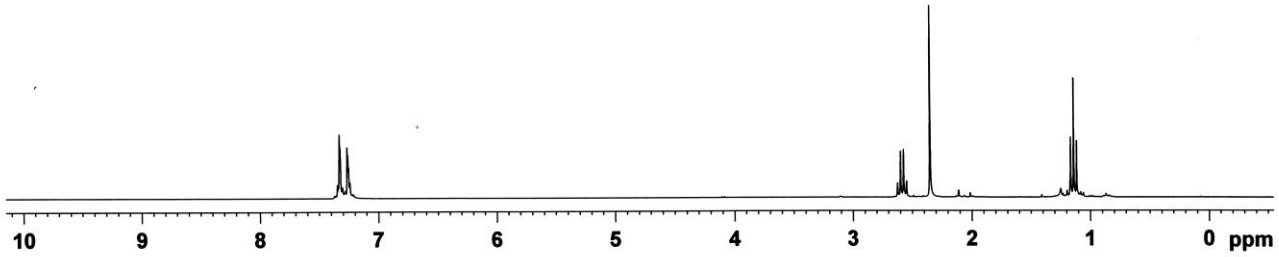
75 MHz, $CDCl_3$



^{1}H NMR of Compound **4b**



300 MHz, $CDCl_3$



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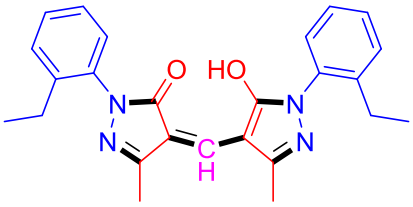

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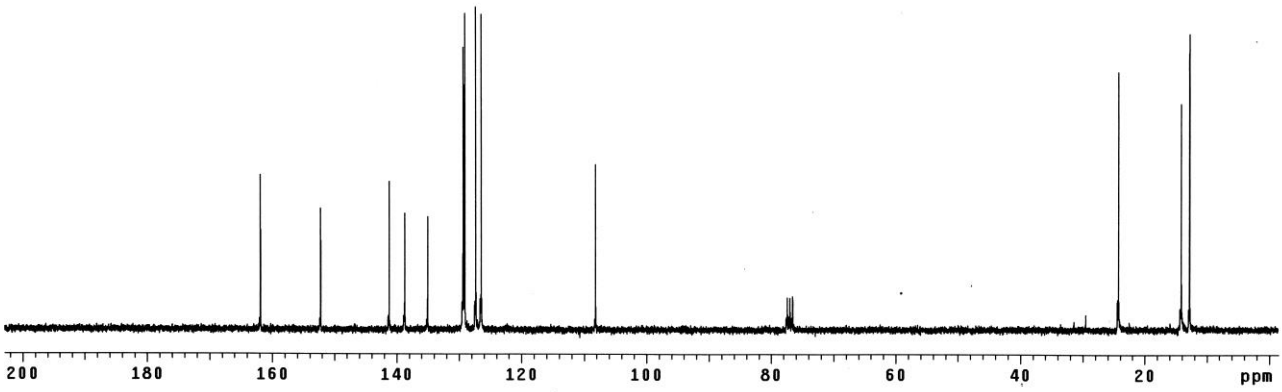
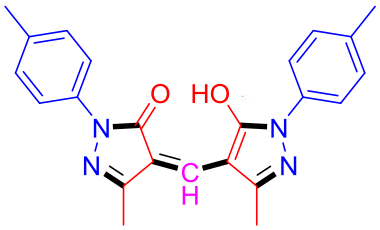


^{1}H NMR of Compound **4c**



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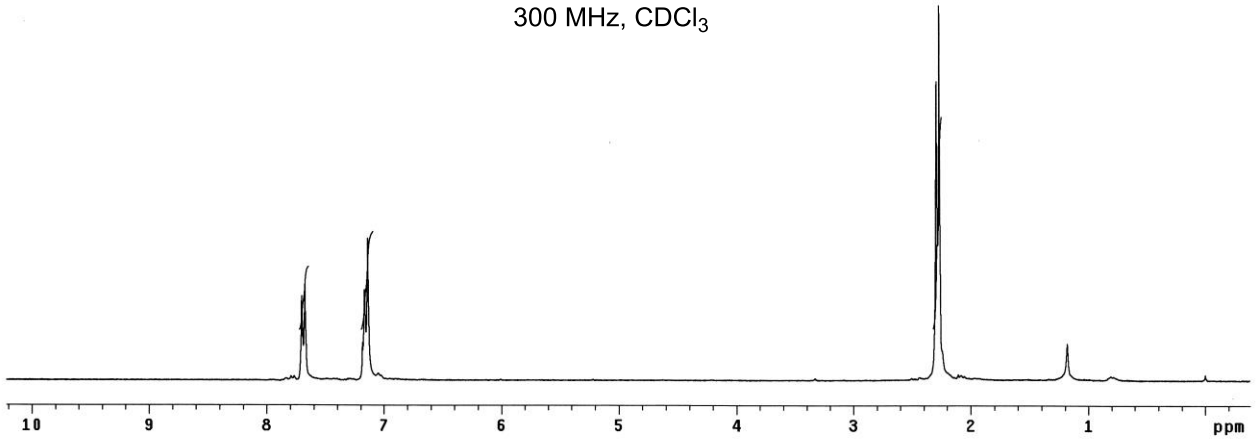
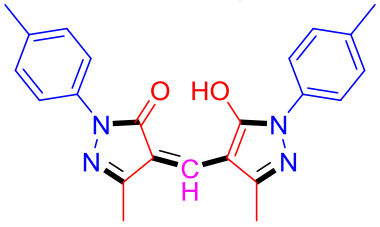
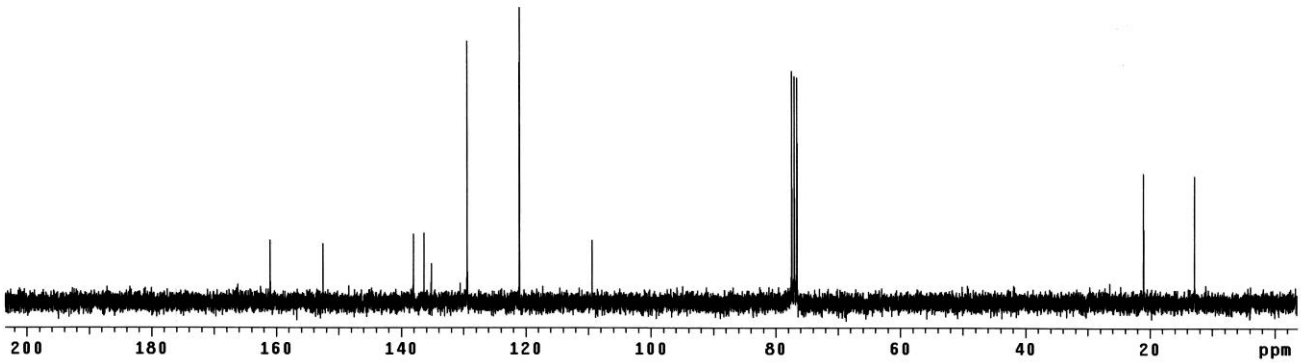


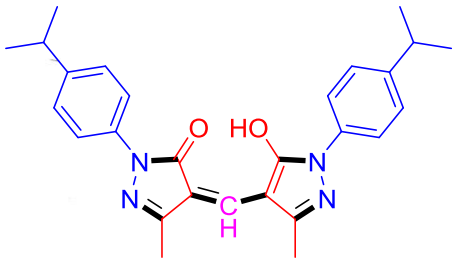
^{13}C NMR of Compound **4c**



75 MHz, $CDCl_3$



^{1}H NMR of Compound **4d**



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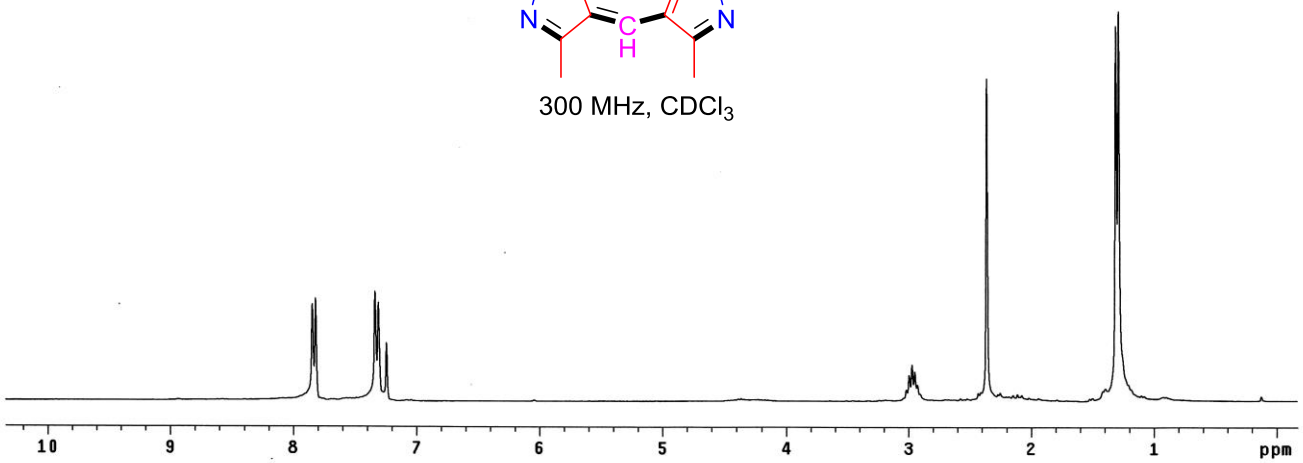

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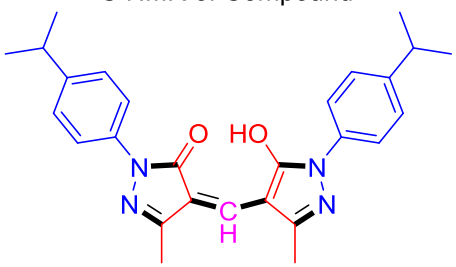

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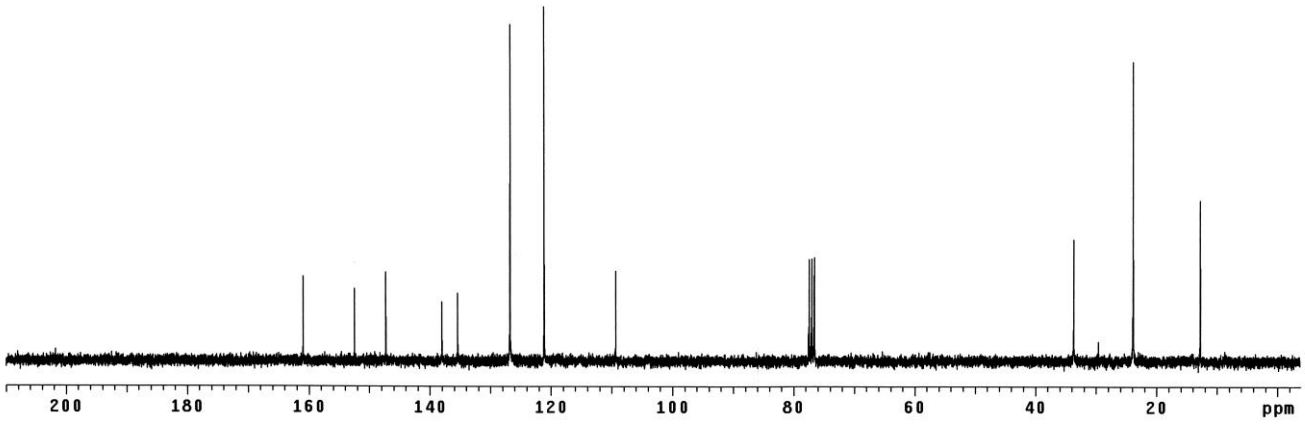
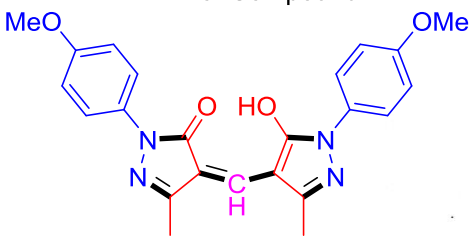


^{1}H NMR of Compound **4e**



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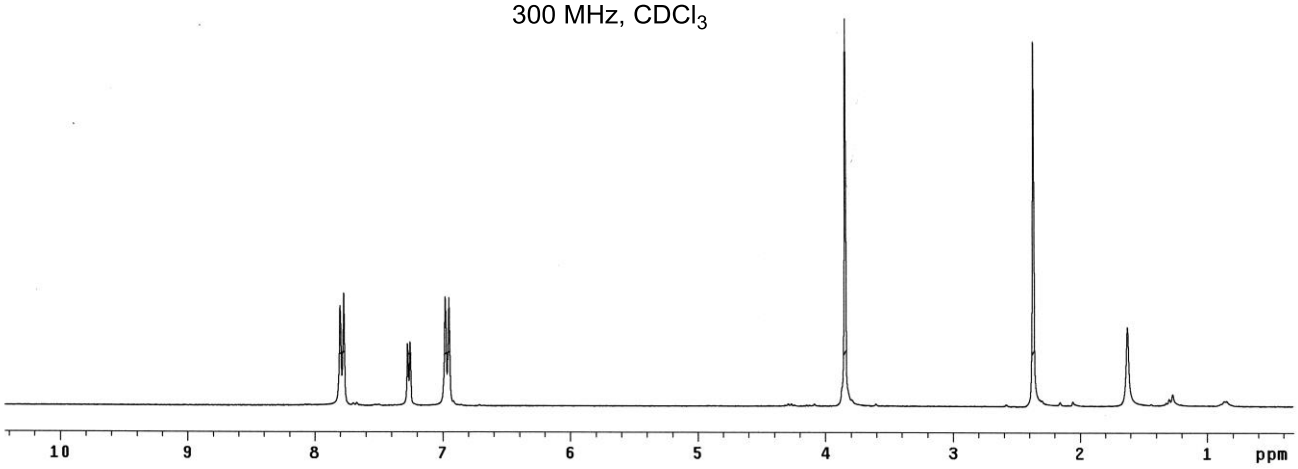

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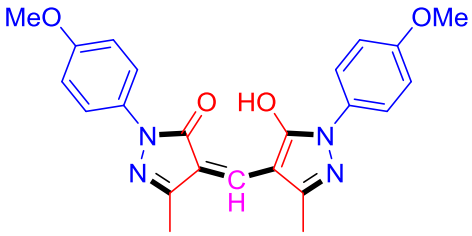

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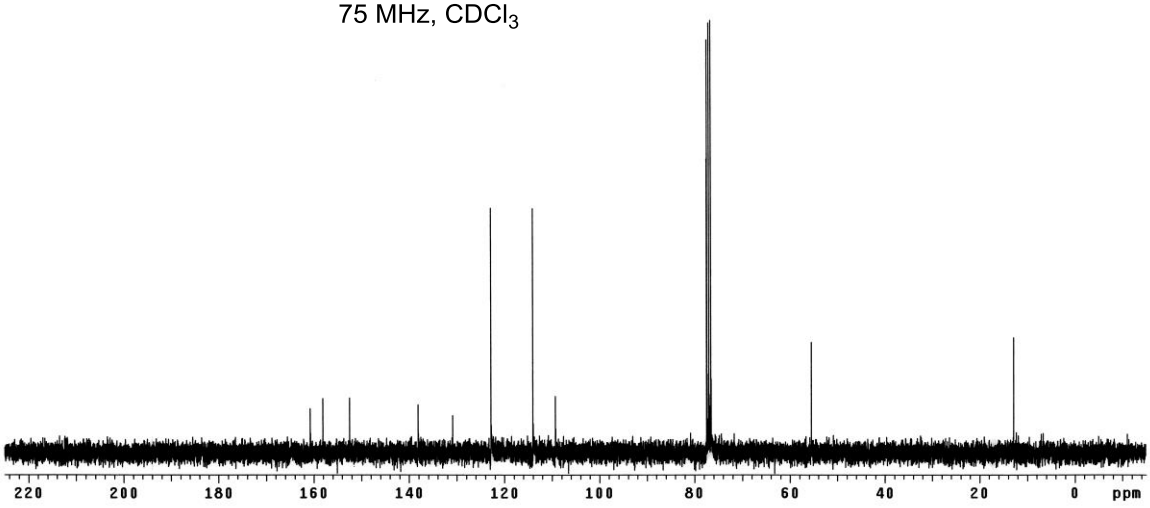
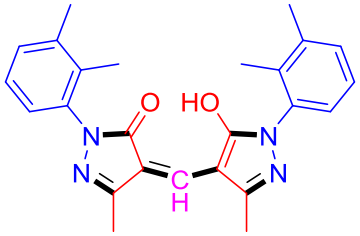


^{1}H NMR of Compound **4f**



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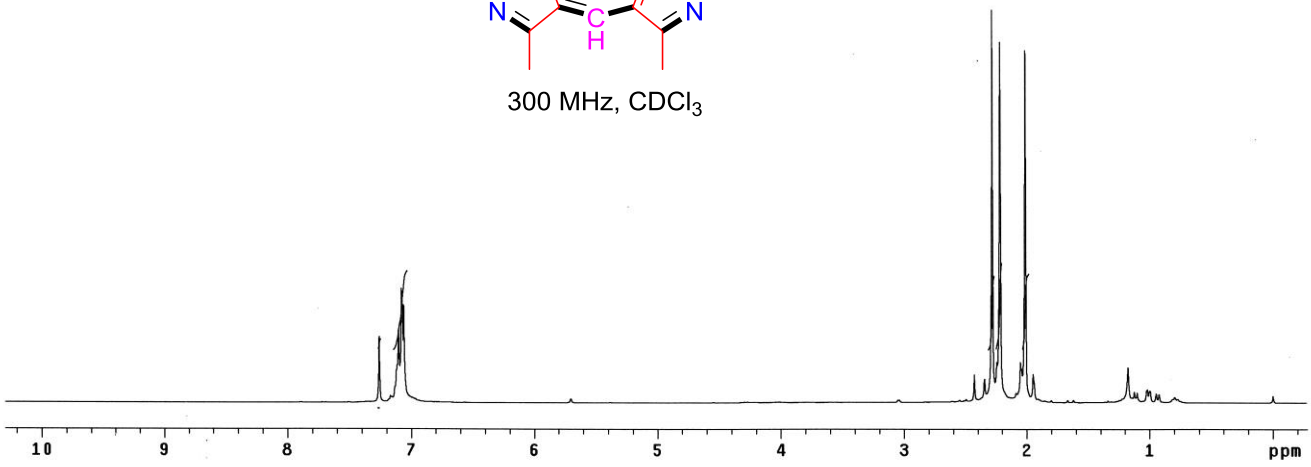

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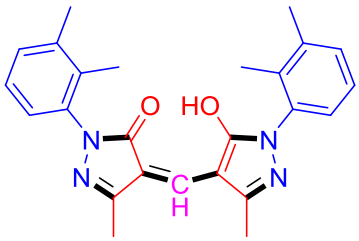

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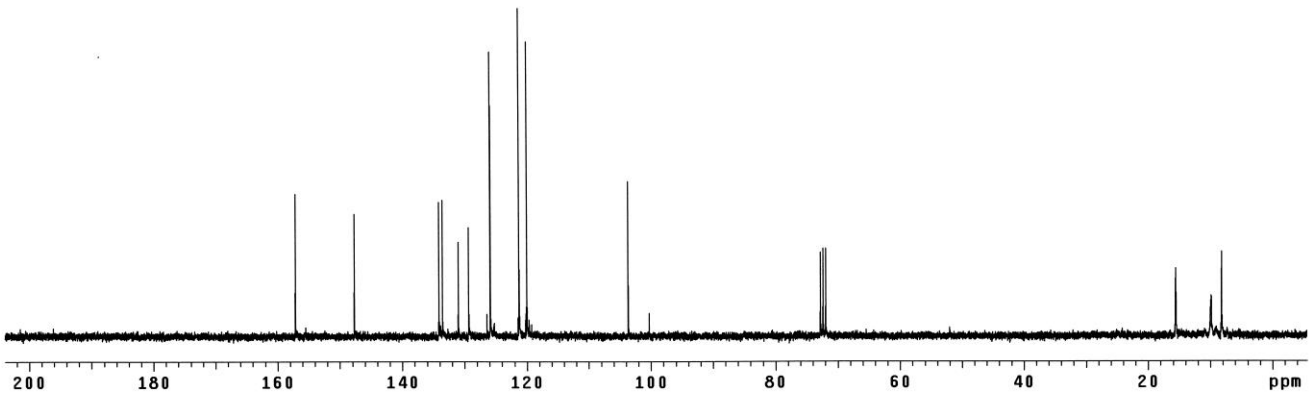
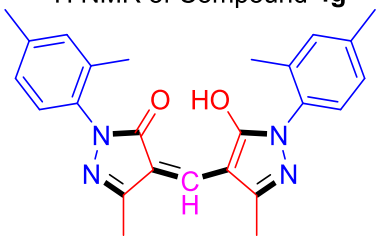
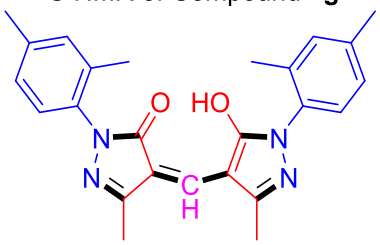


^{1}H NMR of Compound **4g**

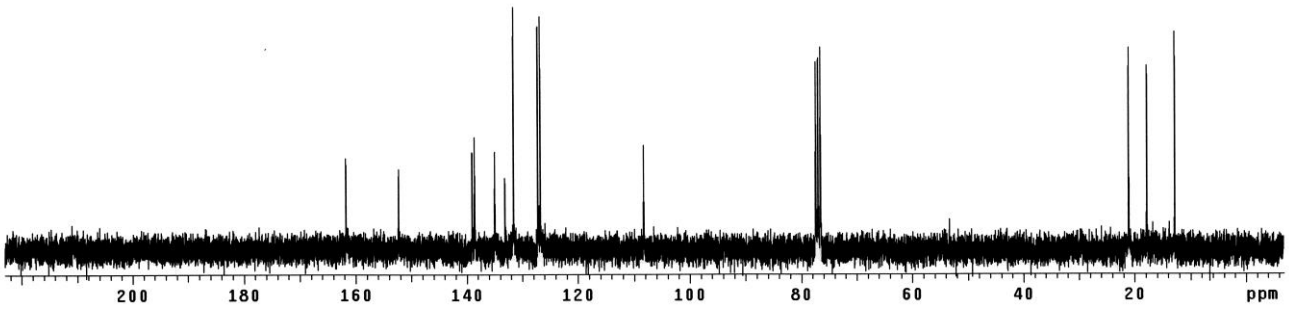


600 MHz, $CDCl_3$

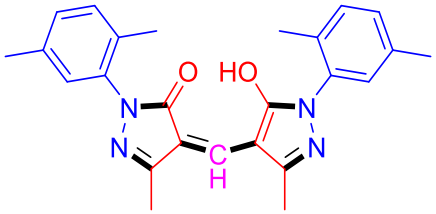
^{13}C NMR of Compound **4g**



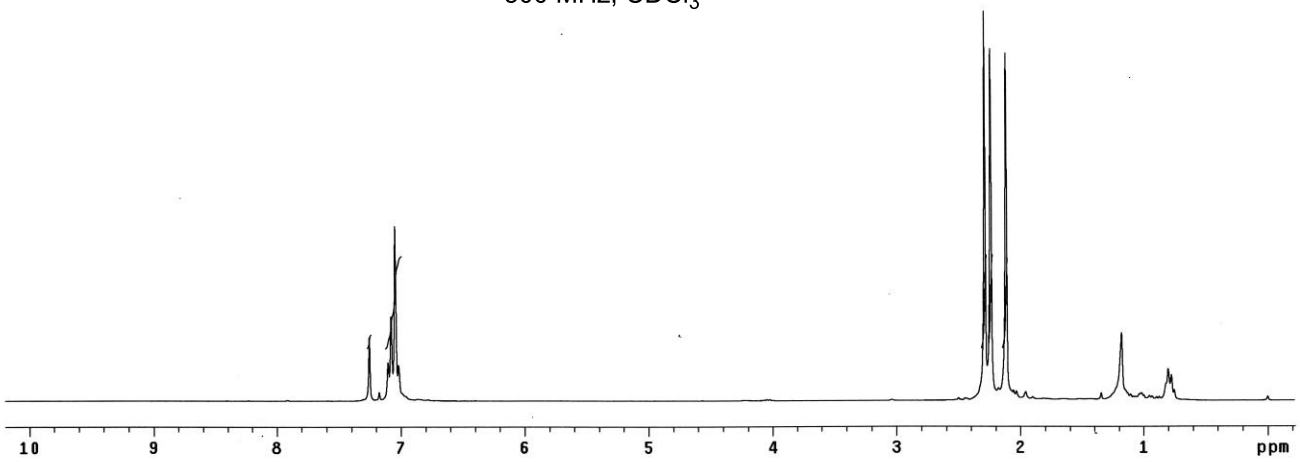
75 MHz, $CDCl_3$



^{1}H NMR of Compound **4h**



300 MHz, $CDCl_3$



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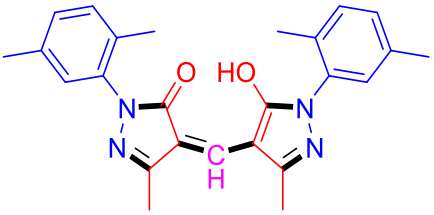

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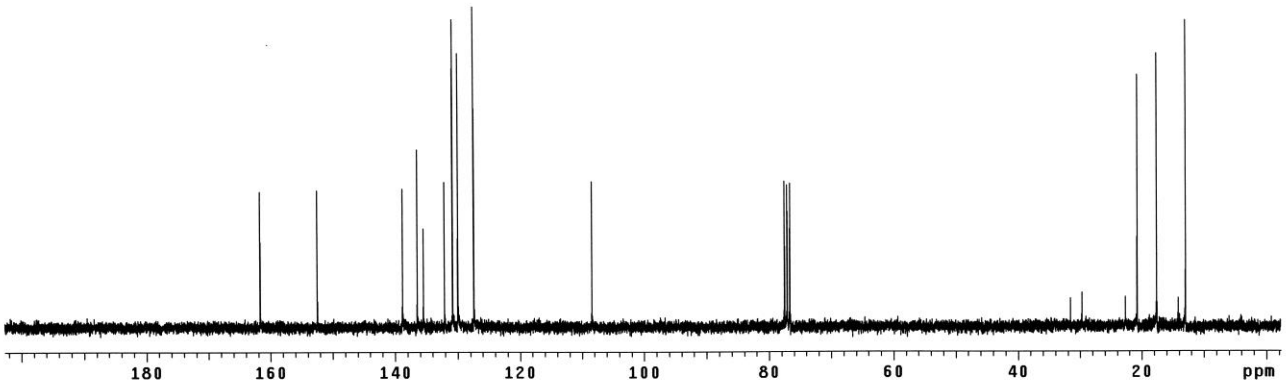
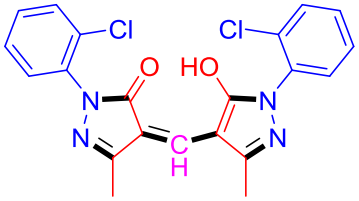


^{1}H NMR of Compound **4i**



300 MHz, $CDCl_3$

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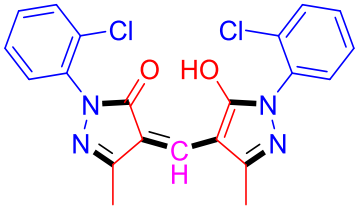

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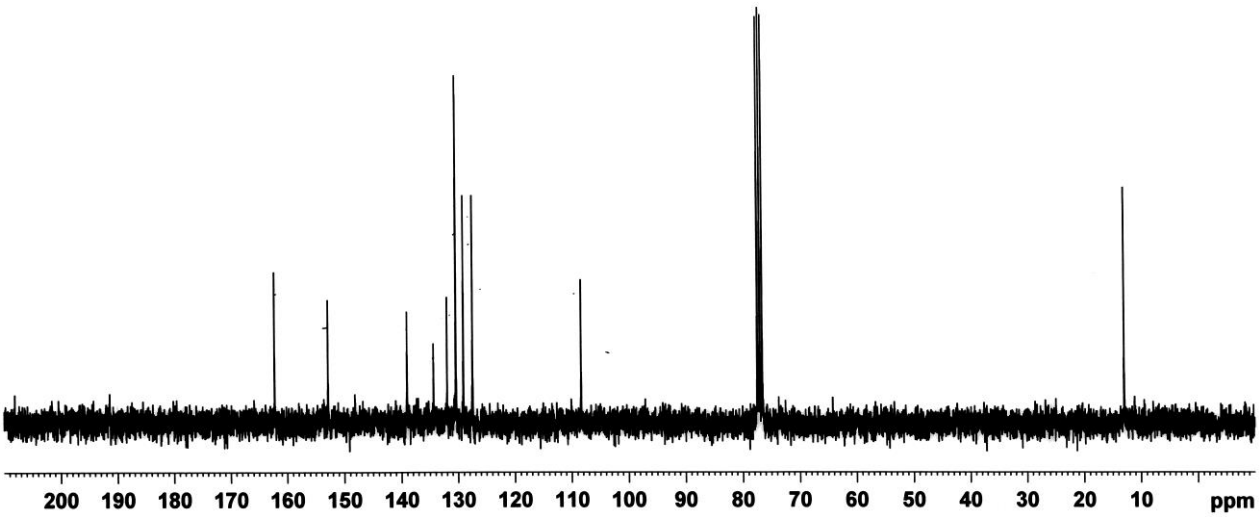

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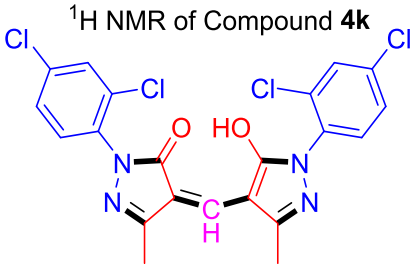
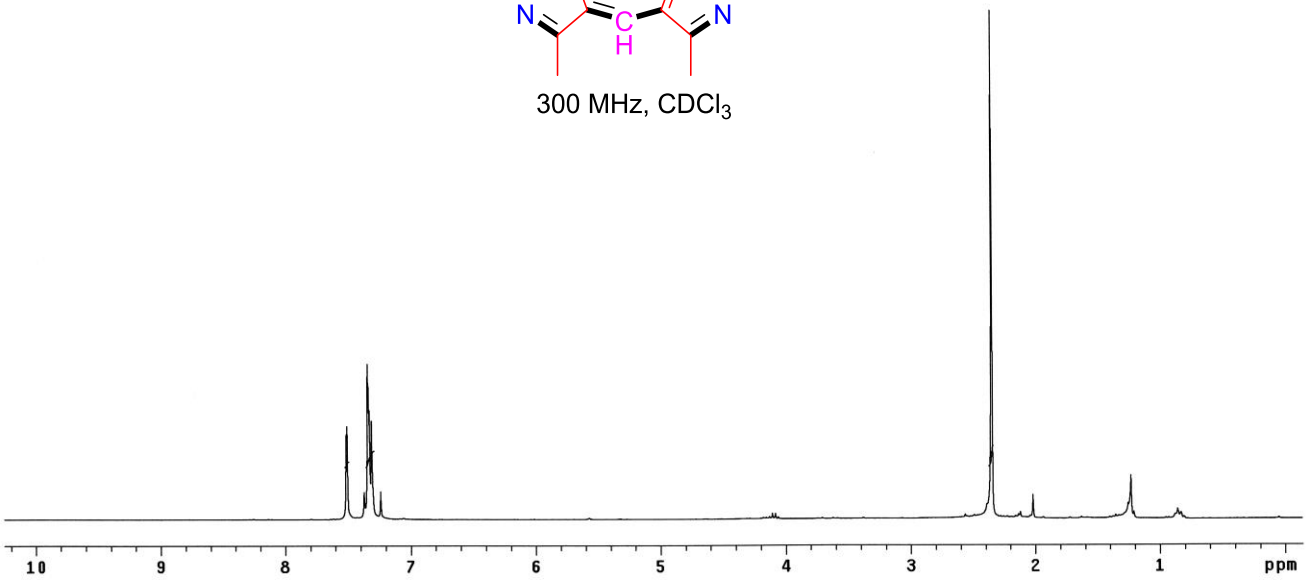


300 MHz, $CDCl_3$



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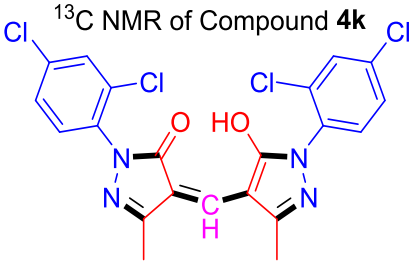

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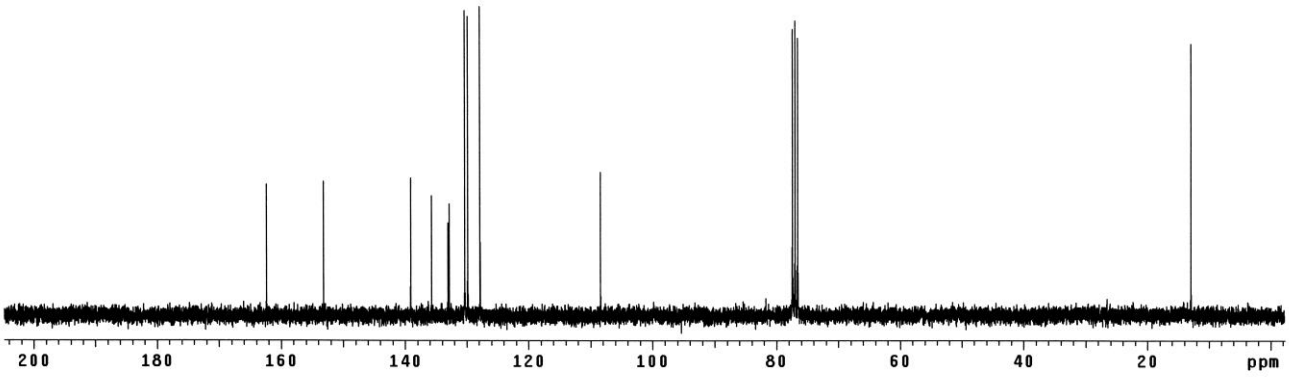

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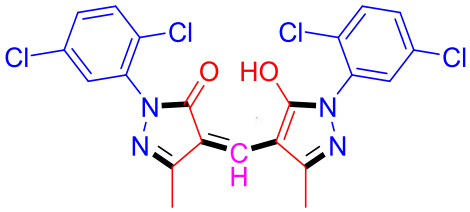
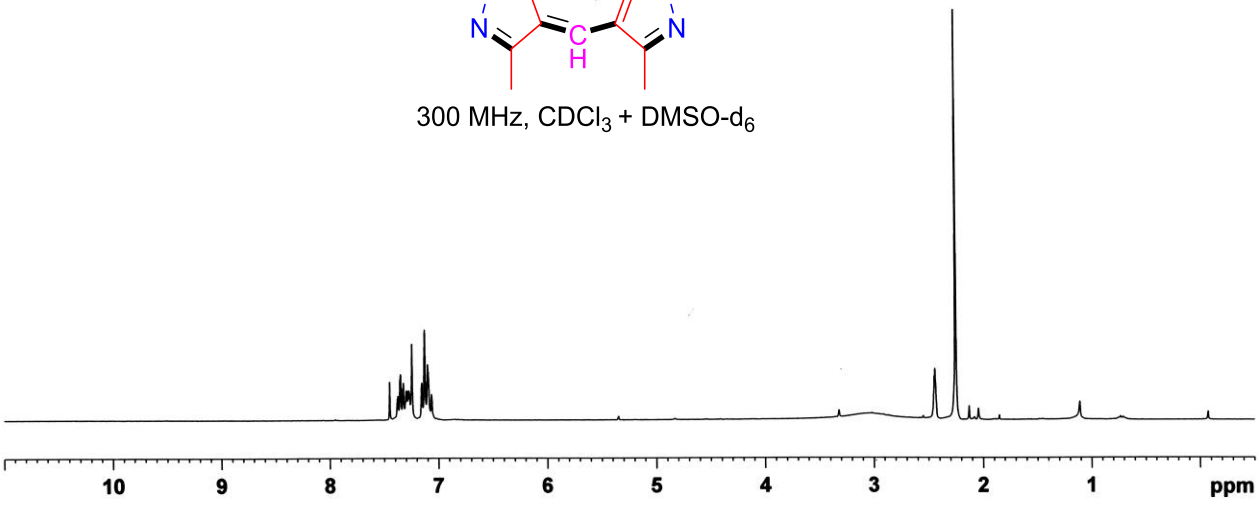


300 MHz, $CDCl_3$ + DMSO-d_6



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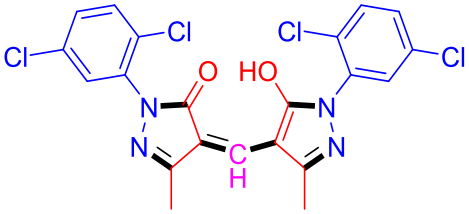
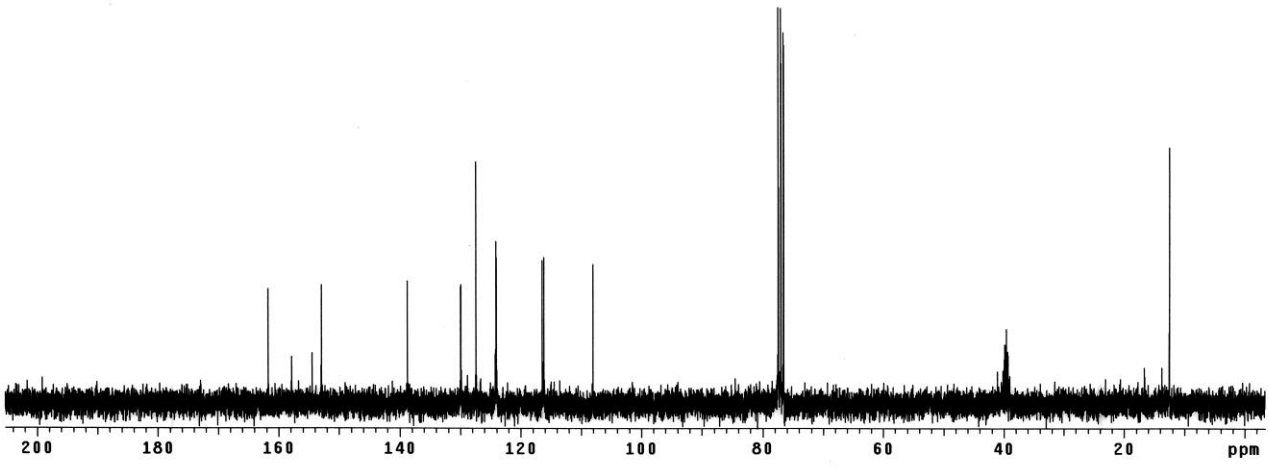
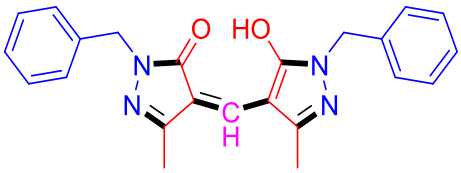


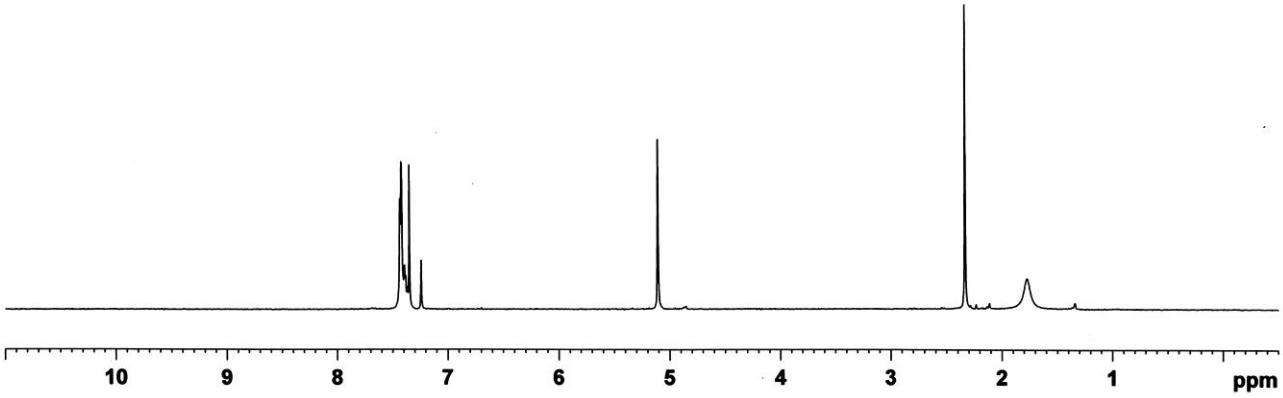
75 MHz, $CDCl_3$ + DMSO-d_6



^{1}H NMR of Compound **4m**



300 MHz, $CDCl_3$



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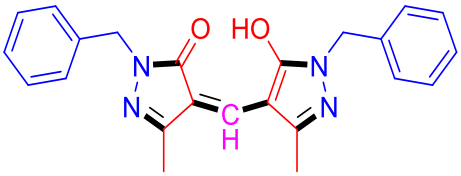
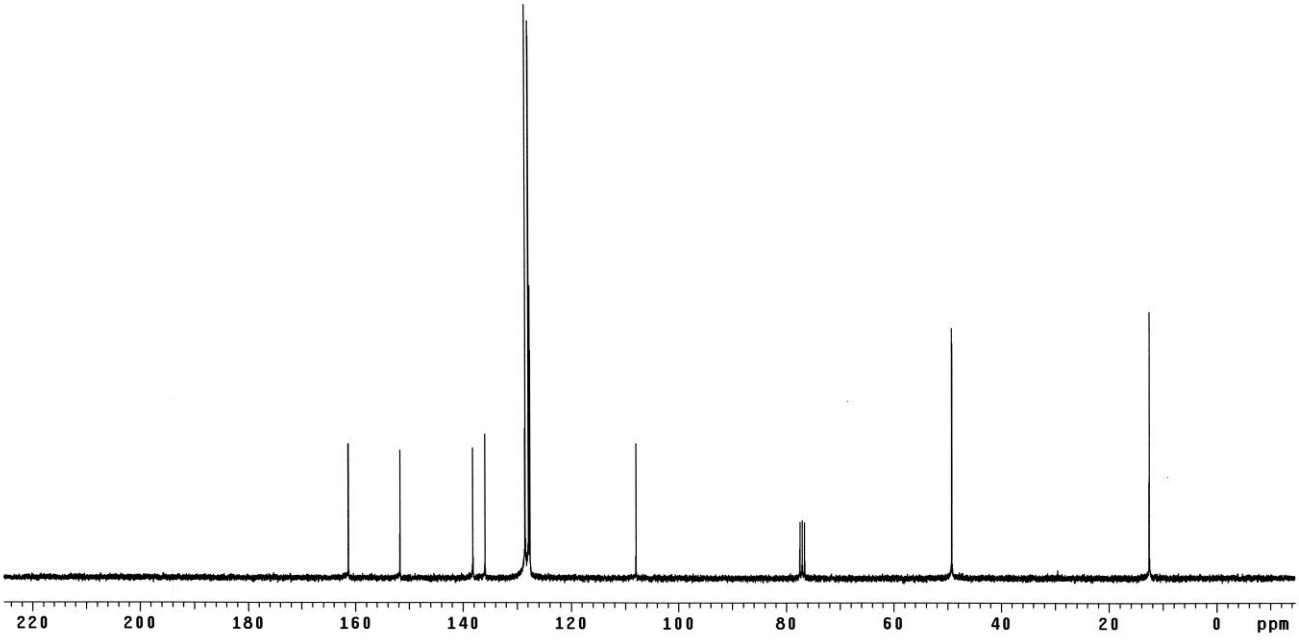
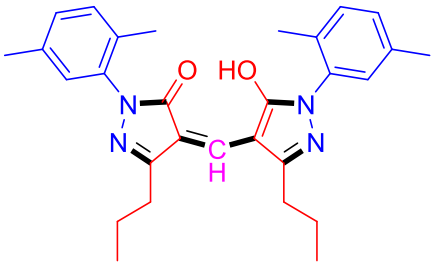


75 MHz, $CDCl_3$



^{1}H NMR of Compound **4n**



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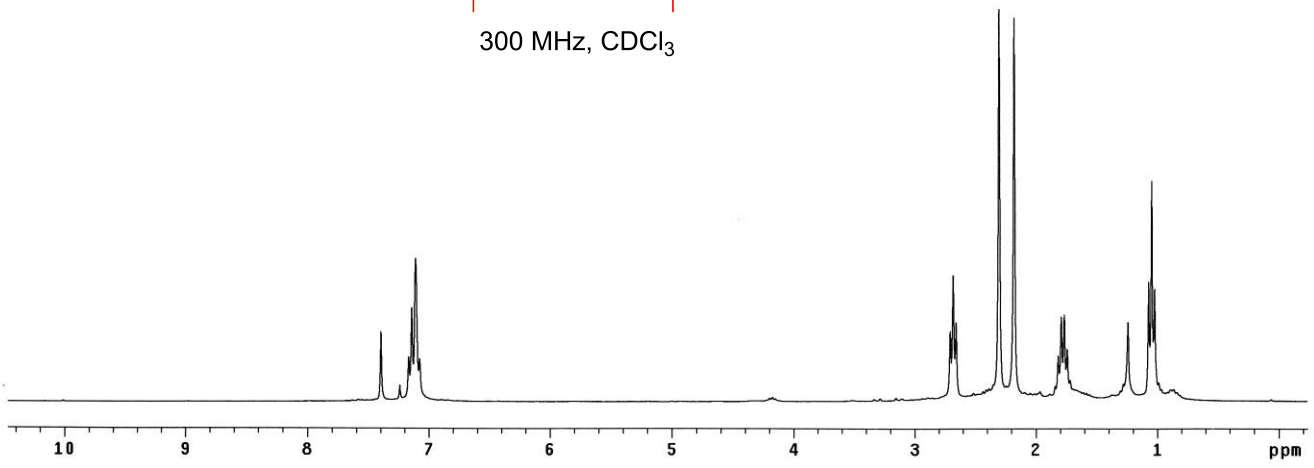

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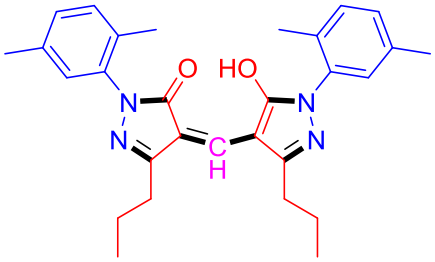

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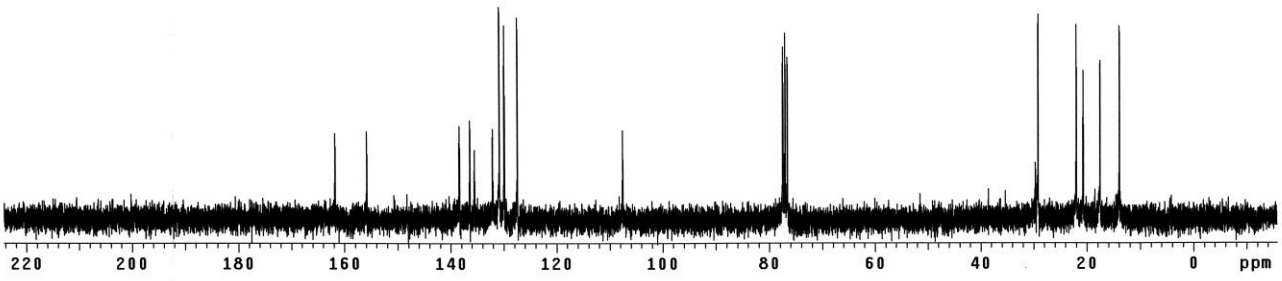
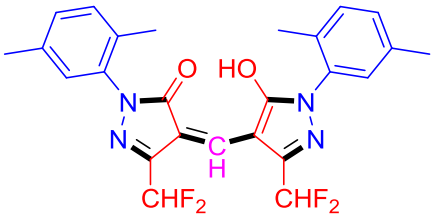


^{1}H NMR of Compound **4o**



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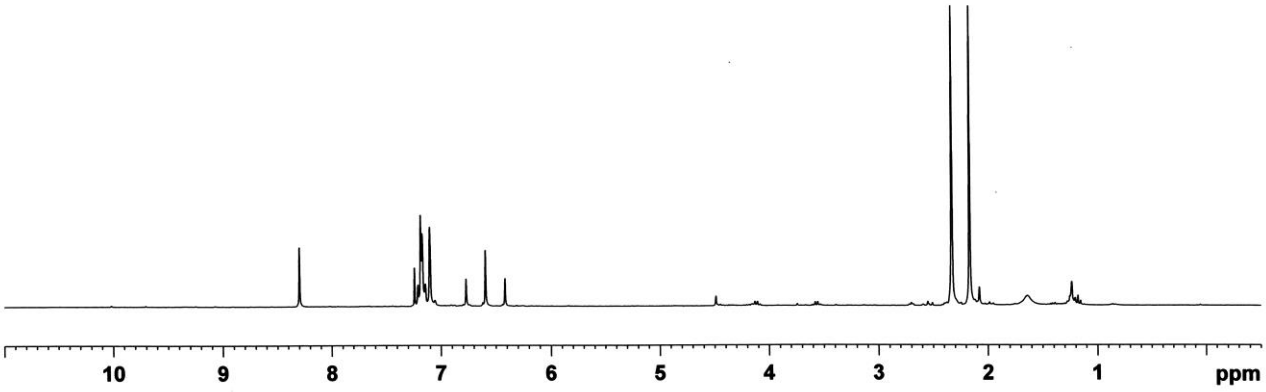
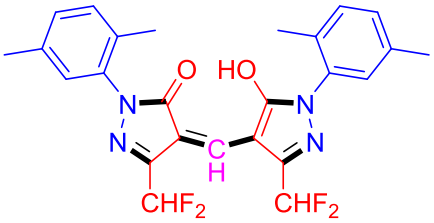
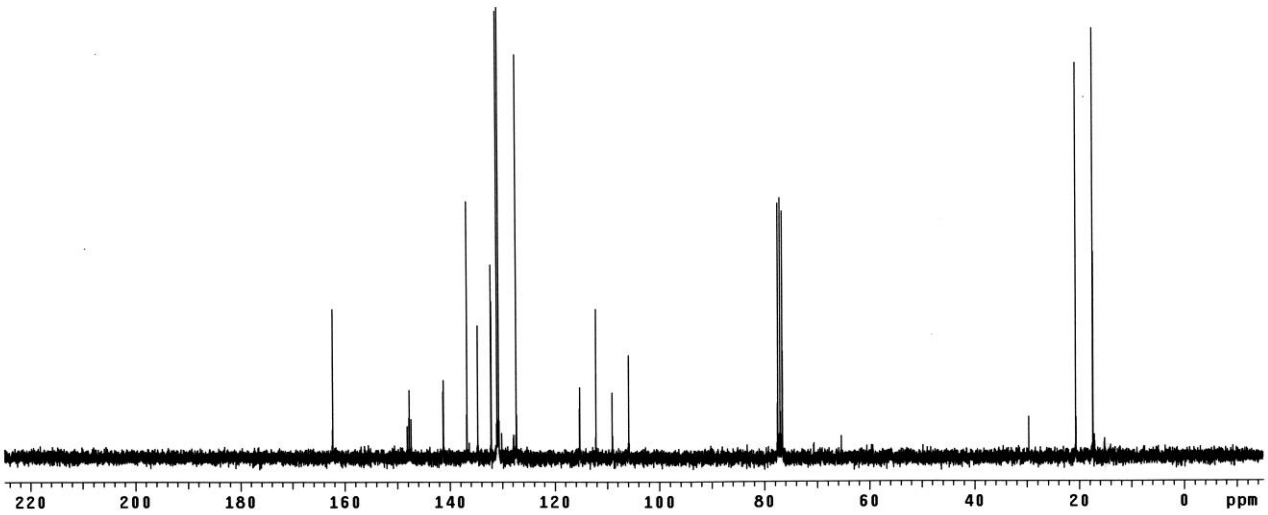


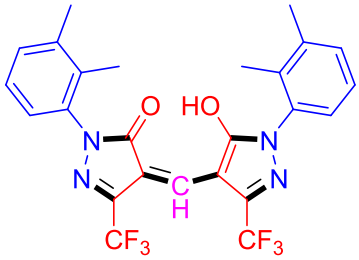
^{13}C NMR of Compound **4o**



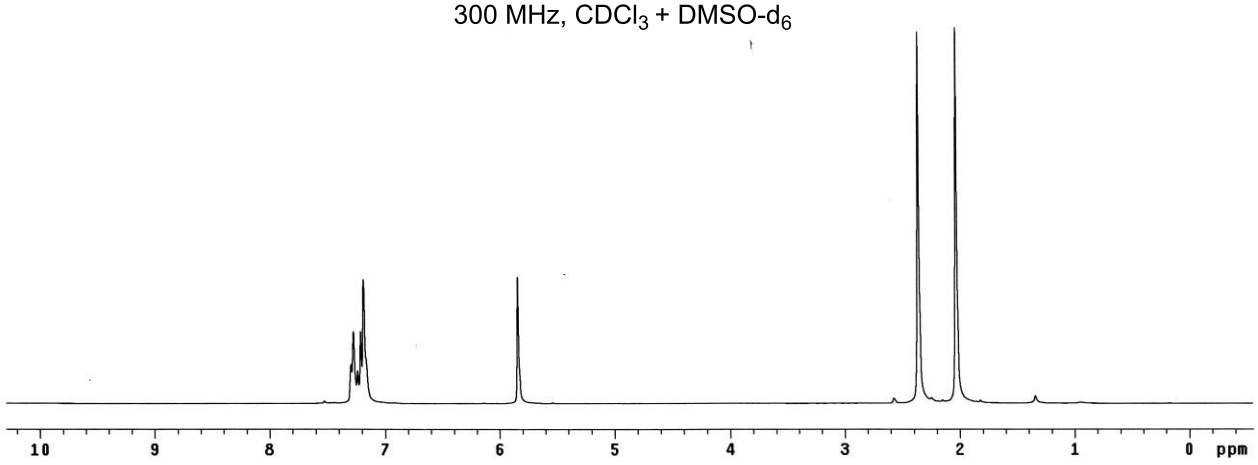
75 MHz, $CDCl_3$



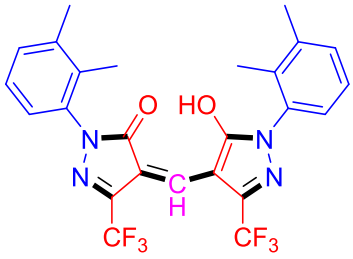
^{1}H NMR of Compound **4p**



300 MHz, $CDCl_3$ + DMSO-d_6



^{13}C NMR of Compound **4p**



75 MHz, $CDCl_3$ + DMSO-d_6