Supporting Information

Direct α-Alkenylation of Cyclic Amines with Maleimides through Fe(III)-Catalyzed C(sp^3)−H/C(sp^2)−H Cross Dehydrogenative Coupling

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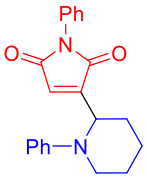
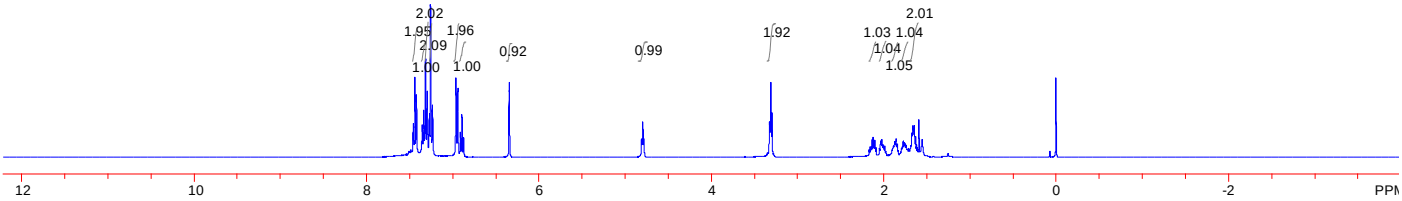
I. Copies of NMR Spectra of 3a-3w

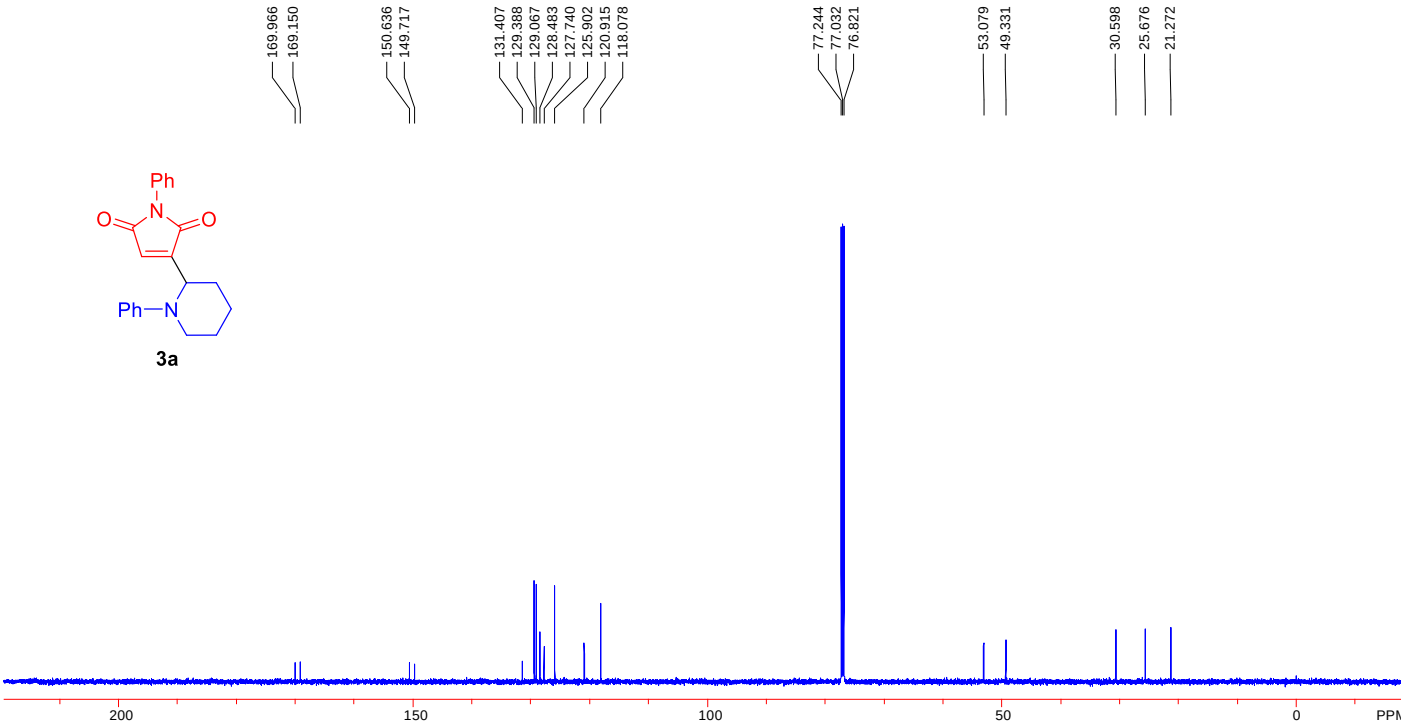


3a

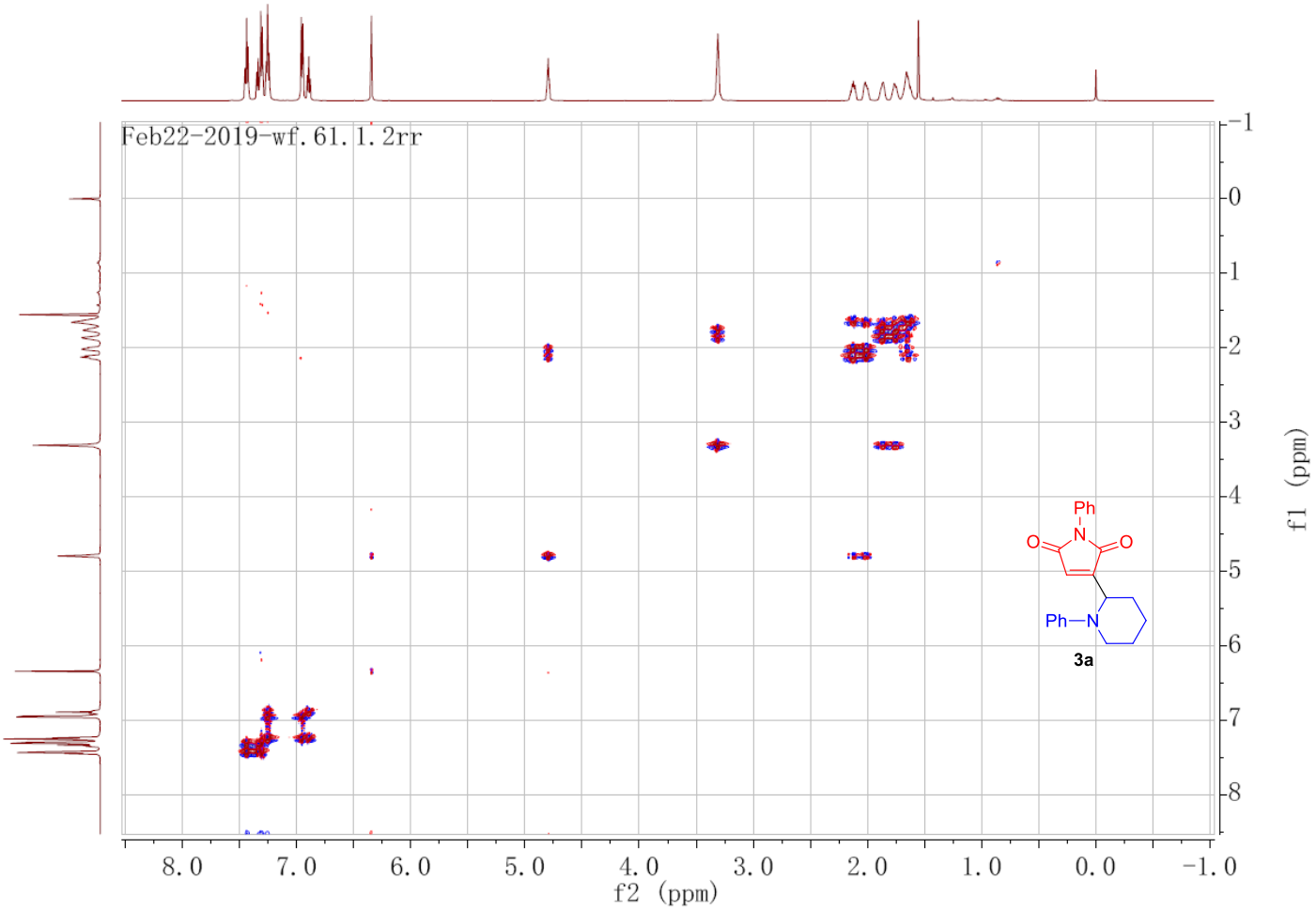


^{1}H NMR (400 MHz, $CDCl_3$) of **3a**

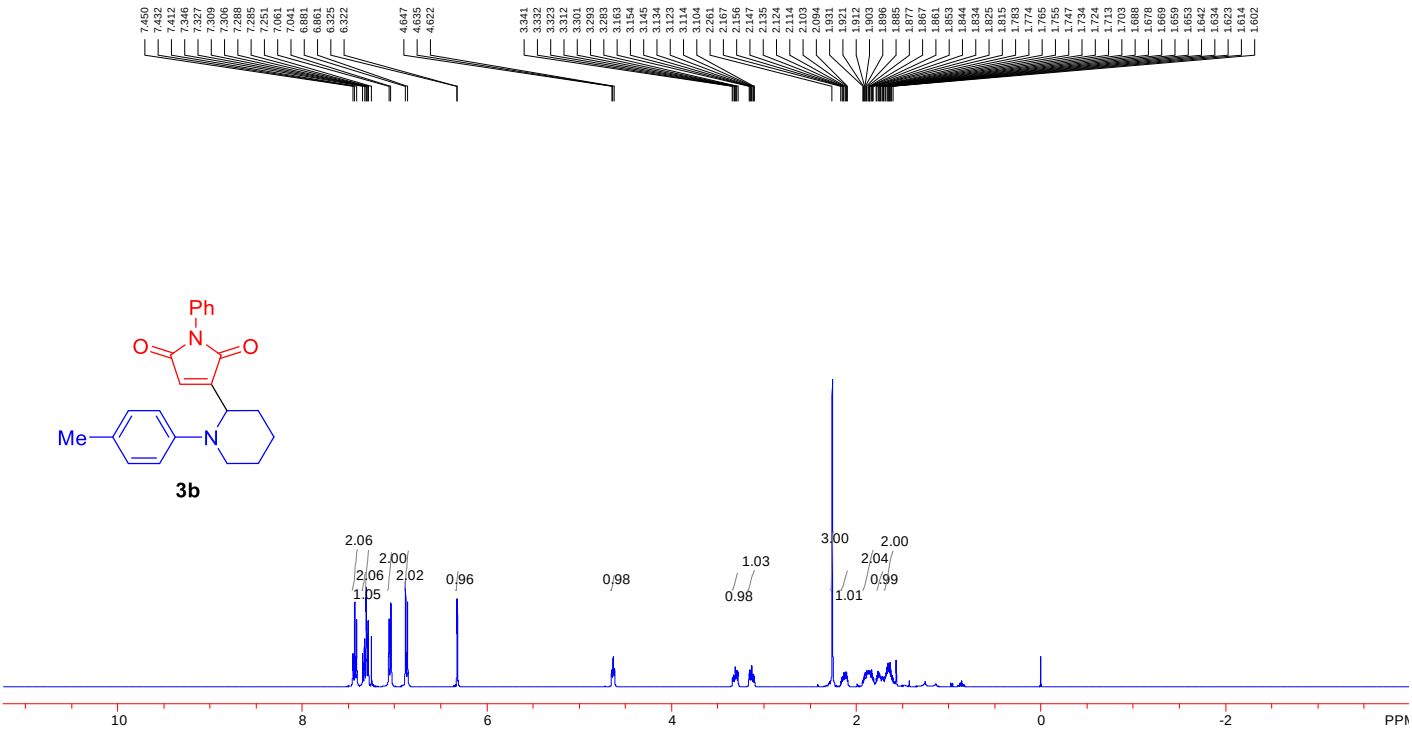
3a



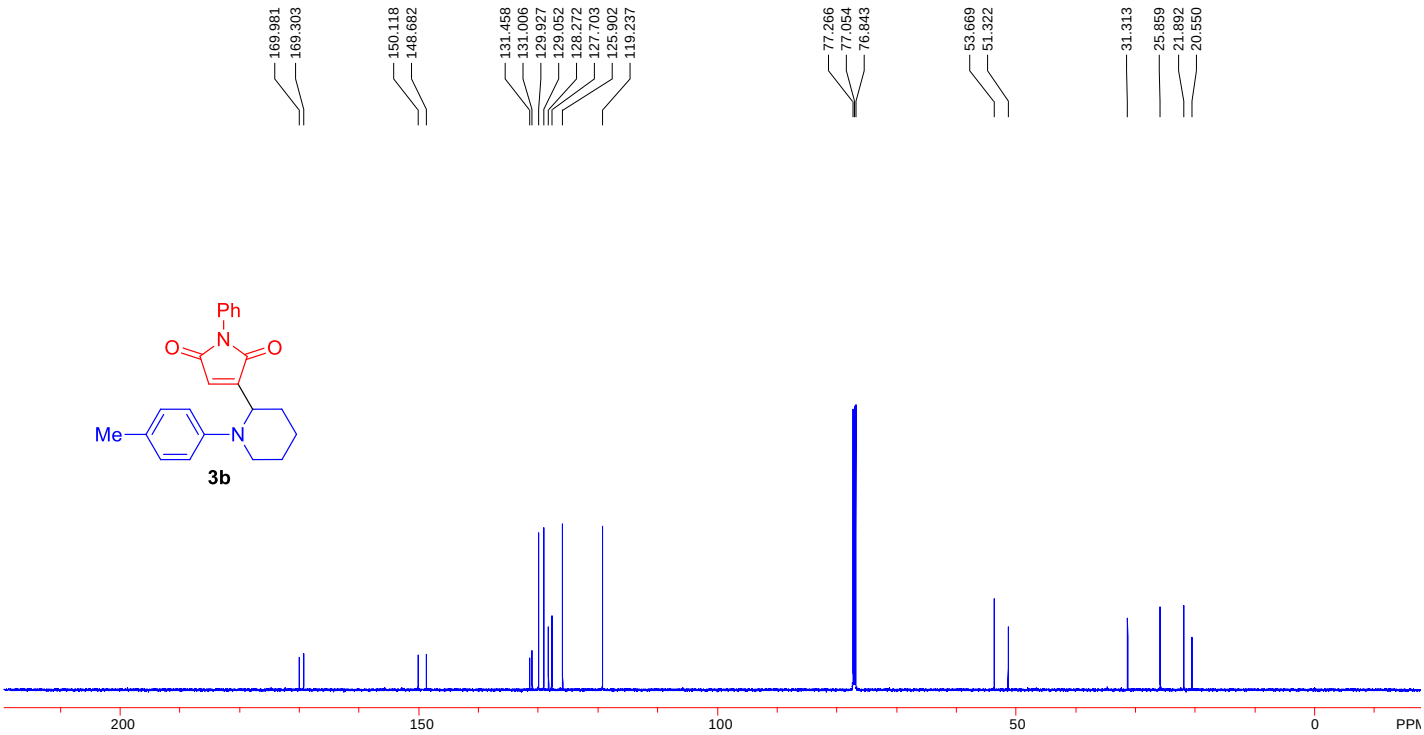
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **3a**



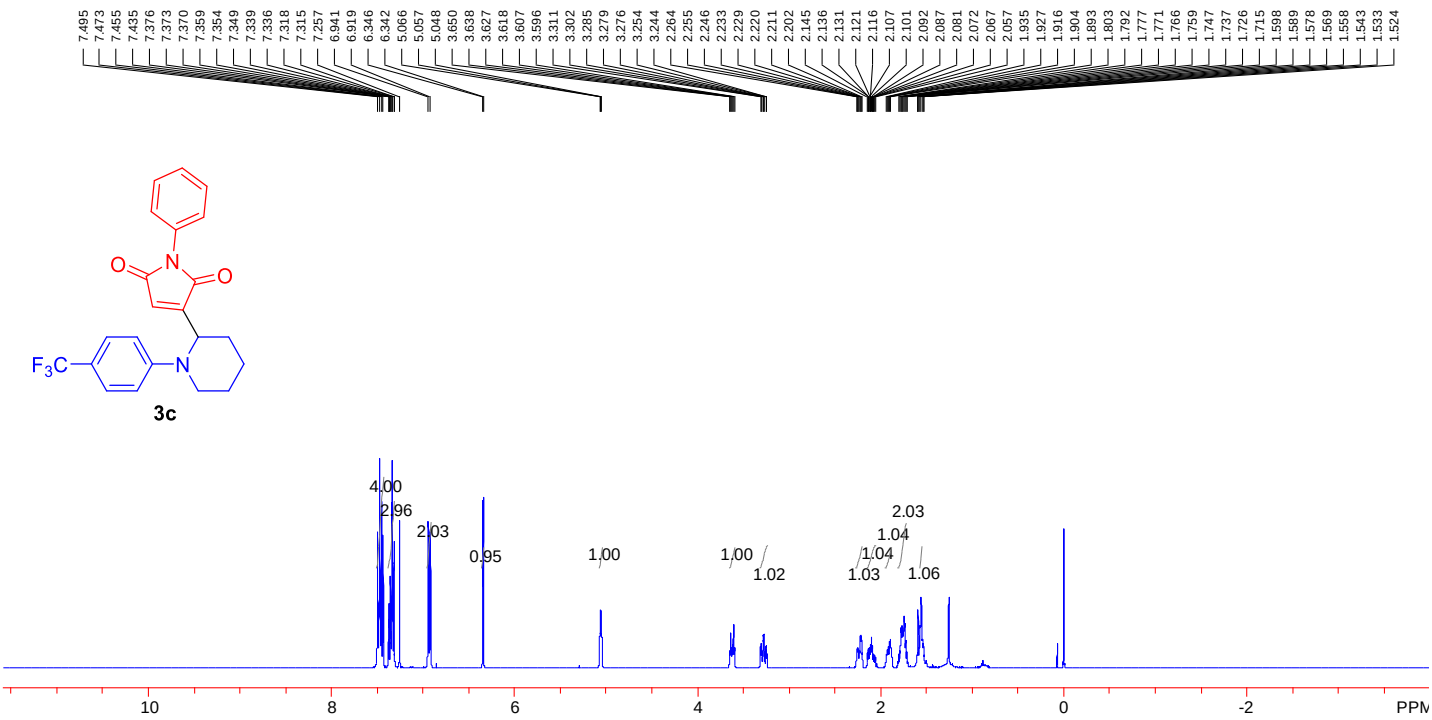
gCOSY NMR (400 MHz, $CDCl_3$) of **3a**



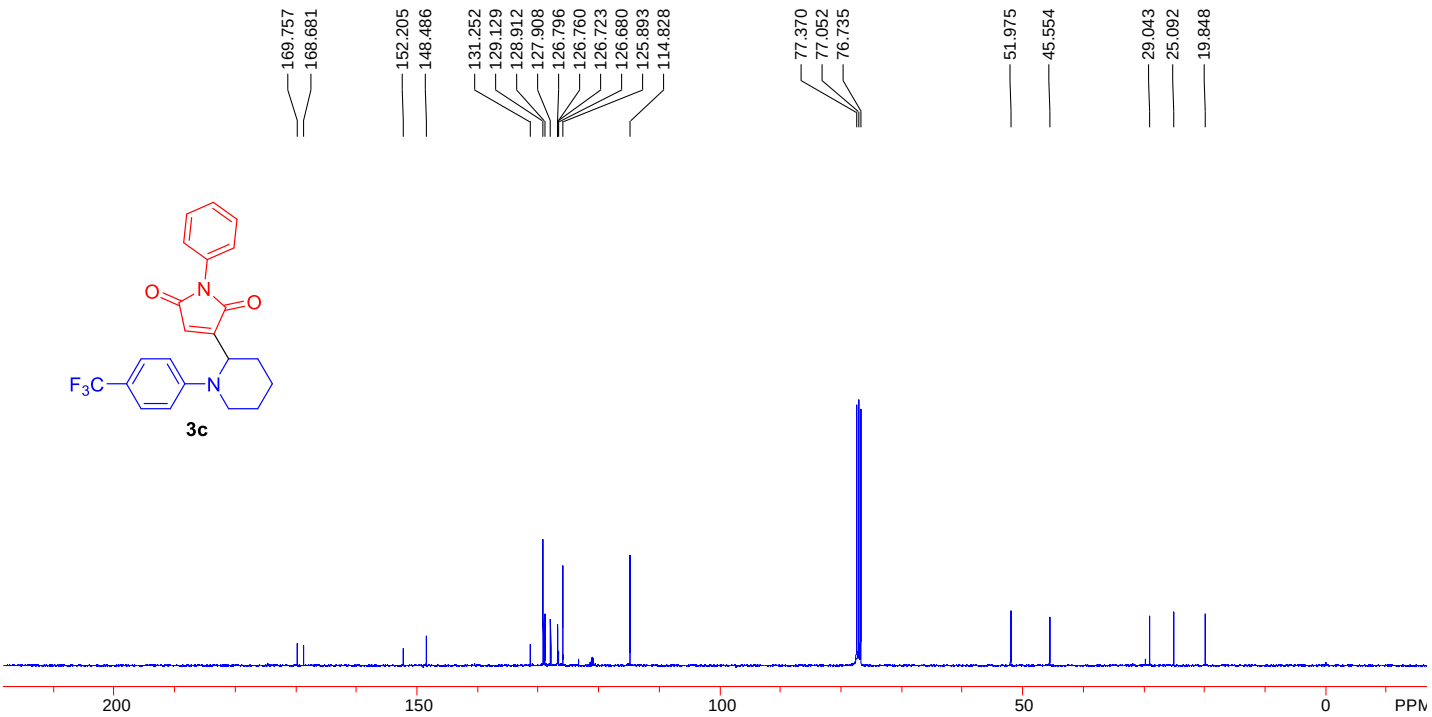
^{1}H NMR (400 MHz, $CDCl_3$) of **3b**



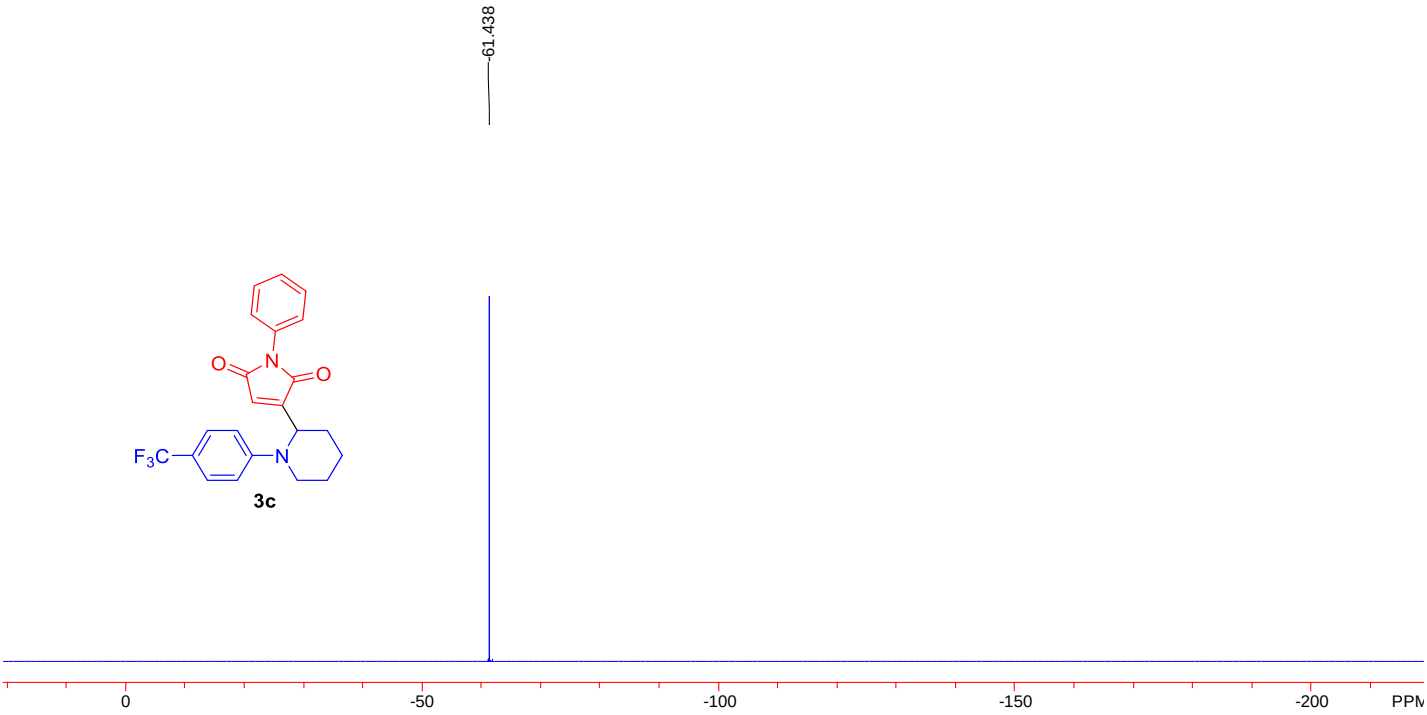
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3b**



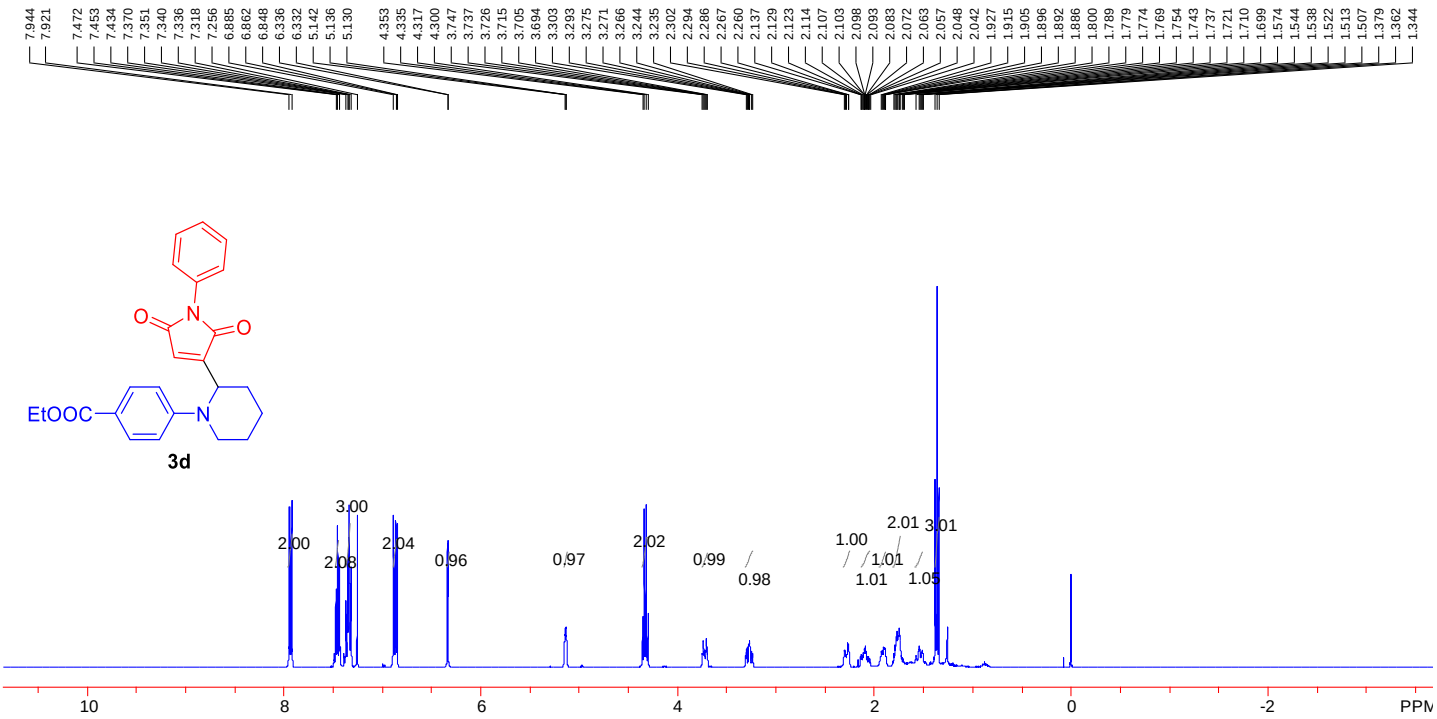
^{1}H NMR (400 MHz, $CDCl_3$) of **3c**



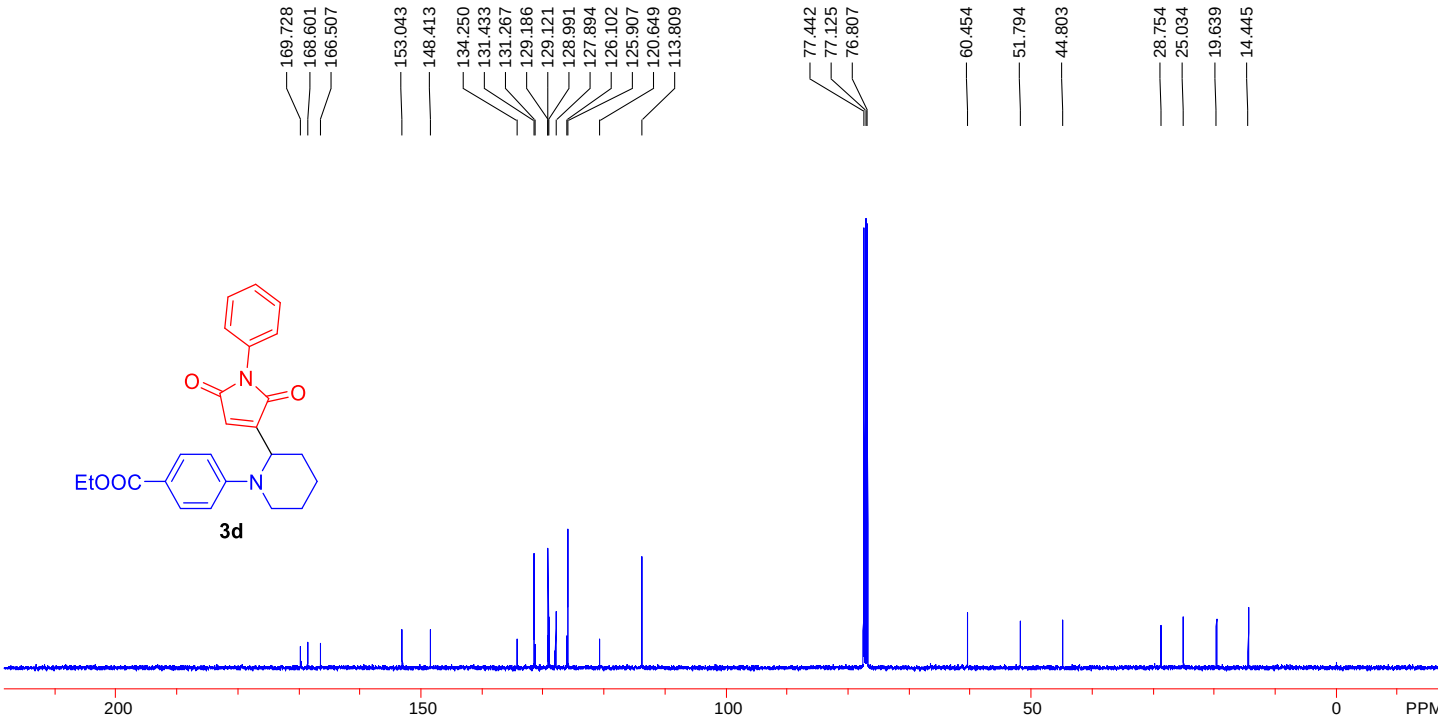
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3c**



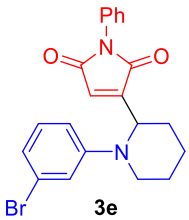
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) of **3c**



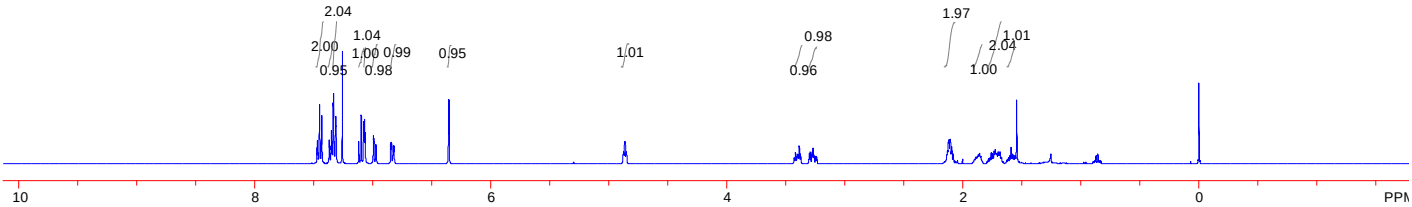
^{1}H NMR (400 MHz, $CDCl_3$) of **3d**



^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3d**

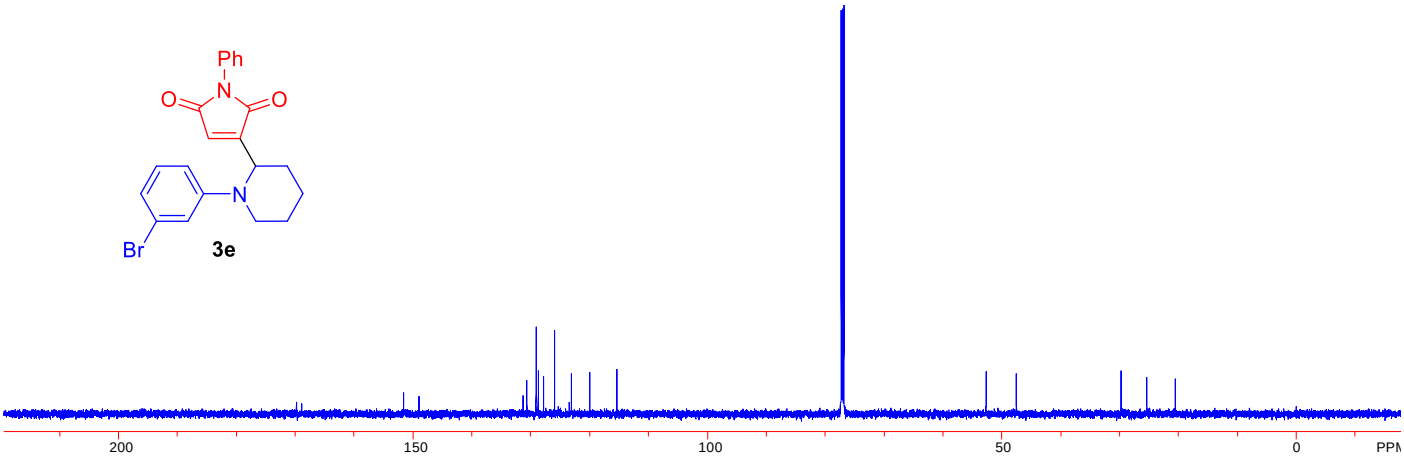


3e

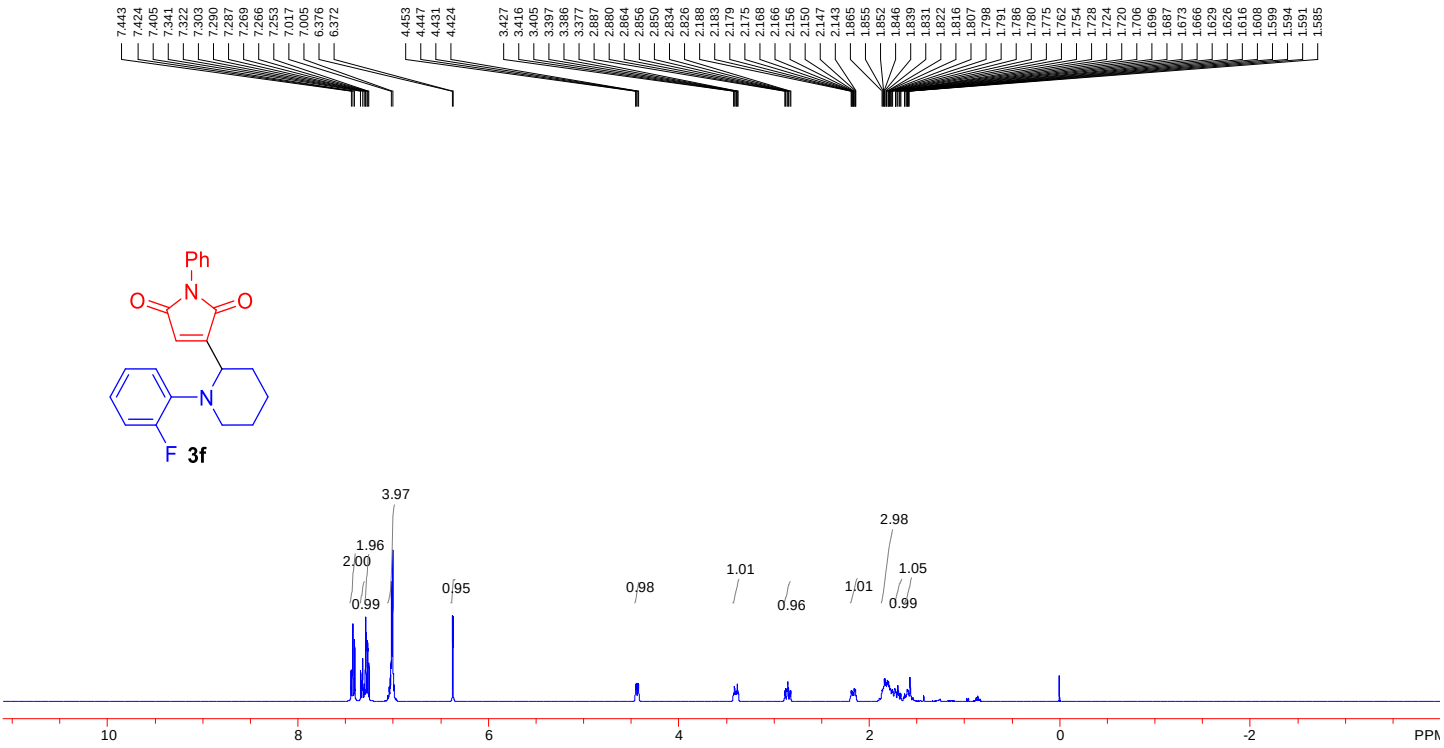


^{1}H NMR (400 MHz, $CDCl_3$) of **3e**

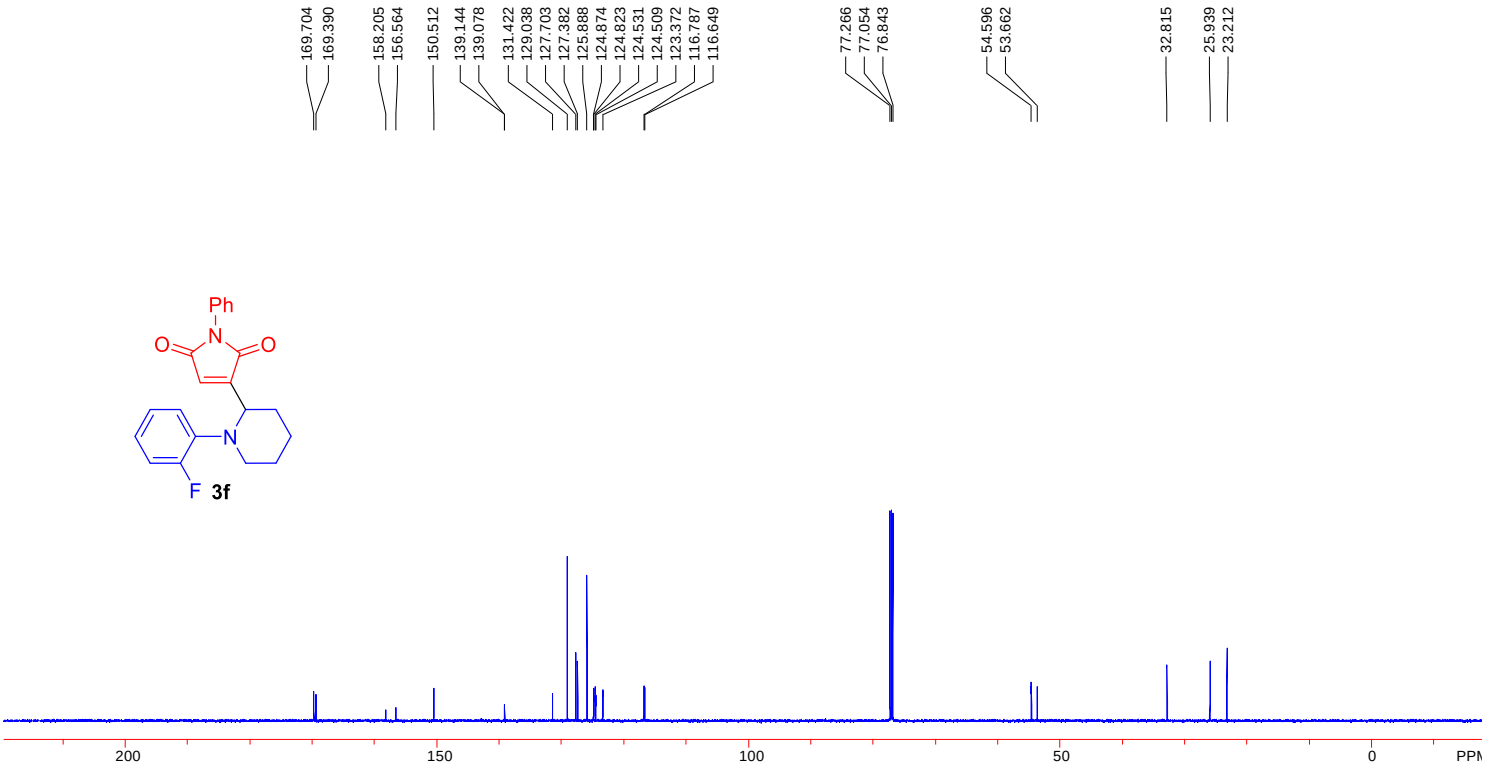
3e



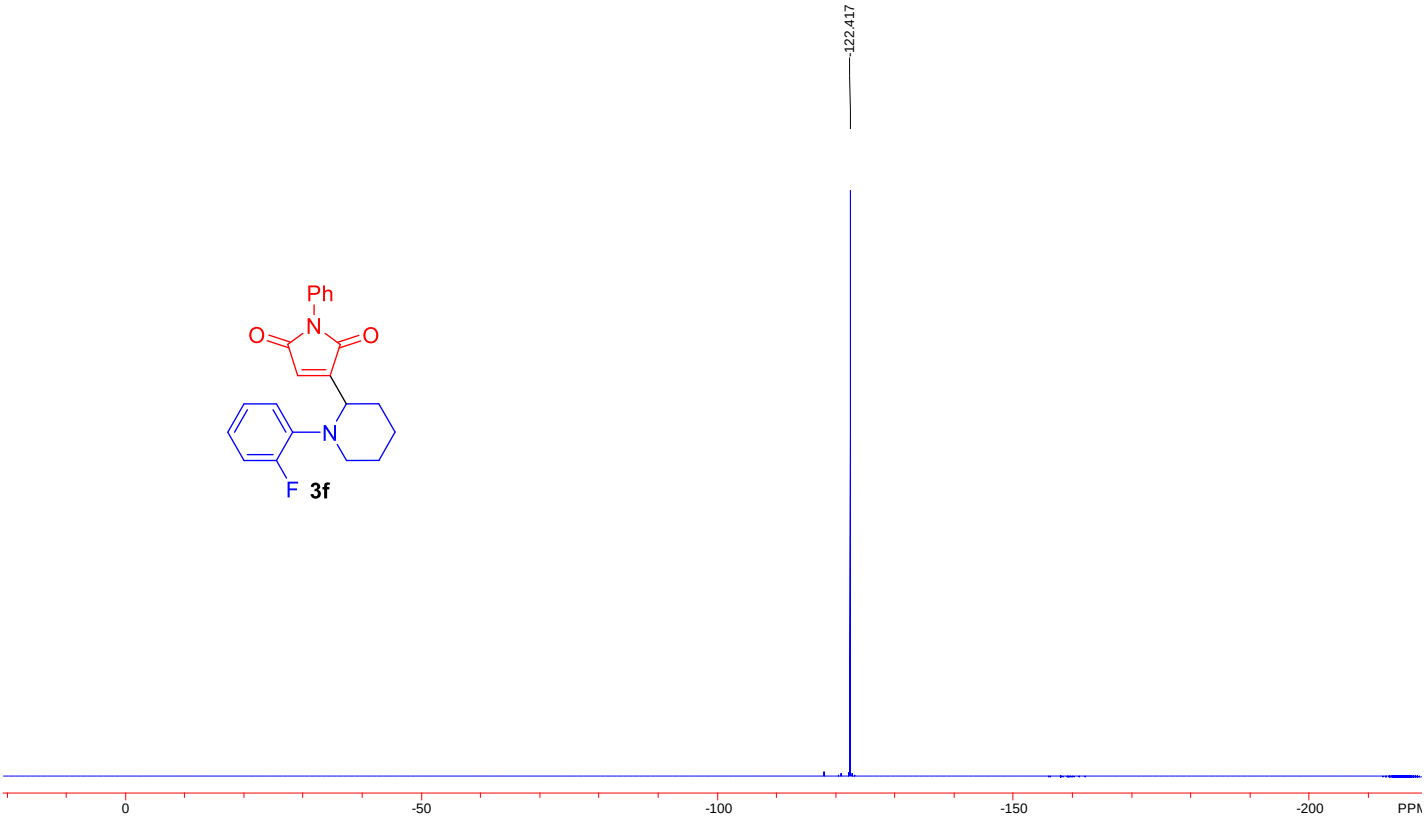
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3e**



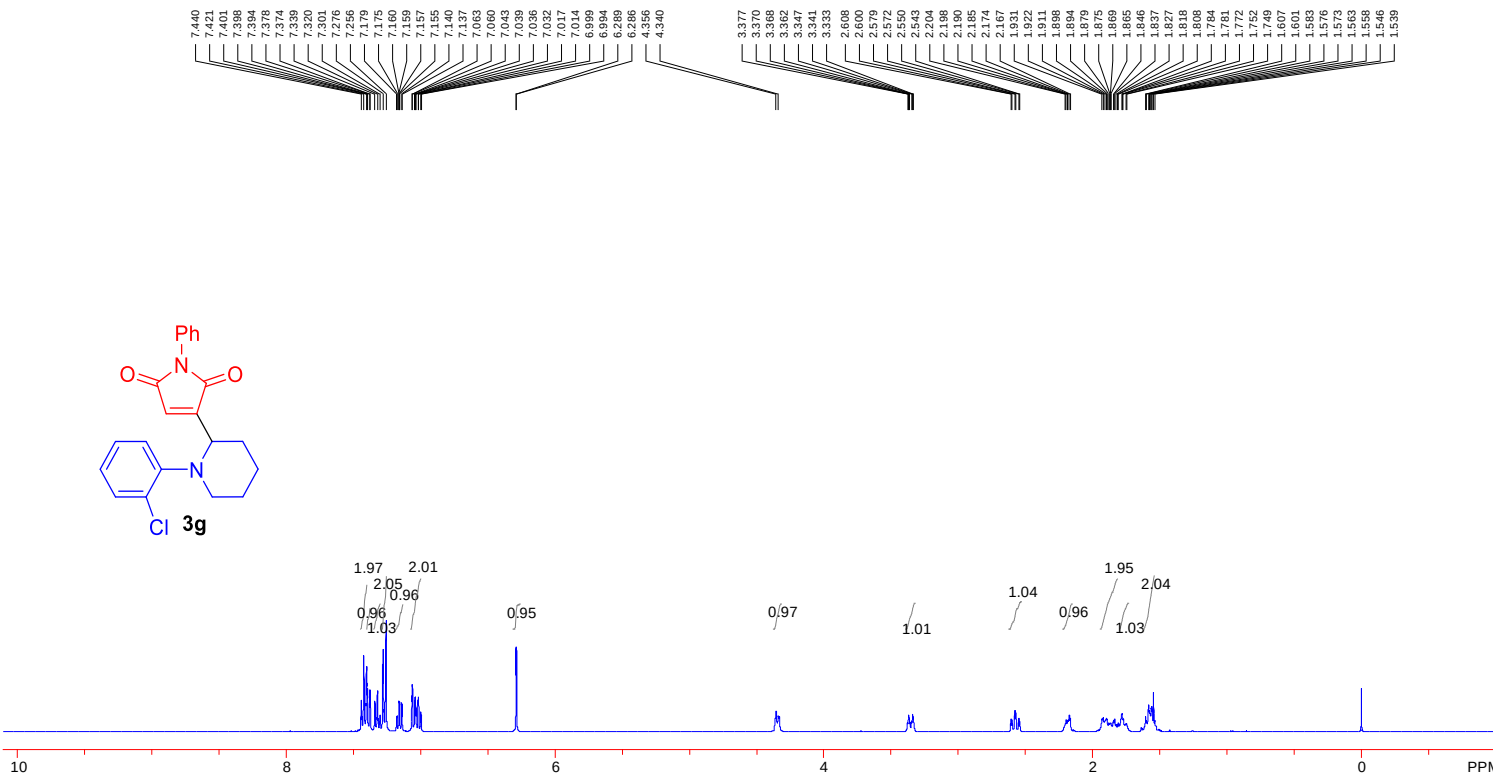
^{1}H NMR (400 MHz, $CDCl_3$) of **3f**



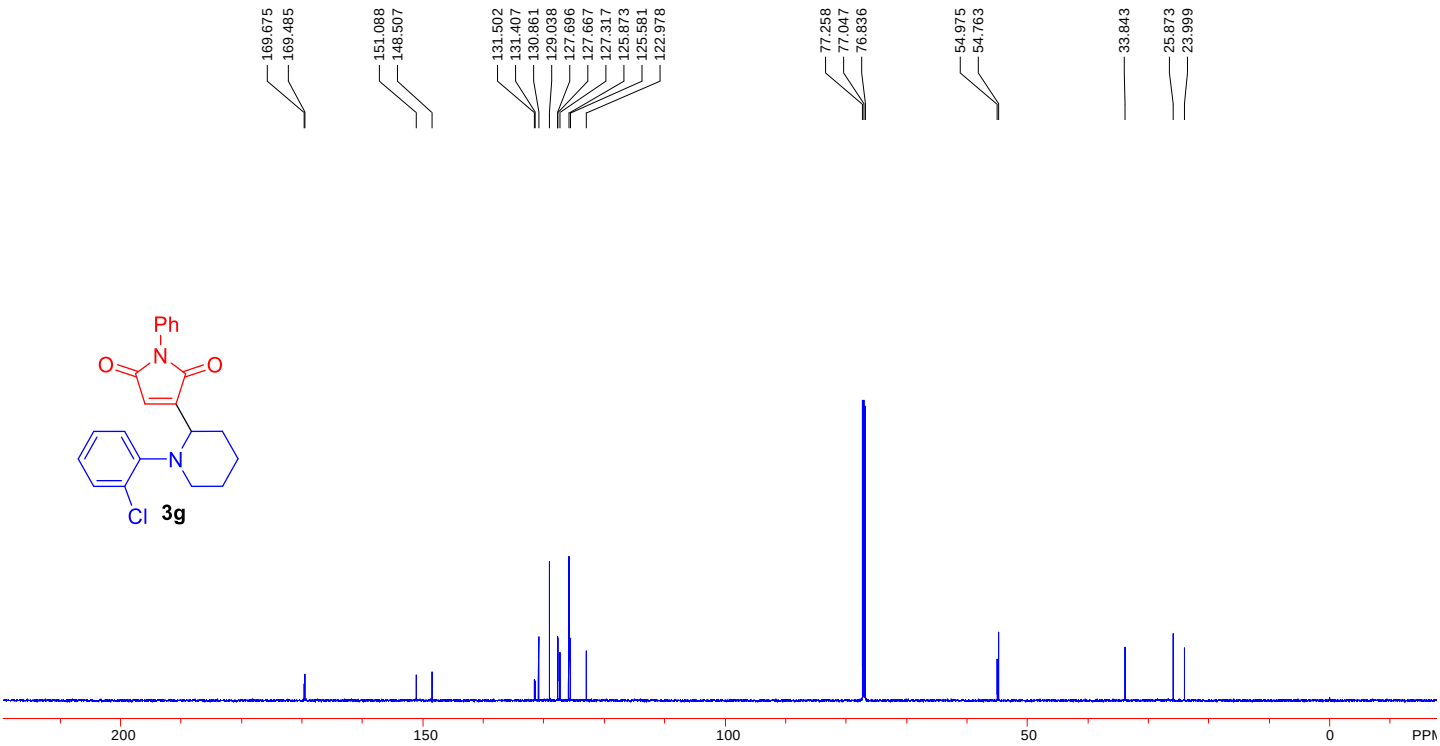
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3f**



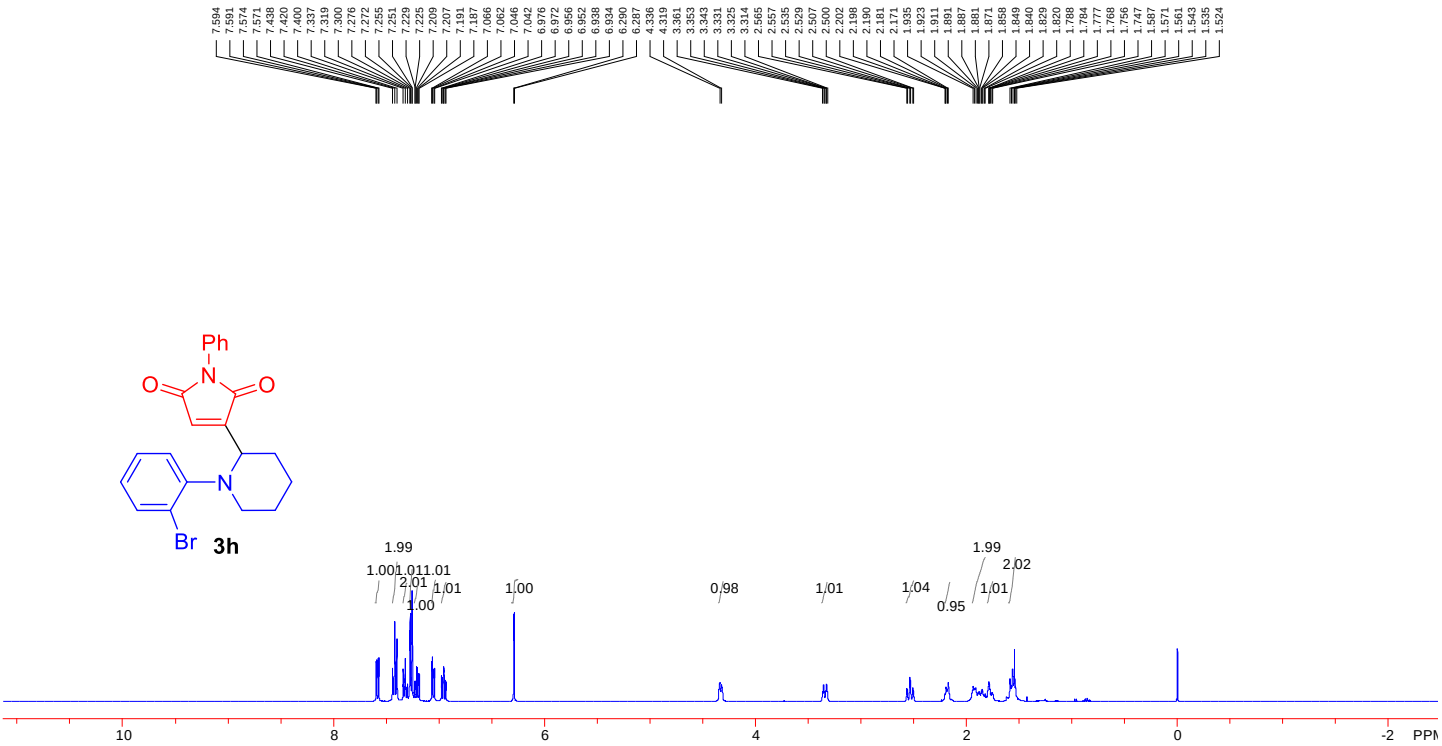
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) of **3f**



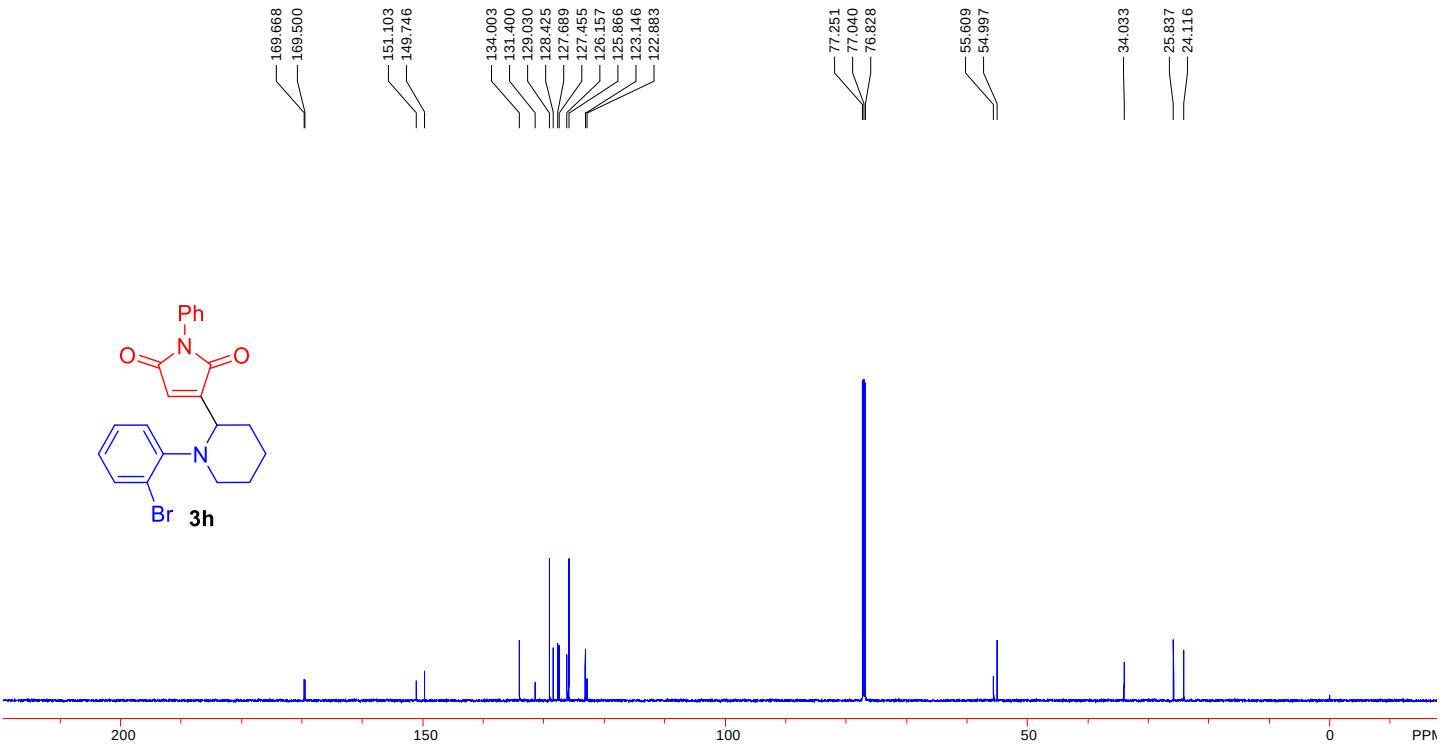
^{1}H NMR (400 MHz, $CDCl_3$) of **3g**



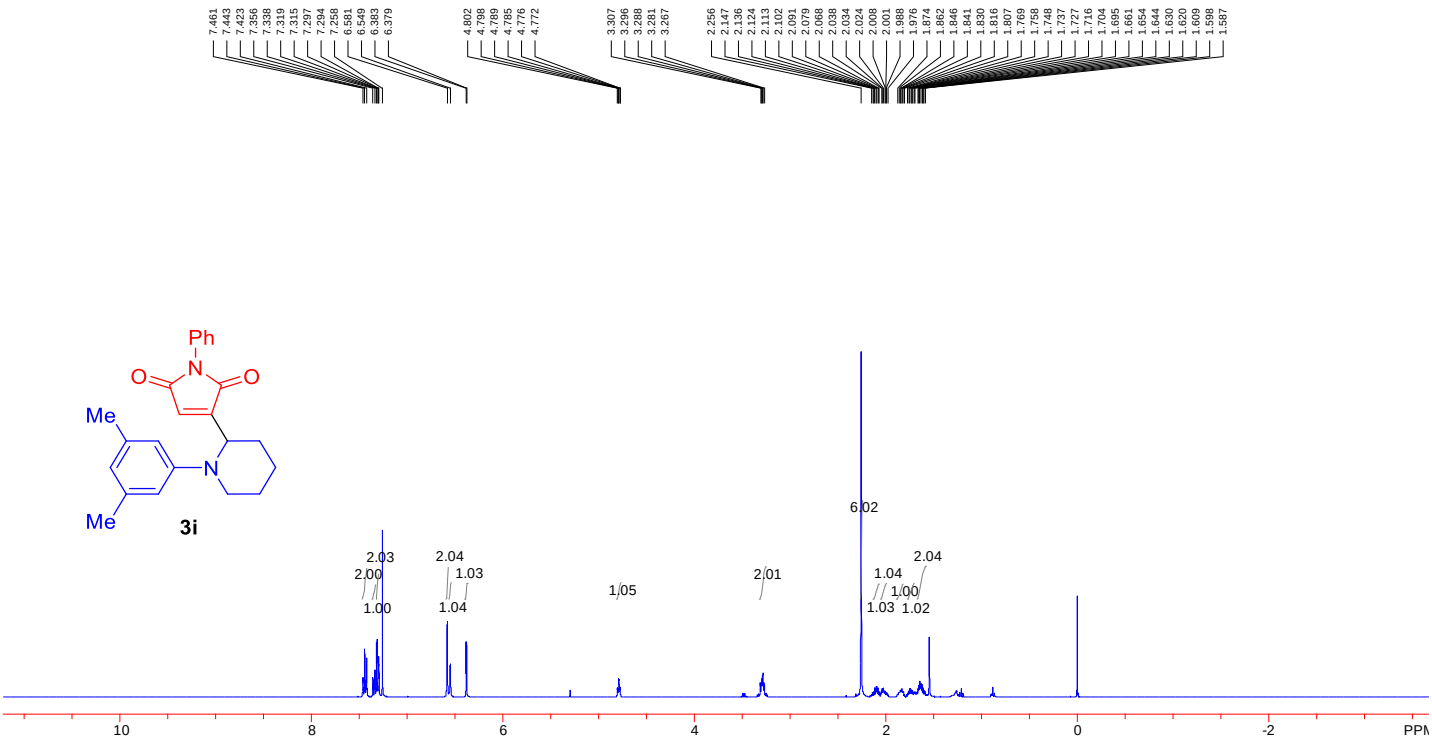
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3g**



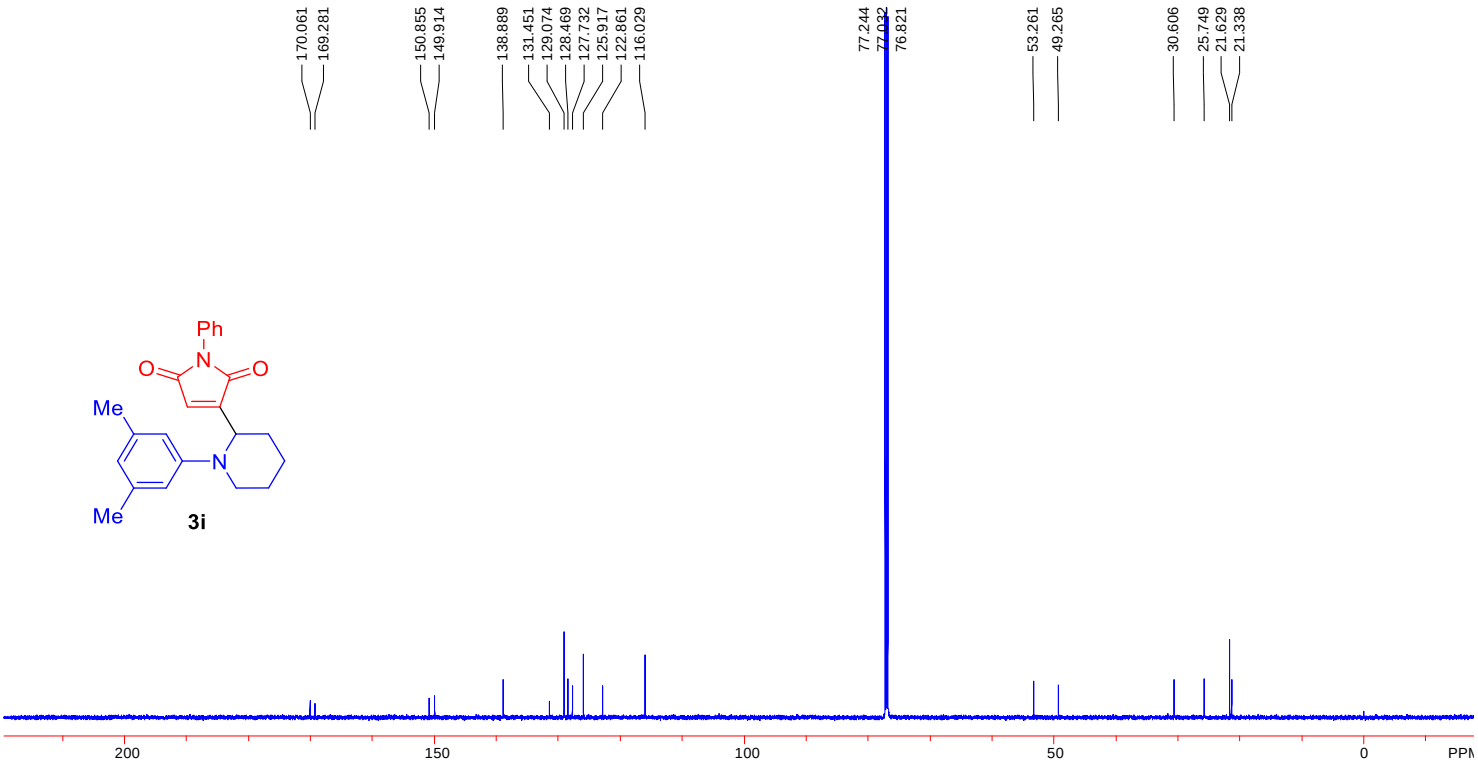
^{1}H NMR (400 MHz, $CDCl_3$) of **3h**



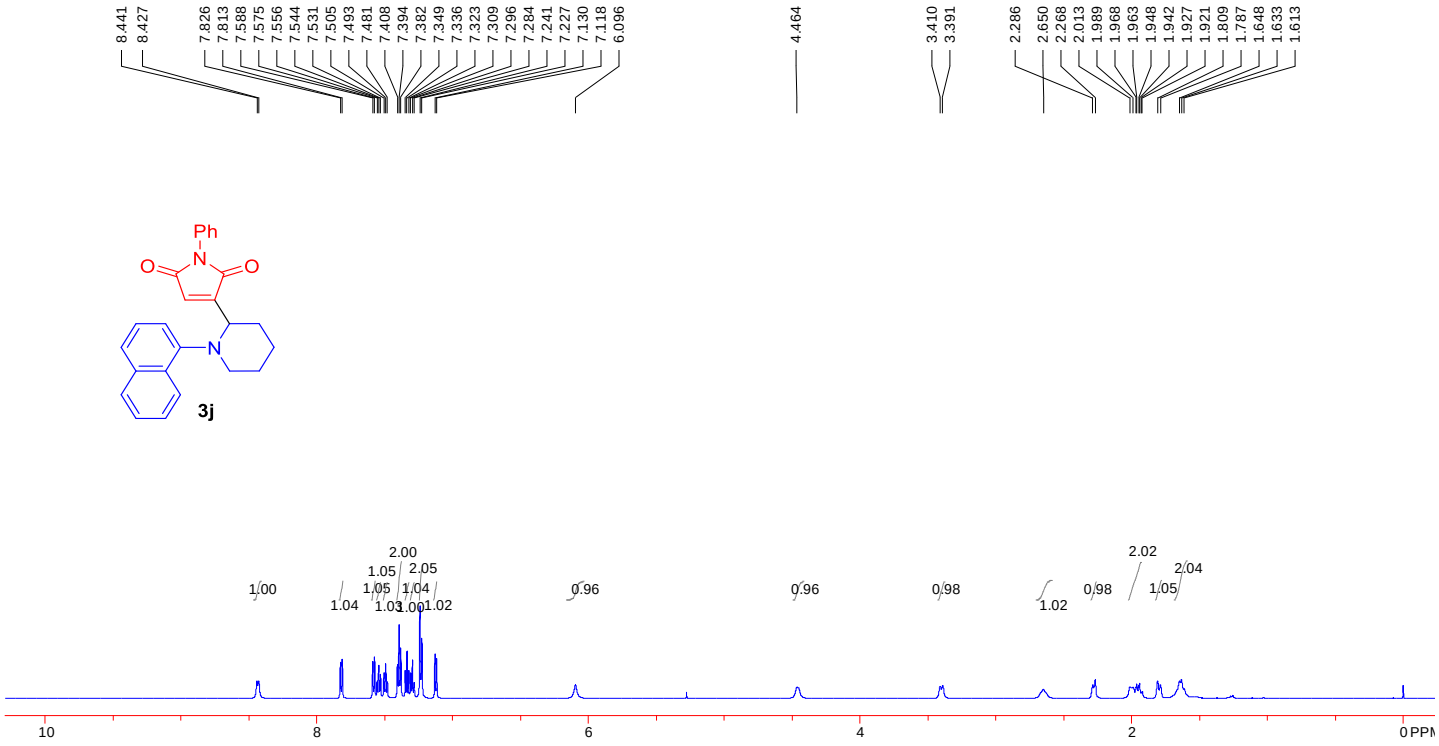
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3h**



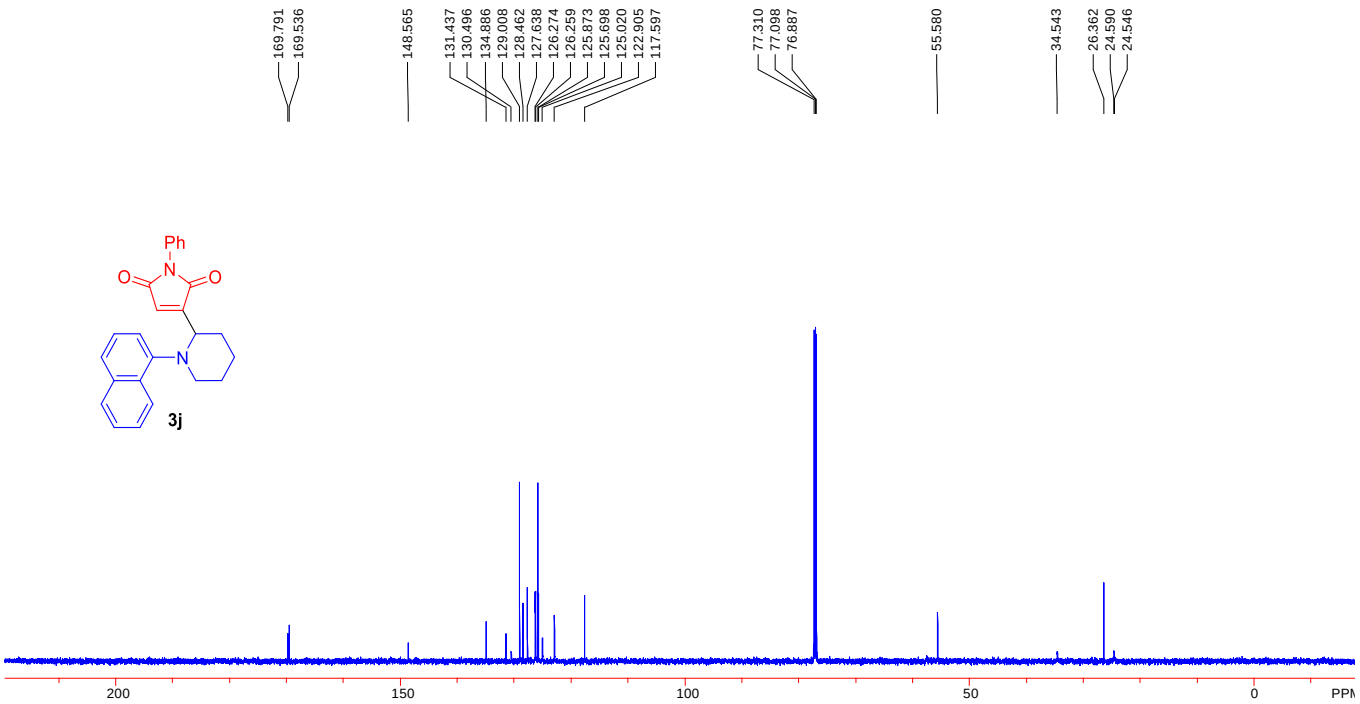
^{1}H NMR (400 MHz, $CDCl_3$) of **3i**



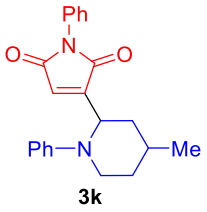
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3i**

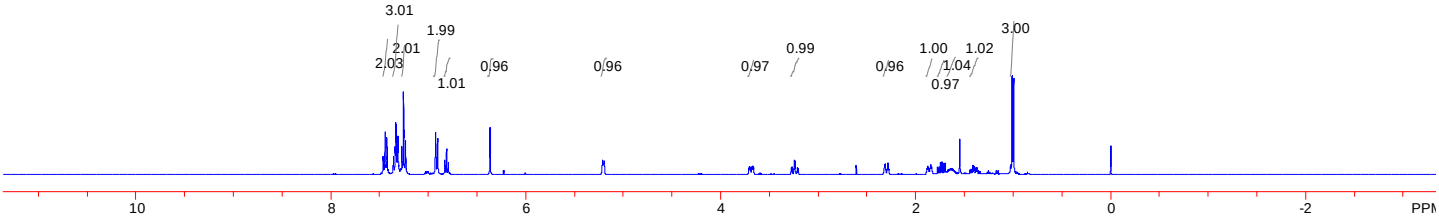


^{1}H NMR (600 MHz, $CDCl_3$) of **3j**

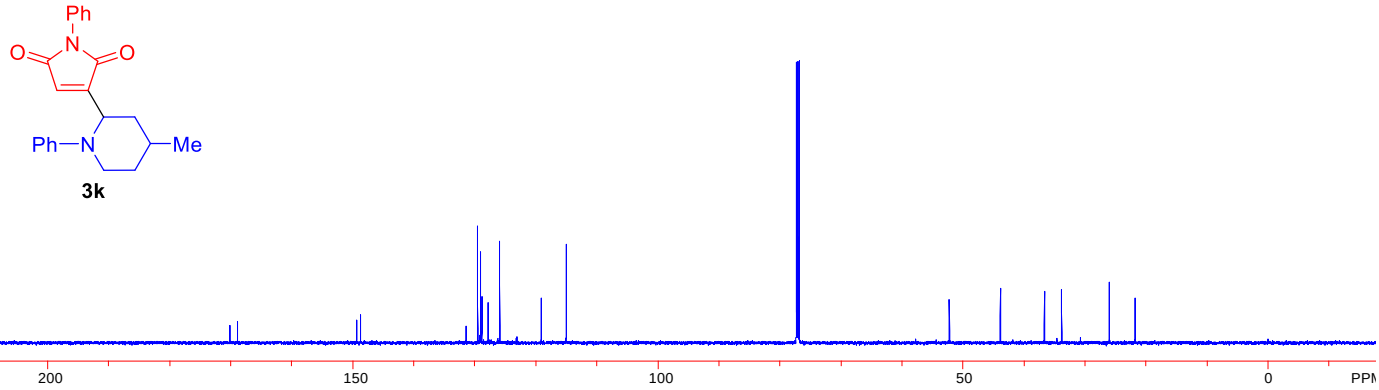


^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3j**

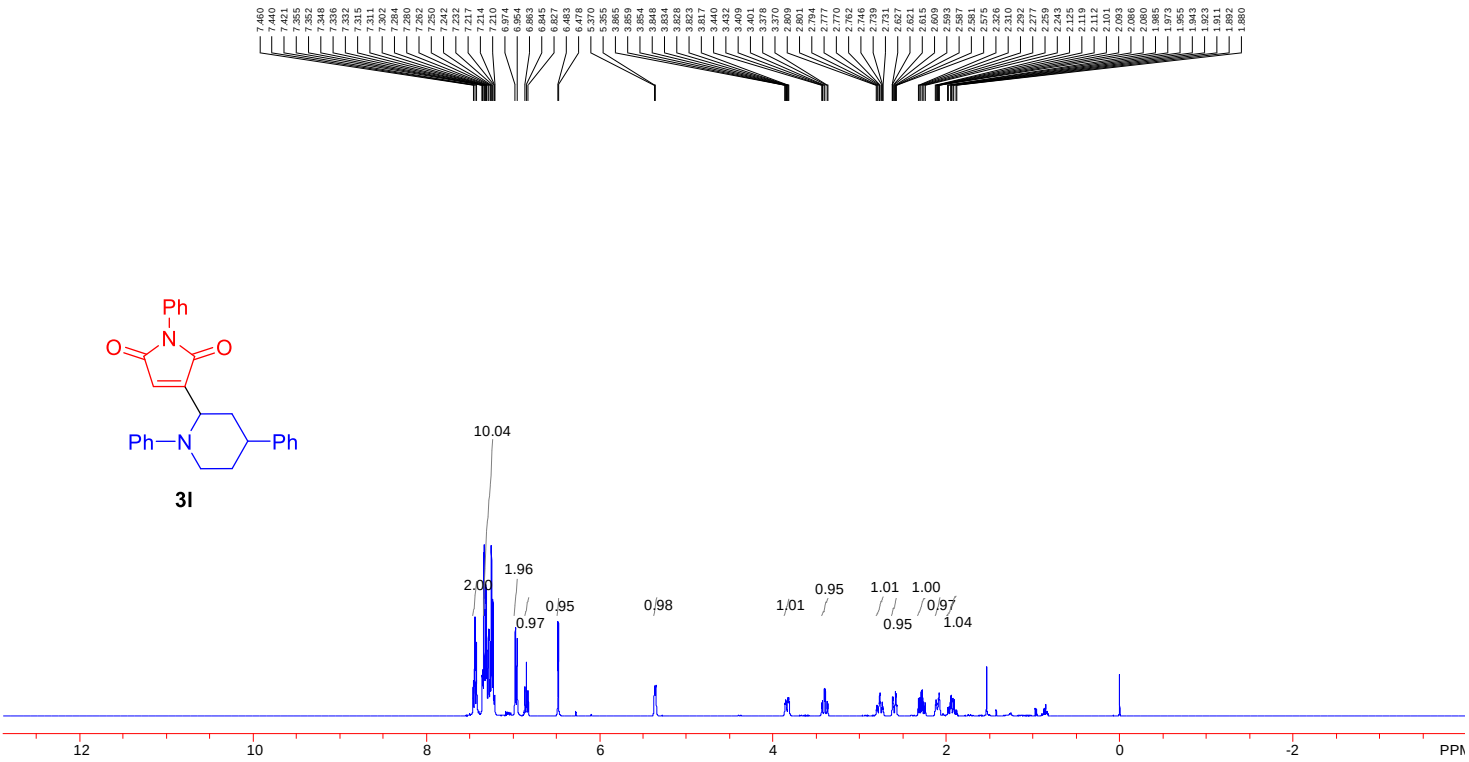




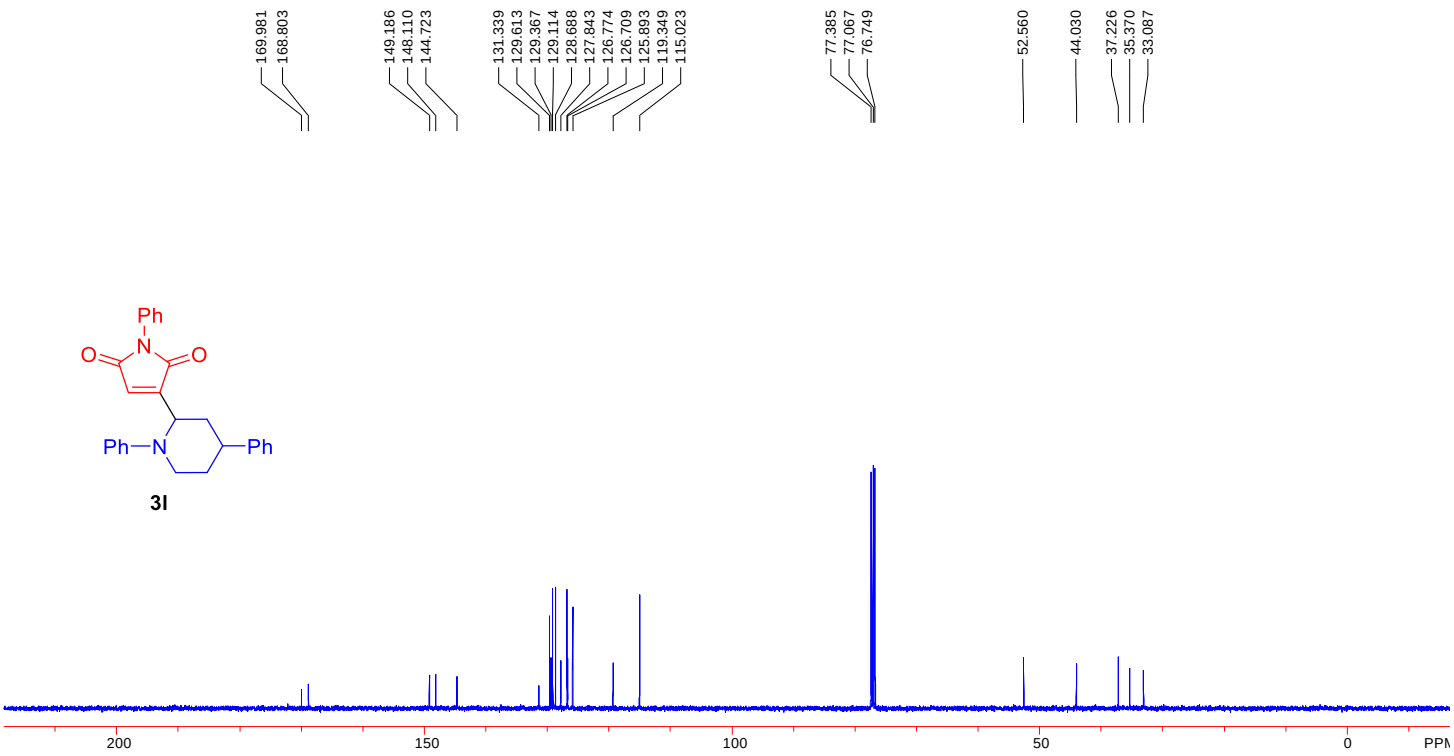
^{1}H NMR (400 MHz, $CDCl_3$) of **3k**



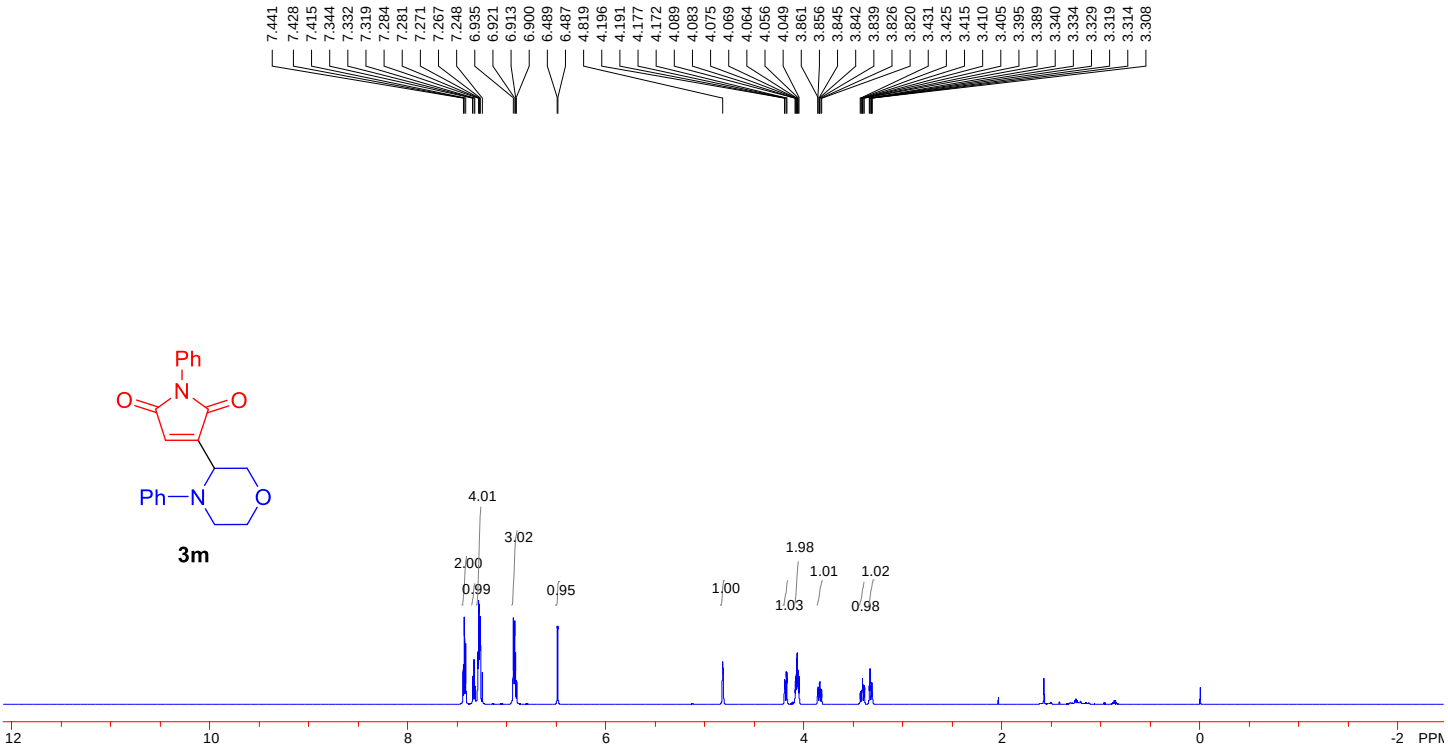
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3k**



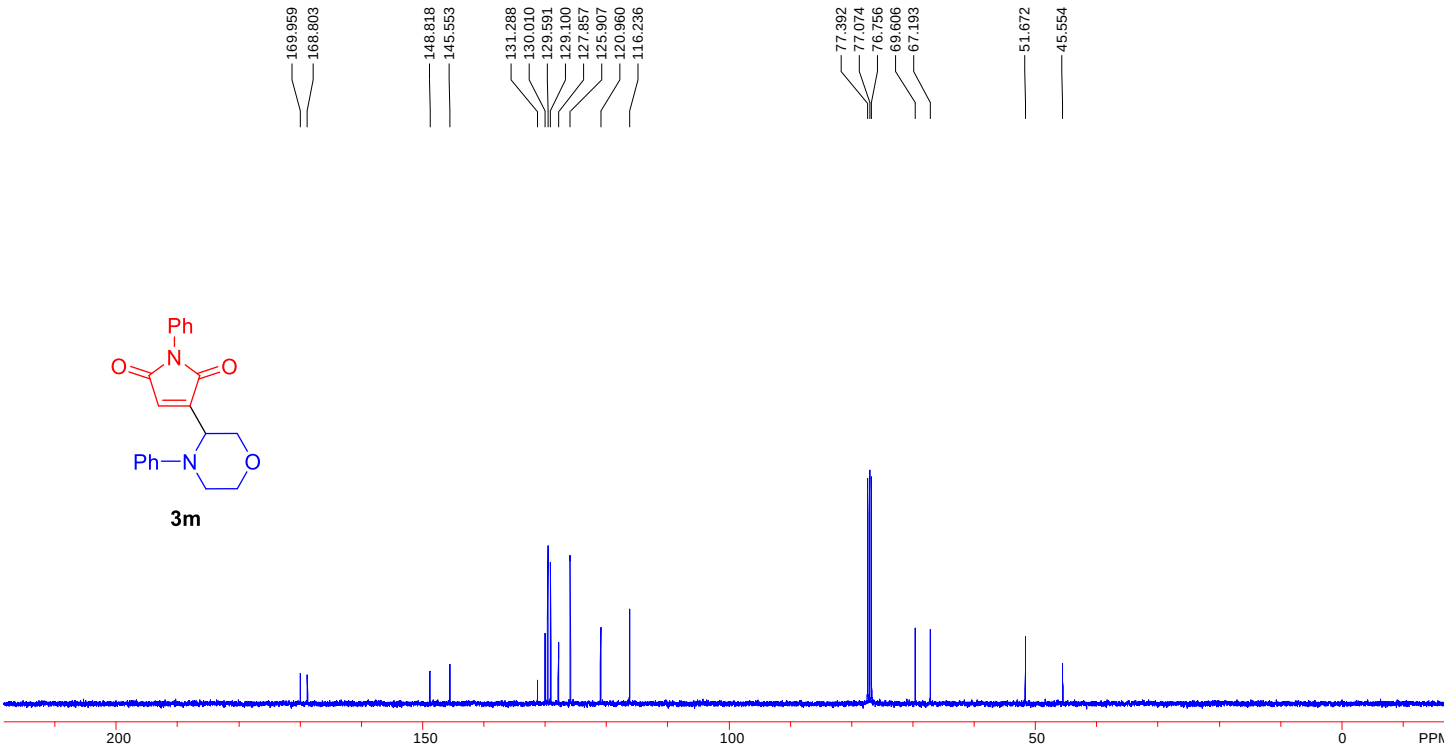
^{1}H NMR (400 MHz, $CDCl_3$) of **3l**



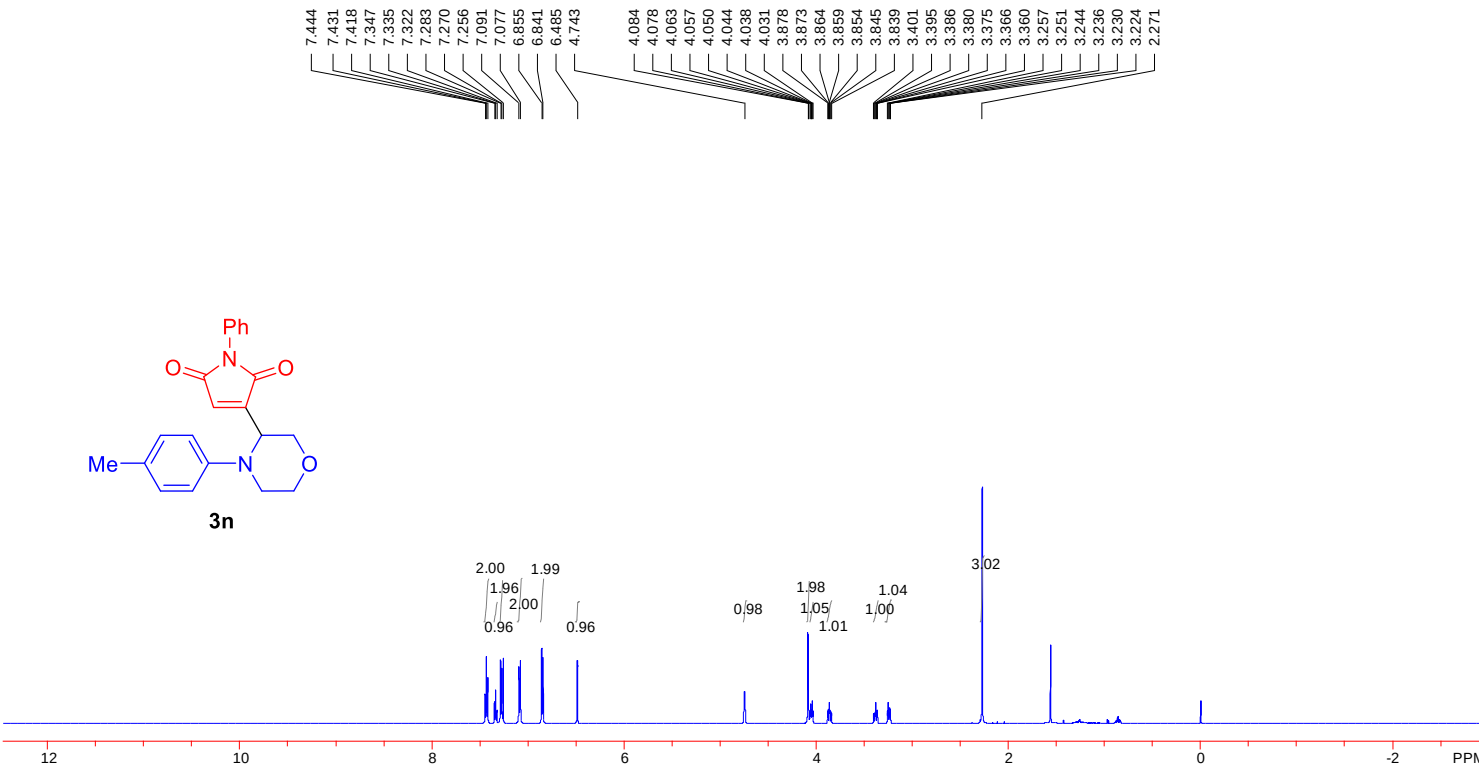
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3l**



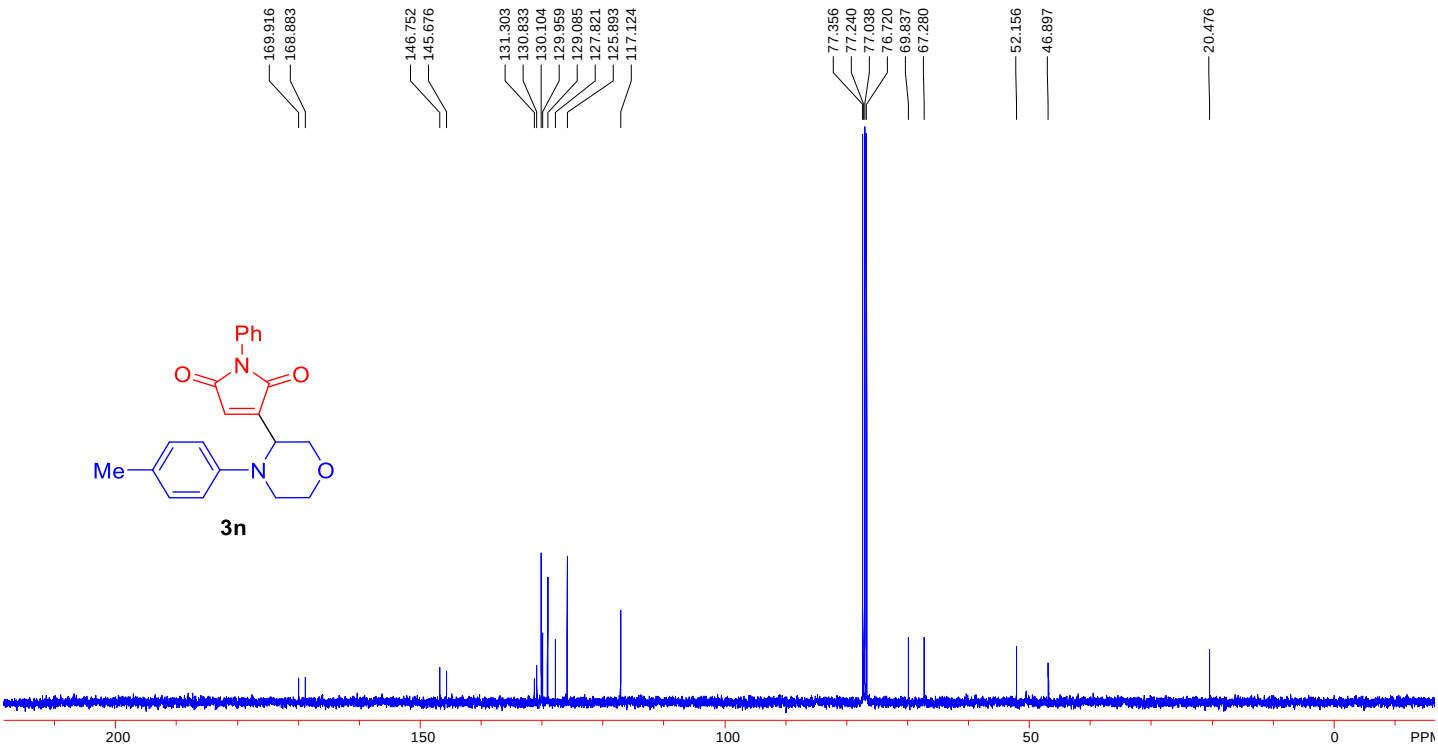
^{1}H NMR (600 MHz, $CDCl_3$) of **3m**



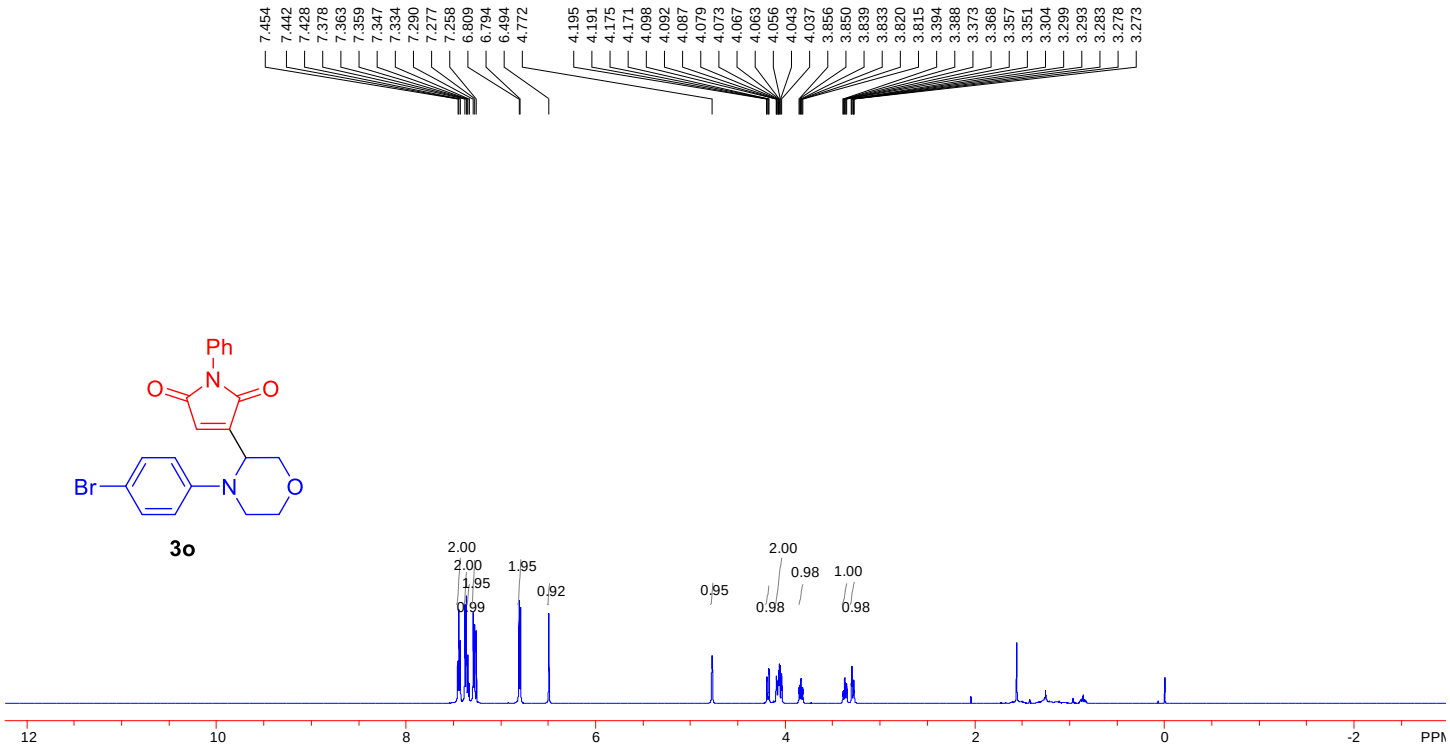
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **3m**



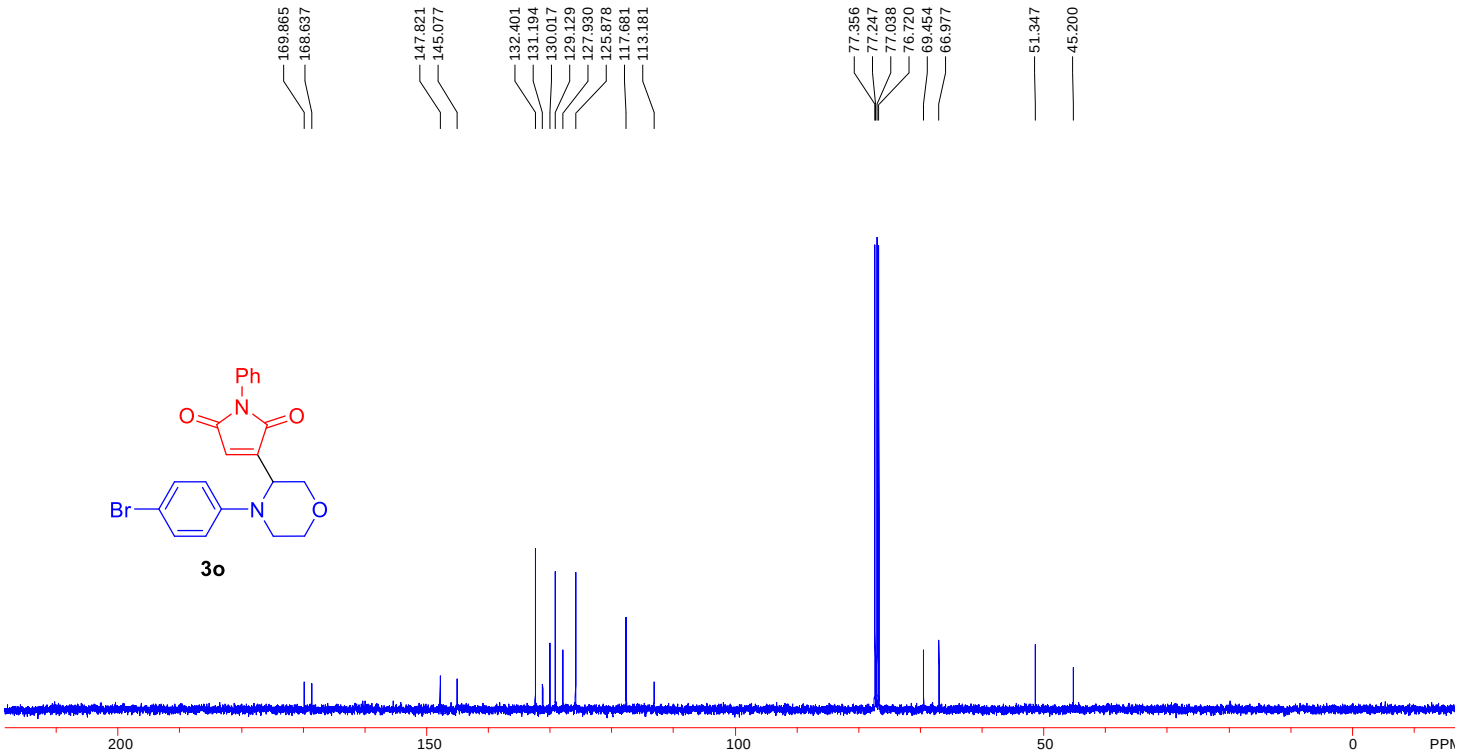
^{1}H NMR (600 MHz, $CDCl_3$) of **3n**



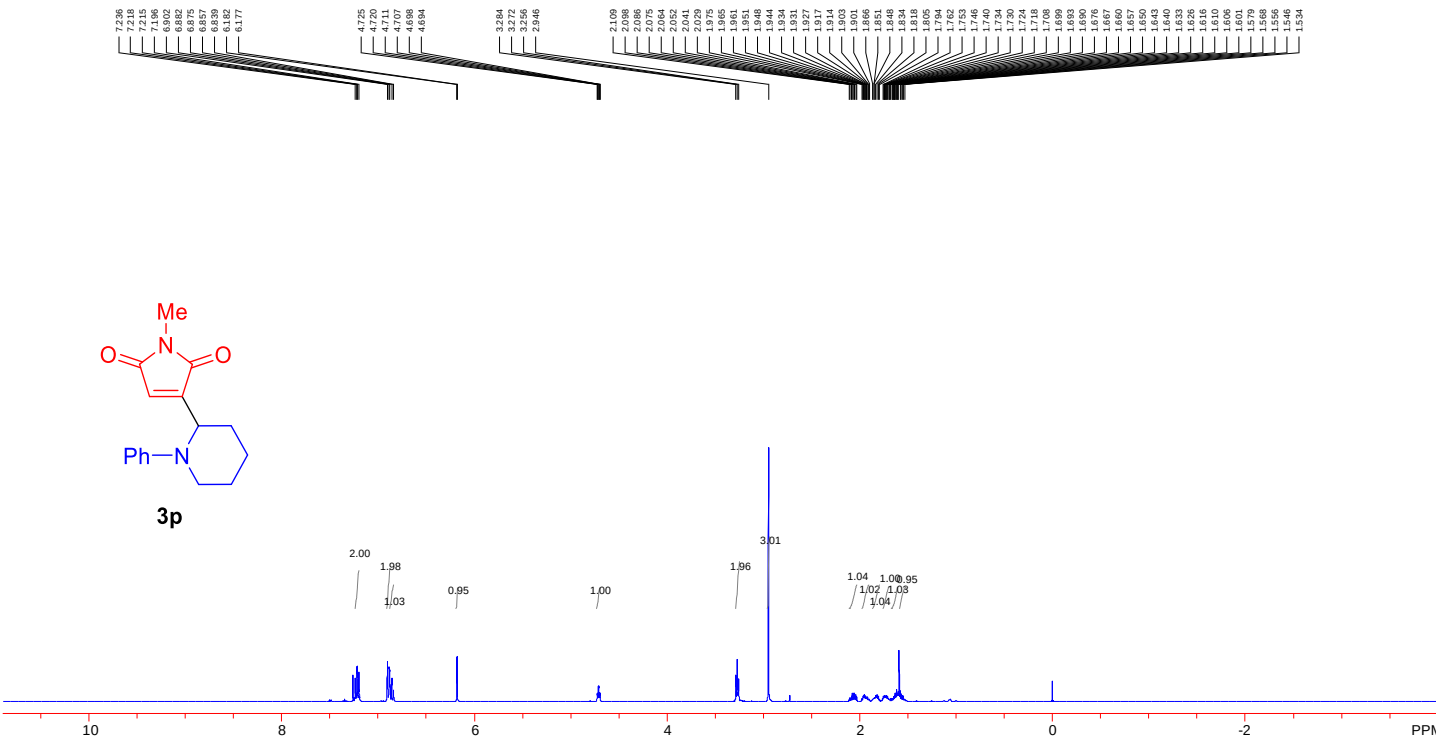
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **3n**



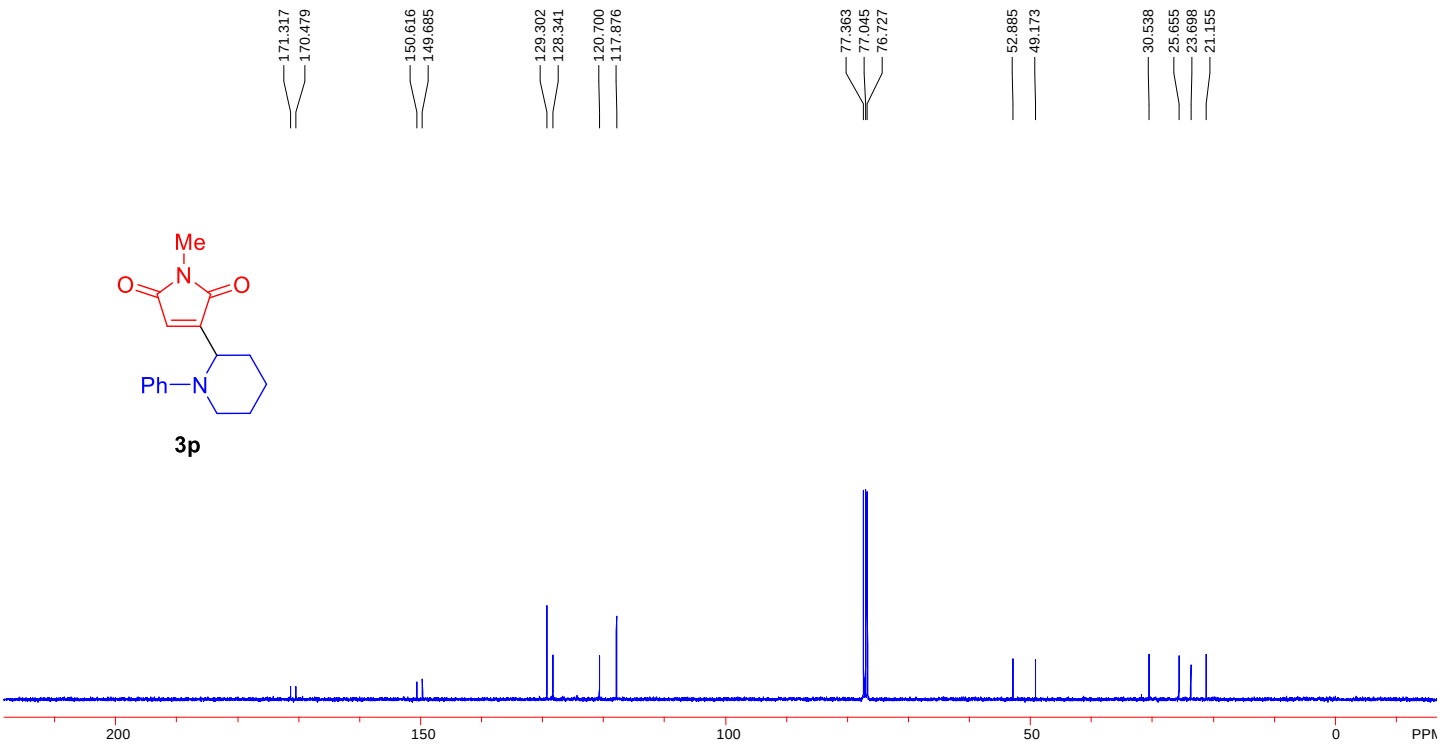
^{1}H NMR (600 MHz, $CDCl_3$) of **3o**



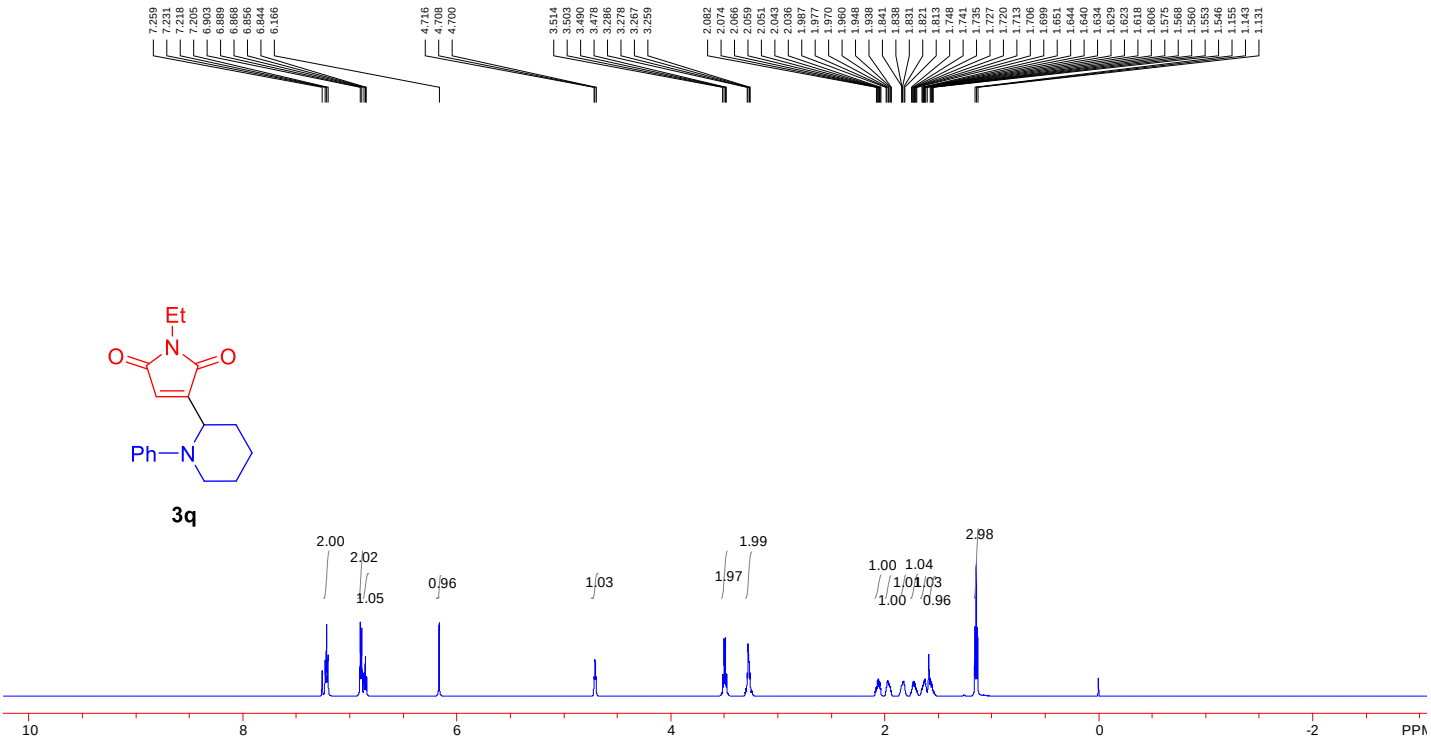
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **3o**



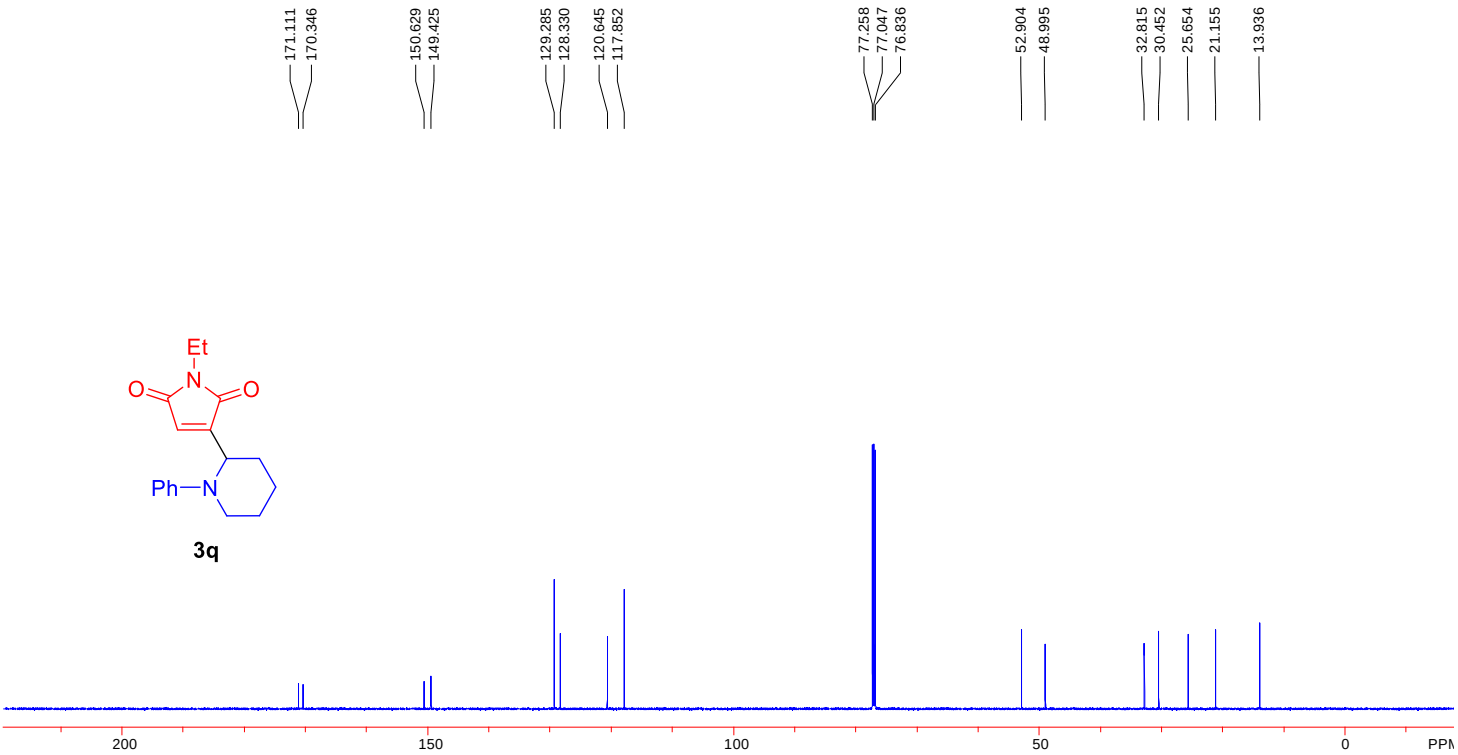
^{1}H NMR (400 MHz, $CDCl_3$) of **3p**



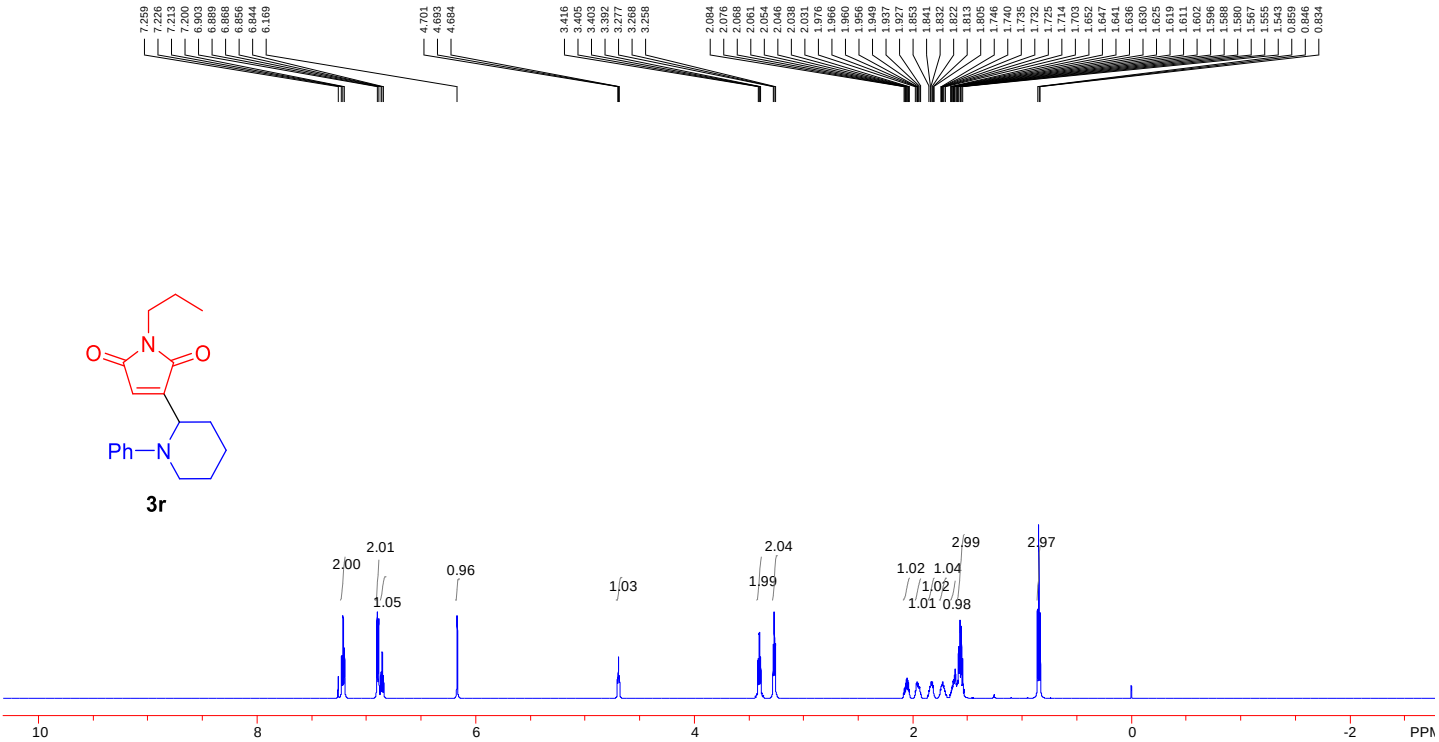
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **3p**



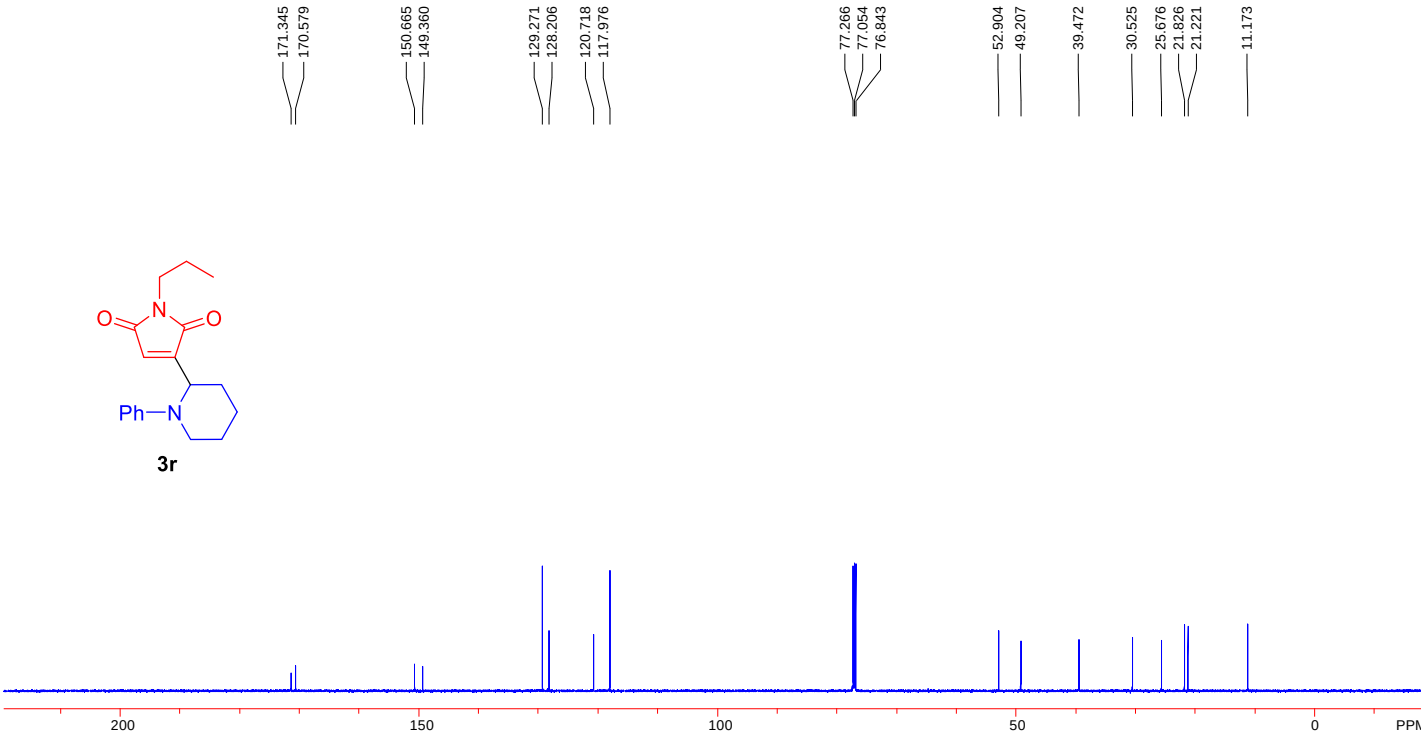
^{1}H NMR (600 MHz, $CDCl_3$) of **3q**



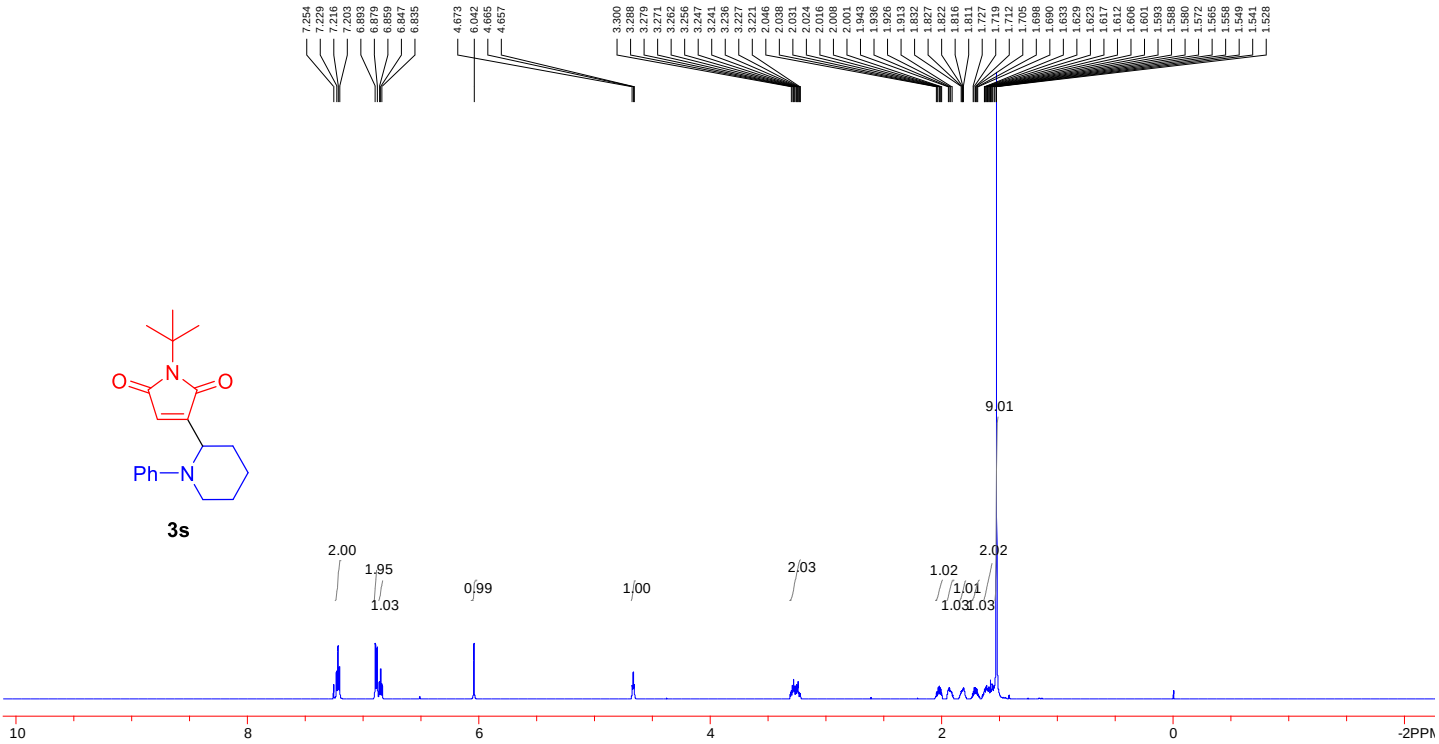
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3q**



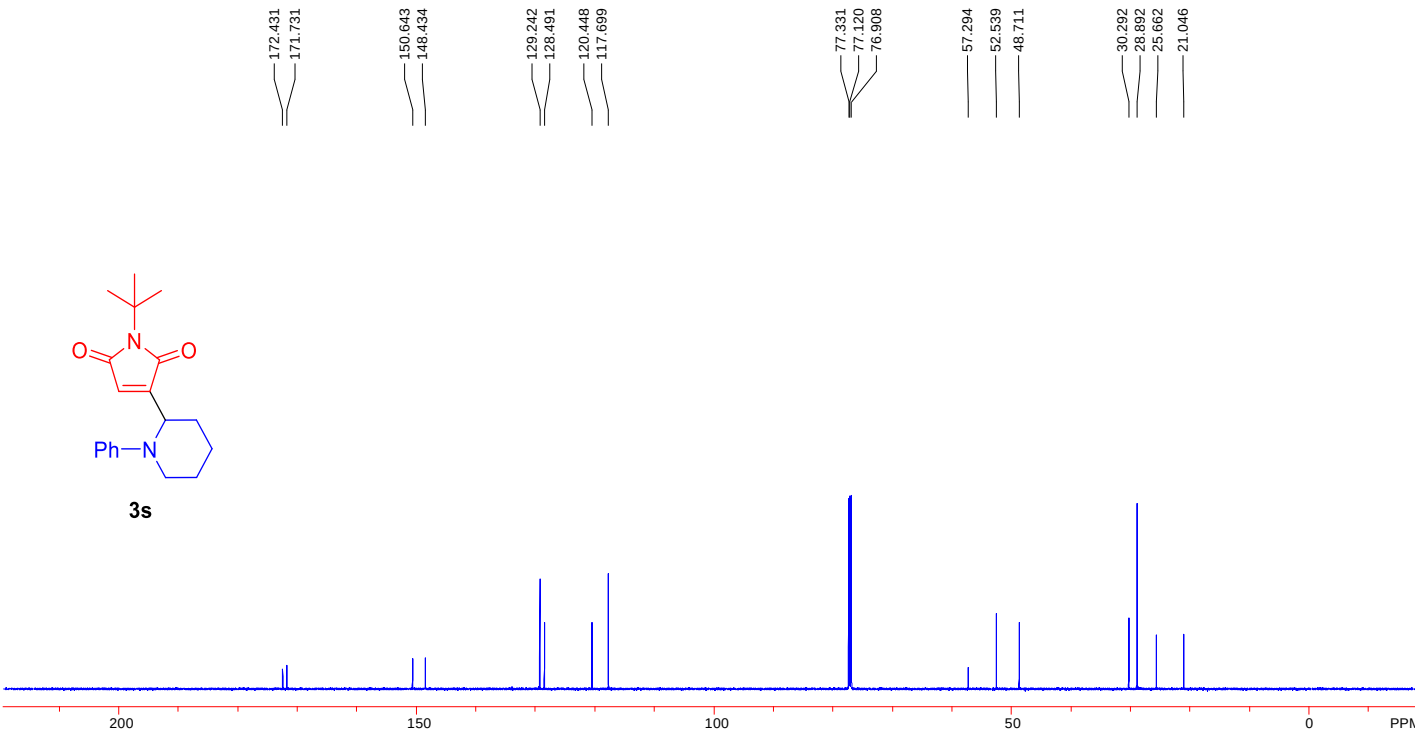
^{1}H NMR (600 MHz, $CDCl_3$) of **3r**



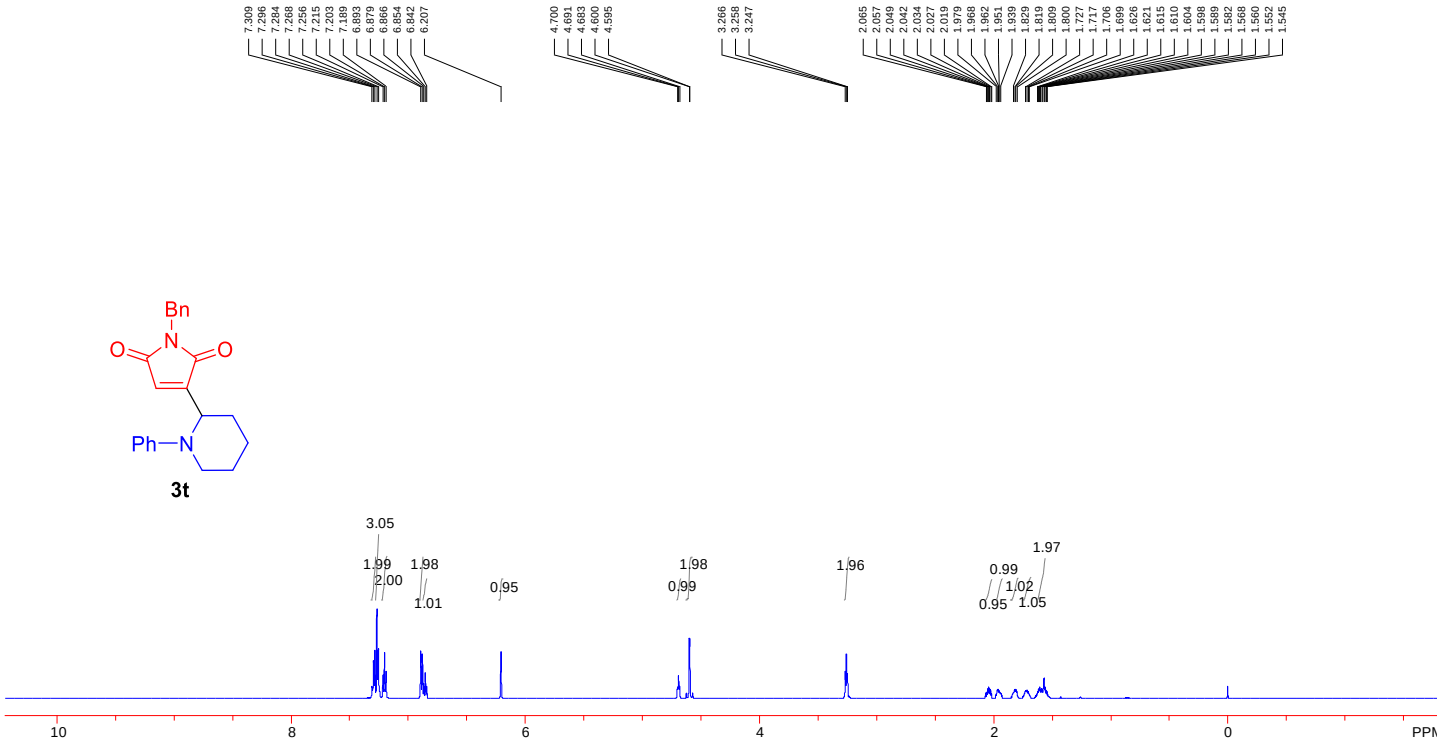
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3r**



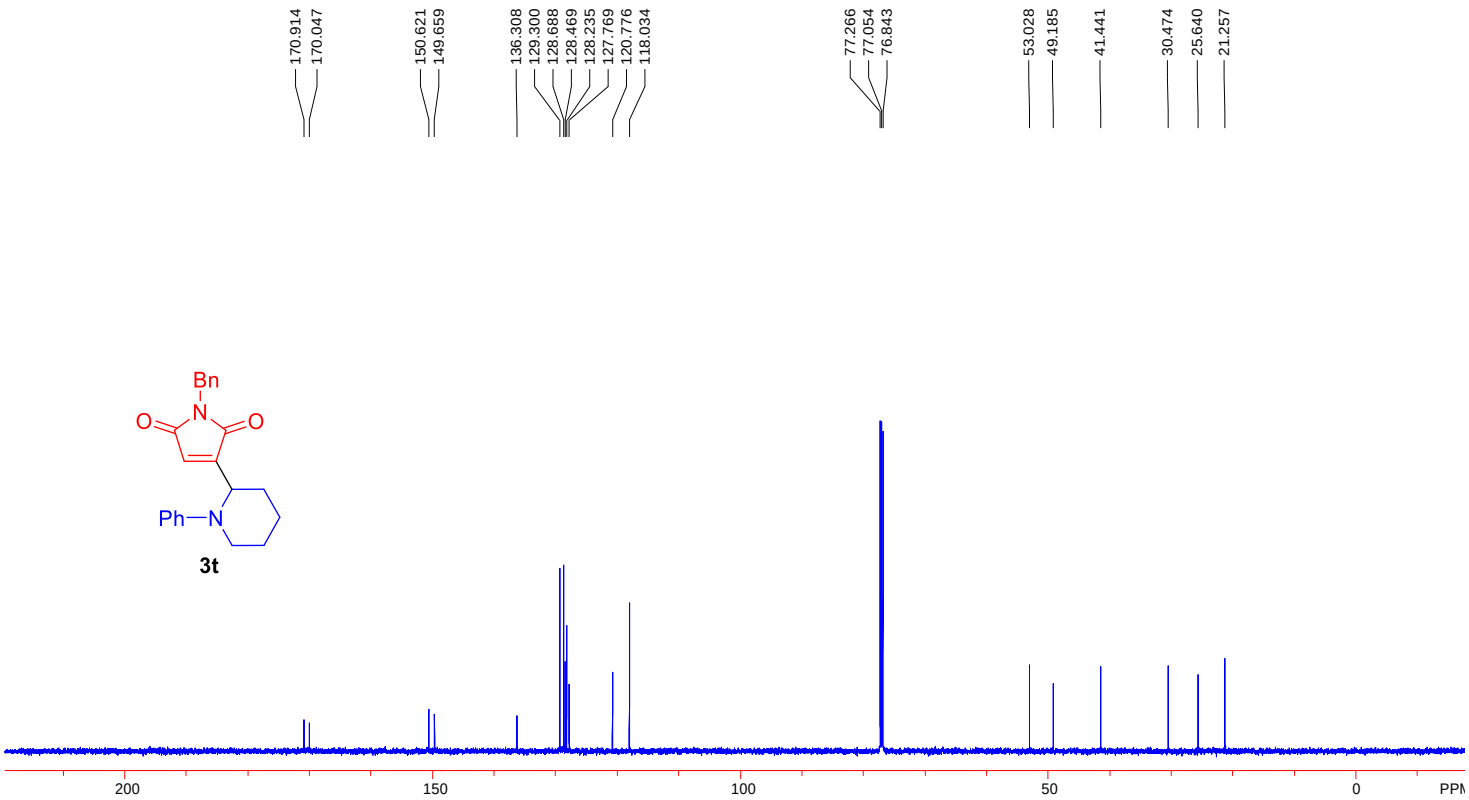
^{1}H NMR (600 MHz, $CDCl_3$) of **3s**



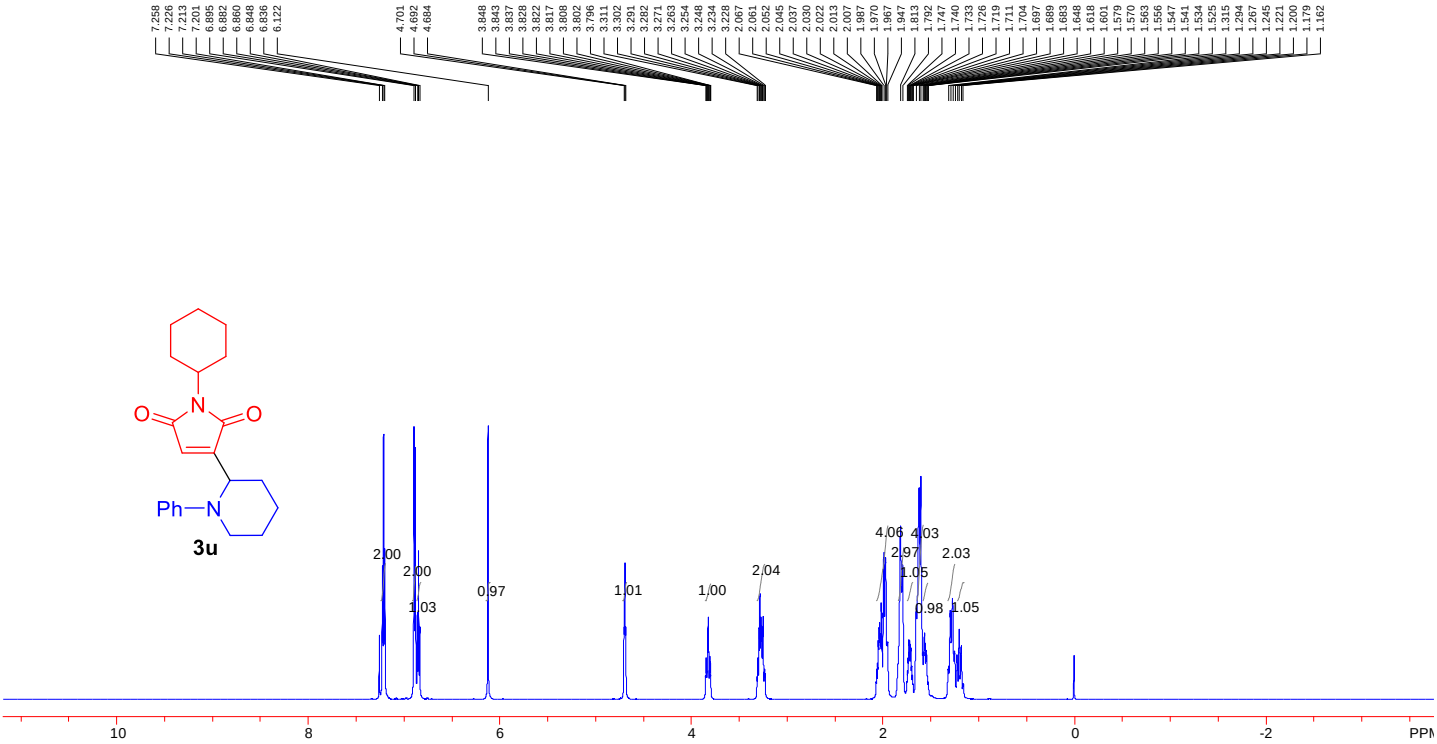
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3s**



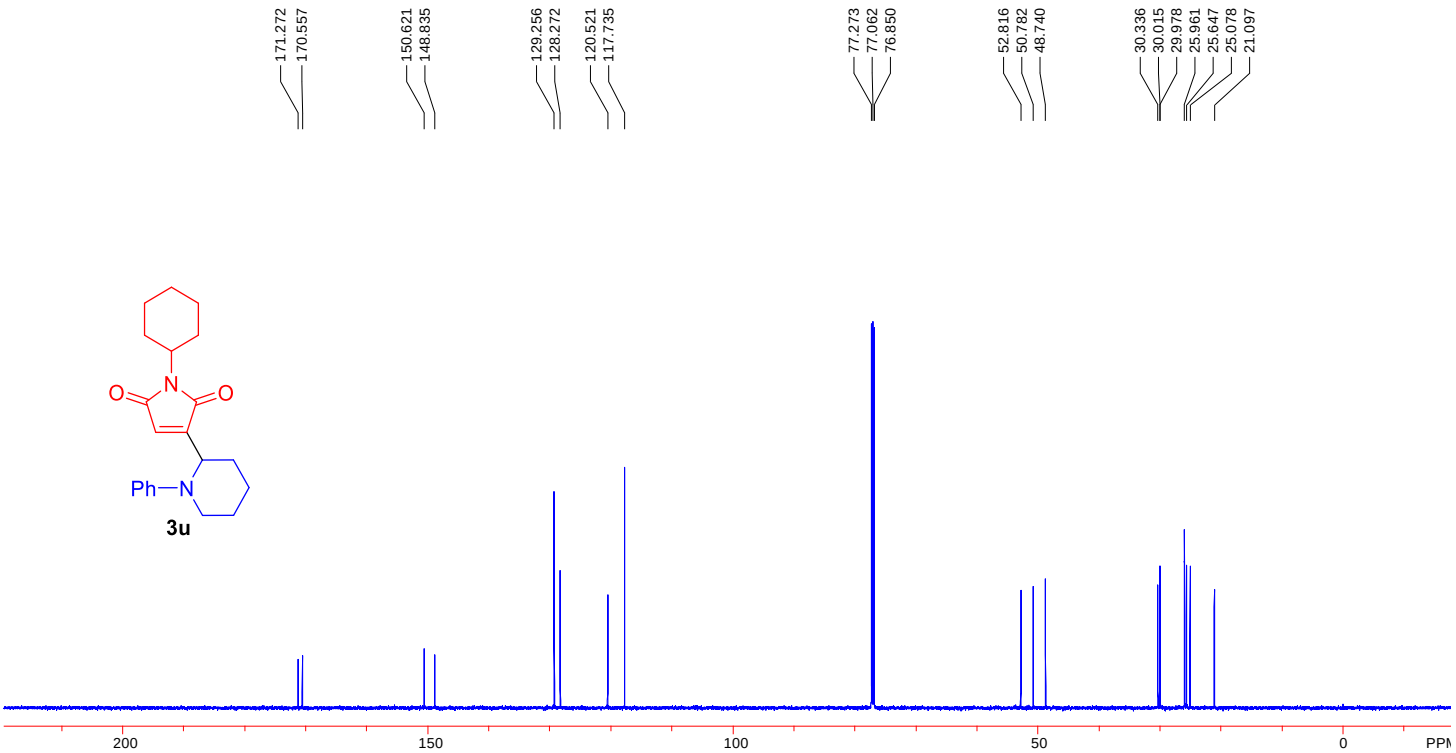
^{1}H NMR (600 MHz, $CDCl_3$) of **3t**



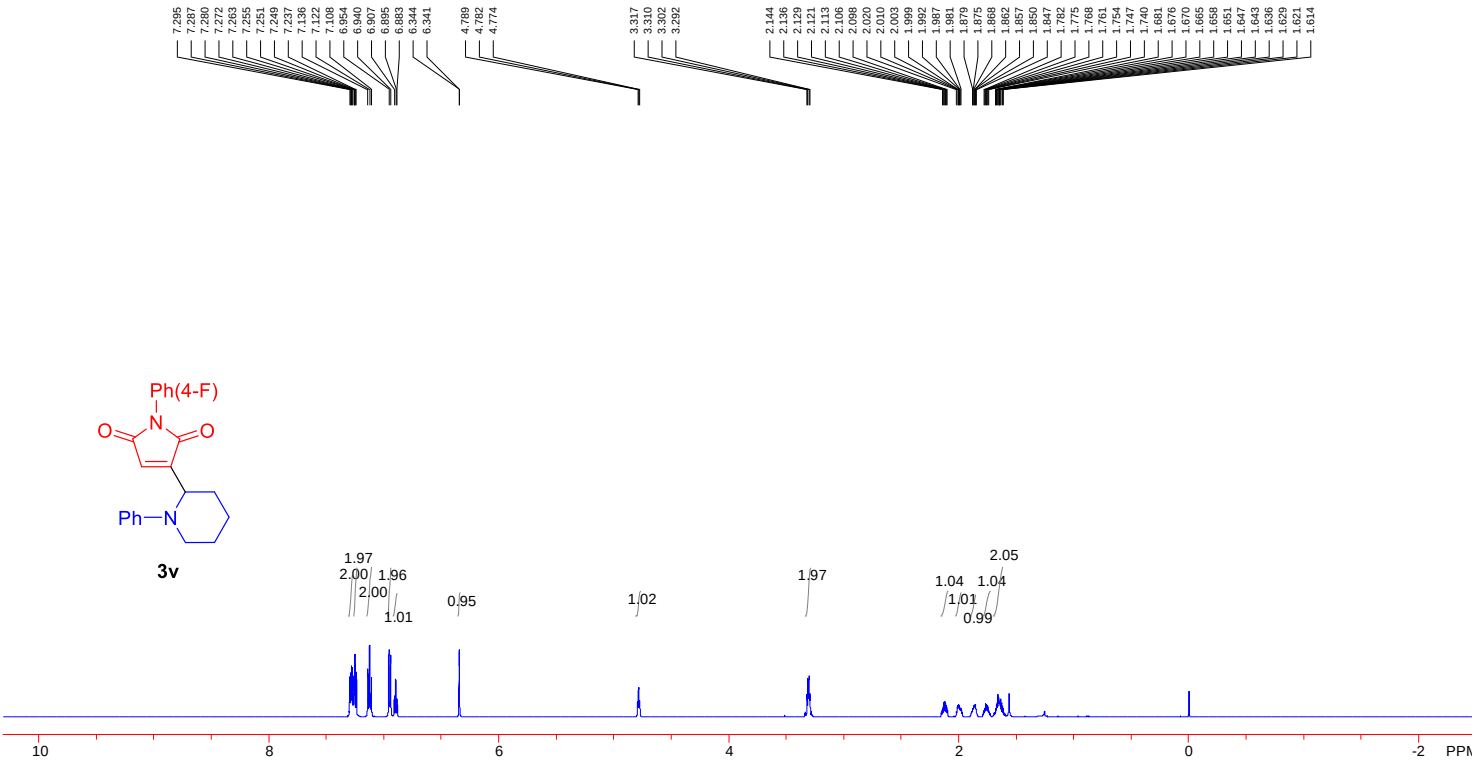
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3t**



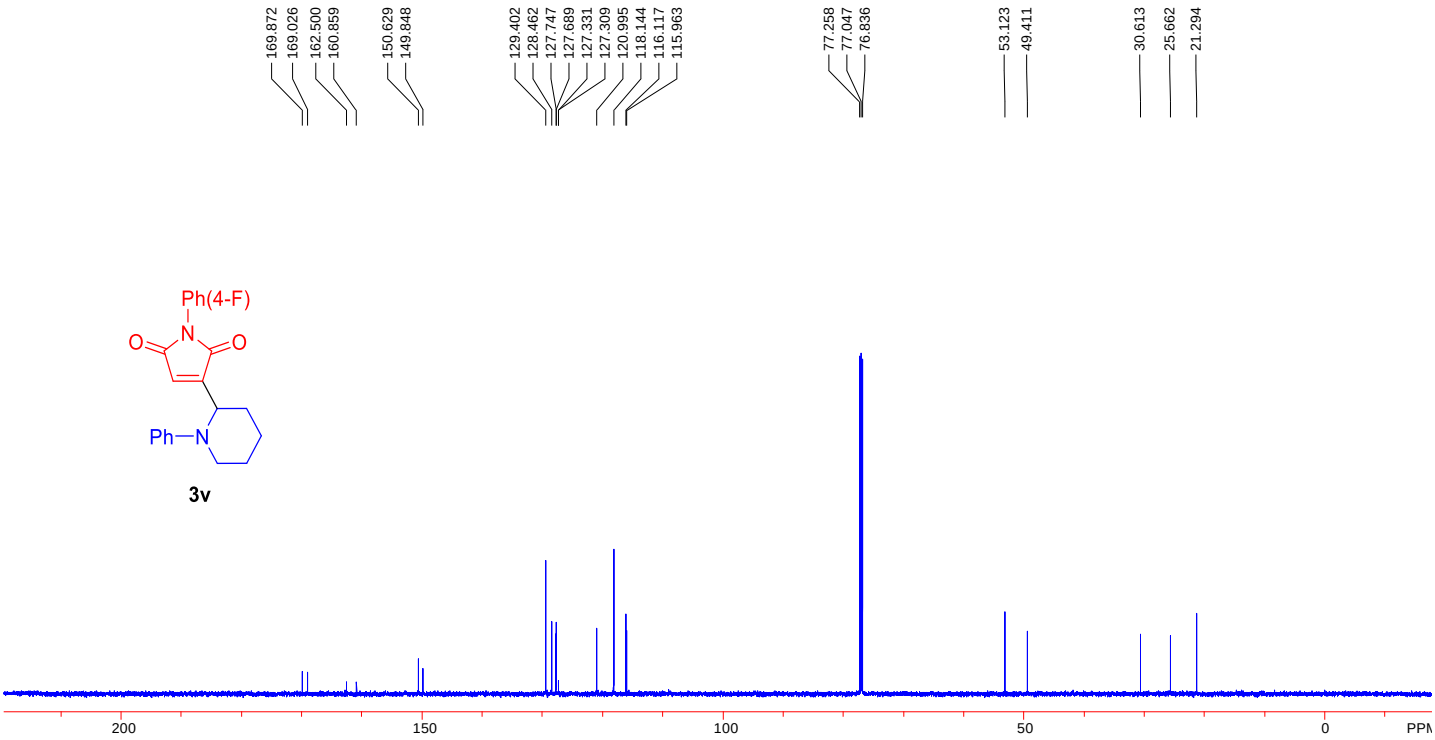
^{1}H NMR (600 MHz, $CDCl_3$) of **3u**



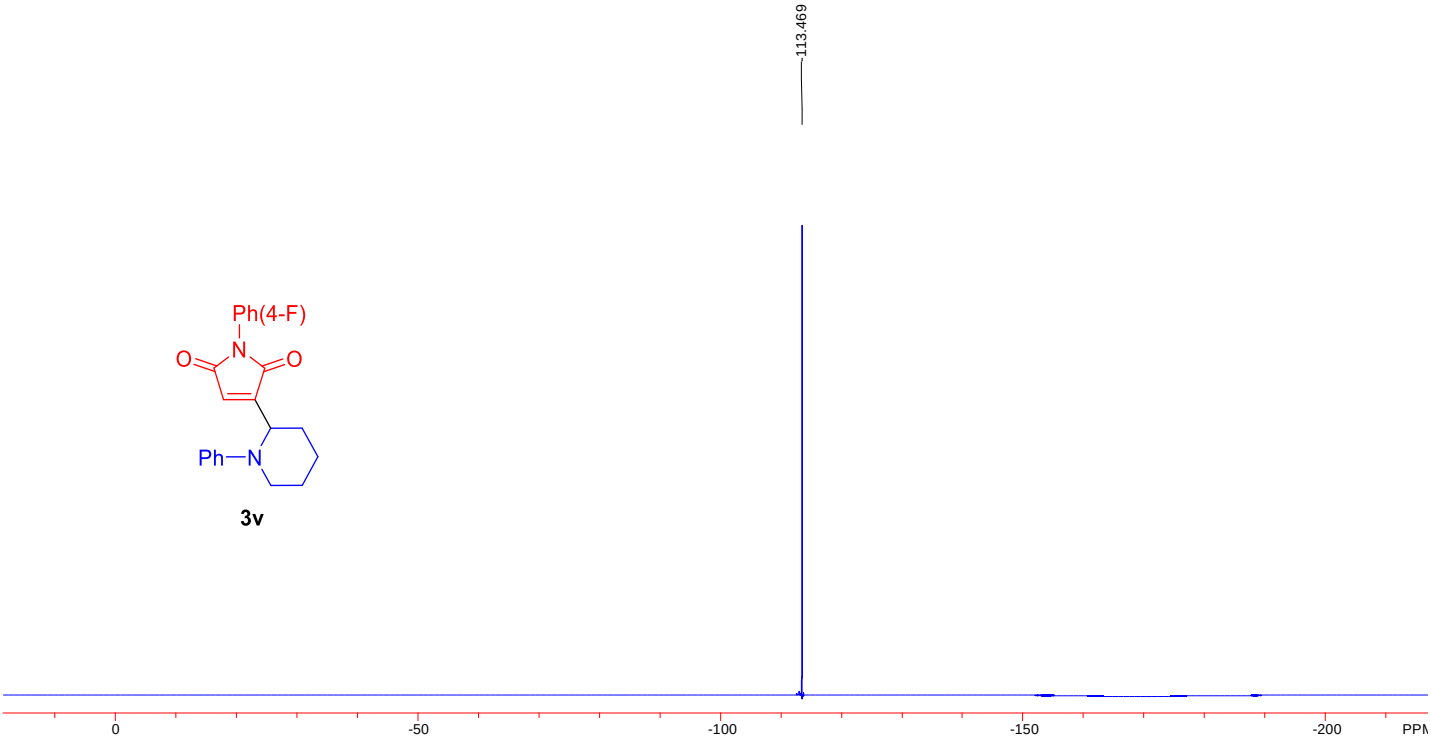
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3u**



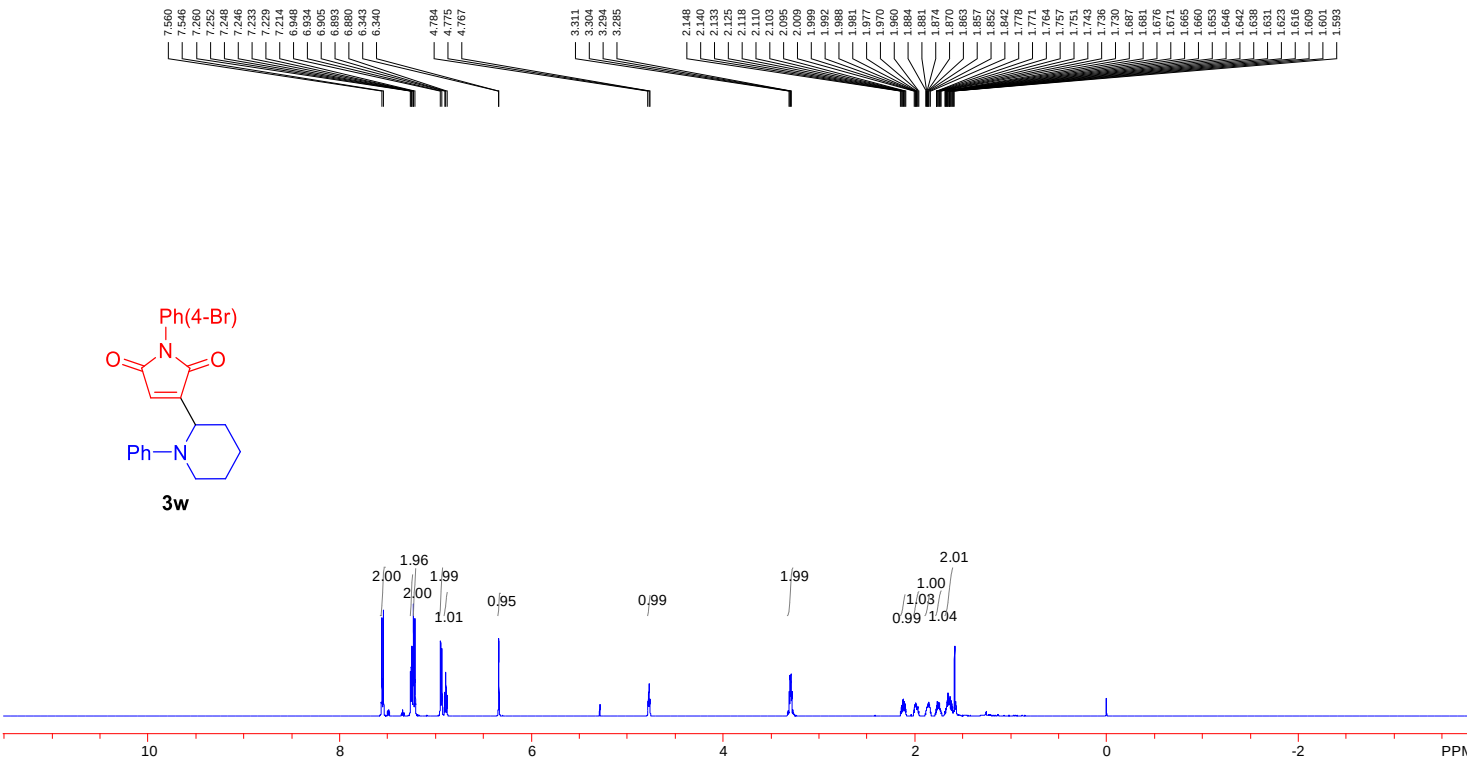
^{1}H NMR (600 MHz, $CDCl_3$) of **3v**



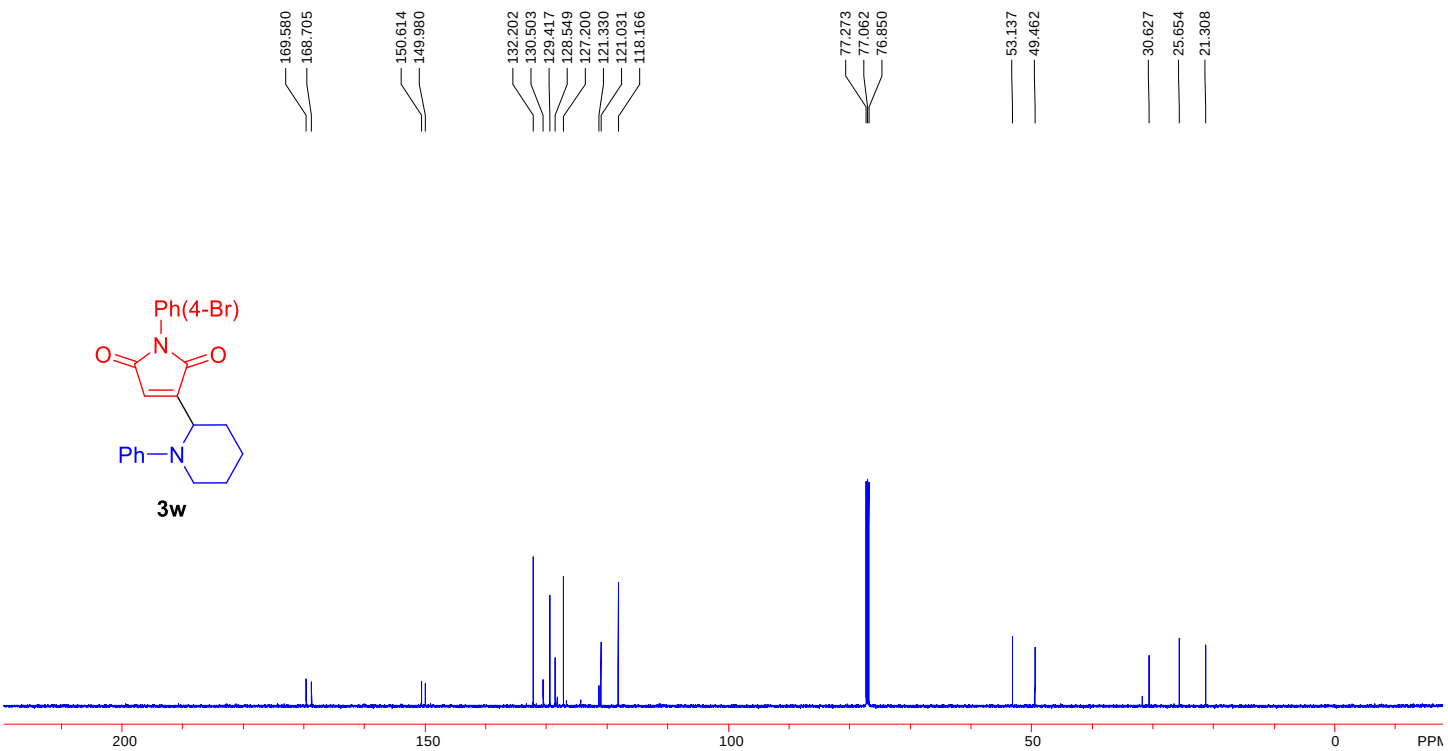
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3v**



$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) of **3v**

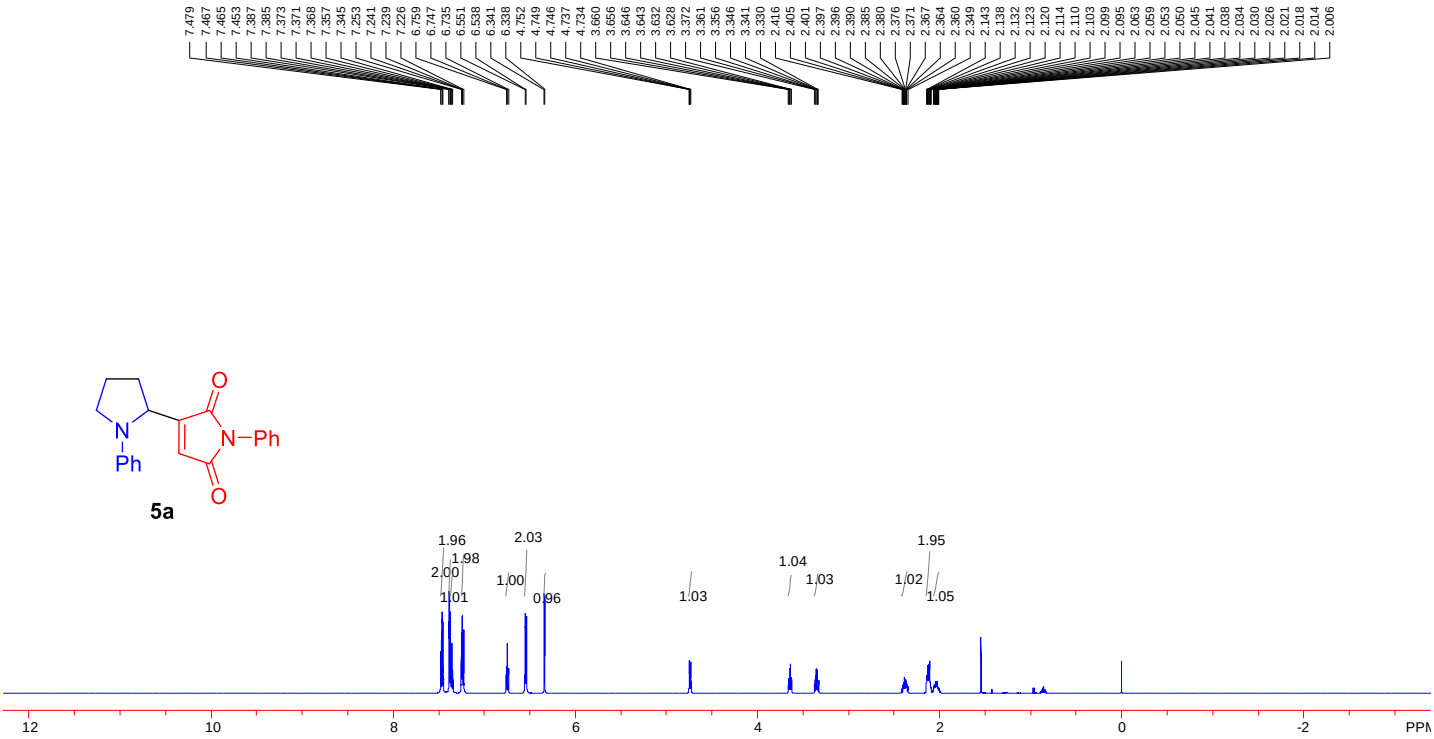


^{1}H NMR (600 MHz, $CDCl_3$) of **3w**

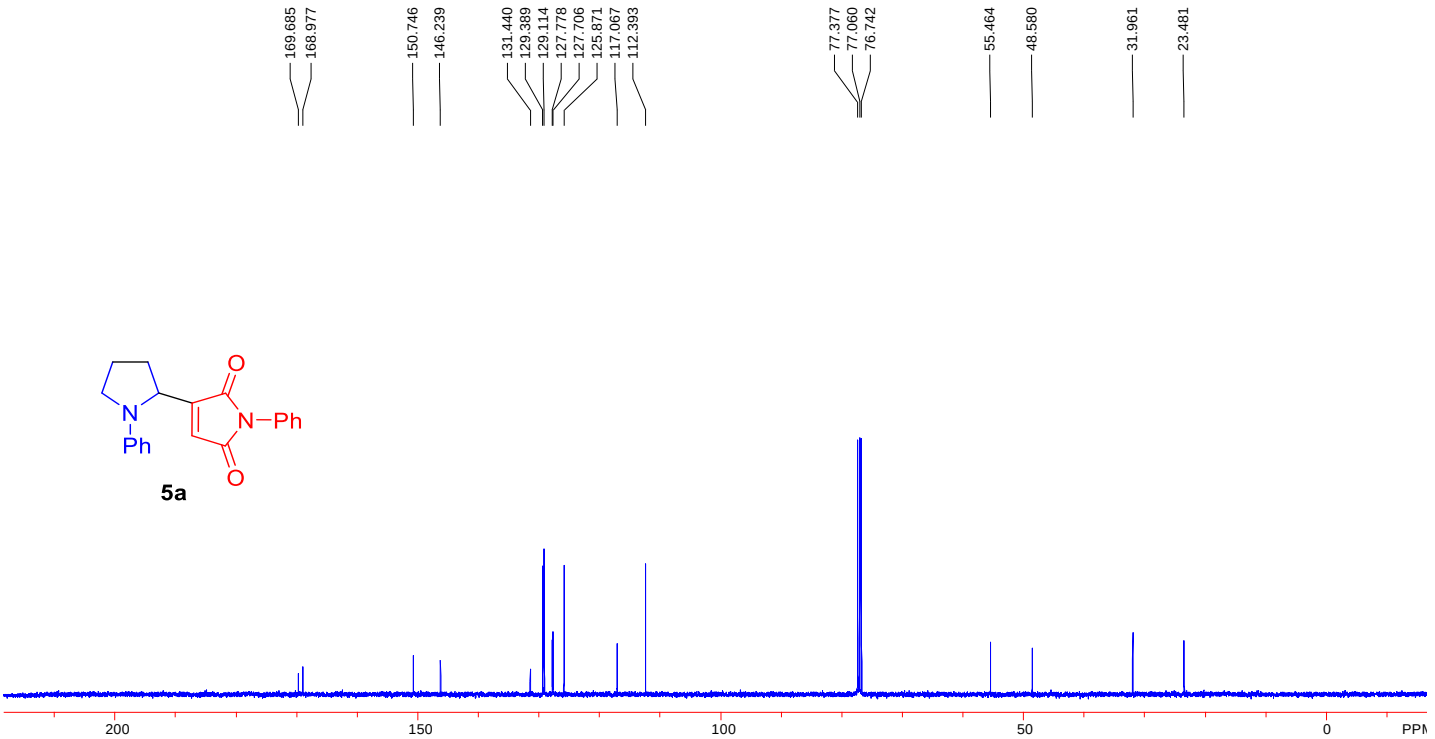


^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **3w**

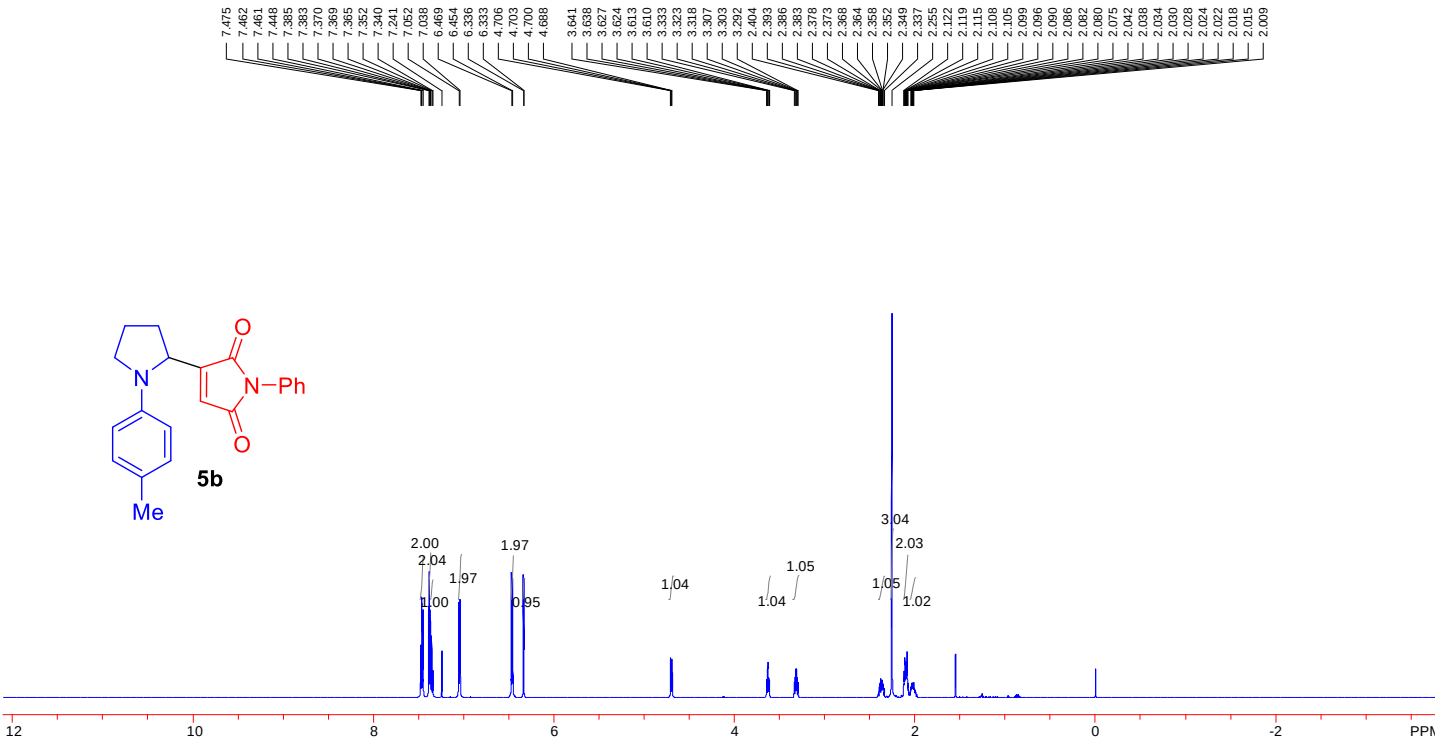
II. Copies of NMR Spectra of 5a-5g



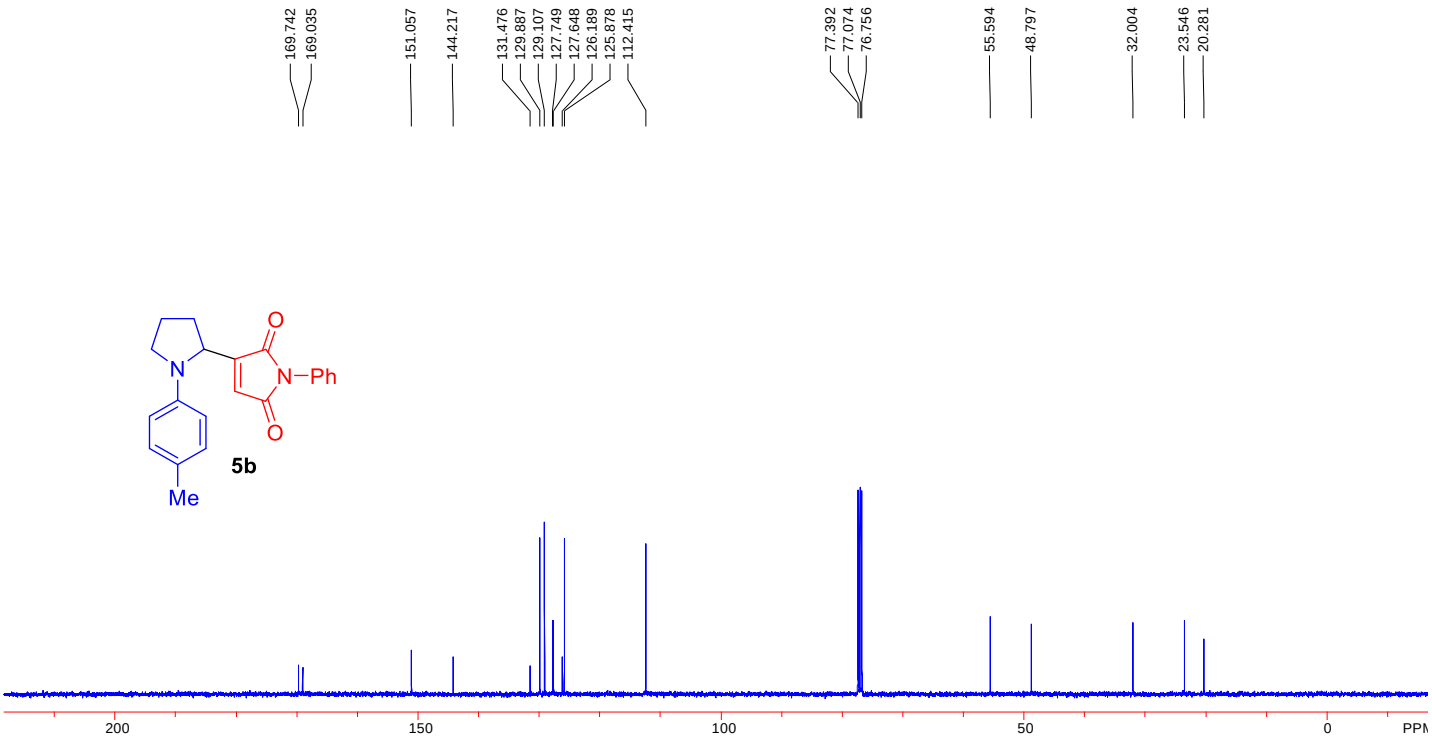
^{1}H NMR (600 MHz, $CDCl_3$) of **5a**



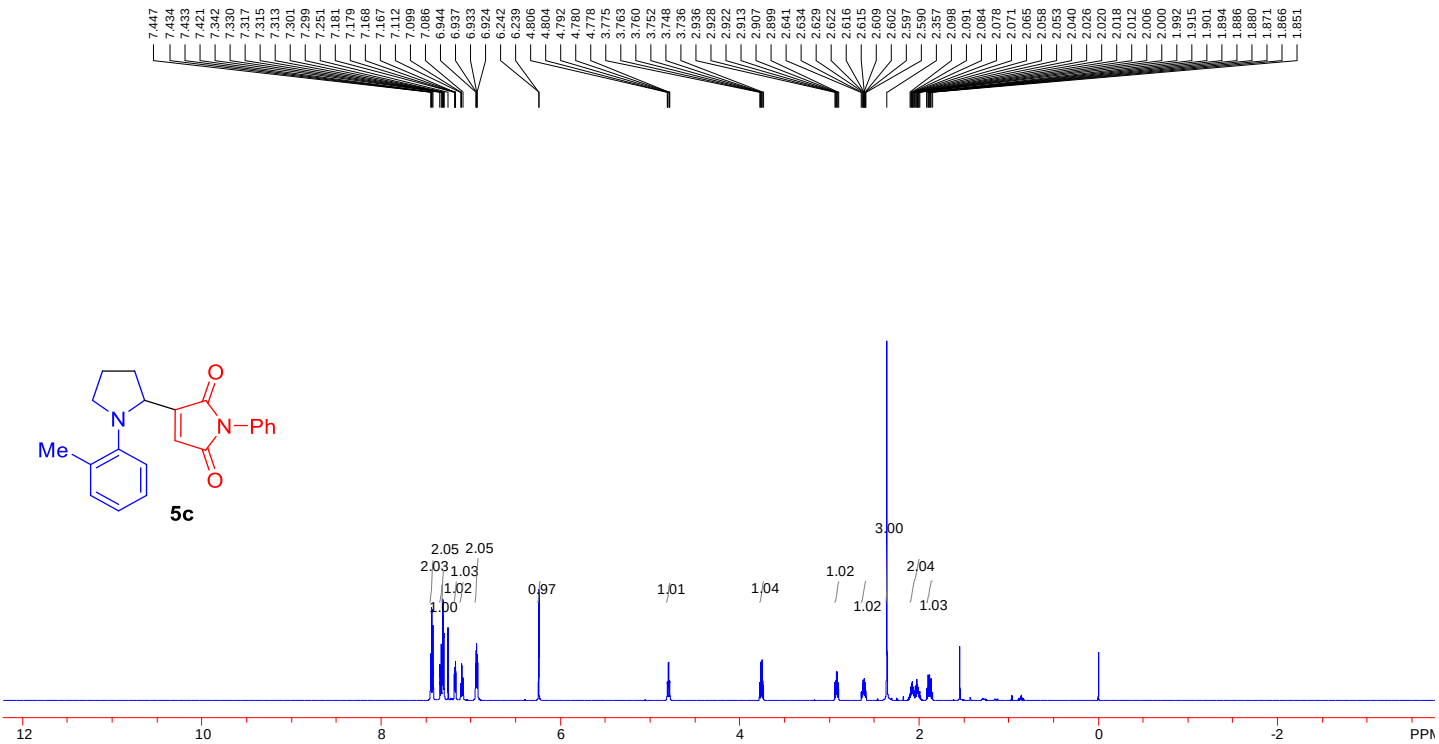
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **5a**



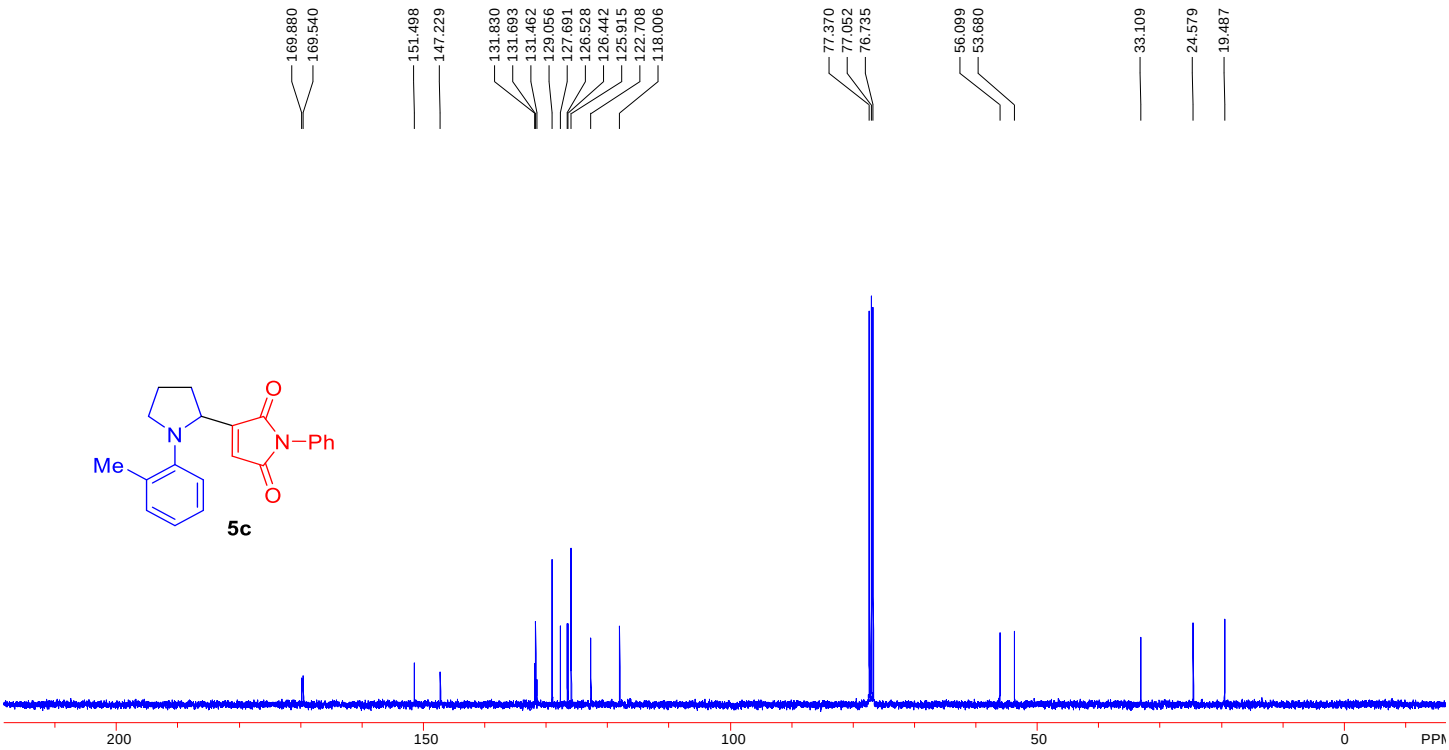
^{1}H NMR (600 MHz, $CDCl_3$) of **5b**



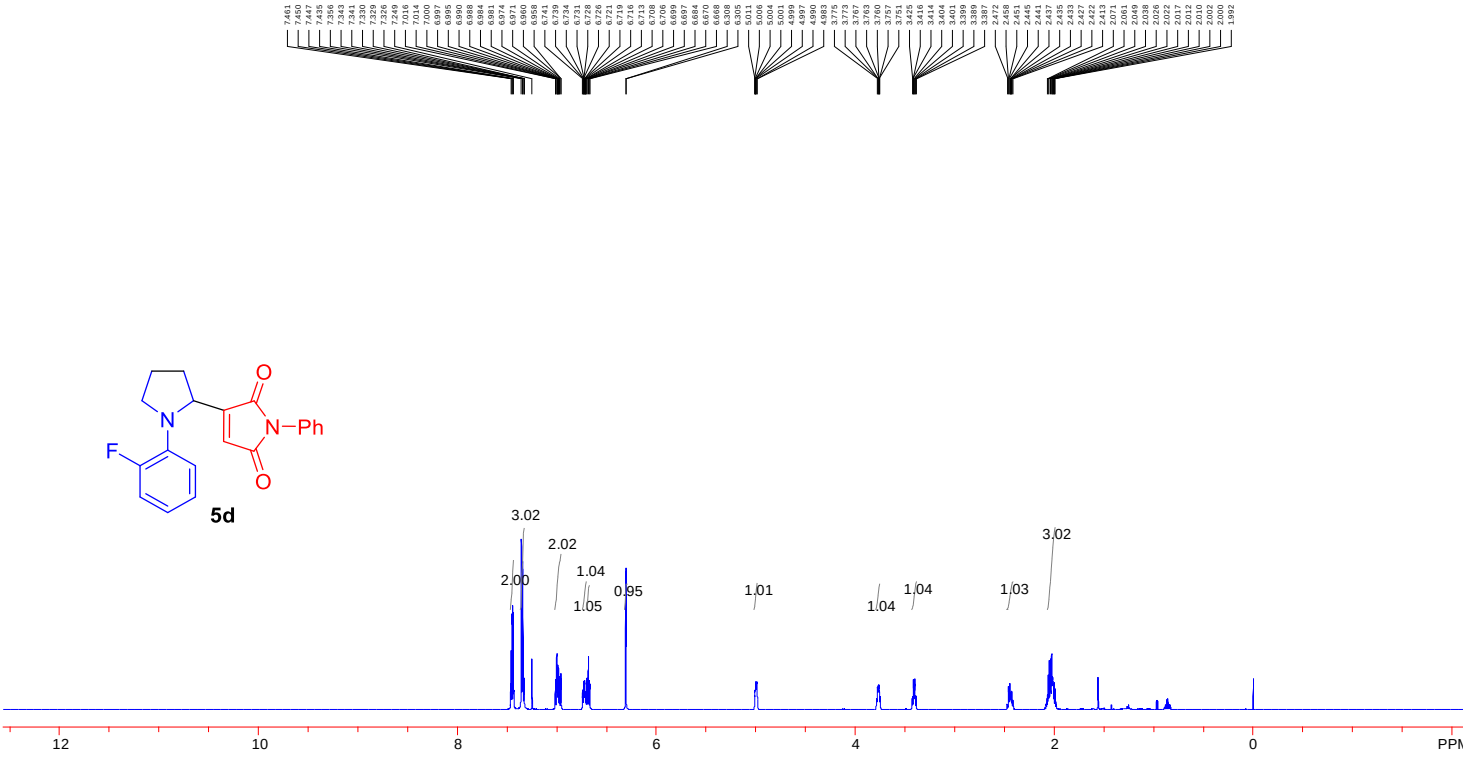
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **5b**



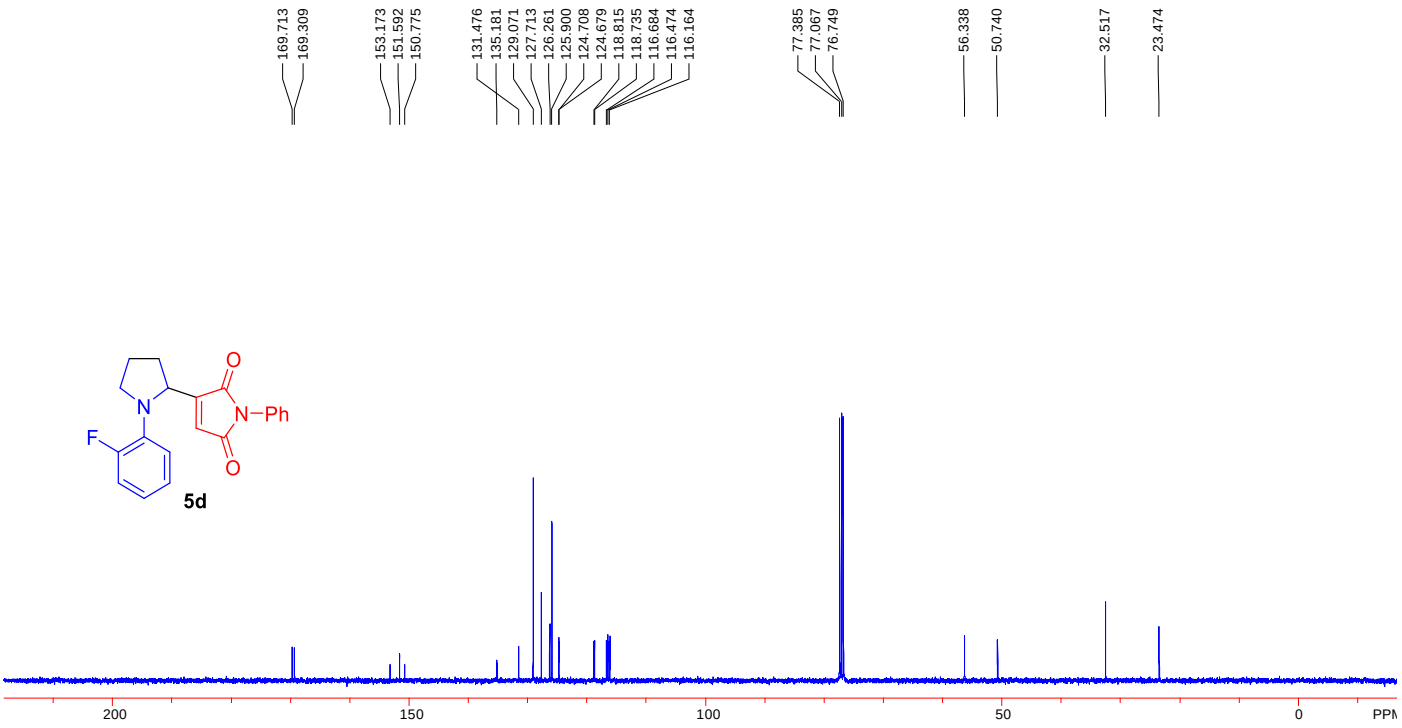
^{1}H NMR (600 MHz, $CDCl_3$) of **5c**



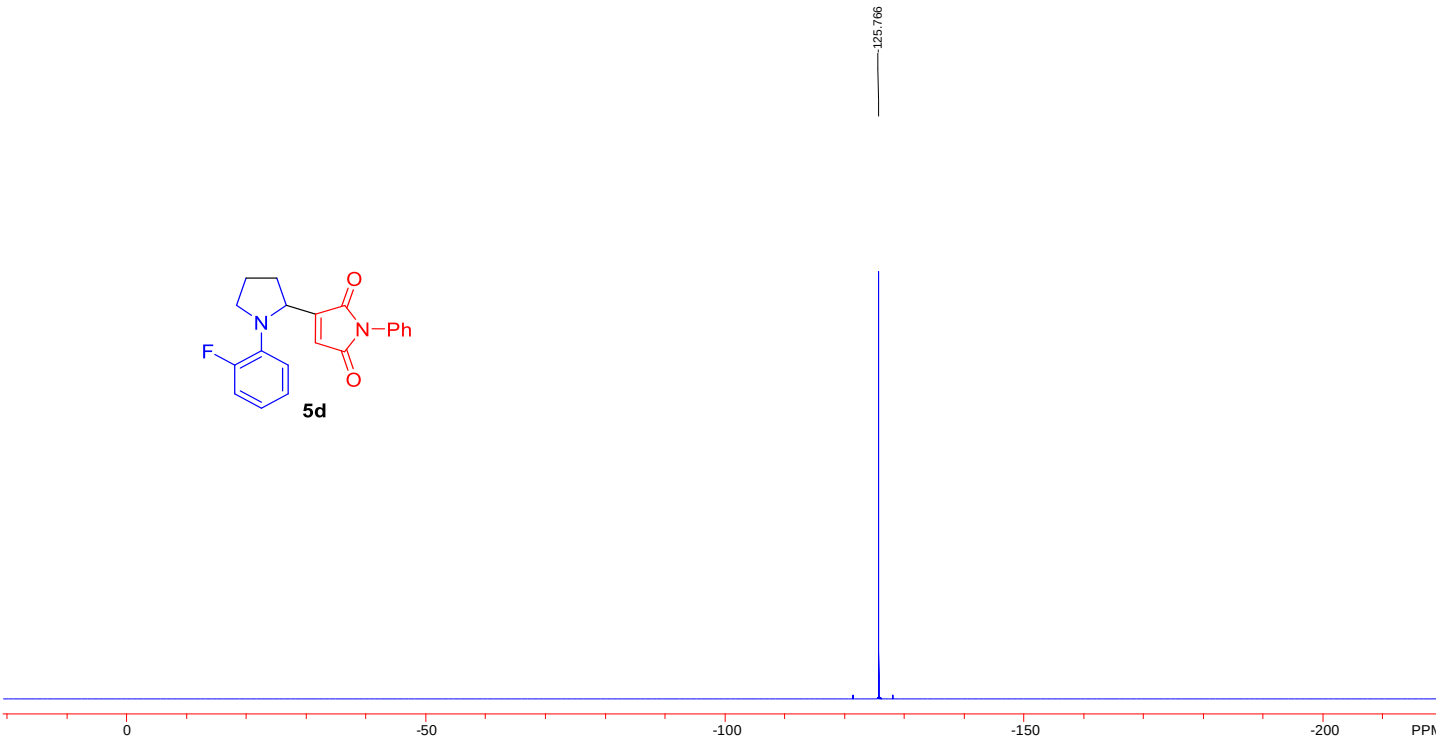
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **5c**



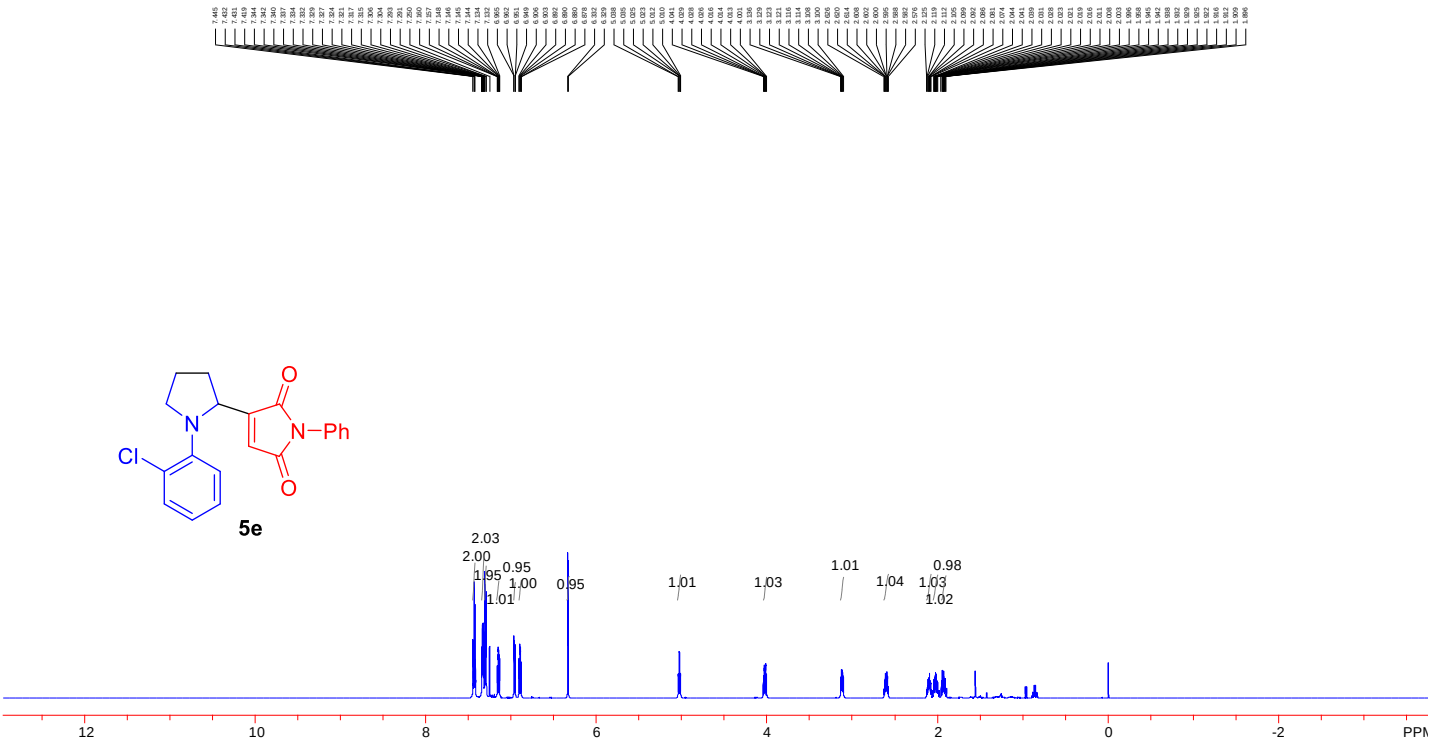
^{1}H NMR (600 MHz, $CDCl_3$) of **5d**



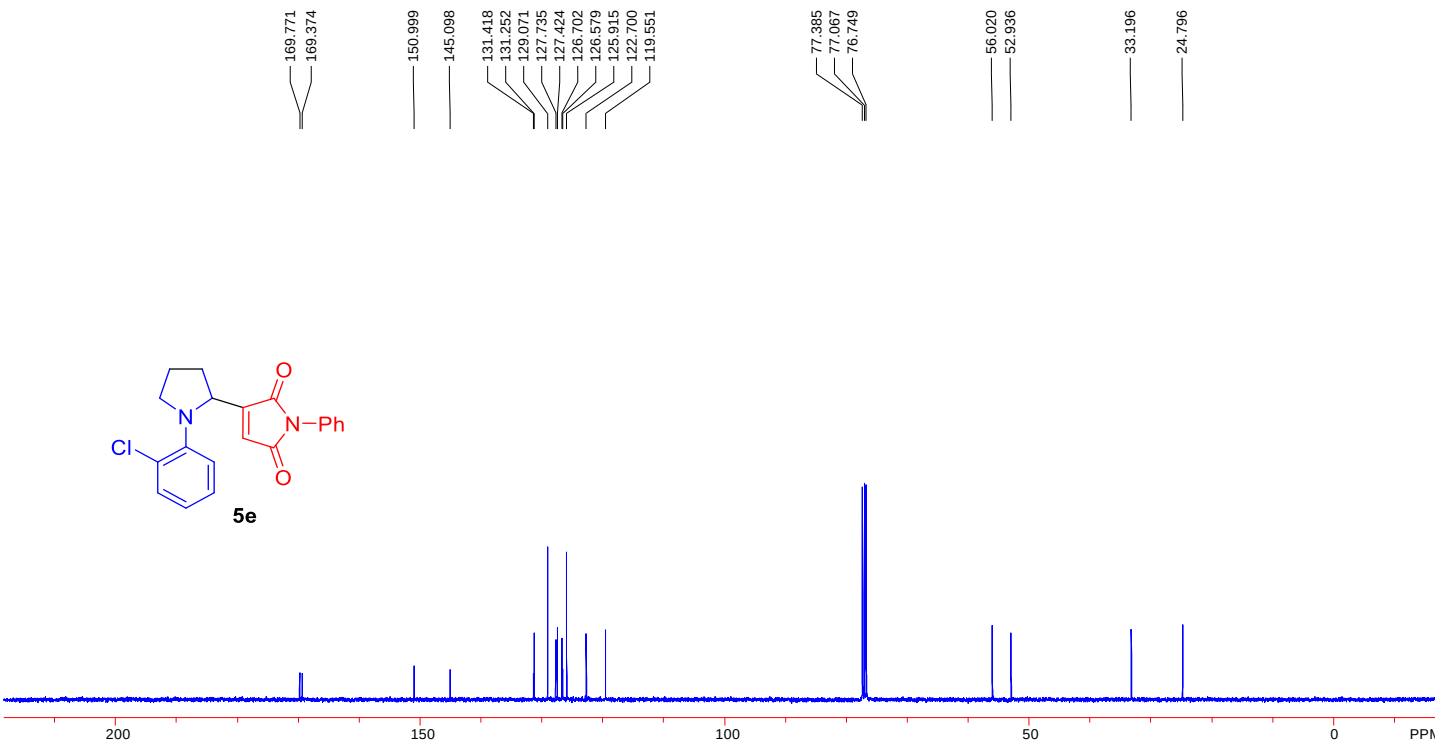
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **5d**



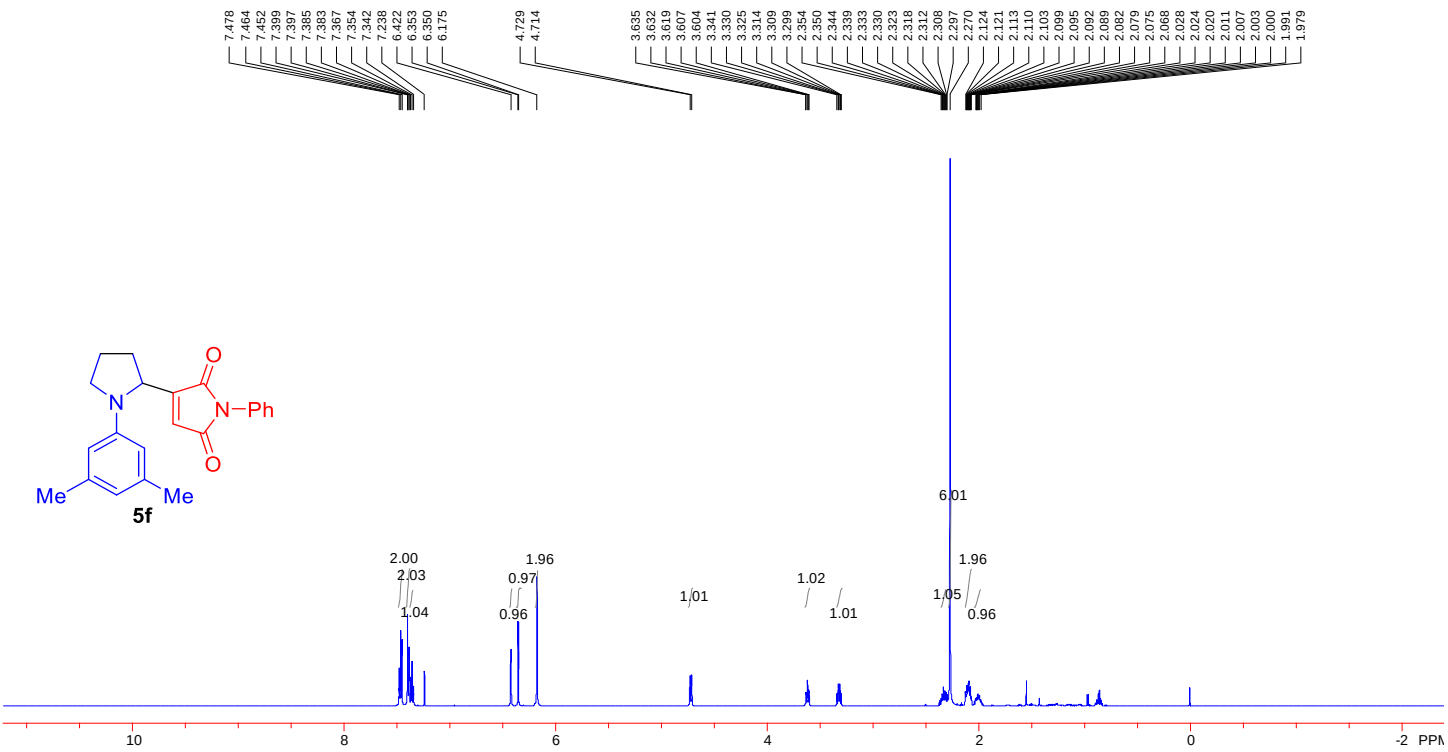
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) of **5d**



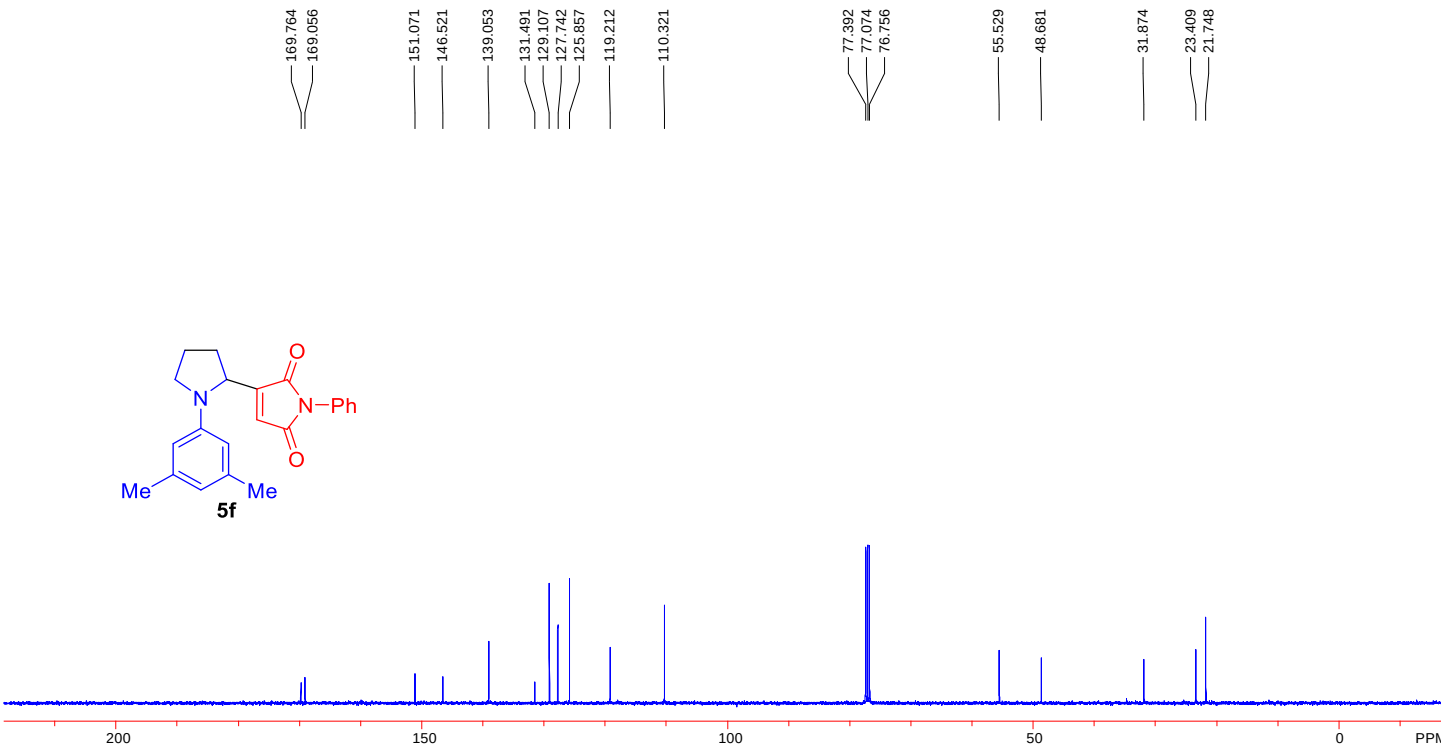
^{1}H NMR (600 MHz, $CDCl_3$) of **5e**



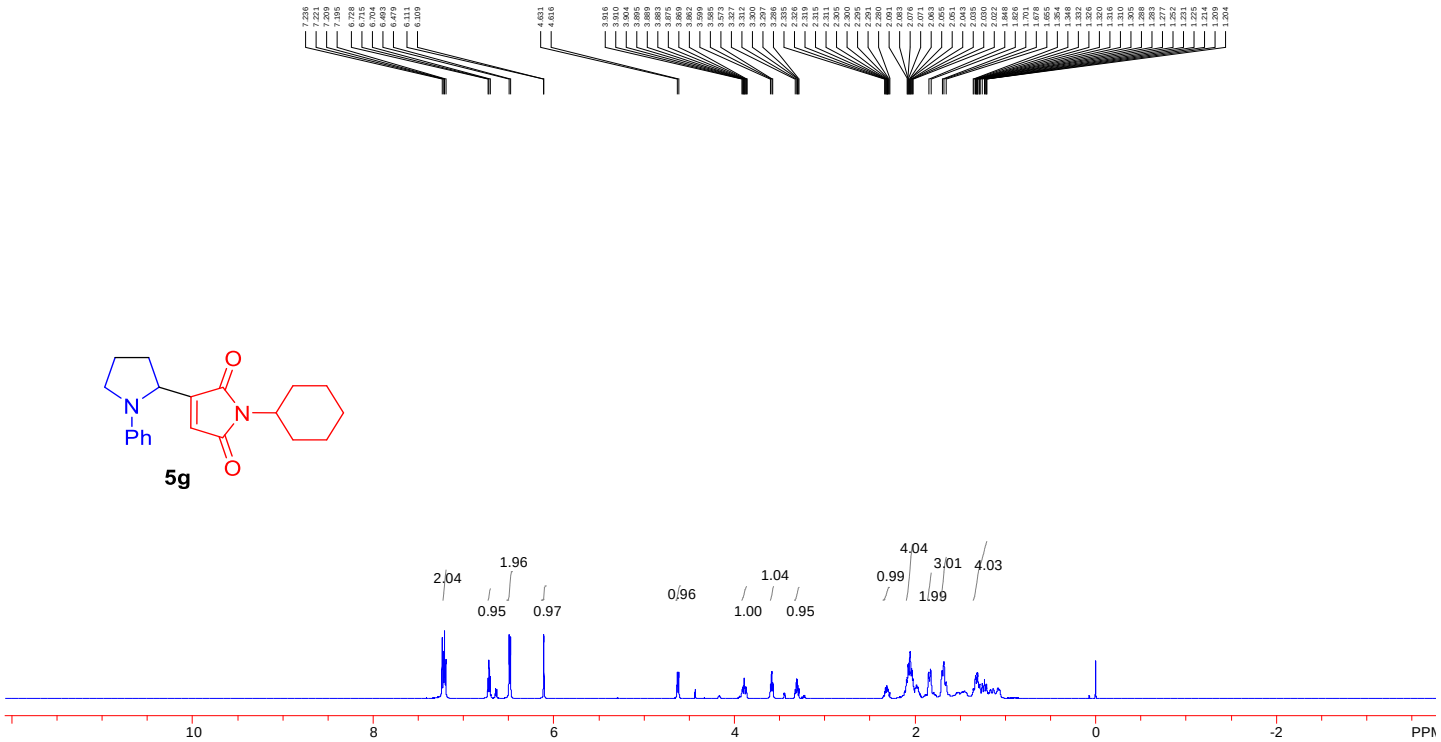
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **5e**



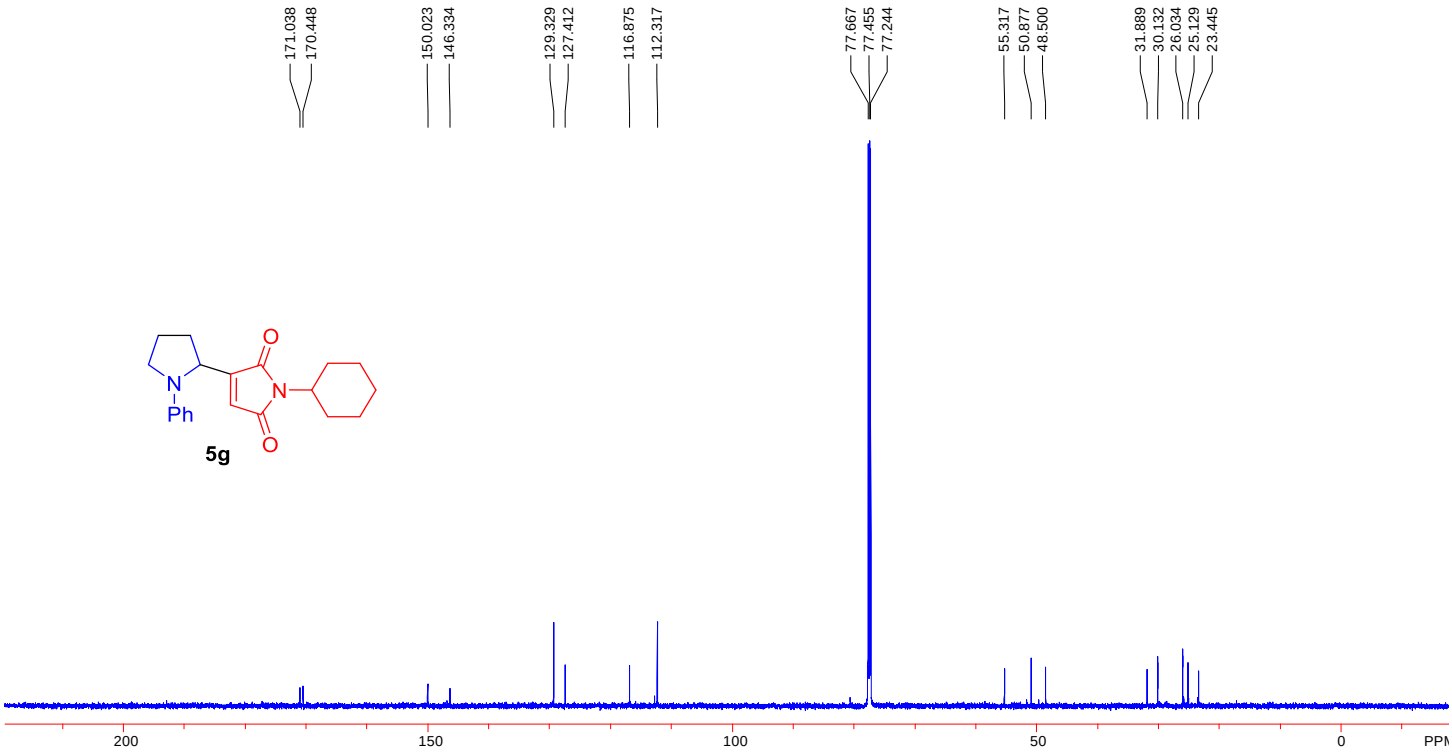
^{1}H NMR (600 MHz, $CDCl_3$) of **5f**



^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **5f**

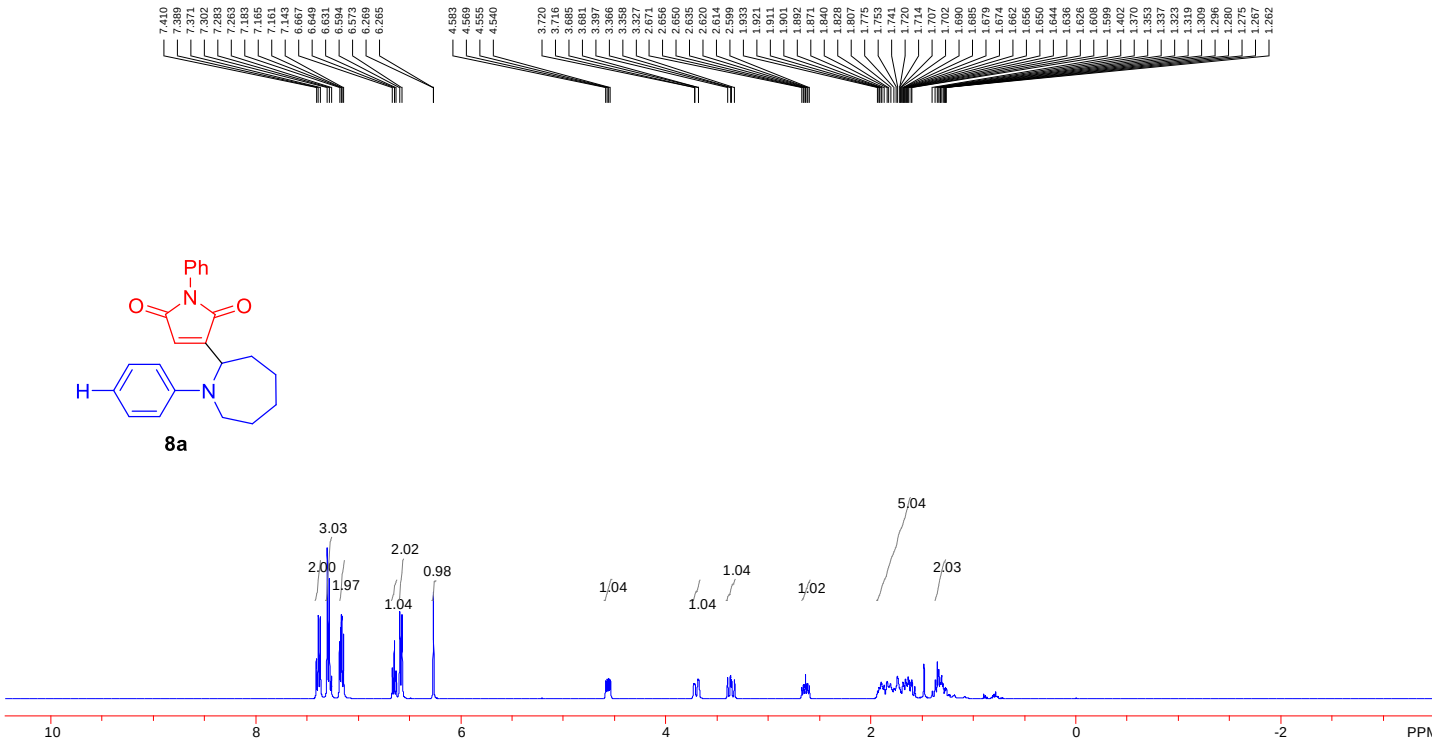


^{1}H NMR (600 MHz, $CDCl_3$) of **5g**

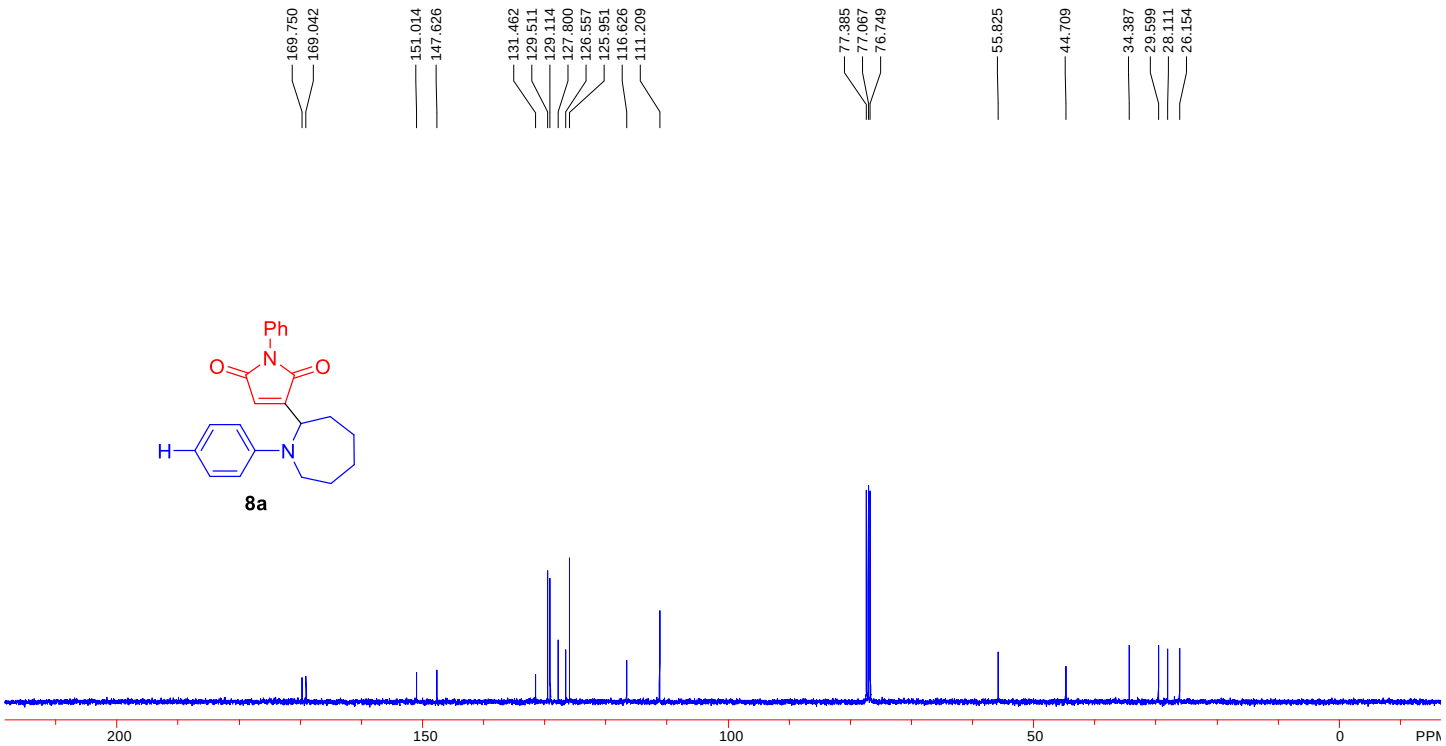


^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **5g**

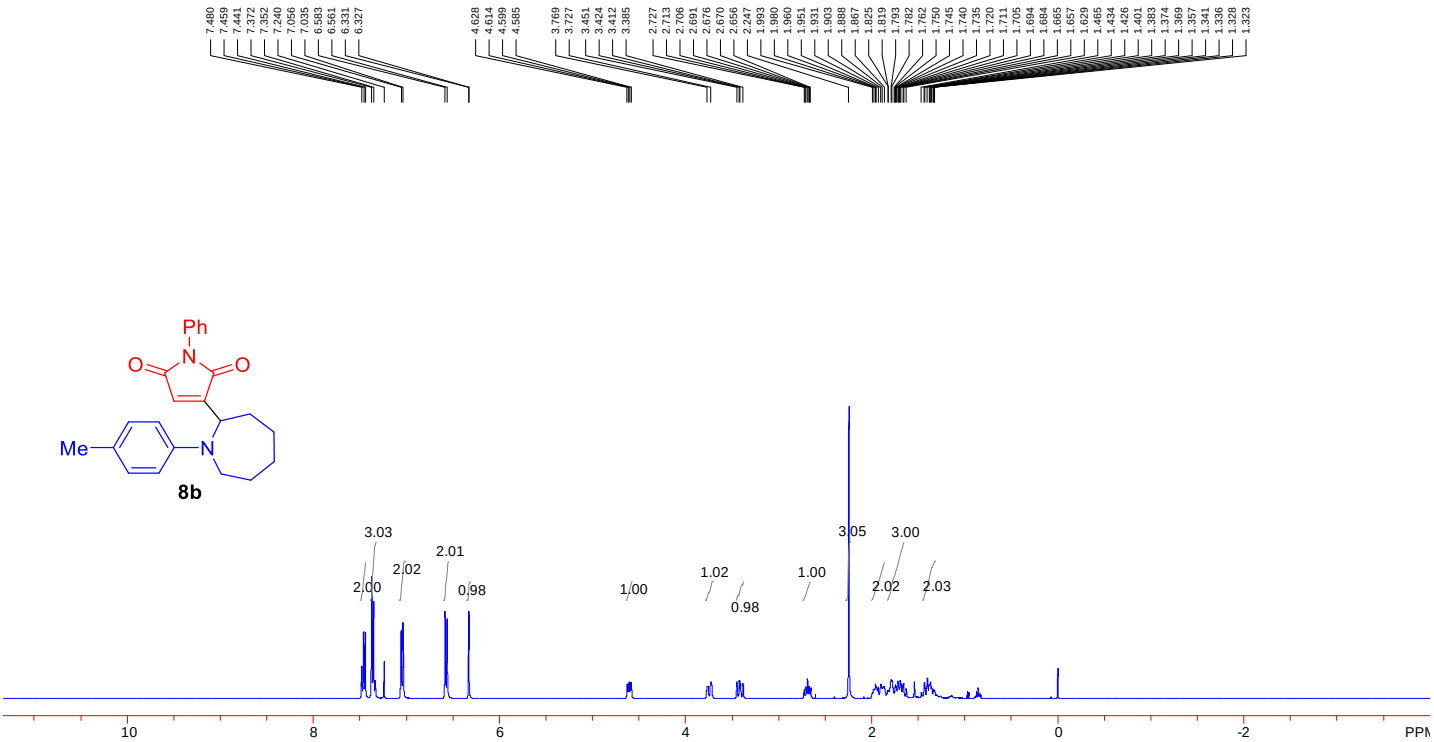
III. Copies of NMR Spectra of 8a-8m



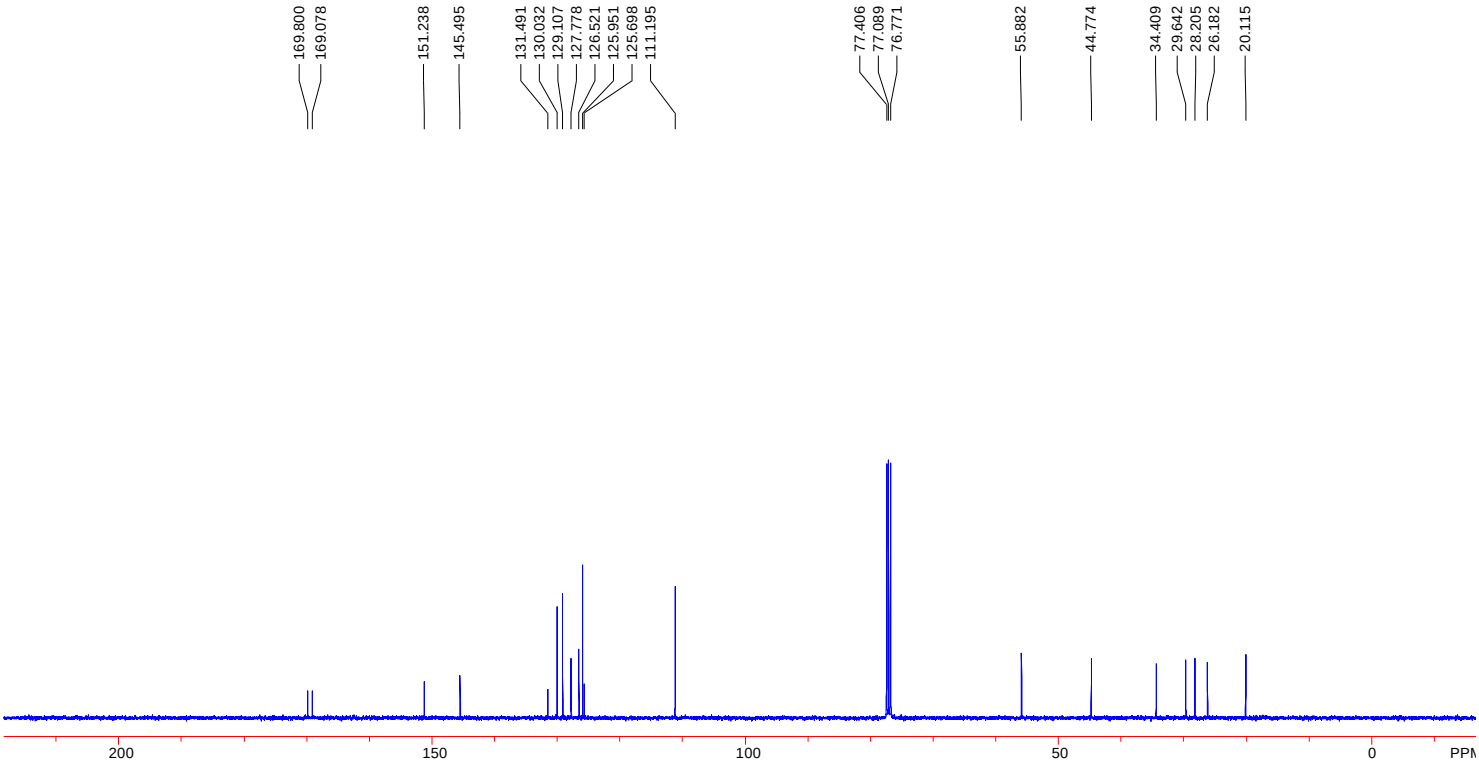
^{1}H NMR (400 MHz, $CDCl_3$) of **8a**



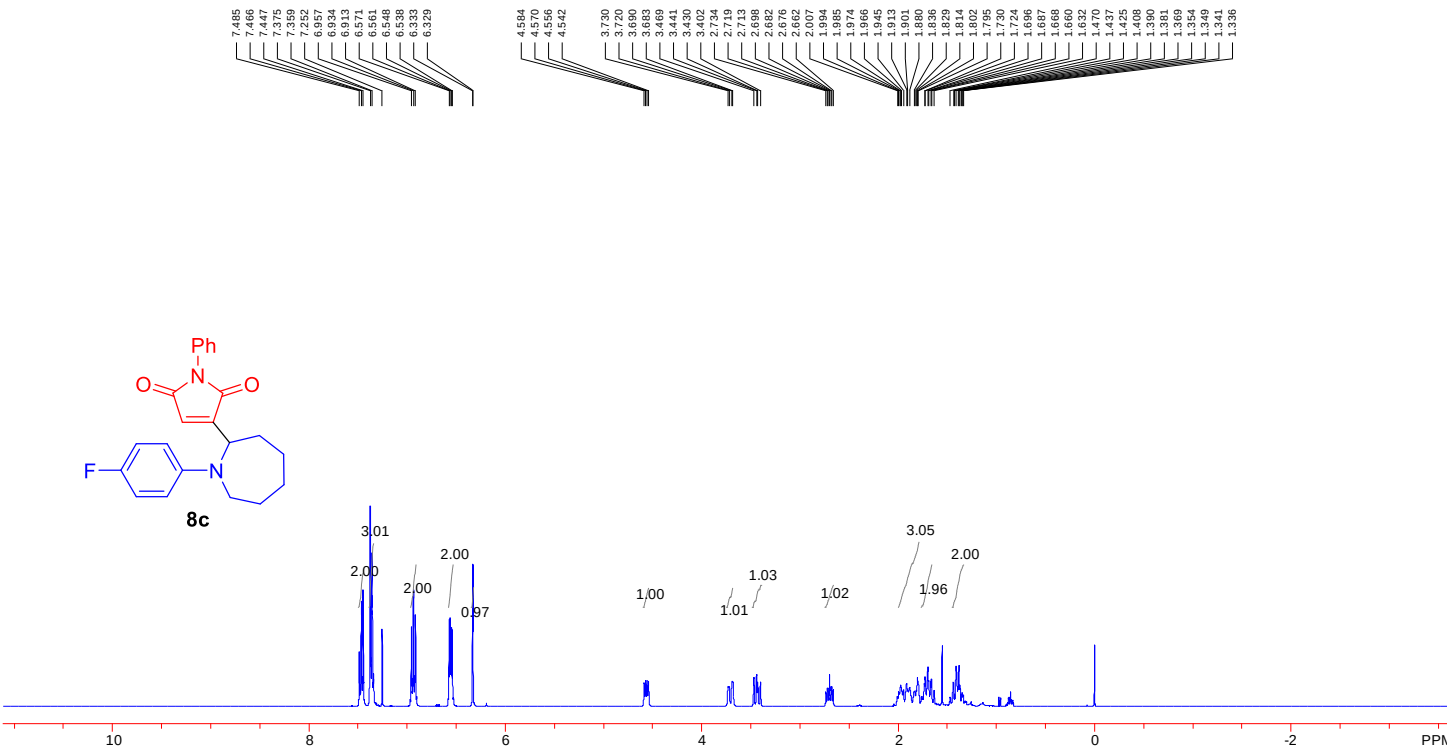
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8a**



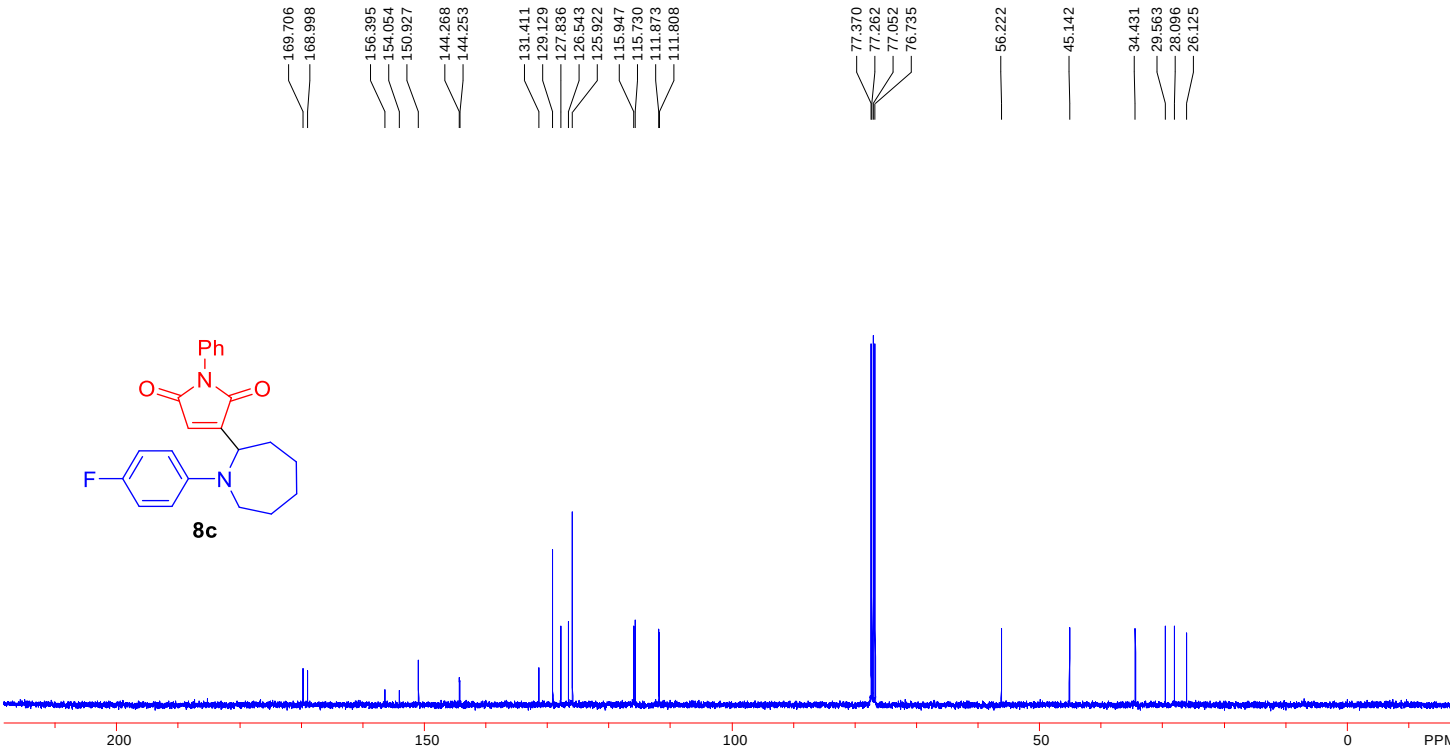
^{1}H NMR (400 MHz, $CDCl_3$) of **8b**



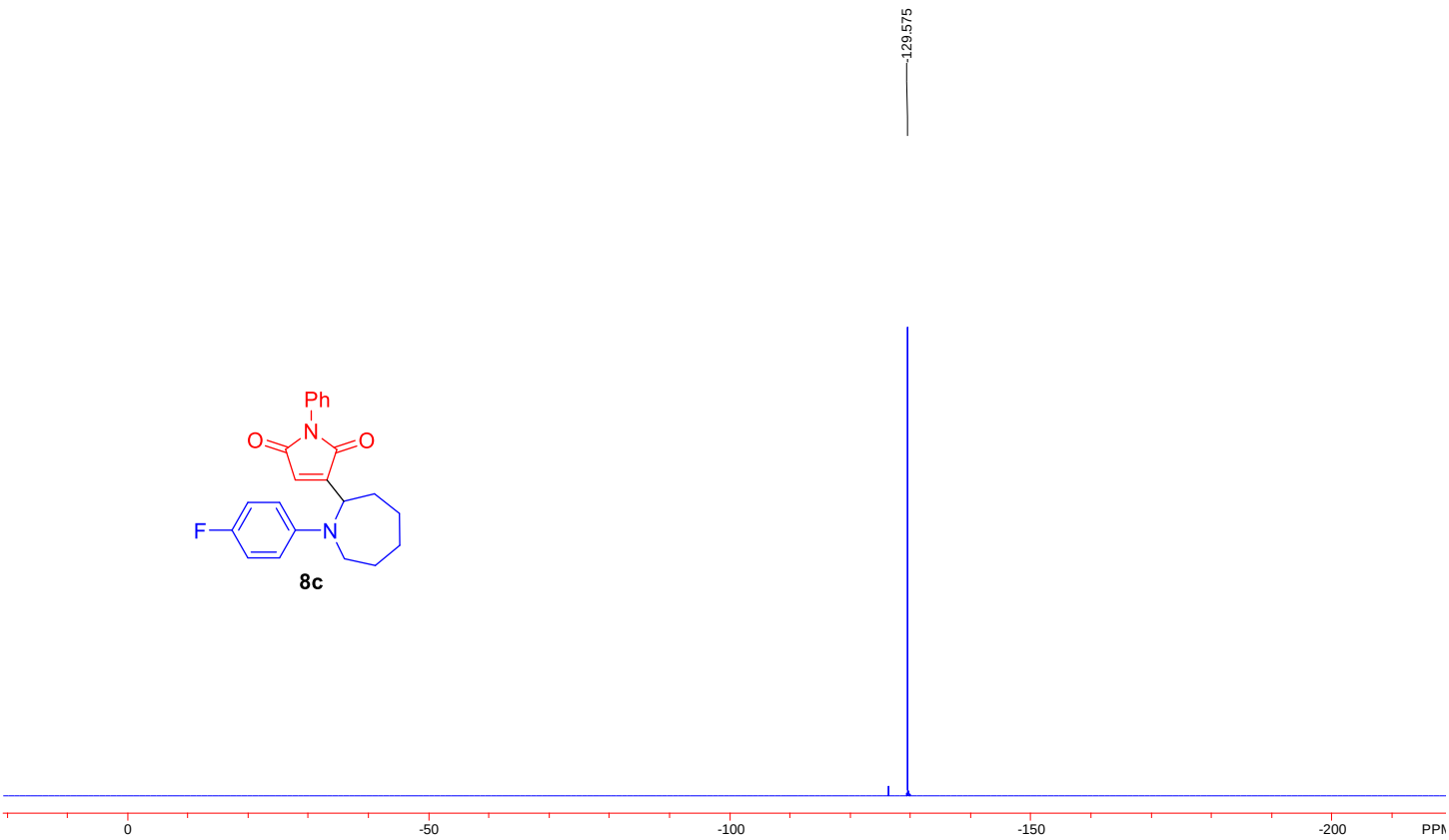
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8b**



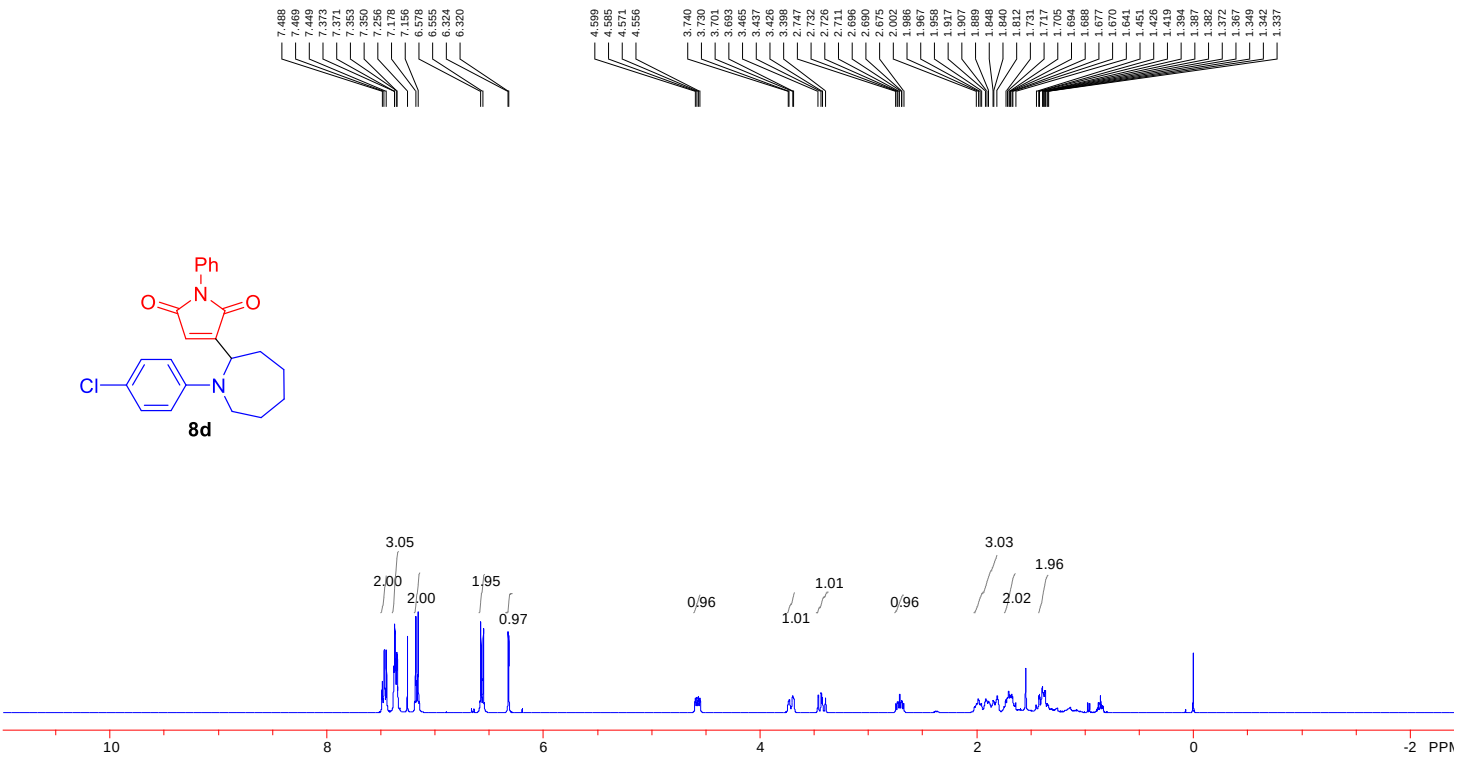
^{1}H NMR (400 MHz, $CDCl_3$) of **8c**



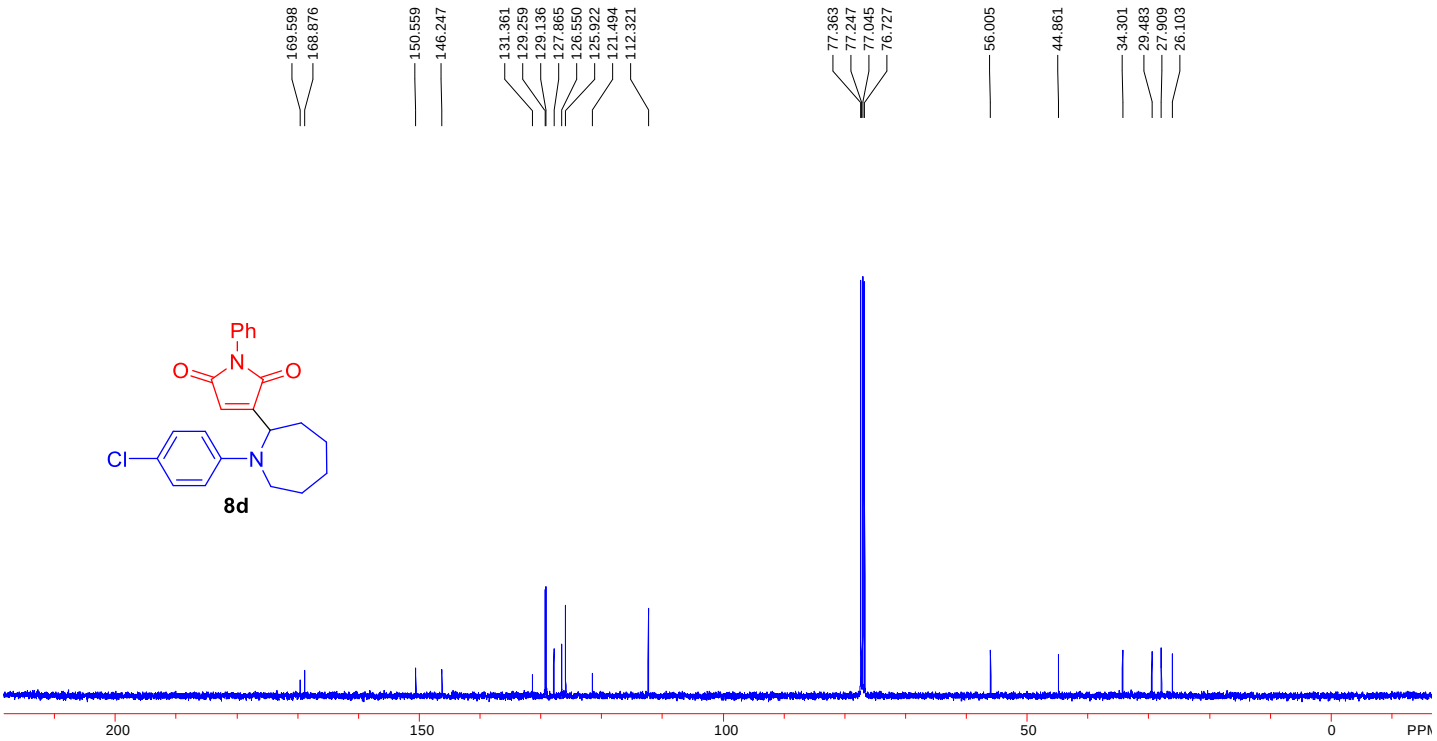
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8c**



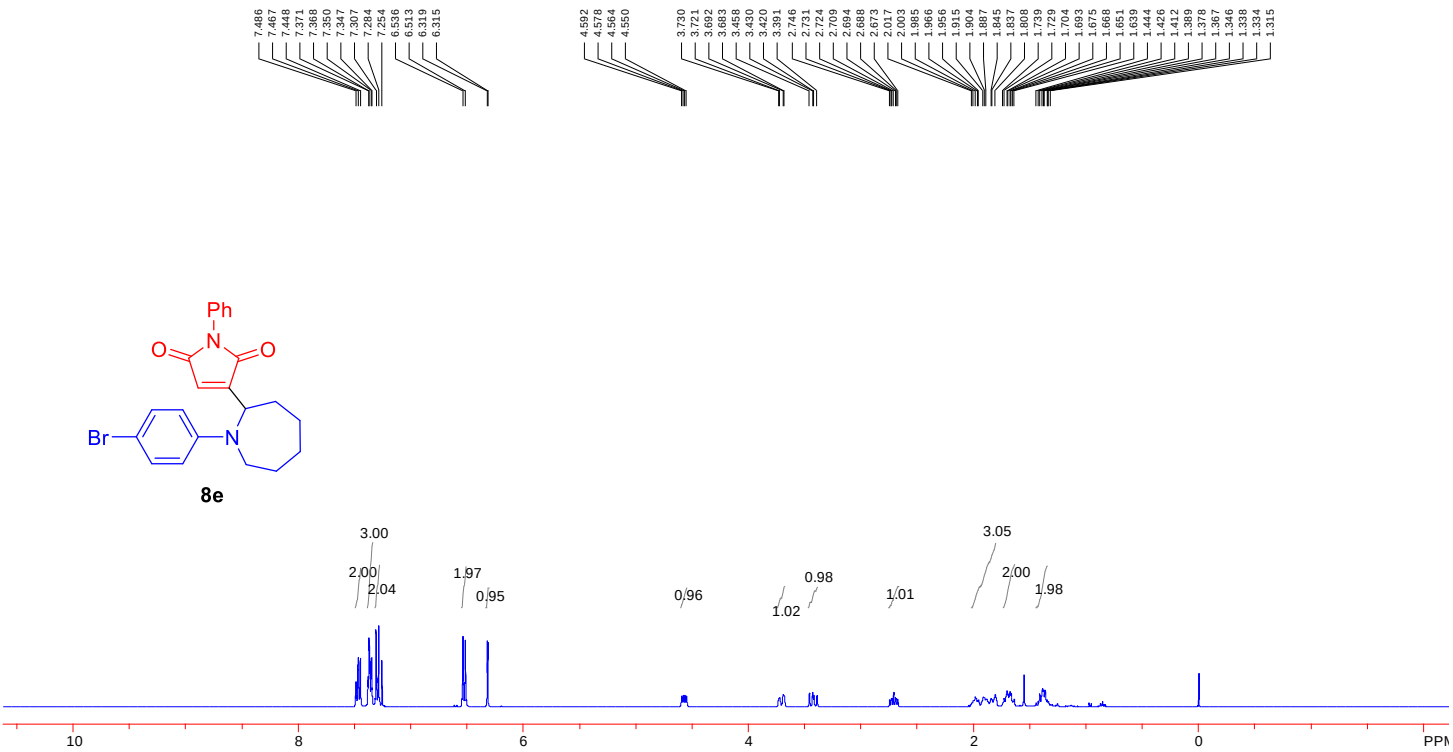
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) of **8c**



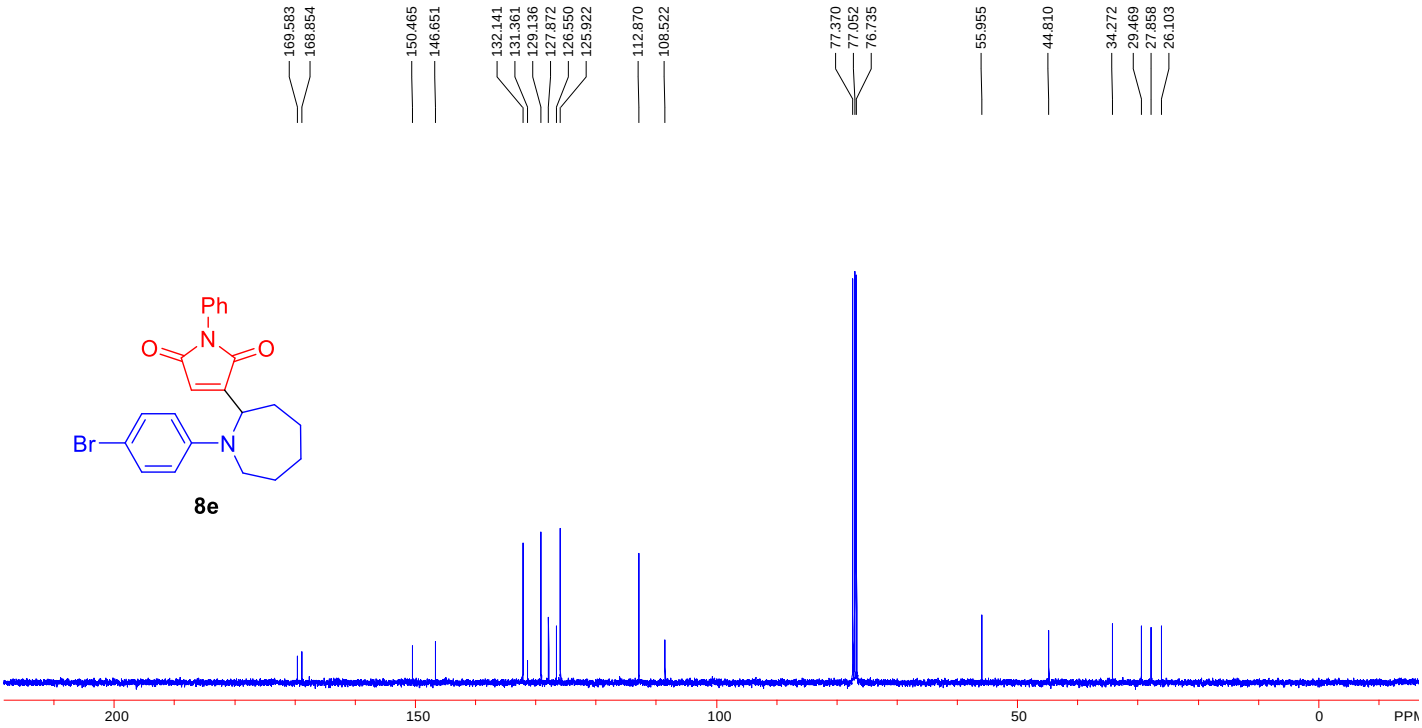
^{1}H NMR (400 MHz, $CDCl_3$) of **8d**



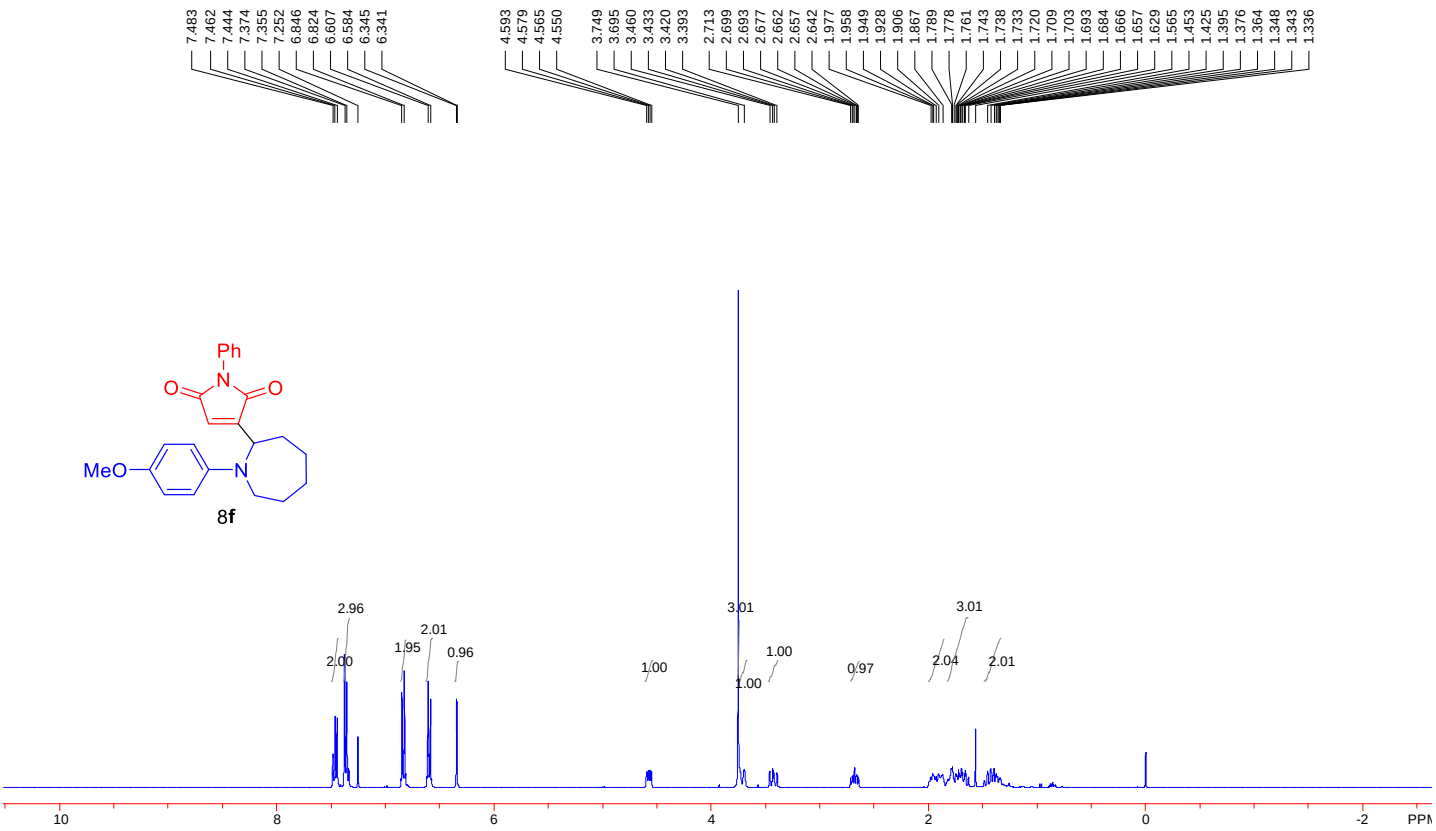
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8d**



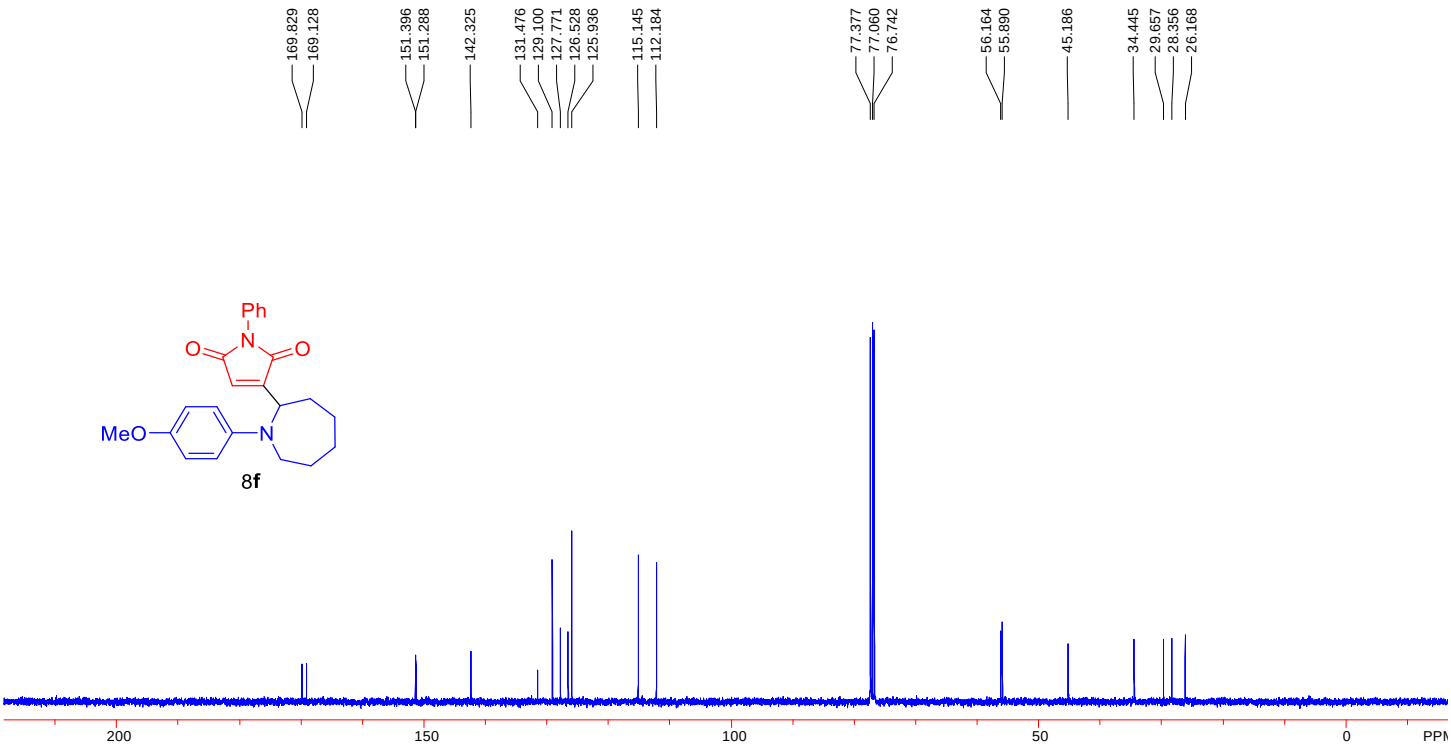
^{1}H NMR (400 MHz, $CDCl_3$) of **8e**



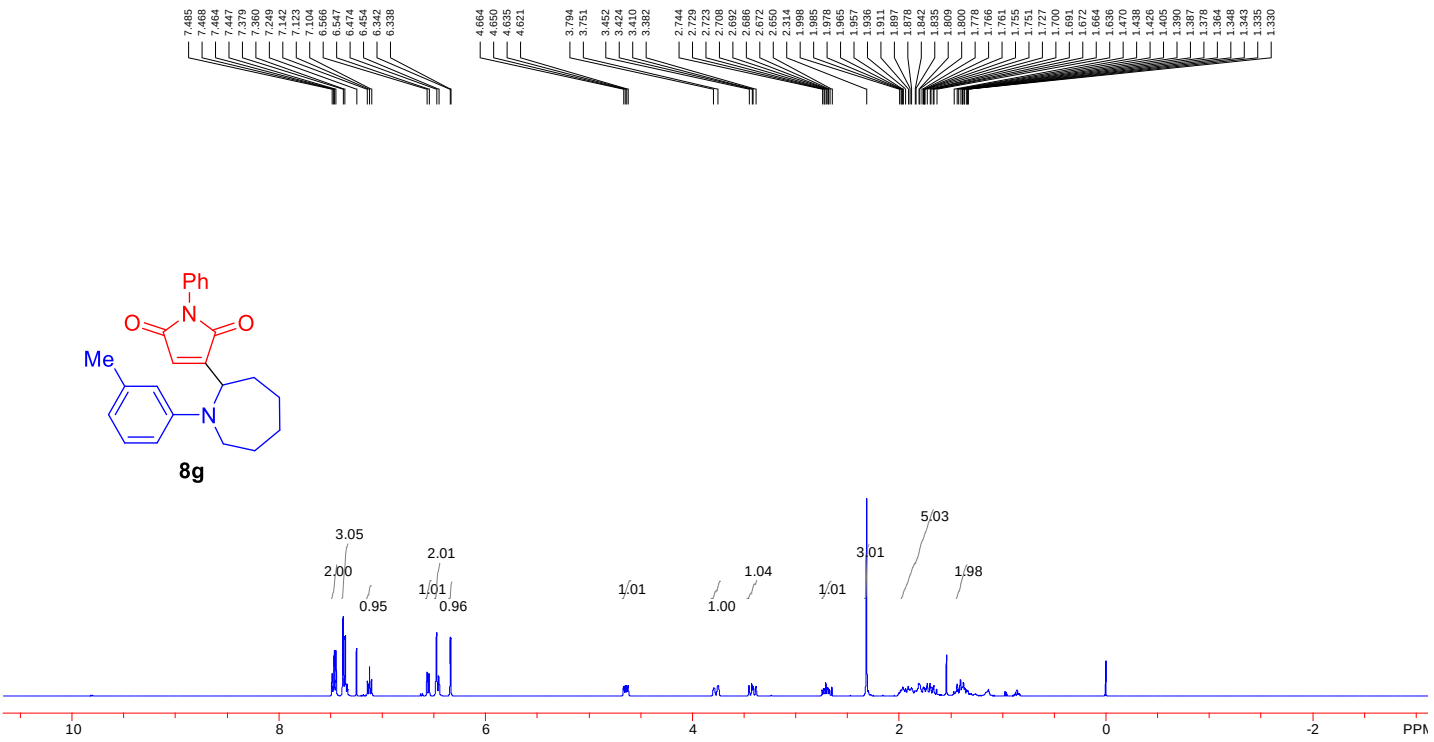
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8e**



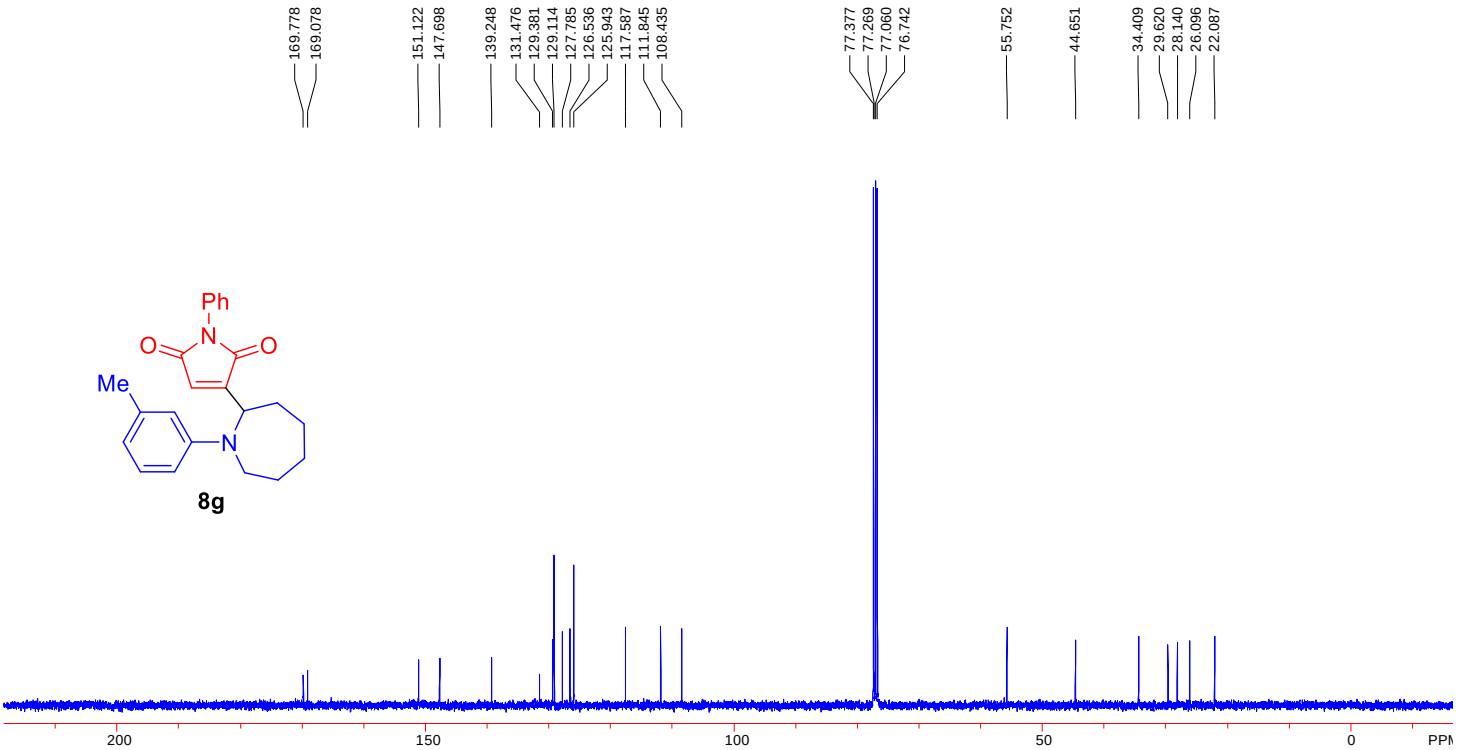
^{1}H NMR (400 MHz, $CDCl_3$) of **8f**



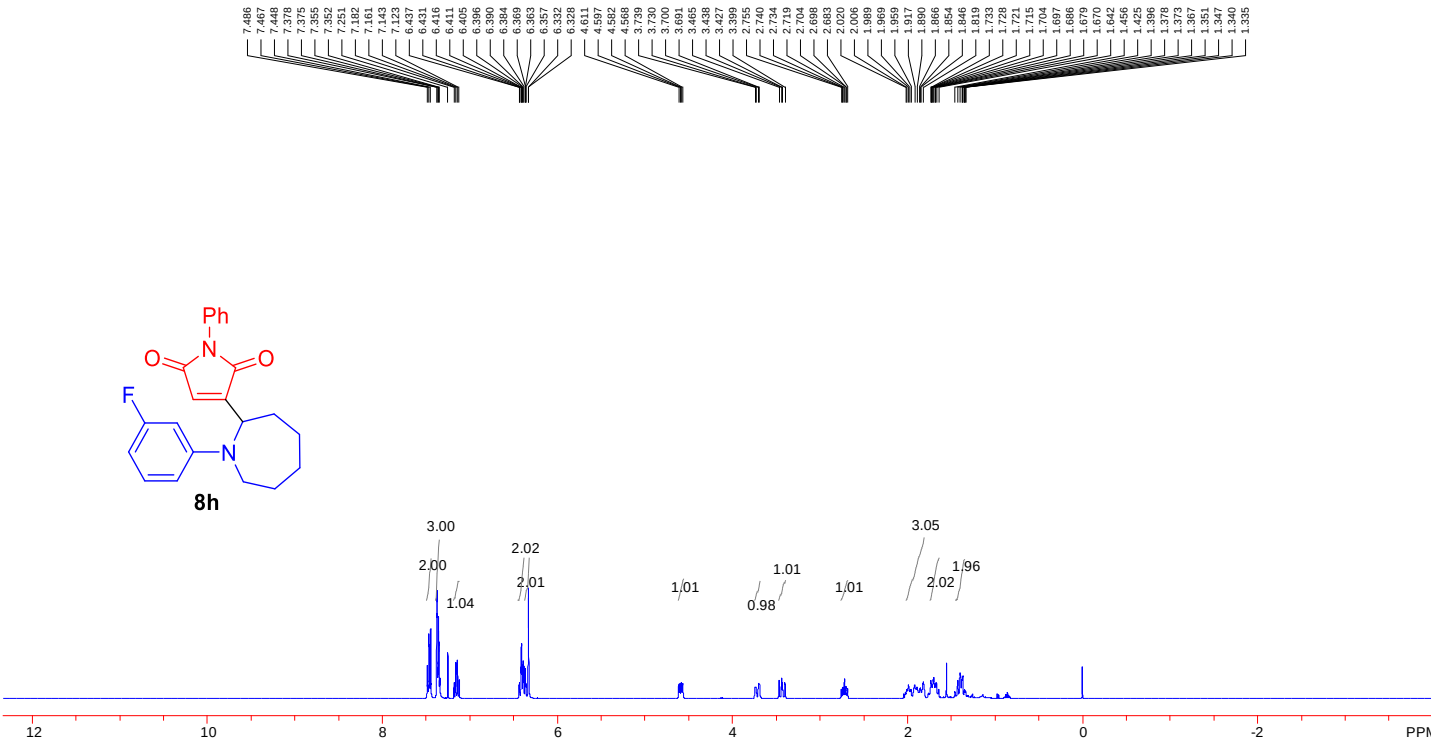
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8f**



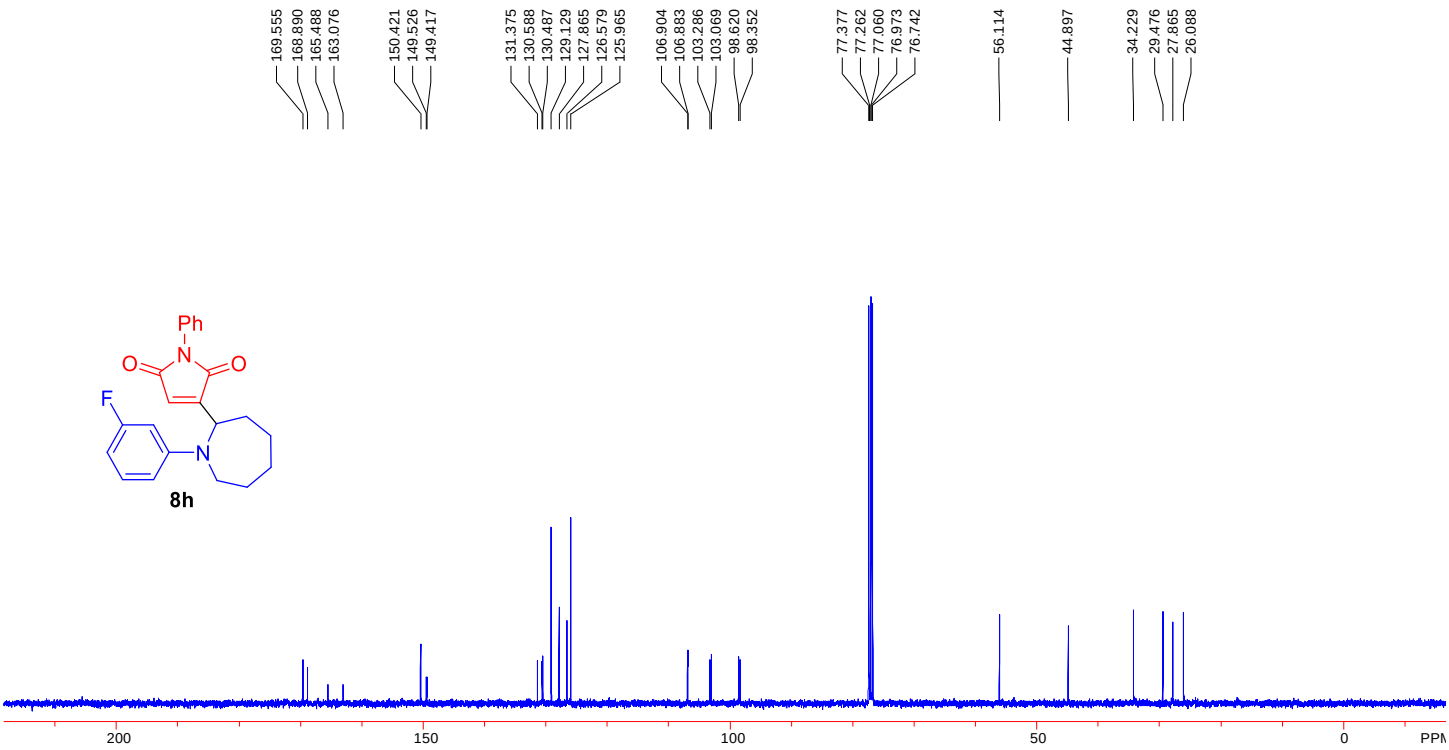
^{1}H NMR (400 MHz, $CDCl_3$) of **8g**



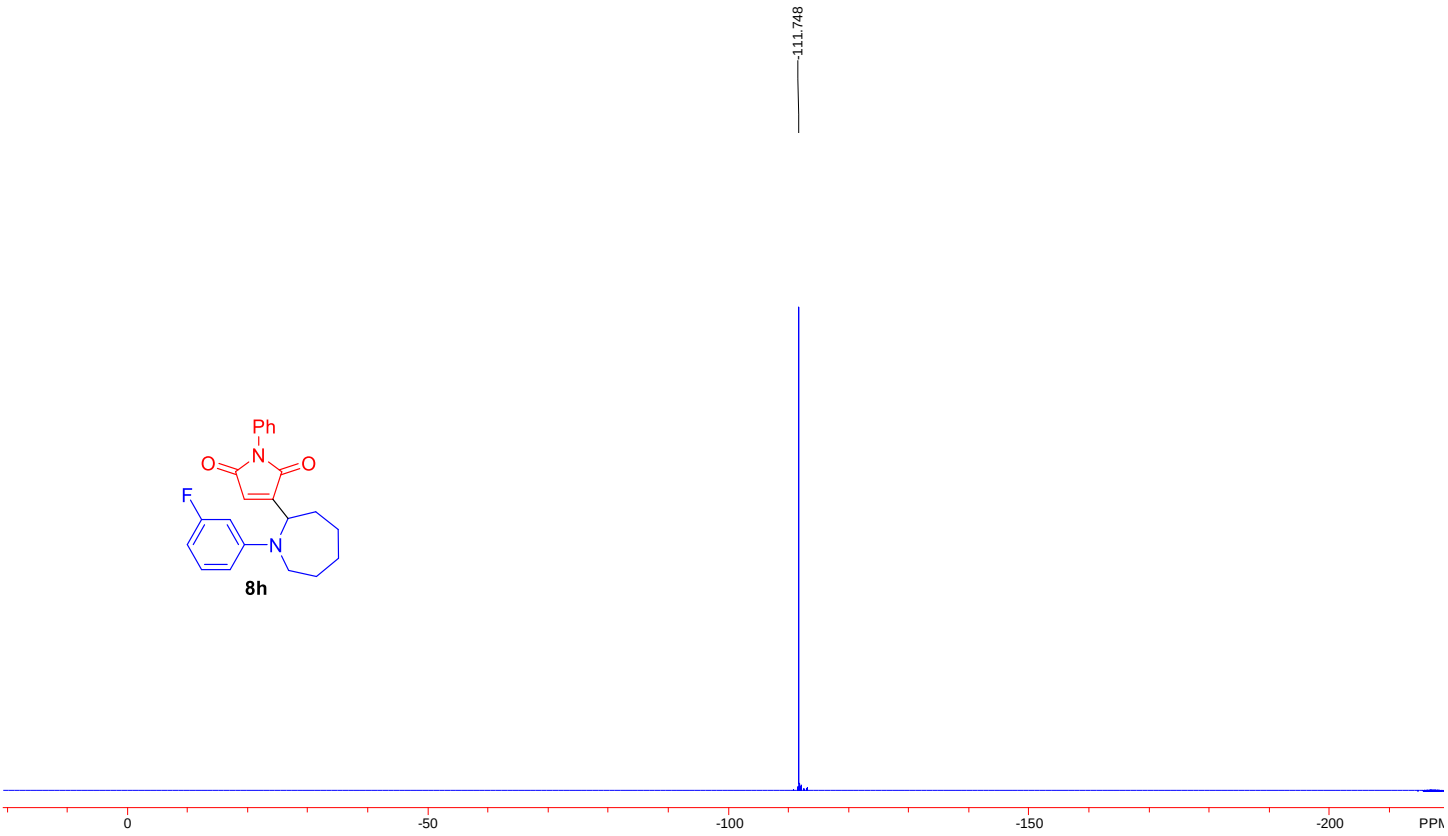
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8g**



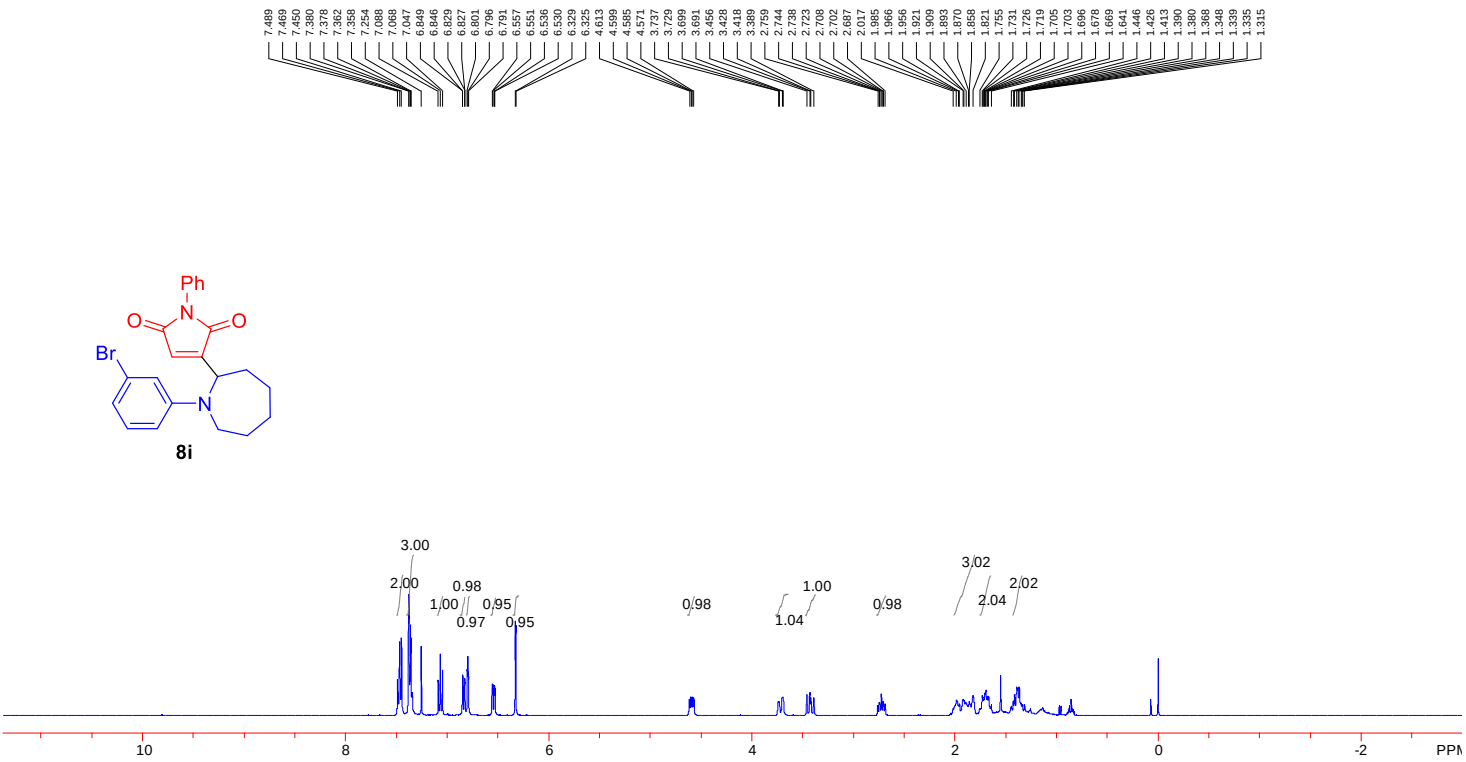
^{1}H NMR (400 MHz, $CDCl_3$) of **8h**



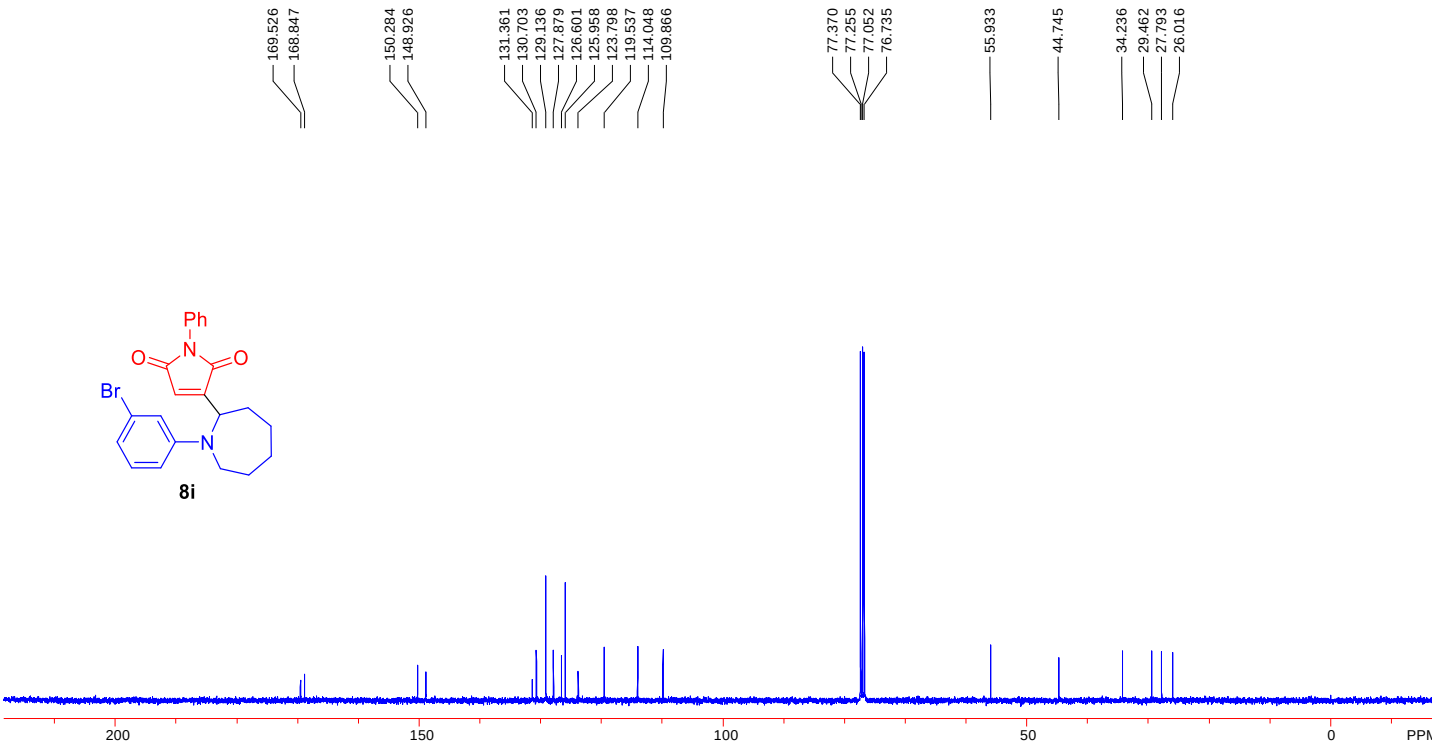
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8h**



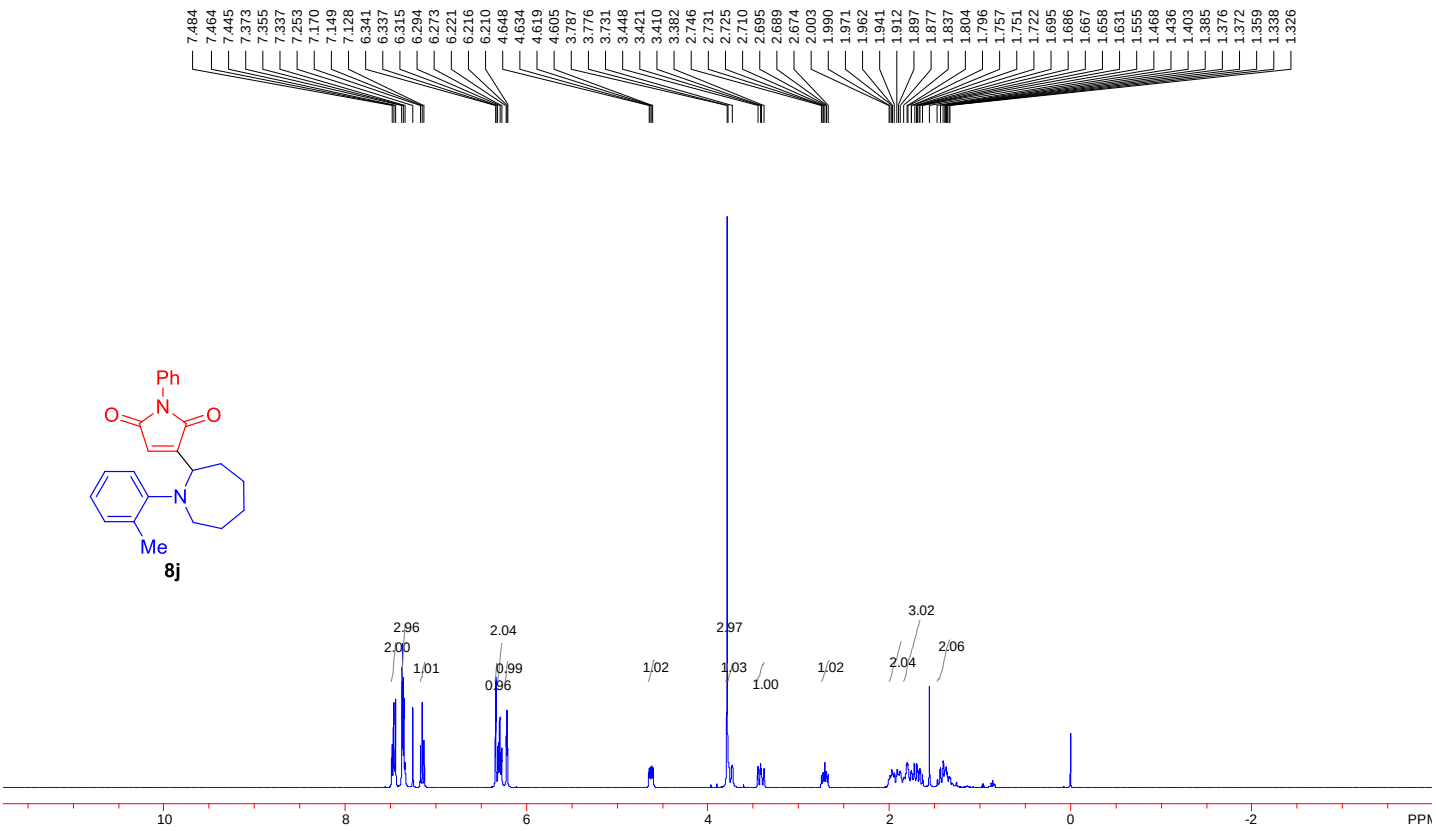
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) of **8h**



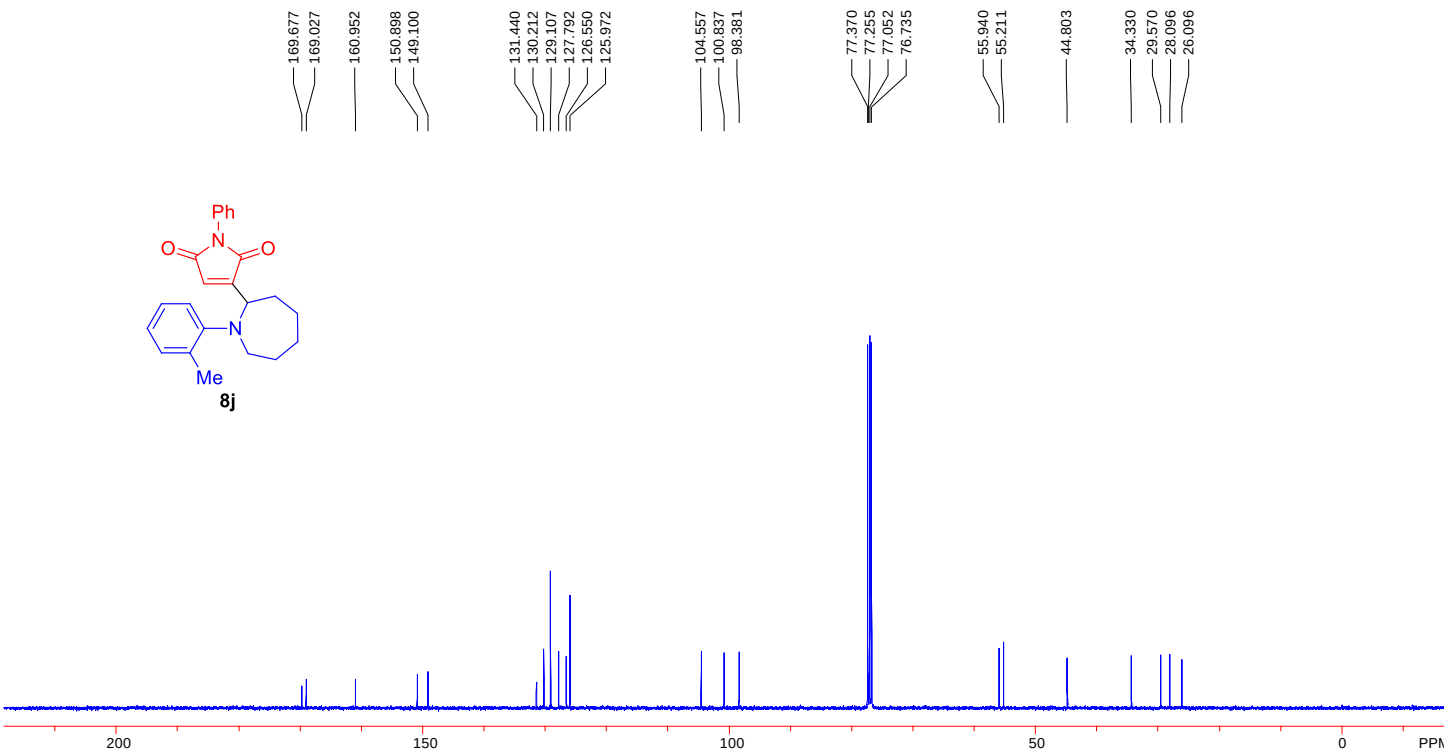
^{1}H NMR (400 MHz, $CDCl_3$) of **8i**



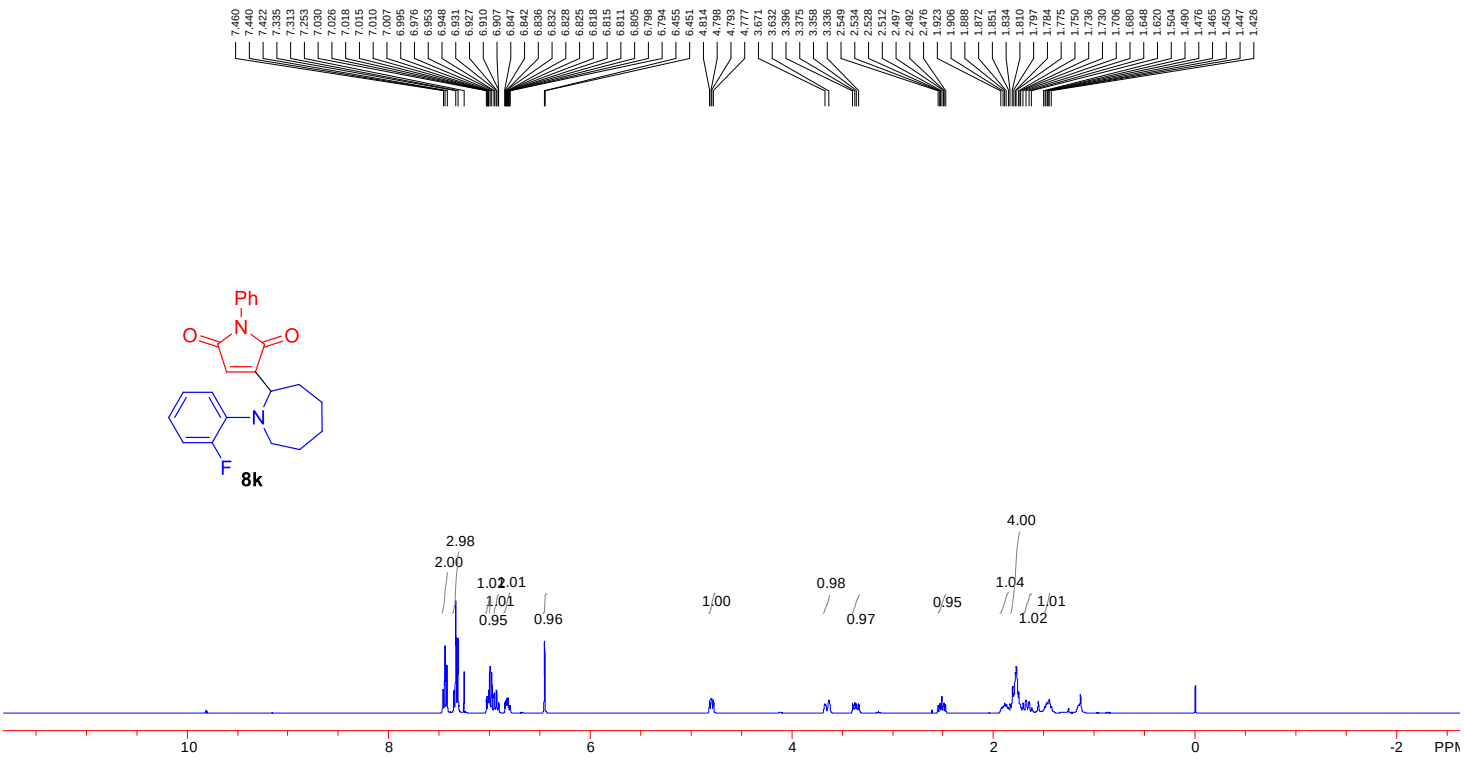
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8i**



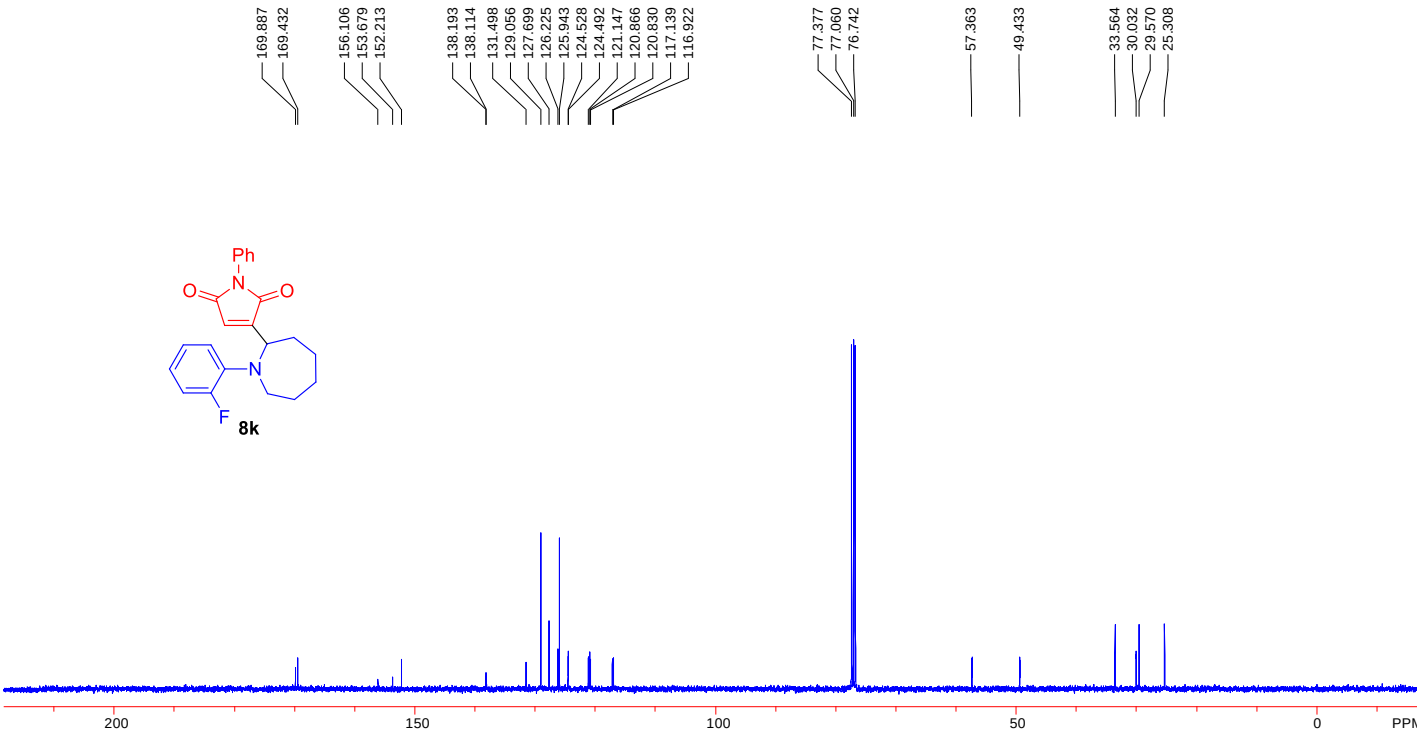
^{1}H NMR (400 MHz, $CDCl_3$) of **8j**



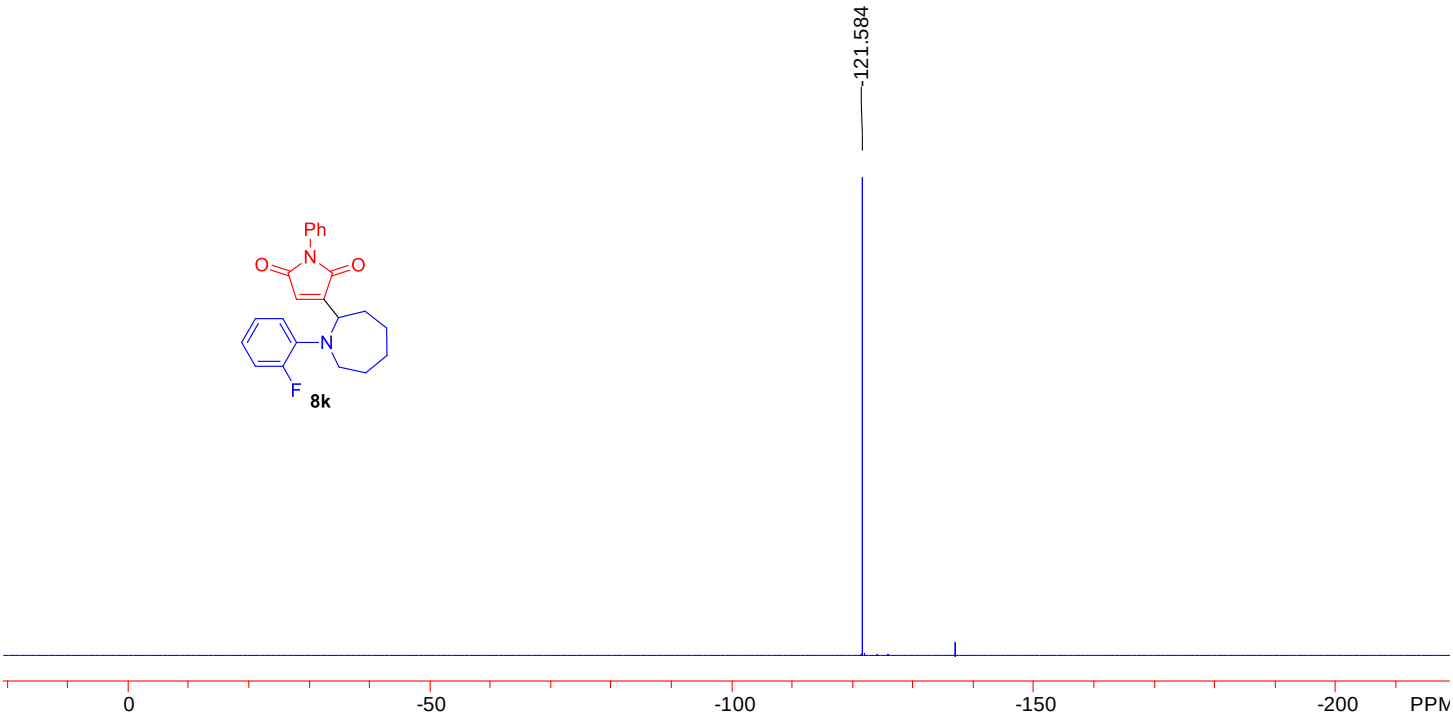
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of 8**j**



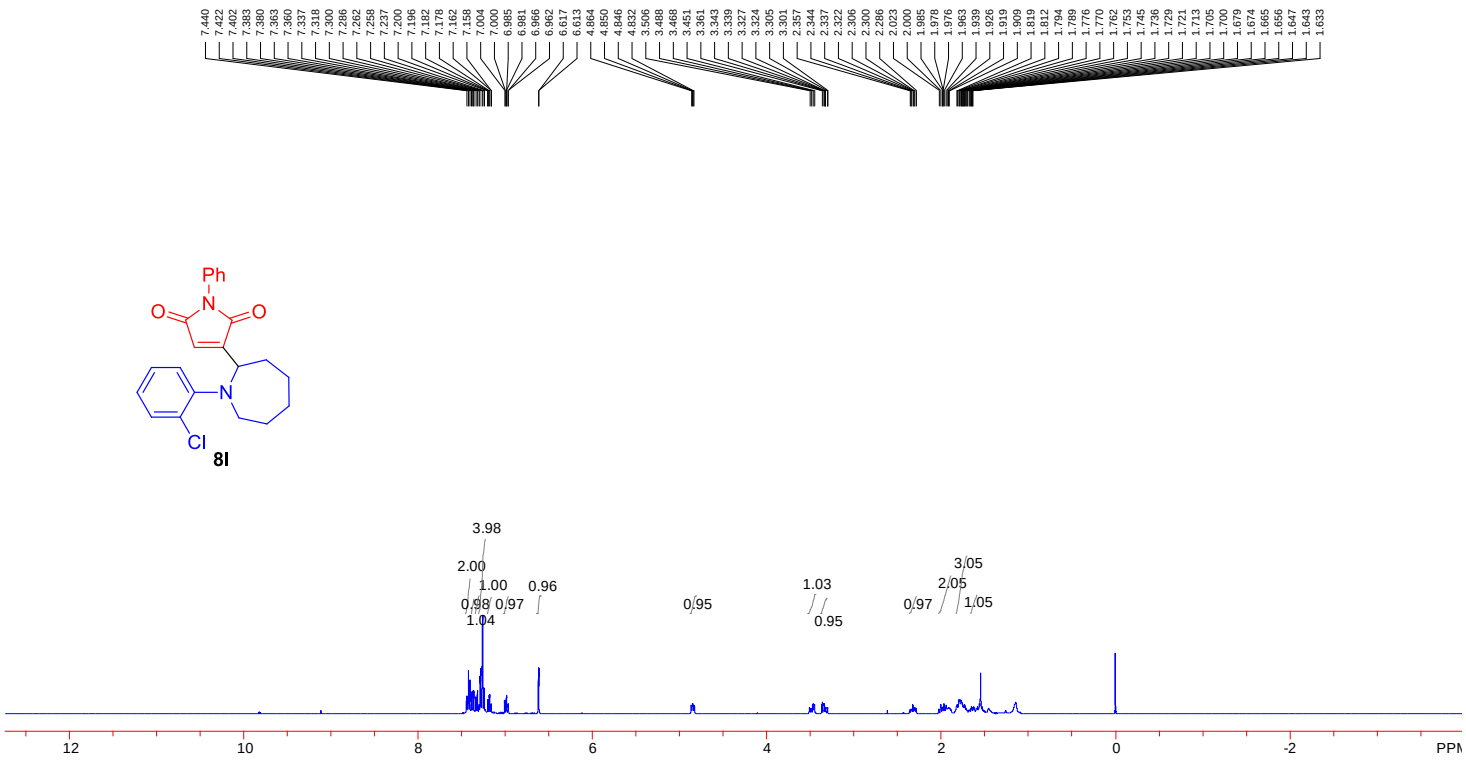
^{1}H NMR (400 MHz, $CDCl_3$) of **8k**



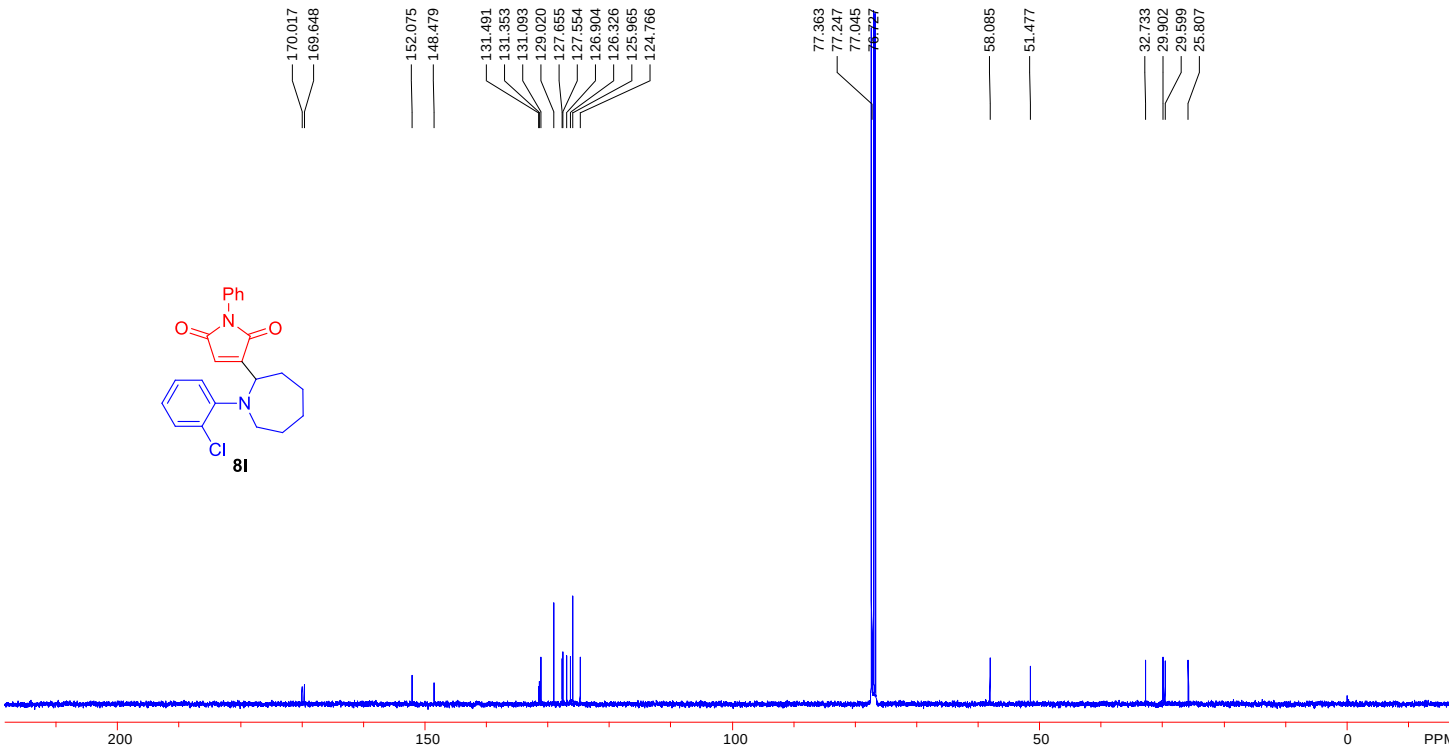
^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8k**



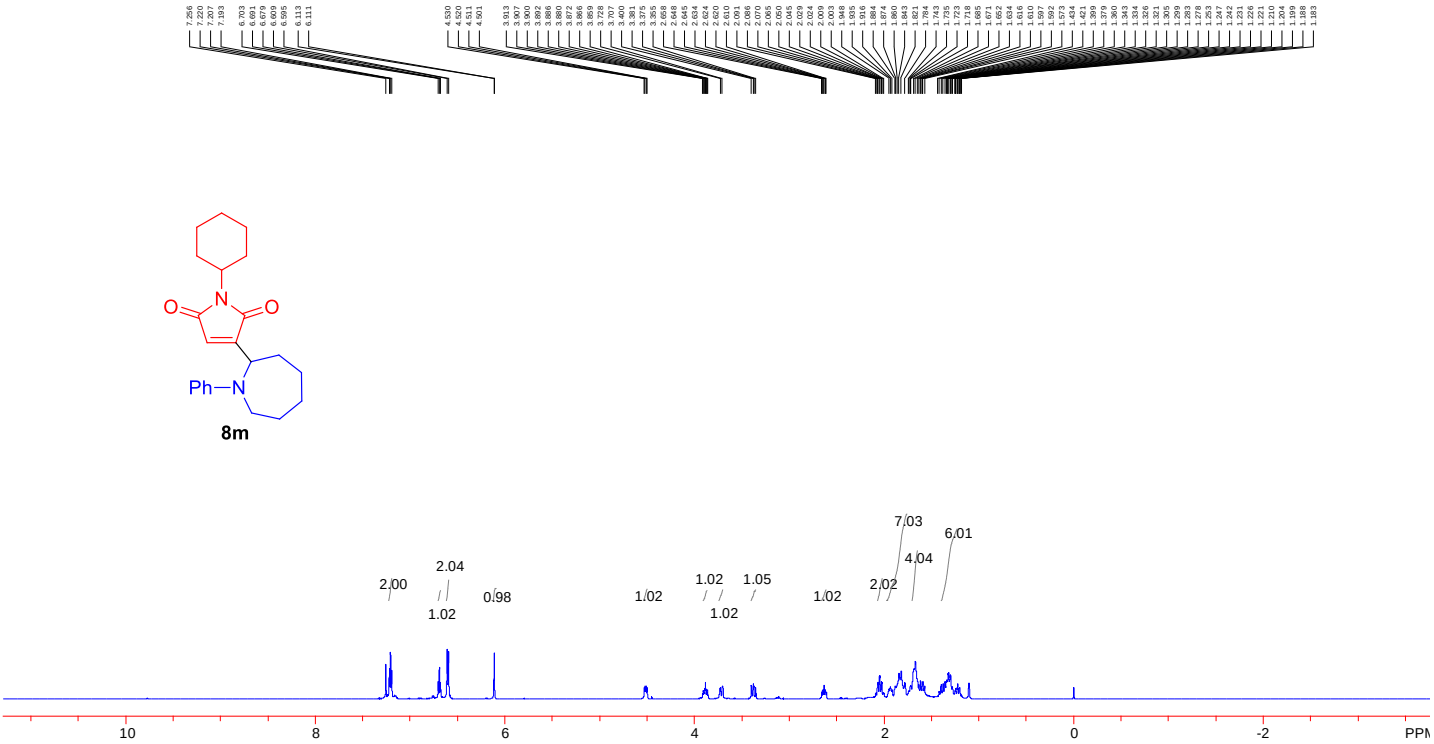
$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) of **8k**



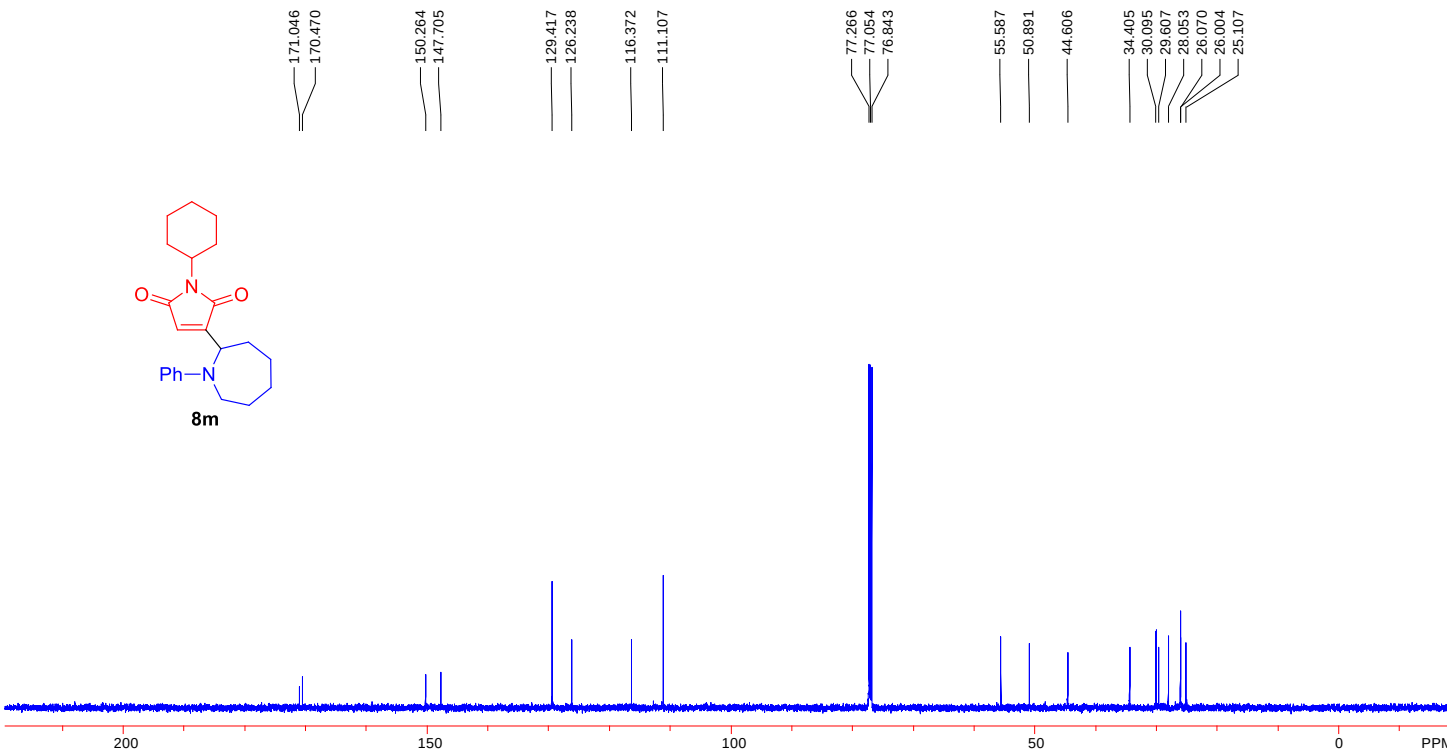
^{1}H NMR (400 MHz, $CDCl_3$) of **8l**



^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8l**

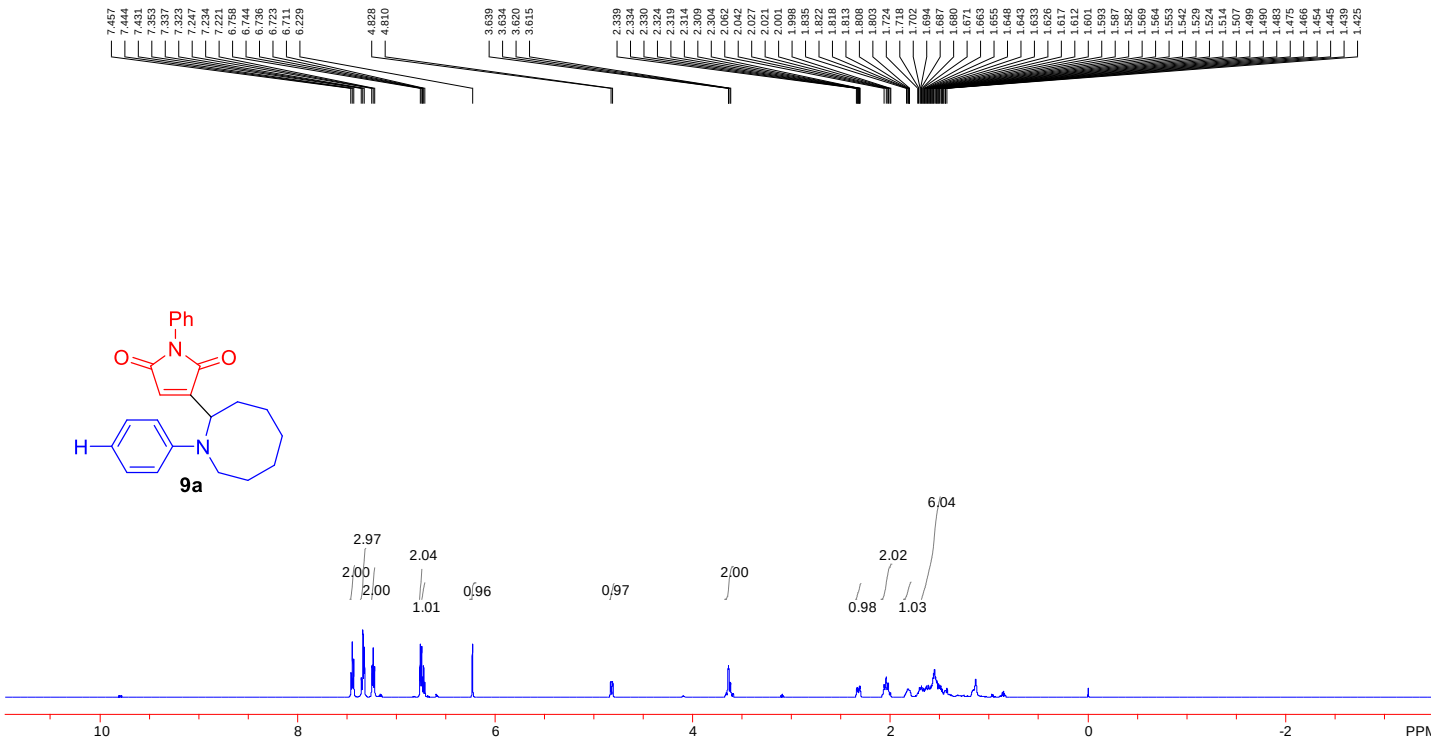


^{1}H NMR (600 MHz, $CDCl_3$) of **8m**

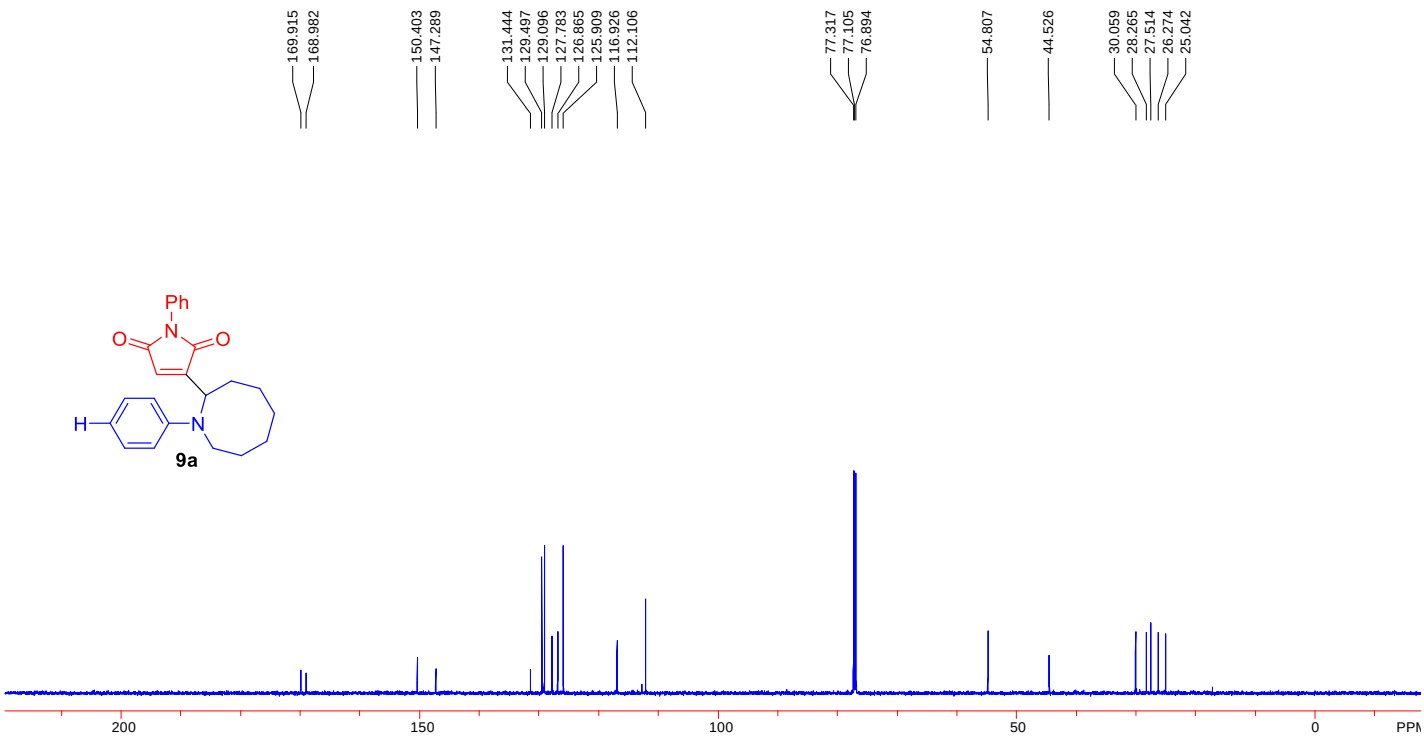


^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **8m**

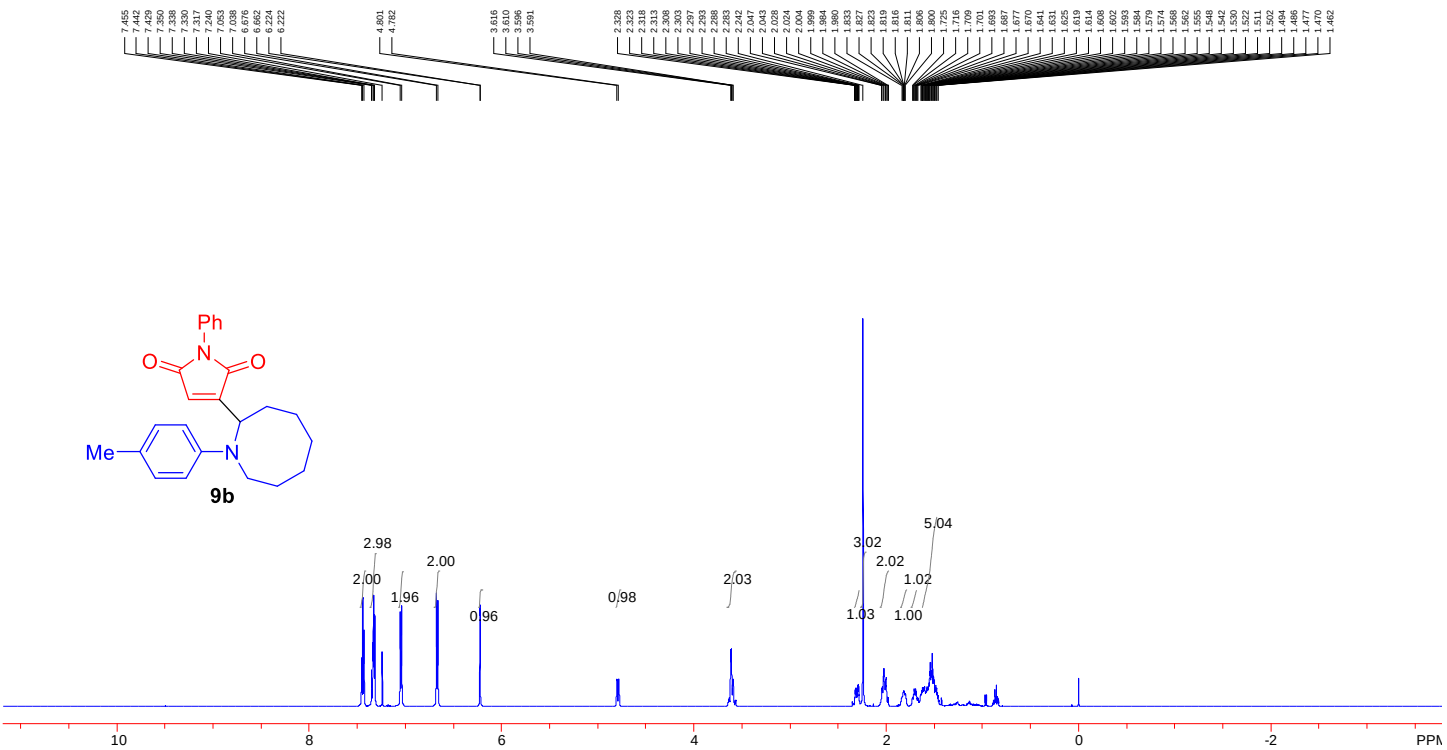
IV. Copies of NMR Spectra of 9a-9f



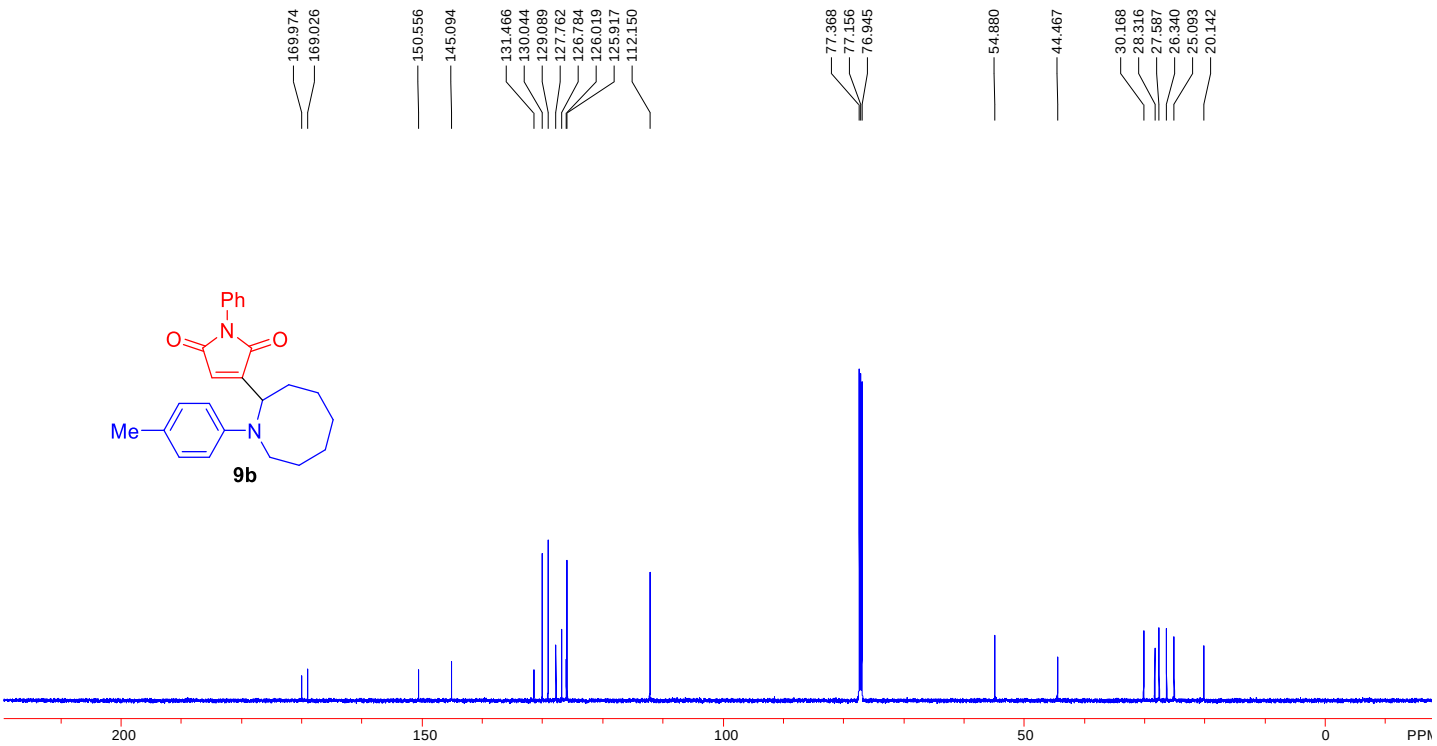
^{1}H NMR (600 MHz, $CDCl_3$) of **9a**



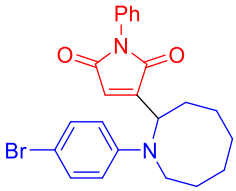
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **9a**



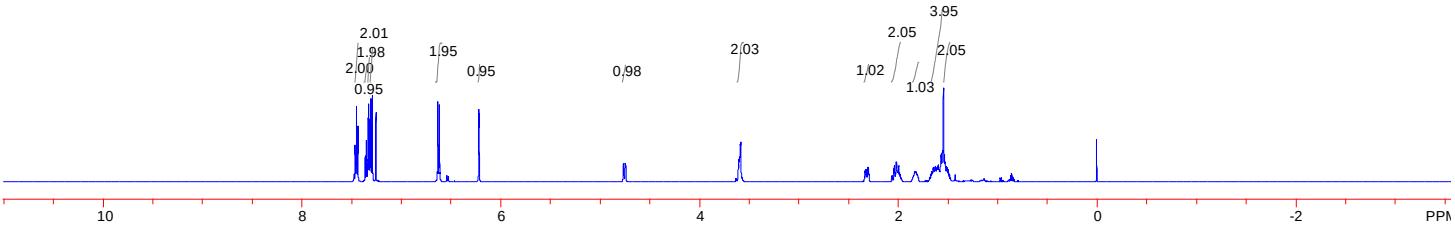
^{1}H NMR (600 MHz, $CDCl_3$) of **9b**



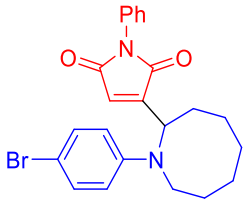
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **9b**



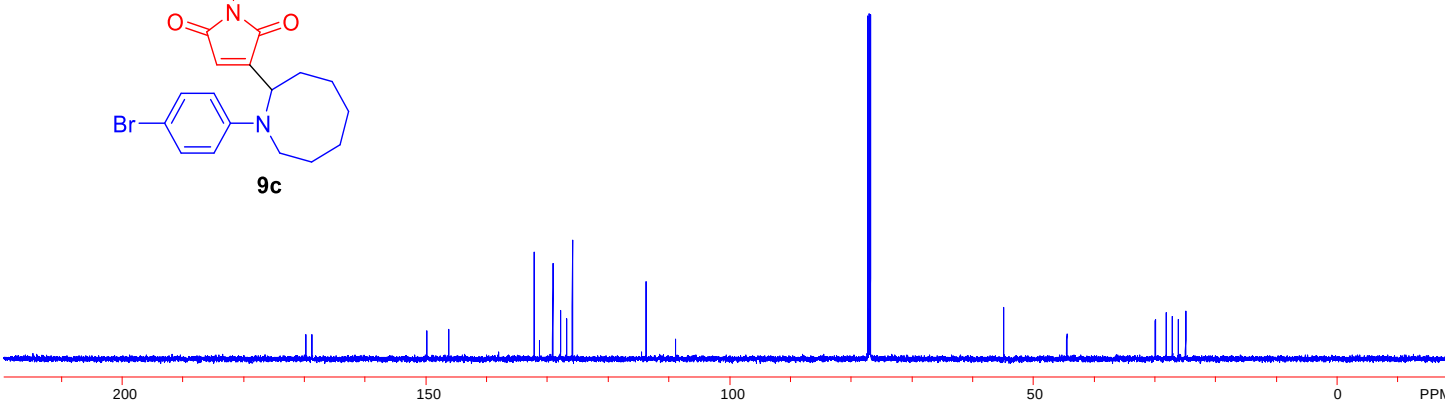
9c



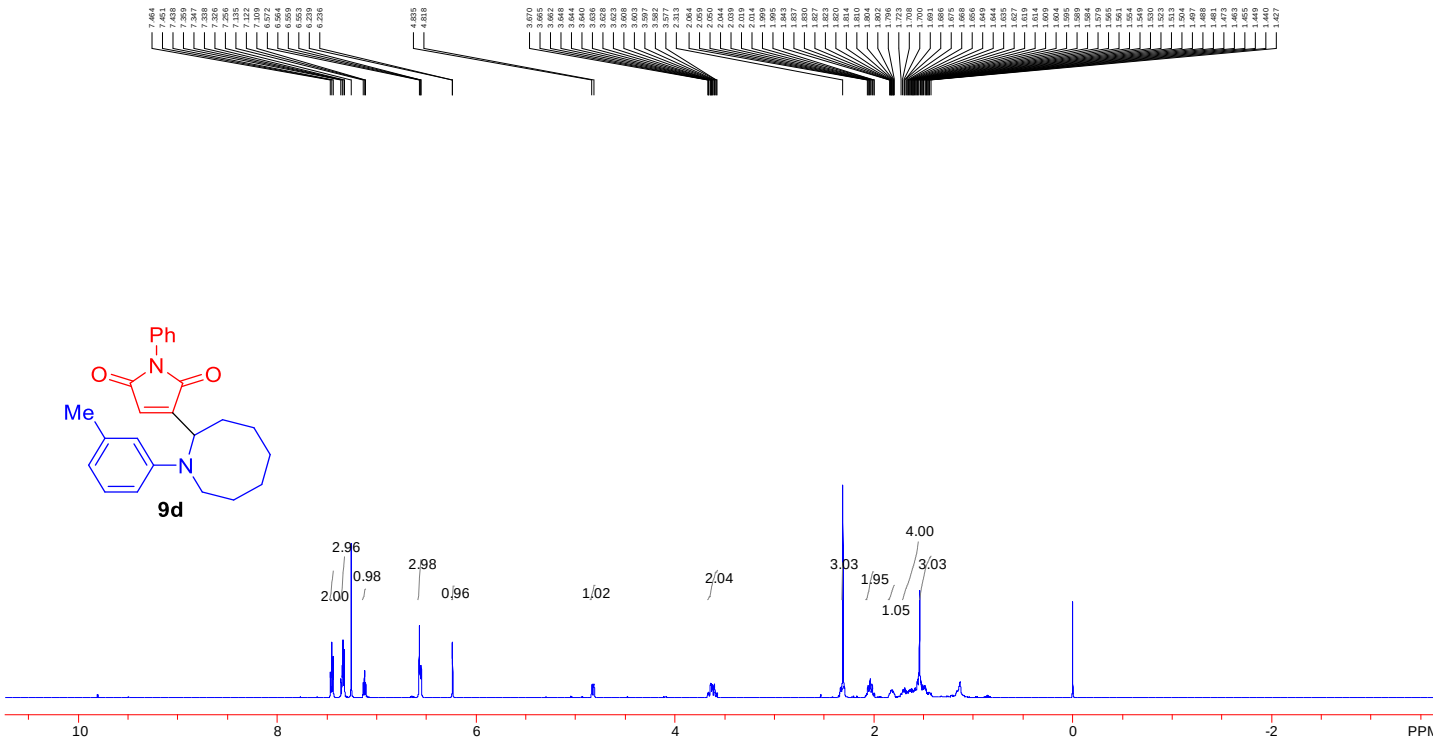
^{1}H NMR (600 MHz, $CDCl_3$) of **9c**



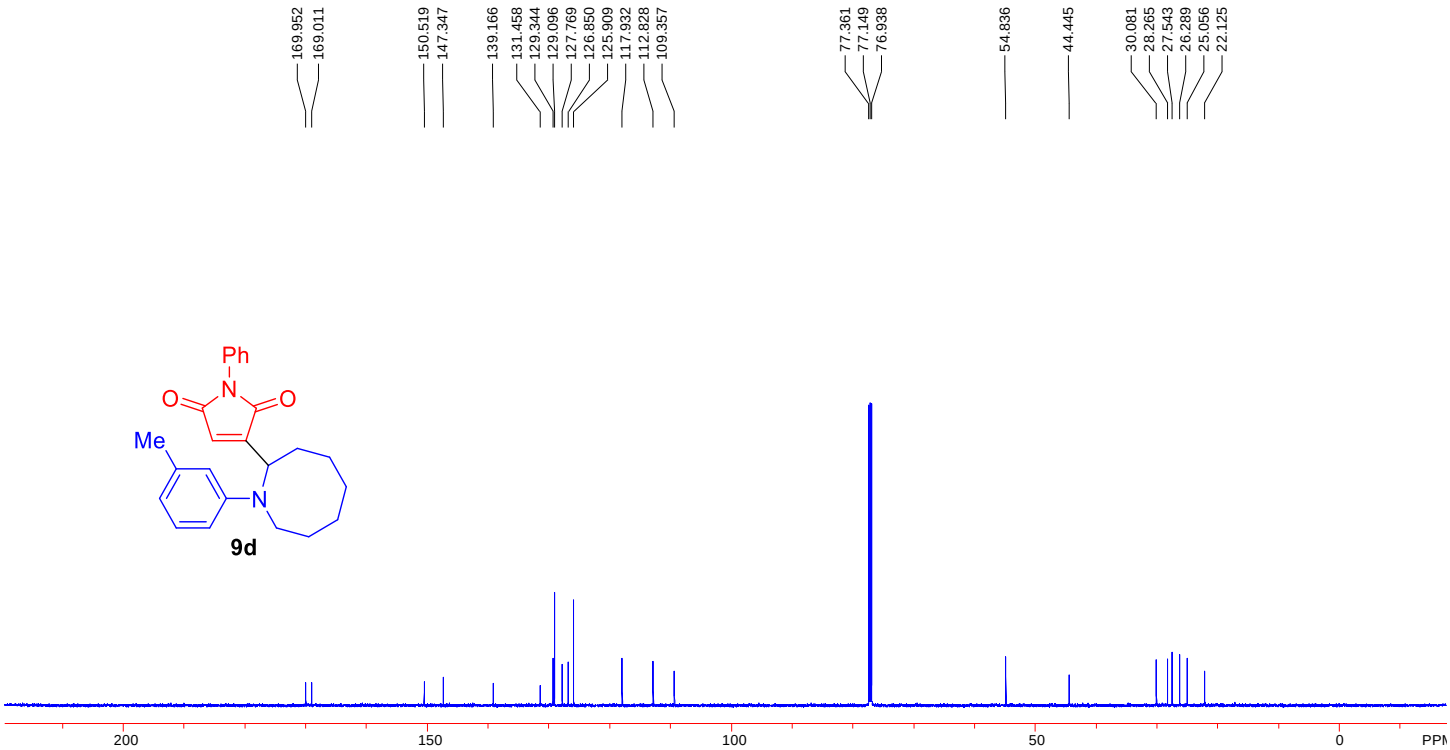
9c



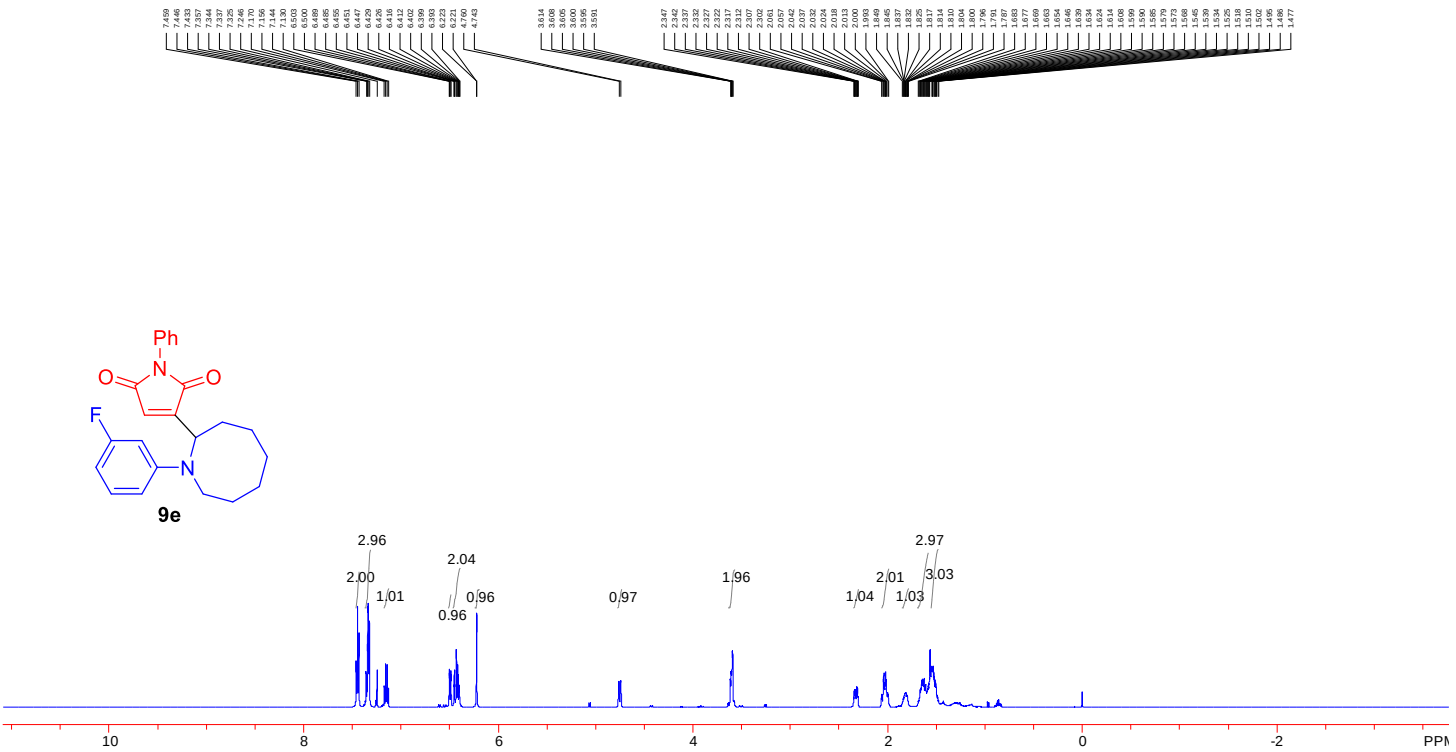
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **9c**



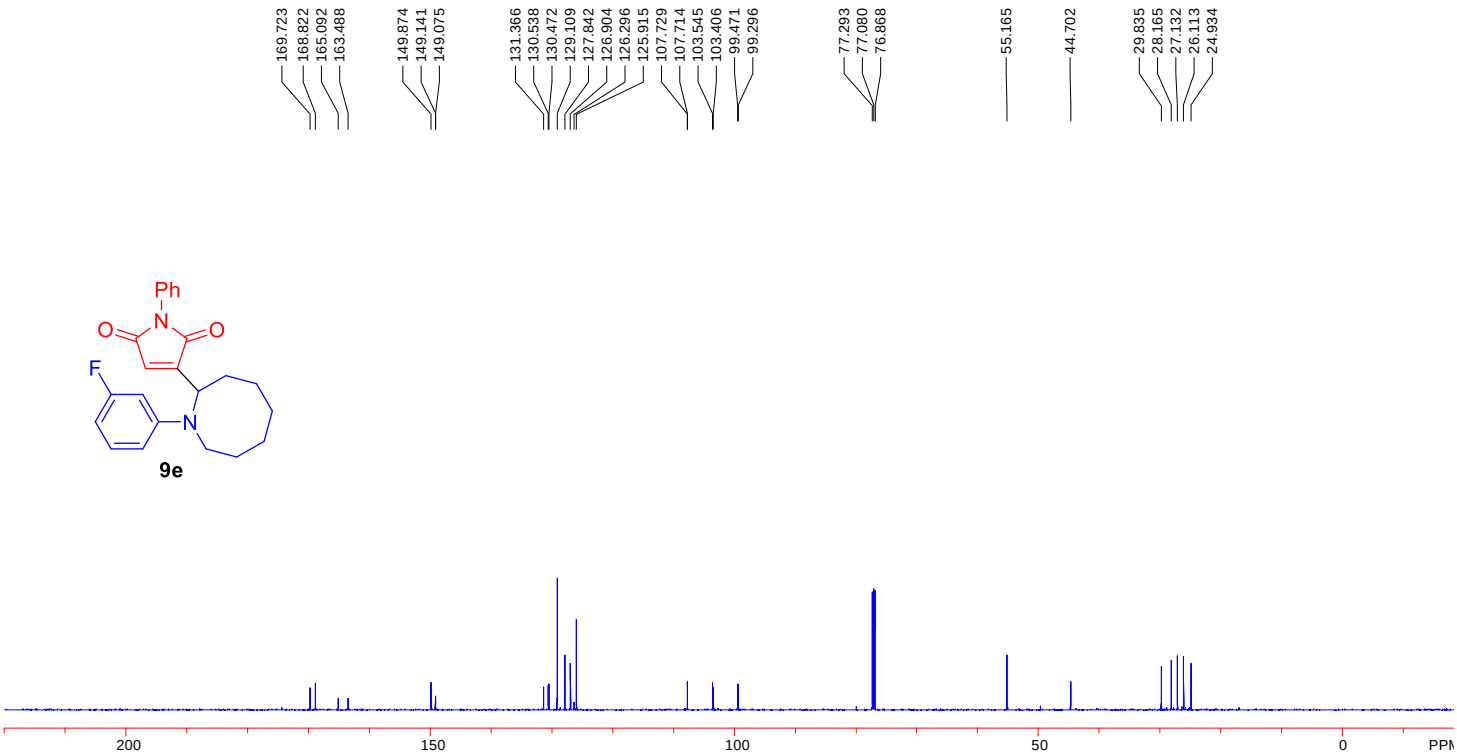
^{1}H NMR (600 MHz, $CDCl_3$) of **9d**



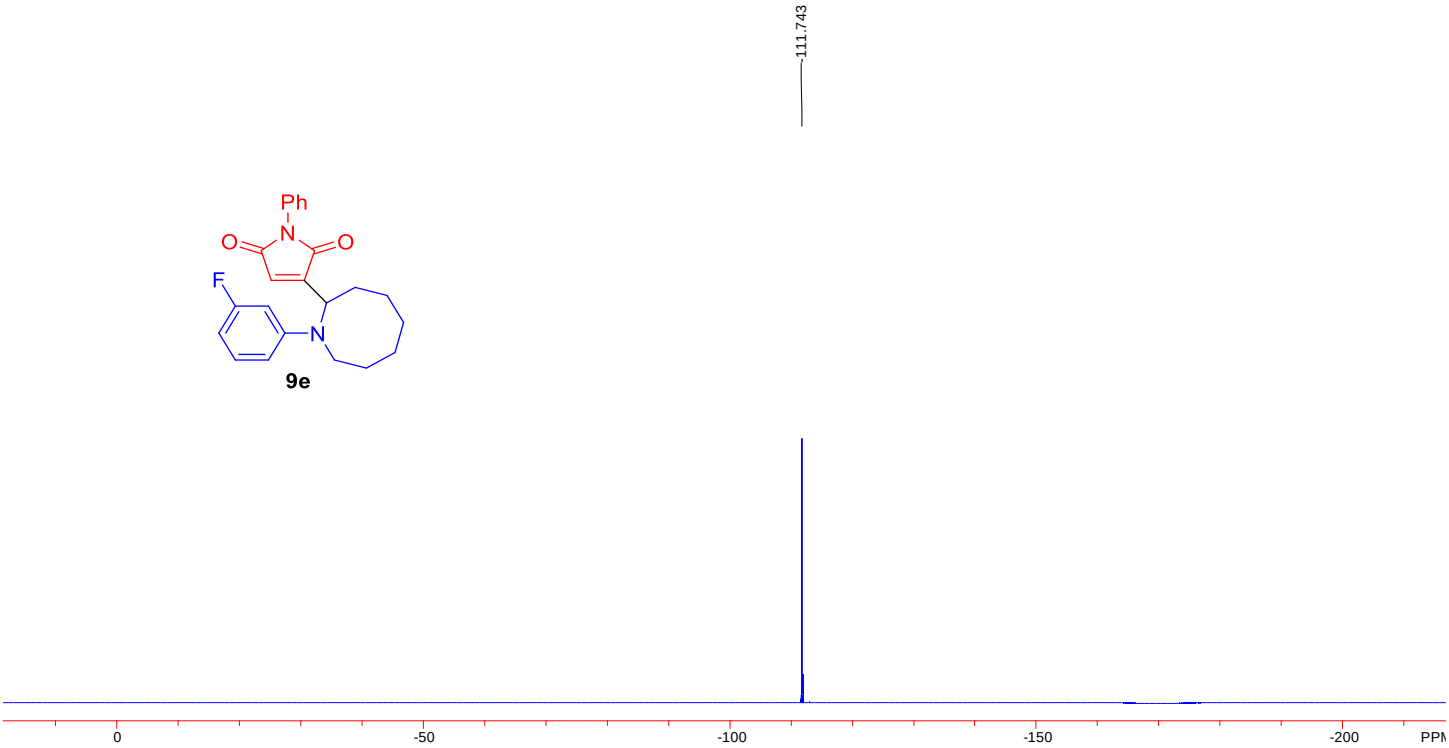
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **9d**



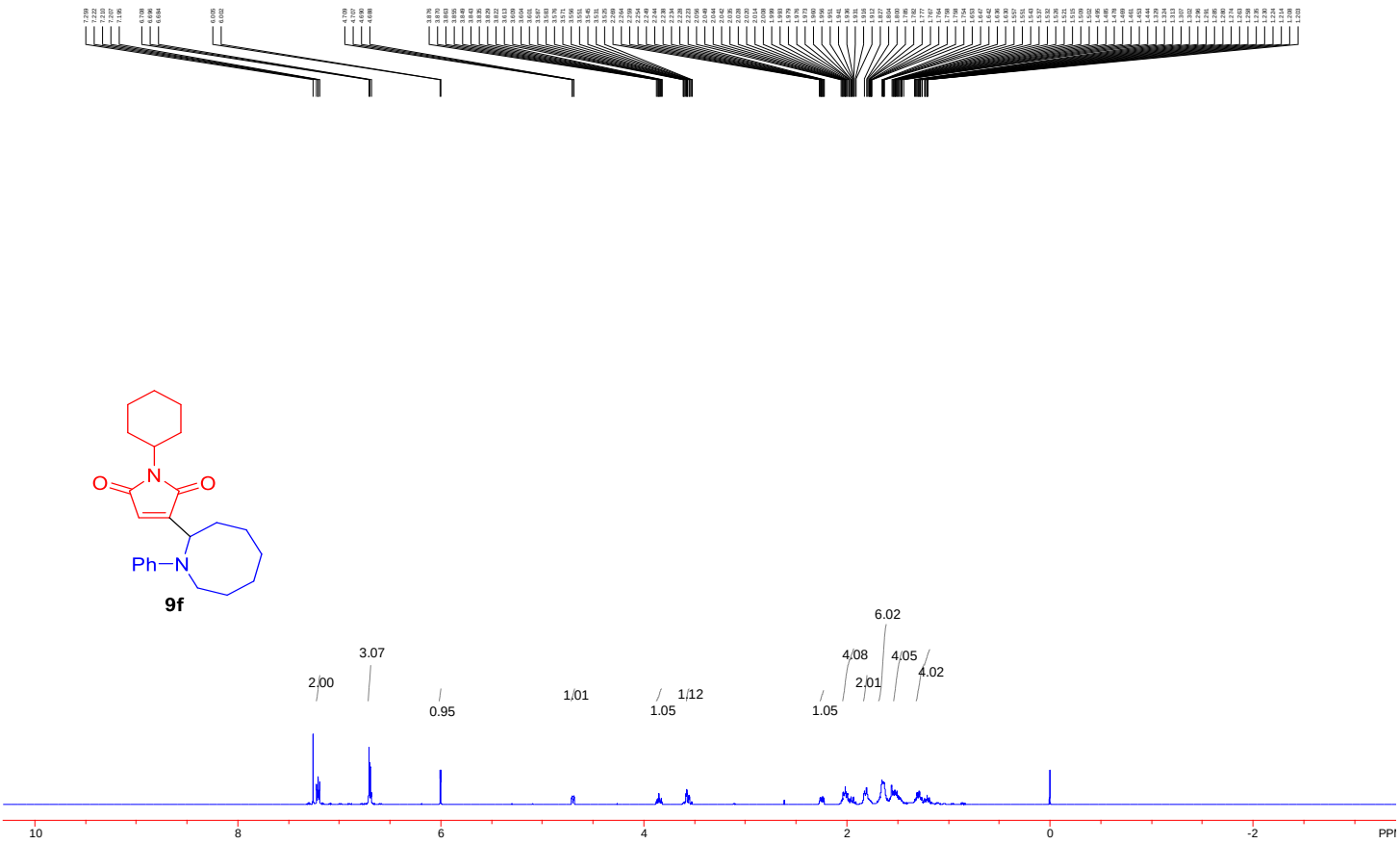
^{1}H NMR (600 MHz, $CDCl_3$) of **9e**



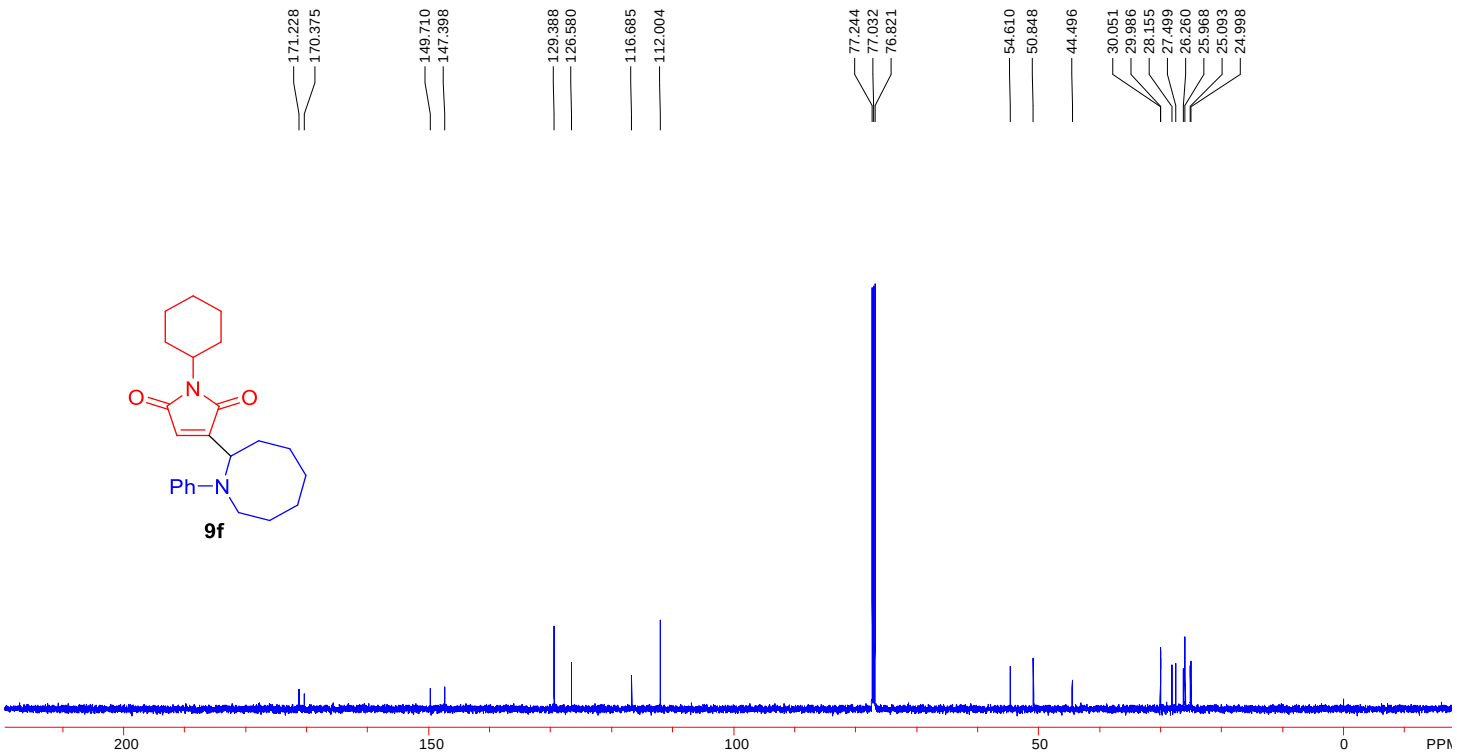
^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **9e**



$^{19}F\{^{1}H\}$ NMR (376 MHz, $CDCl_3$) of **9e**

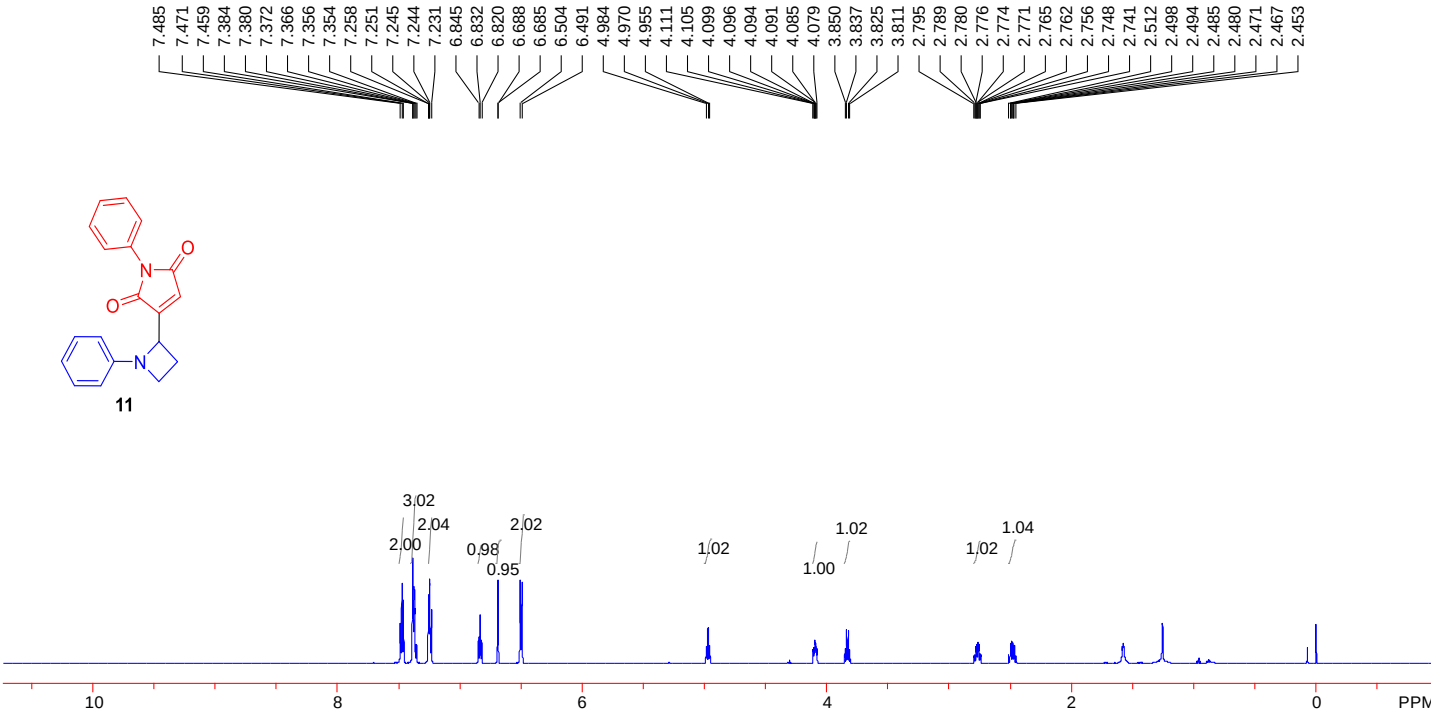


^{1}H NMR (600 MHz, $CDCl_3$) of **9f**

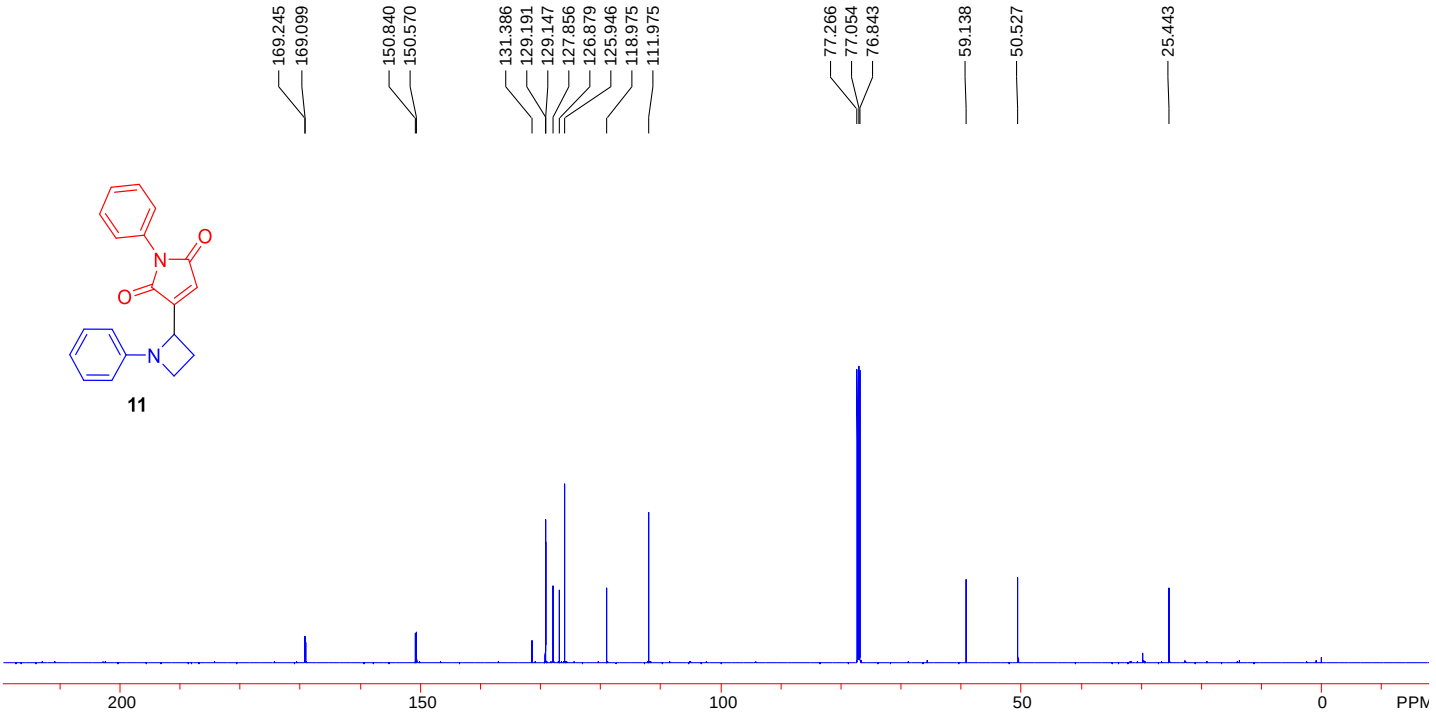


^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **9f**

V. Copies of NMR Spectra of 11

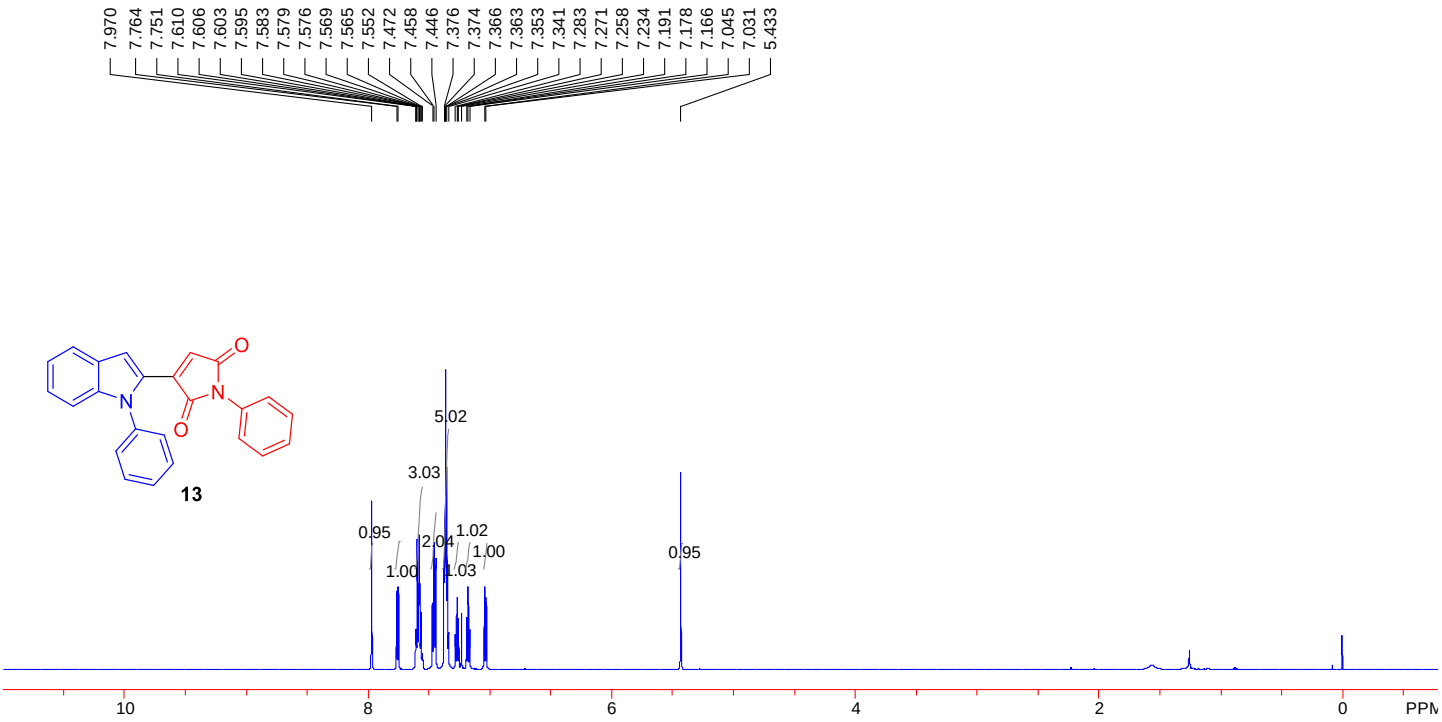


^{1}H NMR (600 MHz, $CDCl_3$) of **11**

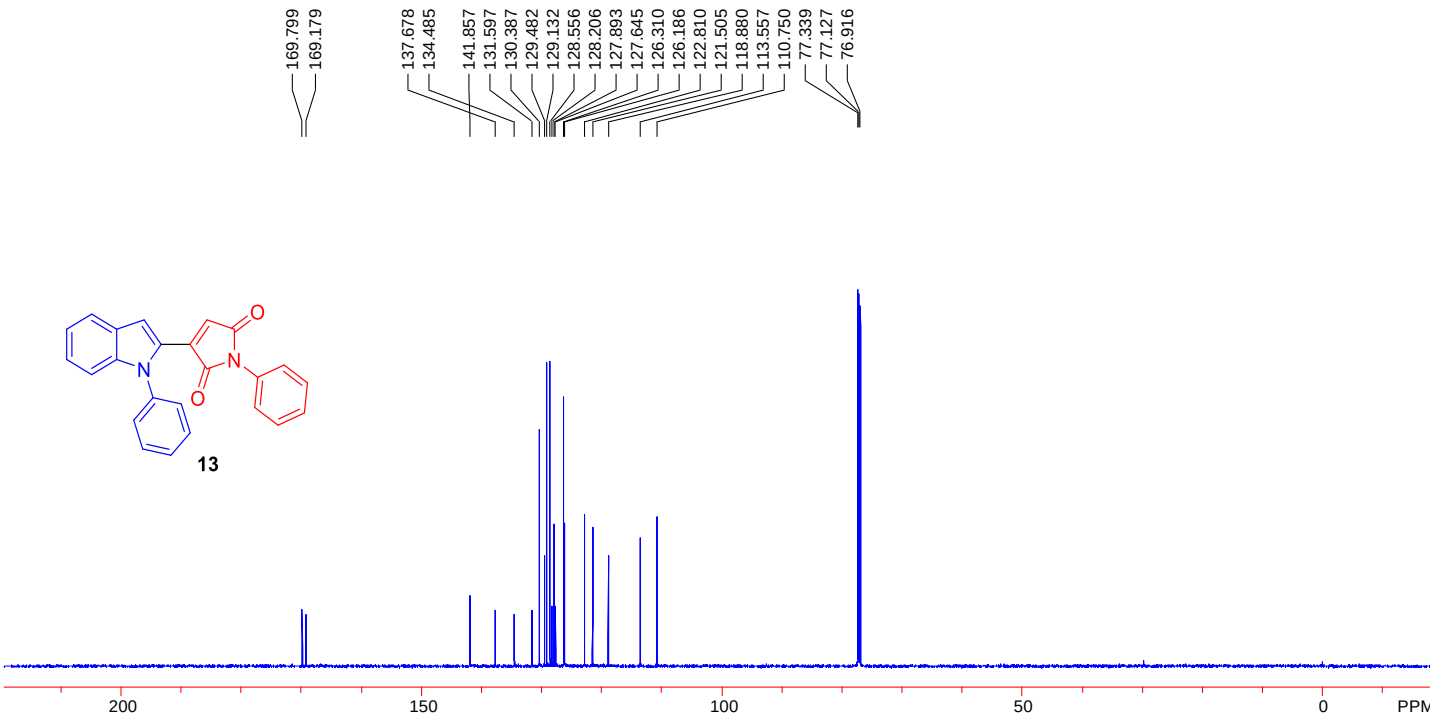


^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) of **11**

VI. Copies of NMR Spectra of 13



^{1}H NMR (600 MHz, $CDCl_3$) of **13**



^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$) of **13**

VII. X-Ray Crystal Structure and Data of 3s

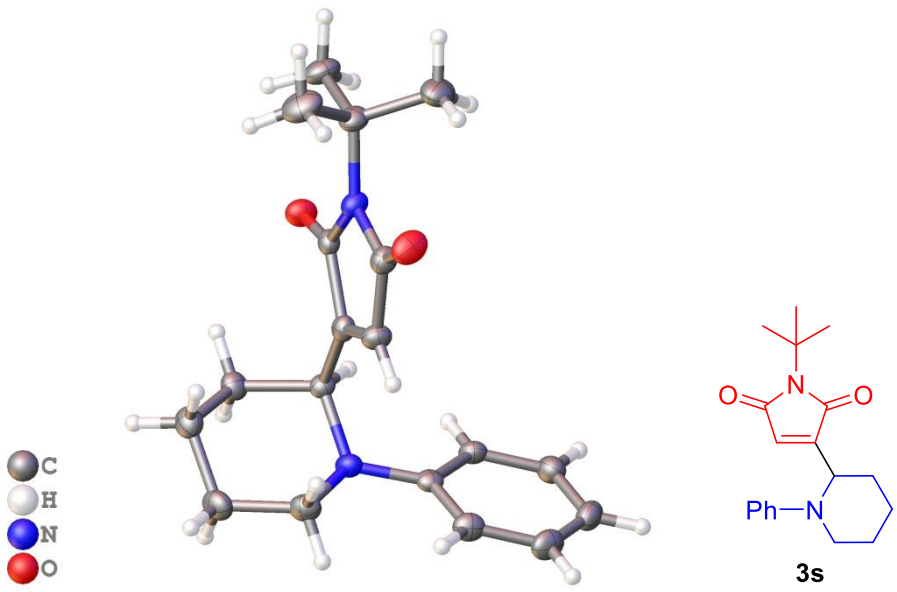
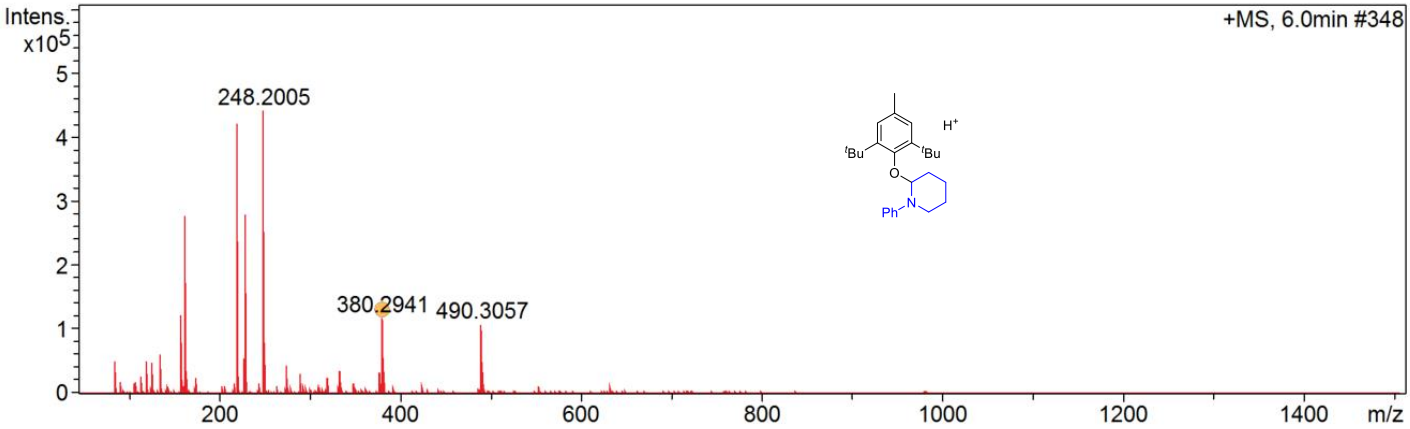
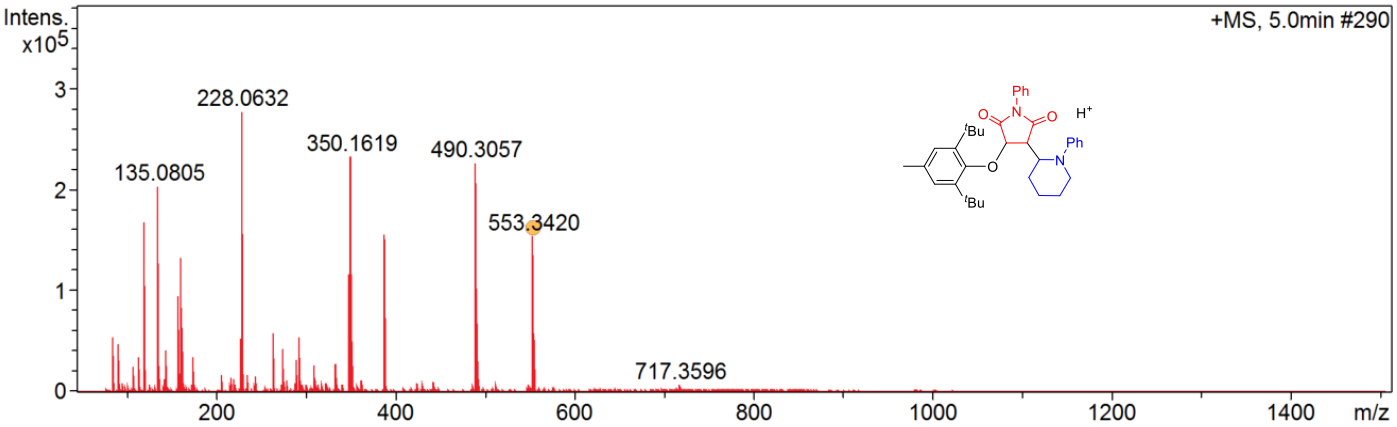


Fig. S1 X-ray crystal structure of **3s** with 50% ellipsoid probability

X-ray structure determination. Single crystals suitable for X-ray diffraction were obtained by slow evaporation of the solvent from an acetonitrile solution of **3s**. Crystal data collection and refinement parameters of **3s** are summarized in Table S1. Intensity data were collected at 150 K on a SuperNova Dual diffractometer using mirror-monochromated Mo Kα radiation, λ = 0.71073 Å. The data were corrected for decay, Lorentz, and polarization effects as well as absorption and beam corrections based on the multi-scan technique. Using Olex2, the structure was solved with the SHELXS structure solution program using Direct Methods and refined with the SHELXL refinement package using Least Squares minimisation. Nonhydrogen atoms were refined with anisotropic displacement parameters. The H-atoms were either located or calculated and subsequently treated with a riding model.

Table S1 Crystallographic data and structure refinement results of **3s**

Empirical formula	$C_{19}H_{24}N_2O_2$
Formula weight	312.40
Temp, K	150.00(10)
Crystal system	monoclinic
Space group	$P2_1$
a, Å	6.9979(4)

b, Å	7.7768(3)
c, Å	15.6987(8)
α (°)	90
β (°)	99.432(5)
γ (°)	90
Volume, Å^3	842.79(7)
Z	2
d_{calc}, g cm^{-3}	1.231
λ, Å	0.71073
μ, mm^{-1}	0.080
No. of data collected	6841
No. of unique data	3817
R_{int}	0.0332
Goodness-of-fit on F^2	1.036
R_1, wR_2 ($I > 2\sigma(I)$)	0.0660, 0.1389
R_1, wR_2 (all data)	0.0872, 0.1578

VIII. HRMS Analysis Reports of the Control Experiment