

Supplementary Material (ESI) for Green Chemistry
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Supporting Information

A facile, water mediated, microwave-assisted synthesis of 4,6-diaryl-2,3,3a,4-tetrahydro-1H-pyrido[3,2,1-jk]carbazoles by domino Fischer indole reaction-intramolecular cyclization sequence

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Experimental

General

Melting points were measured in open capillary tubes and are uncorrected. A CEM Discover microwave synthesizer (Model No: 908010) operating at 180/264 V and 50/60 Hz with microwave power maximum level of 300 W and microwave frequency of 2455 MHz was employed for the microwave-assisted experiments. The ¹H, ¹³C and 2D NMR were recorded on a Bruker (Avance) 300 MHz NMR instrument using TMS as internal standard and CDCl₃ as solvent. Standard Bruker software was used throughout. Chemical shifts are given in parts per million (δ-scale) and the coupling constants are given in Hertz. Silica gel-G plates (Merck) were used for TLC analysis with a mixture of petroleum ether (60–80 °C) and ethyl acetate as eluent. Elemental analyses were performed on a Perkin Elmer 2400 Series II Elemental CHNS analyzer. IR spectra were recorded on a JASCO FT IR instrument (KBr pellet).

General procedure for synthesis of 4,6-diphenyl-2,3,3a,4-tetrahydro-1H-pyrido[3,2,1-jk]carbazole (3)

Conventional heating method. A mixture of 2-(3-oxo-1,3-diarylpropyl)-1-cyclohexanones (1 mmol) and phenylhydrazine hydrochloride (1 mmol) in water

was refluxed for 2 h. After completion of the reaction (TLC), the yellow solid was filtered off and washed with water and further purified by recrystallisation from ethanol (5ml) to yield pure 3.

Microwave irradiation method. A vial containing a mixture of 2-(3-oxo-1,3-diarylpropyl)-1-cyclohexanones (1 mmol) and phenylhydrazine hydrochloride (1 mmol) in water (4 ml) was sealed and subjected to microwave irradiation, programmed at 140 °C and 120 W. After a period of 3-5 minutes, the temperature reached a plateau, 130 °C, and remained constant. After completion of the reaction (15 min), the vial was cooled to room temperature. The pure solid product was filtered off washed with water and further purified by recrystallisation from ethanol to yield pure compound 3.

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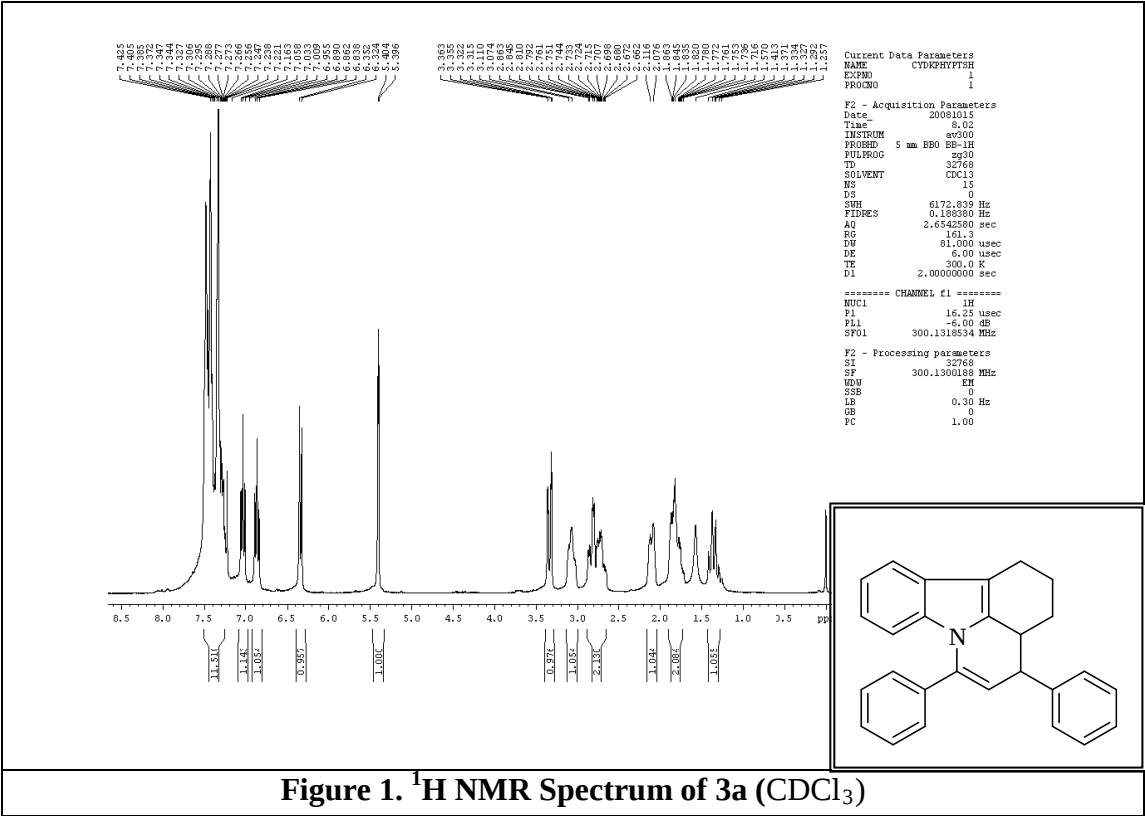


Figure 1. ¹H NMR Spectrum of 3a (CDCl₃)

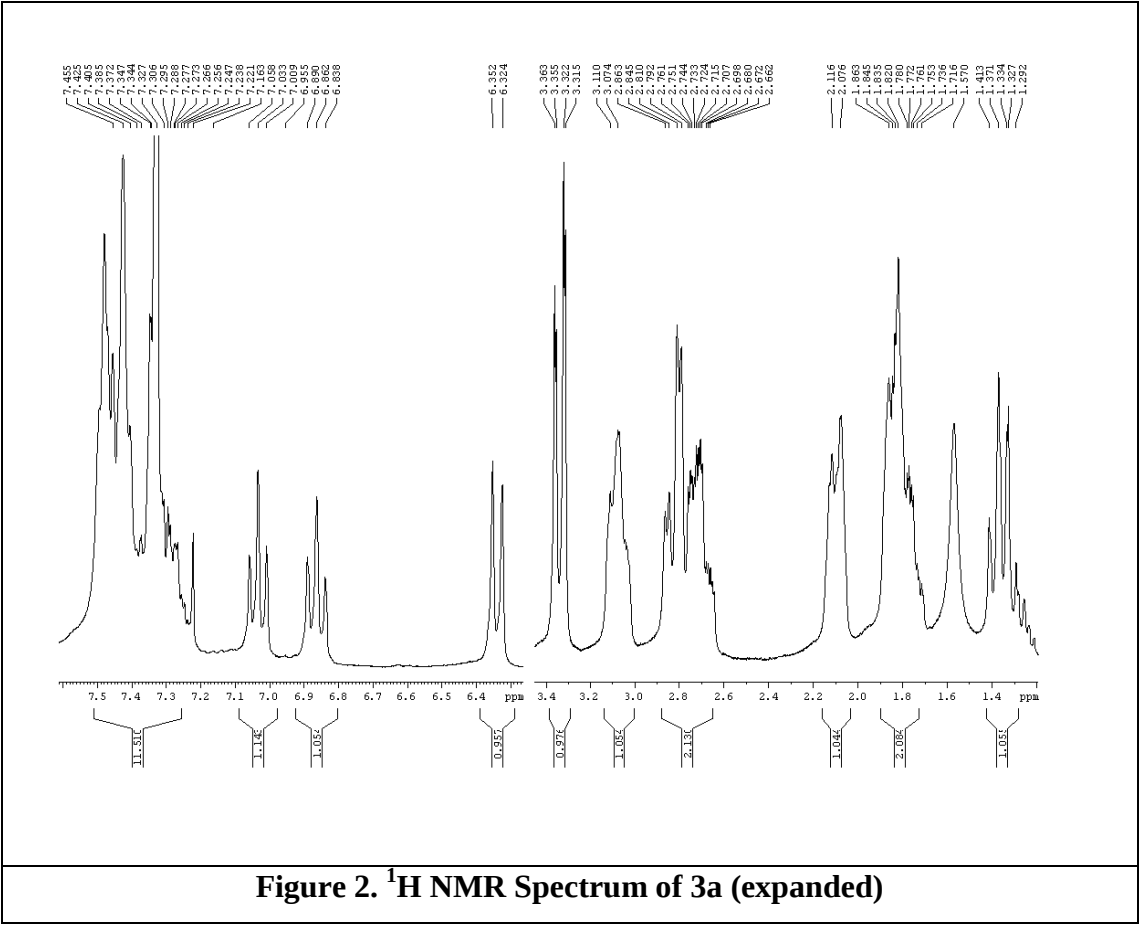
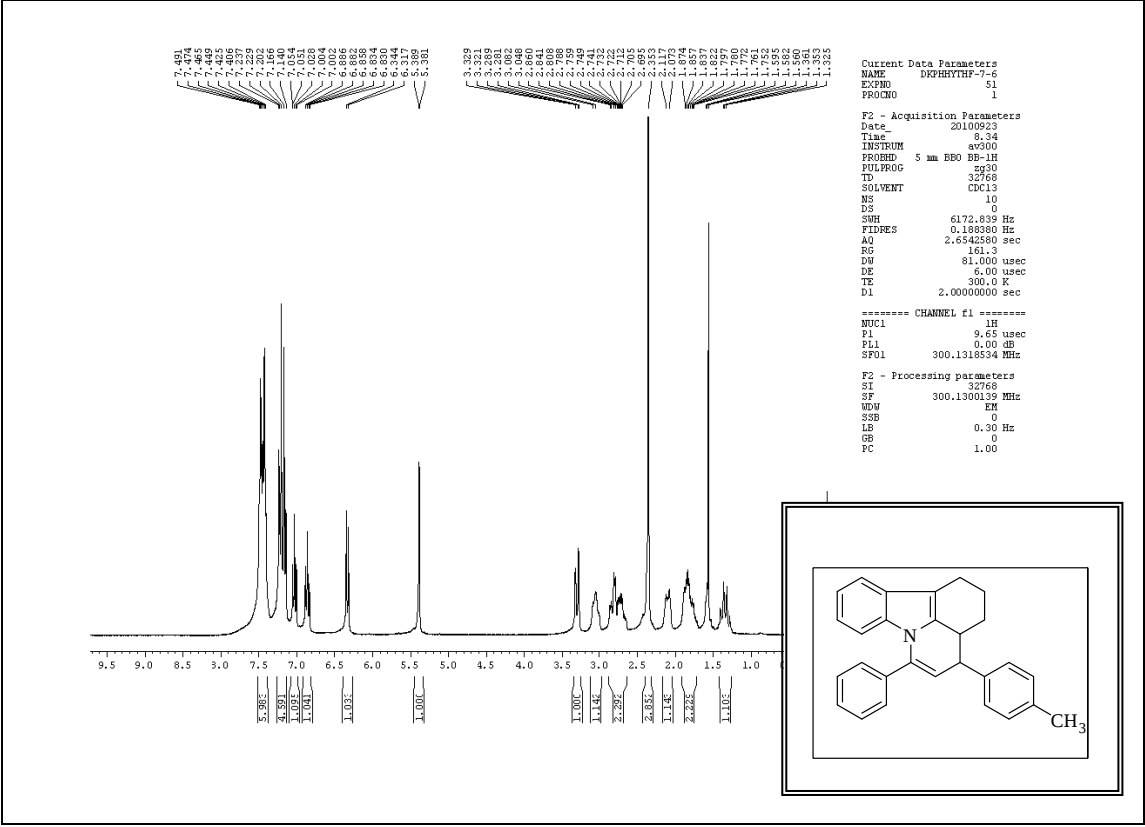
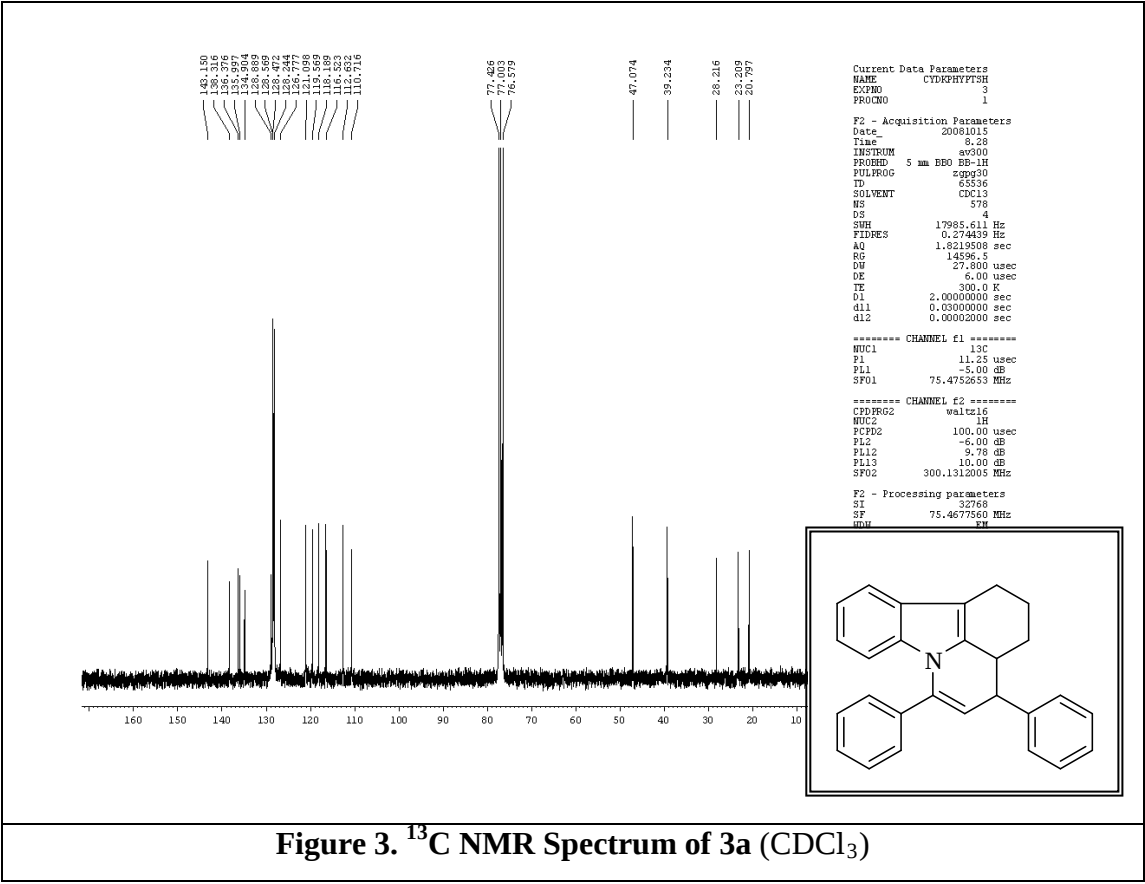


Figure 2. ¹H NMR Spectrum of 3a (expanded)



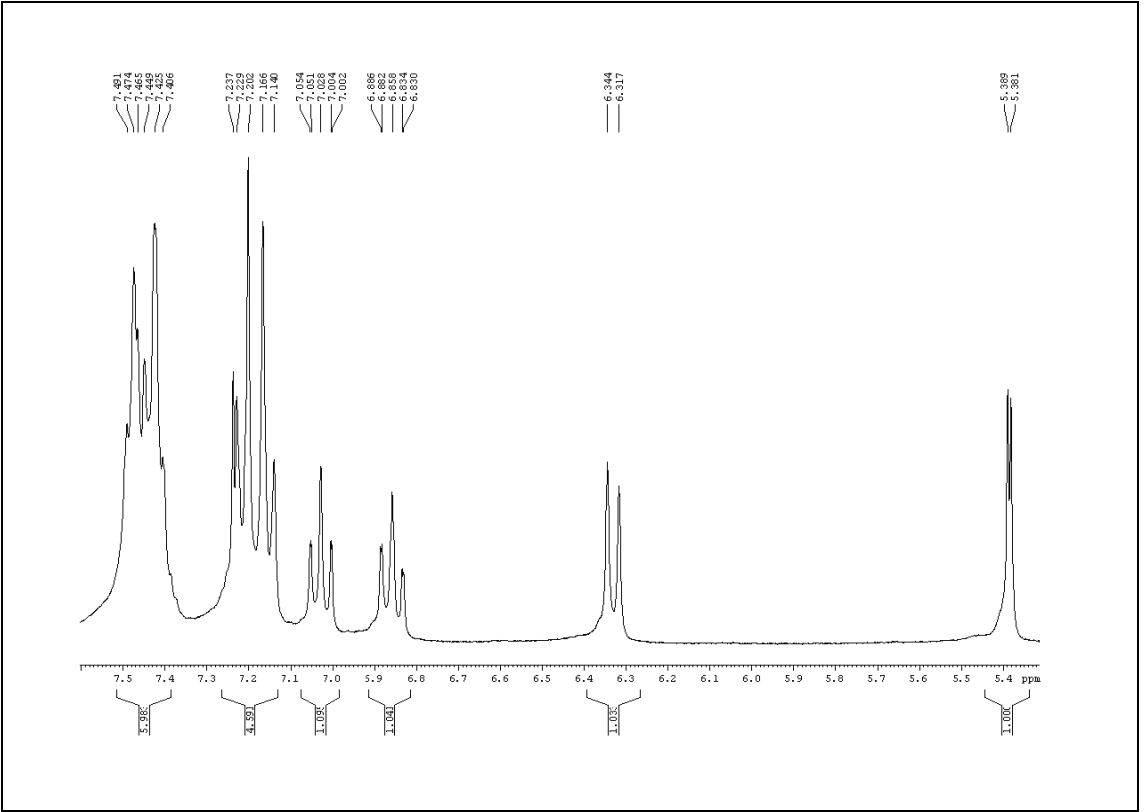
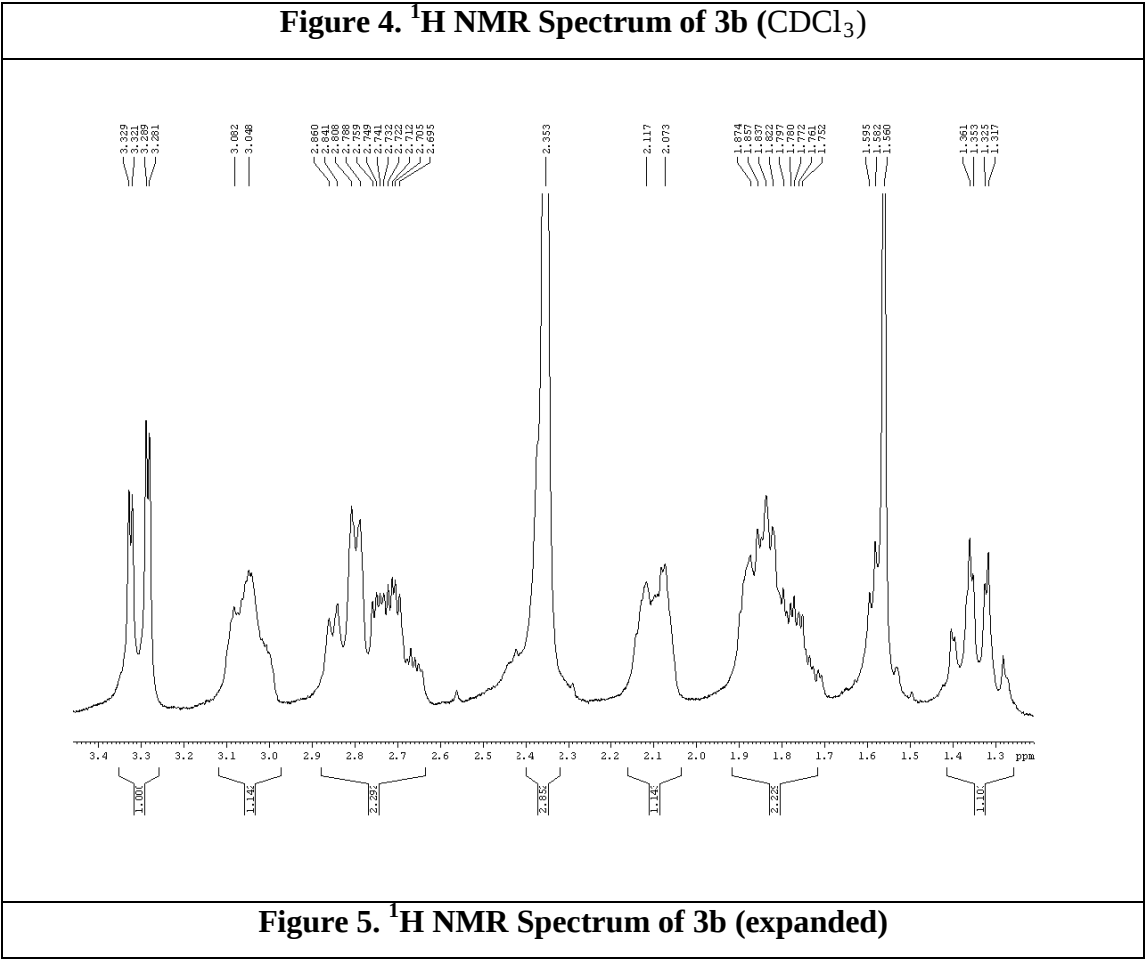


Figure 6. ^1H NMR Spectrum of 3b (expanded)

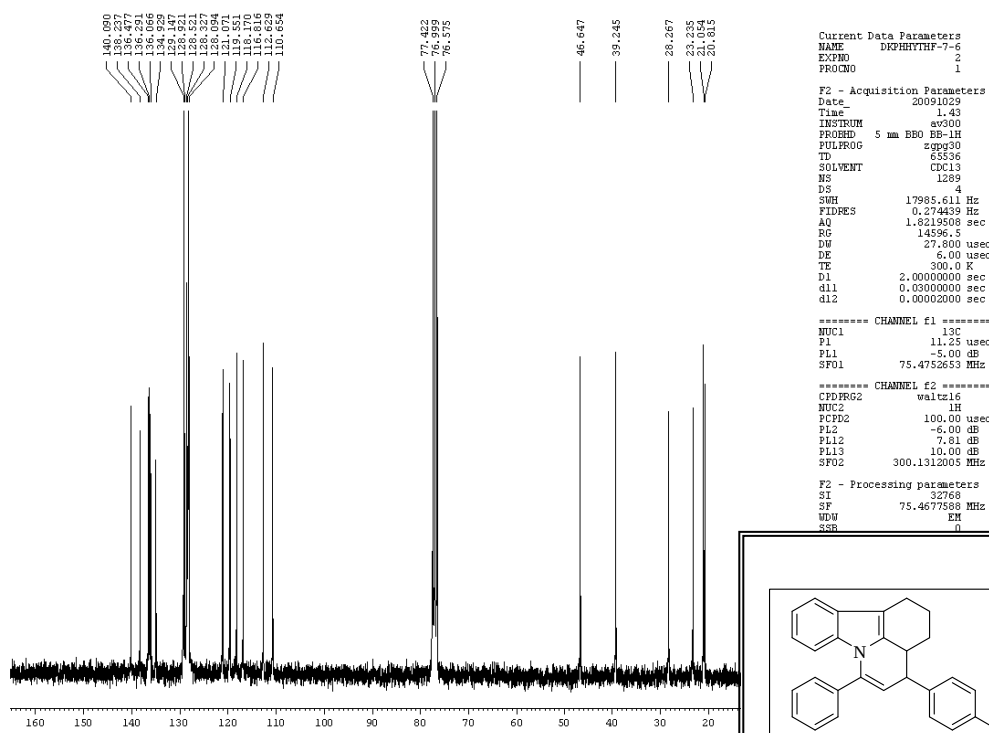
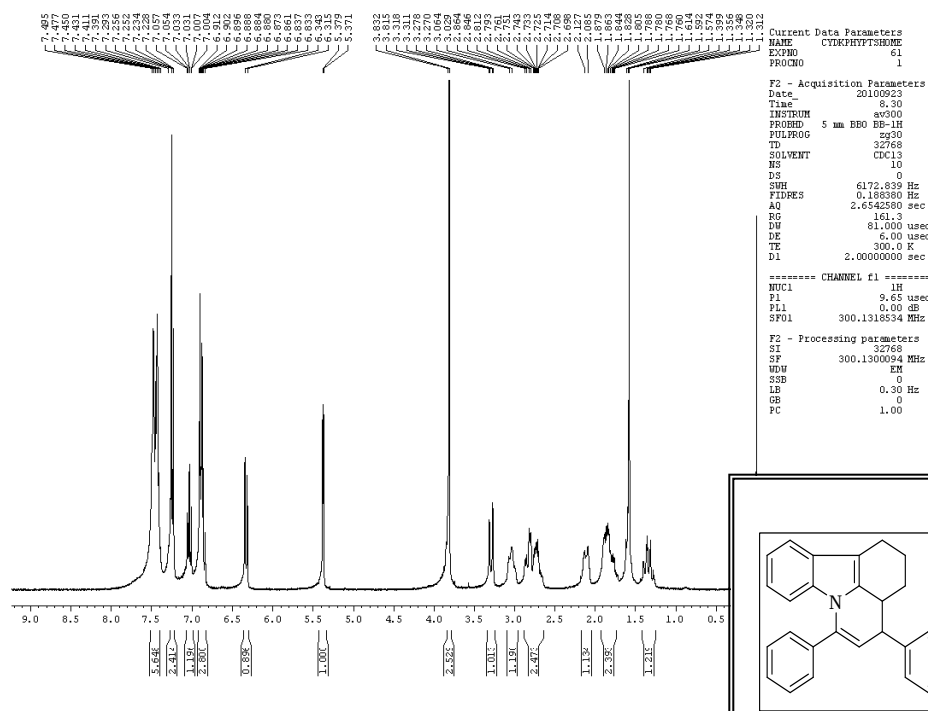


Figure 7. ^{13}C NMR Spectrum of 3b (CDCl_3)



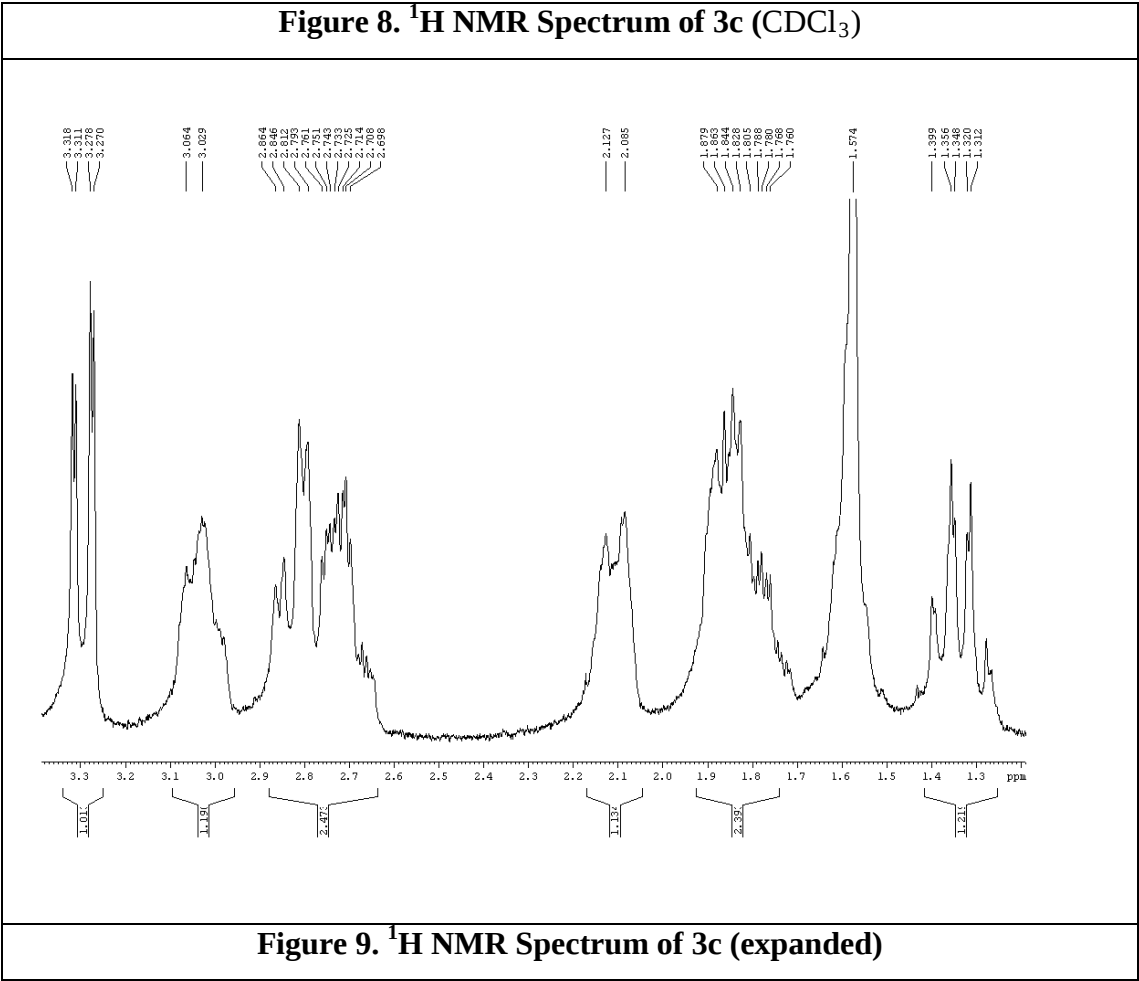


Figure 10. ^1H NMR Spectrum of 3c (expanded)

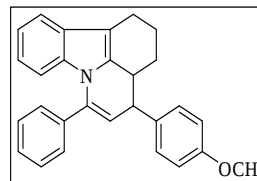
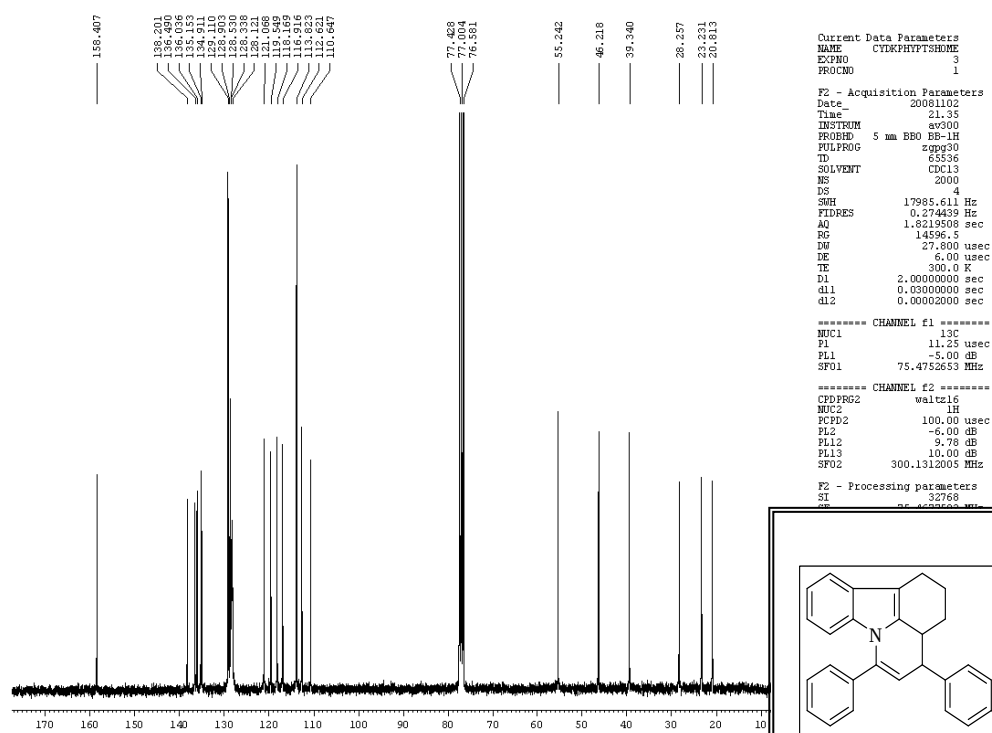
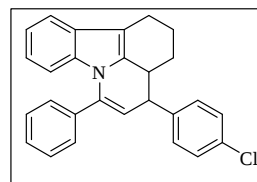
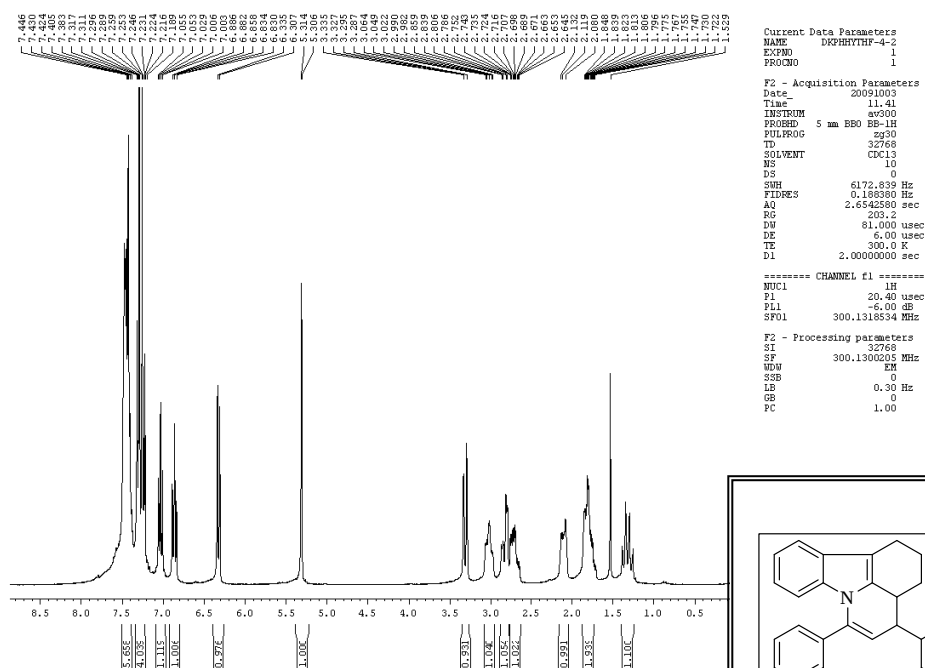


Figure 11. ^{13}C NMR Spectrum of 3c (CDCl_3)



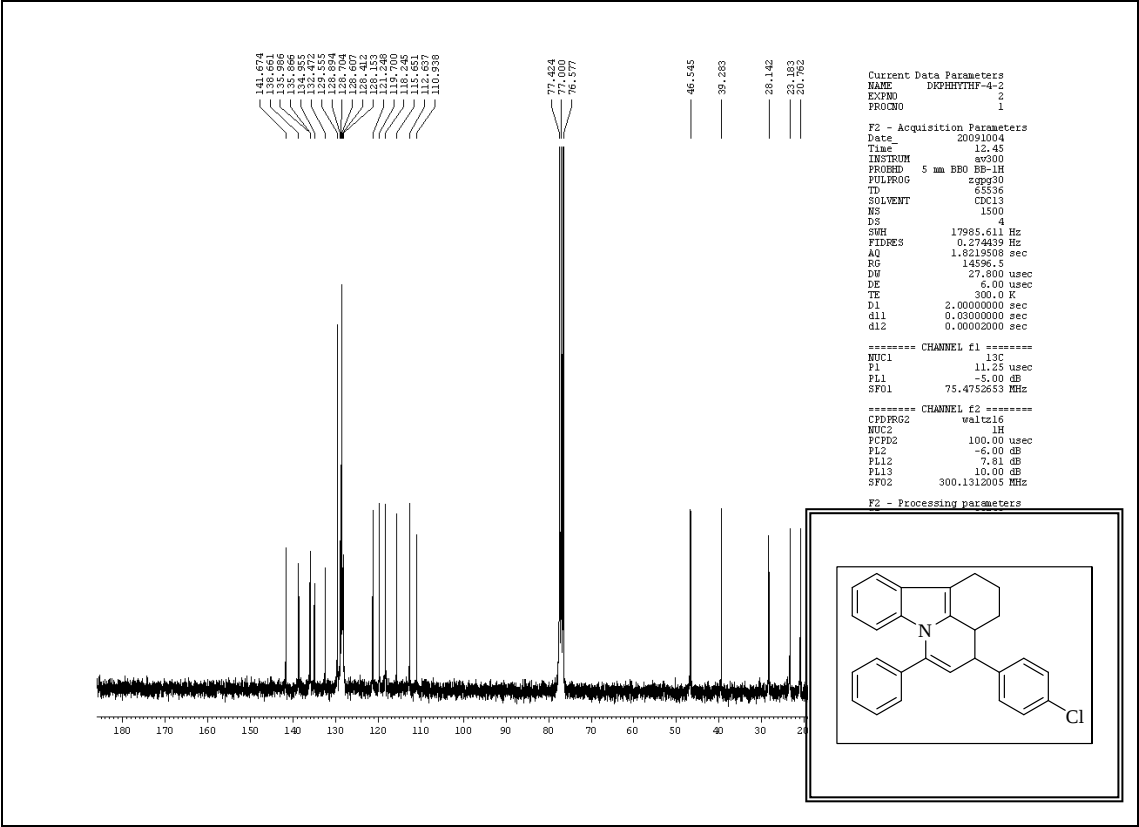
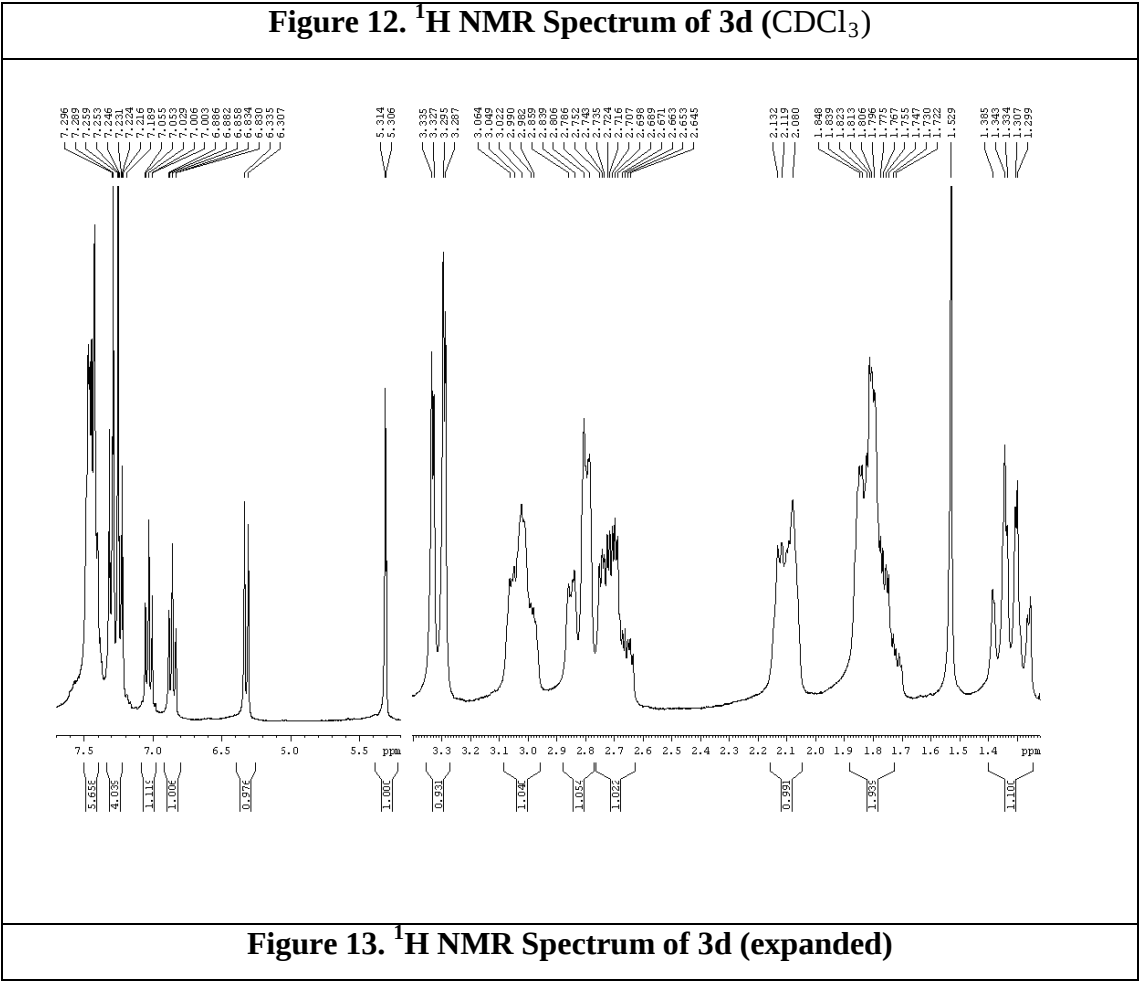


Figure 14. ^{13}C NMR Spectrum of 3d (CDCl_3)

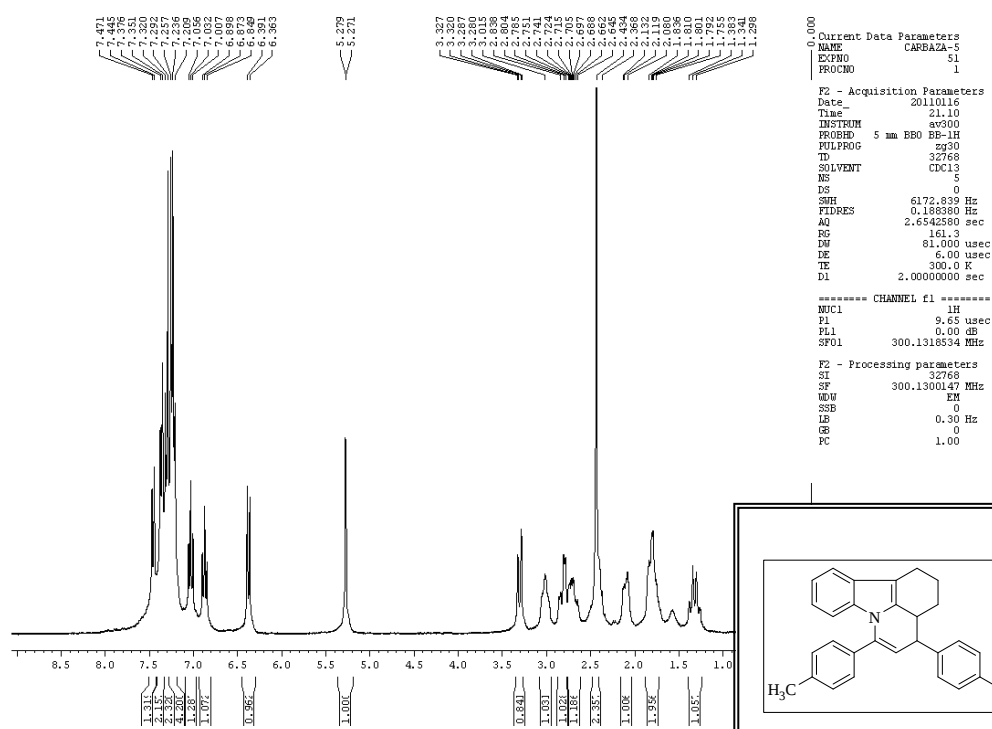


Figure 15. ^1H NMR Spectrum of 3e (CDCl_3)

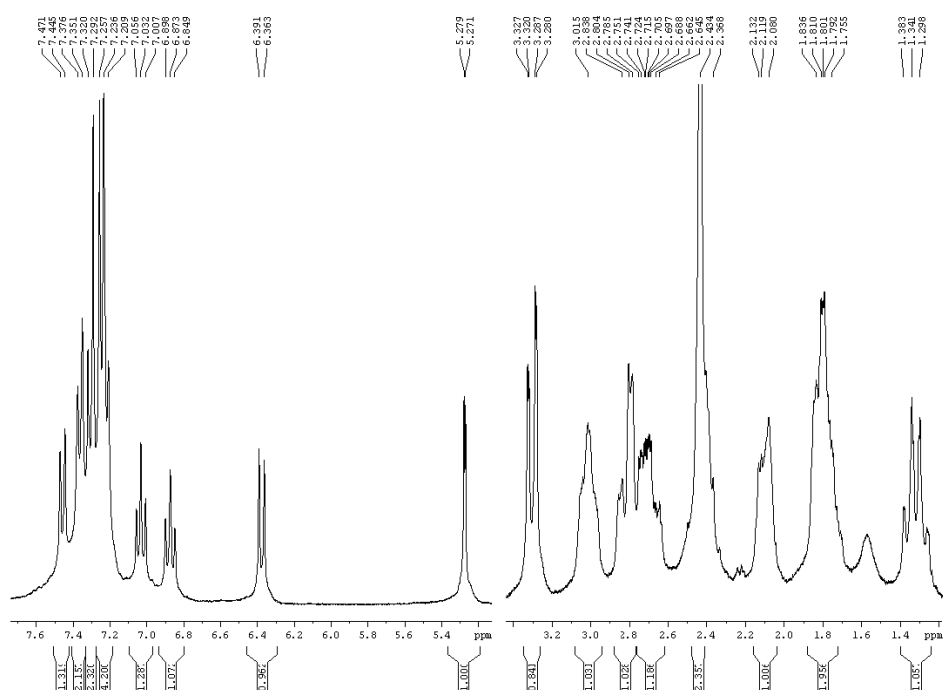


Figure 16. ^1H NMR Spectrum of 3e (expanded)

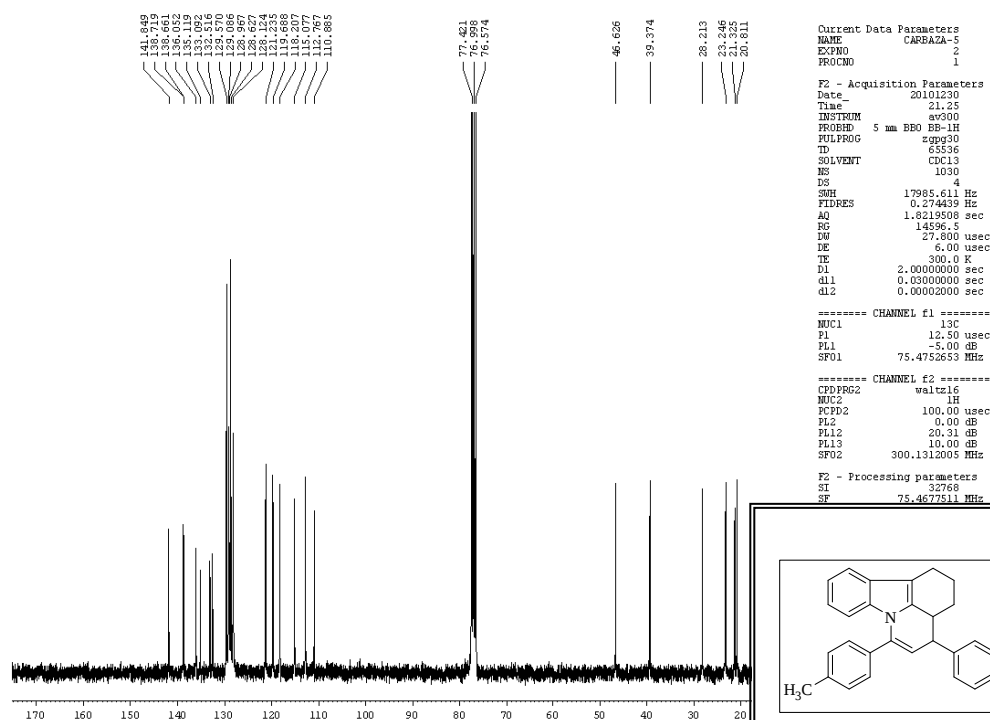


Figure 17. ^{13}C NMR Spectrum of 3e (CDCl_3)

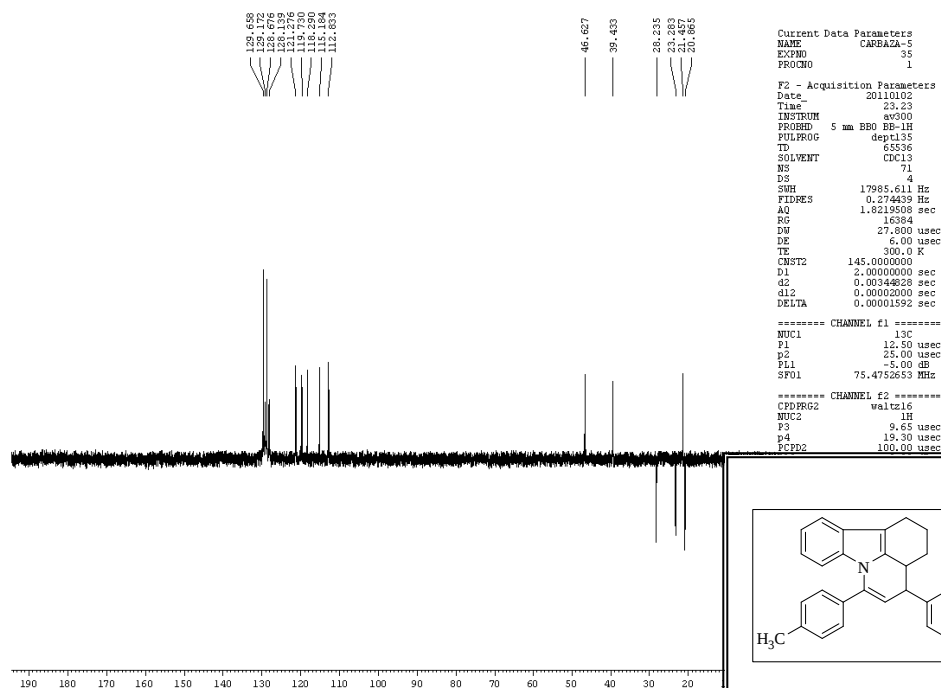


Figure 18. DEPT-135 Spectrum of 3e (CDCl₃)

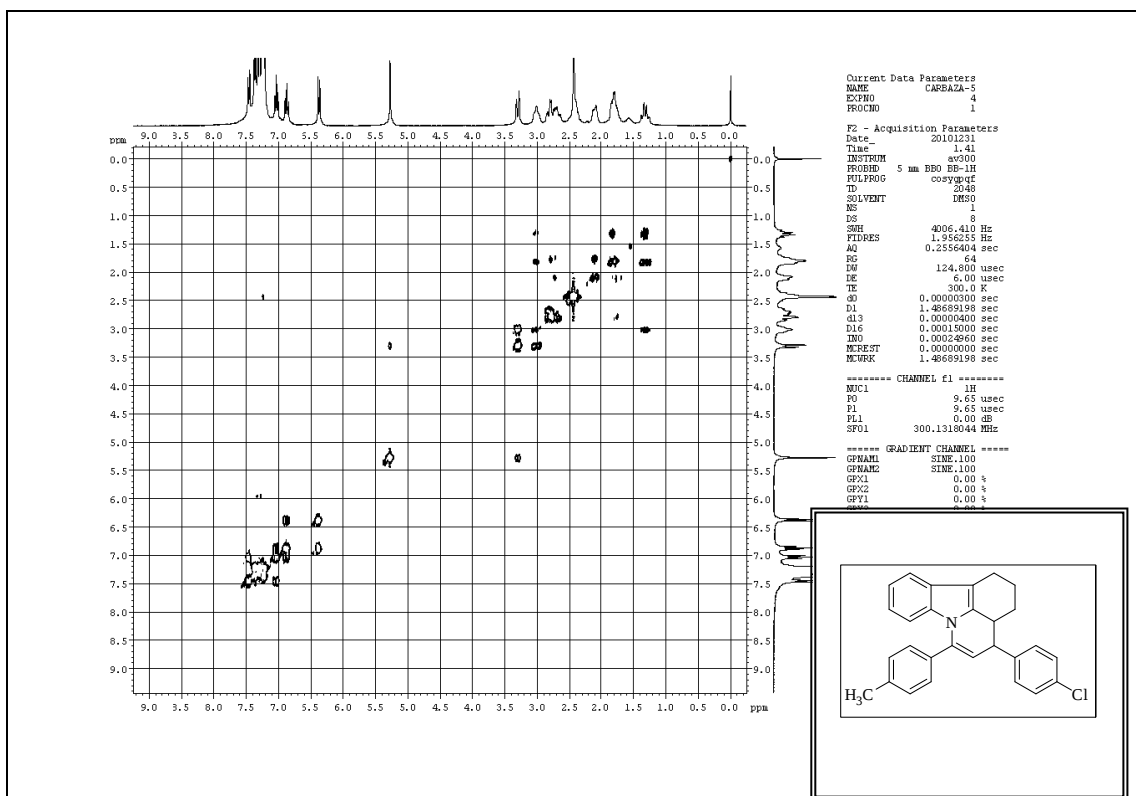


Figure 19. H,H-COSY Spectrum of 3e

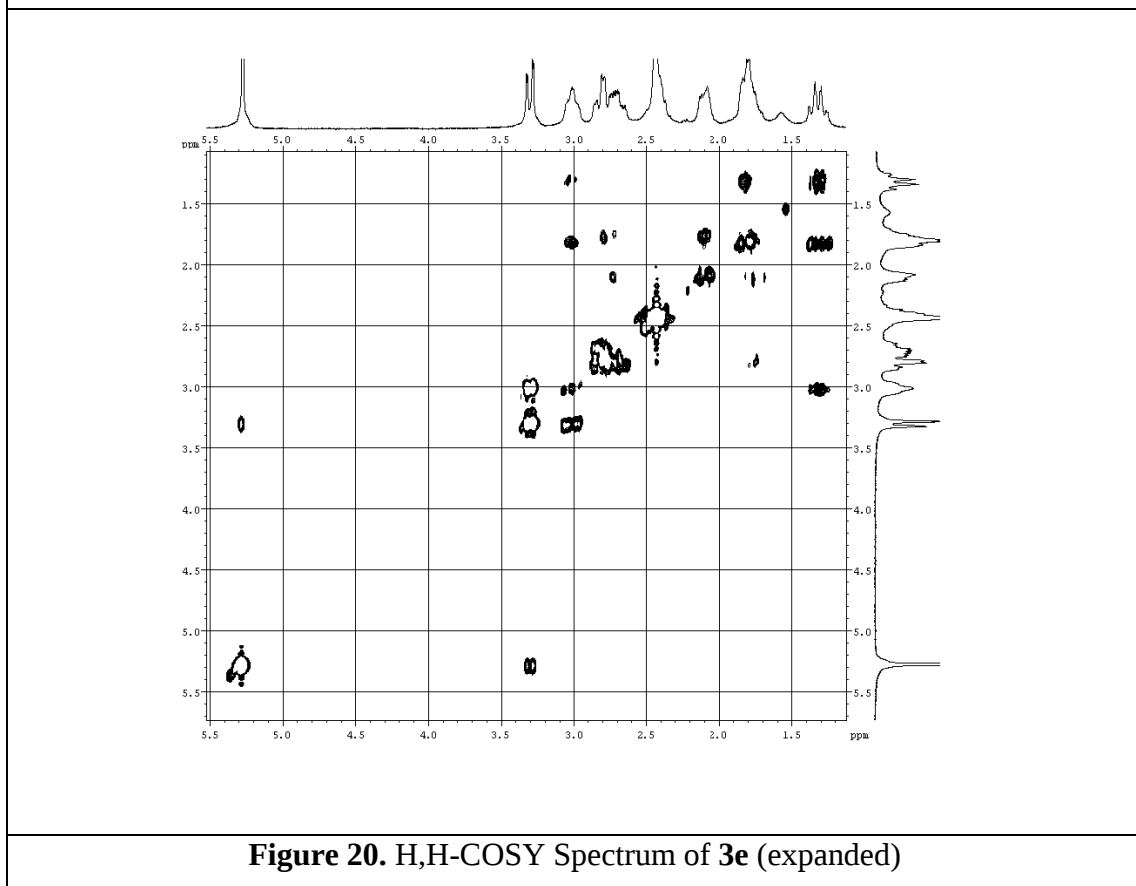


Figure 20. H,H-COSY Spectrum of 3e (expanded)

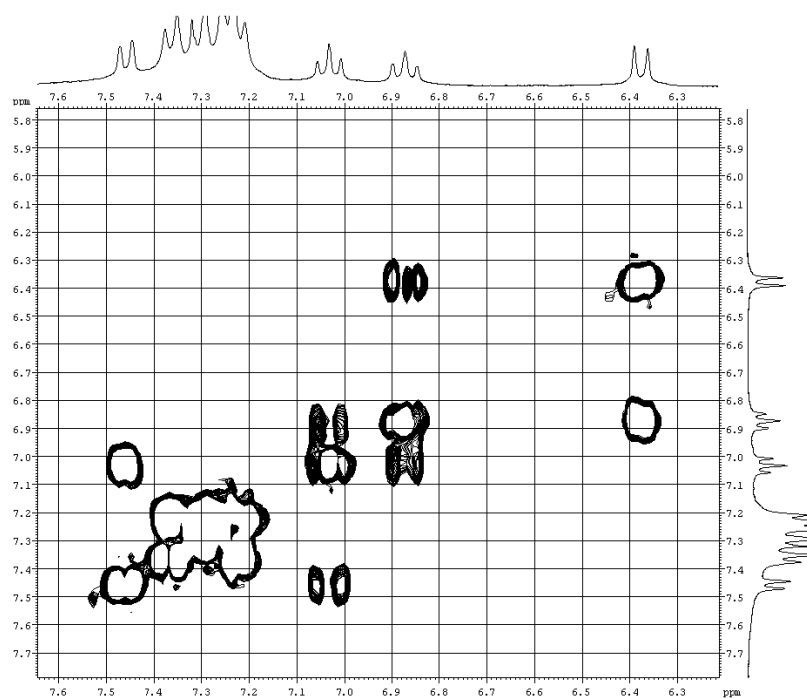


Figure 21. H,H-COSY Spectrum of 3e (expanded)

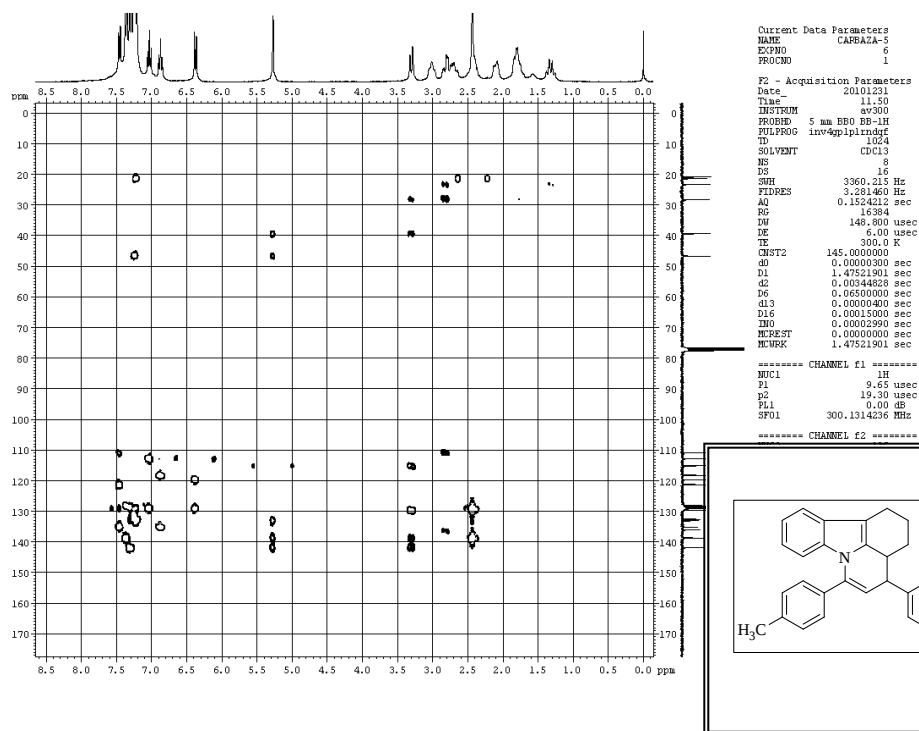


Figure 22. HMBC Spectrum of 3e

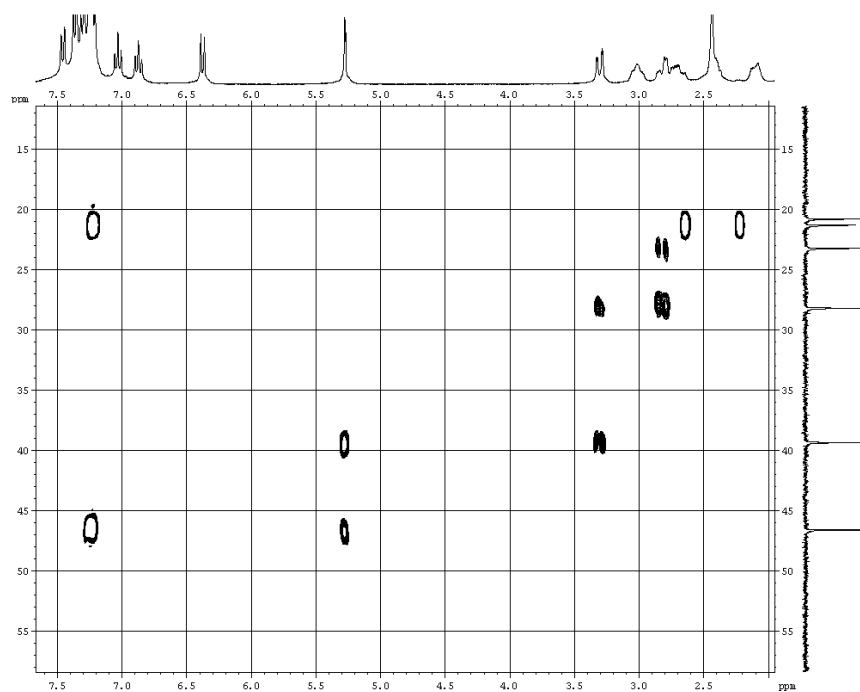


Figure 23. HMBC Spectrum of 3e (expanded)

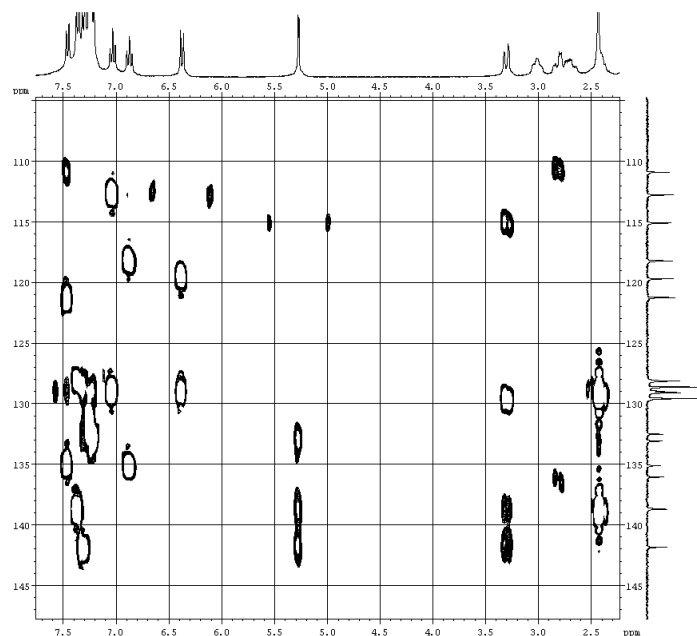


Figure 24. HMBC Spectrum of 3e (expanded)

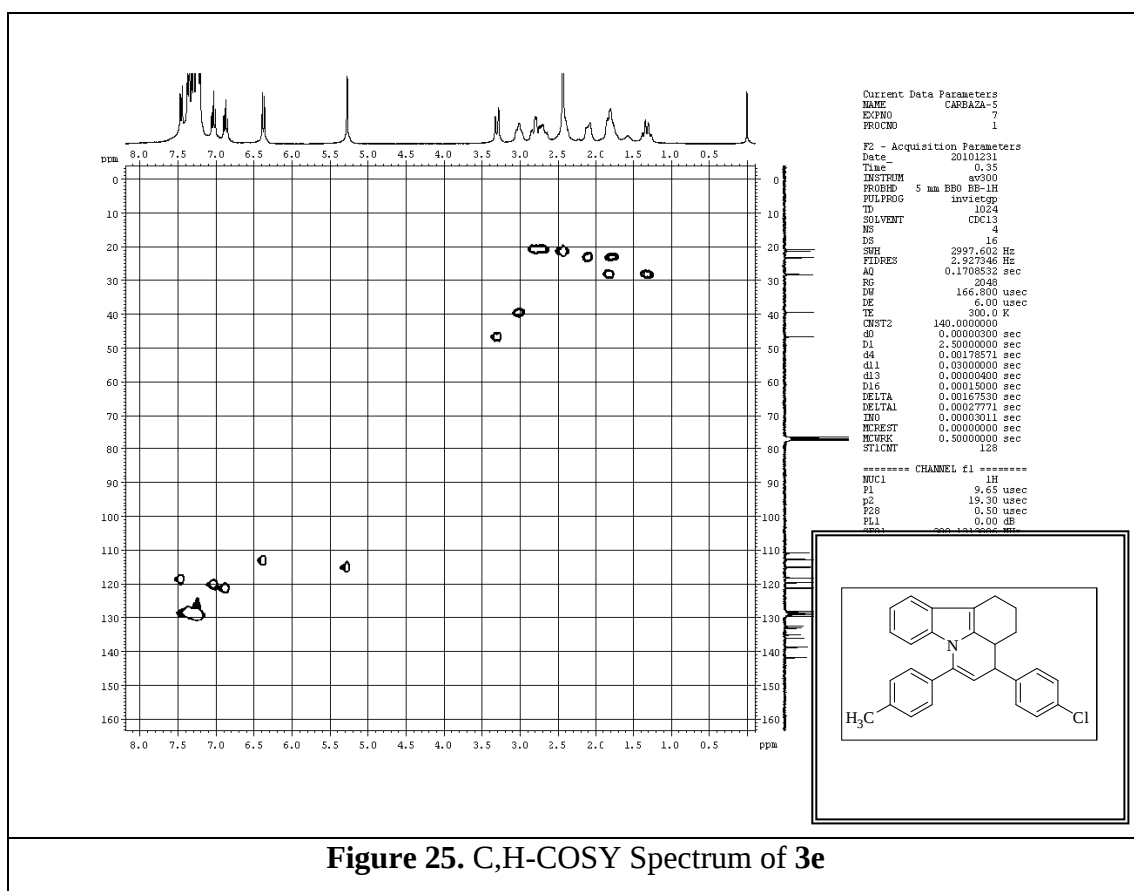


Figure 25. C,H-COSY Spectrum of 3e

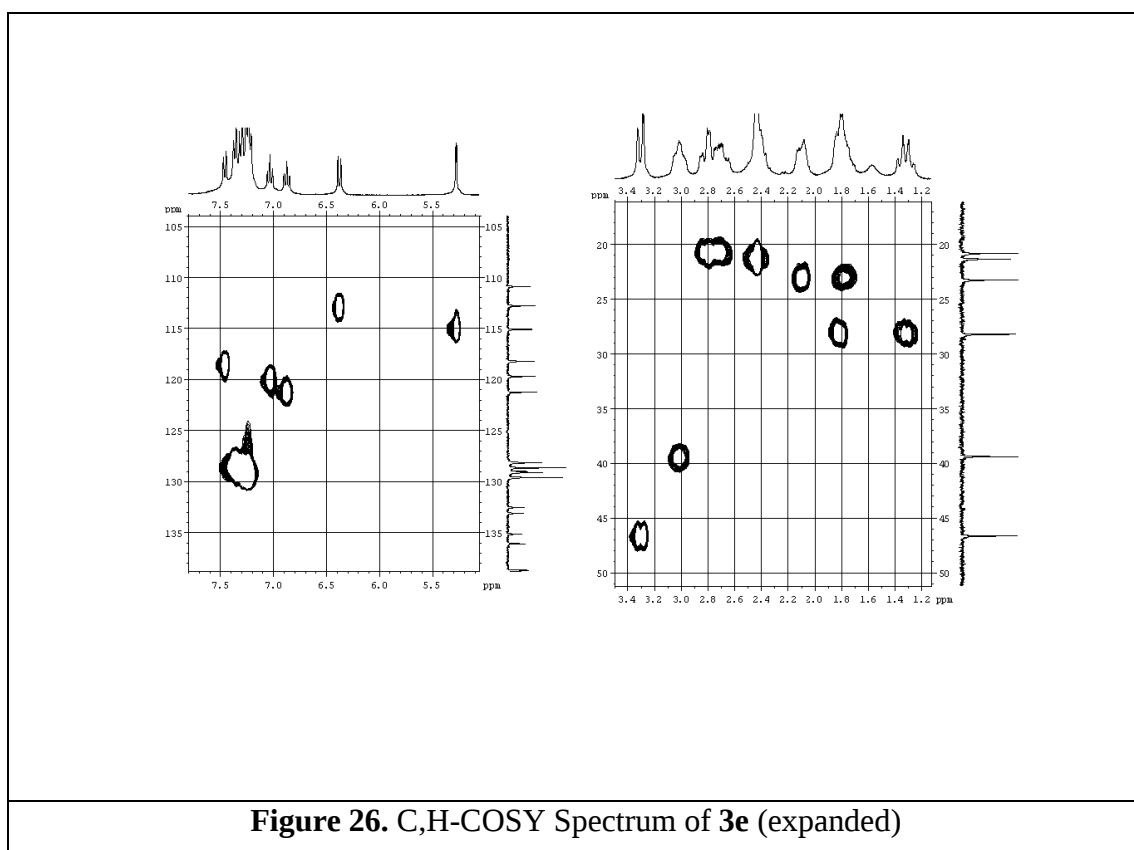


Figure 26. C,H-COSY Spectrum of 3e (expanded)

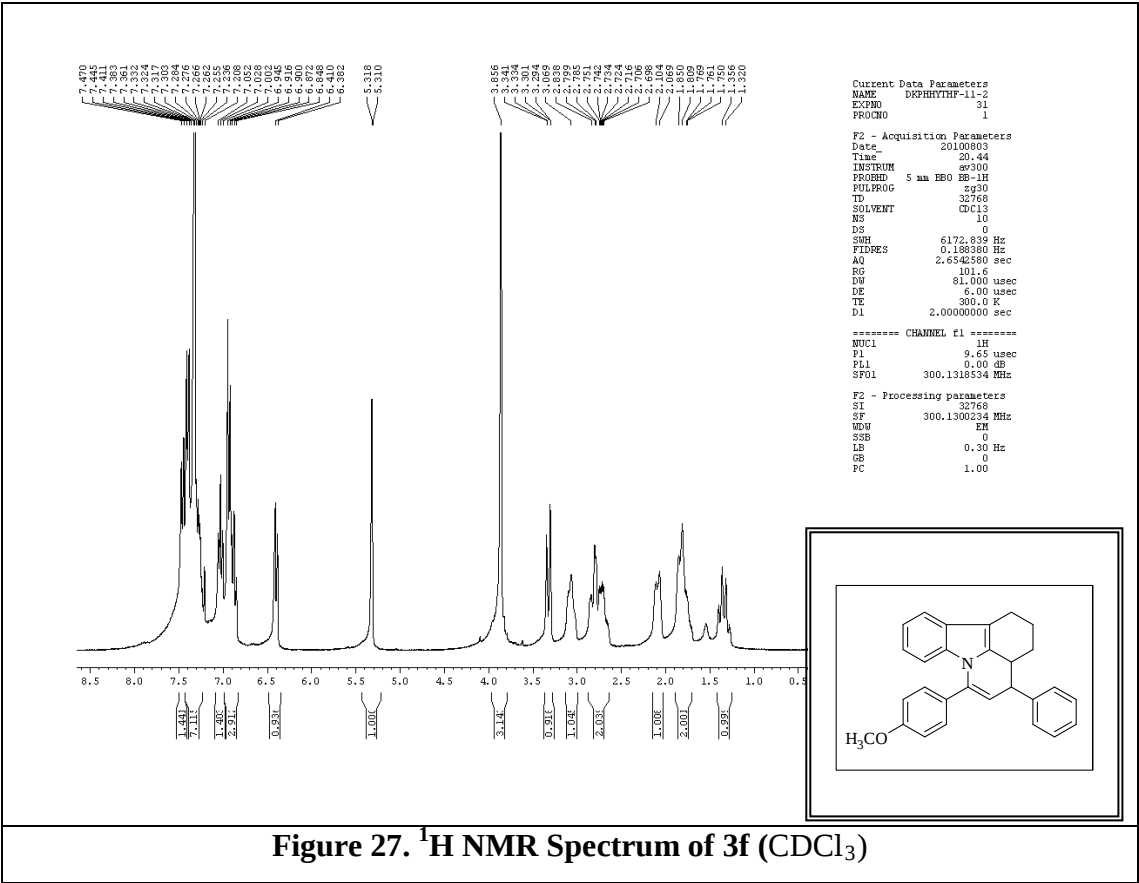


Figure 27. ¹H NMR Spectrum of 3f (CDCl₃)

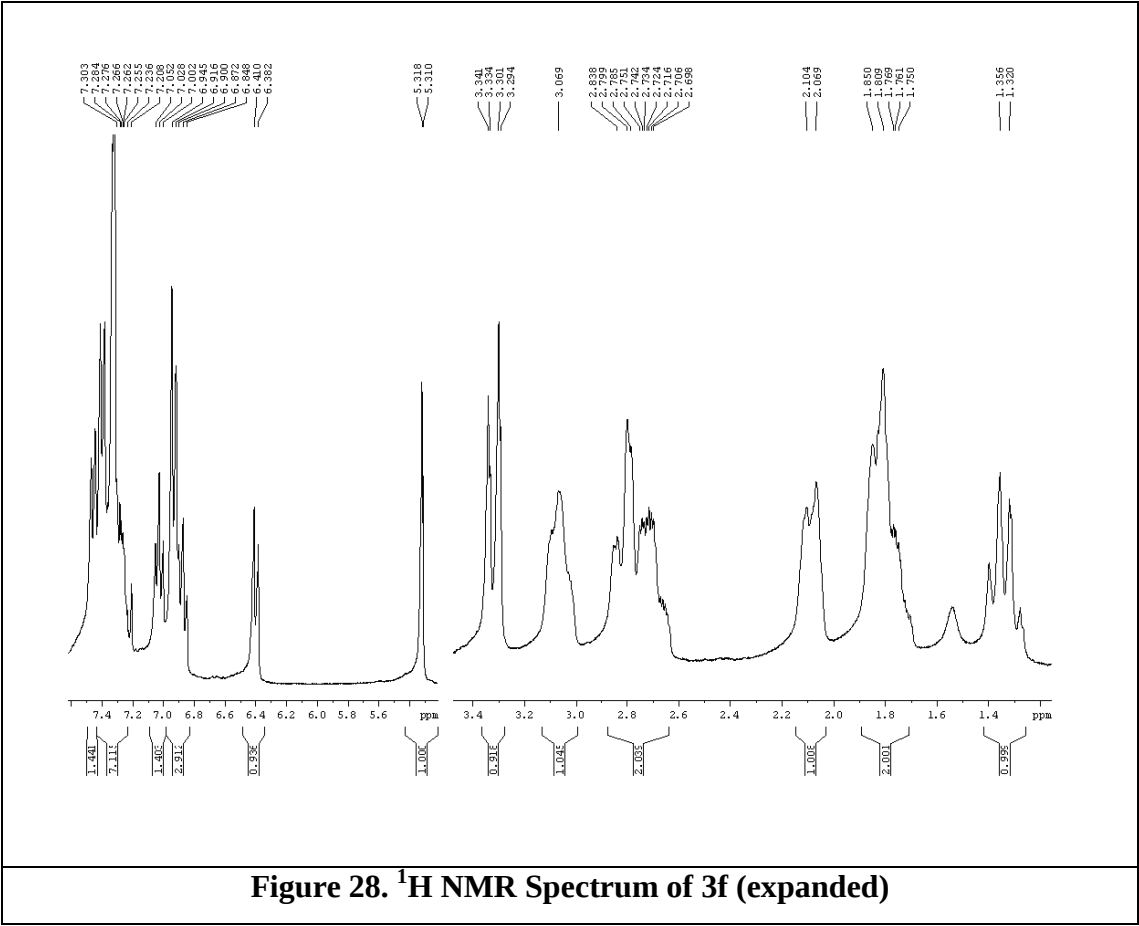
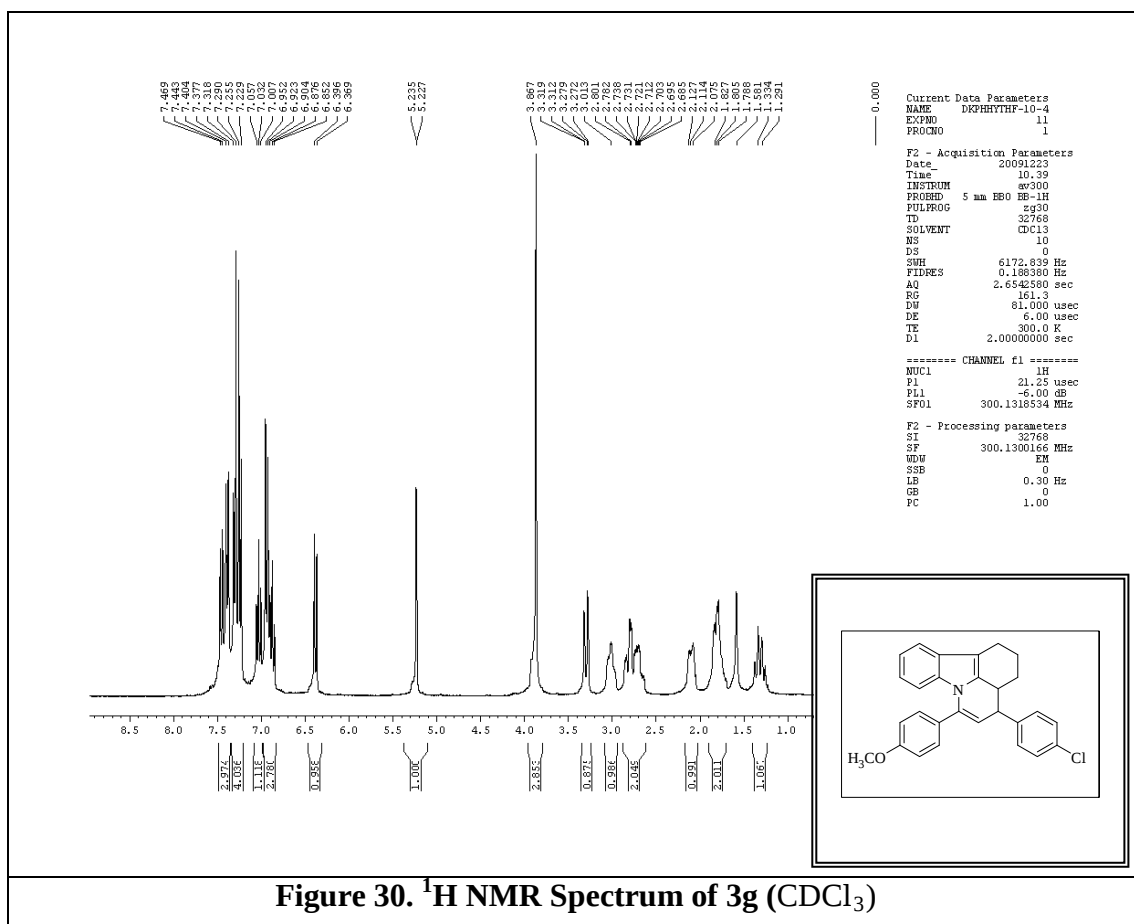
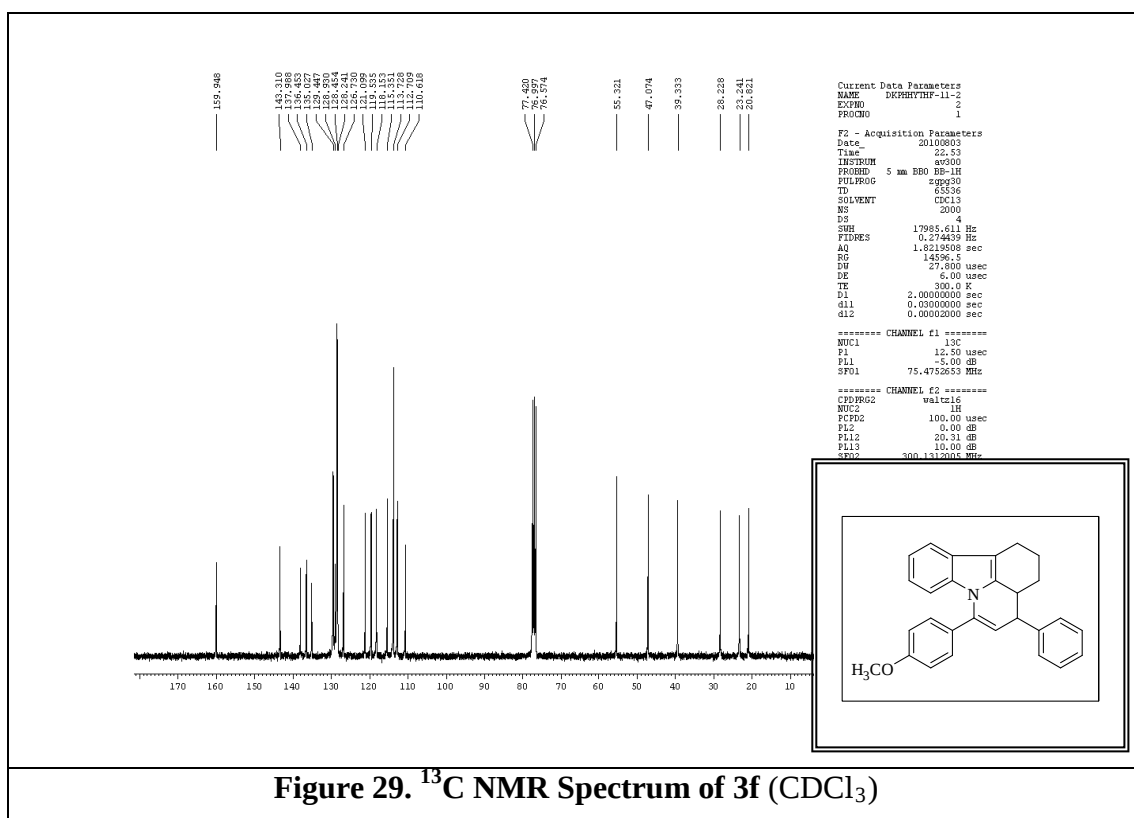


Figure 28. ¹H NMR Spectrum of 3f (expanded)



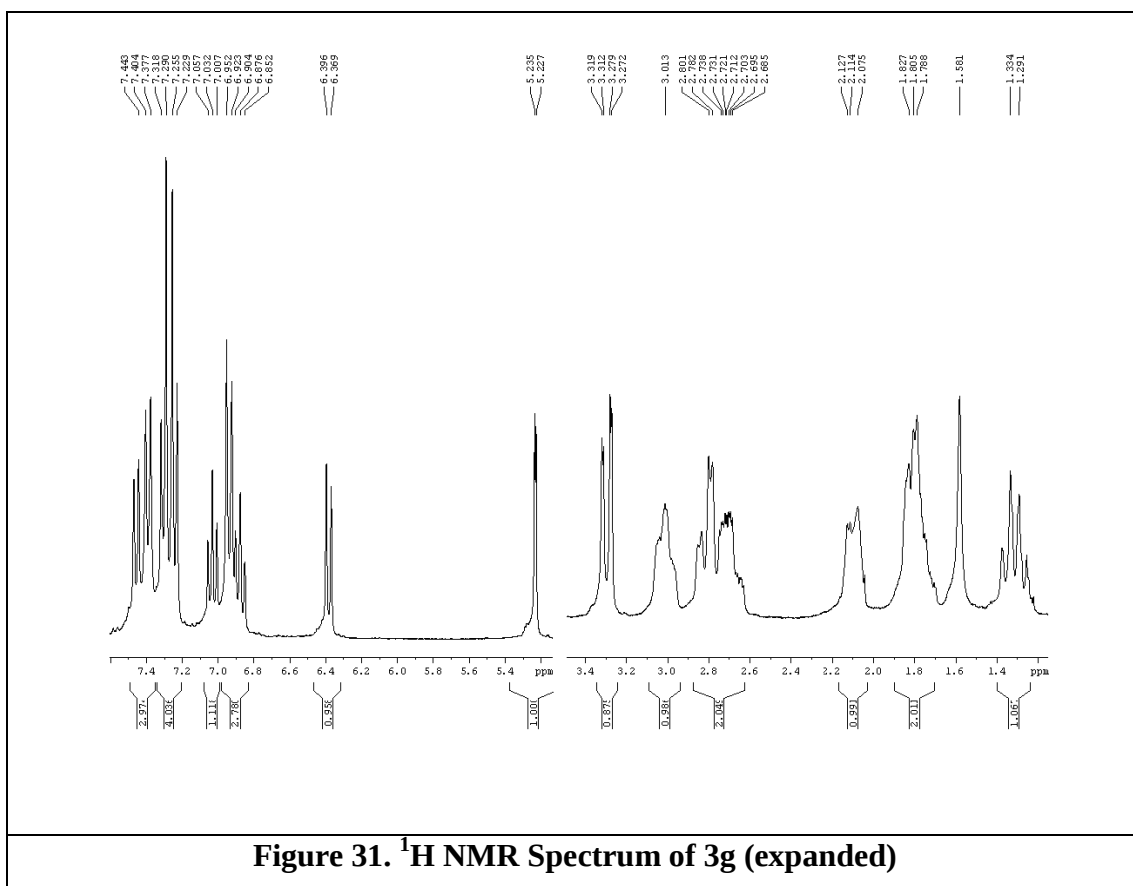


Figure 31. ^1H NMR Spectrum of 3g (expanded)

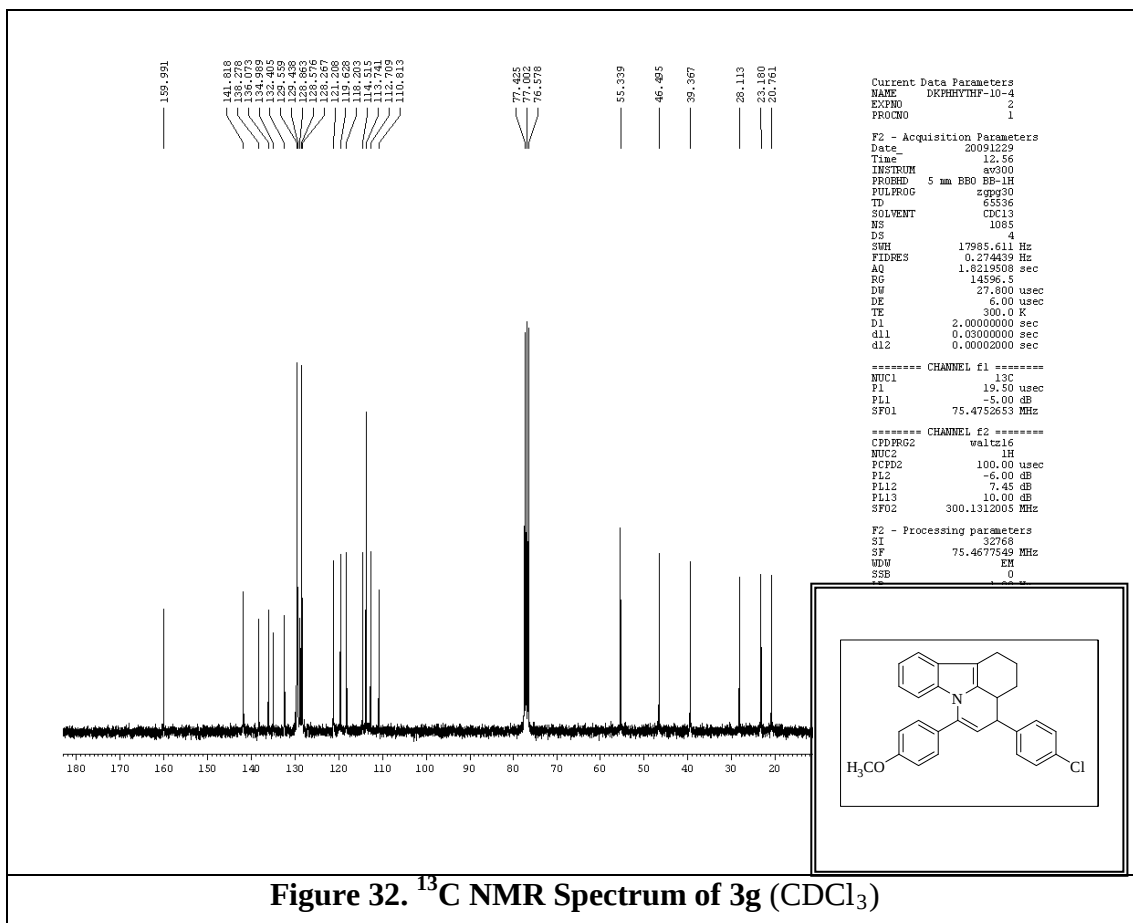
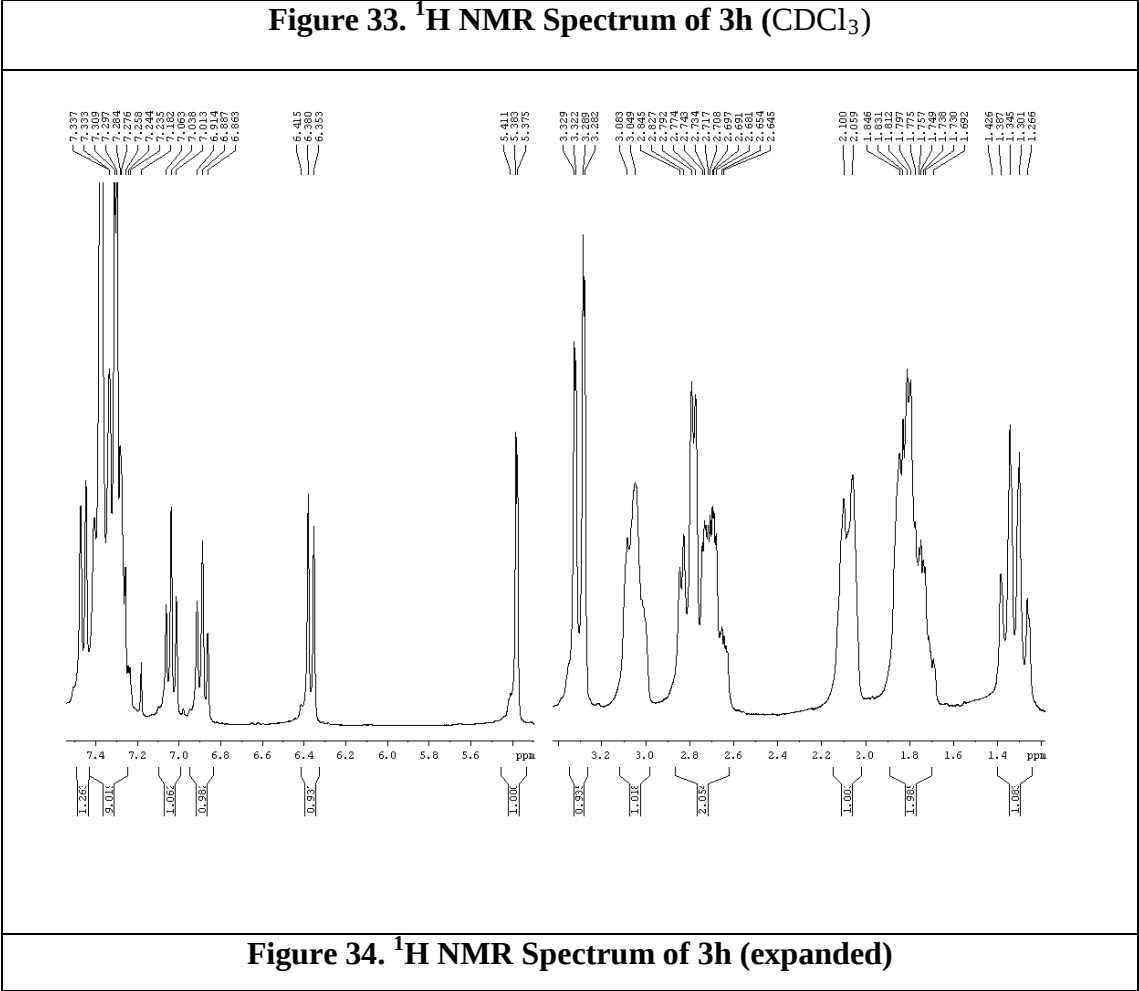
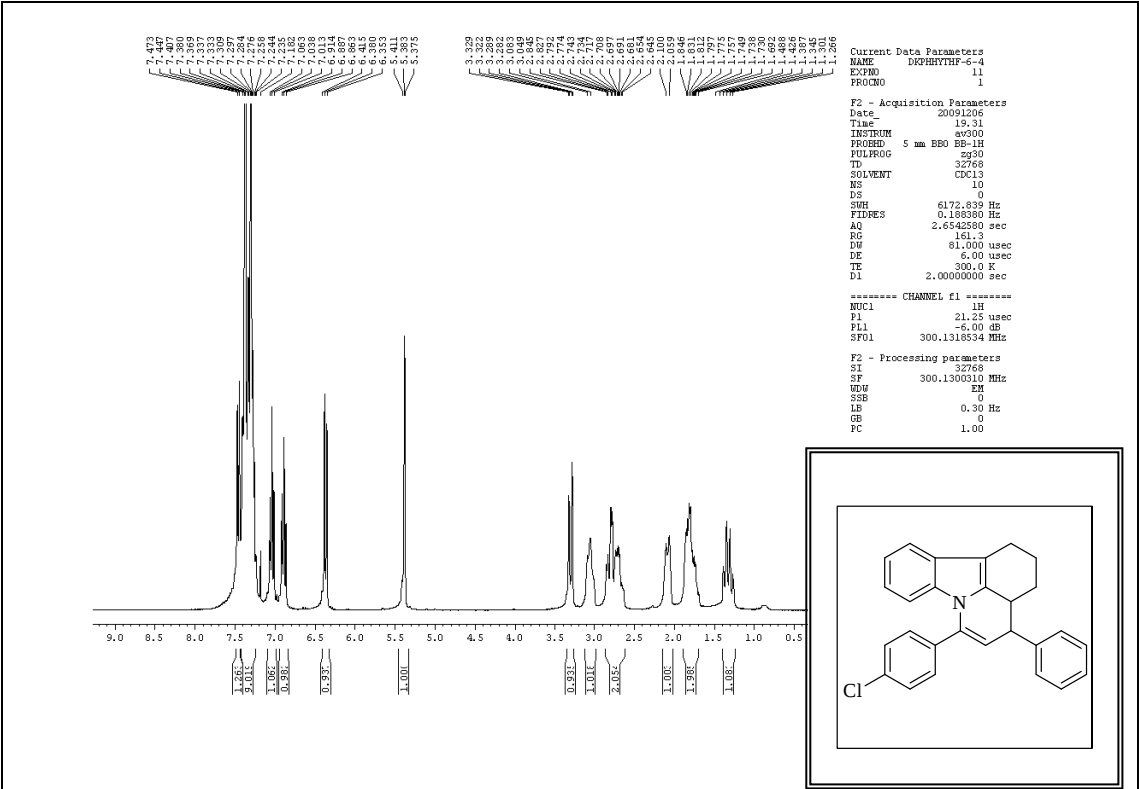
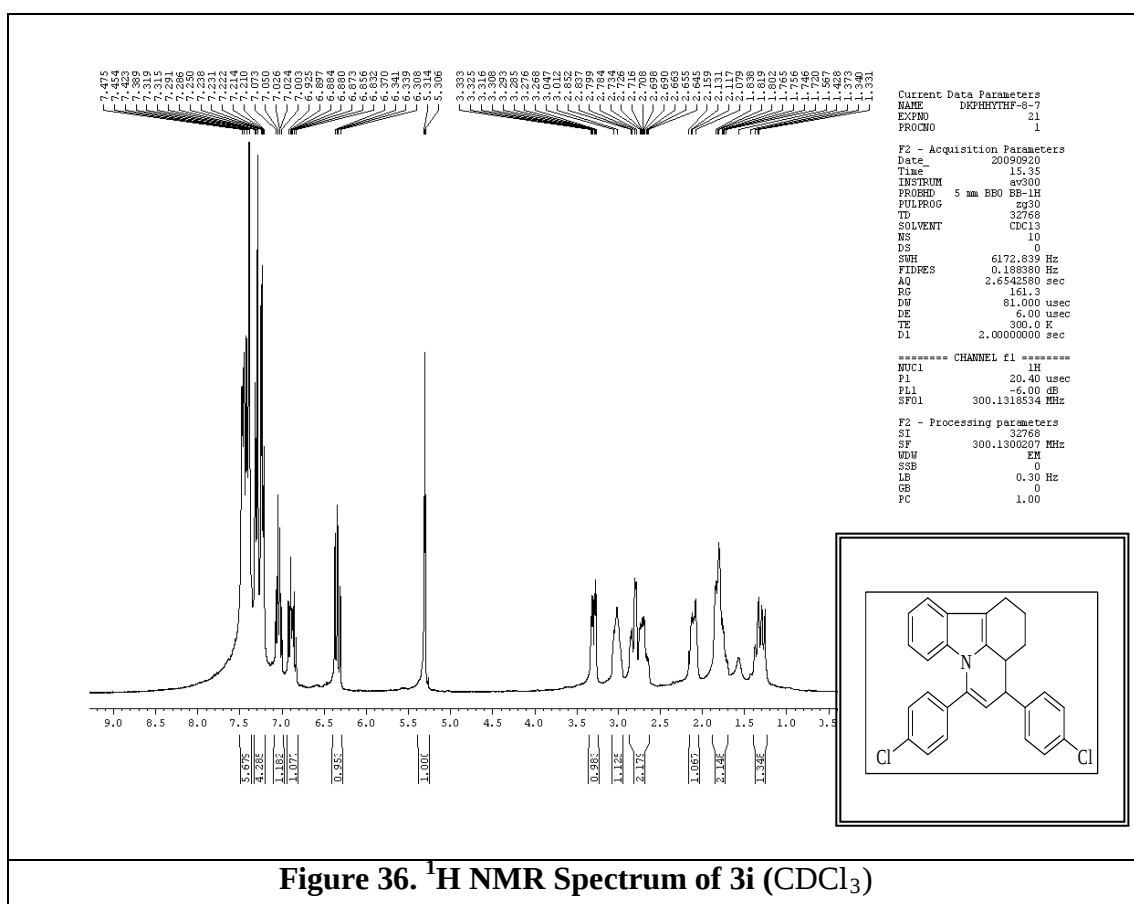
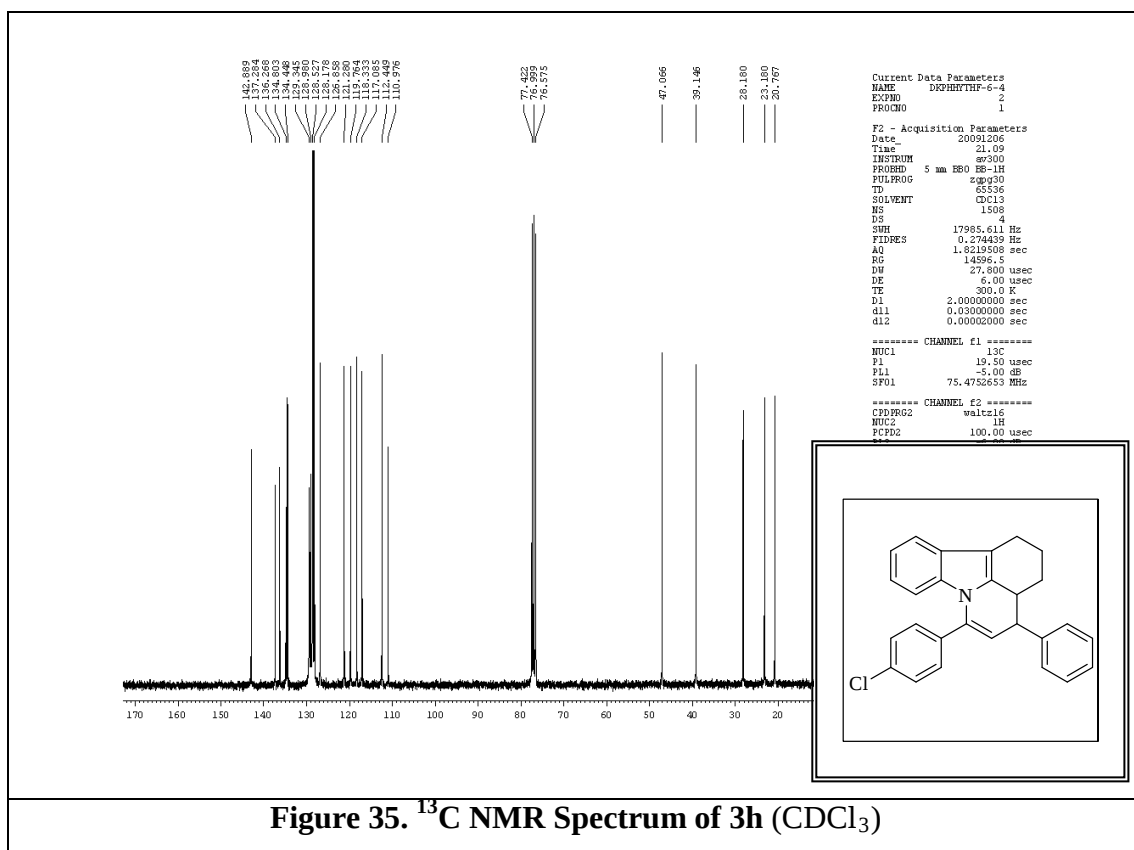
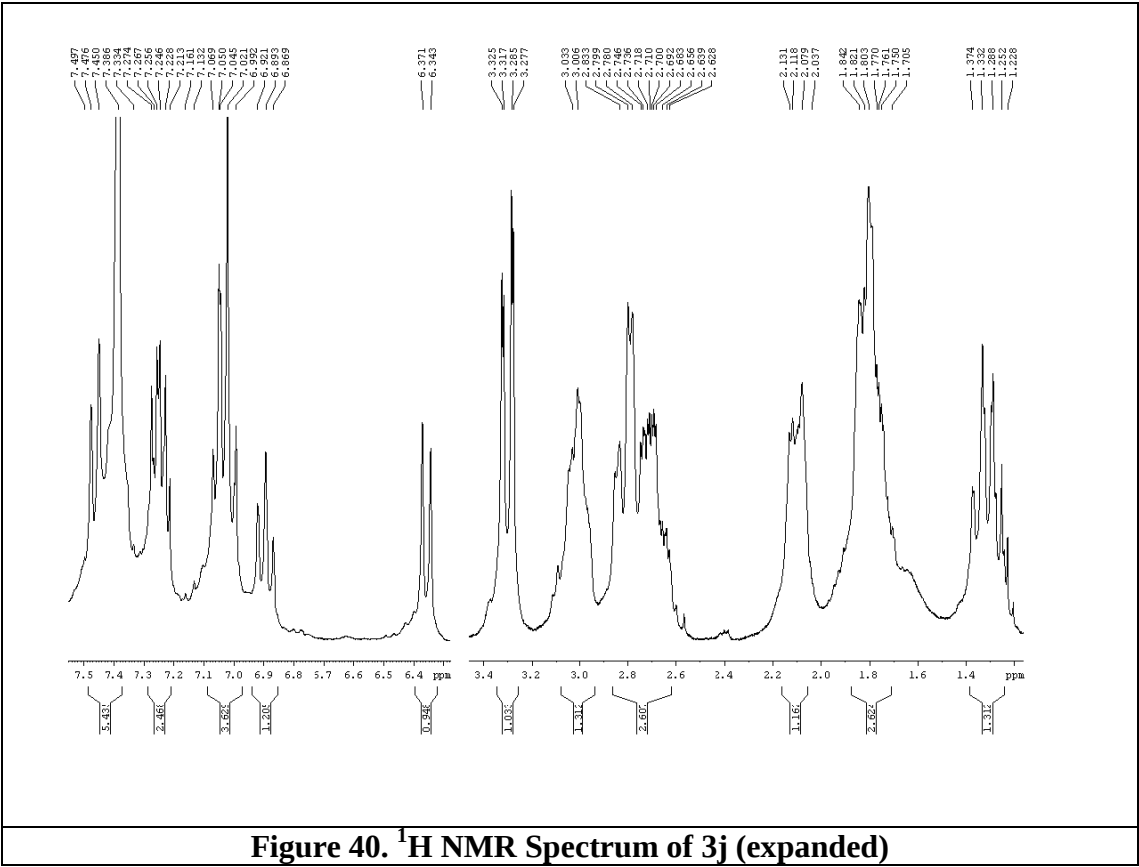
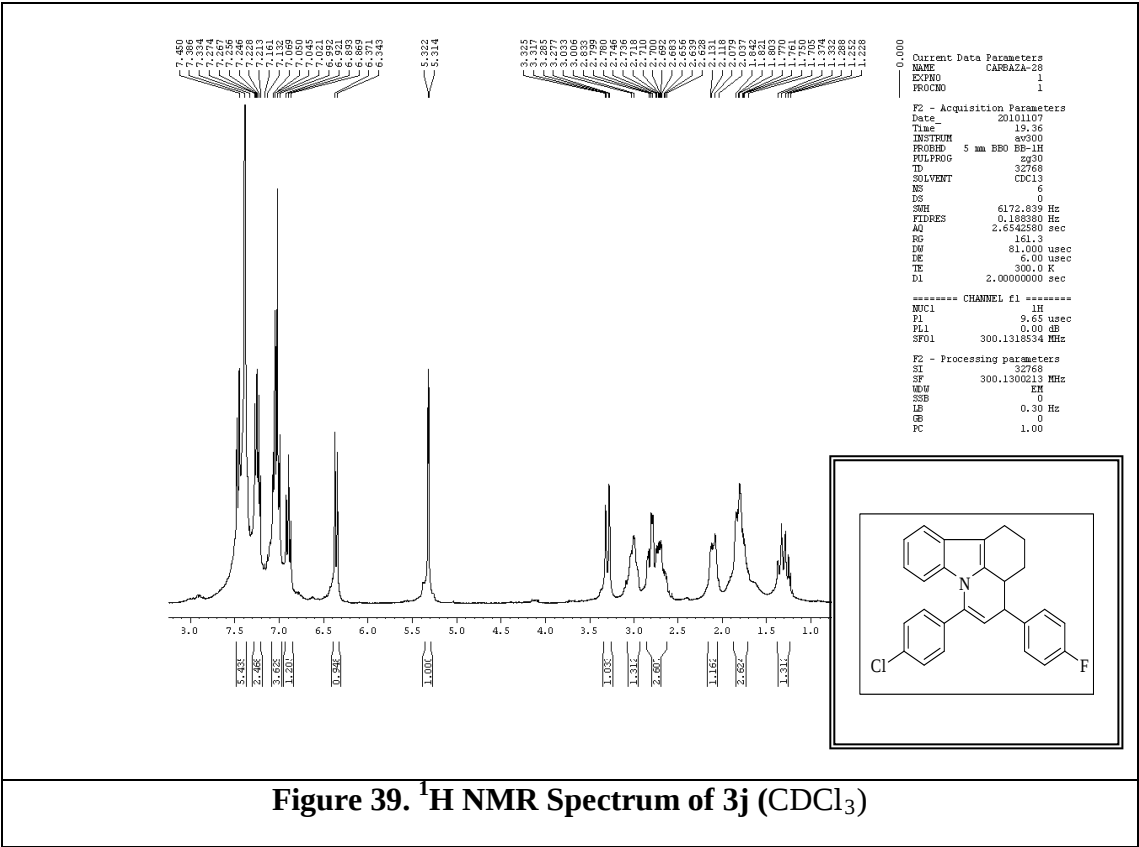


Figure 32. ^{13}C NMR Spectrum of 3g (CDCl_3)









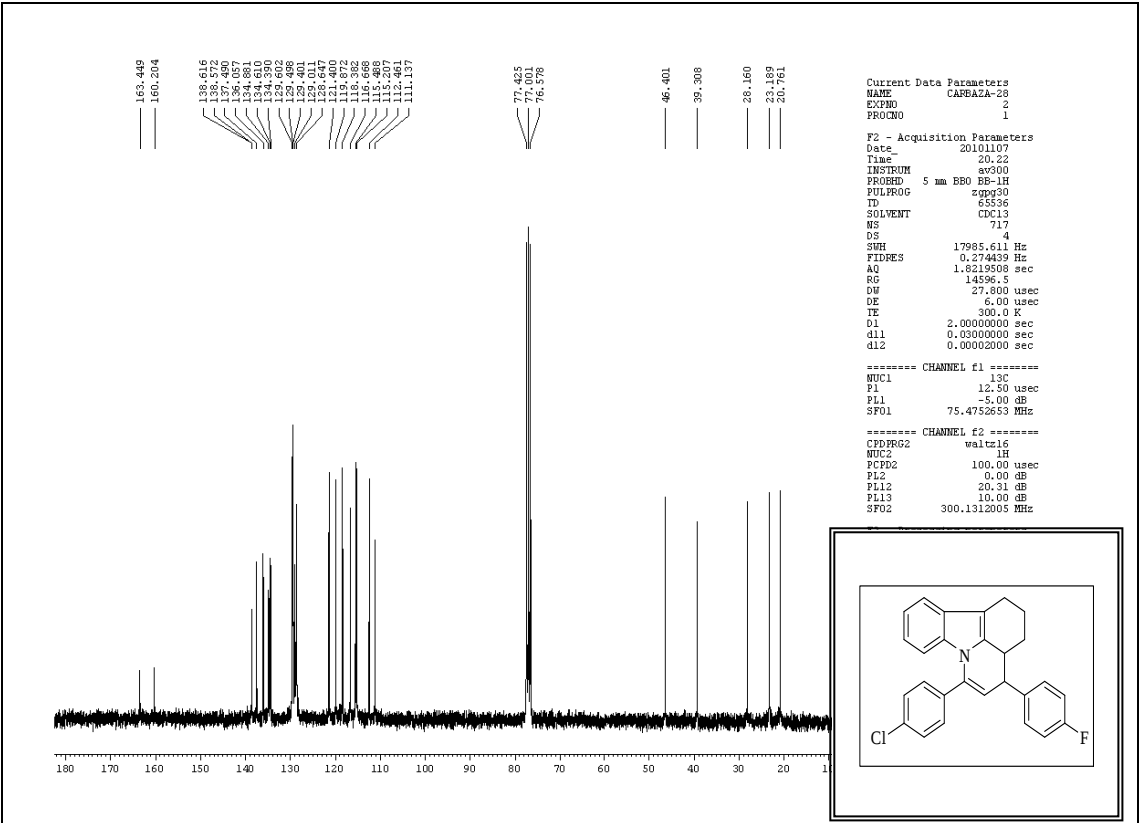


Figure 41. ¹³C NMR Spectrum of 3j (CDCl₃)

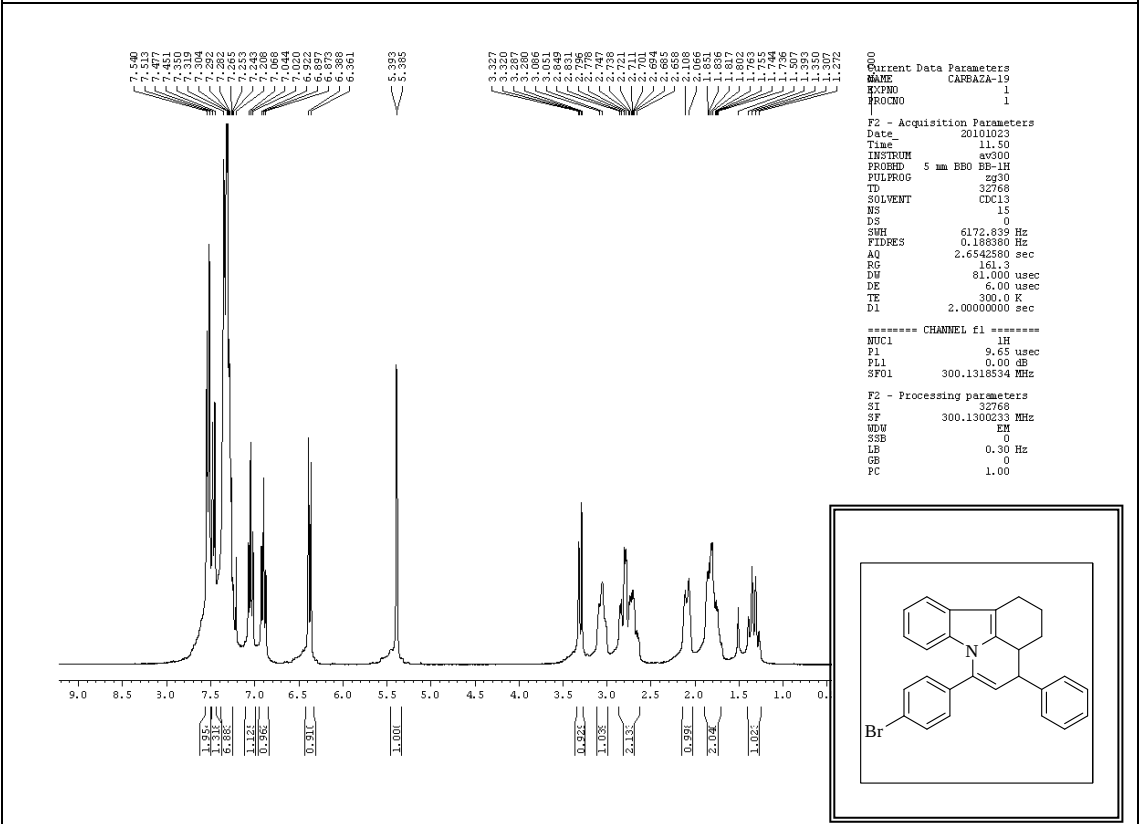


Figure 42. ¹H NMR Spectrum of 3k (CDCl₃)

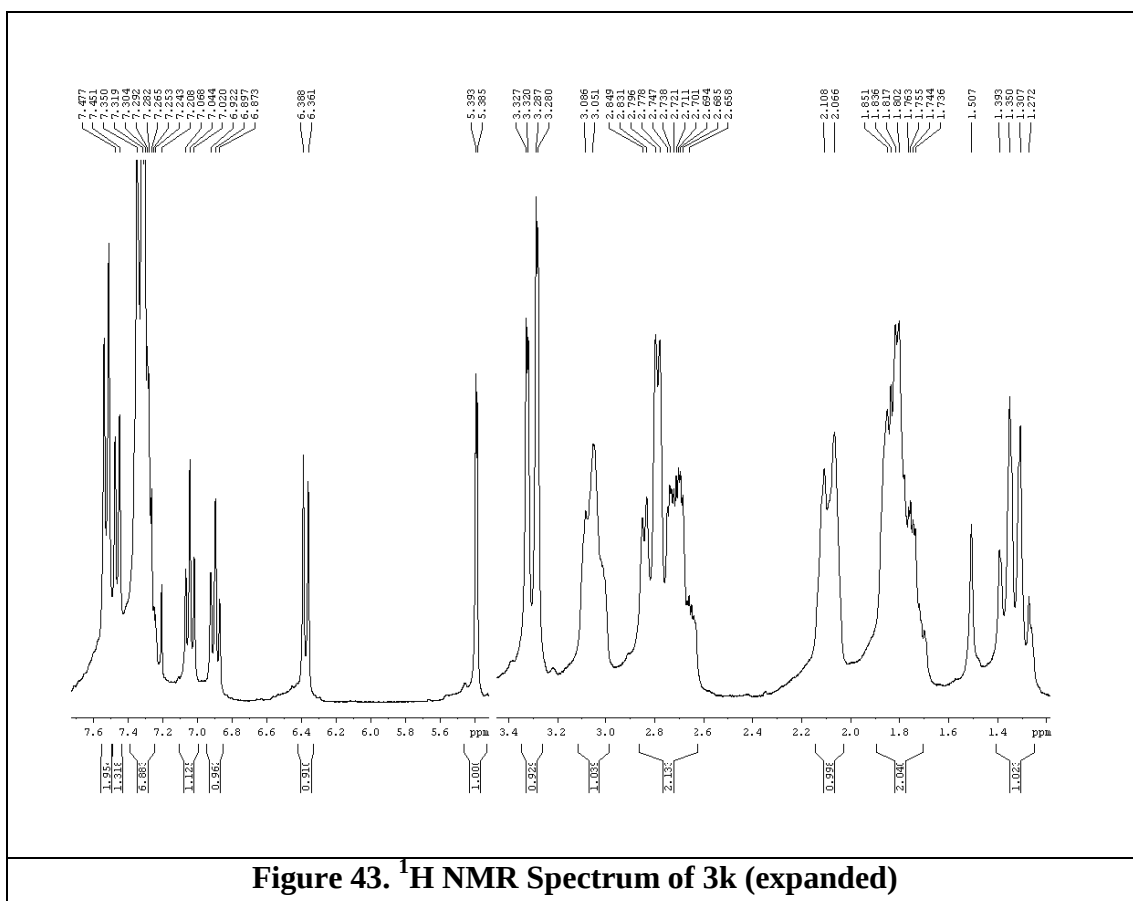


Figure 43. ^1H NMR Spectrum of 3k (expanded)

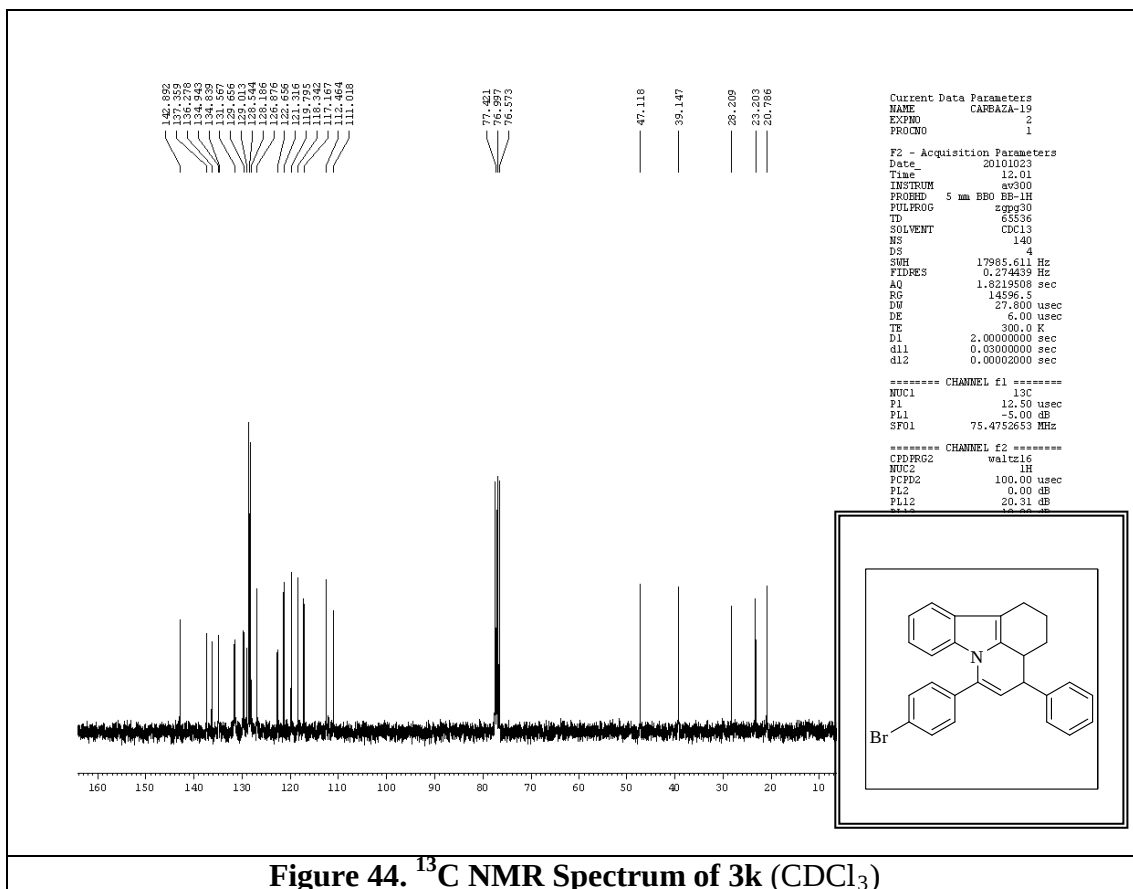


Figure 44. ^{13}C NMR Spectrum of 3k (CDCl₃)

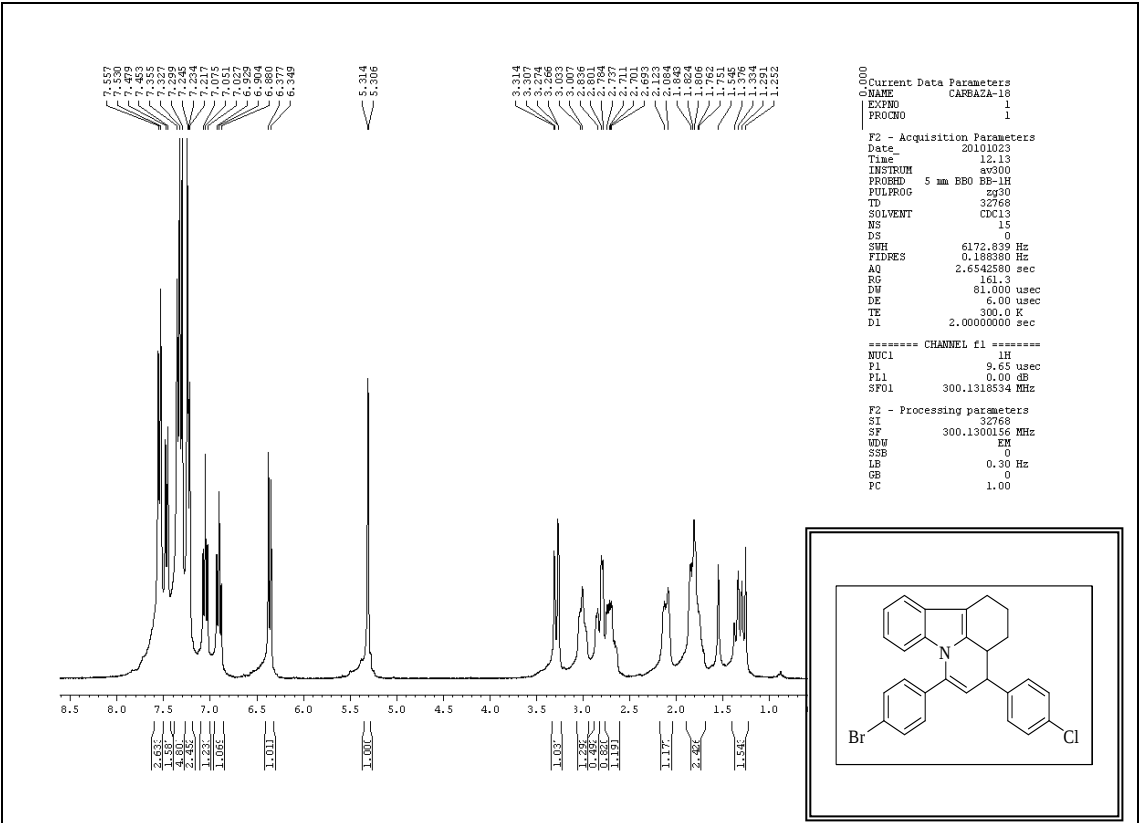


Figure 45. ¹H NMR Spectrum of 3l (CDCl₃)

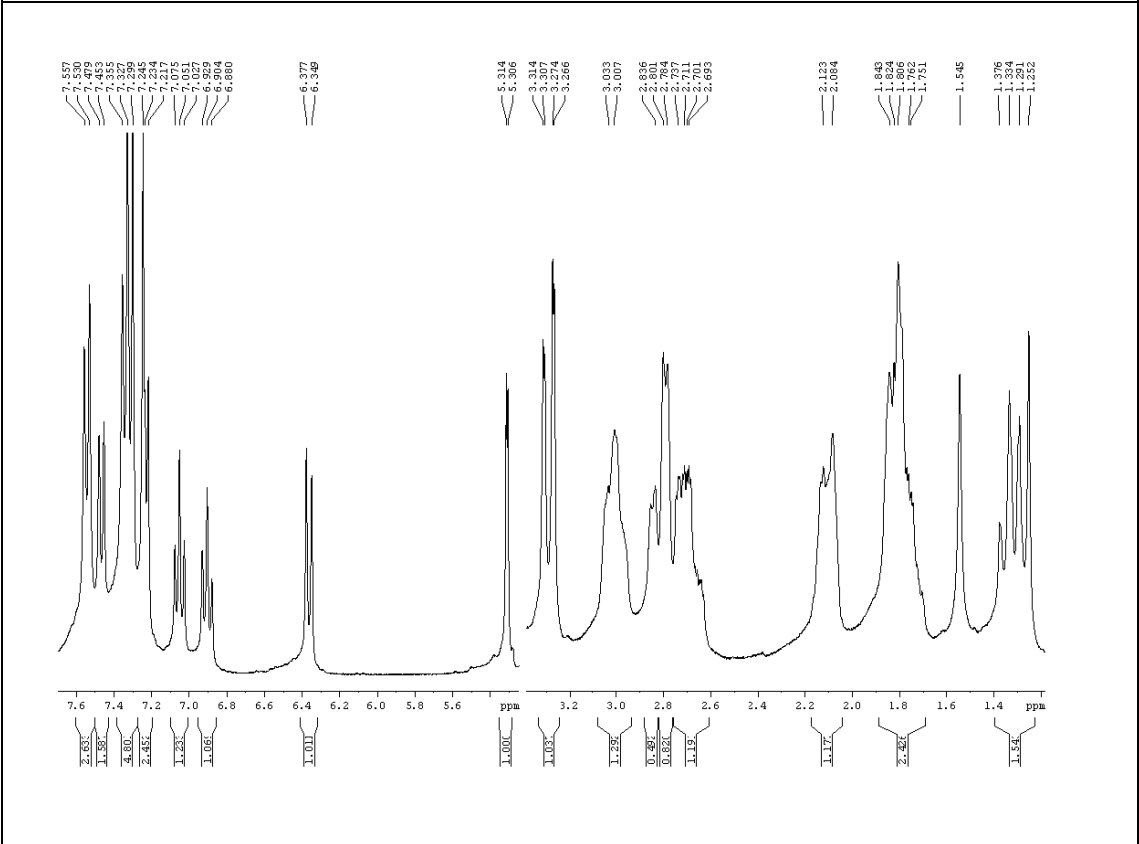


Figure 46. ¹H NMR Spectrum of 3l (expanded)

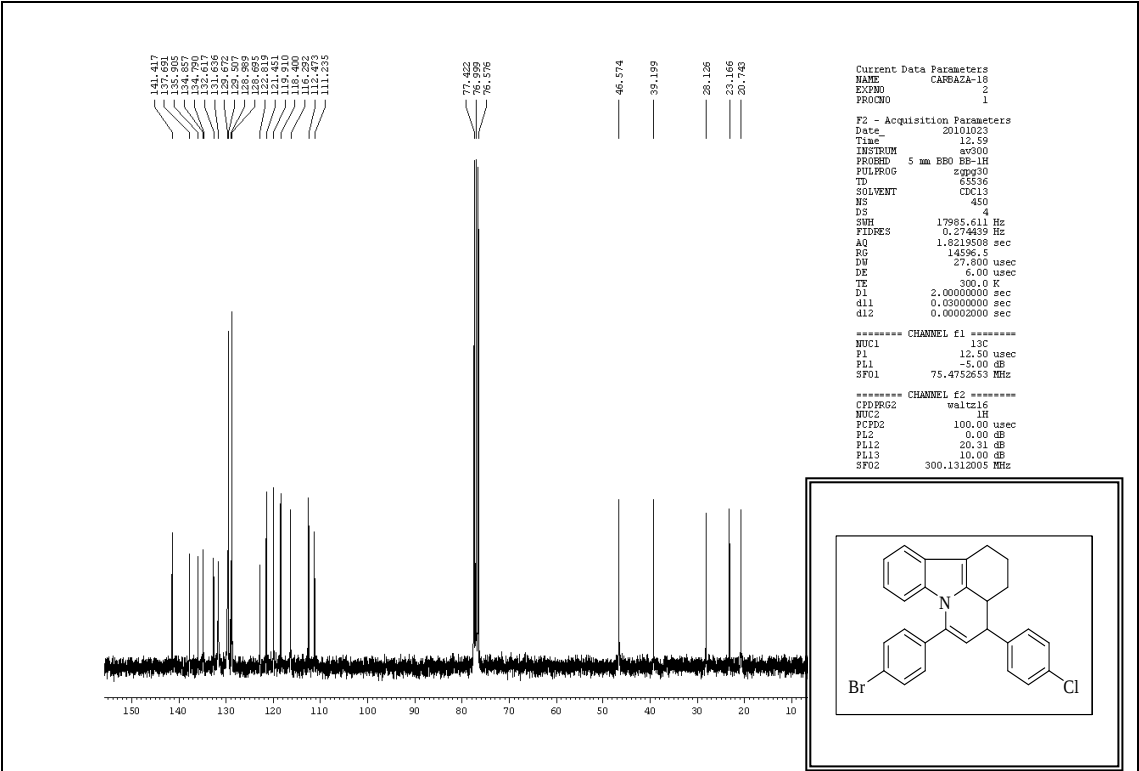


Figure 47. ¹³C NMR Spectrum of 3l (CDCl₃)

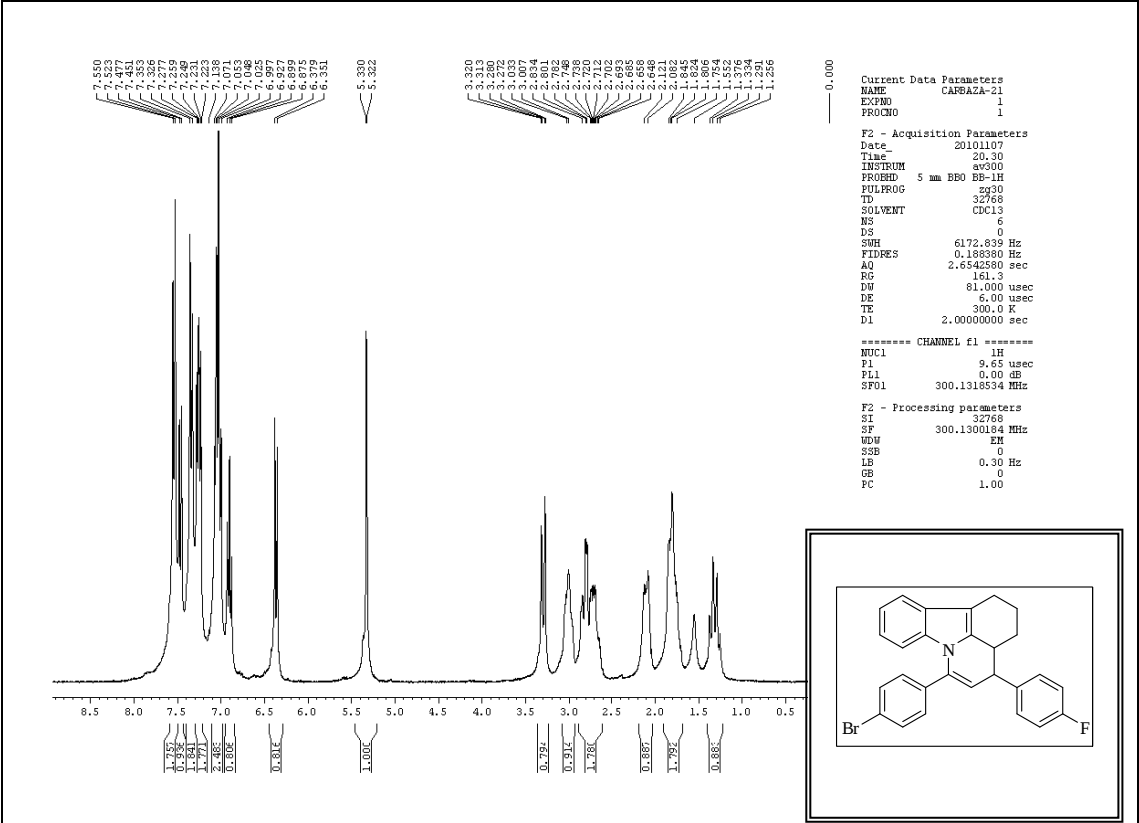


Figure 48. ¹H NMR Spectrum of 3m (CDCl₃)

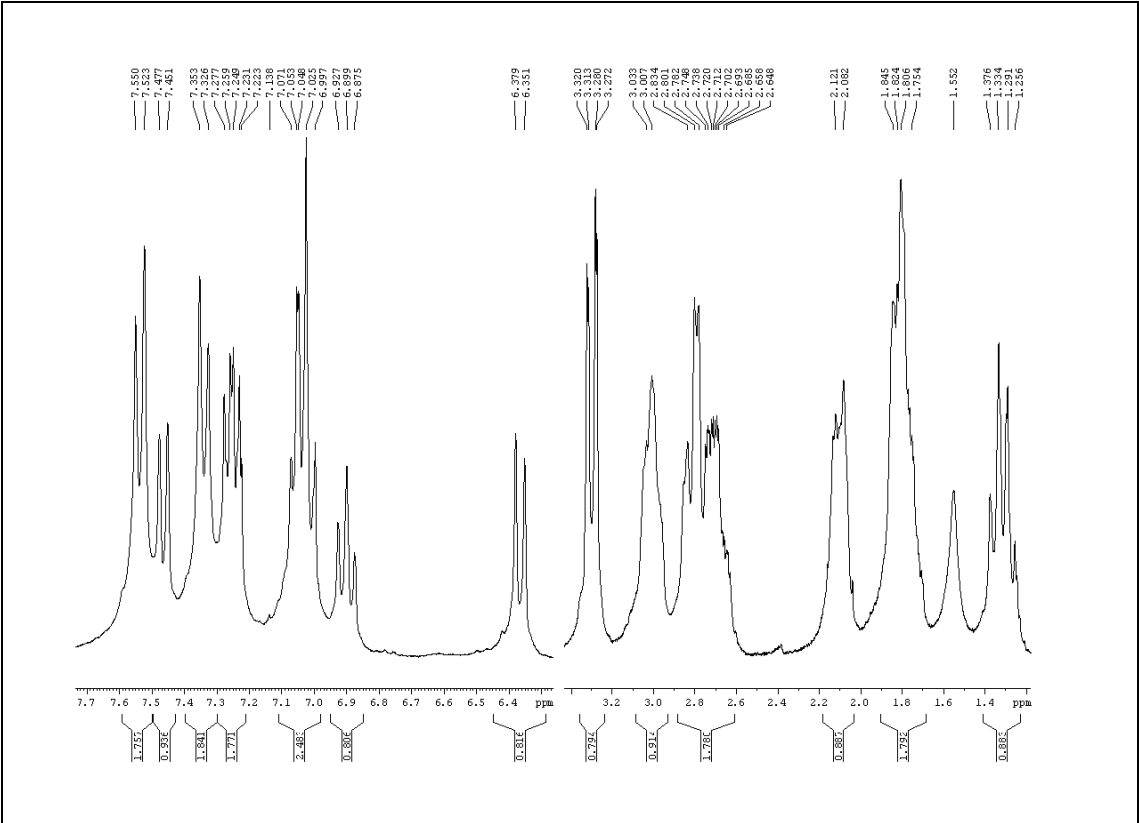


Figure 49. ¹H NMR Spectrum of 3m (expanded)

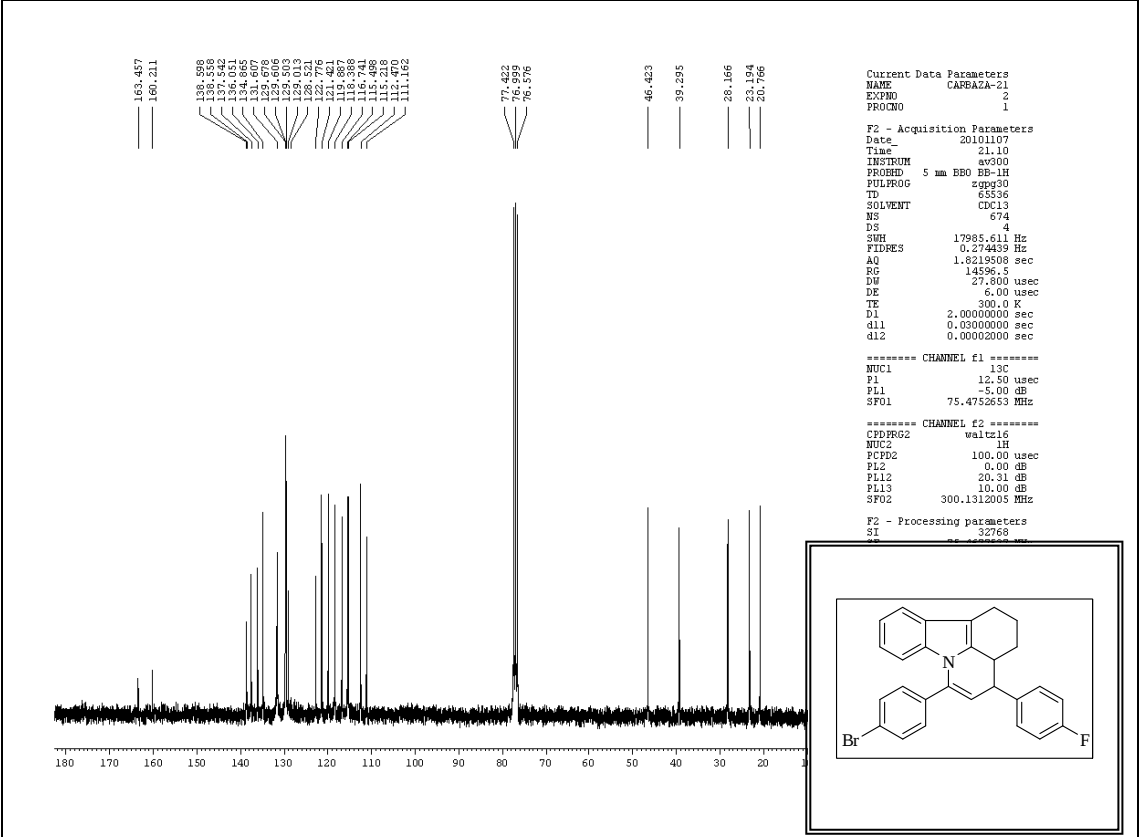
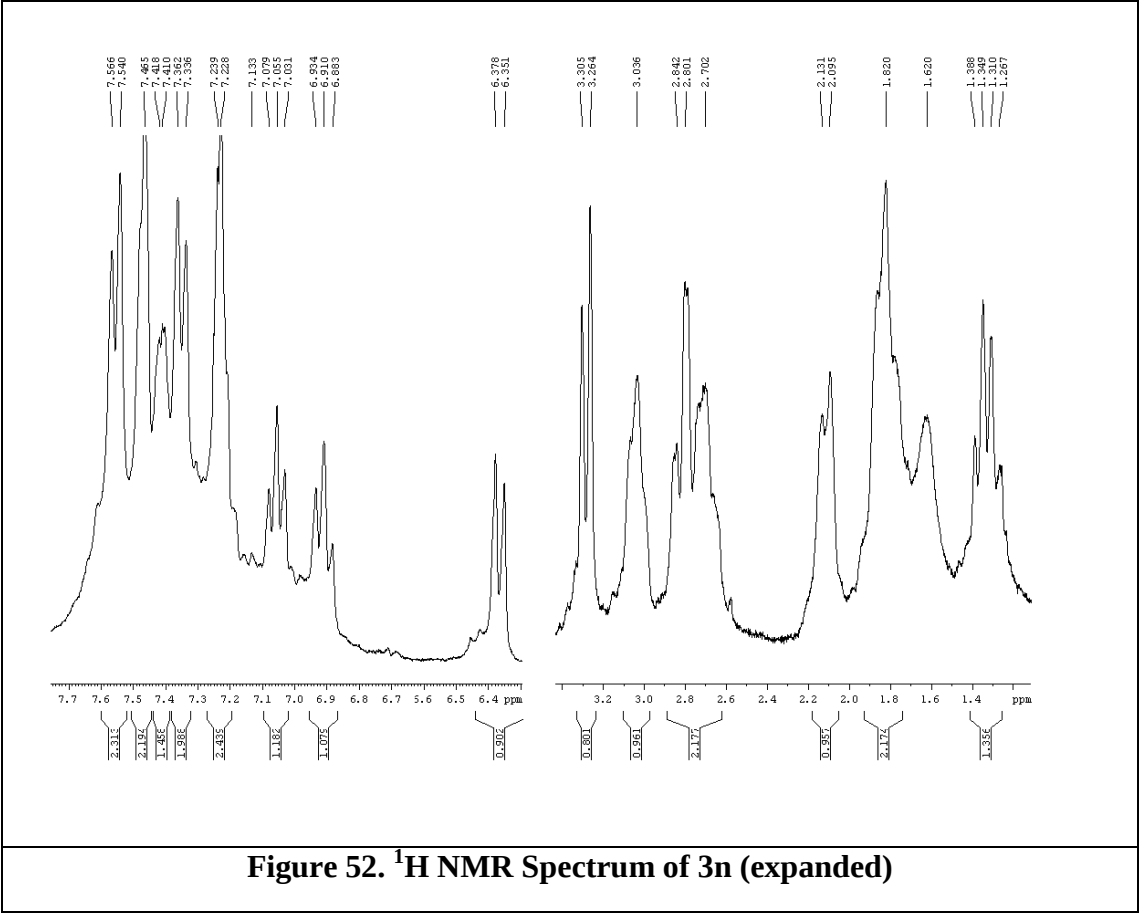
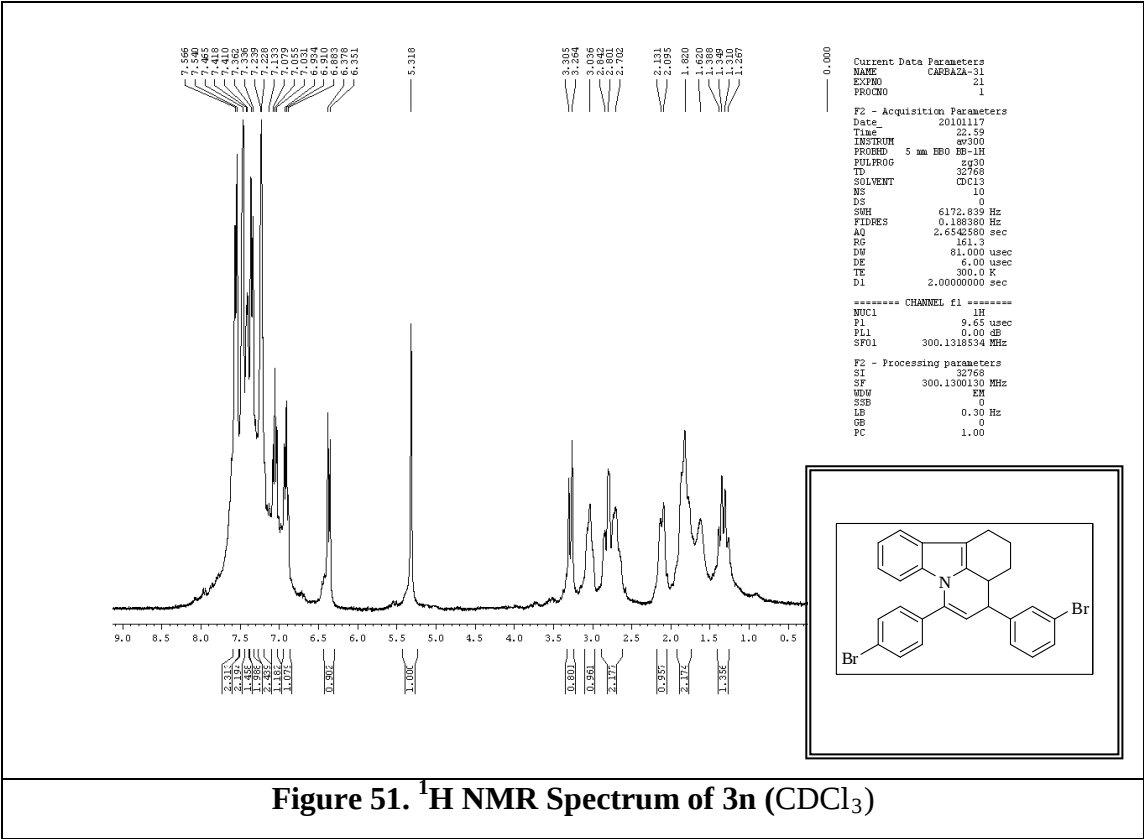


Figure 50. ¹³C NMR Spectrum of 3m (CDCl₃)



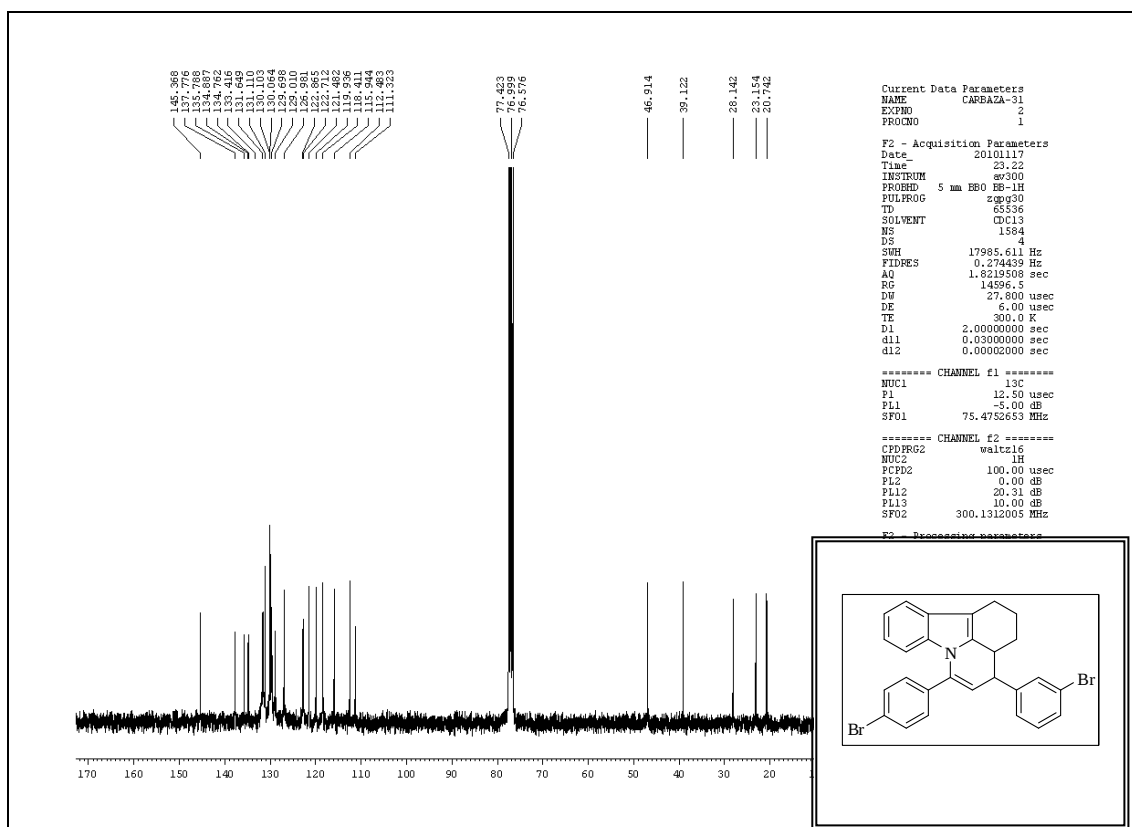


Figure 53. ^{13}C NMR Spectrum of 3n (CDCl_3)

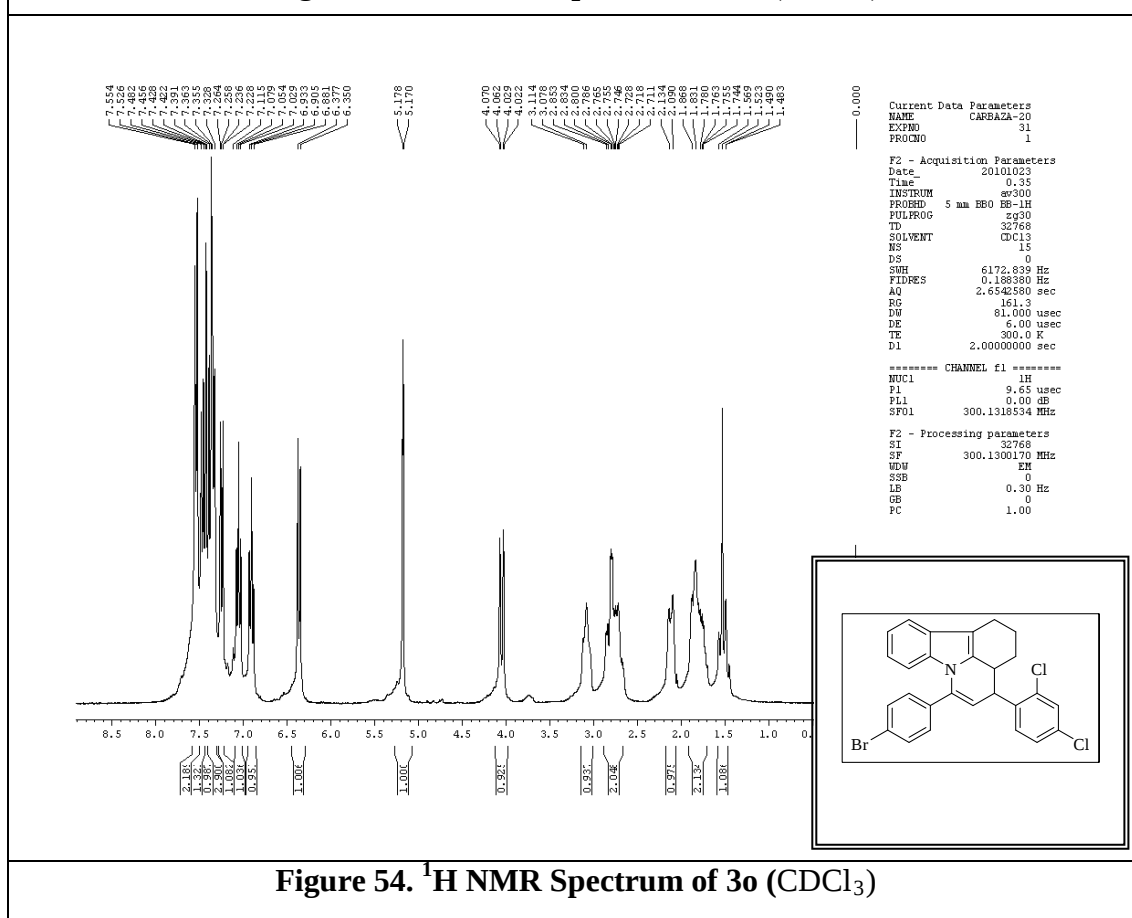


Figure 54. ^1H NMR Spectrum of 3o (CDCl_3)

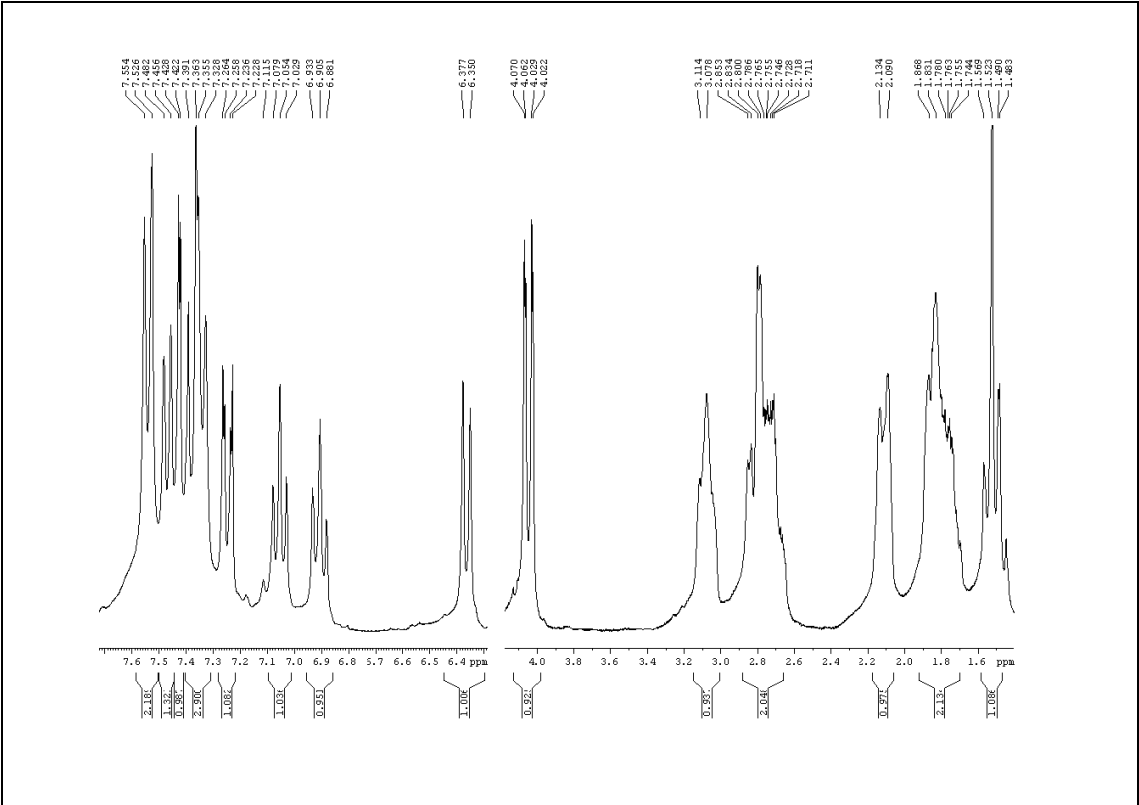


Figure 55. ¹H NMR Spectrum of 3o (expanded)

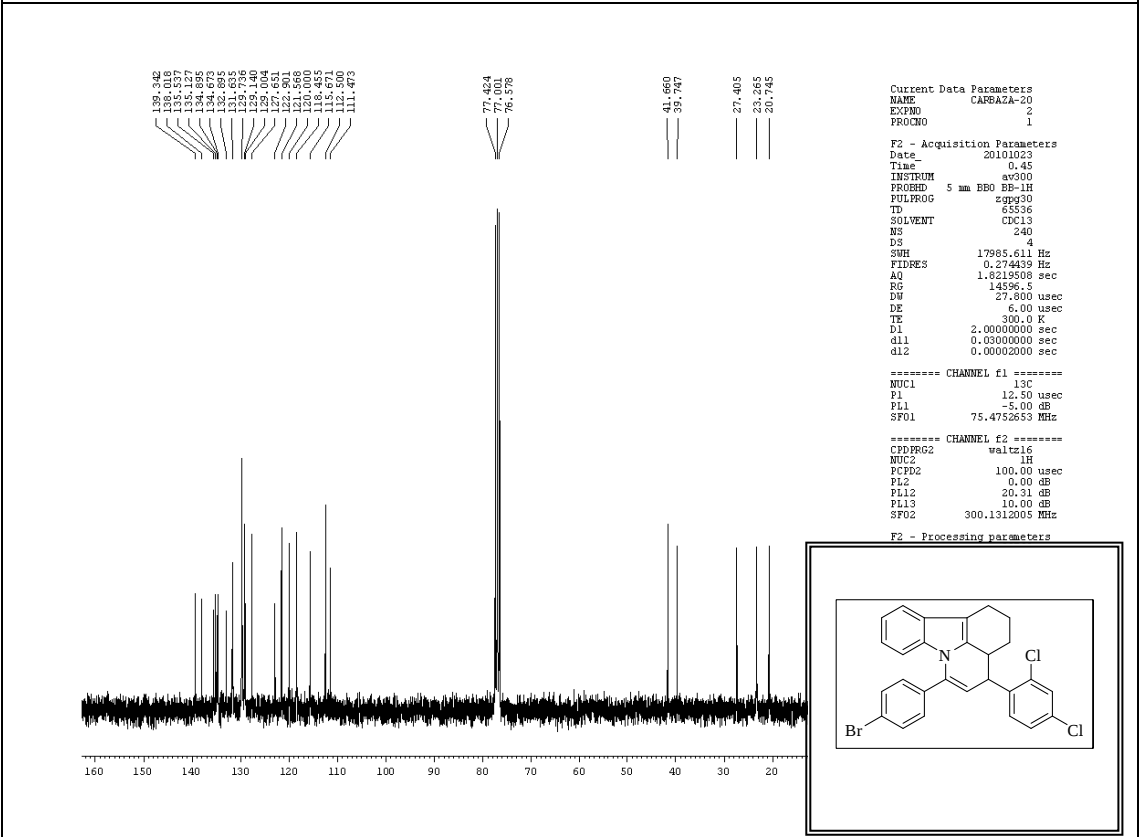


Figure 56. ¹³C NMR Spectrum of 3o (CDCl₃)

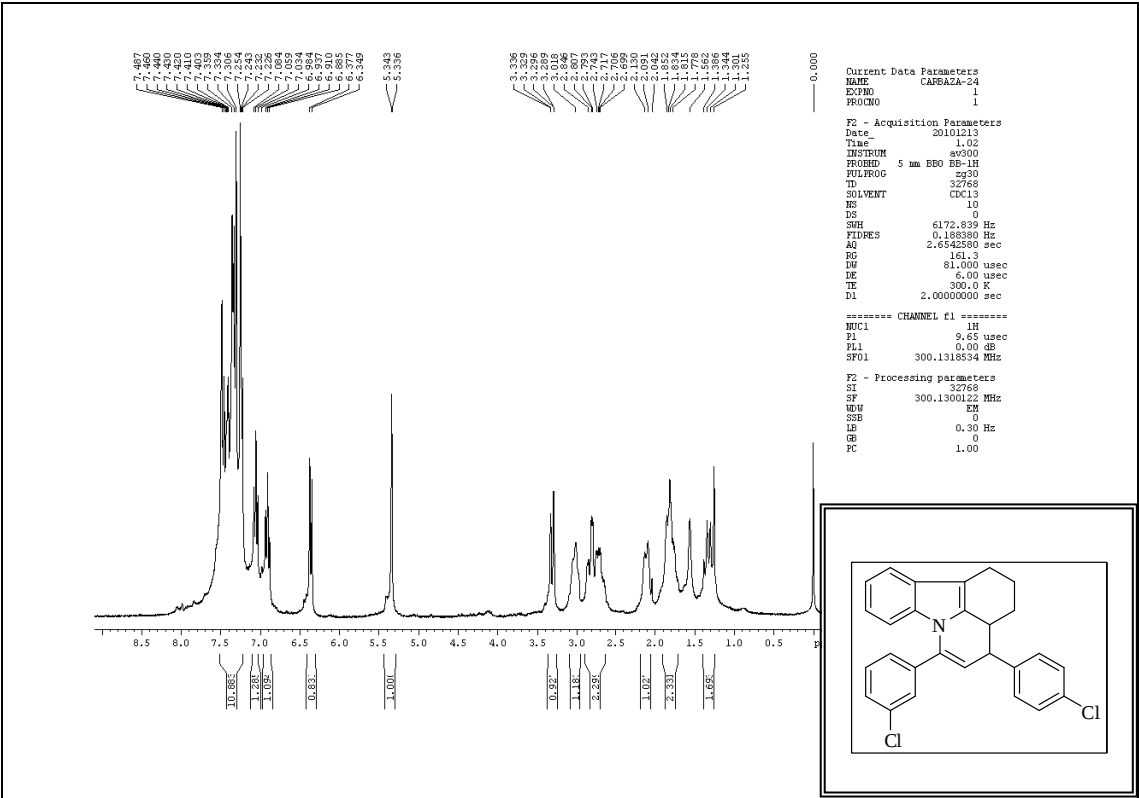


Figure 57. ¹H NMR Spectrum of 3p (CDCl₃)

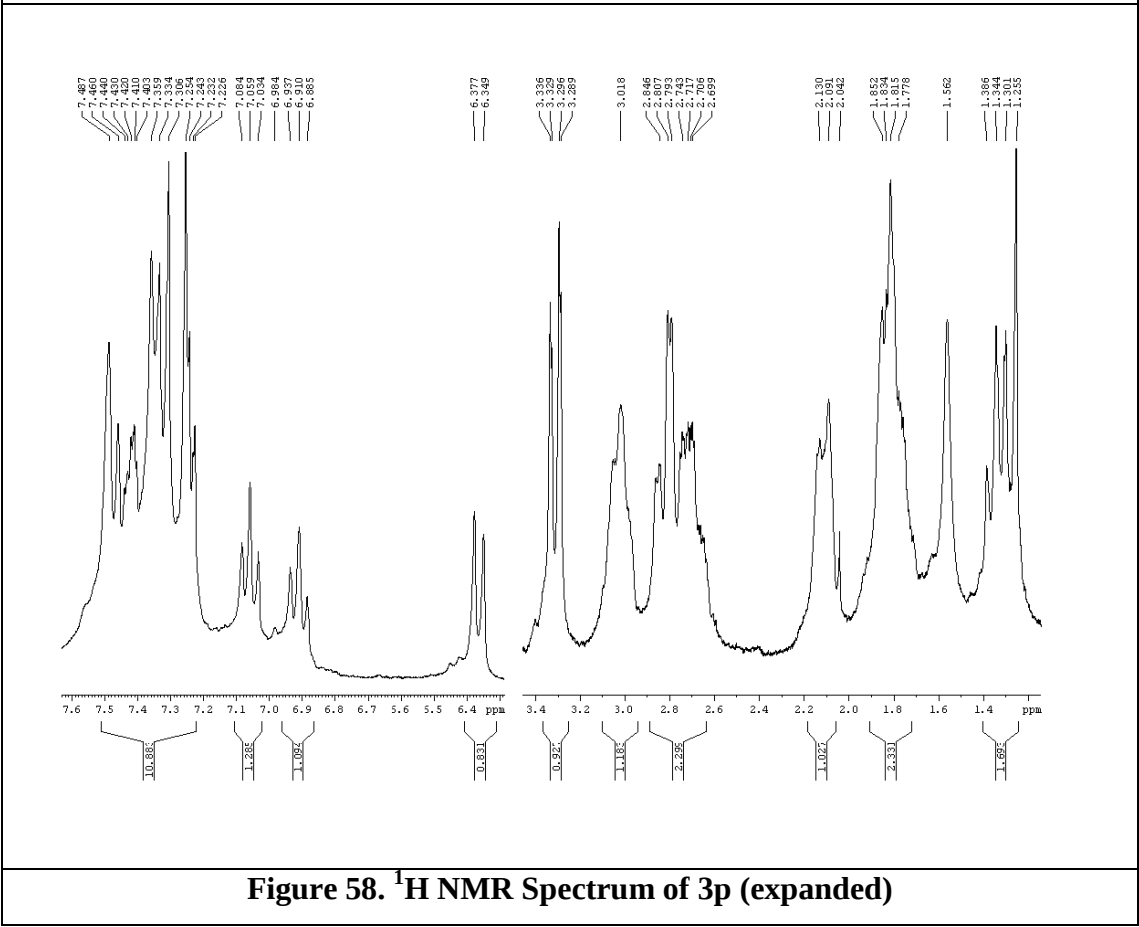


Figure 58. ¹H NMR Spectrum of 3p (expanded)

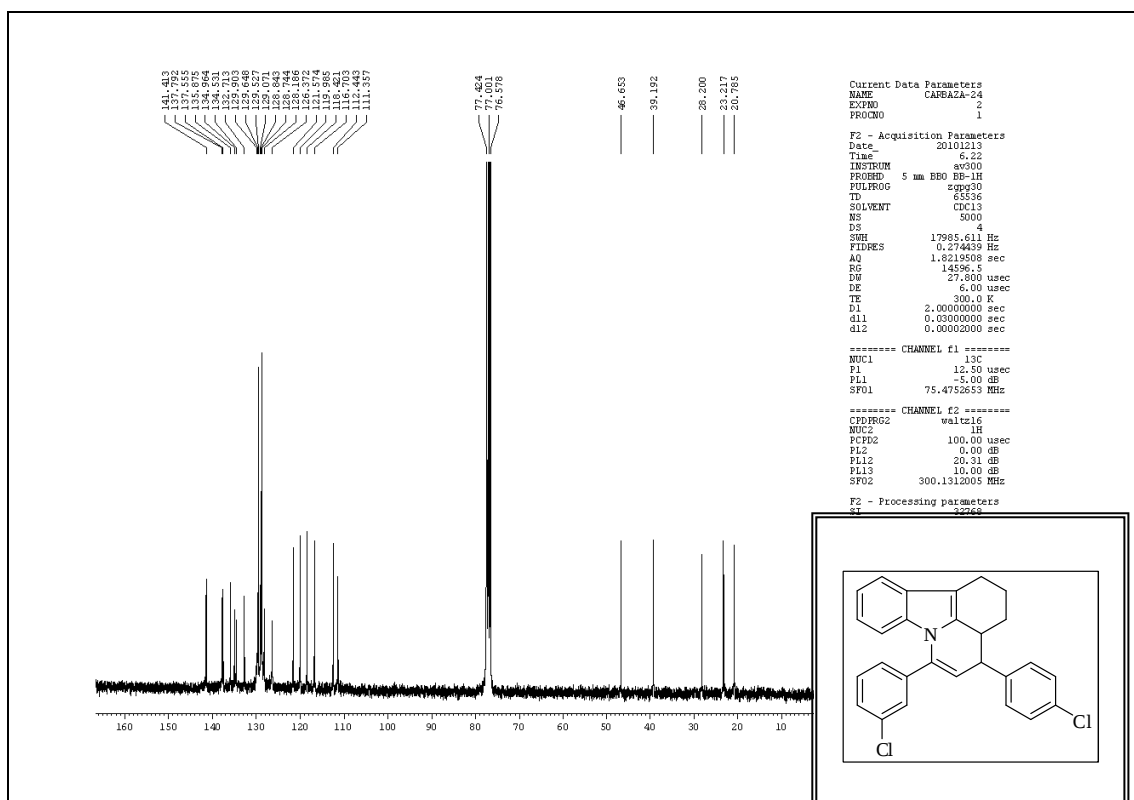


Figure 59. ^{13}C NMR Spectrum of 3p (CDCl_3)

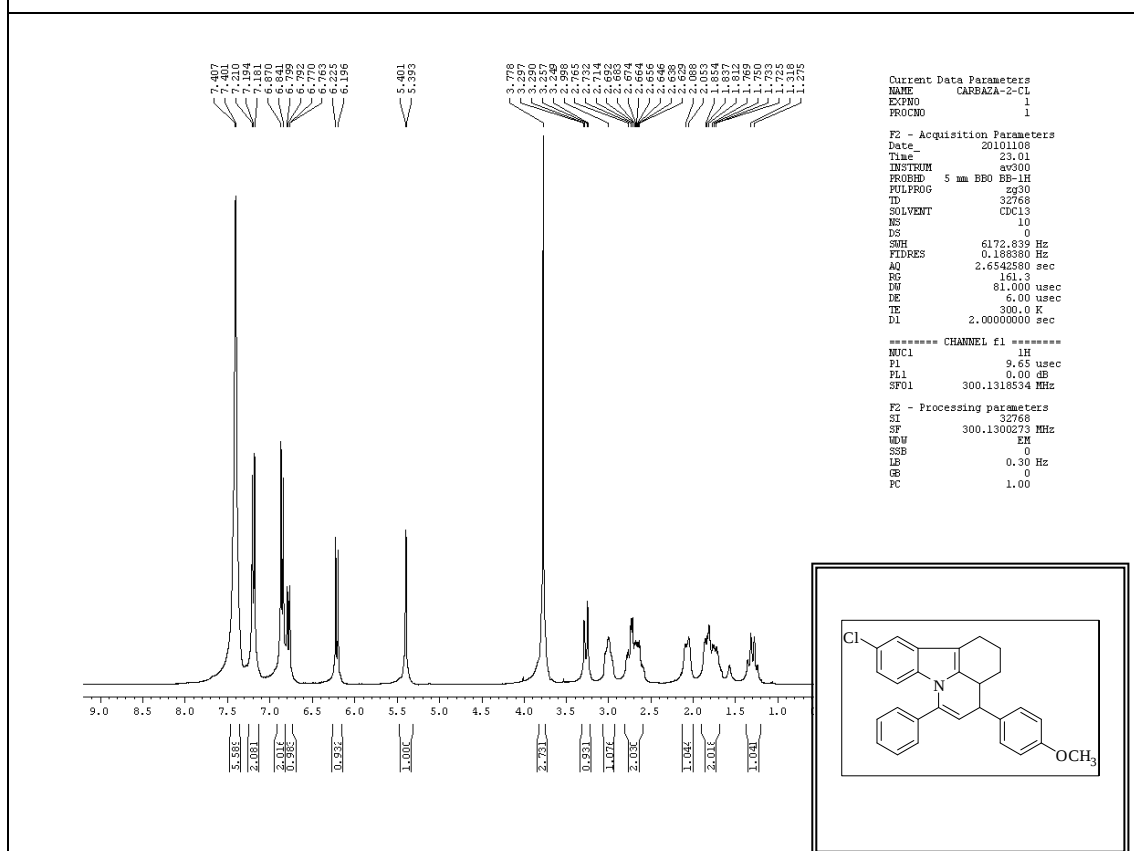


Figure 60. ^1H NMR Spectrum of 3q (CDCl_3)

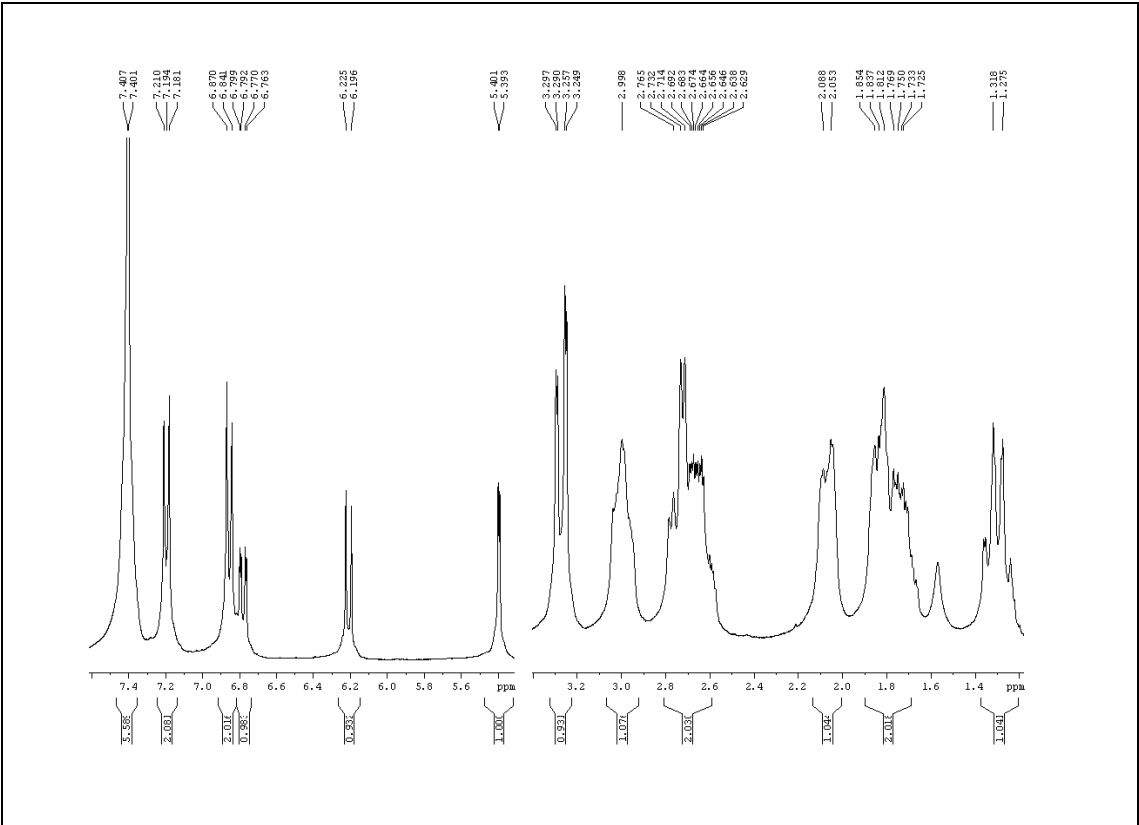


Figure 61. ¹H NMR Spectrum of 3q (expanded)

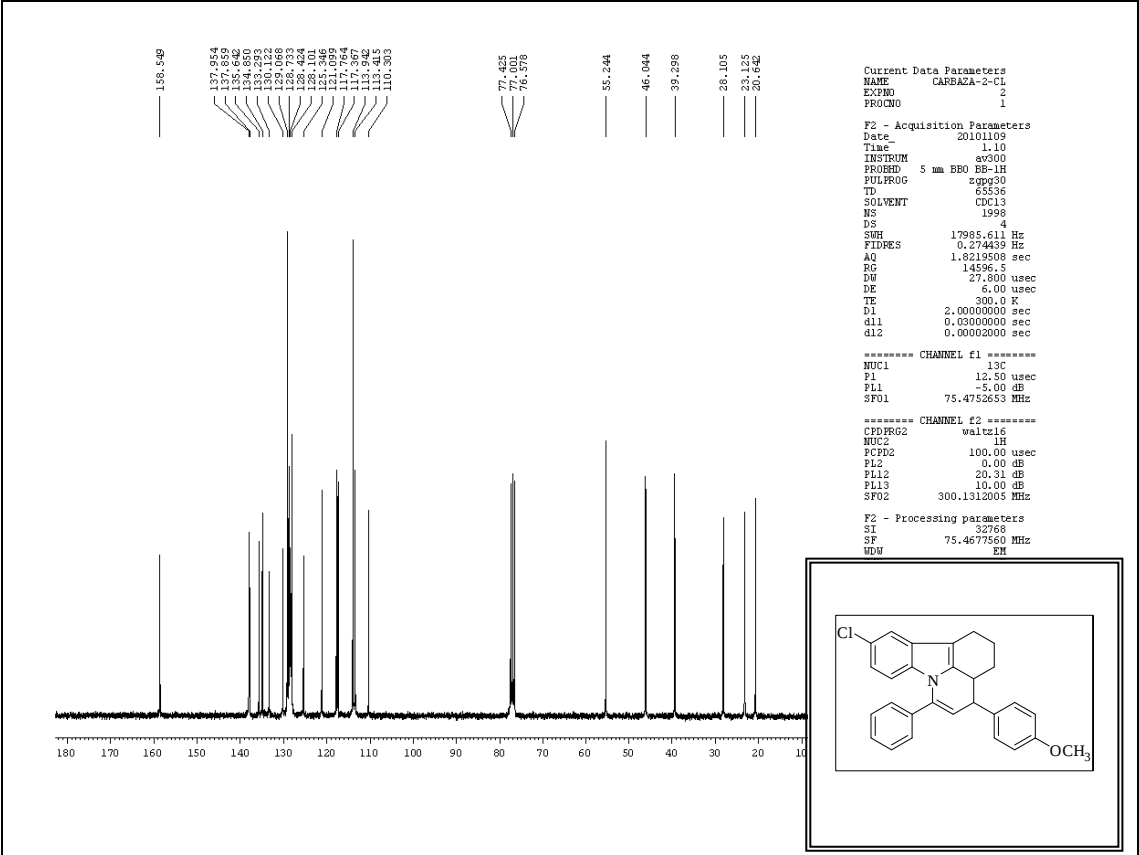
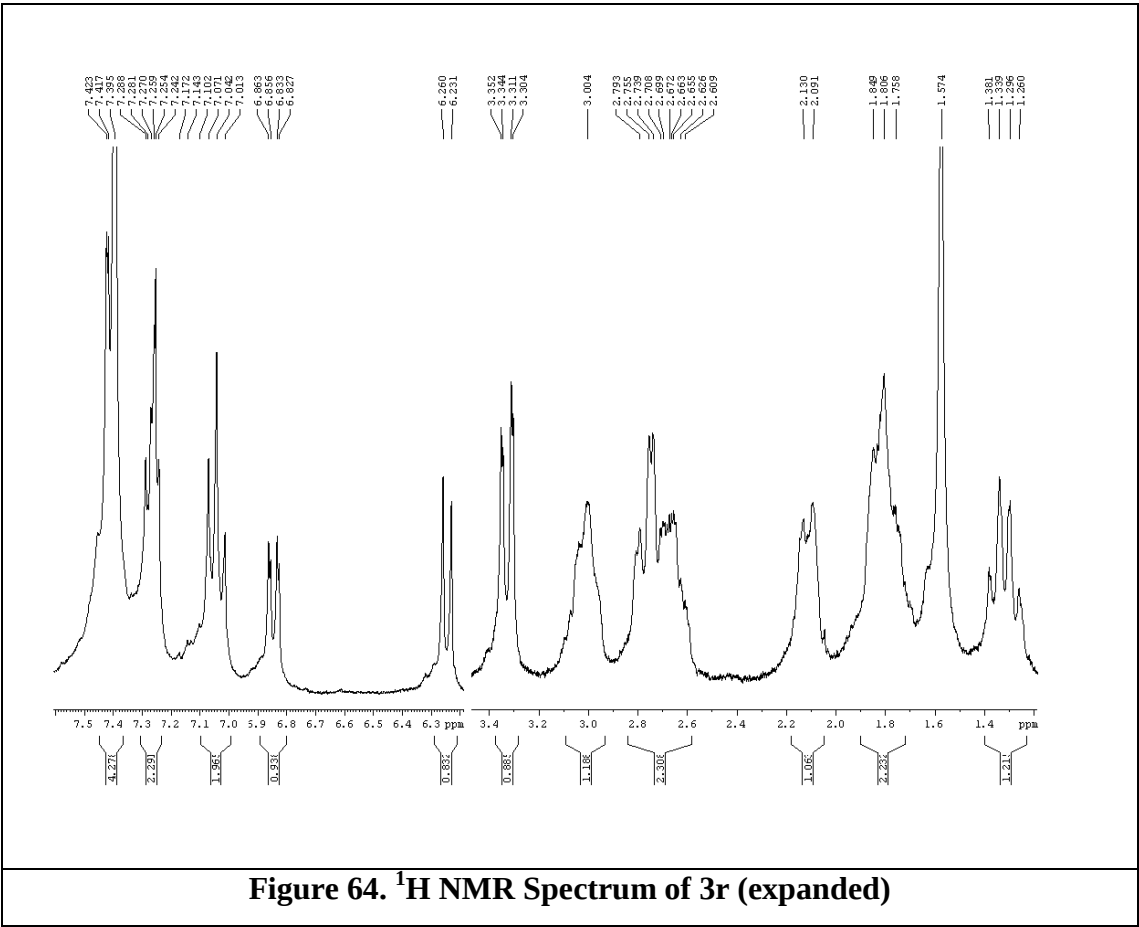
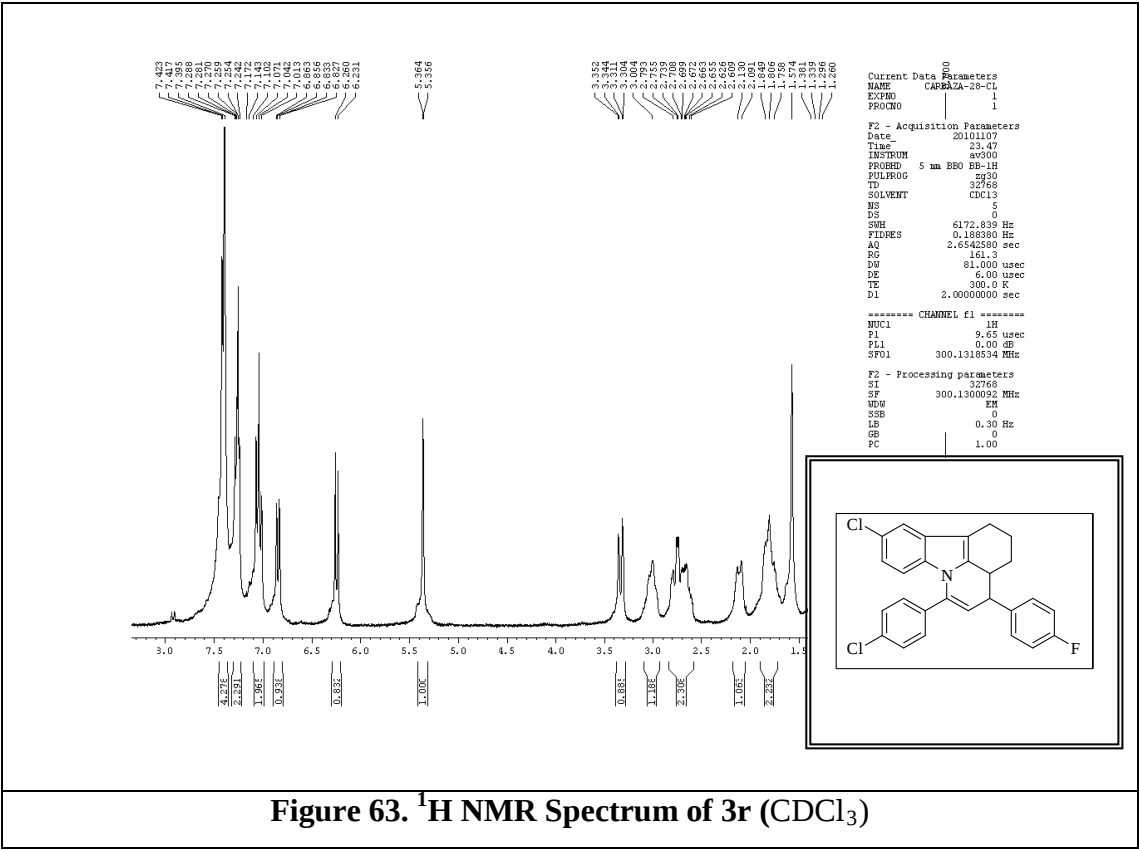
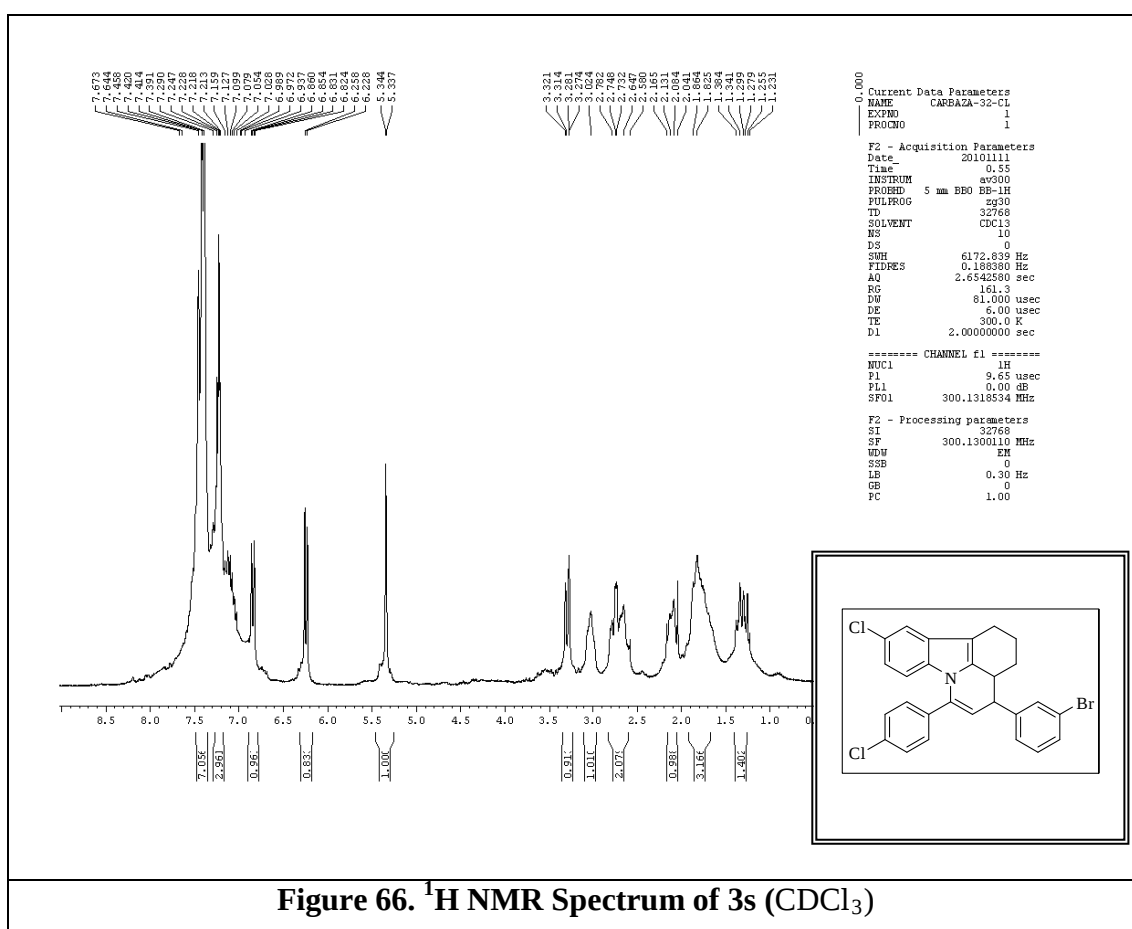
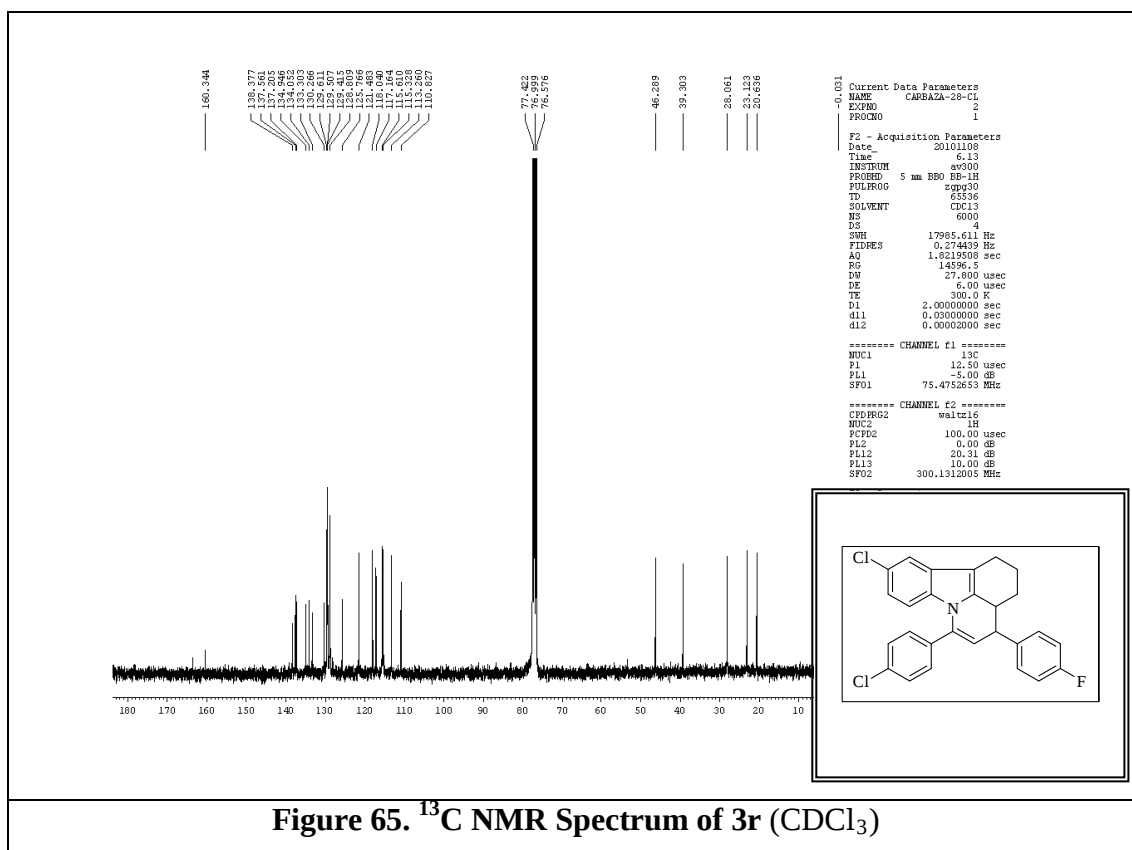


Figure 62. ¹³C NMR Spectrum of 3q (CDCl₃)





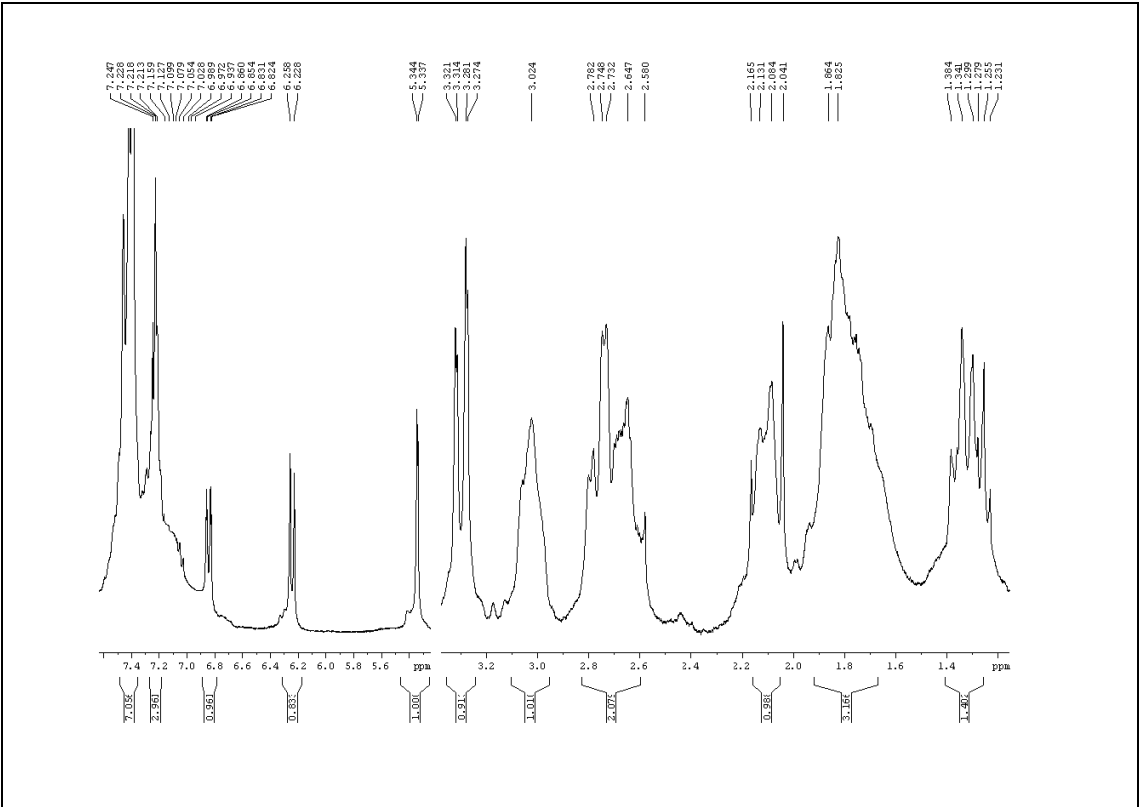


Figure 67. ¹H NMR Spectrum of 3s (expanded)

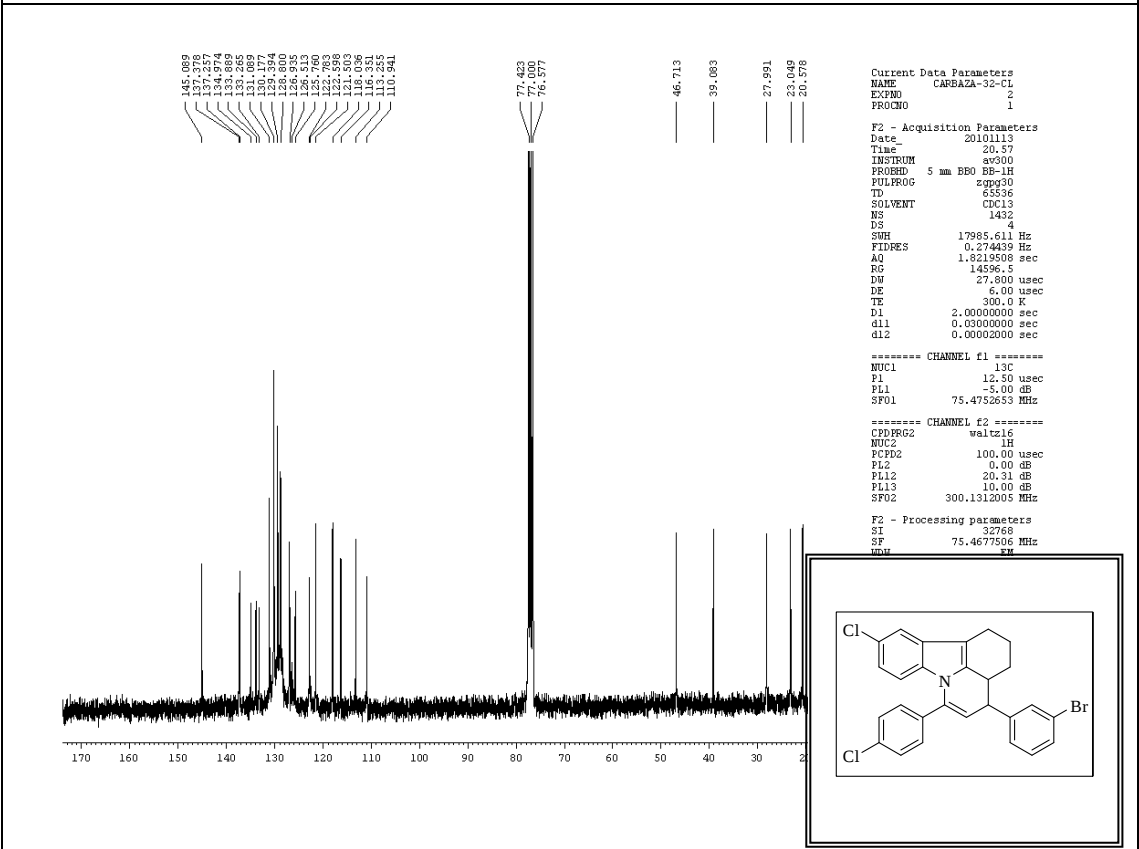
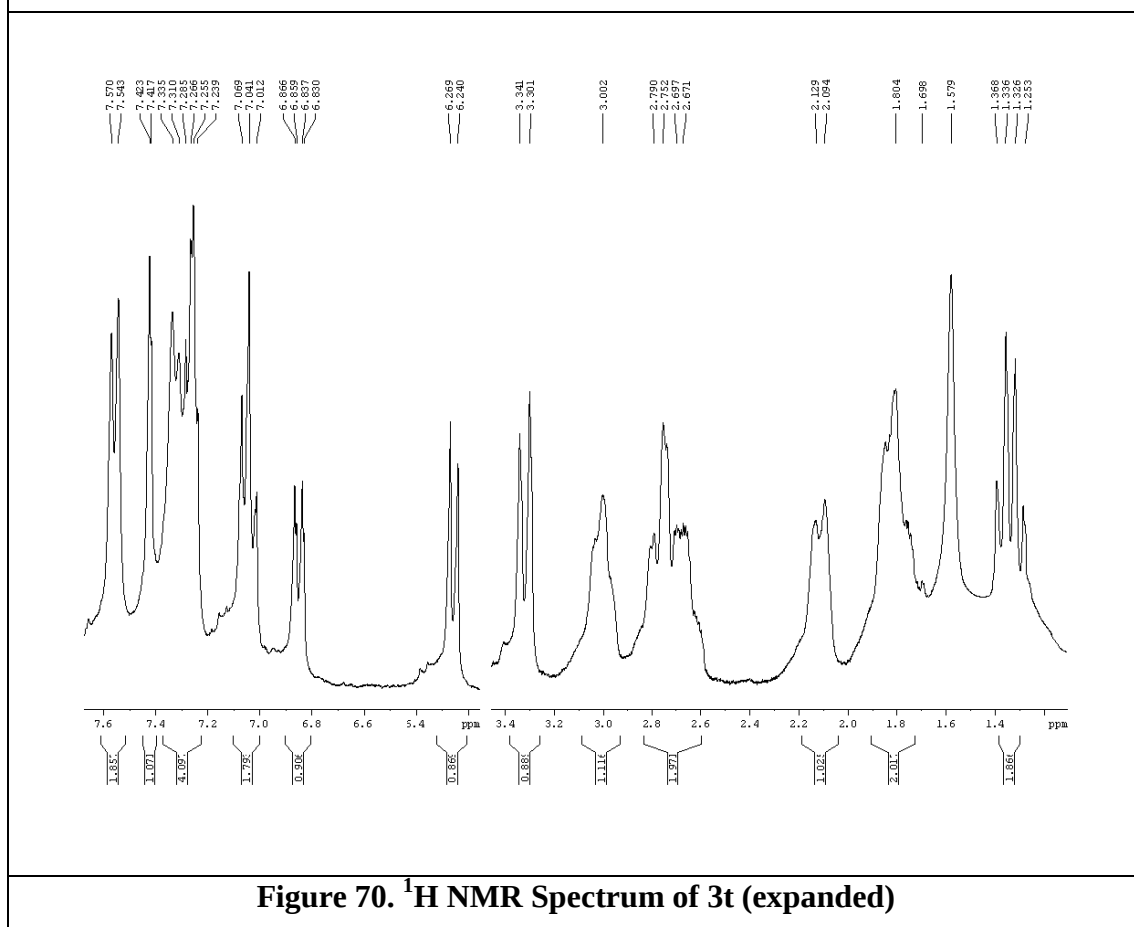
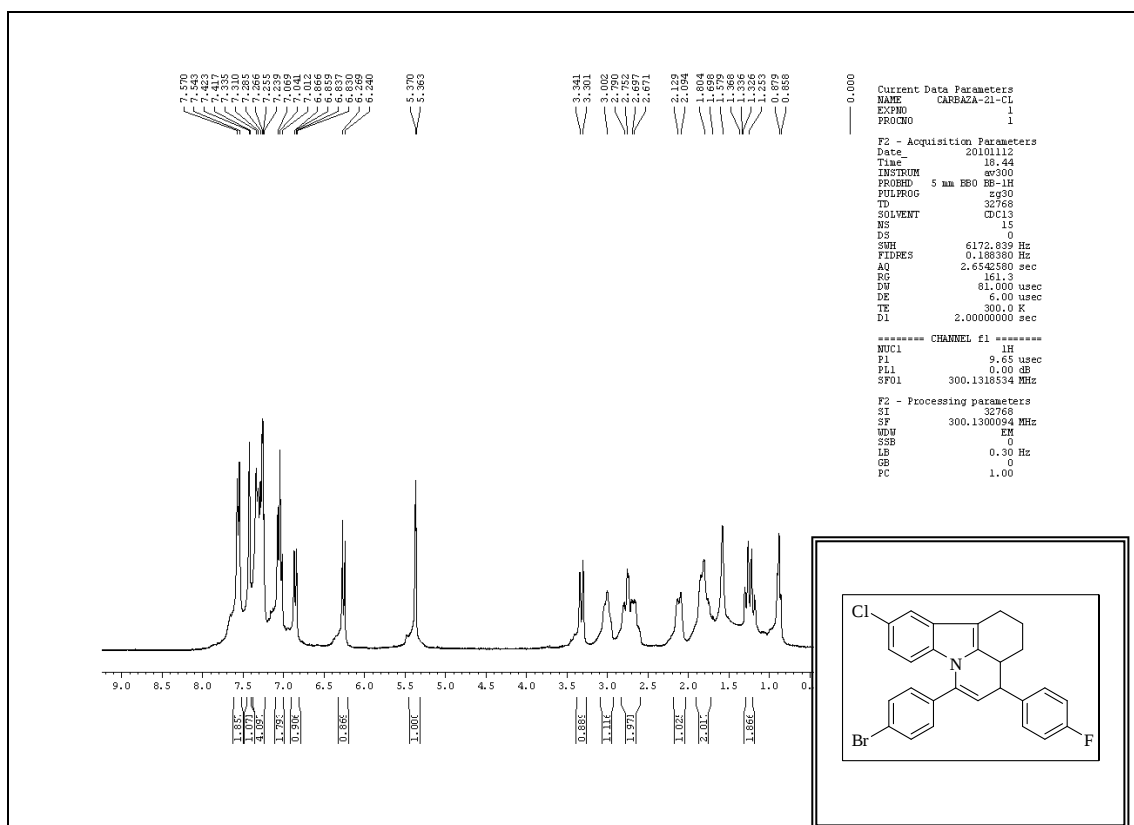


Figure 68. ¹³C NMR Spectrum of 3s (CDCl₃)



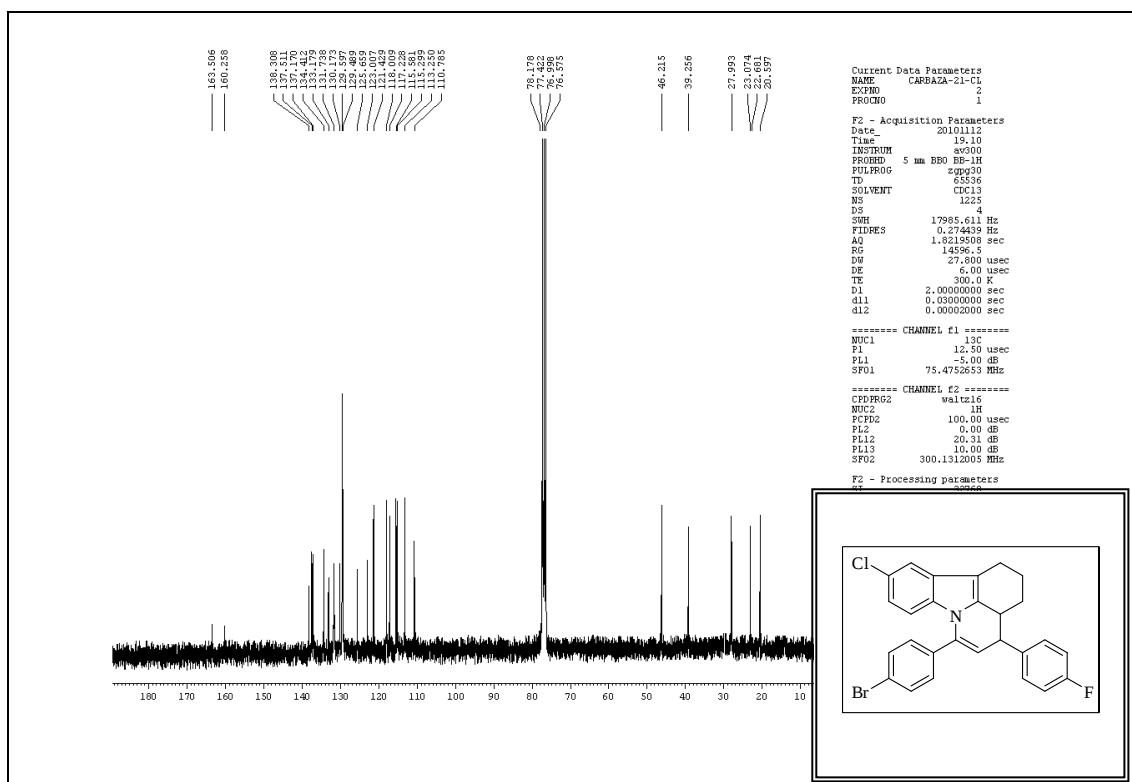


Figure 71. ^{13}C NMR Spectrum of 3t (CDCl_3)

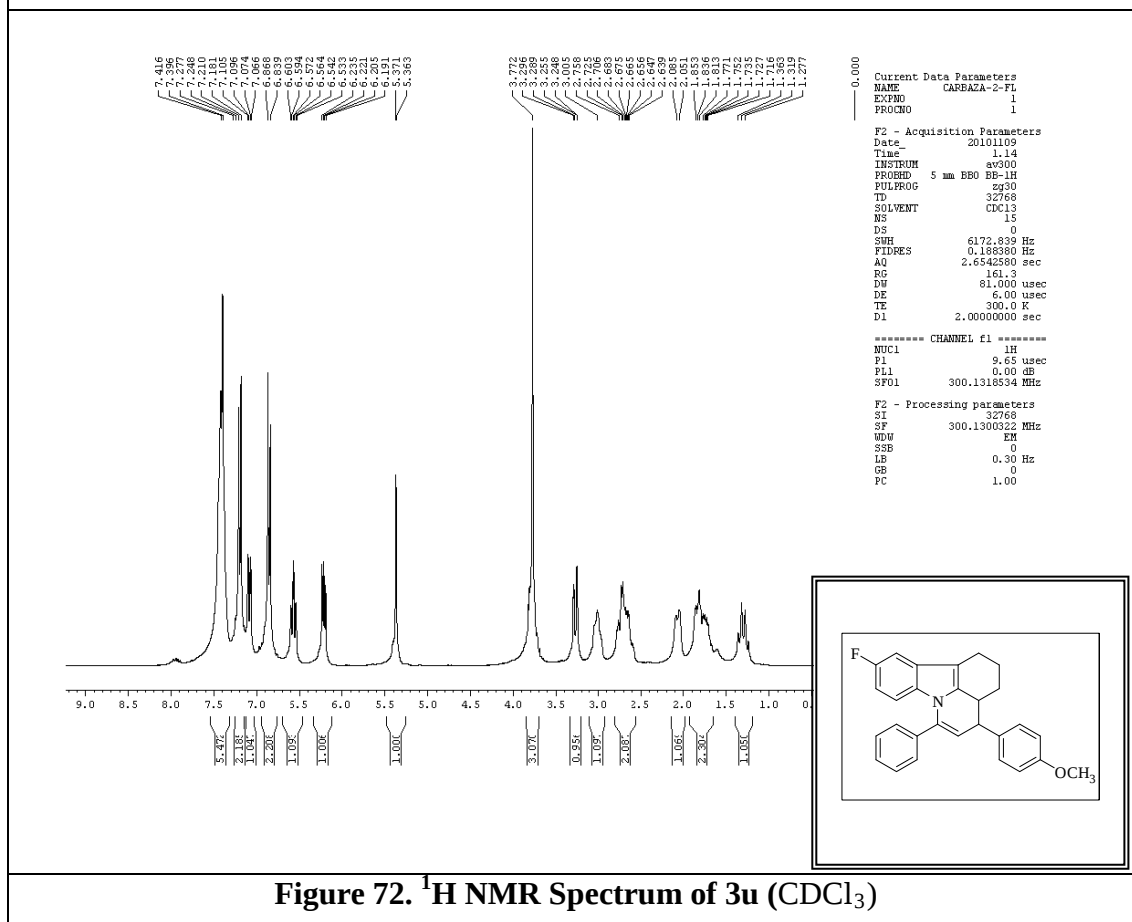


Figure 72. ^1H NMR Spectrum of 3u (CDCl_3)

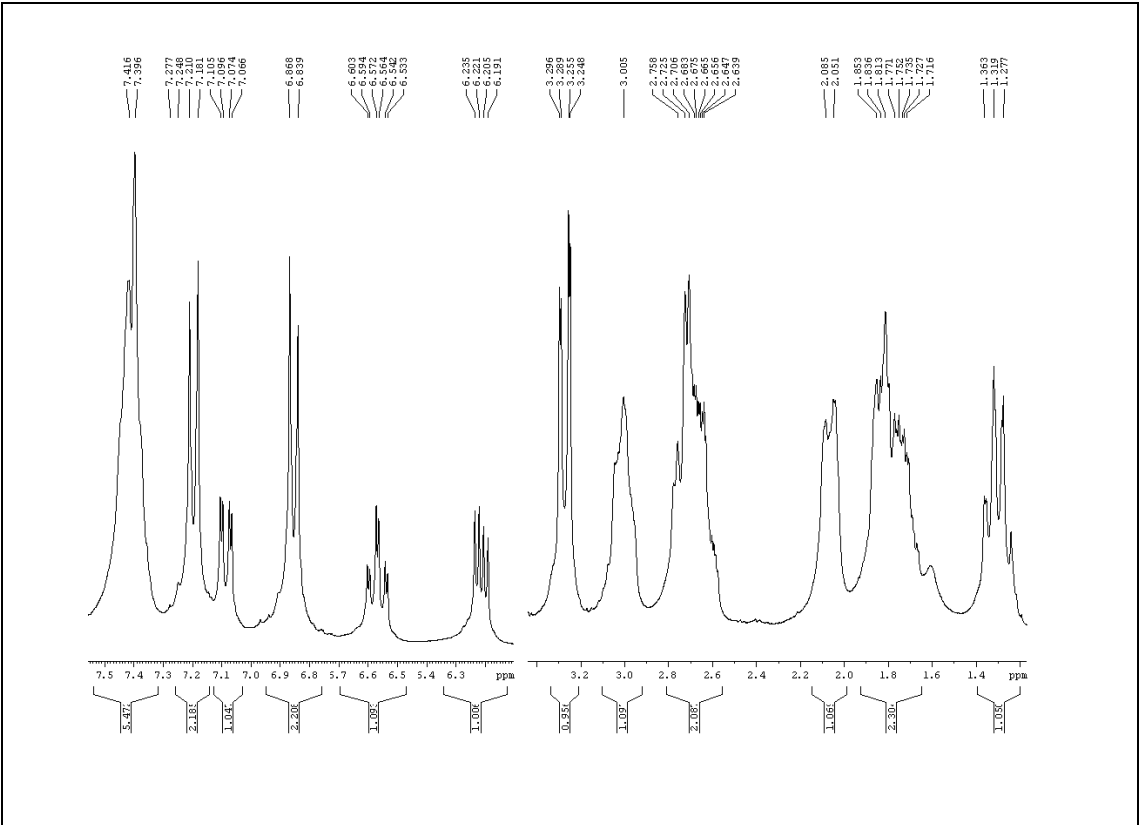


Figure 73. ¹H NMR Spectrum of 3u (expanded)

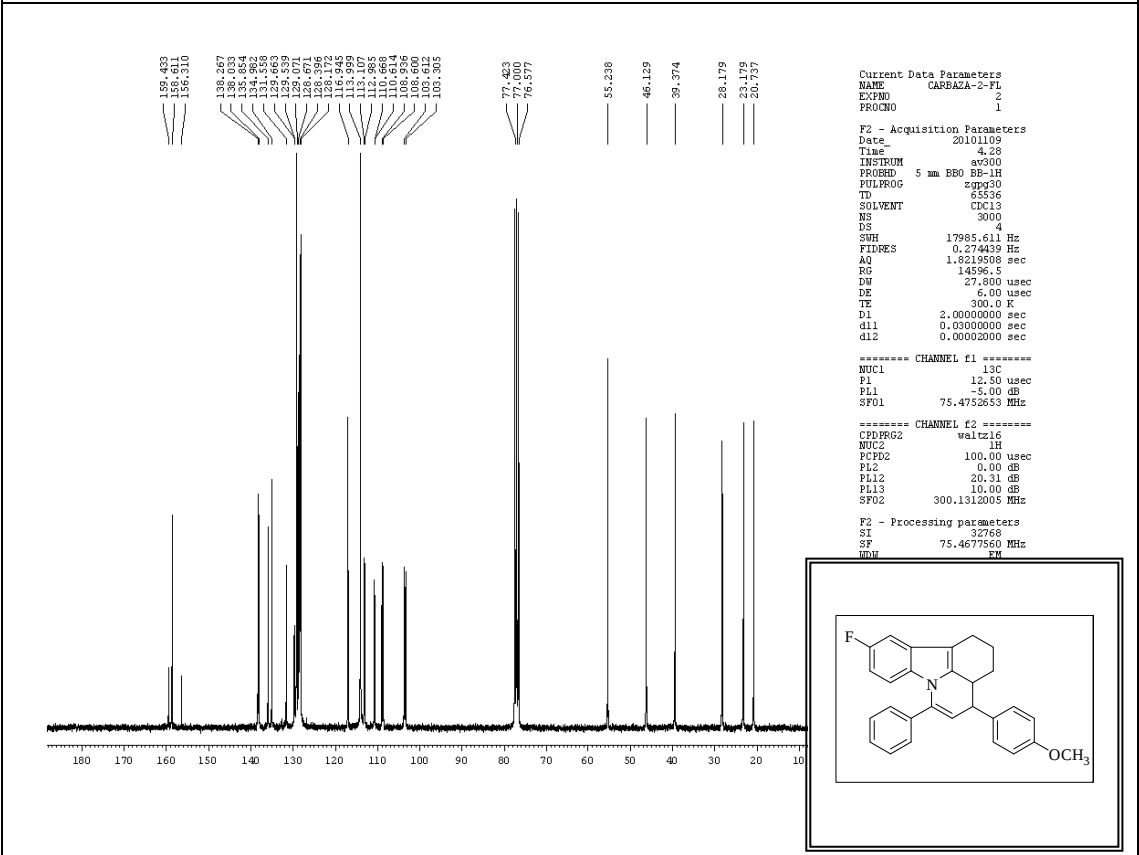


Figure 74. ¹³C NMR Spectrum of 3u (CDCl₃)

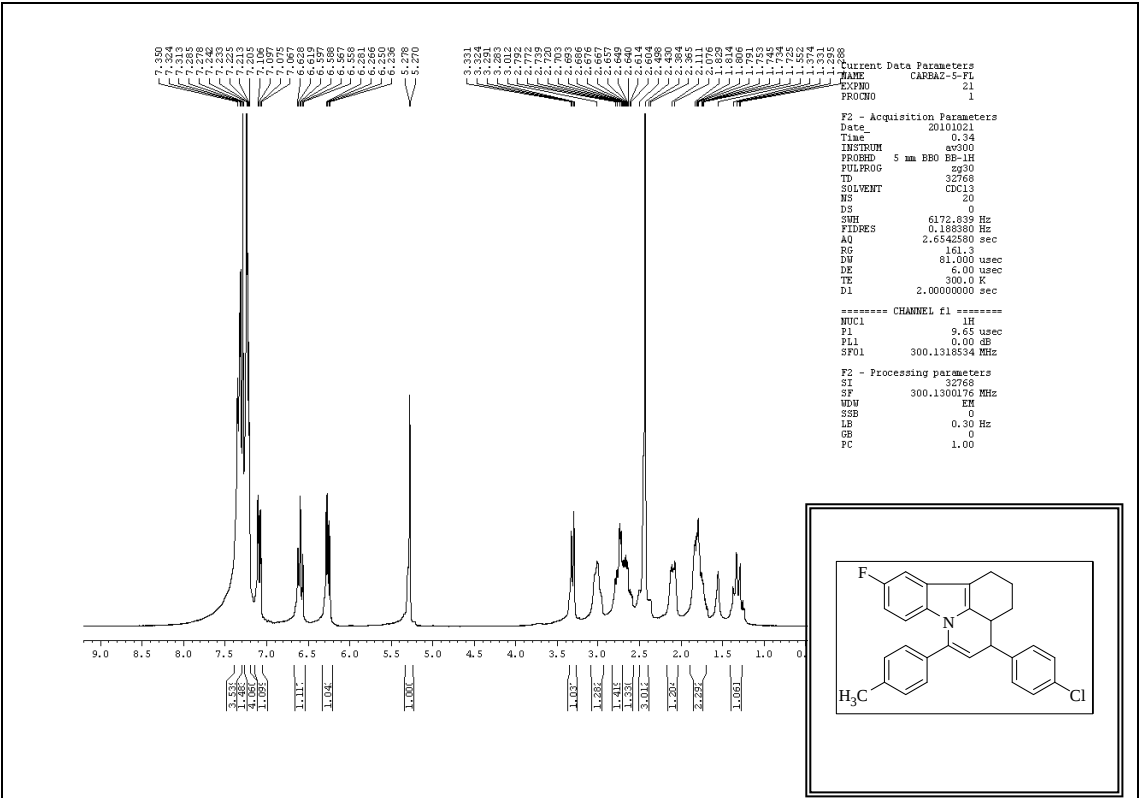


Figure 75. ¹H NMR Spectrum of 3v (CDCl₃)

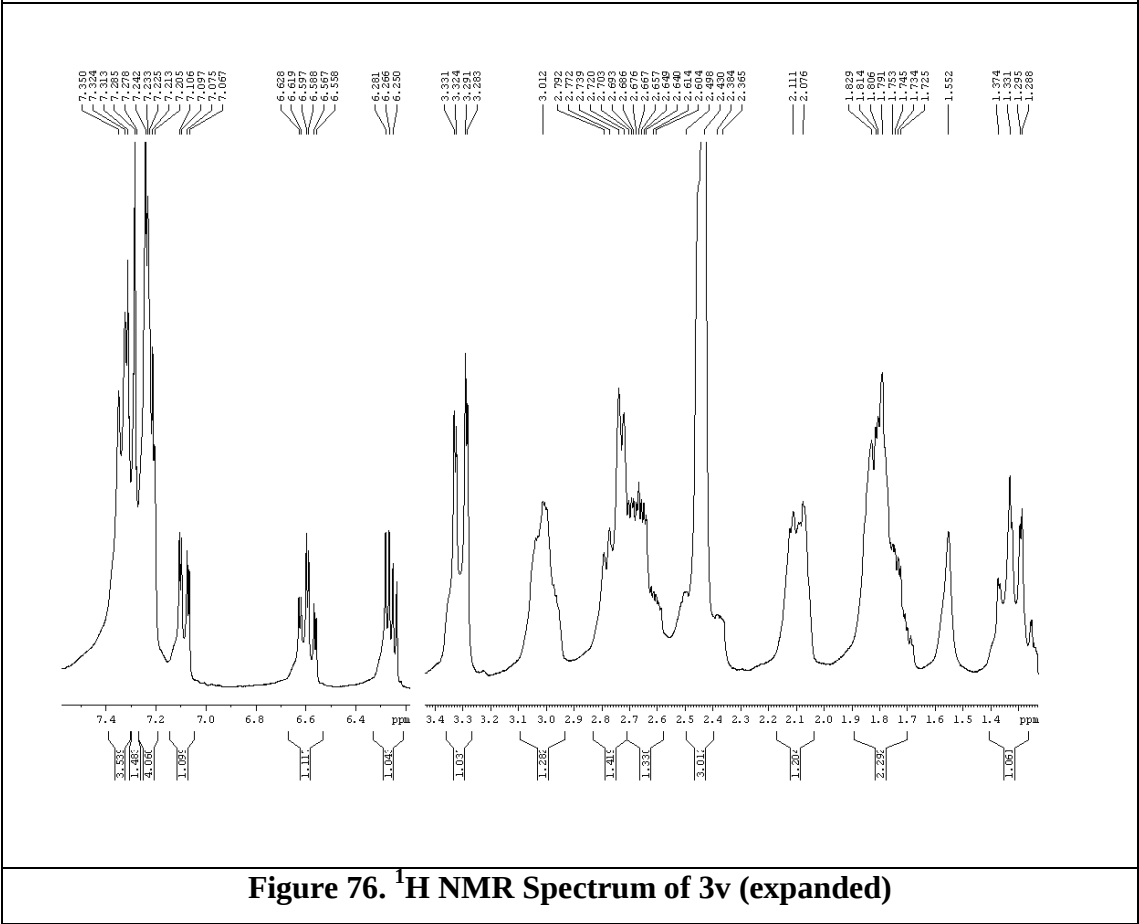


Figure 76. ¹H NMR Spectrum of 3v (expanded)

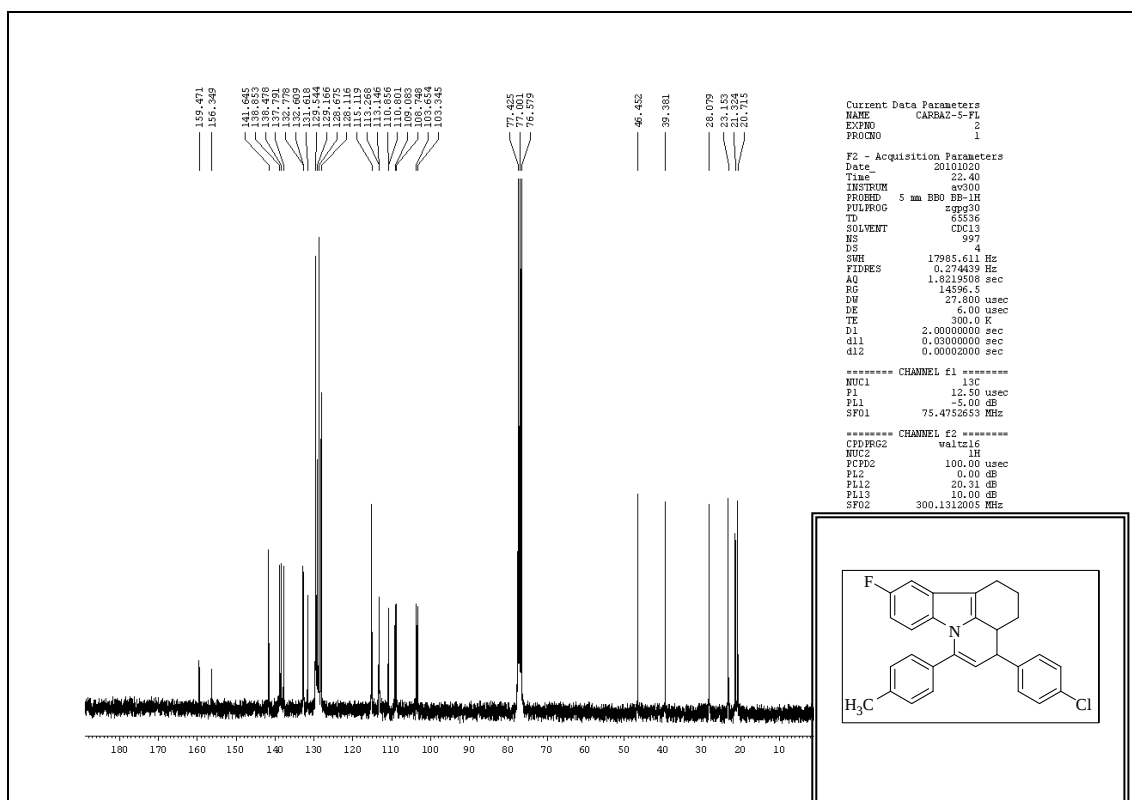


Figure 77. ^{13}C NMR Spectrum of 3v (CDCl_3)

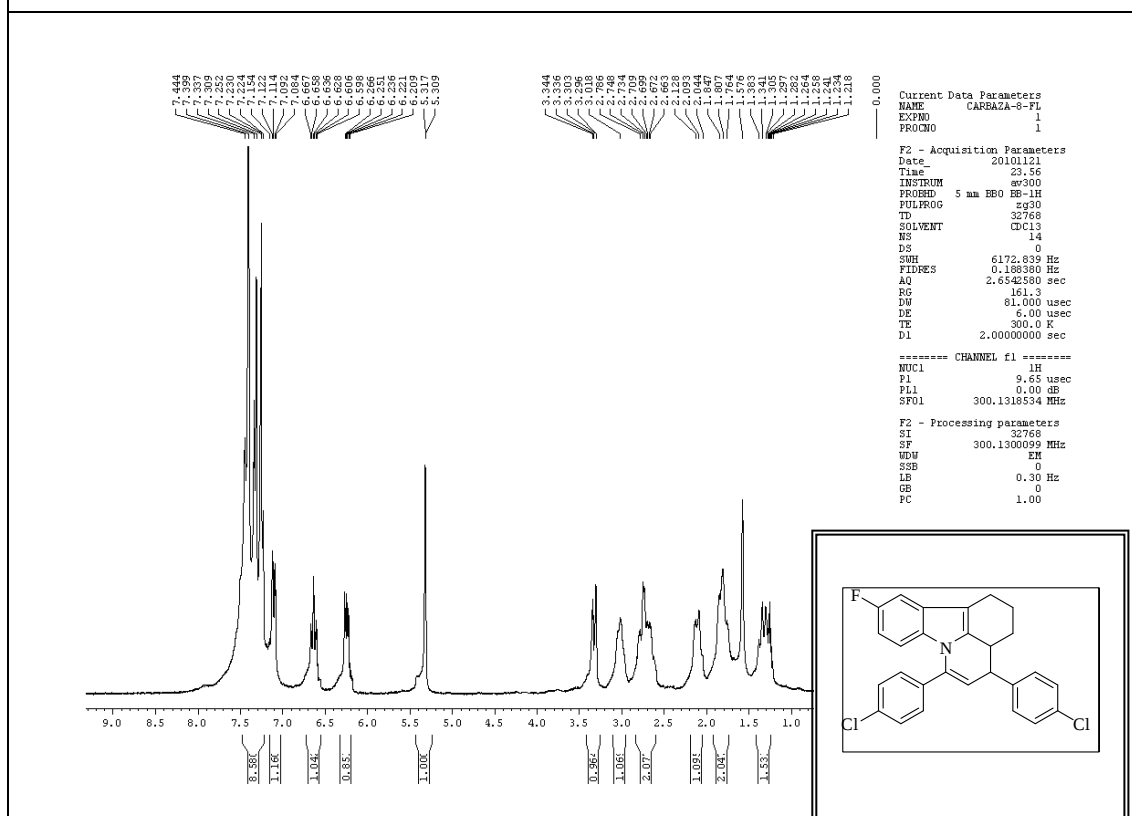


Figure 78. ^1H NMR Spectrum of 3w (CDCl_3)

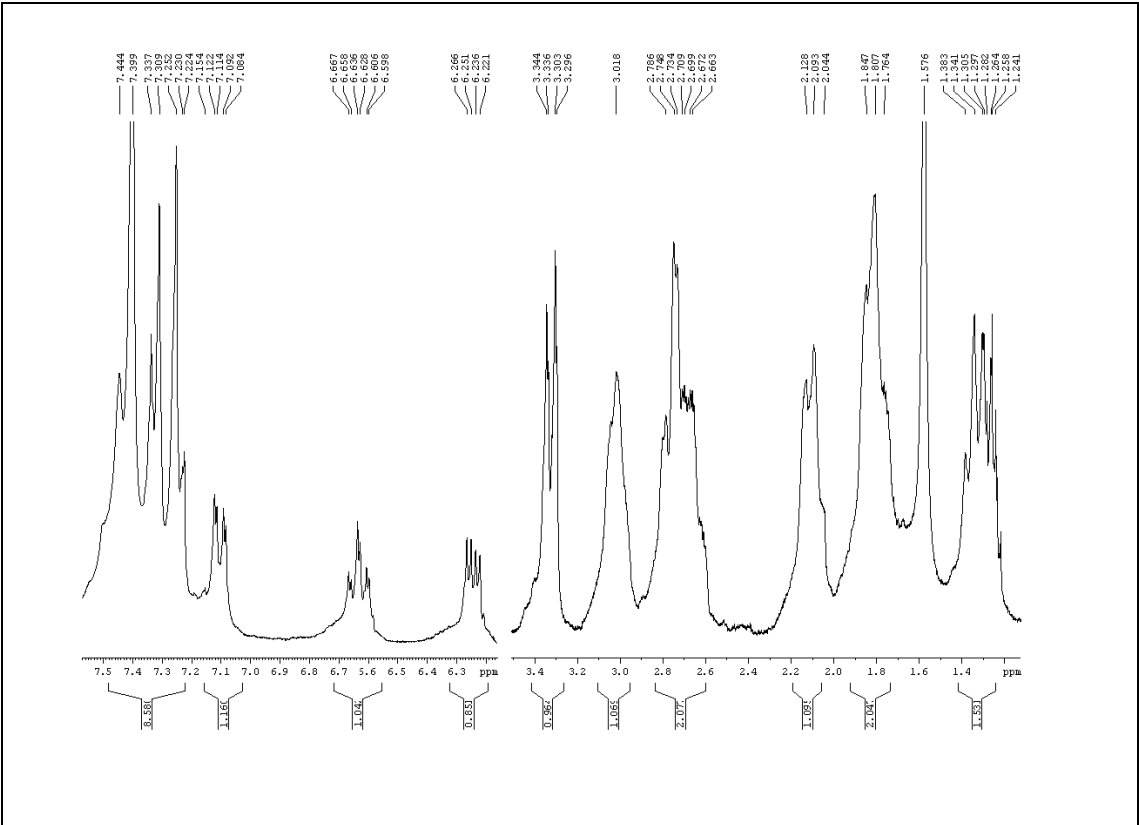


Figure 79. ¹H NMR Spectrum of 3w (expanded)

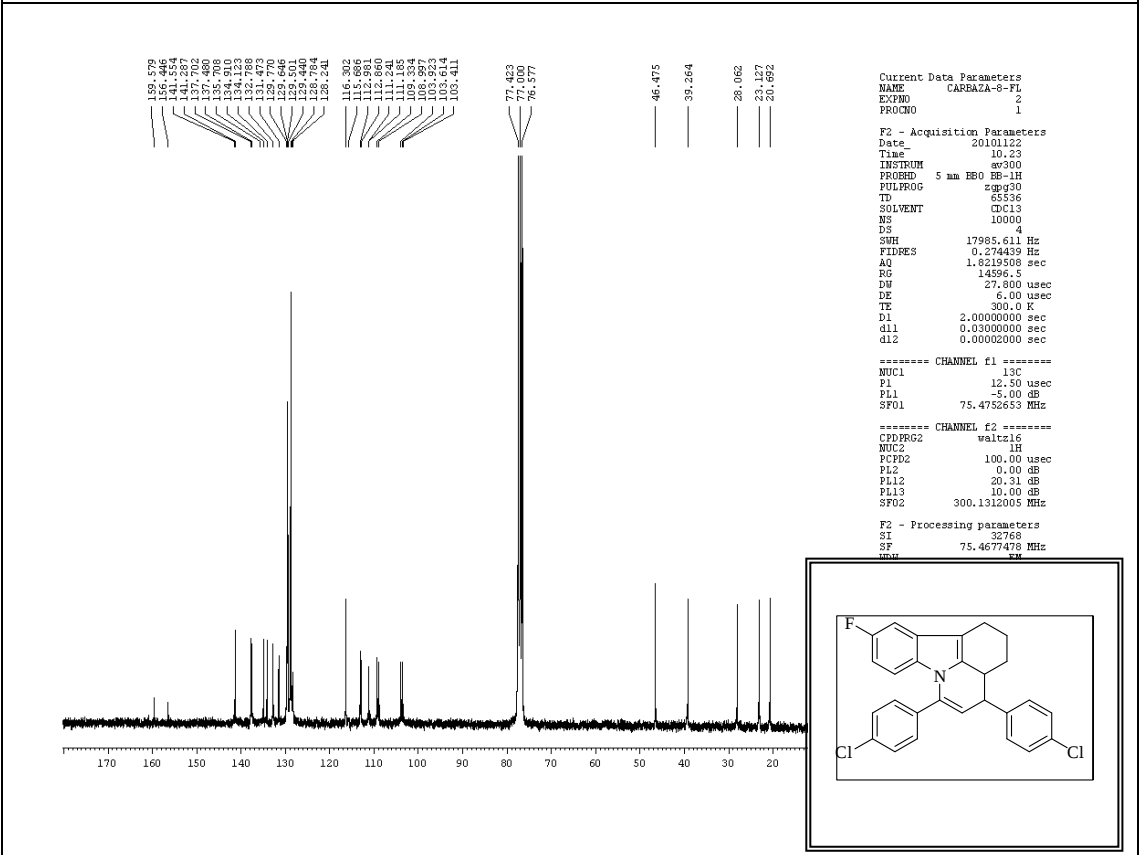


Figure 80. ¹³C NMR Spectrum of 3w (CDCl₃)

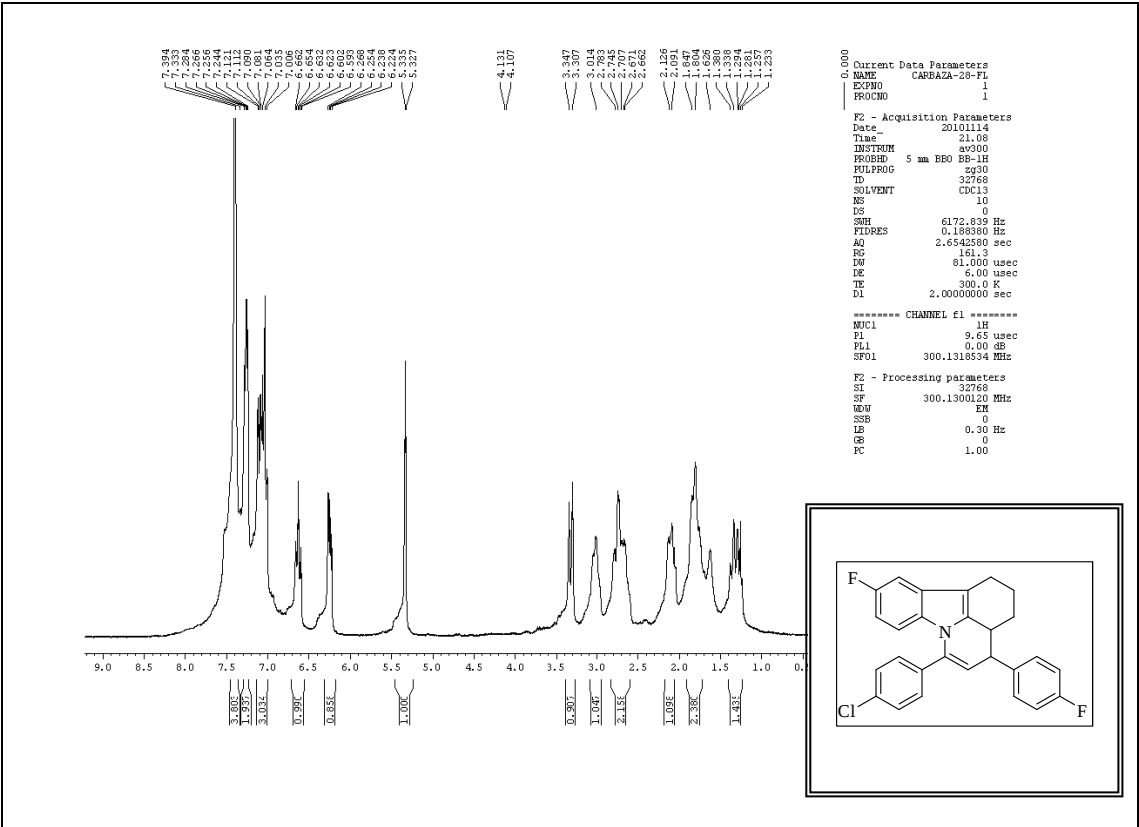


Figure 81. ¹H NMR Spectrum of 3x (CDCl₃)

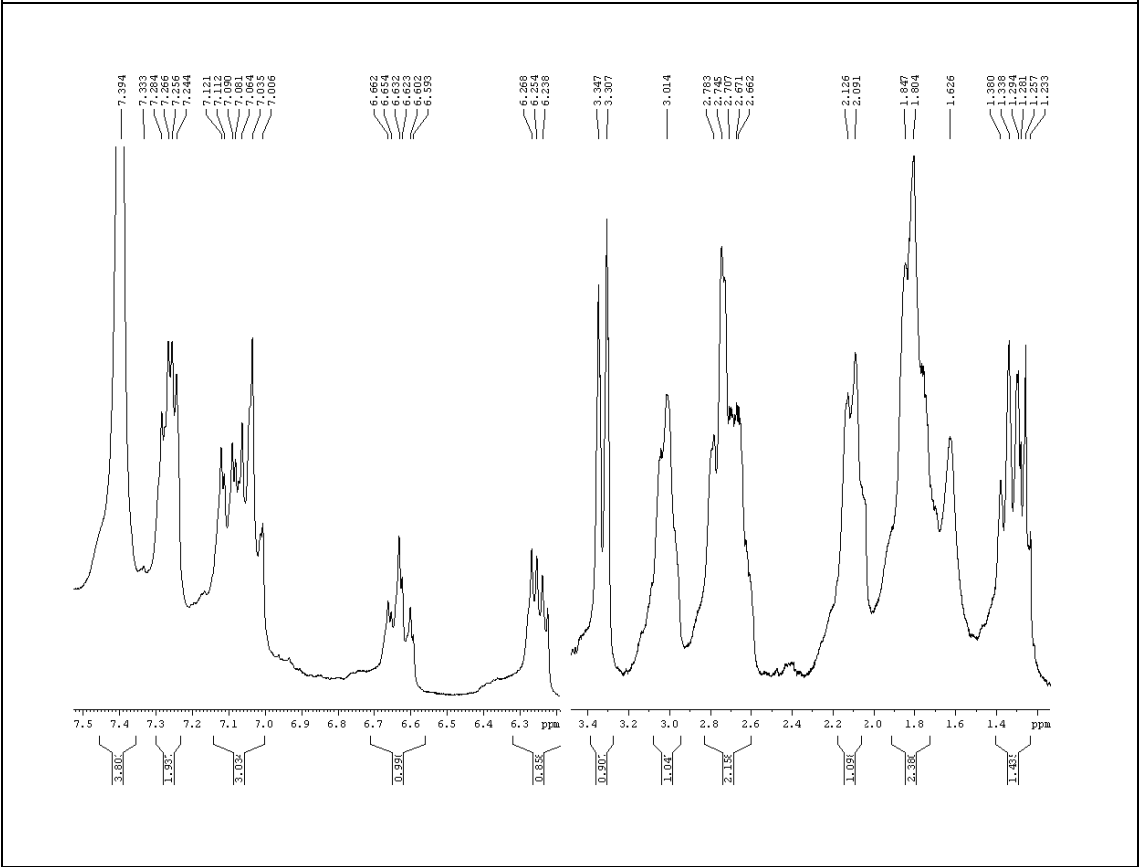


Figure 82. ¹H NMR Spectrum of 3x (expanded)

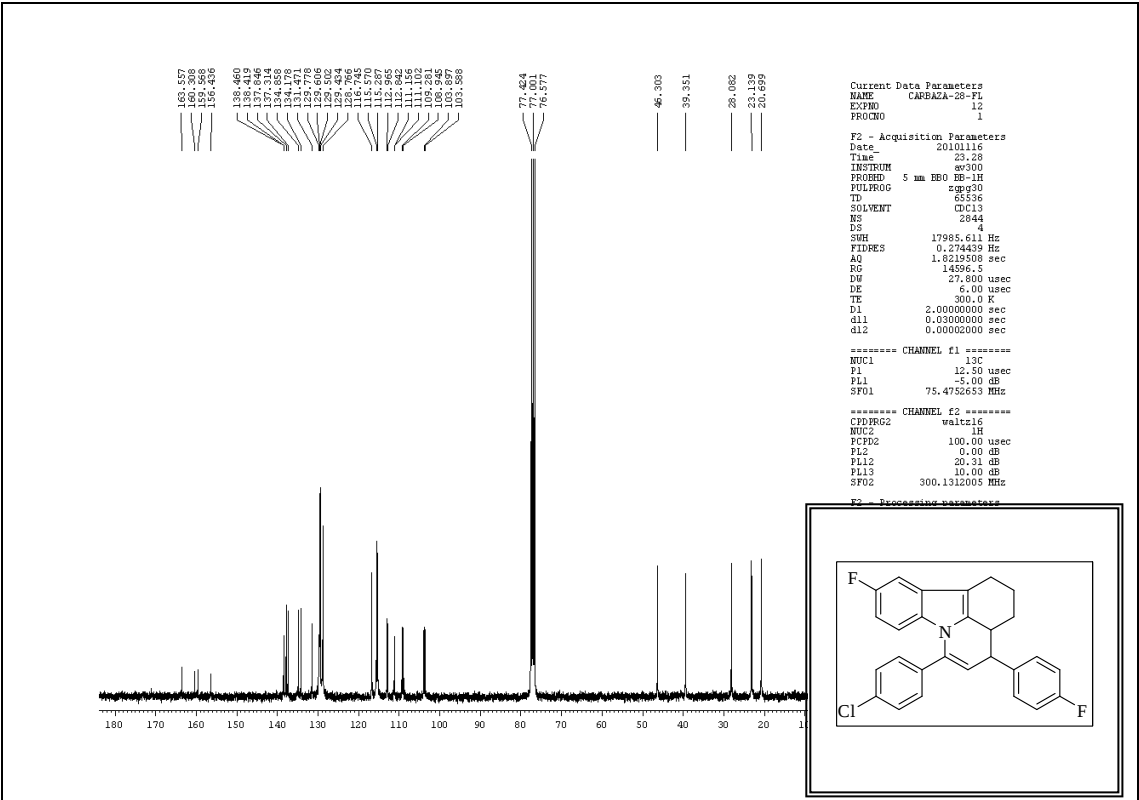


Figure 83. ¹³C NMR Spectrum of 3x (CDCl₃)

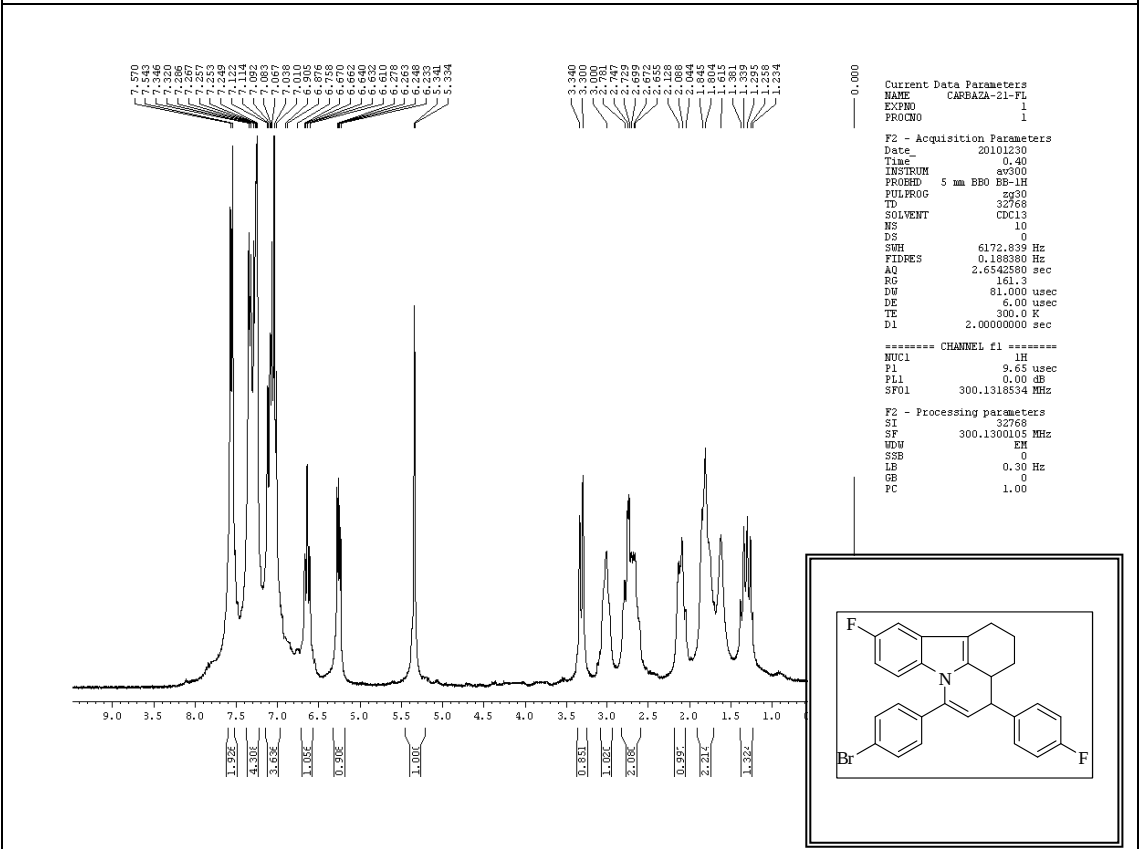


Figure 84. ¹H NMR Spectrum of 3y (CDCl₃)

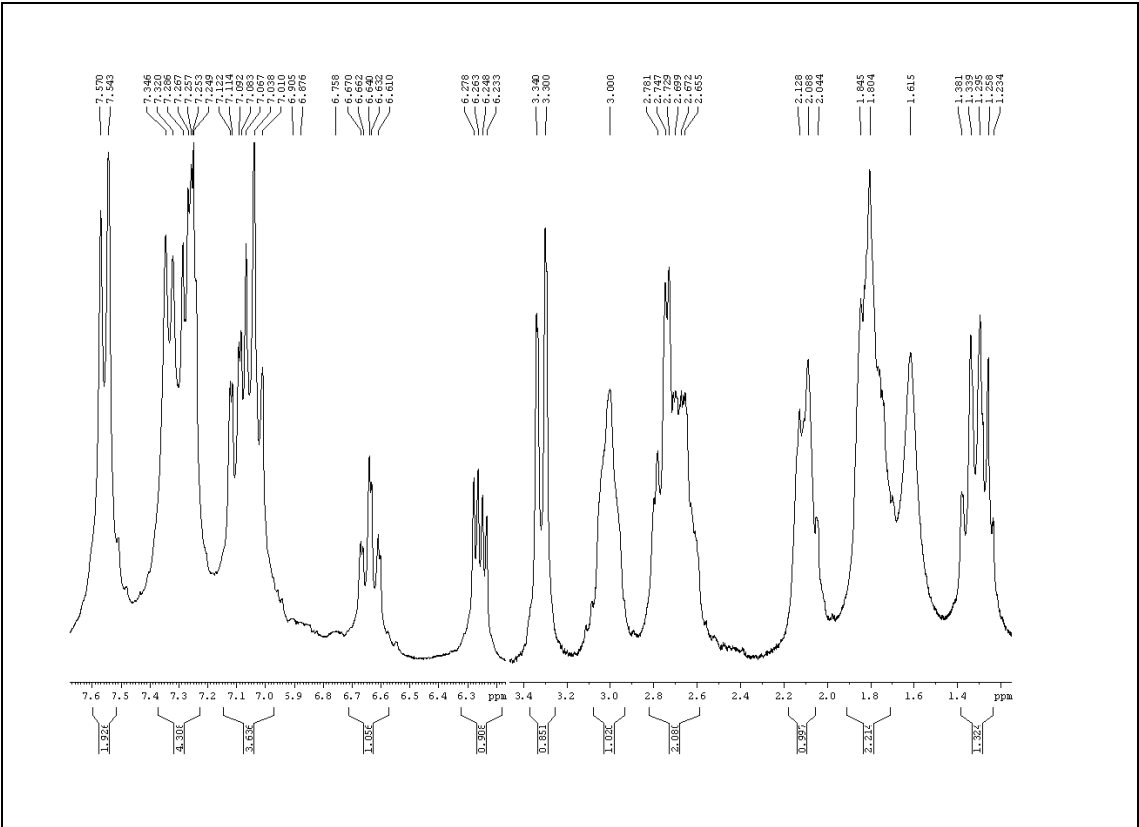


Figure 85. ¹H NMR Spectrum of 3y (expanded)

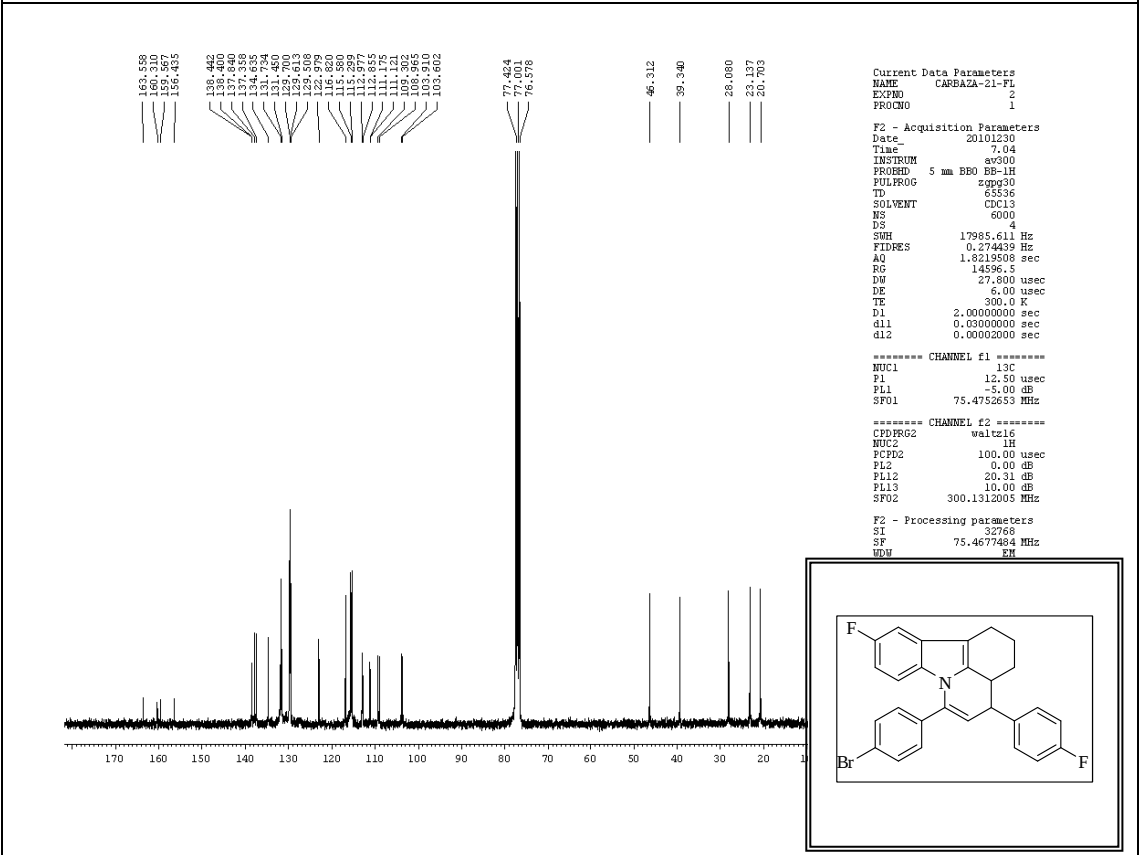


Figure 86. ¹³C NMR Spectrum of 3y (CDCl₃)

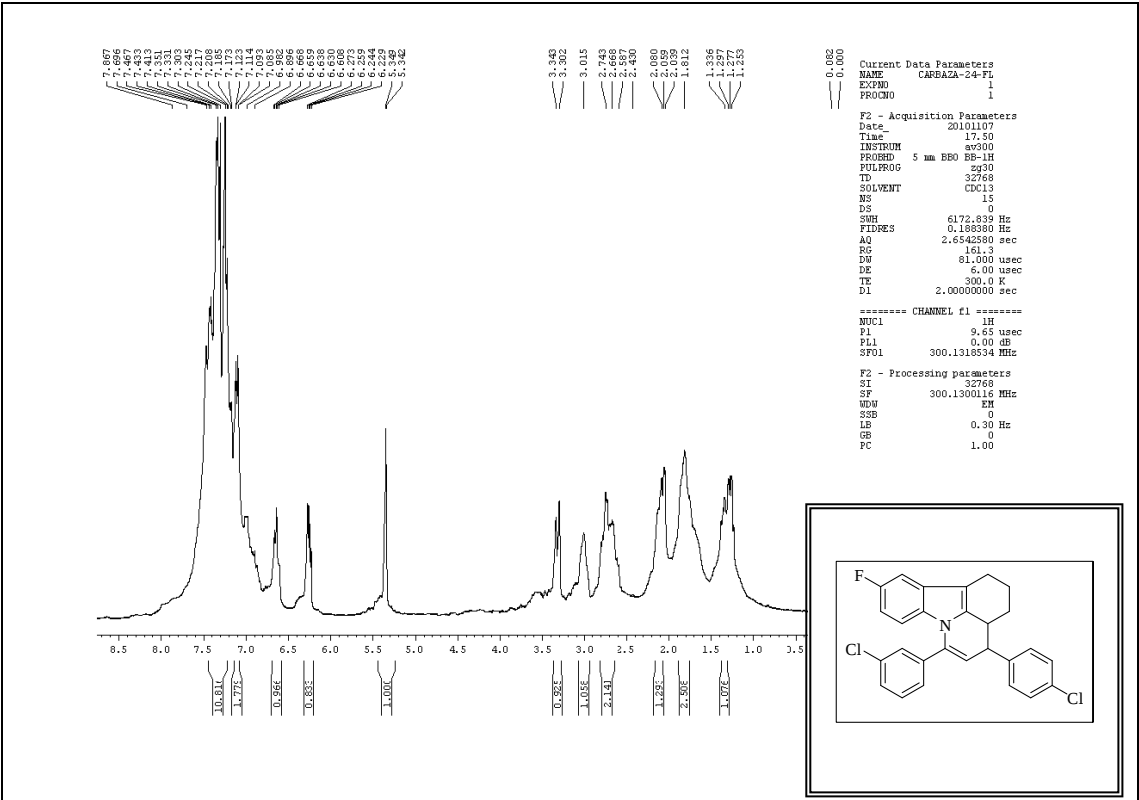


Figure 87. ¹H NMR Spectrum of 3z (CDCl₃)

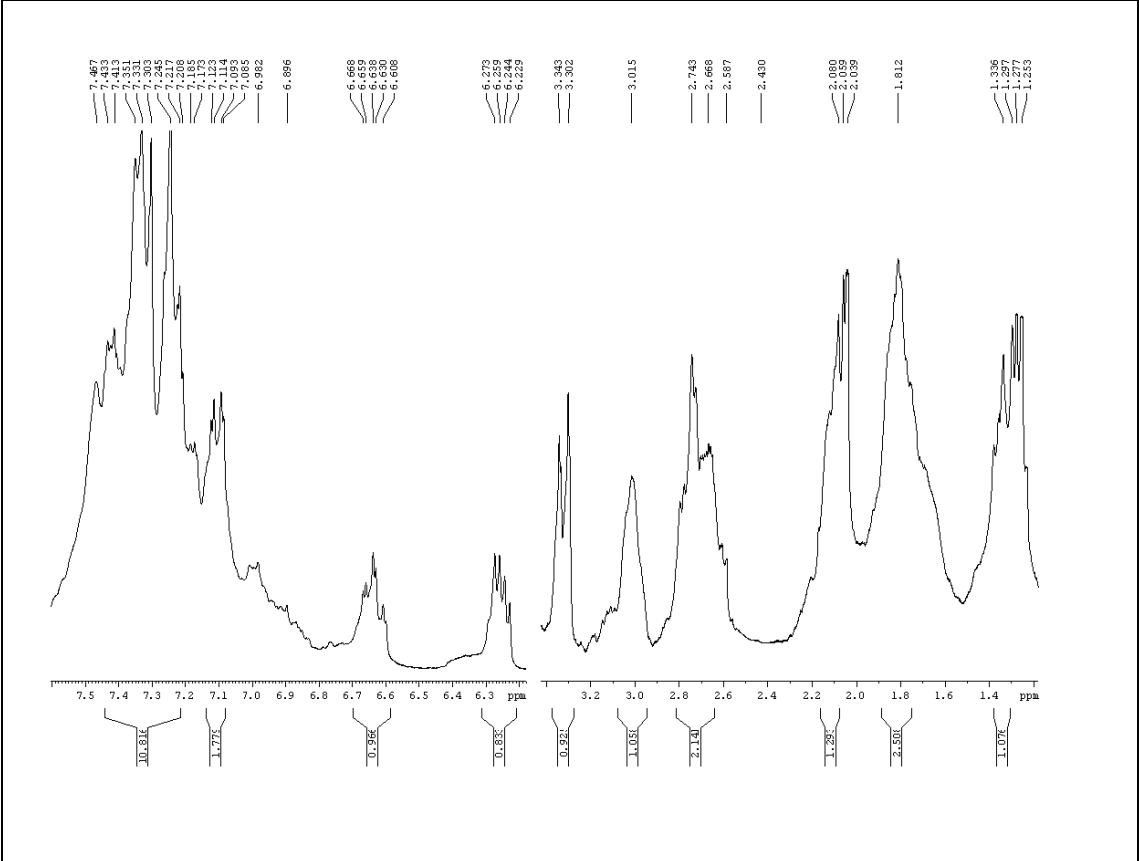
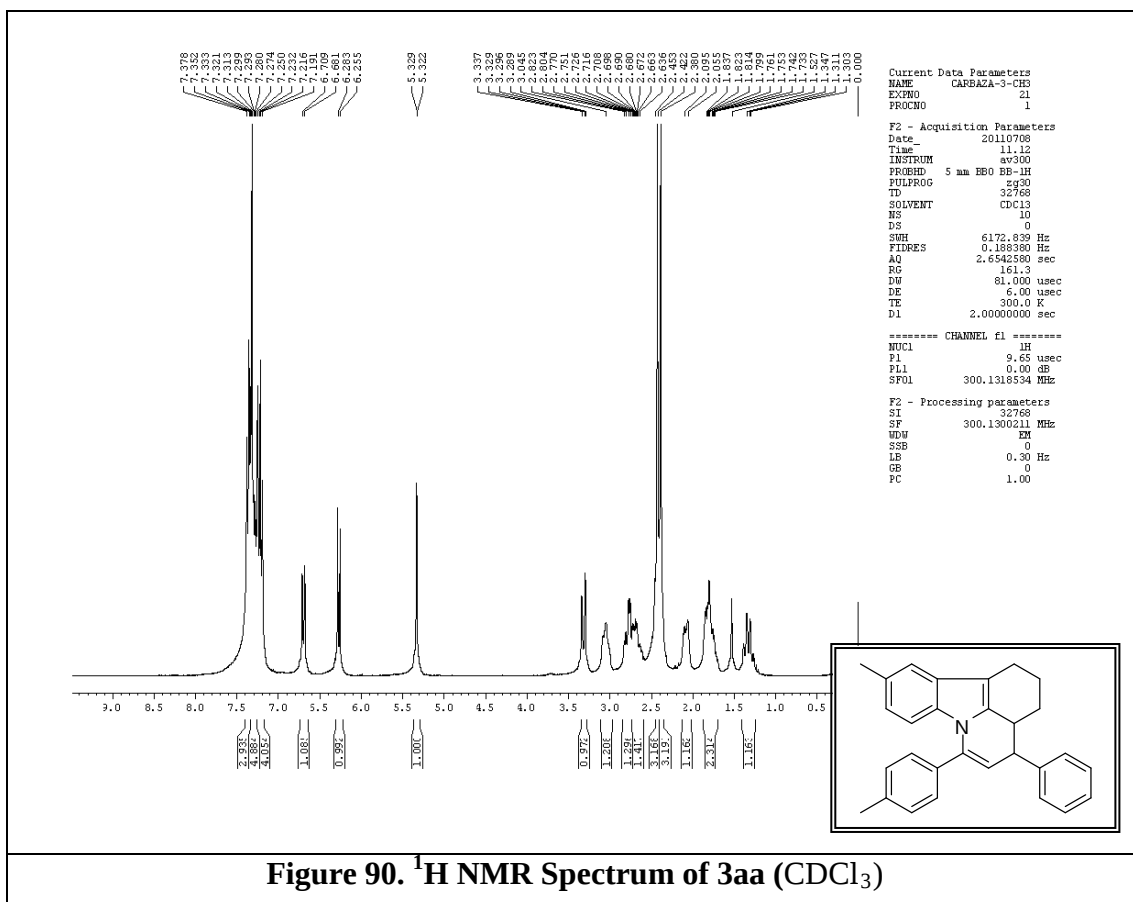
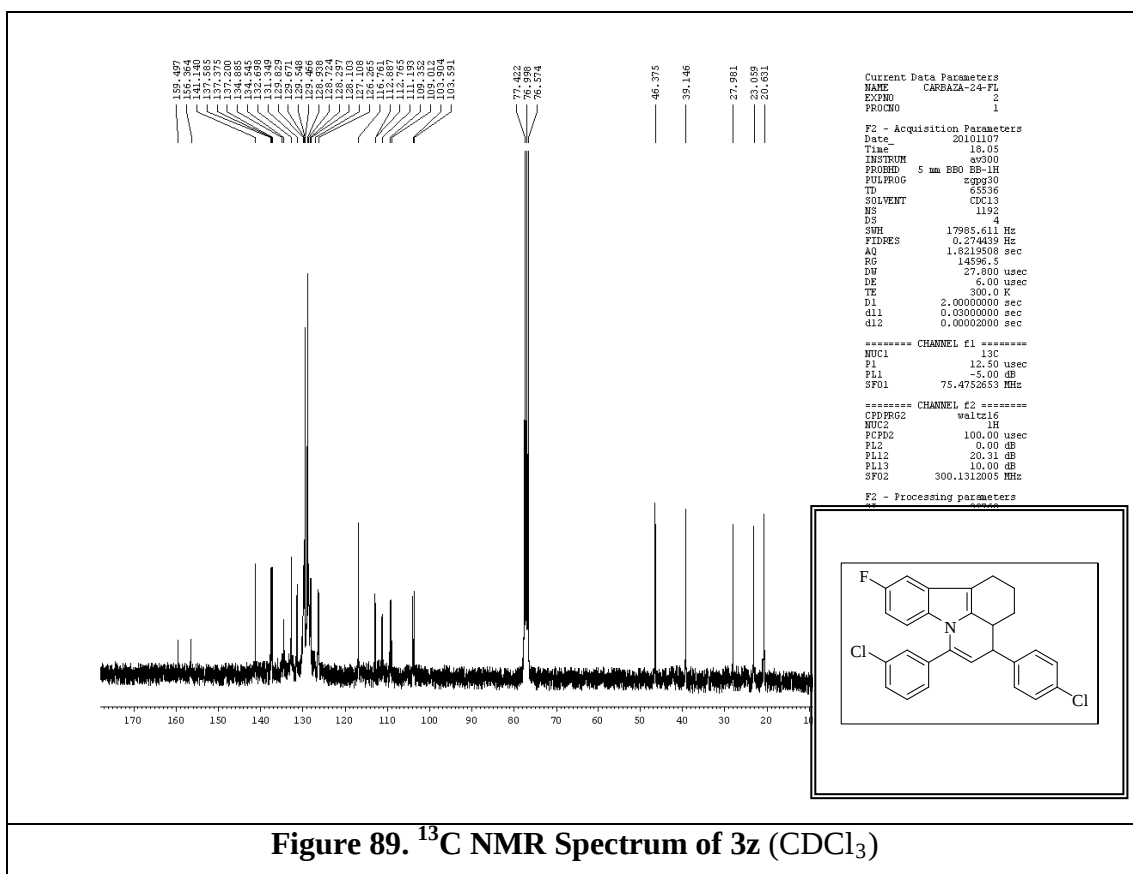


Figure 88. ¹H NMR Spectrum of 3z (expanded)



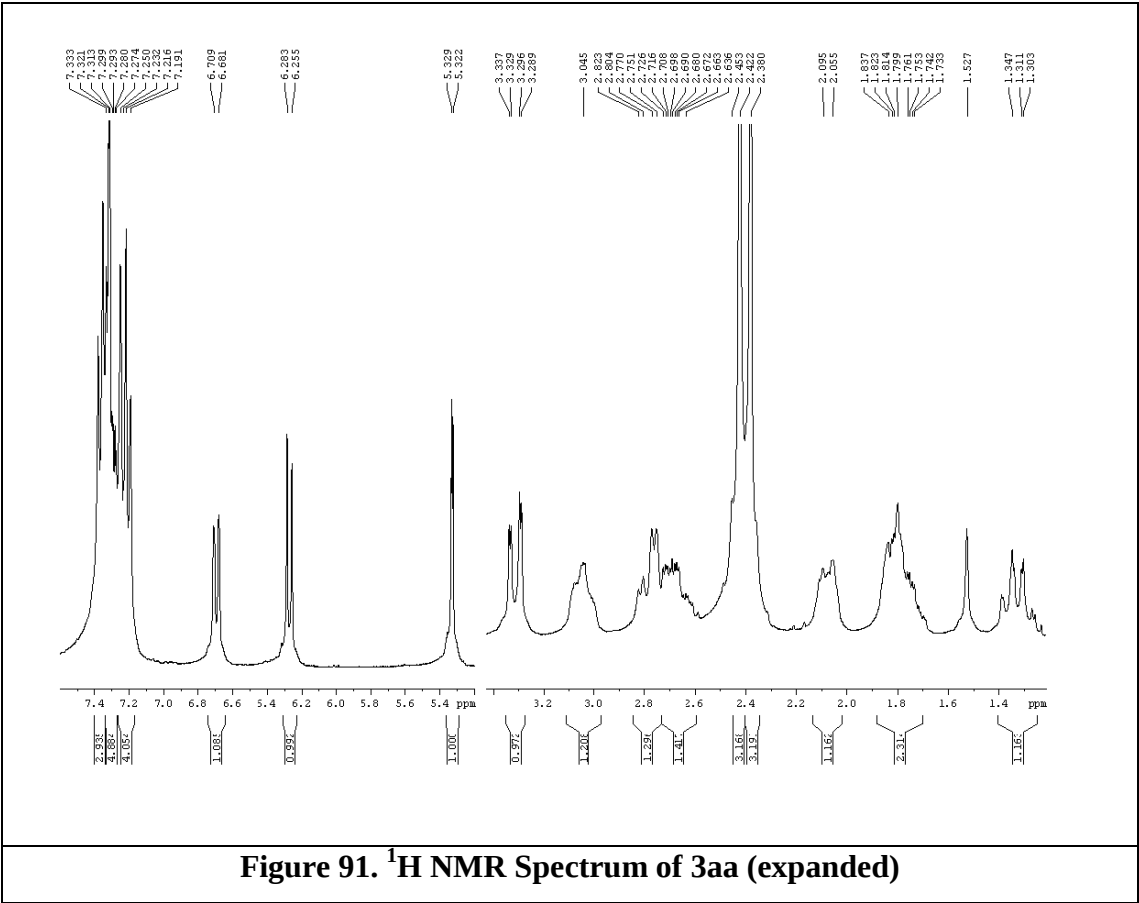


Figure 91. ¹H NMR Spectrum of 3aa (expanded)

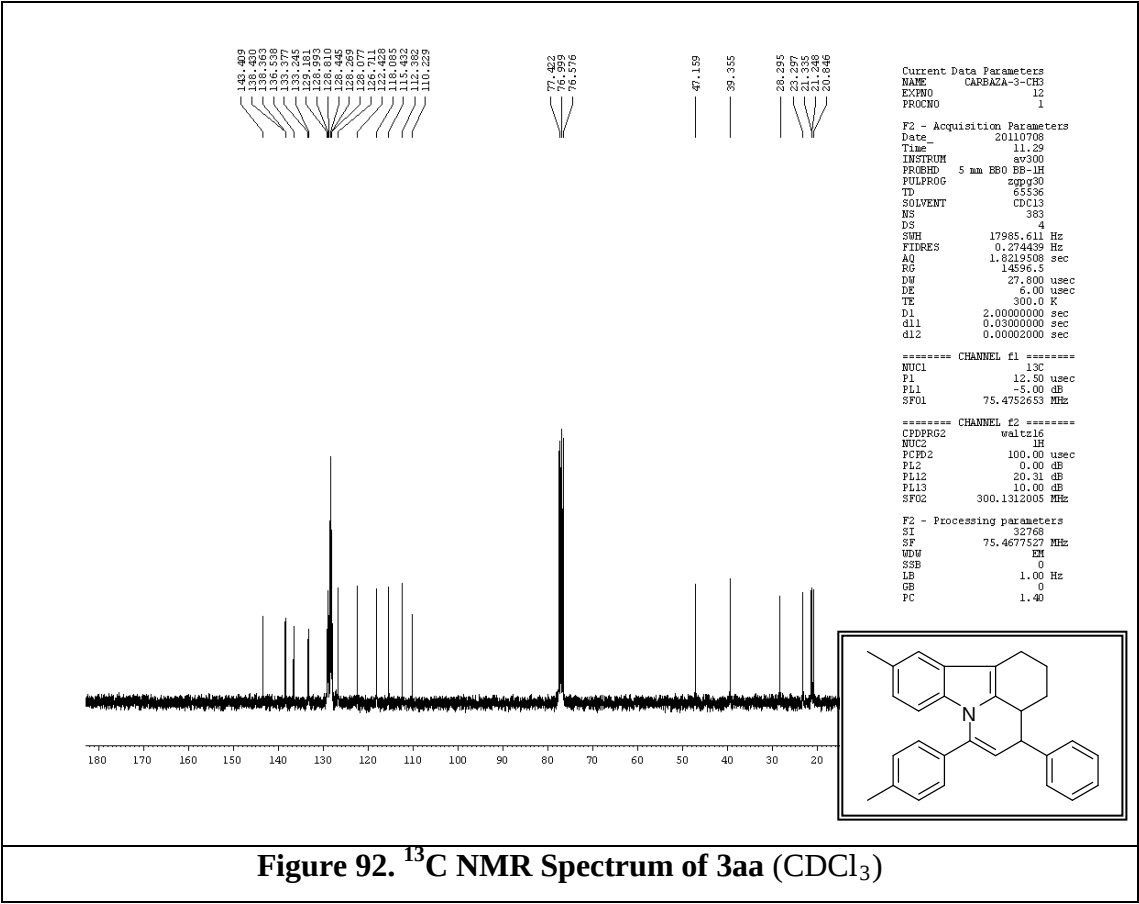
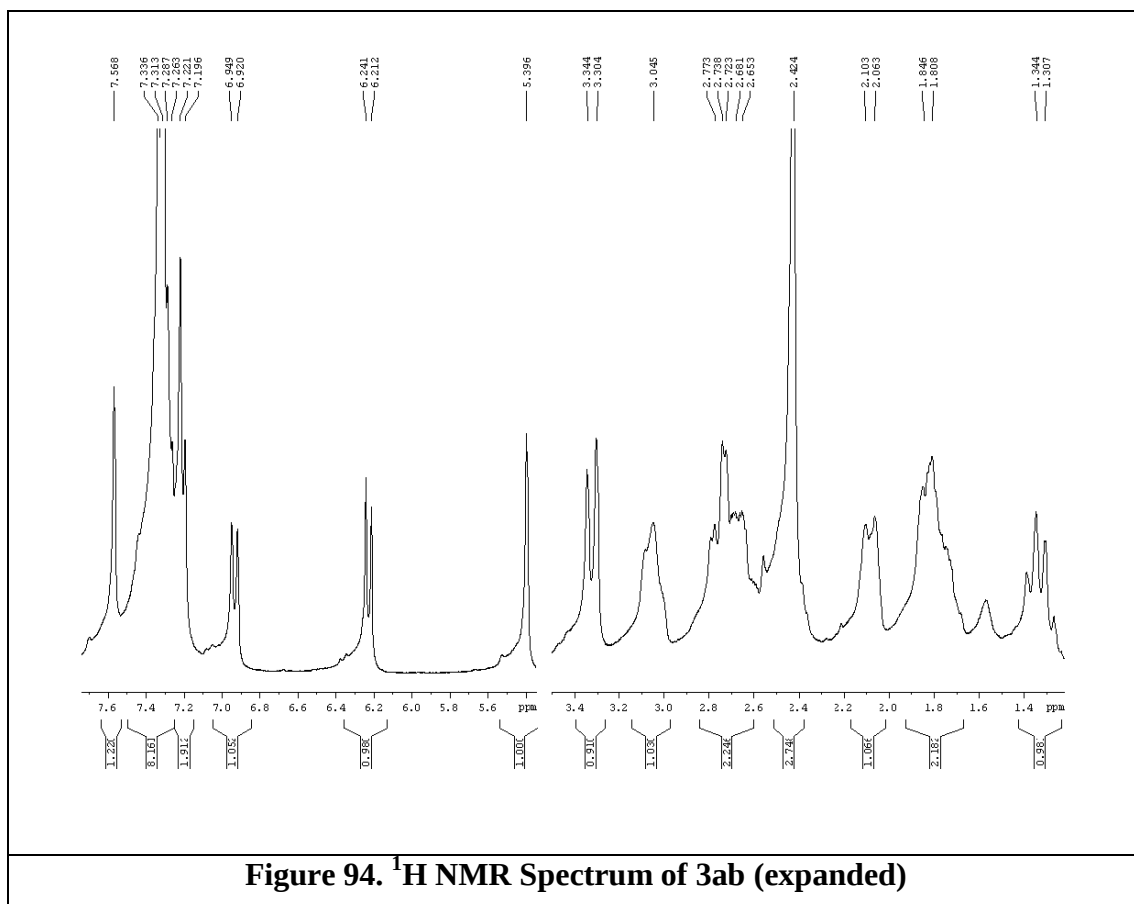
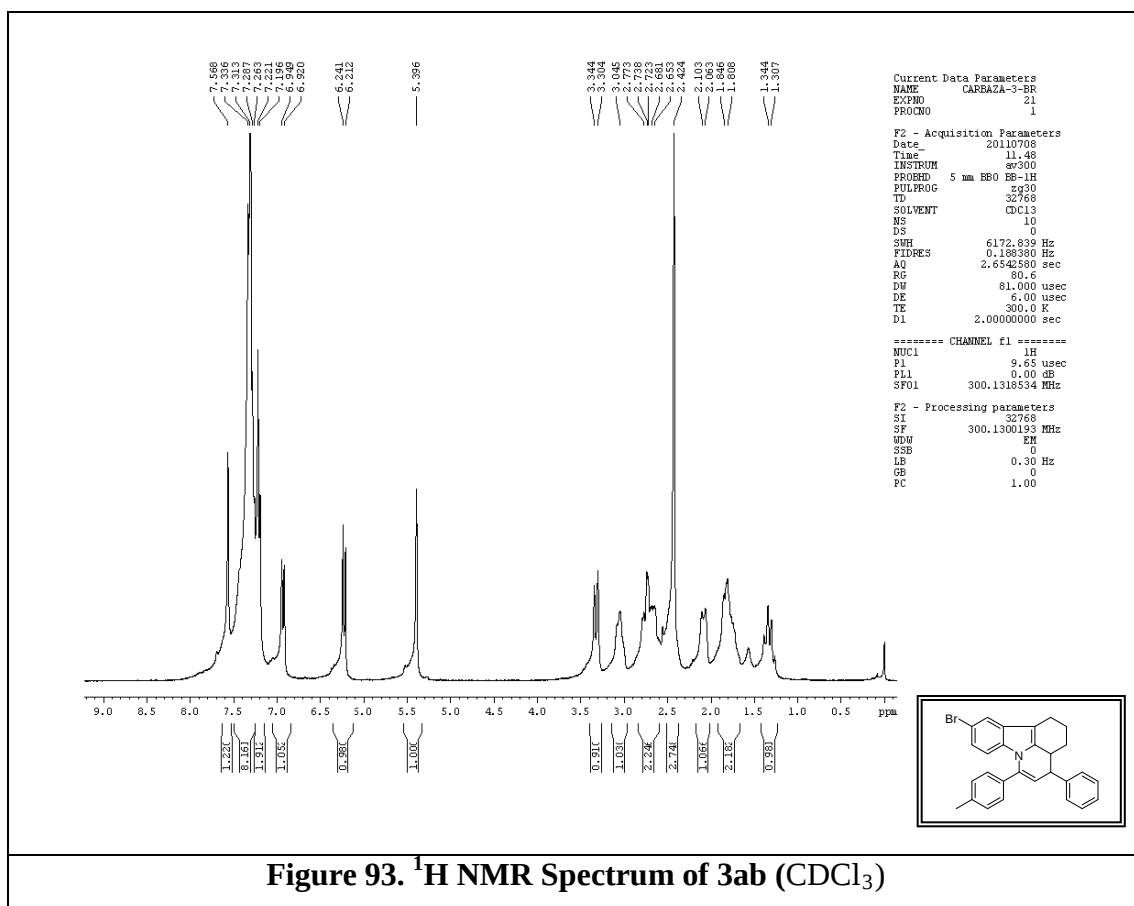
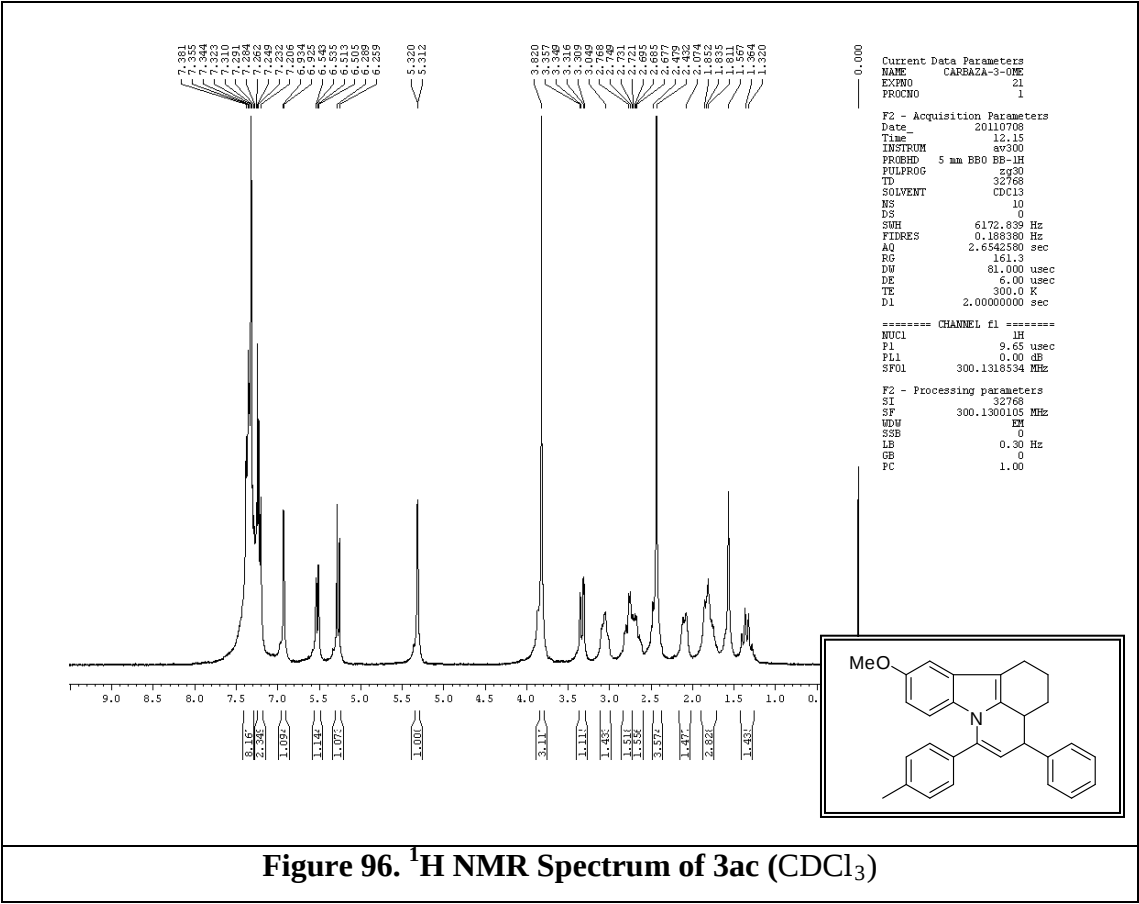
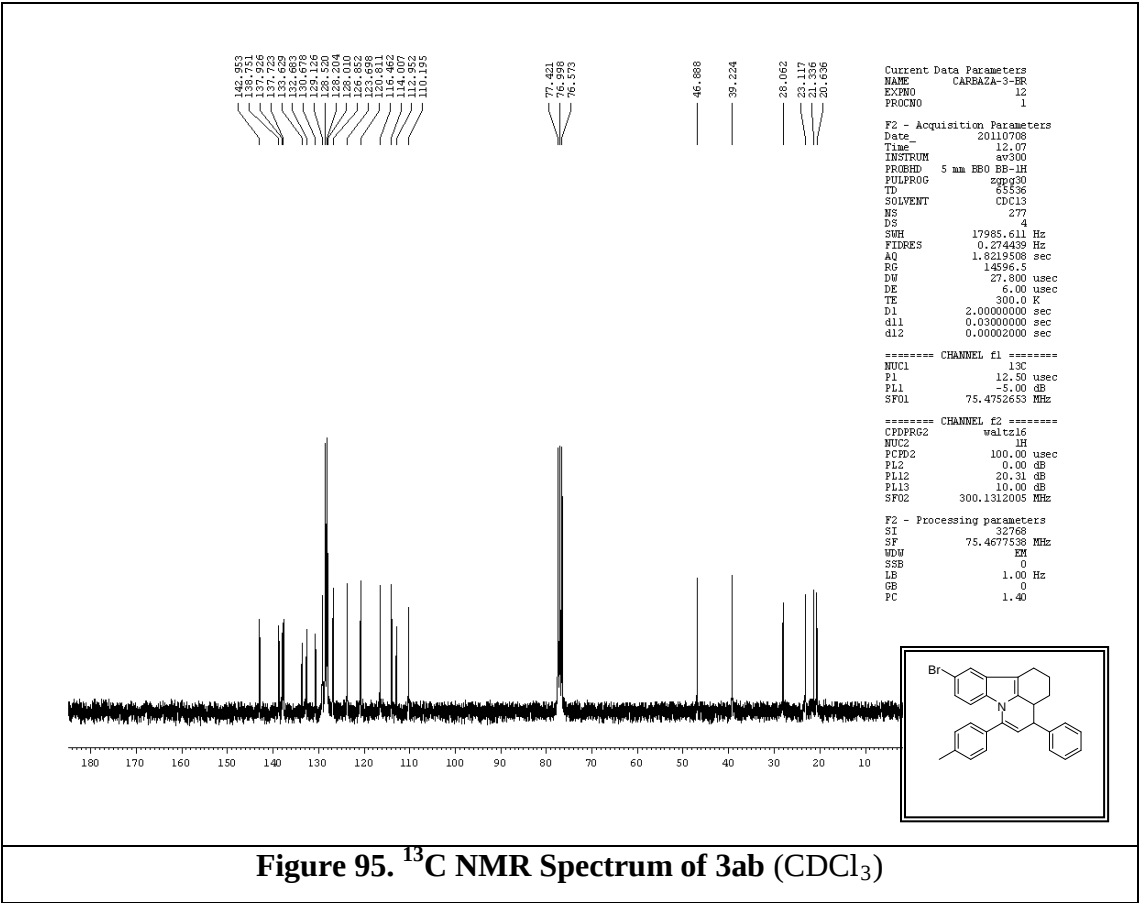
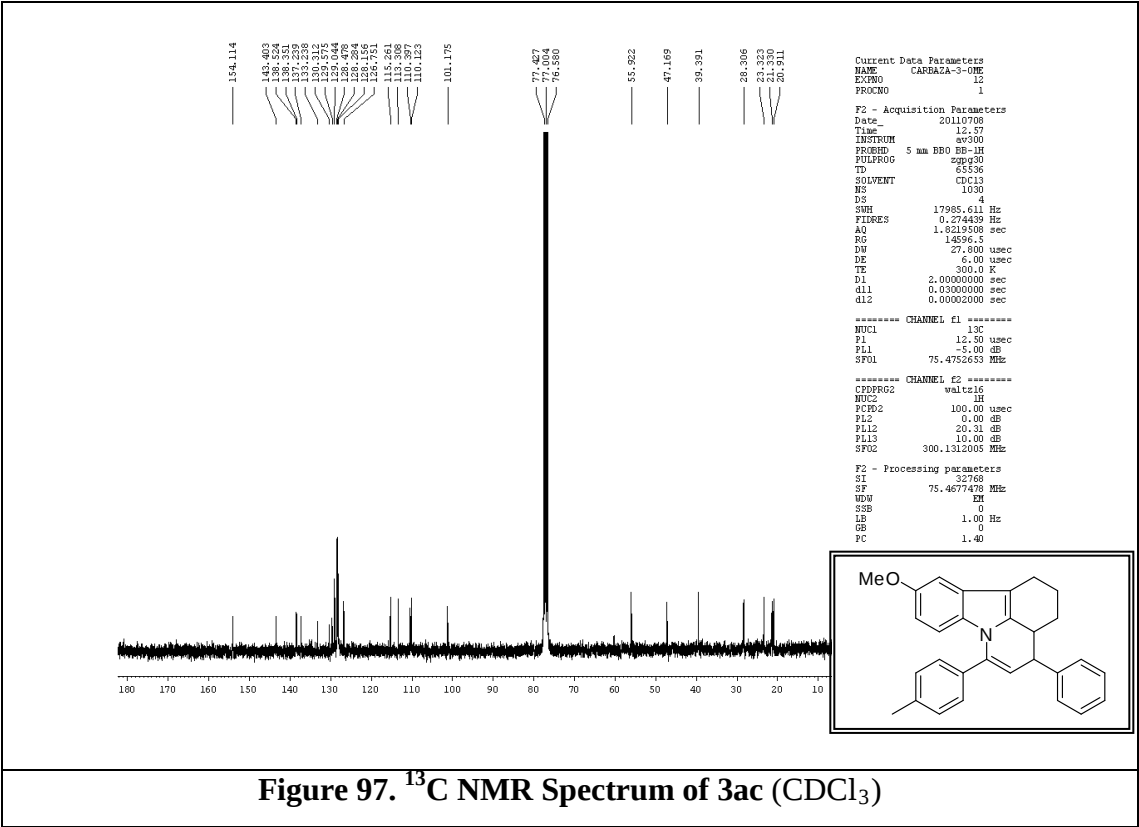
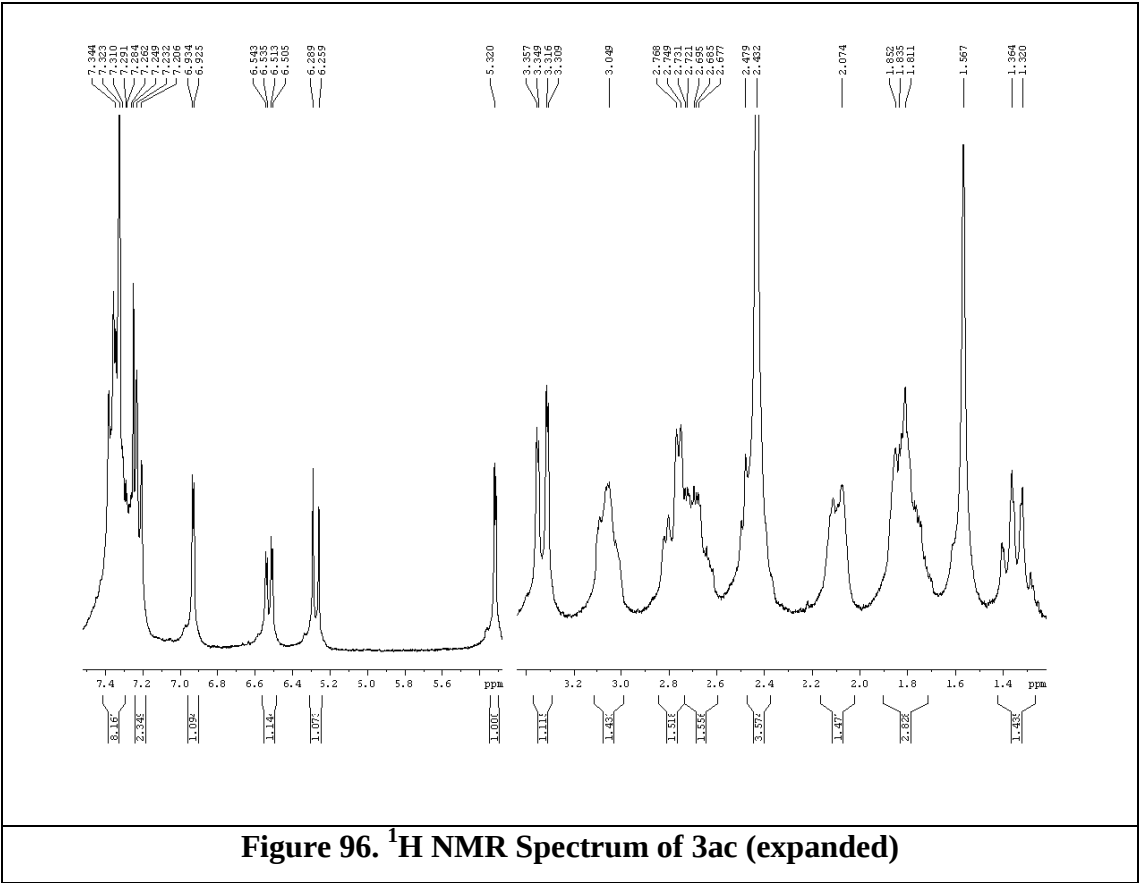
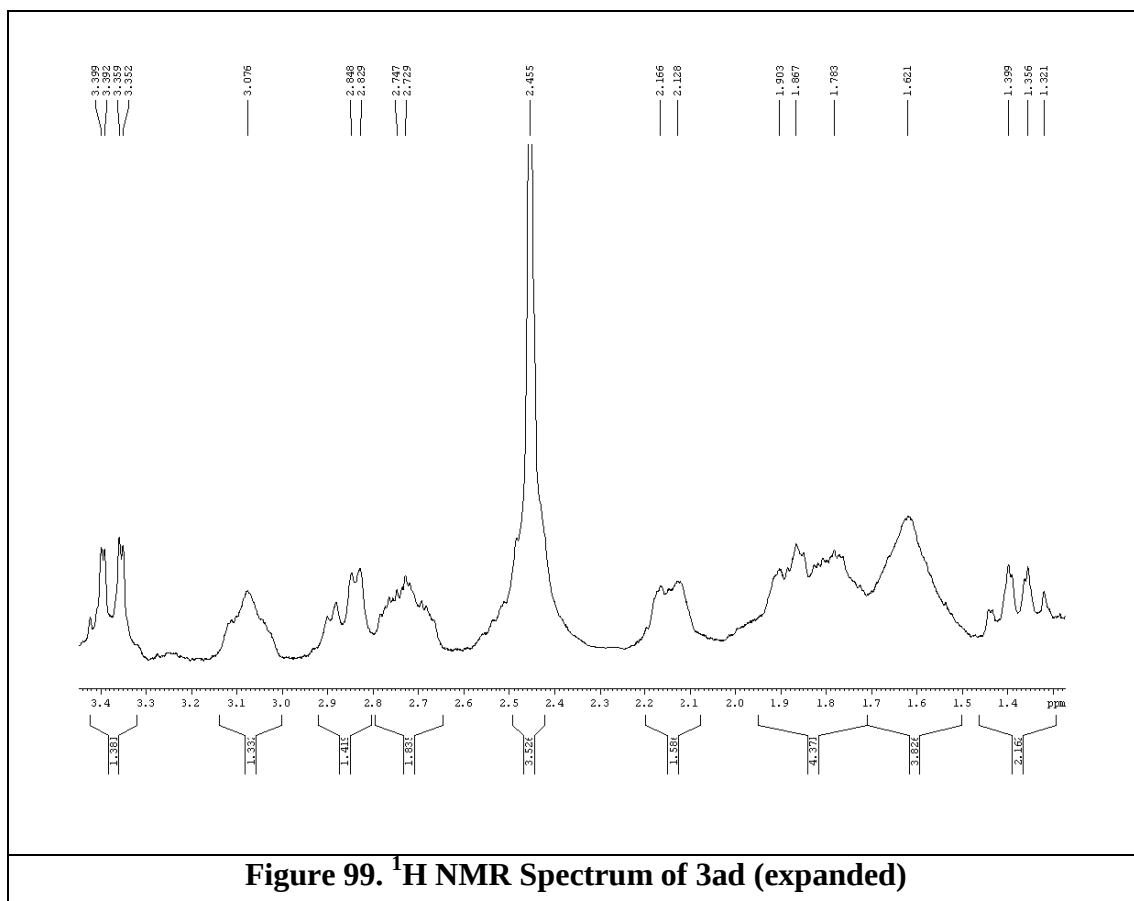
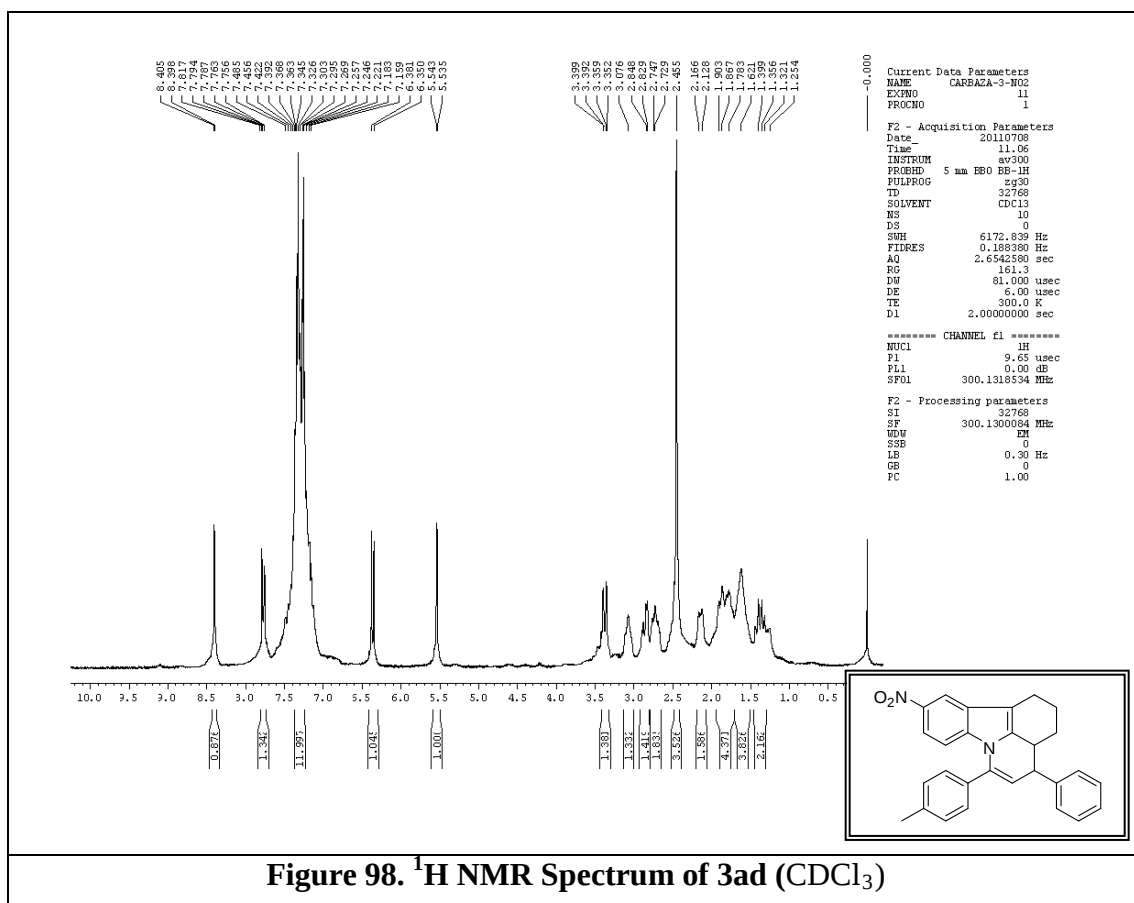


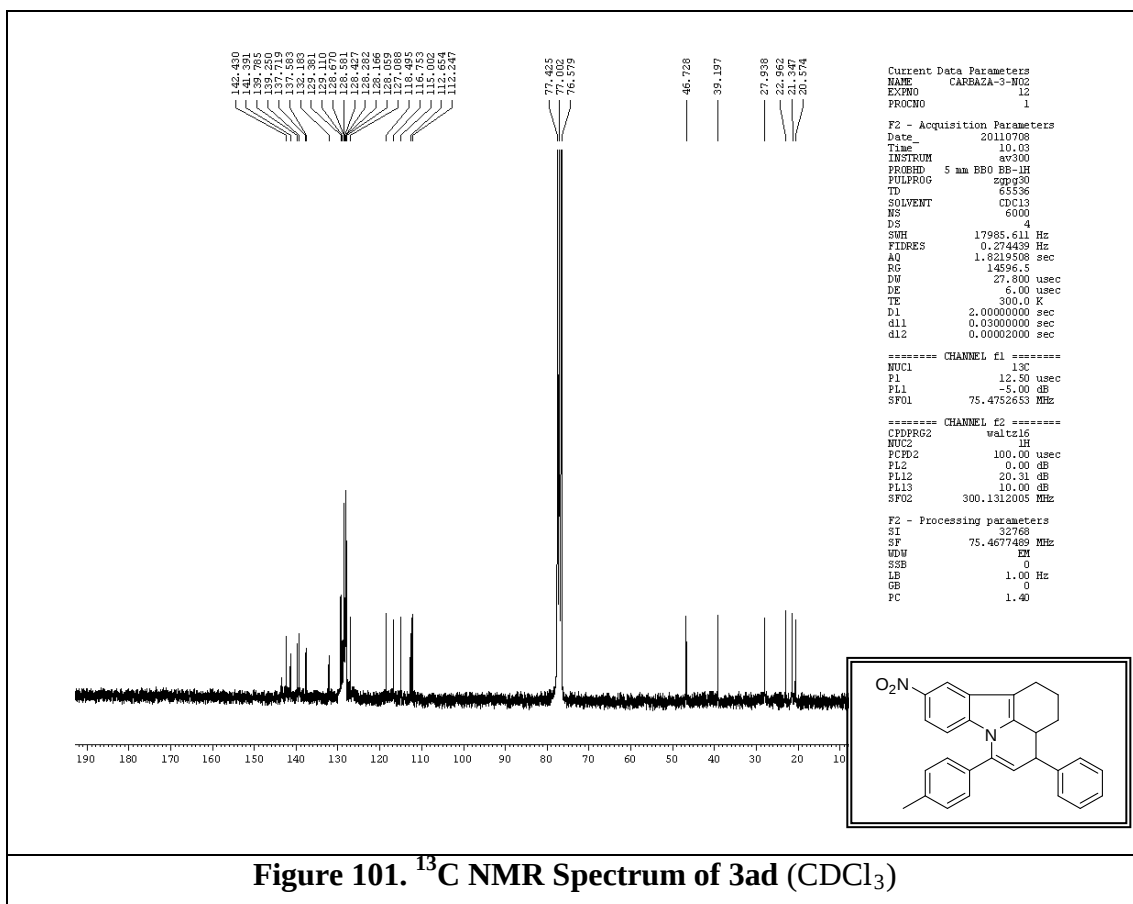
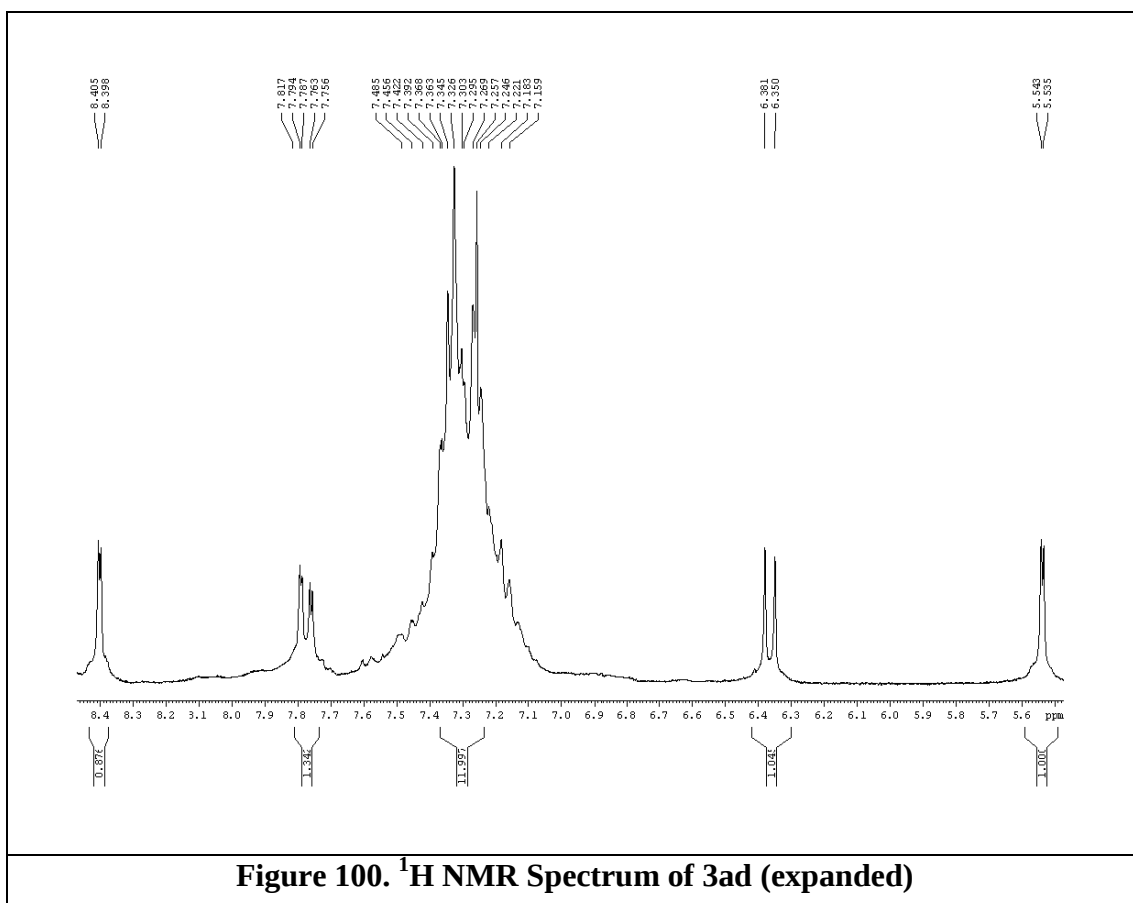
Figure 92. ¹³C NMR Spectrum of 3aa (CDCl₃)

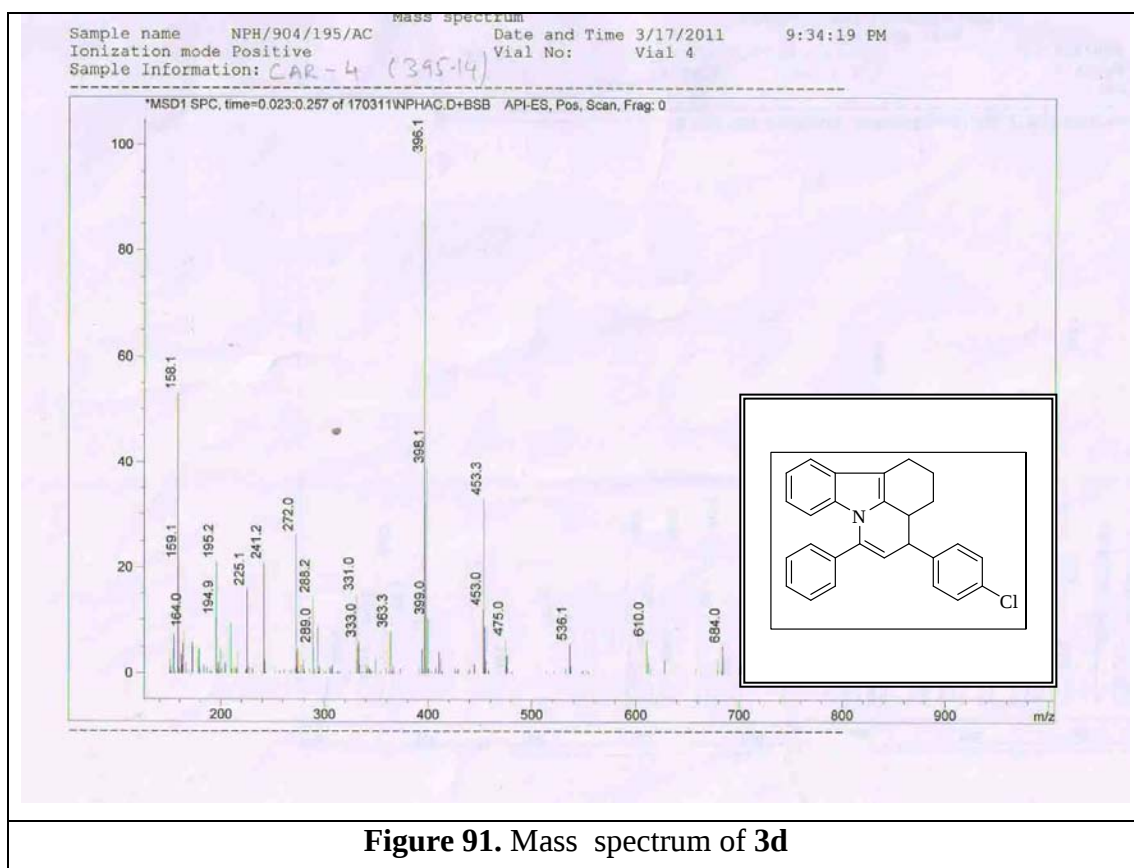
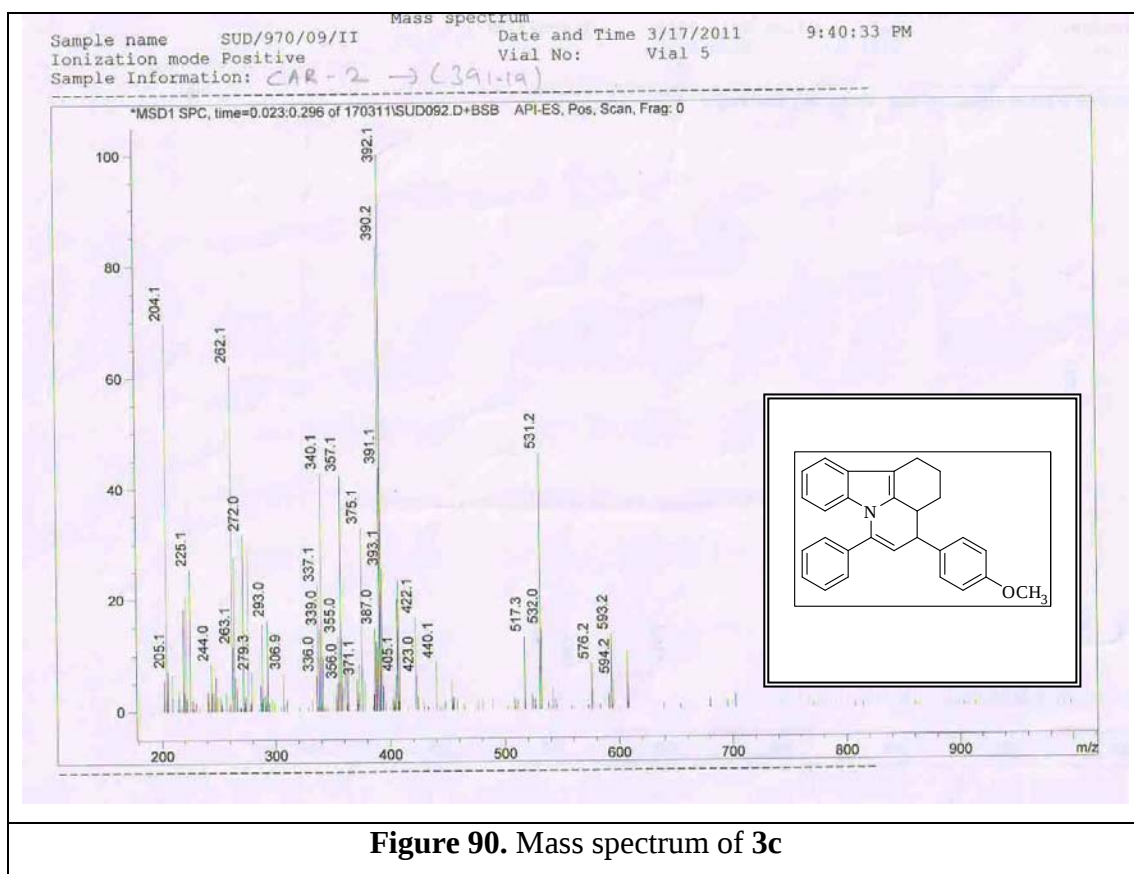












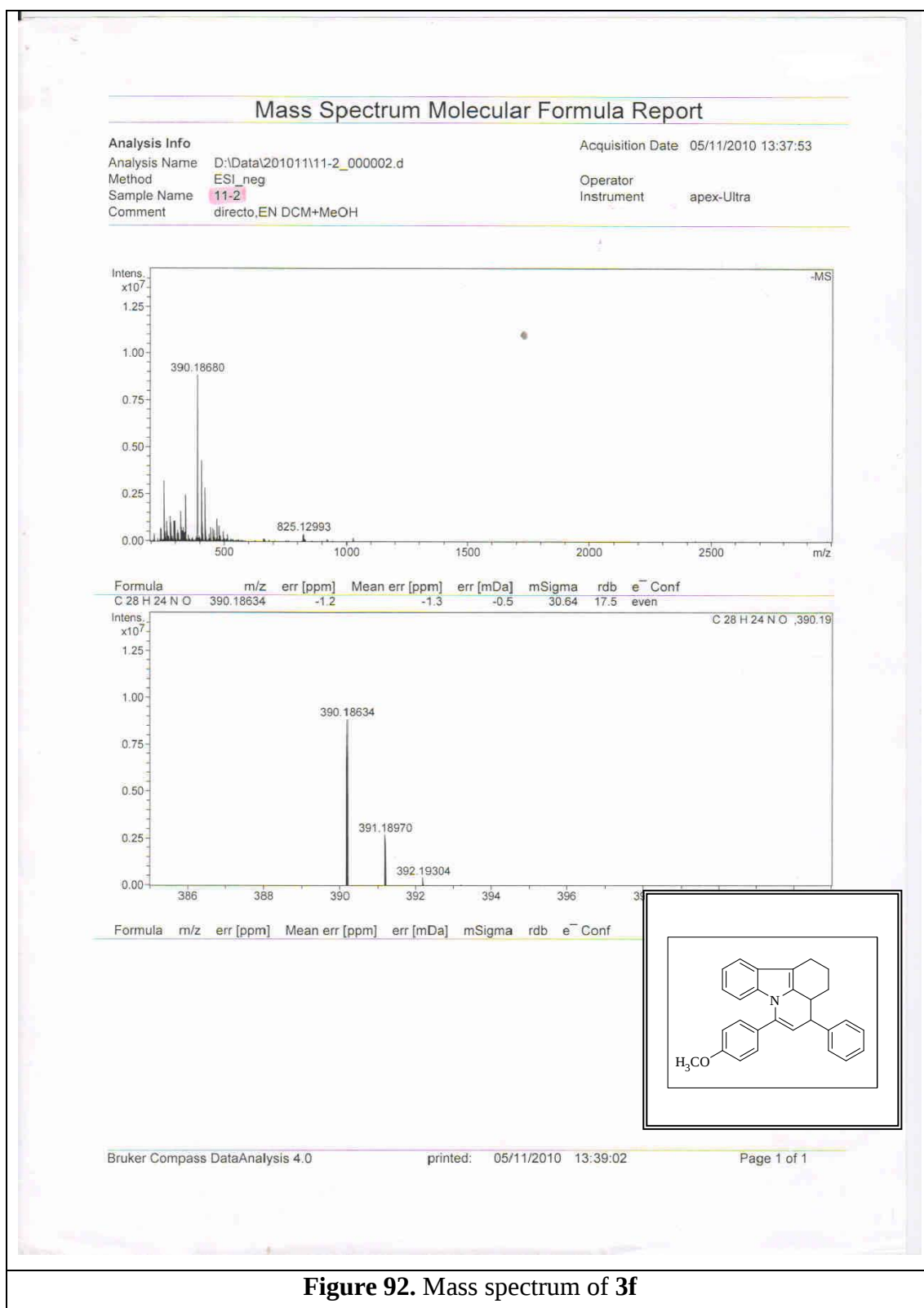


Figure 92. Mass spectrum of **3f**

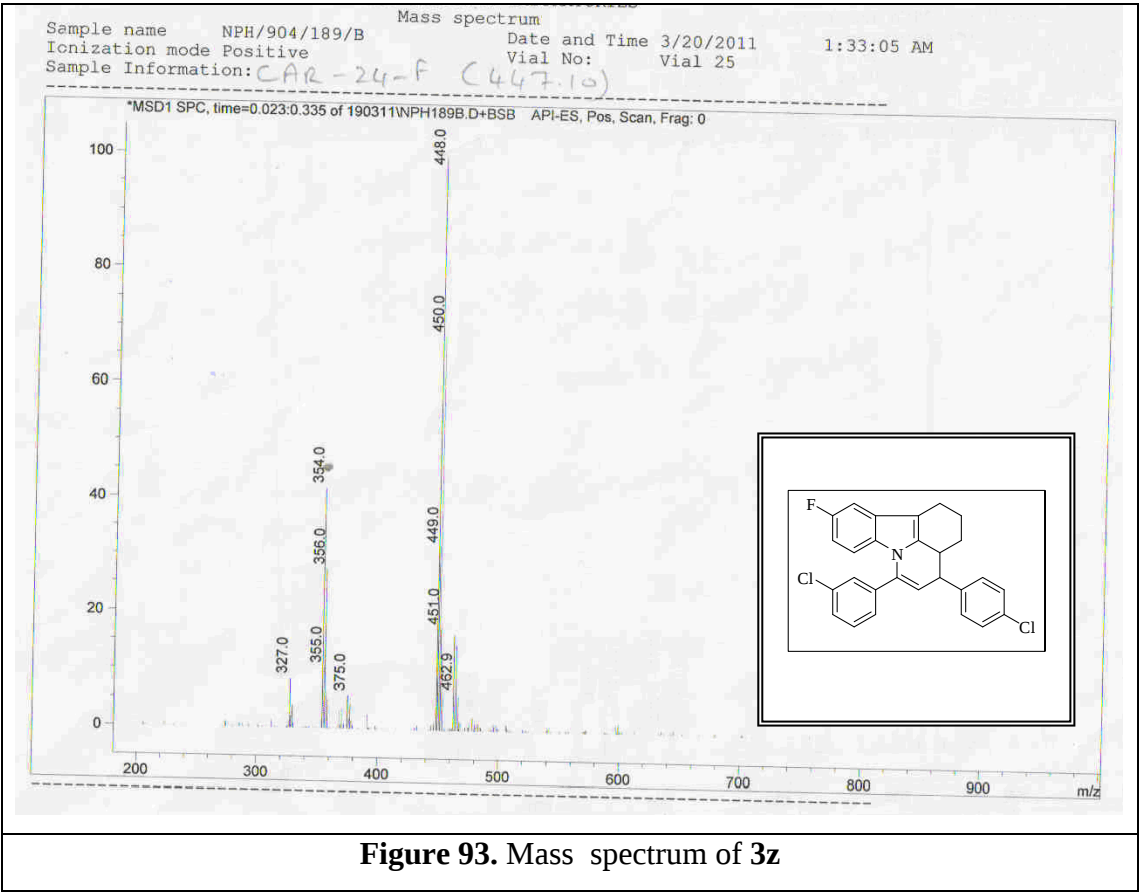


Figure 93. Mass spectrum of 3z