Supporting Information

For

Concise and Efficient Synthesis of Indole-Indolone Scaffolds through MeOTf-Induced Annulation of *N*-(2-Cyanoaryl)indoles

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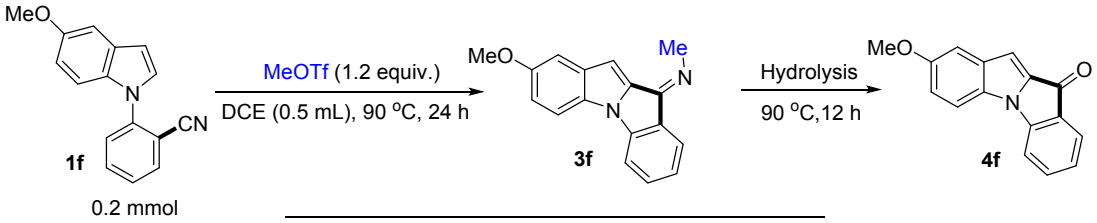
1. *Optimized the hydrolyzation reaction conditions*



Entry	Hydrolysis	Yield (%)	
		3f	**4f**
1	Na_2CO_3 (.aq)	63	24
2	K_2CO_3 (.aq)	58	26
3	Na_2OAc (.aq)	54	25
4	NaOH (.aq)	56	28
5	1M HCl	49	30
6	3M HCl	32	47
7	6M HCl	0	87

Figure S1. Optimized the hydrolyzation reaction conditions

We further explored the hydrolyzation of reaction conditions to give the final target **4f**. Acidic and basic conditions were applied from entry 1 to entry 7 after first step. Finally, the best hydrolyzation reaction condition in hand is entry 7 achieving 87% isolated yield.

2. NOESY (Nuclear Overhauser Effect Spectroscopy 3b and 3m)

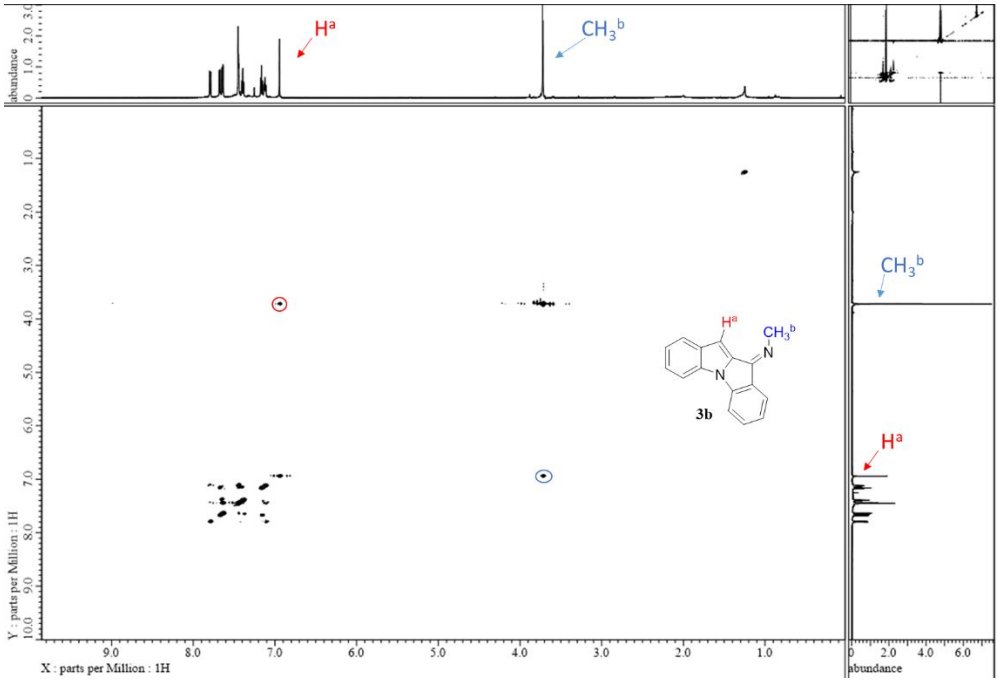


Figure S2. NOESY for **3b**

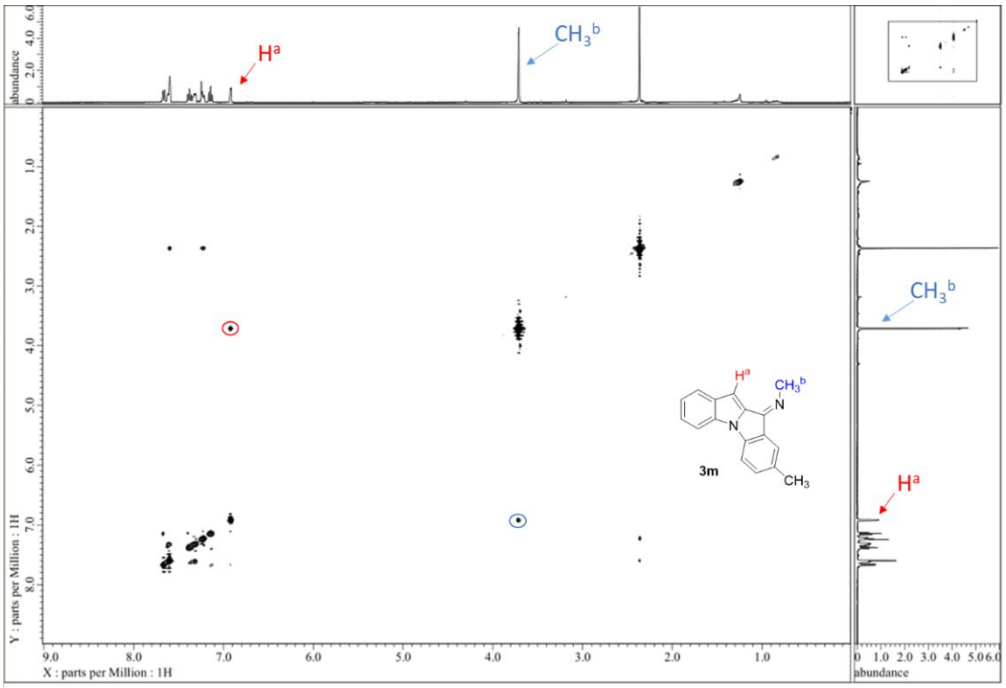


Figure S3. NOESY for **3m**

To decide the configuration of C=N bonds in compound **3**, we also did the NOESY experiment. The strong coupling was demonstrated between H^a and H_3^b in the

compound **3b** and **3m** from NOESY experiment chart, hence, the configuration of C=N bonds in compound **3** is *Z* configuration.

3. Copies of ^{1}H, $^{13}C\{^{1}H\}$, and ^{19}F NMR Spectra

3-1 Copies of ^{1}H NMR Spectra for Starting Materials 1 and Copies of ^{13}C NMR Spectra for Starting Materials 1.

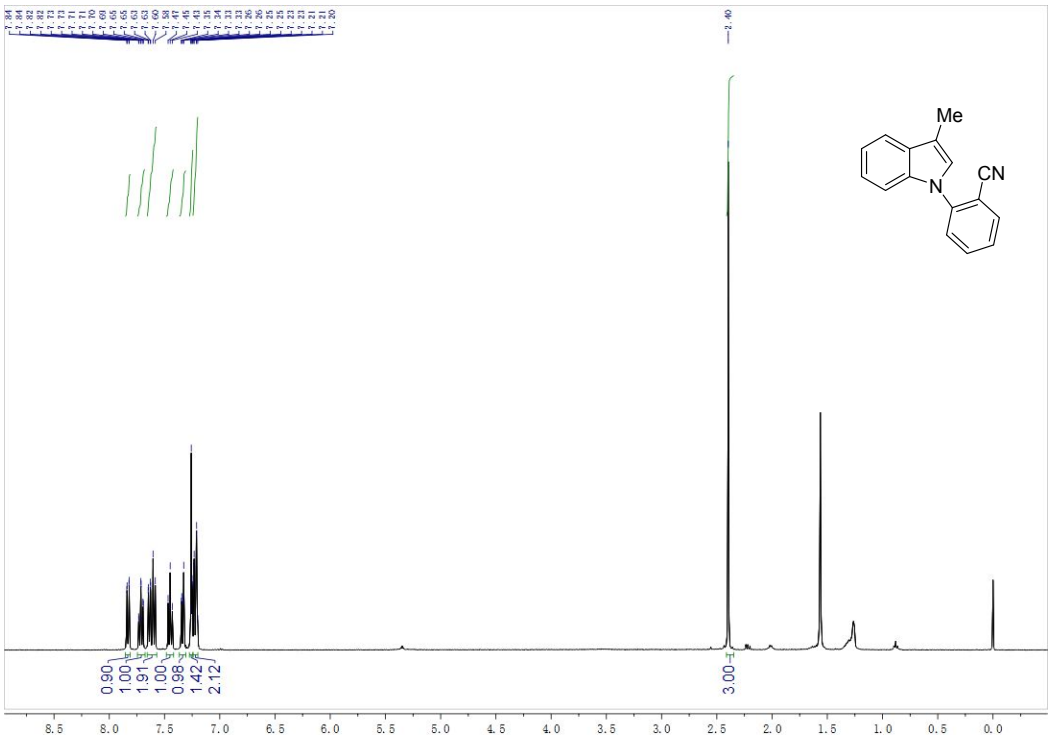


Figure S4. ^{1}H NMR for compound **1a**

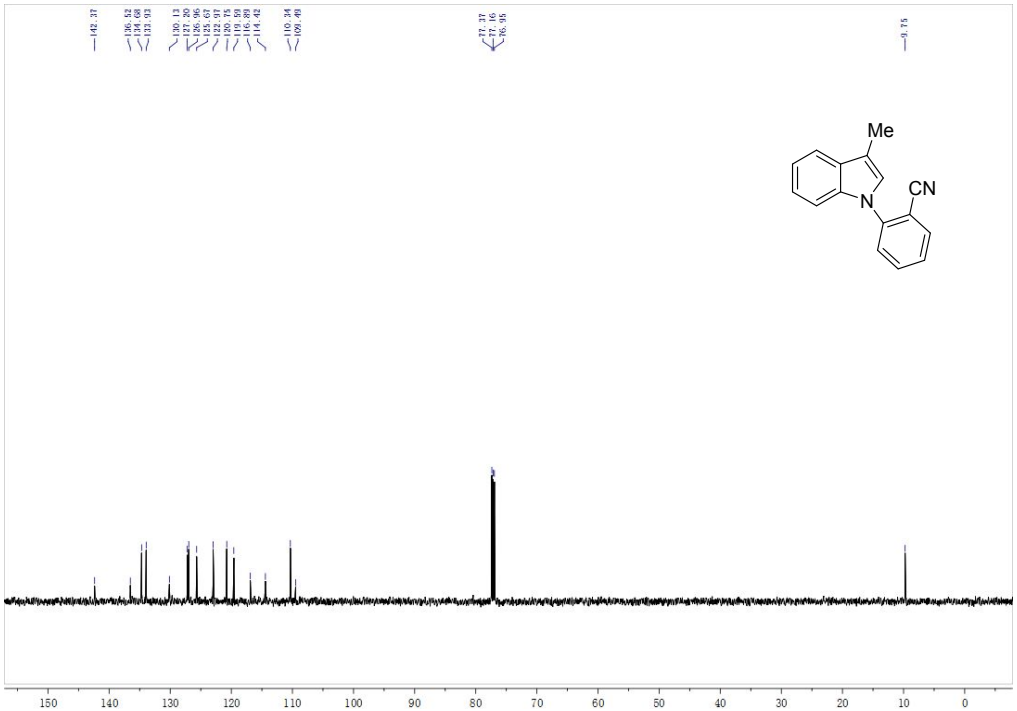


Figure S5. $^{13}C\{^{1}H\}$ NMR for compound **1a**

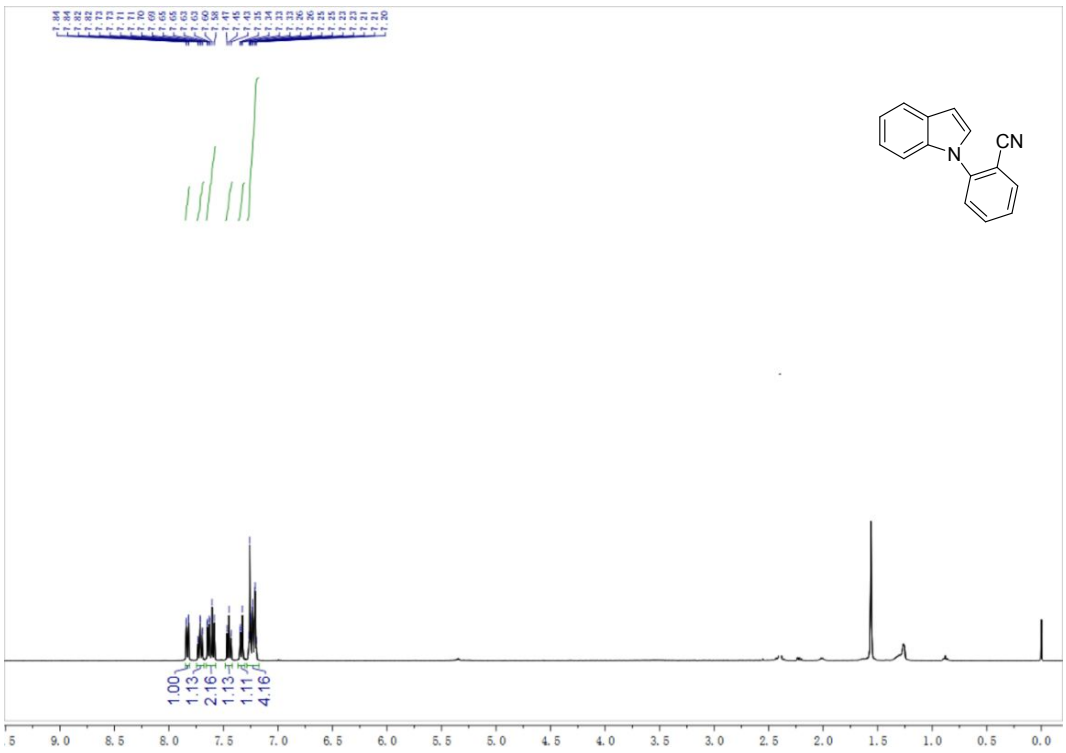


Figure S6. ^{1}H NMR for compound **1b**

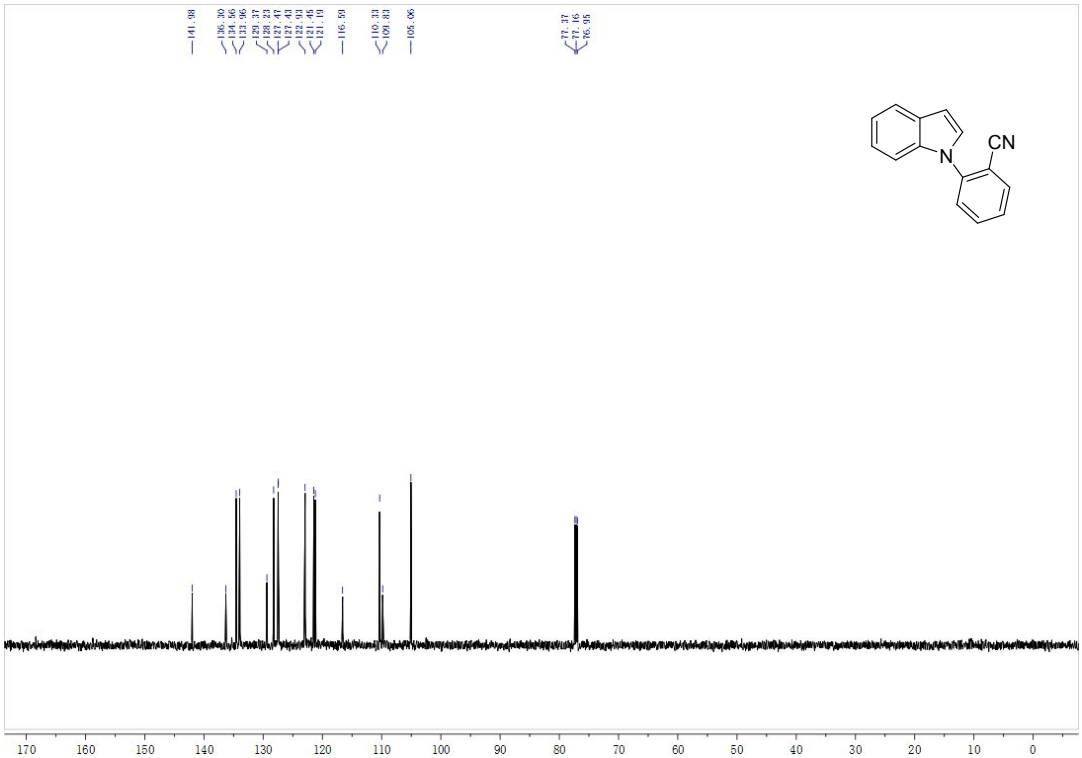


Figure S7. $^{13}C\{^{1}H\}$ NMR for compound **1b**

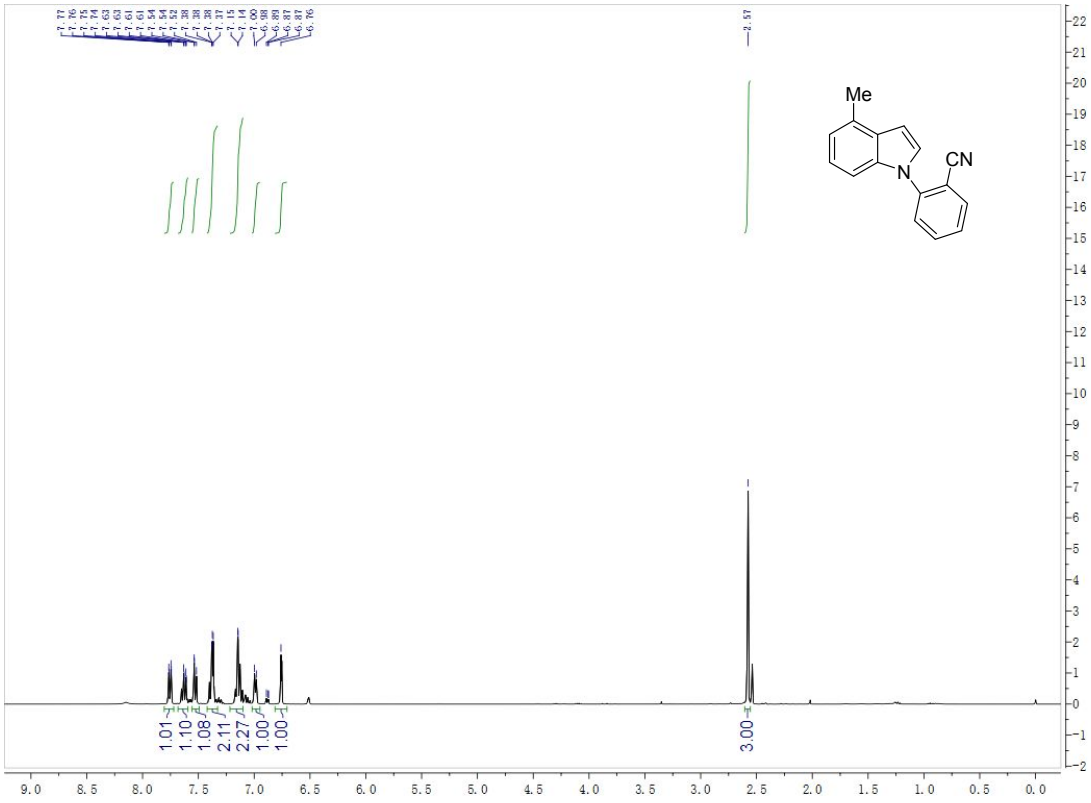


Figure S8. ^{1}H NMR for compound **1c**

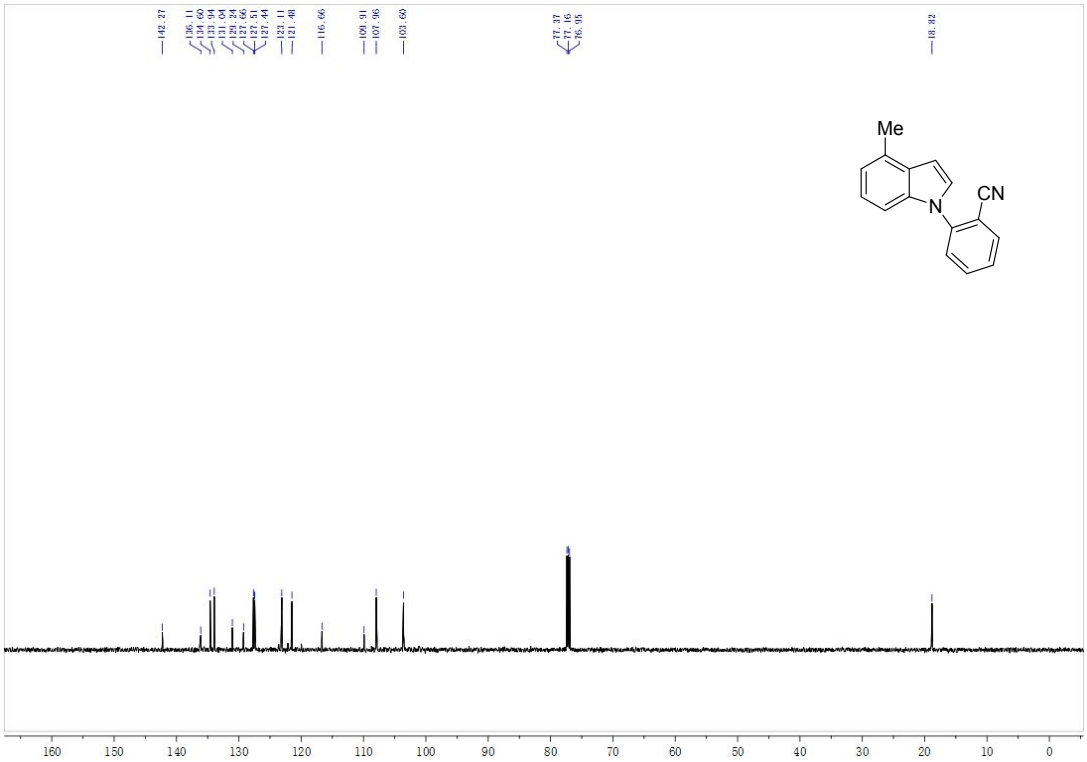


Figure S9. ^{13}C{^{1}H} NMR for compound **1c**

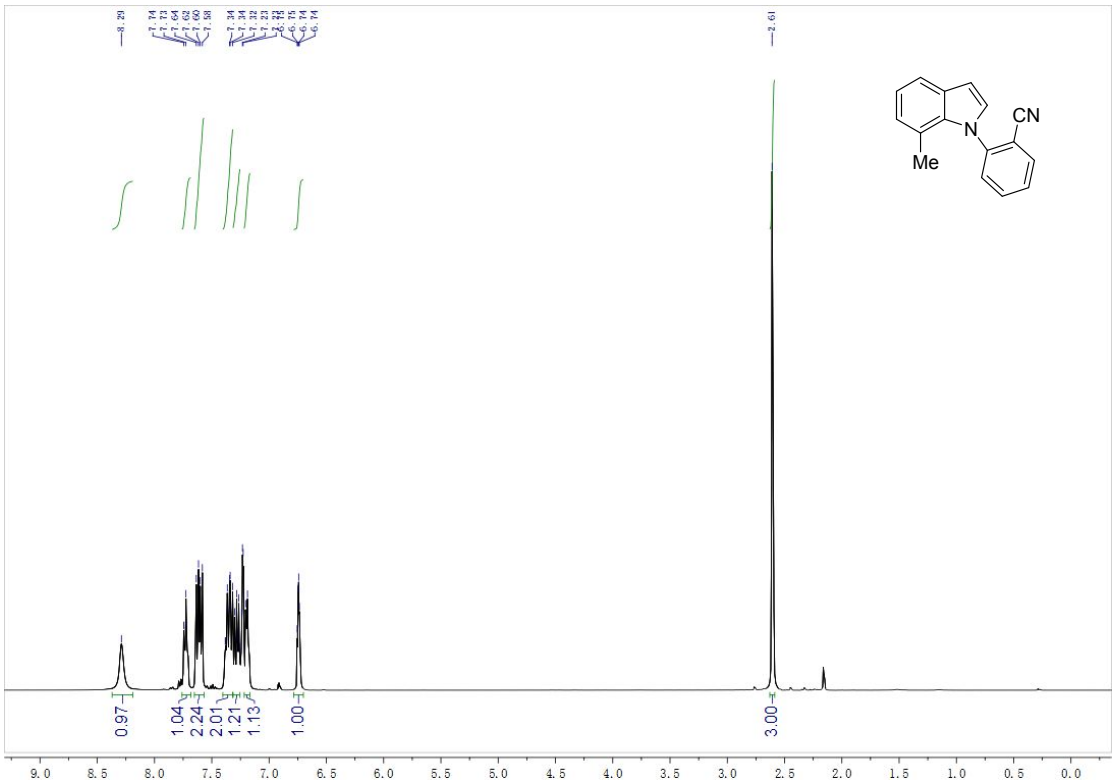


Figure S10. ^{1}H NMR for compound **1d**

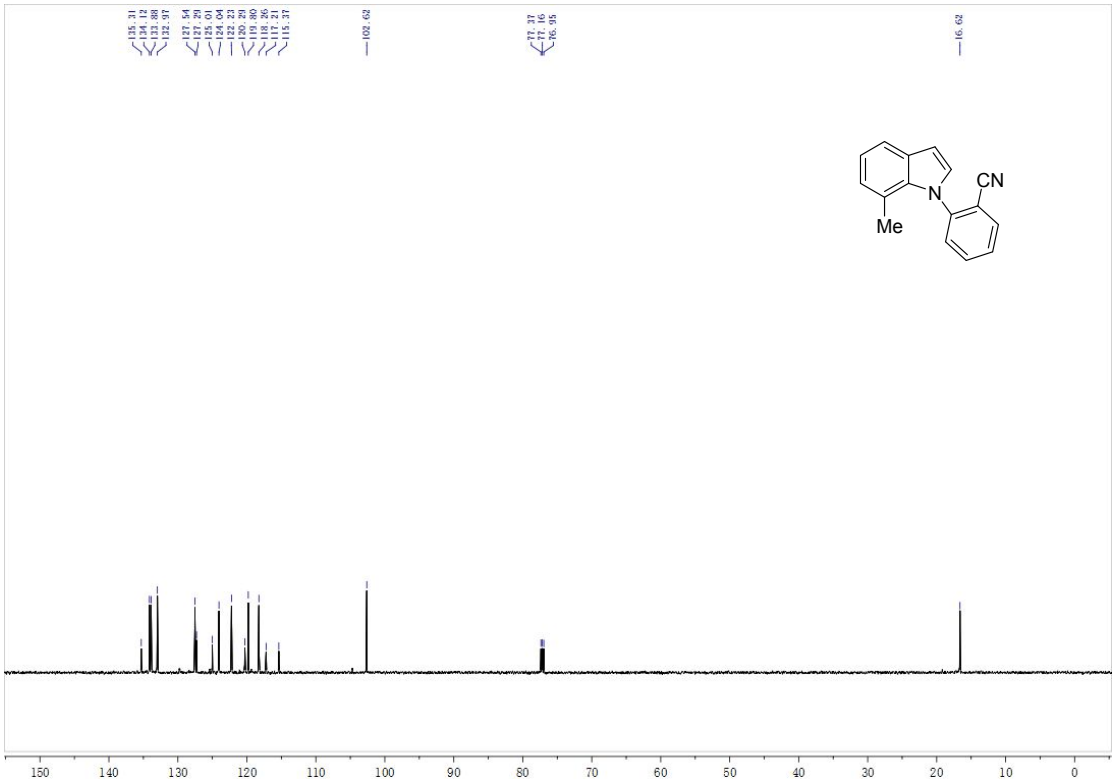


Figure S11. $^{13}C\{^{1}H\}$ NMR for compound **1d**

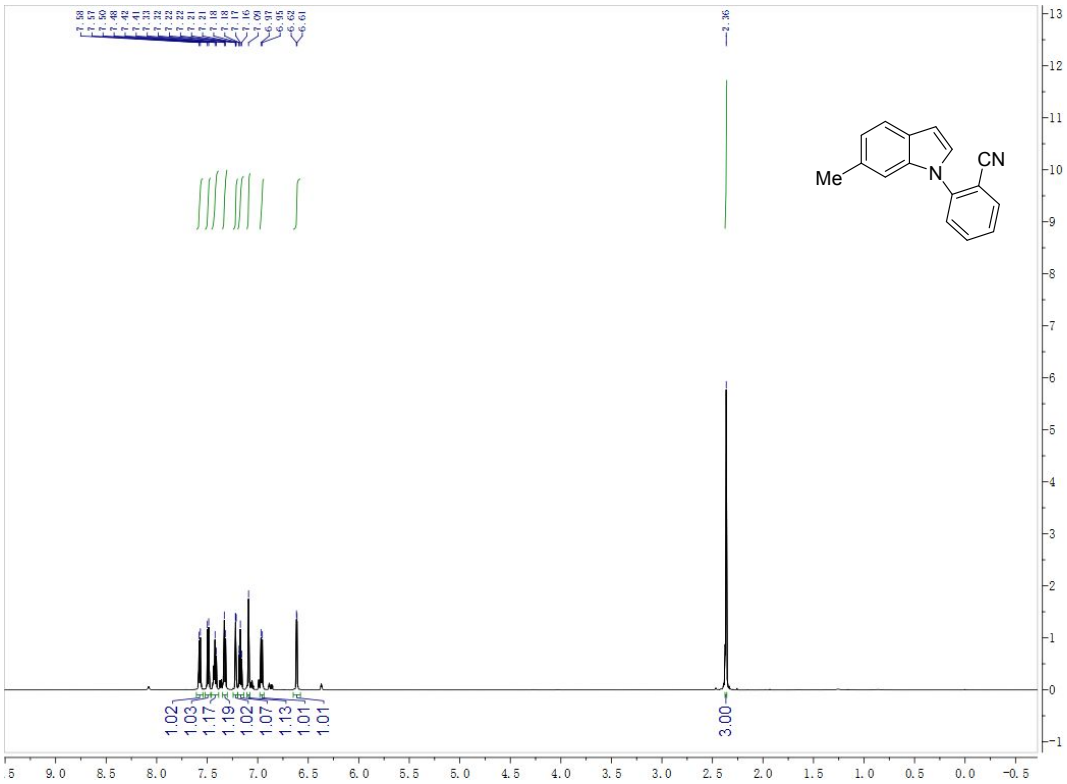


Figure S12. ^{1}H NMR for compound **1e**

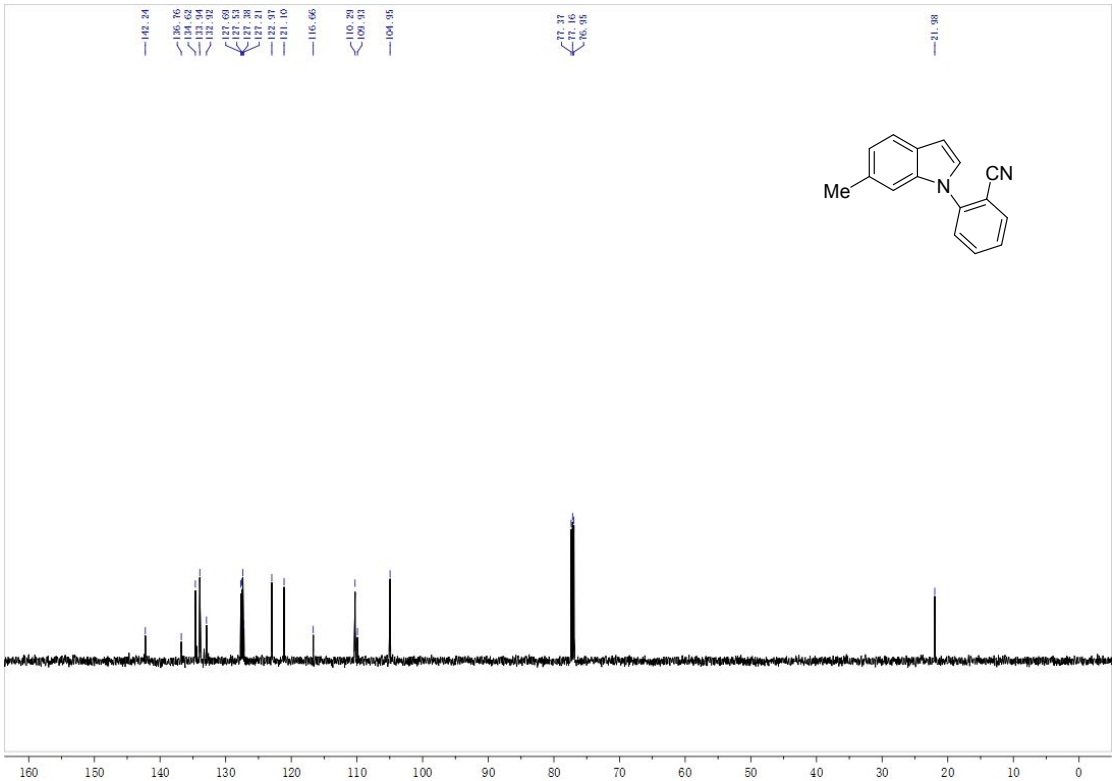


Figure S13. $^{13}C\{^{1}H\}$ NMR for compound **1e**

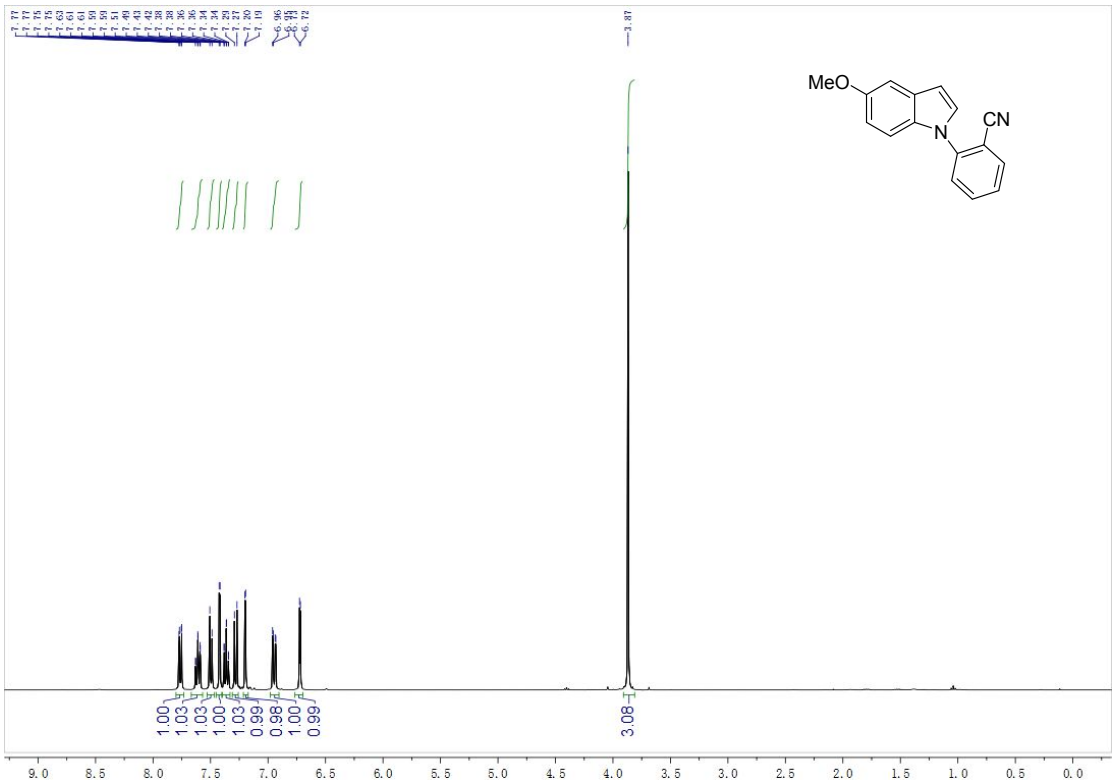


Figure S14. ^{1}H NMR for compound **1f**

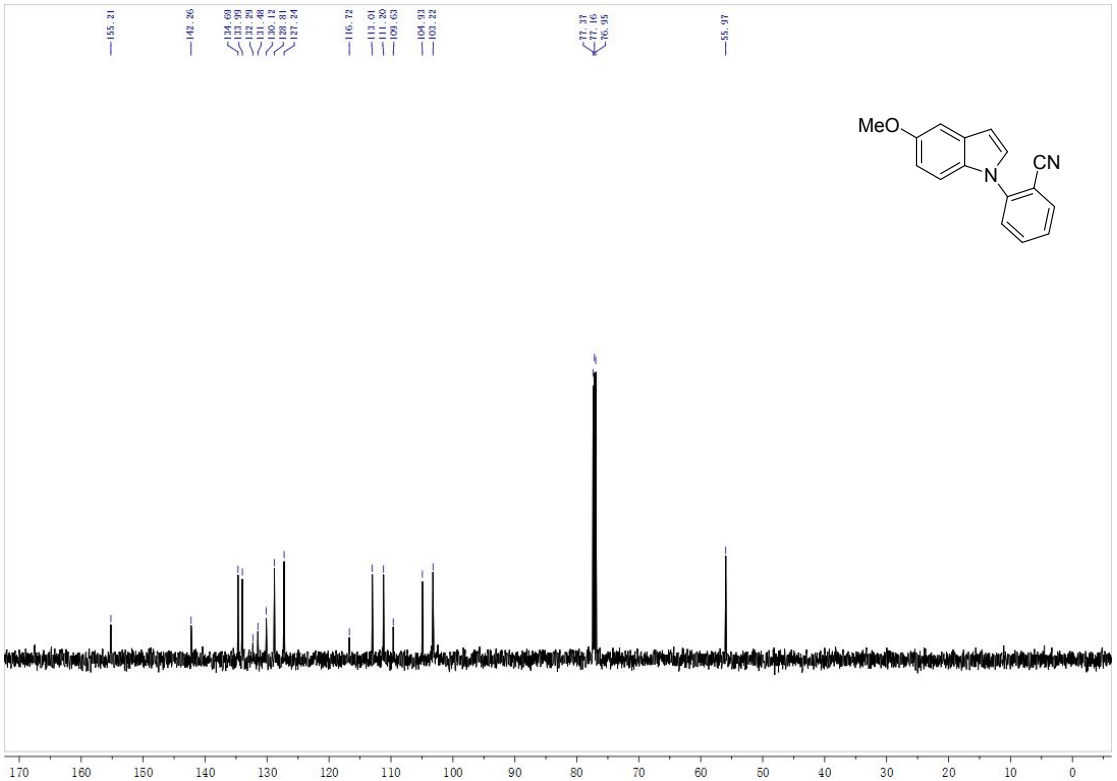


Figure S15. ^{13}C{^{1}H} NMR for compound **1f**

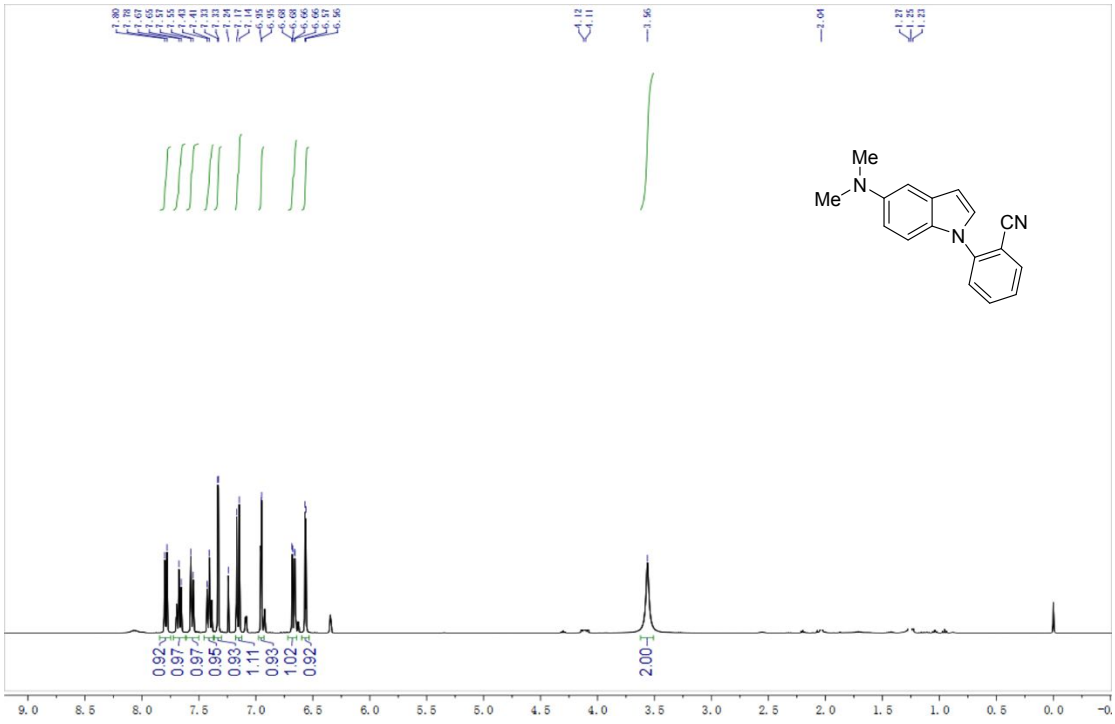


Figure S16. ^{1}H NMR for compound **1g**

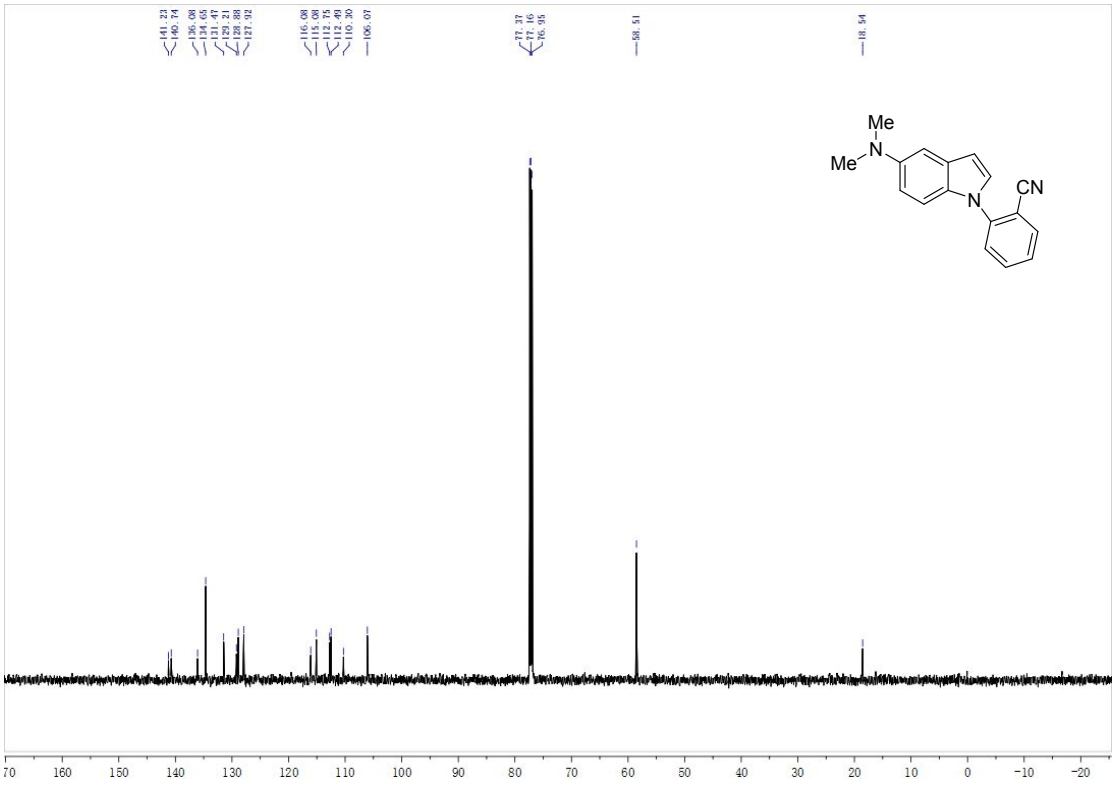


Figure S17. $^{13}C\{^{1}H\}$ NMR for compound **1g**

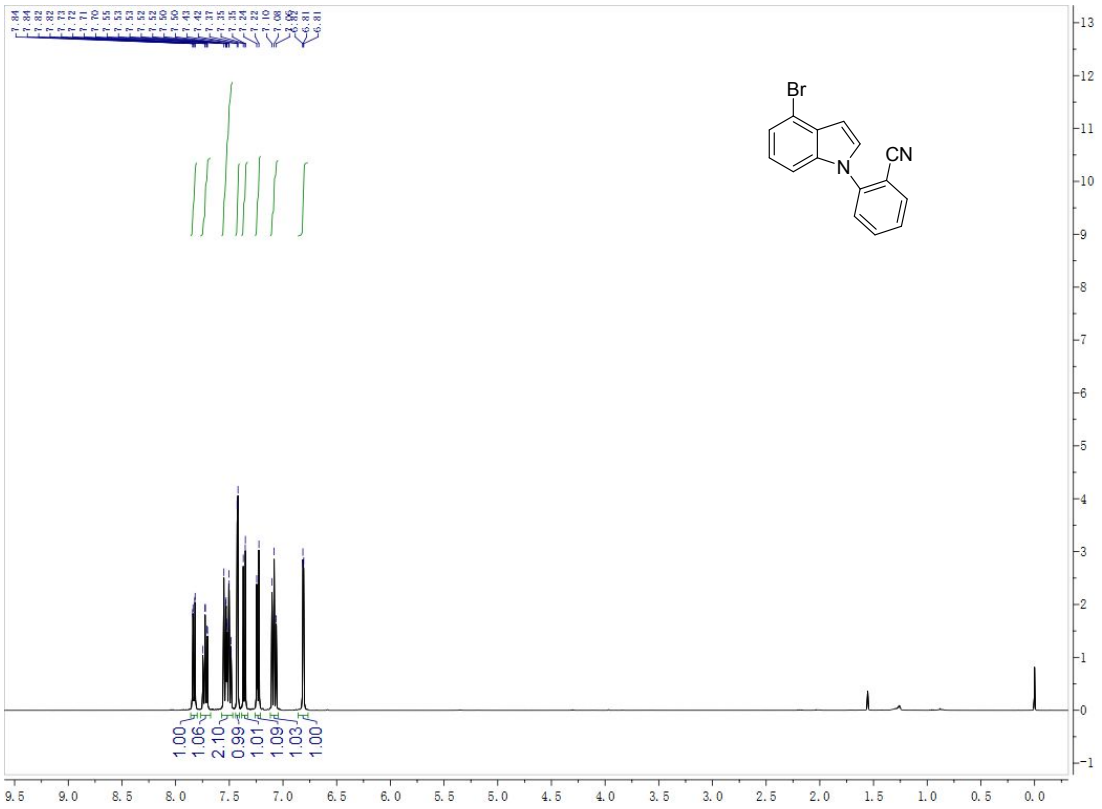


Figure S18. ^{1}H NMR for compound **1h**

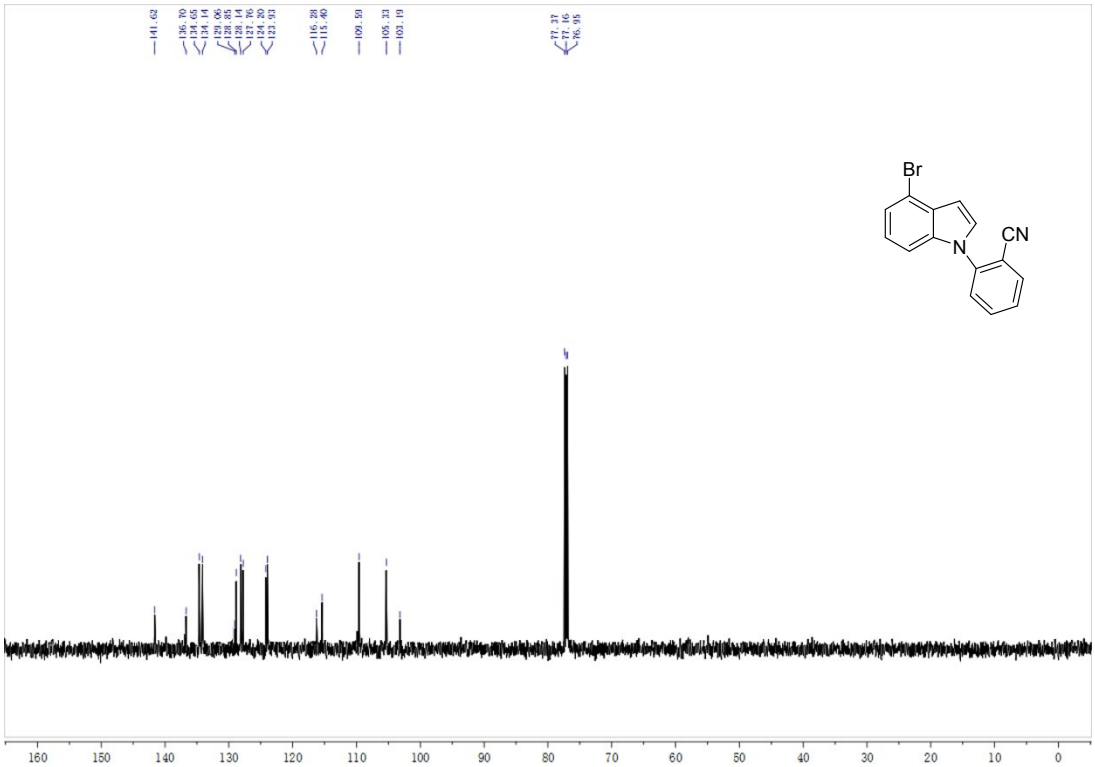


Figure S19. $^{13}C\{^{1}H\}$ NMR for compound **1h**

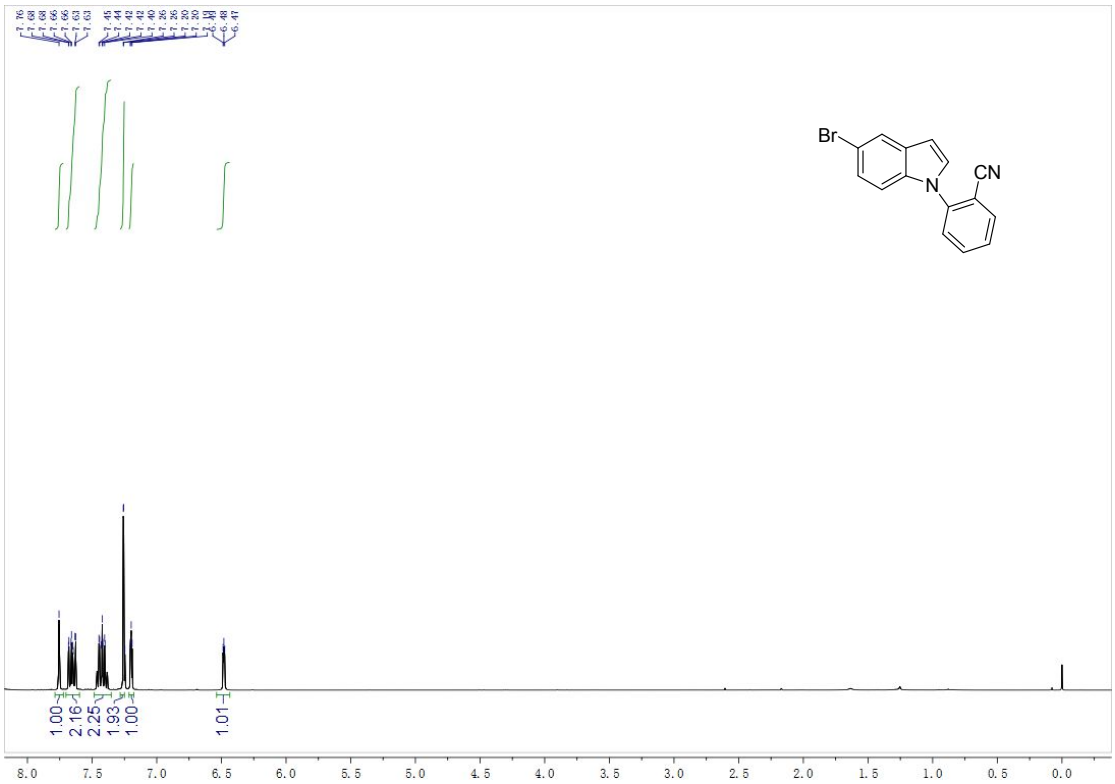


Figure S20. ^{1}H NMR for compound **1i**

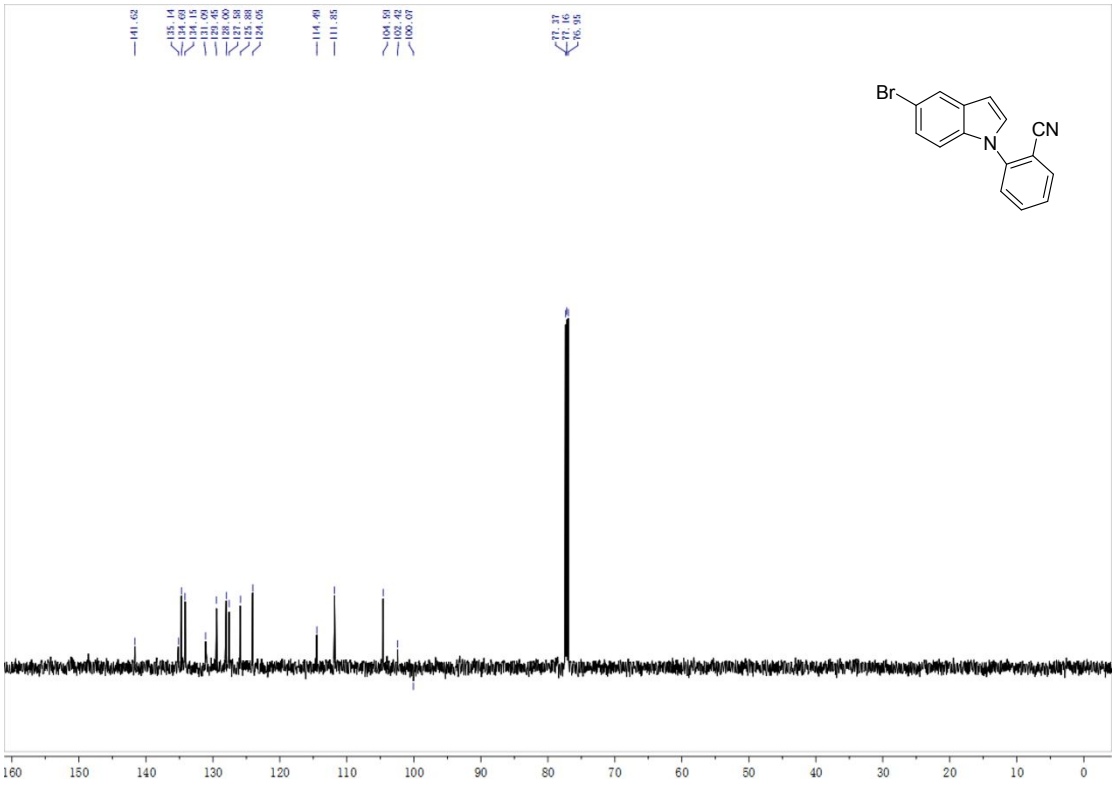


Figure S21. $^{13}C\{^{1}H\}$ NMR for compound **1i**

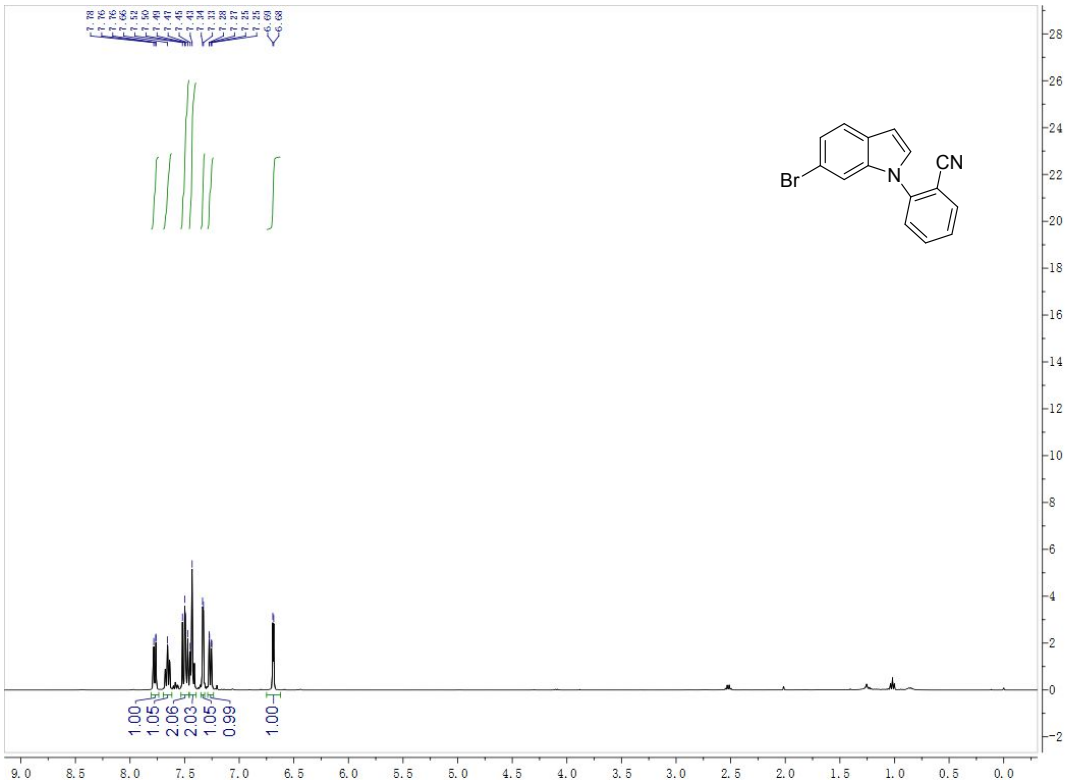


Figure S22. ^{1}H NMR for compound **1j**

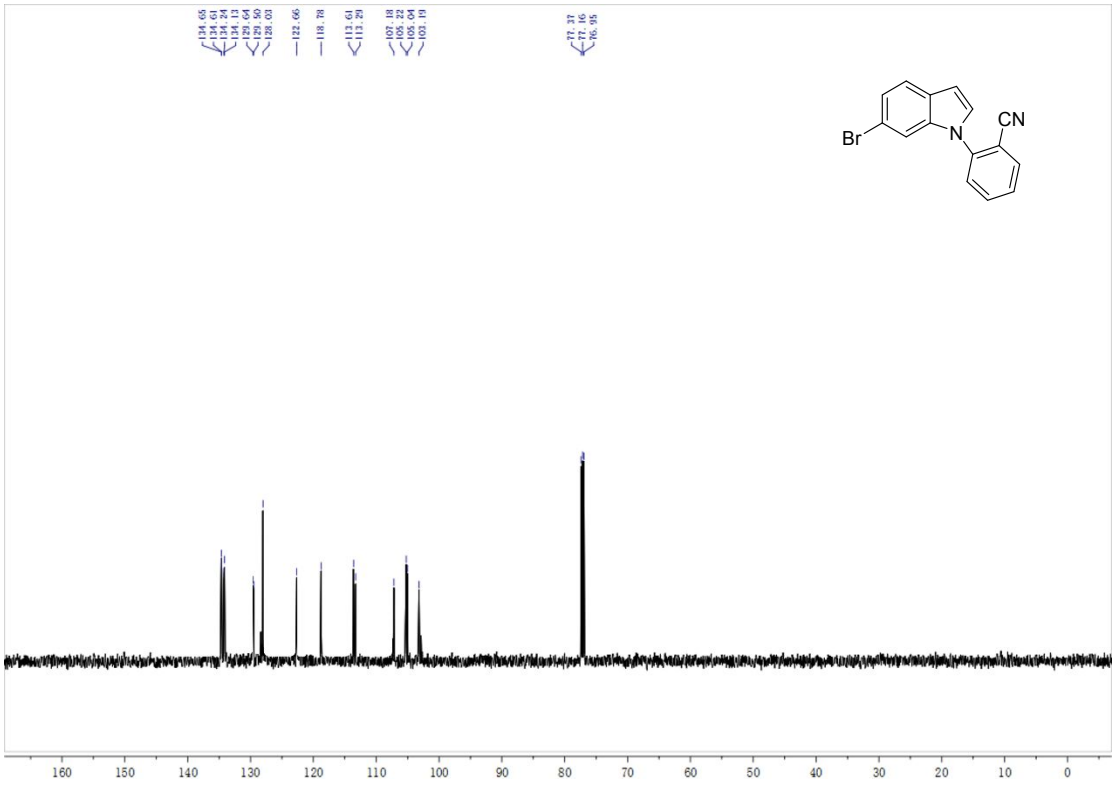


Figure S23. $^{13}C\{^{1}H\}$ NMR for compound **1j**

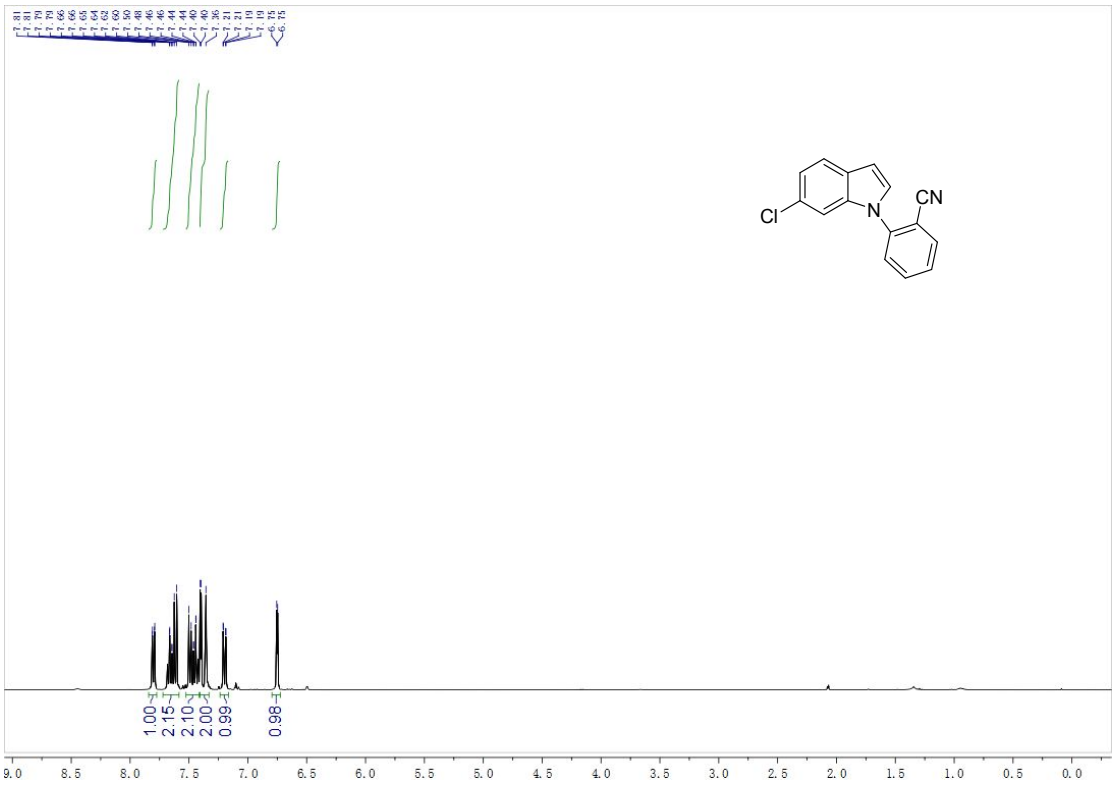


Figure S24. ^{1}H NMR for compound **1k**

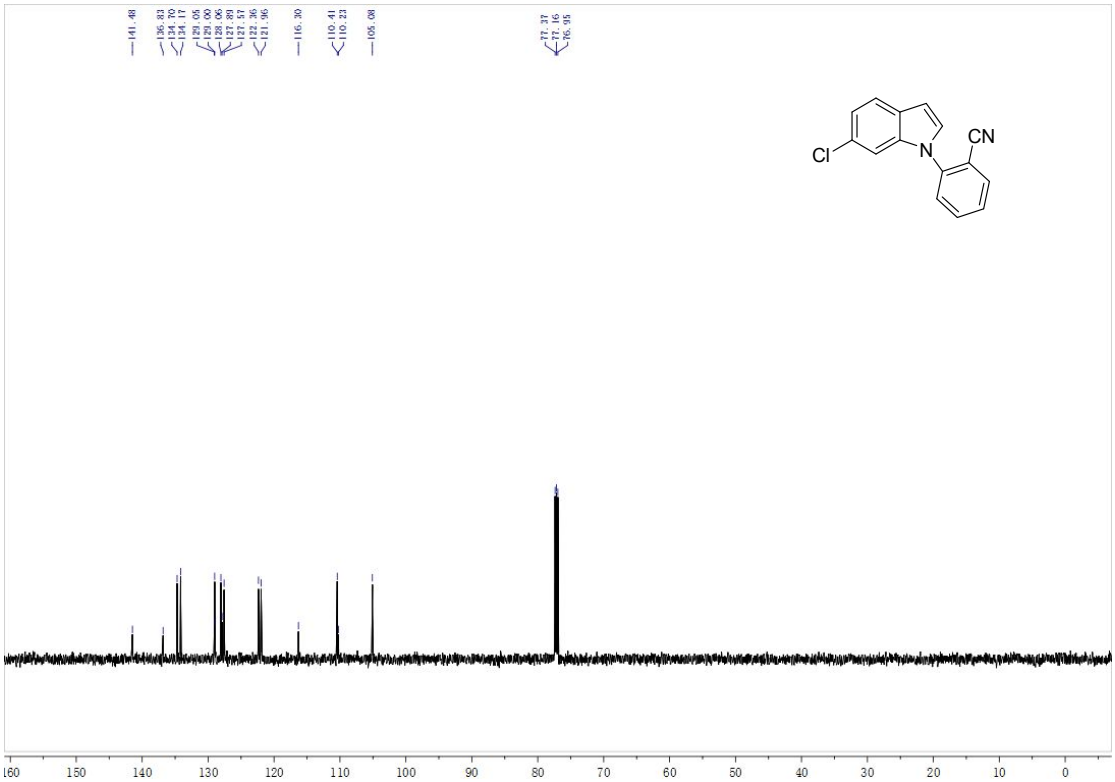


Figure S25. $^{13}C\{^{1}H\}$ NMR for compound **1k**

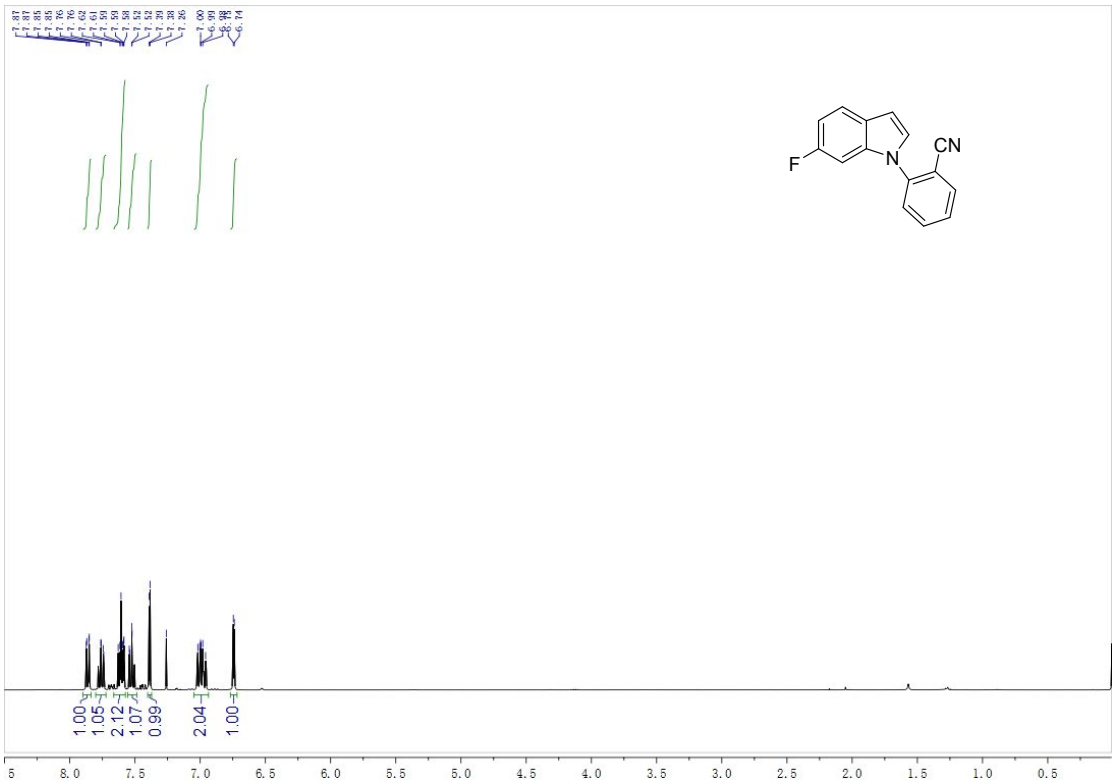


Figure S26. ^{1}H NMR for compound **1l**

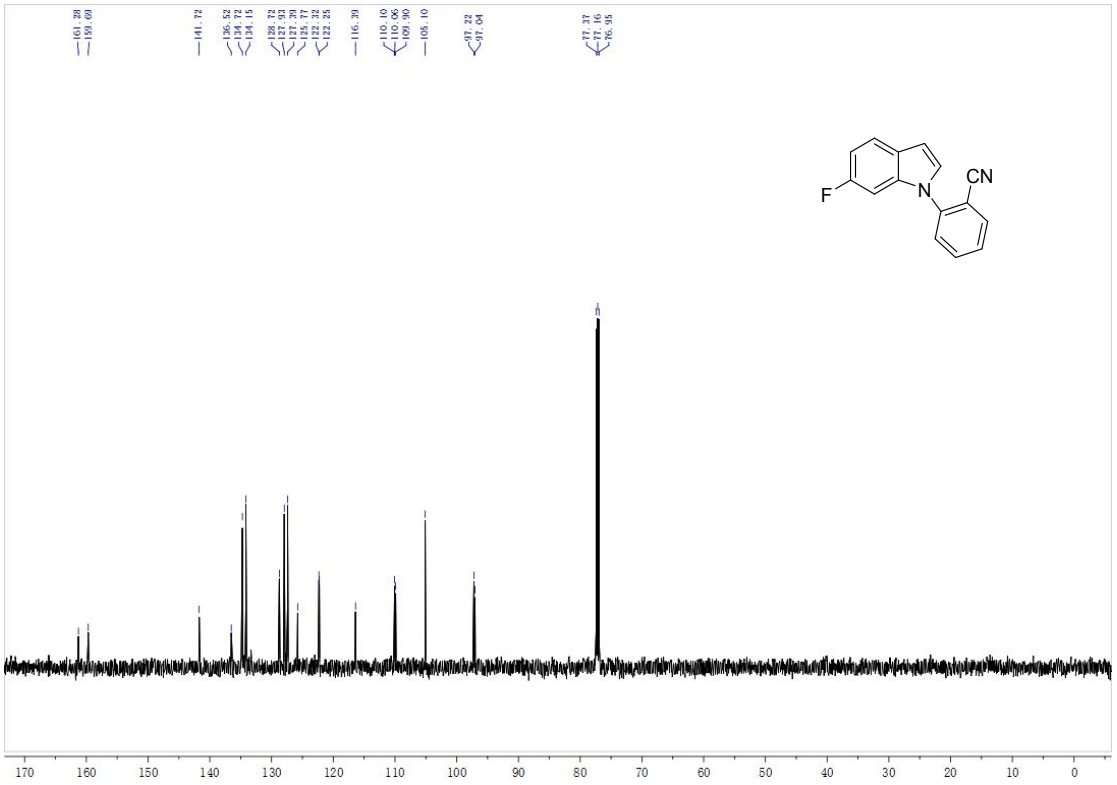


Figure S27. $^{13}C\{^{1}H\}$ NMR for compound **1l**

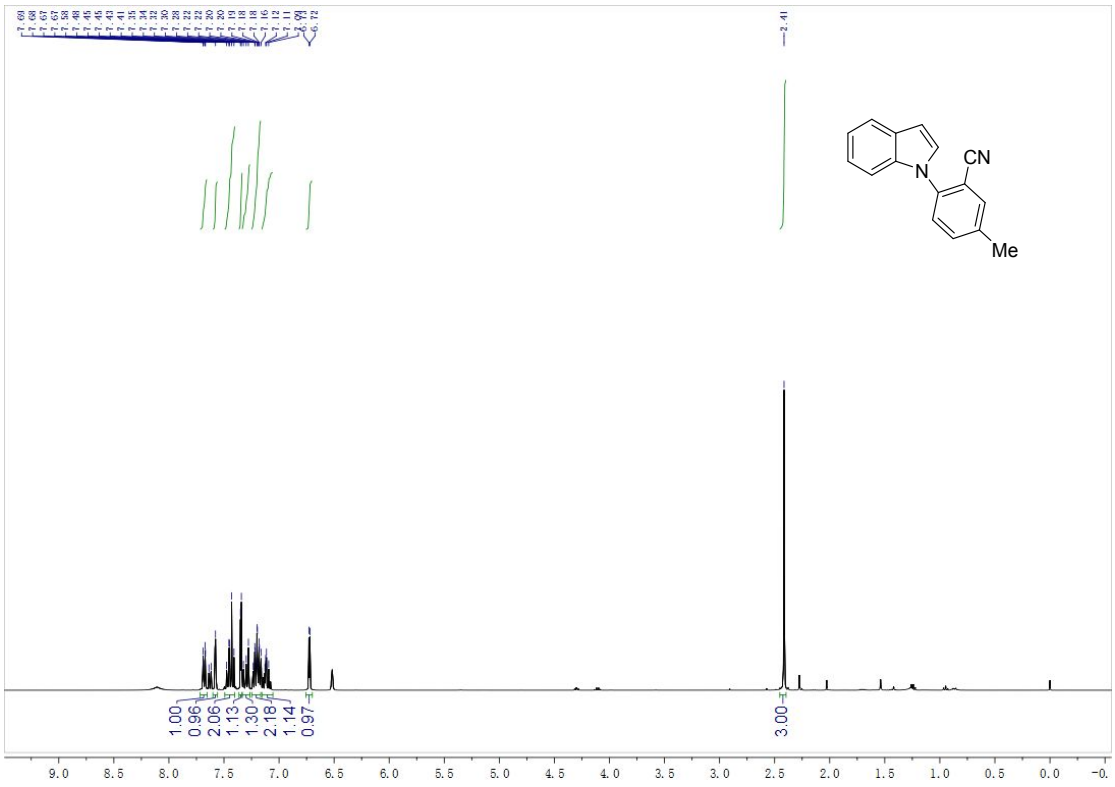


Figure S28. ^{1}H NMR for compound **1m**

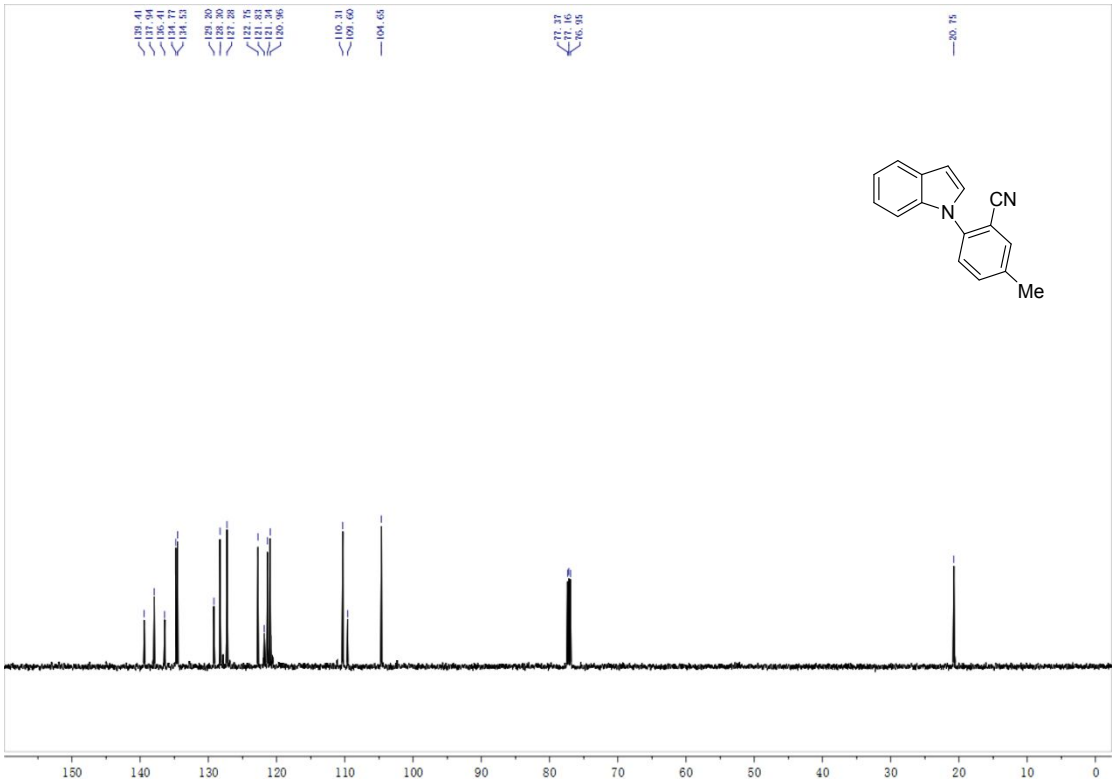


Figure S29. $^{13}C\{^{1}H\}$ NMR for compound **1m**

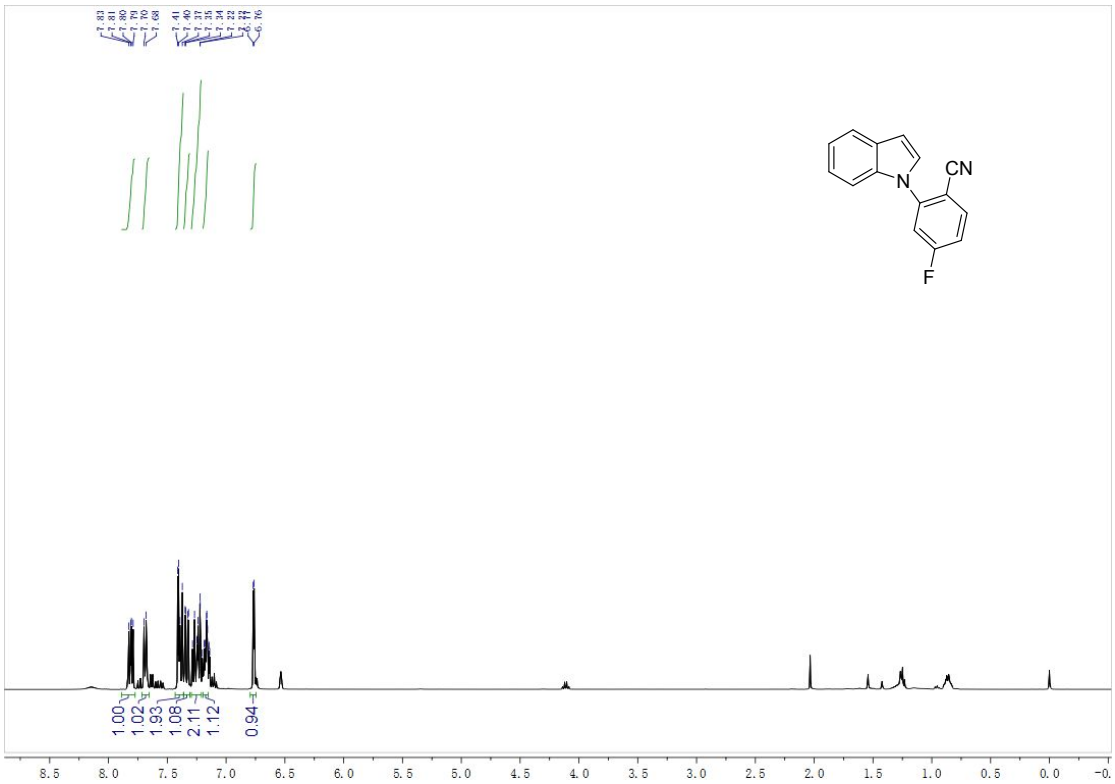


Figure S30. ^{1}H NMR for compound **1n**

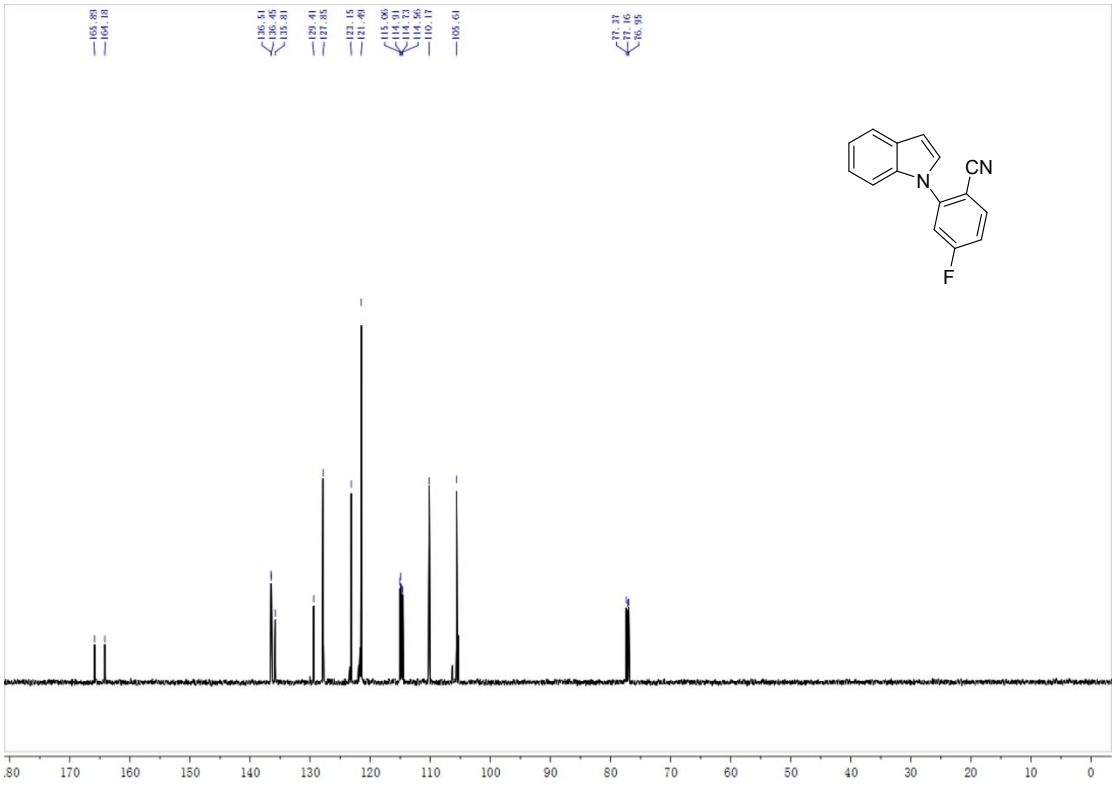


Figure S31. $^{13}C\{^{1}H\}$ NMR for compound **1n**

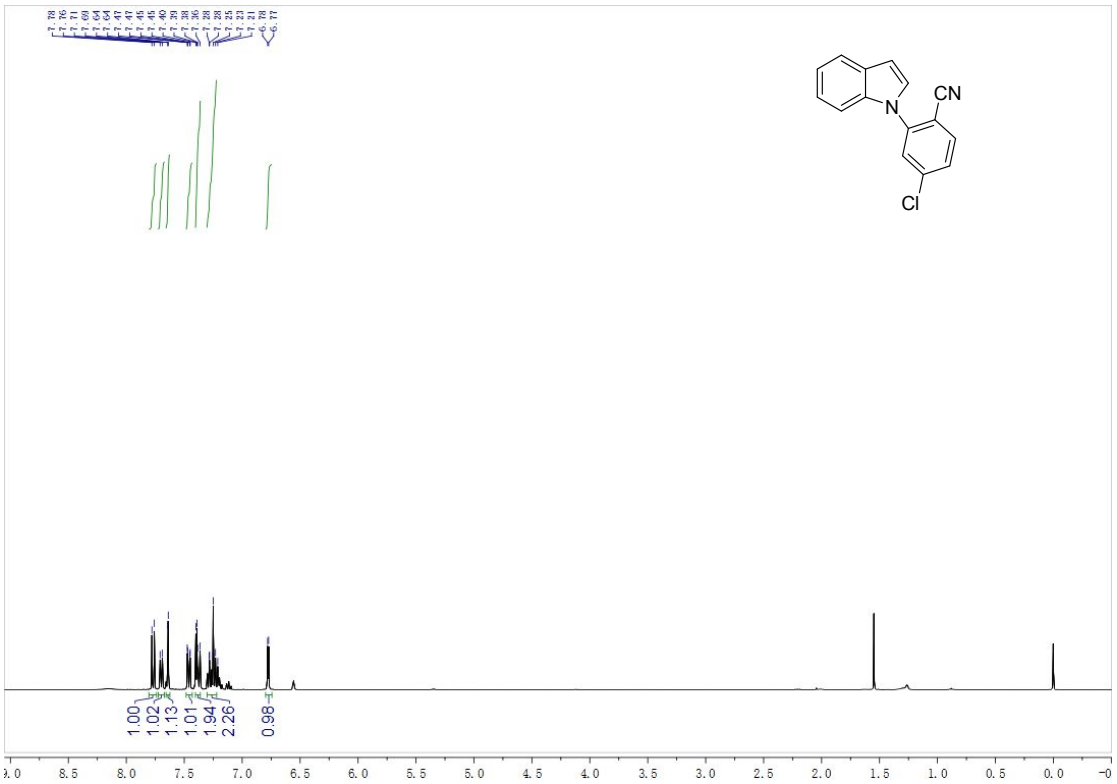


Figure S32. ^{1}H NMR for compound **1o**

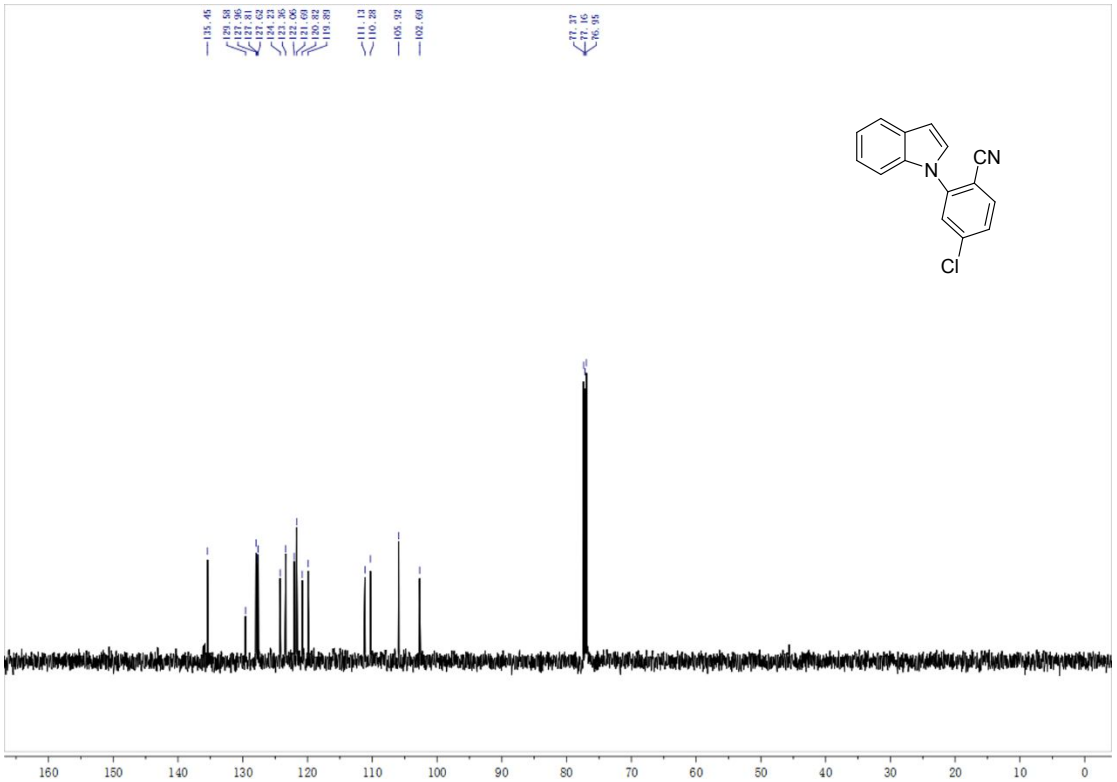


Figure S33. $^{13}C\{^{1}H\}$ NMR for compound **1o**

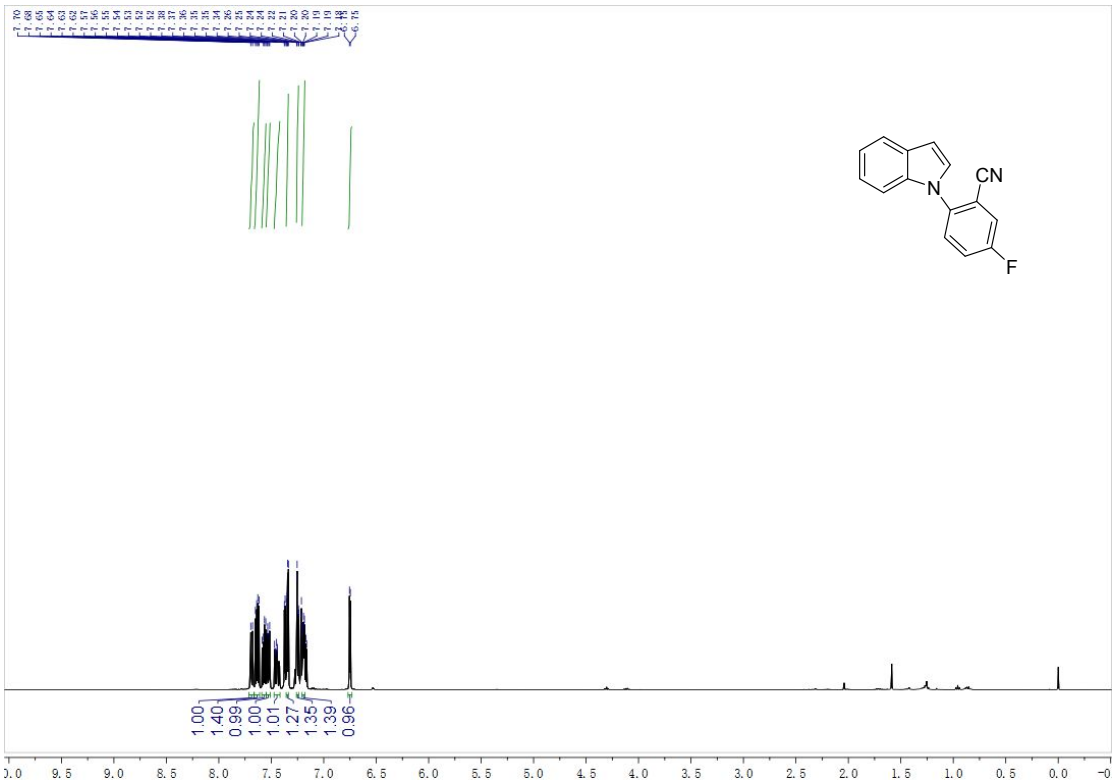


Figure S34. ^{1}H NMR for compound **1p**

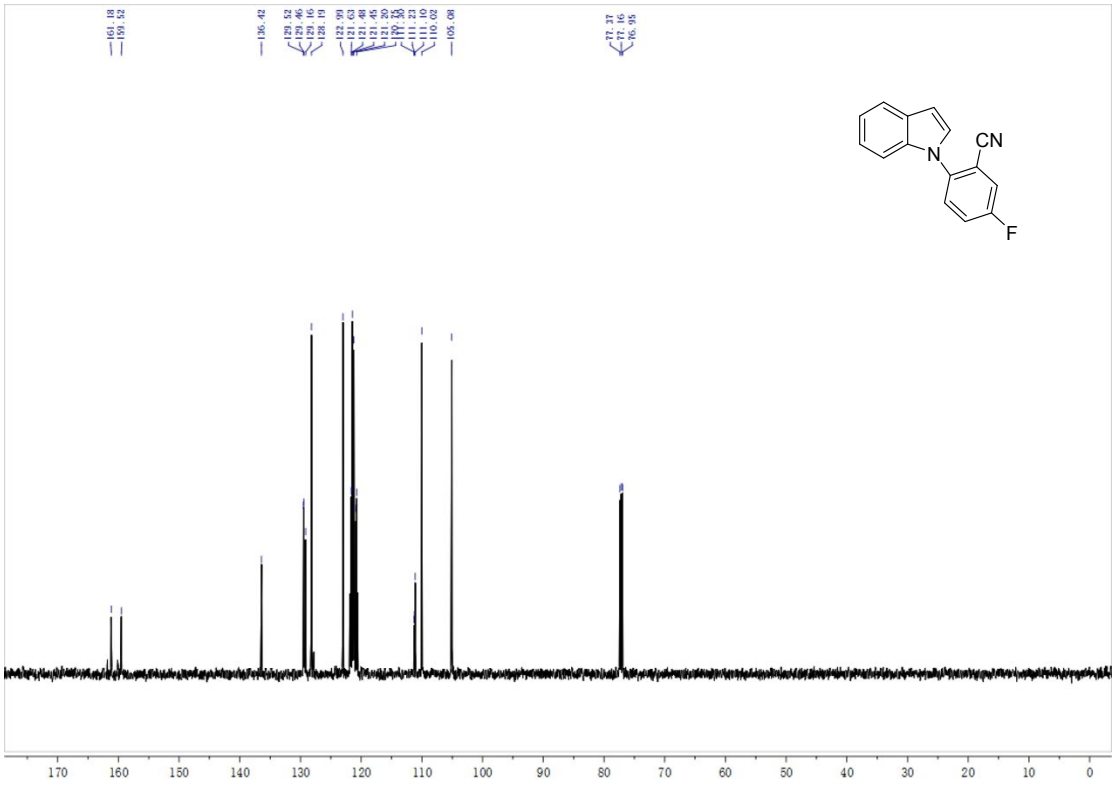


Figure S35. $^{13}C\{^{1}H\}$ NMR for compound **1p**

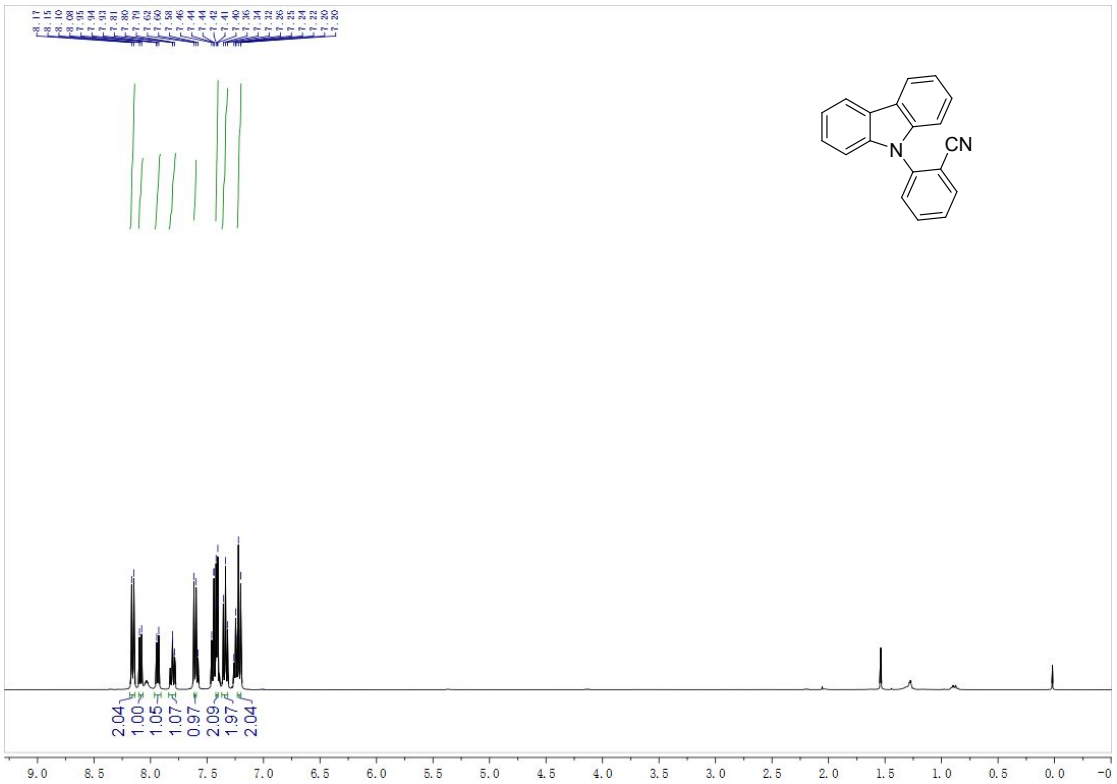


Figure S36. ^{1}H NMR for compound **1q**

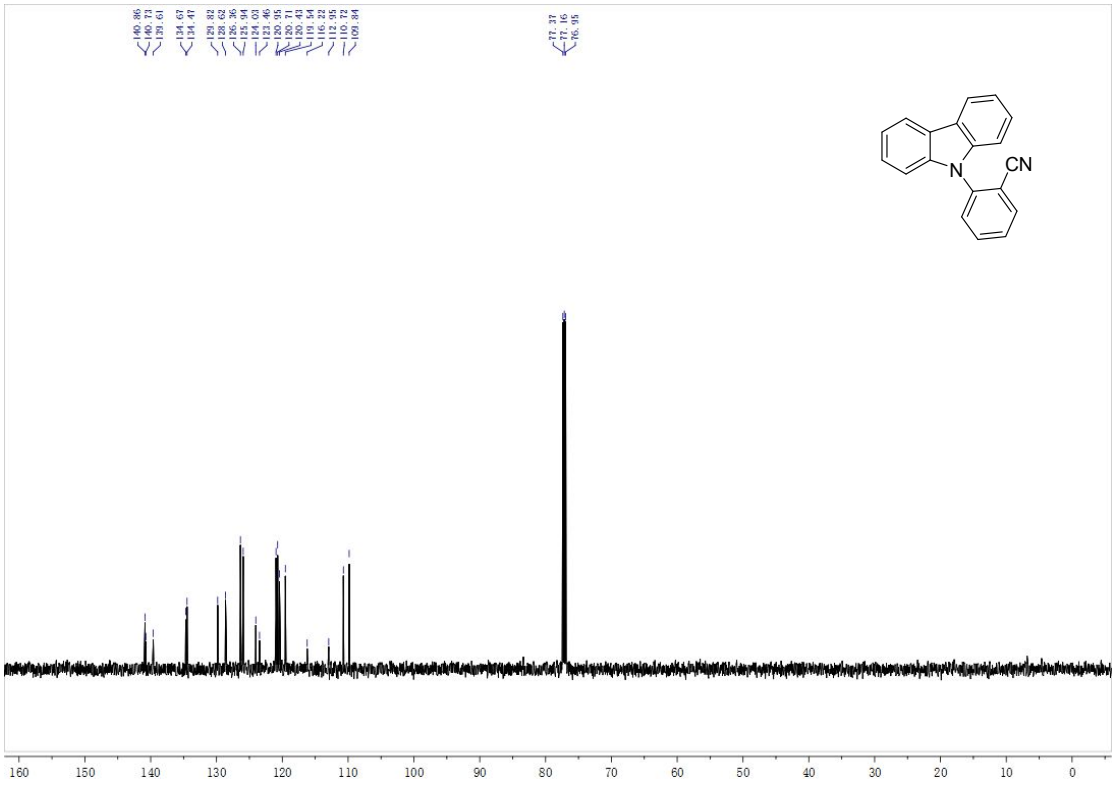


Figure S37. $^{13}C\{^{1}H\}$ NMR for compound **1q**

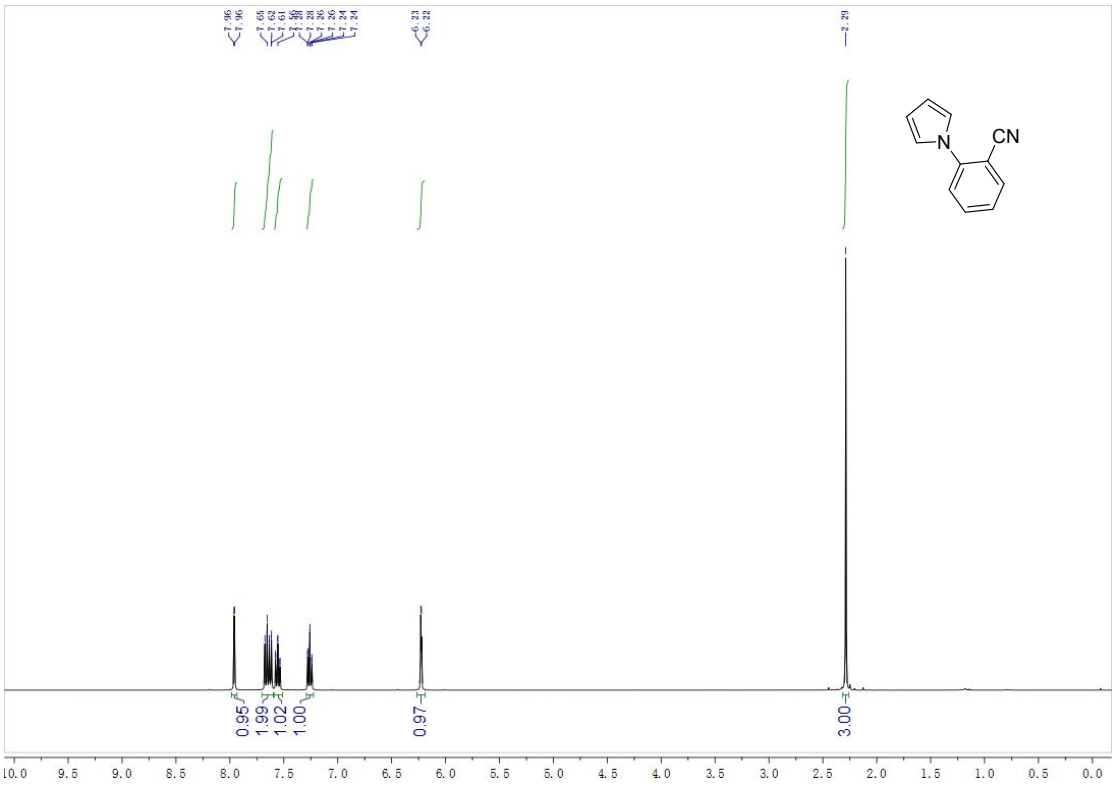


Figure S38. ^{1}H NMR for compound **1r**

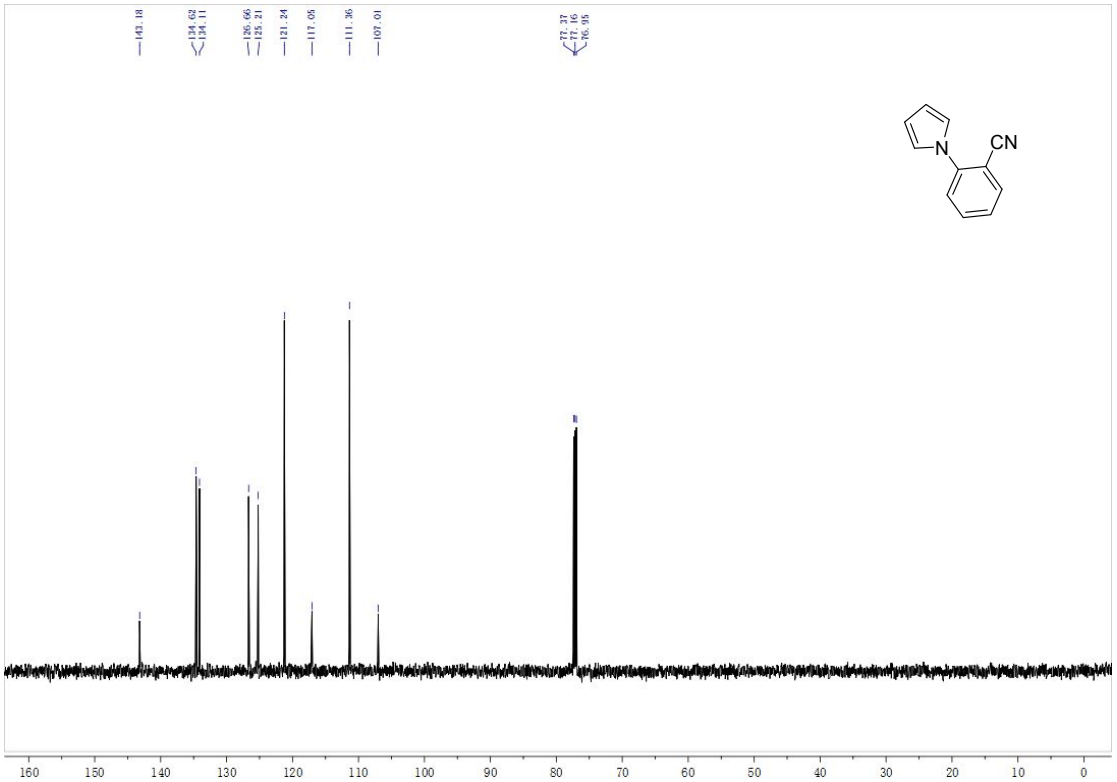


Figure S39. $^{13}C\{^{1}H\}$ NMR for compound **1r**

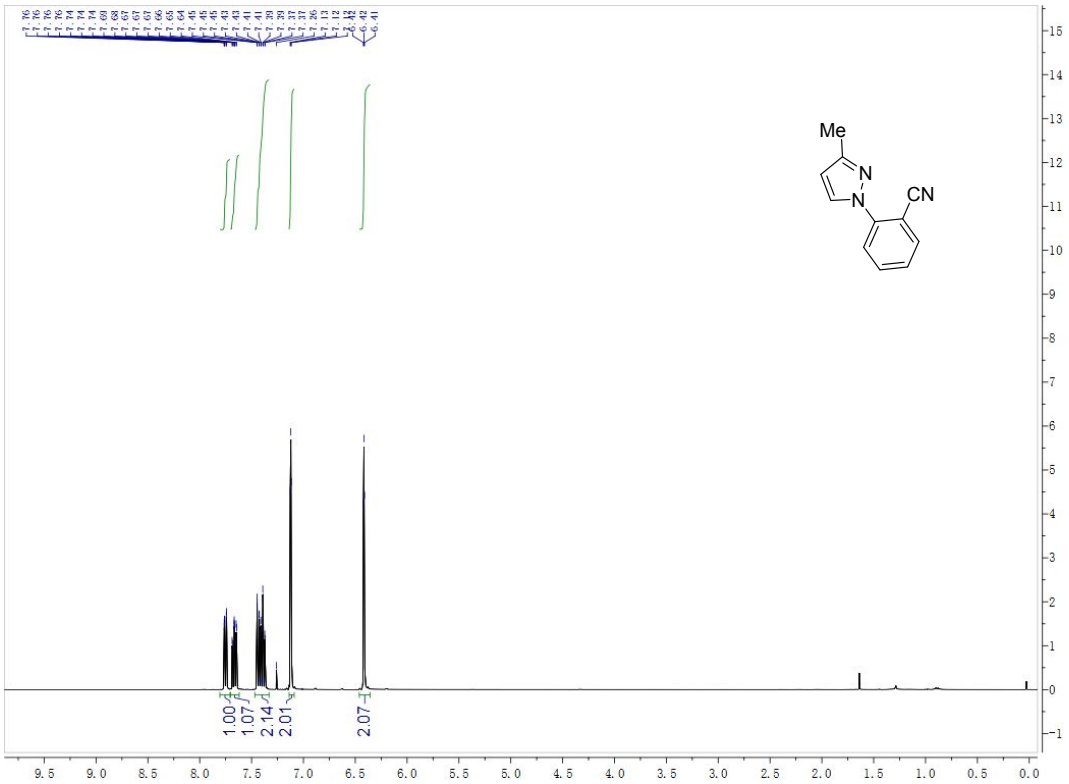


Figure S40. ^{1}H NMR for compound **1s**

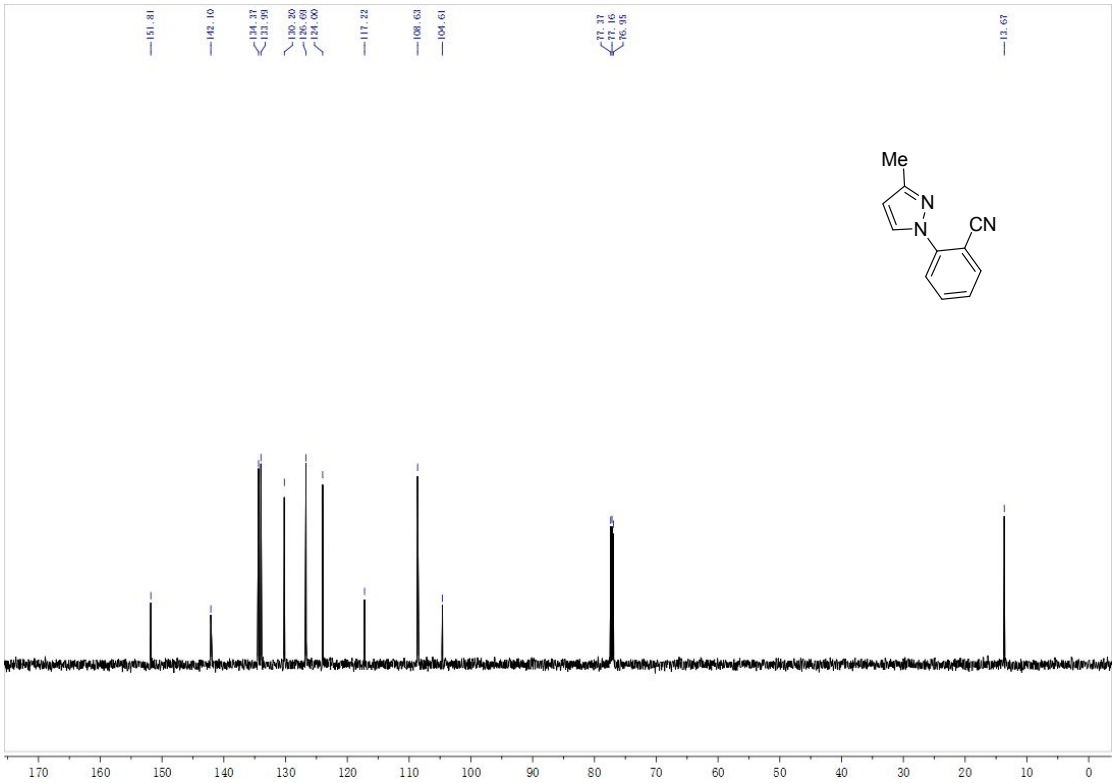


Figure S41. $^{13}C\{^{1}H\}$ NMR for compound **1s**

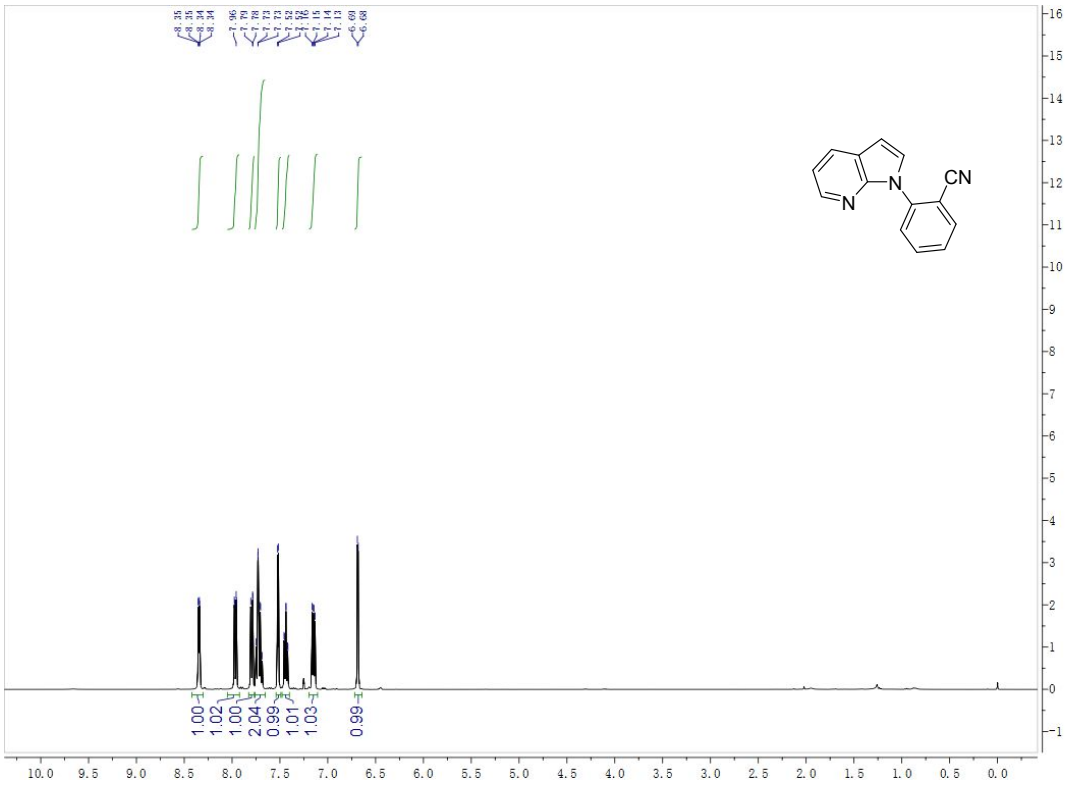


Figure S42. ^{1}H NMR for compound **1t**

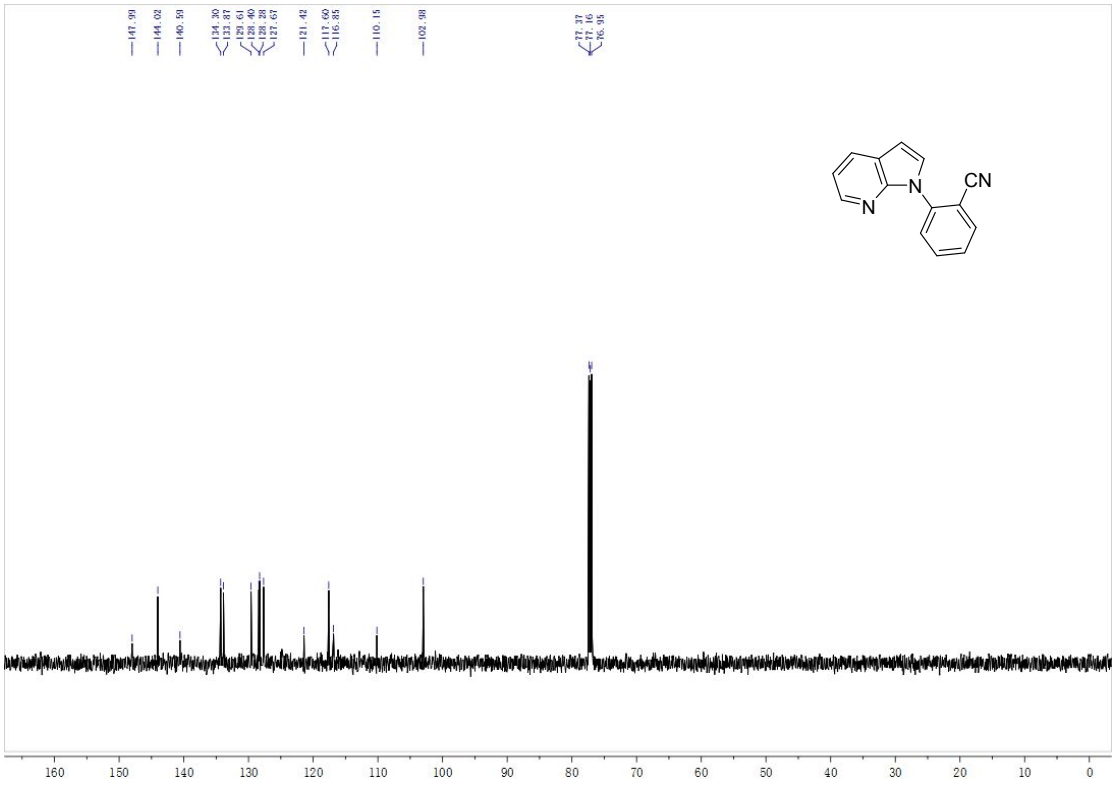


Figure S43. ^{13}C{^{1}H} NMR for compound **1t**

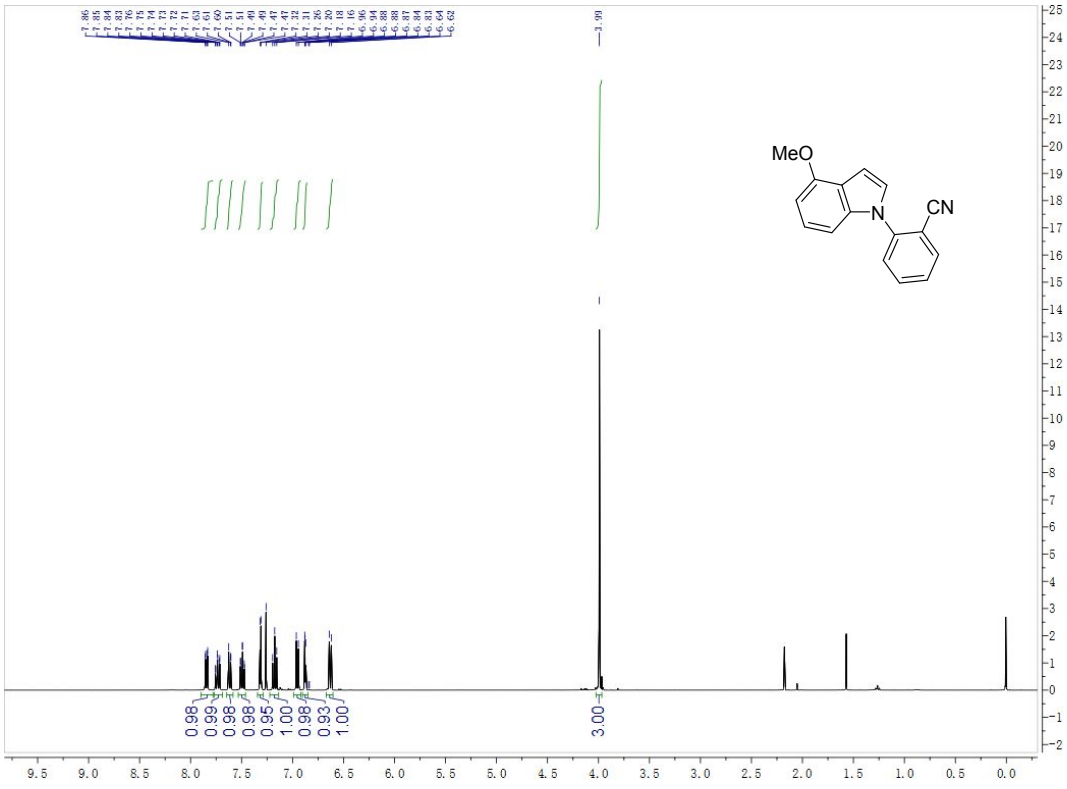


Figure S44. ^{1}H NMR for compound **1u**

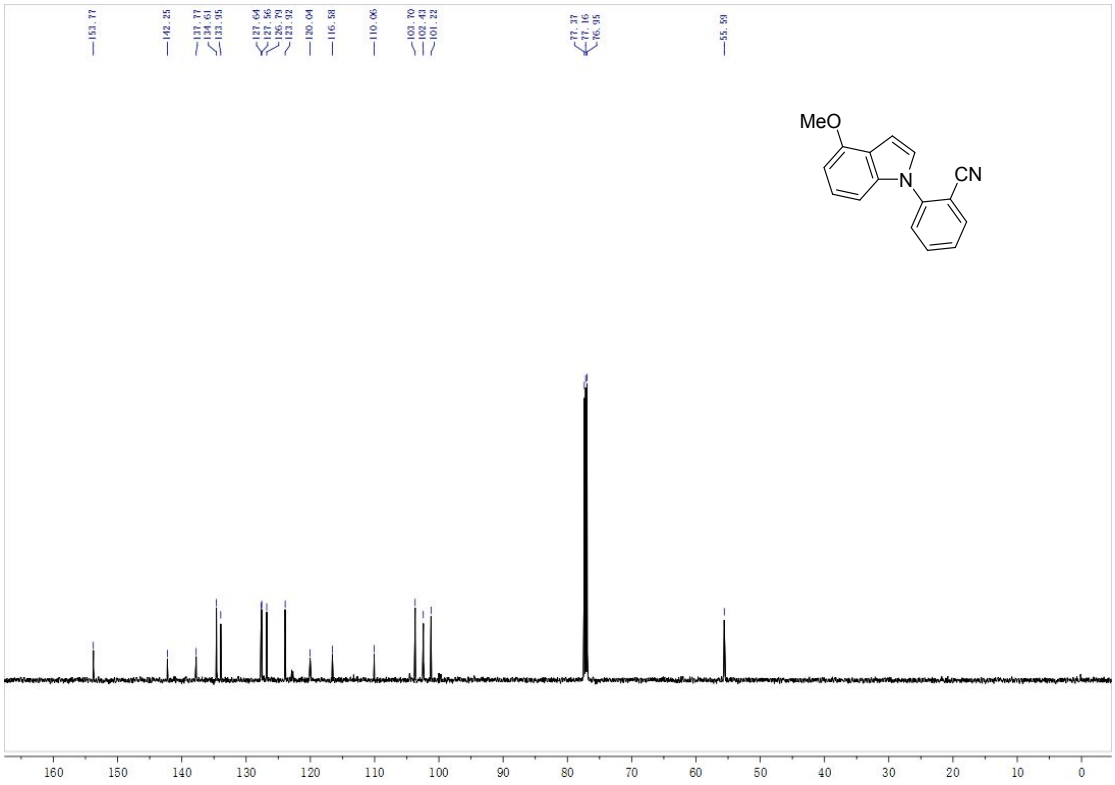


Figure S45. $^{13}C\{^{1}H\}$ NMR for compound **1u**

3-2. Copies of [1] H, [13] C{[1]H}, and [19]F NMR Spectra for products

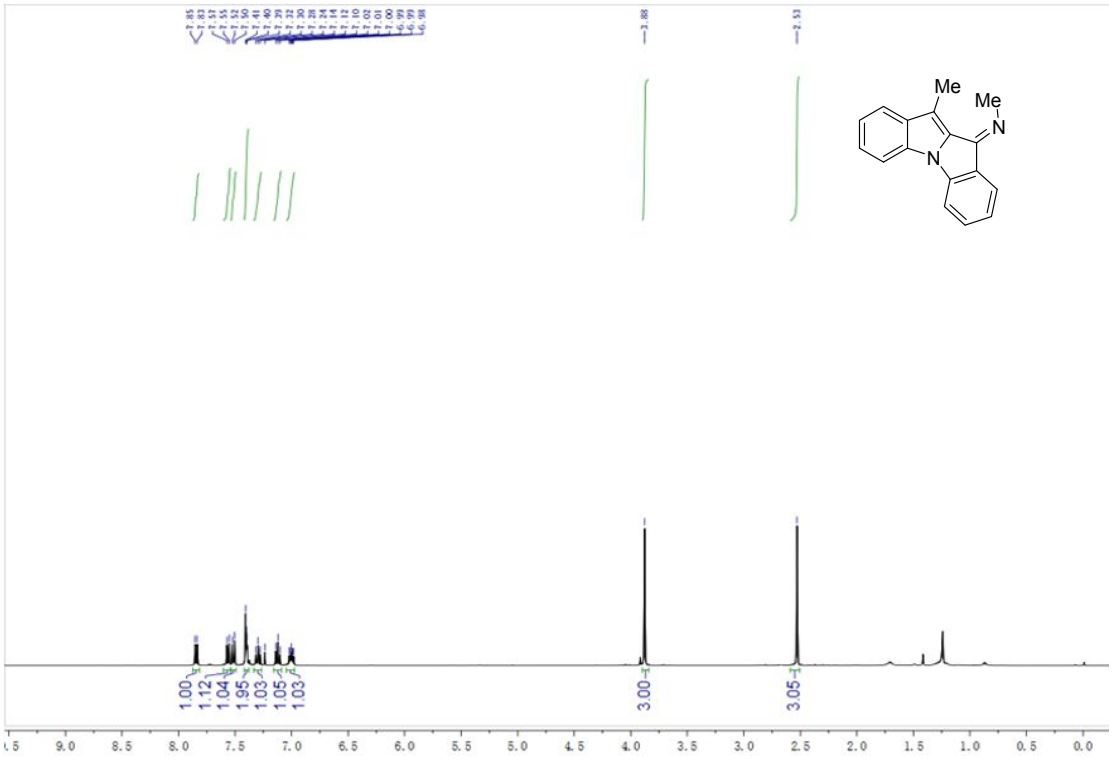


Figure S46. ^{1}H NMR for compound **3a**

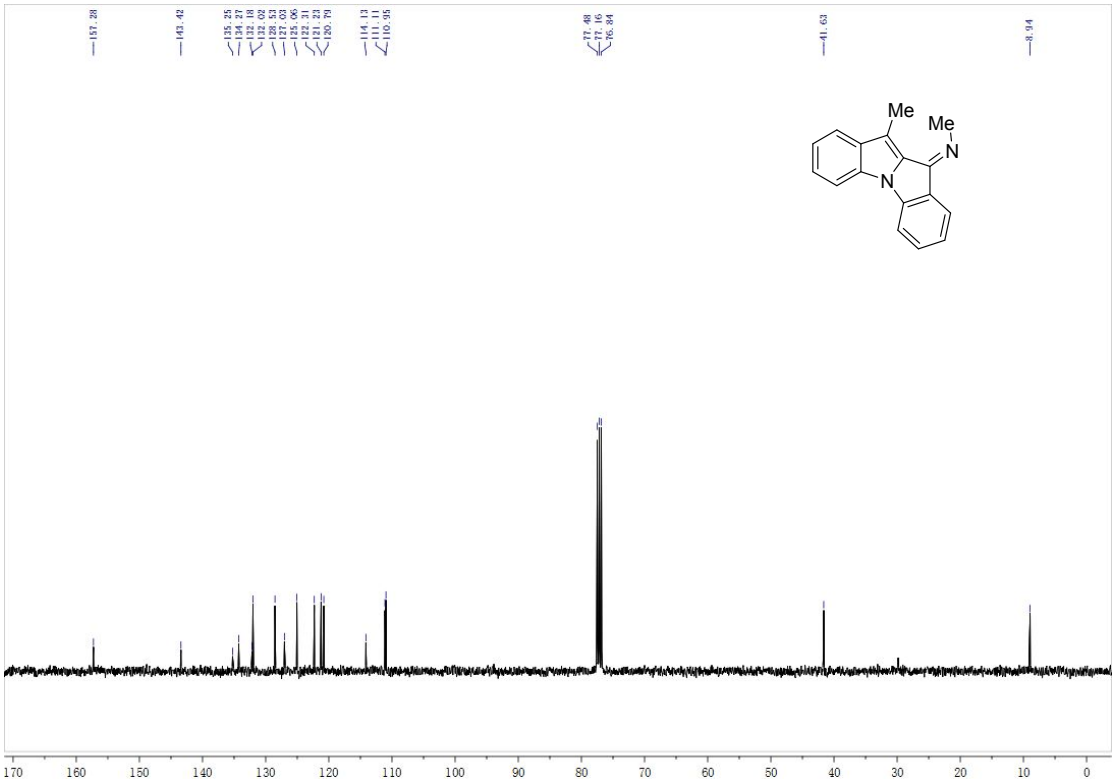


Figure S47. $^{13}C\{^{1}H\}$ NMR for compound **3a**

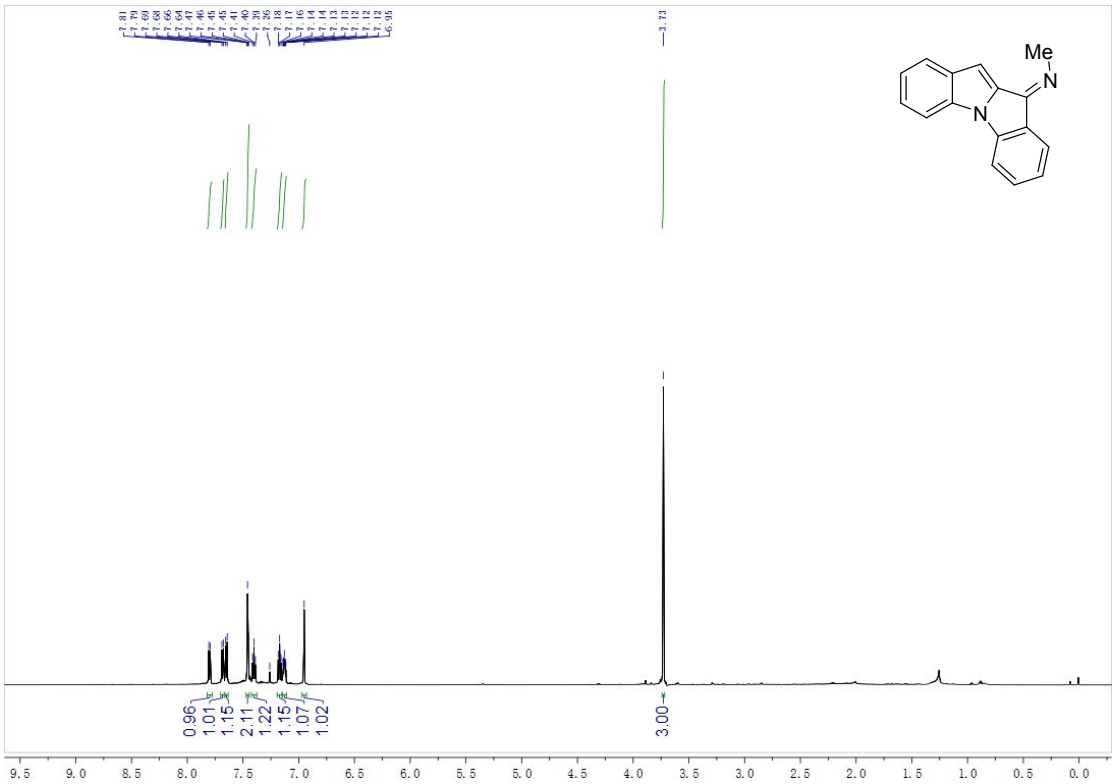


Figure S48. ^{1}H NMR for compound **3b**

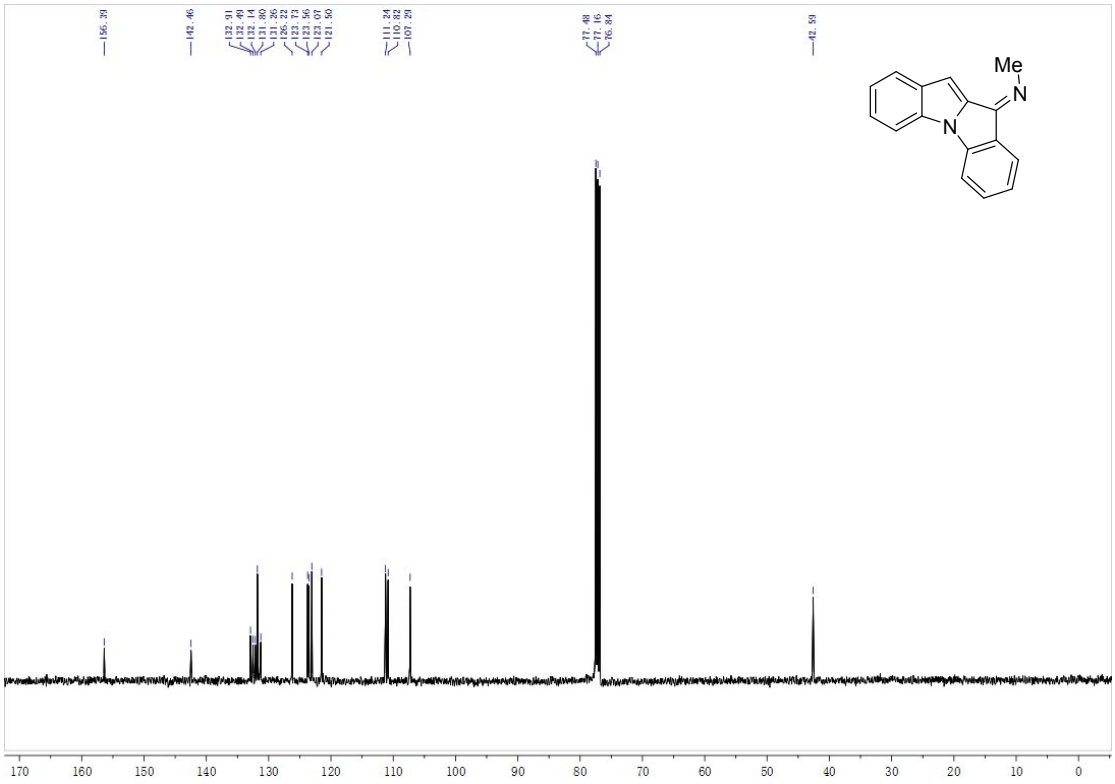


Figure S49. $^{13}C\{^{1}H\}$ NMR for compound **3b**

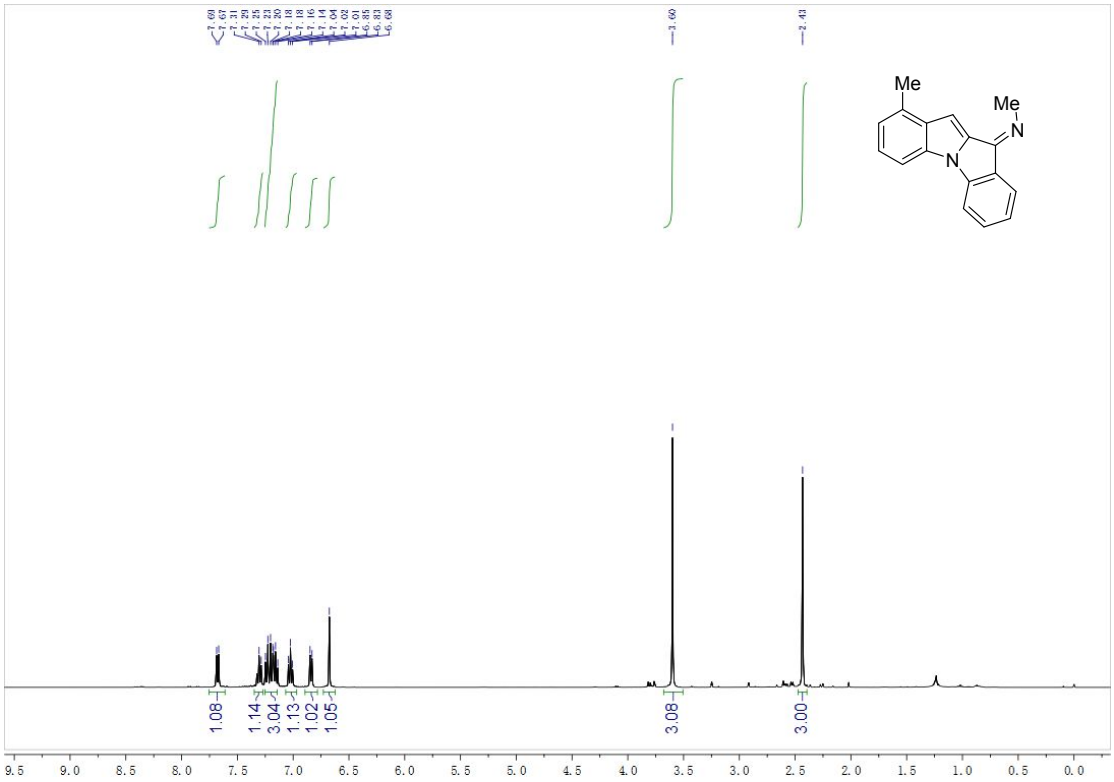


Figure S50. ^{1}H NMR for compound **3c**

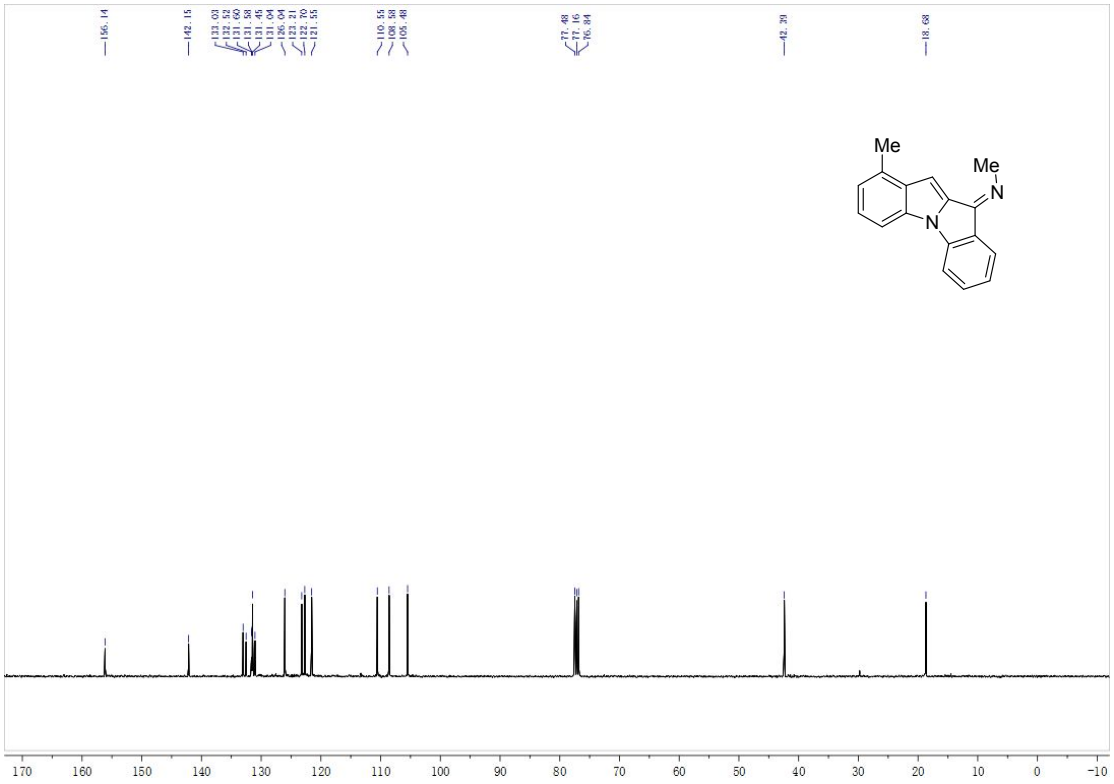


Figure S51. $^{13}C\{^{1}H\}$ NMR for compound **3c**

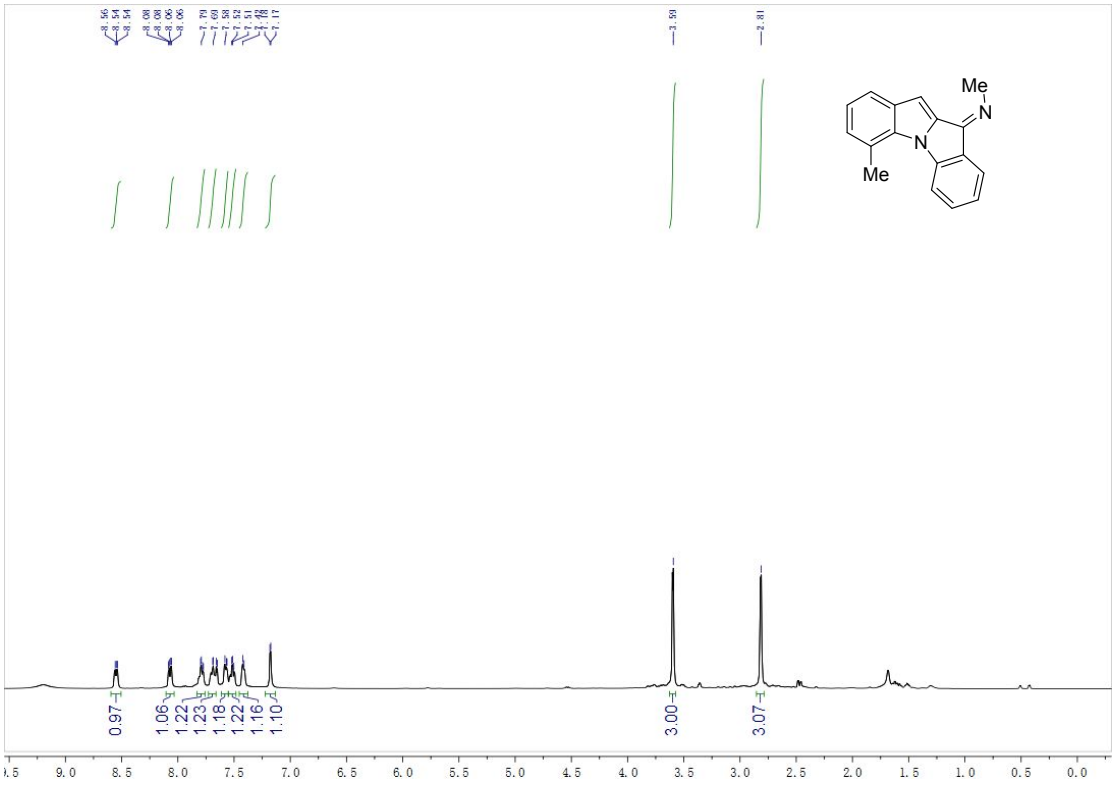


Figure S52. ^{1}H NMR for compound **3d**

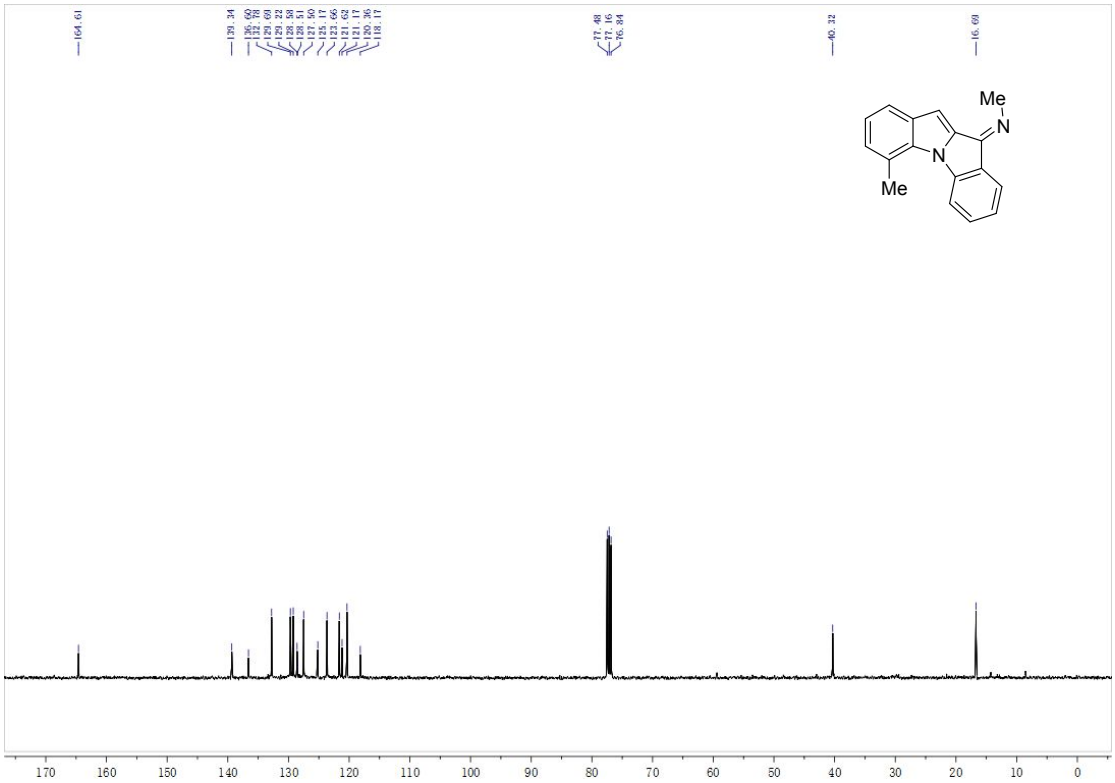


Figure S53. $^{13}C\{^{1}H\}$ NMR for compound **3d**

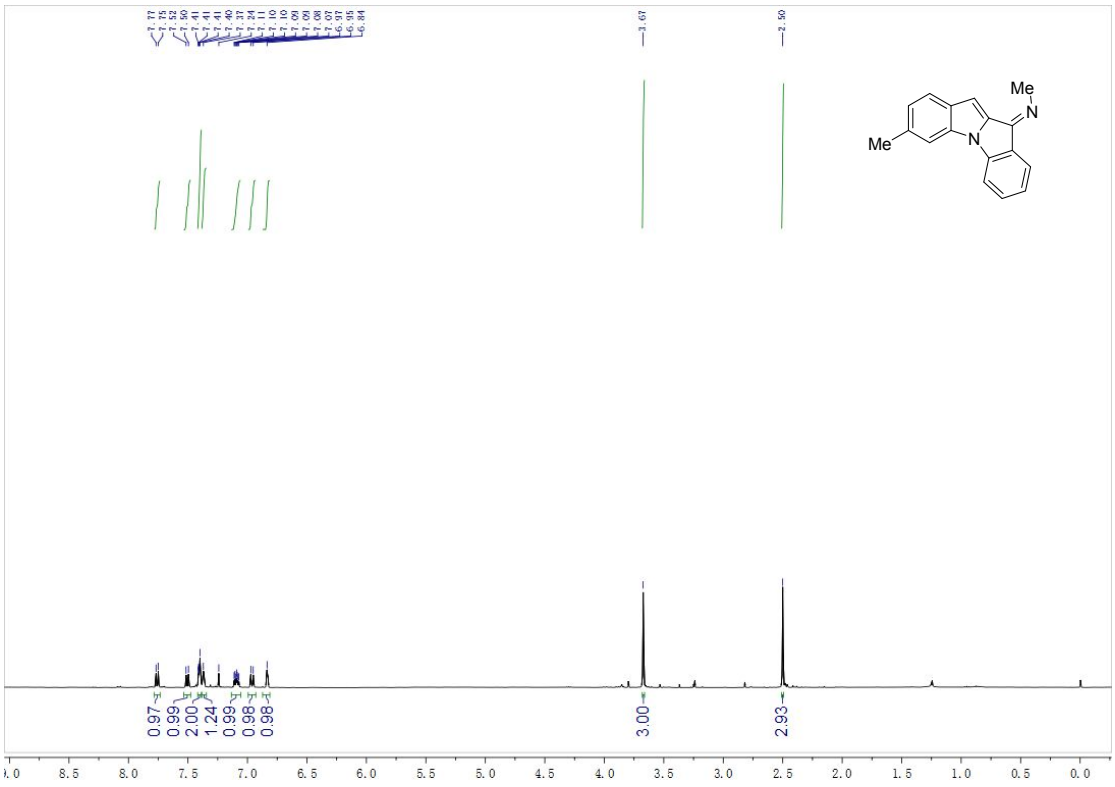


Figure S54. ^{1}H NMR for compound **3e**

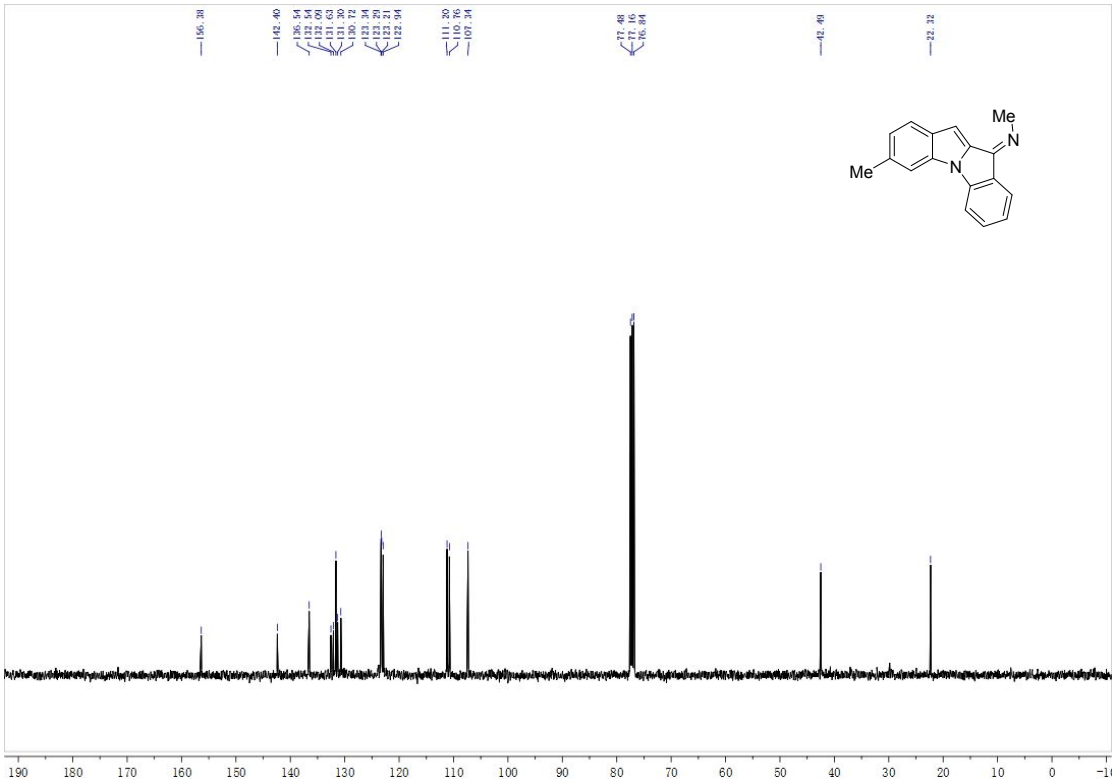


Figure S55. $^{13}C\{^{1}H\}$ NMR for compound **3e**

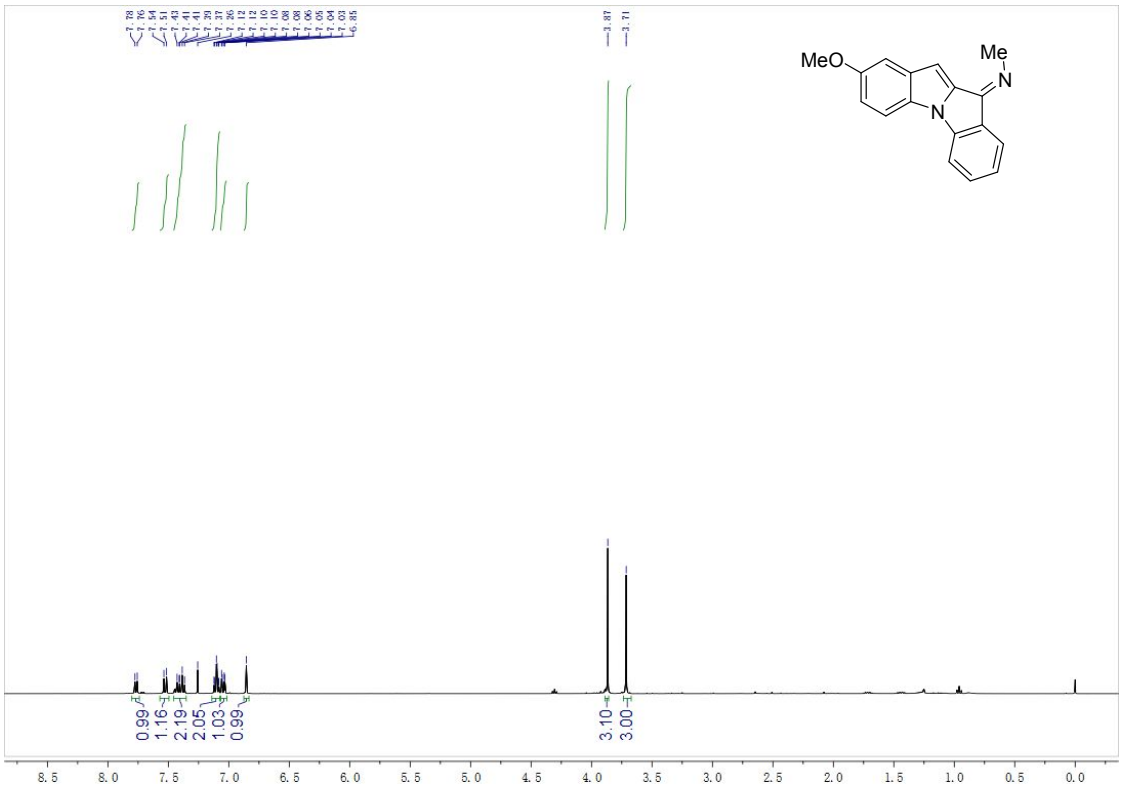


Figure S56. ^{1}H NMR for compound **3f**

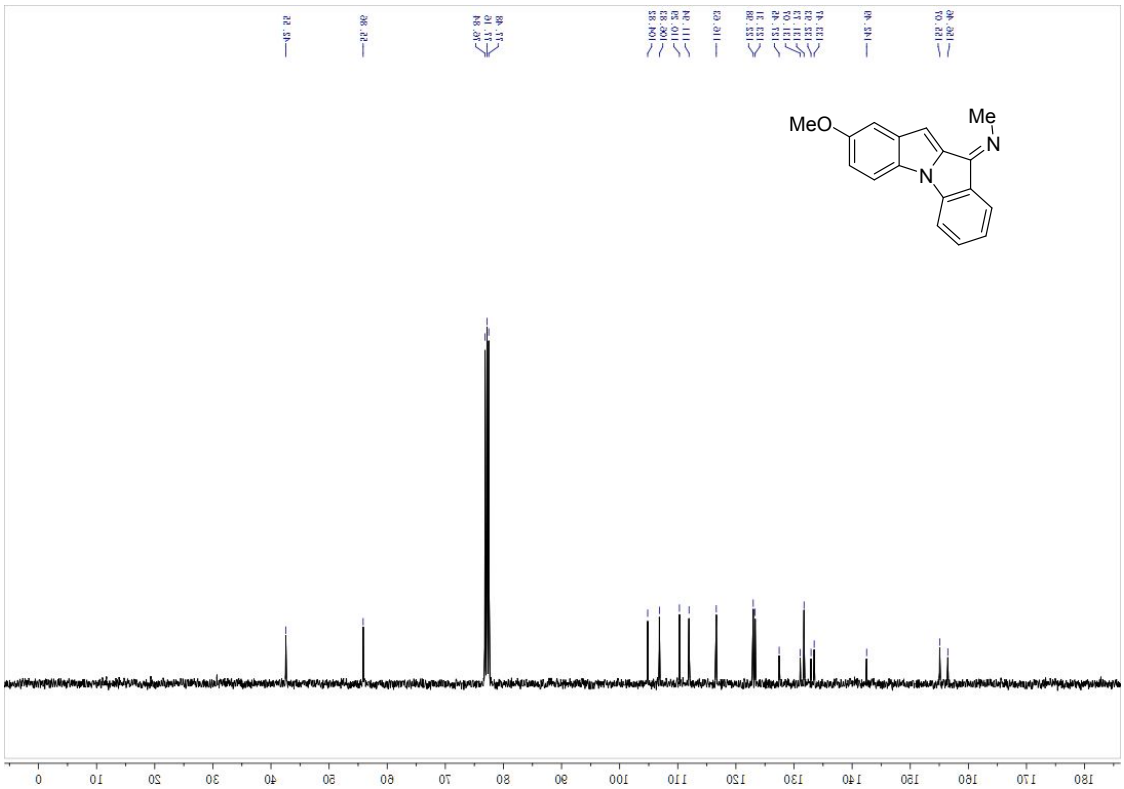


Figure S57. $^{13}C\{^{1}H\}$ NMR for compound **3f**

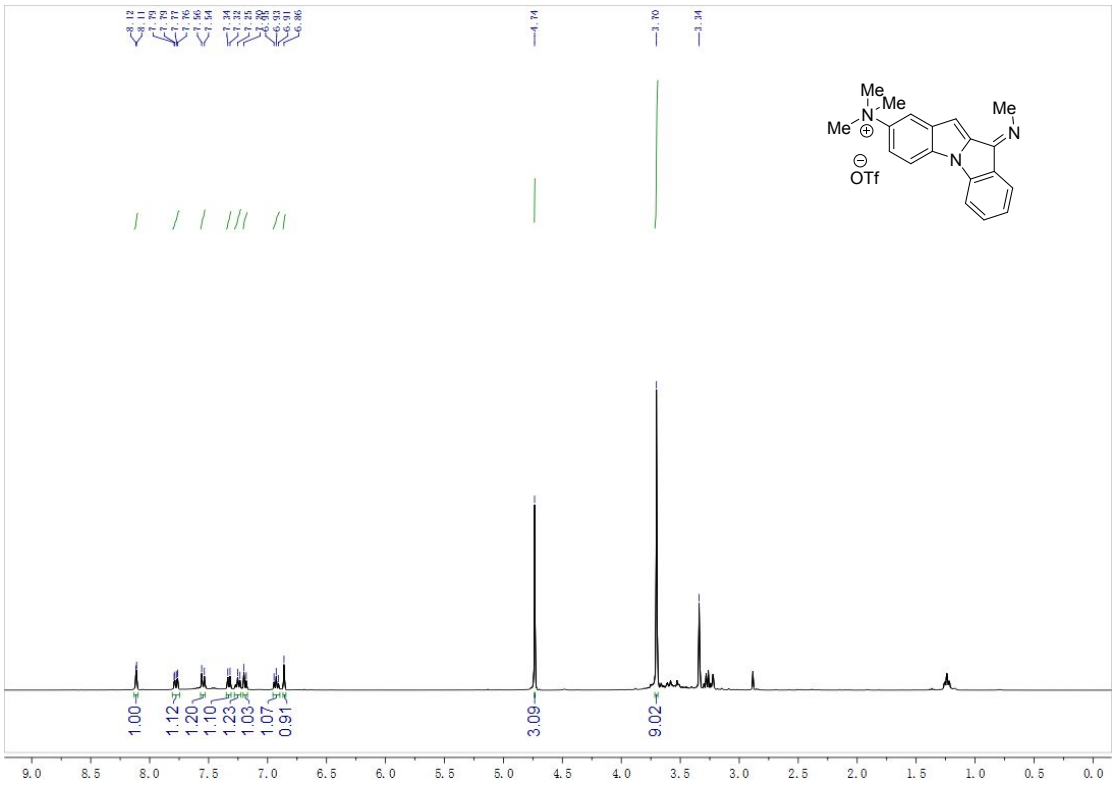


Figure S58. ^{1}H NMR for compound **3g**

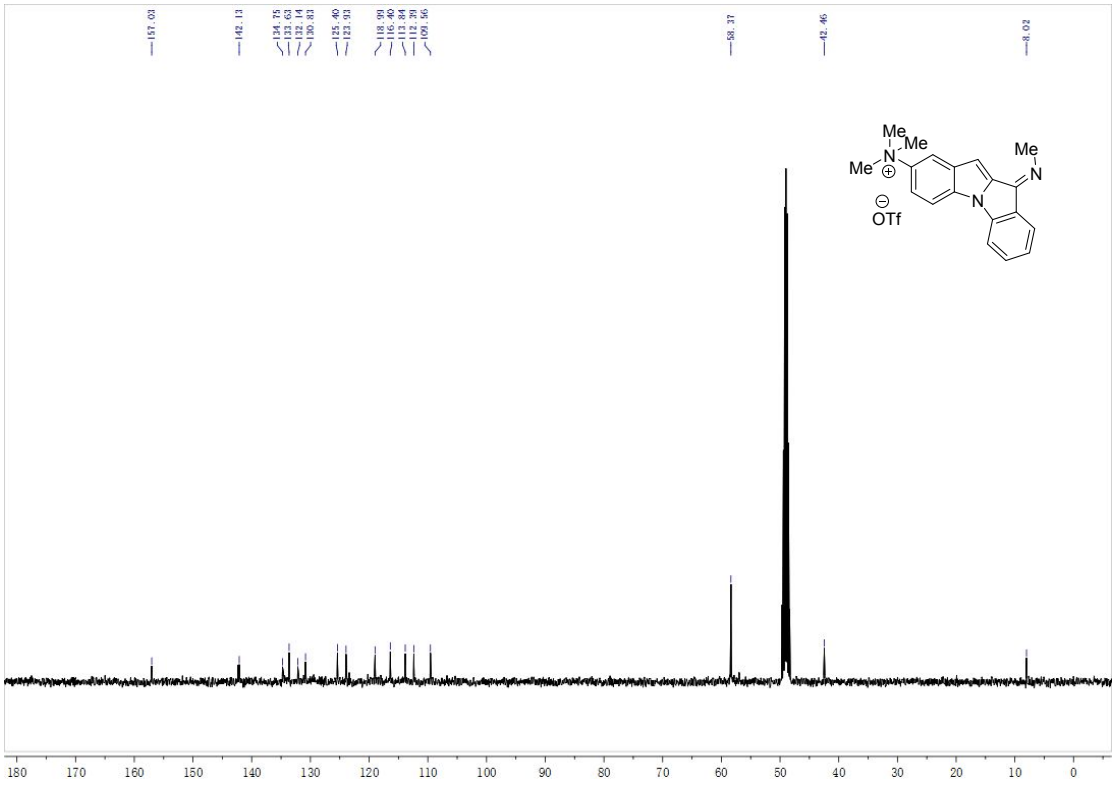


Figure S59. $^{13}C\{^{1}H\}$ NMR for compound **3g**

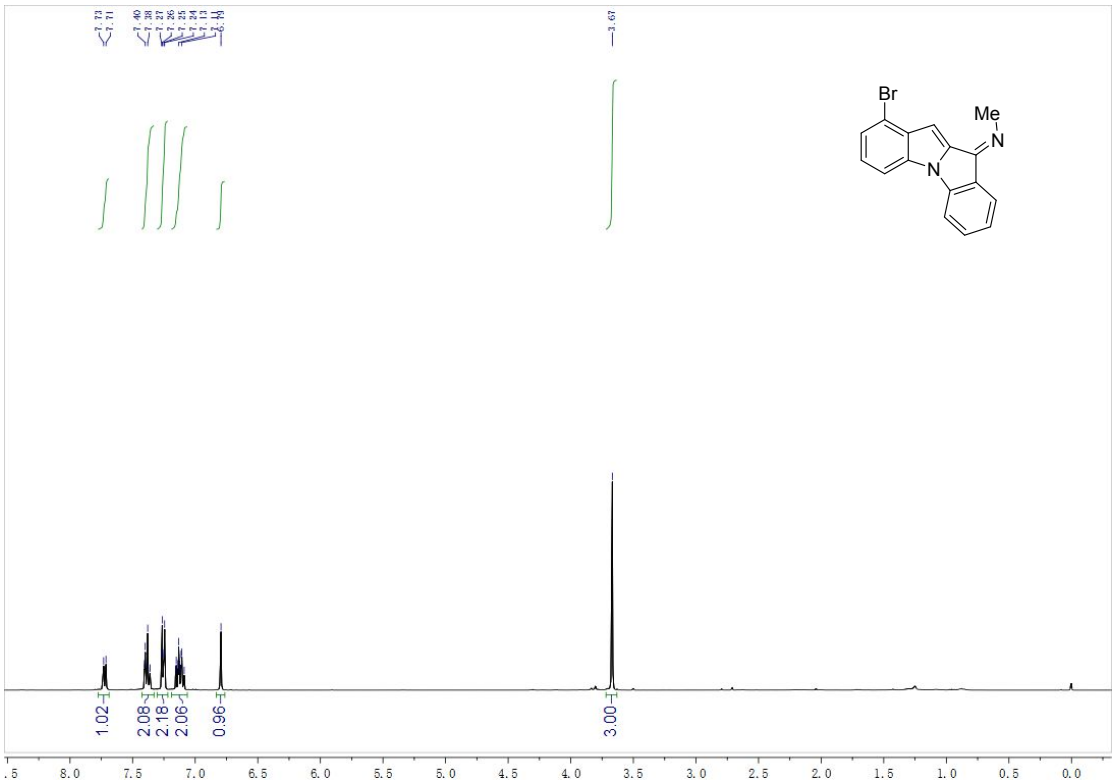


Figure S60. ^{1}H NMR for compound **3h**

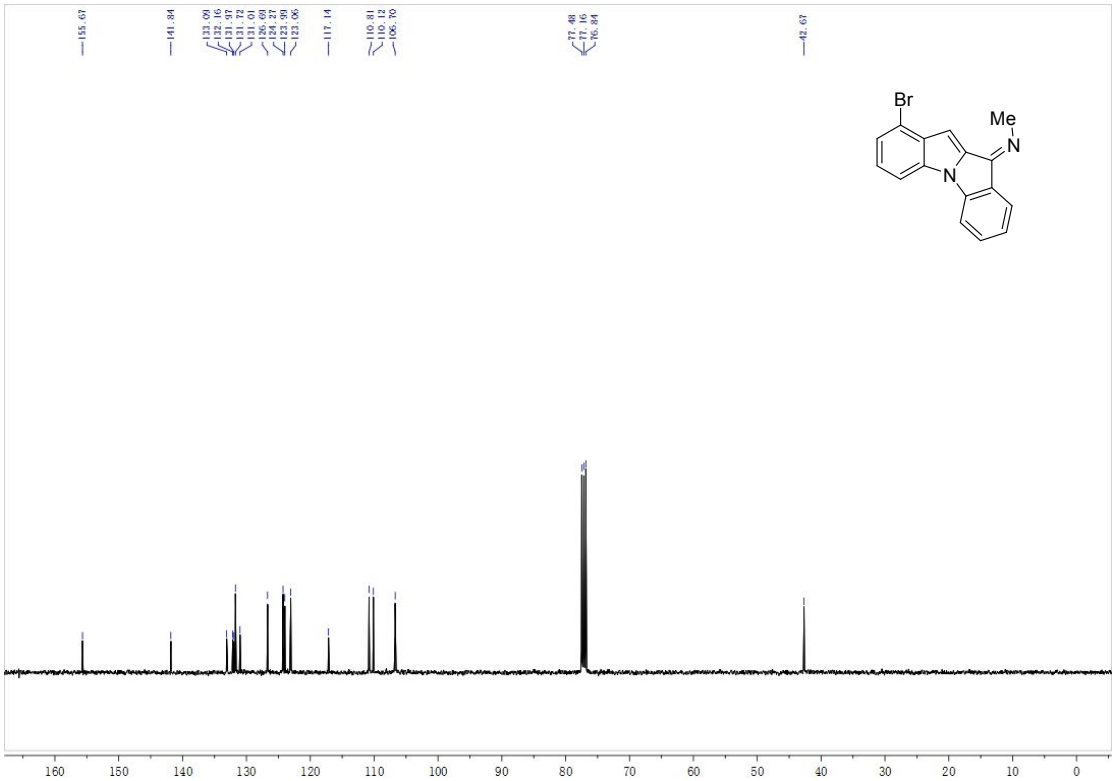


Figure S61. $^{13}C\{^{1}H\}$ NMR for compound **3h**

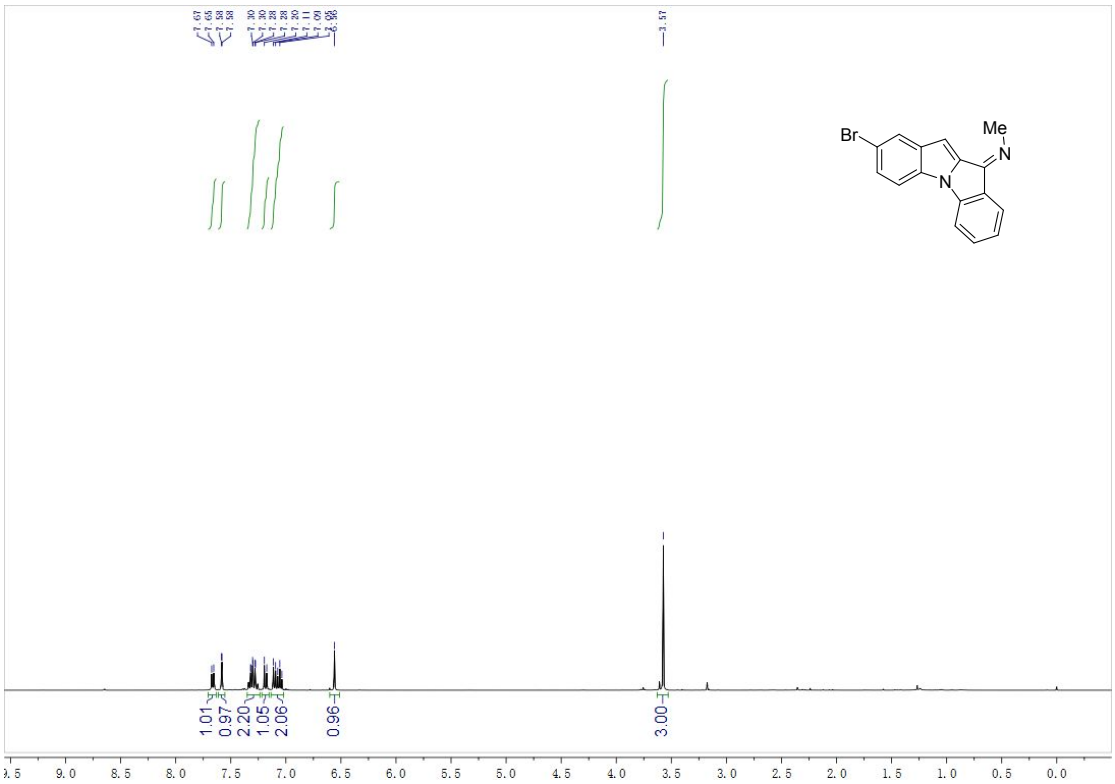


Figure S62. ^{1}H NMR for compound **3i**

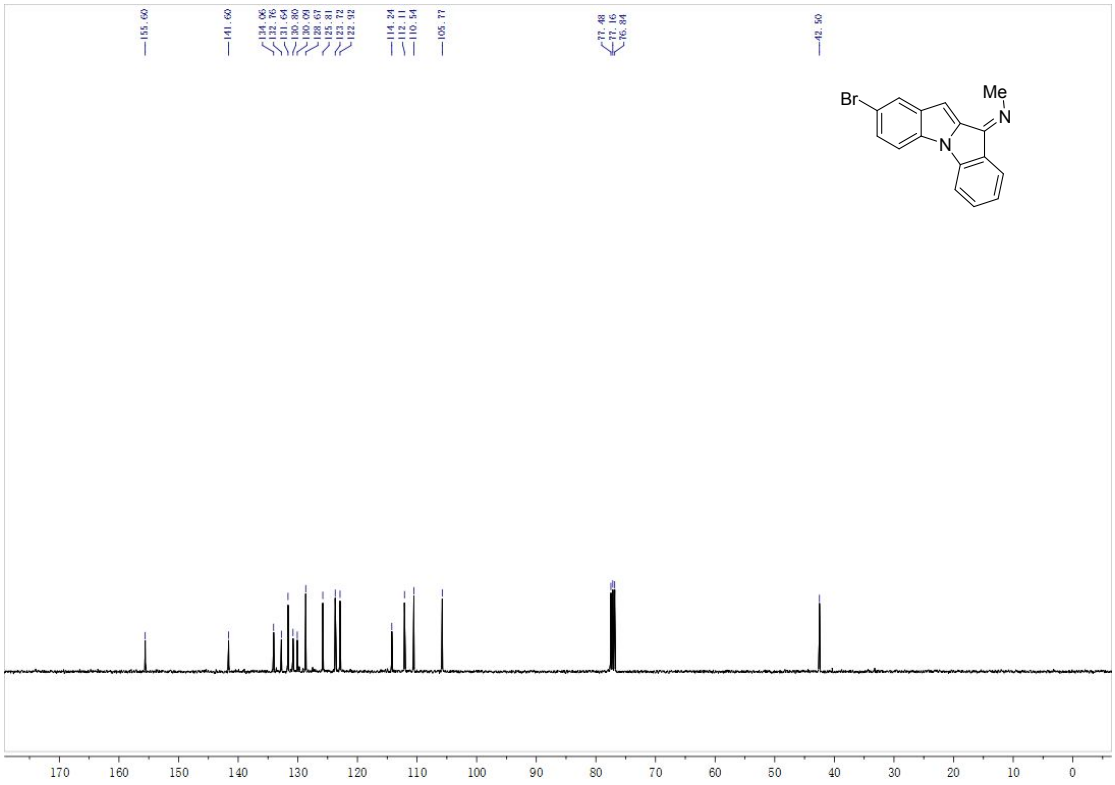


Figure S63. $^{13}C\{^{1}H\}$ NMR for compound **3i**

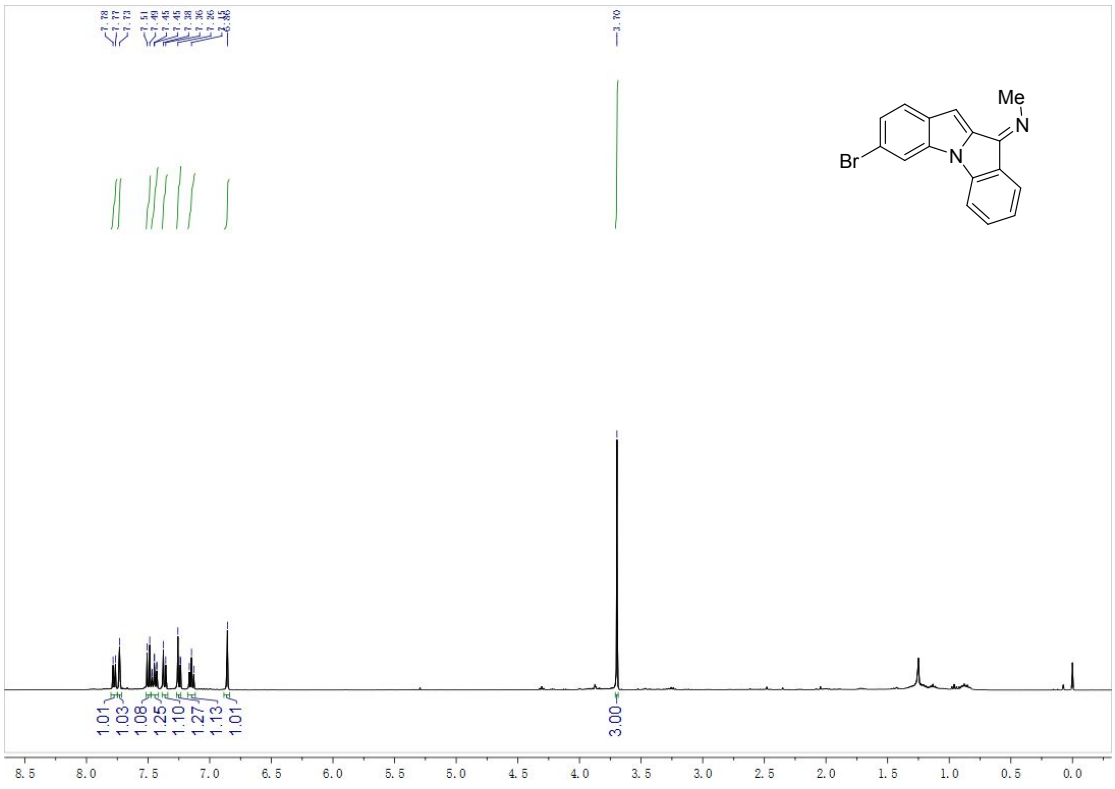


Figure S64. ^{1}H NMR for compound **3j**

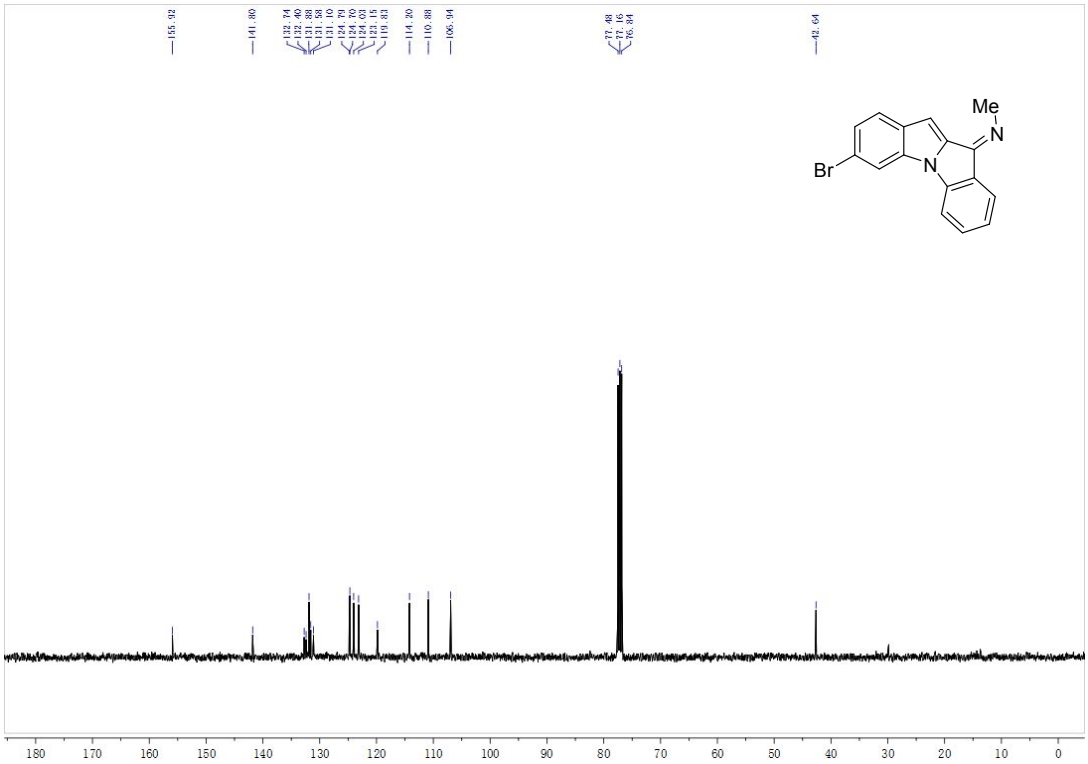


Figure S65. $^{13}C\{^{1}H\}$ NMR for compound **3j**

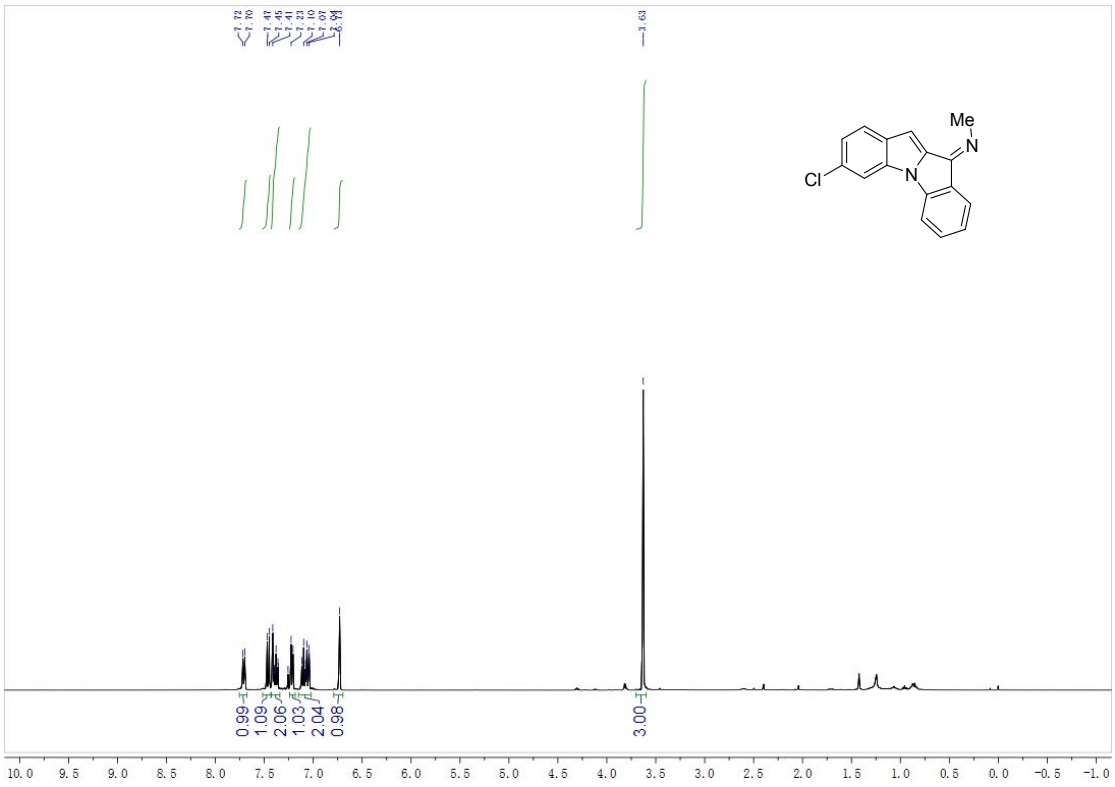


Figure S66. ^{1}H NMR for compound **3k**

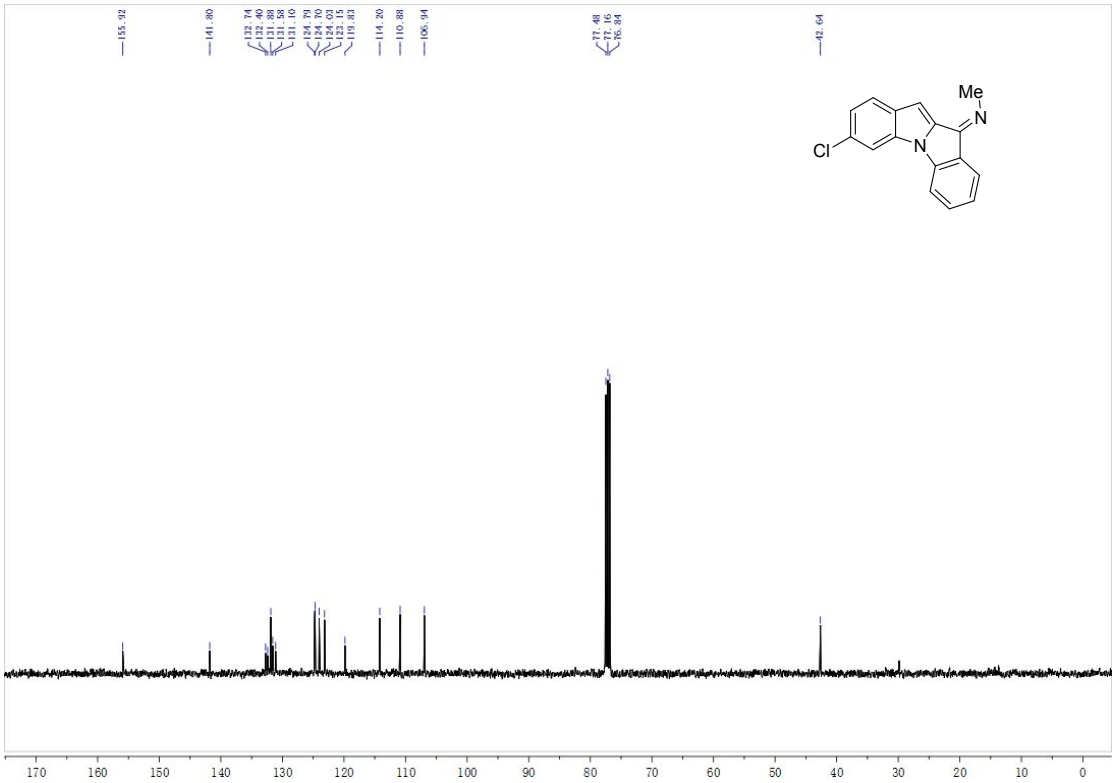


Figure S67. $^{13}C\{^{1}H\}$ NMR for compound **3k**

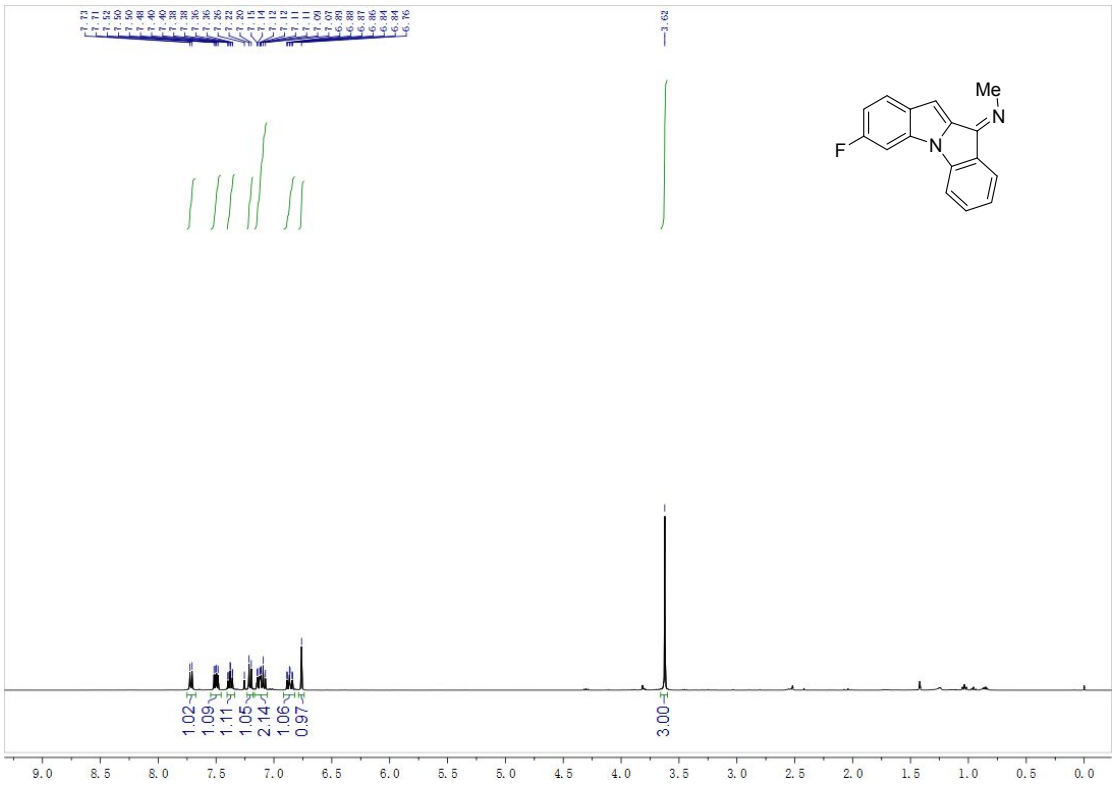


Figure S68. ^{1}H NMR for compound **3l**

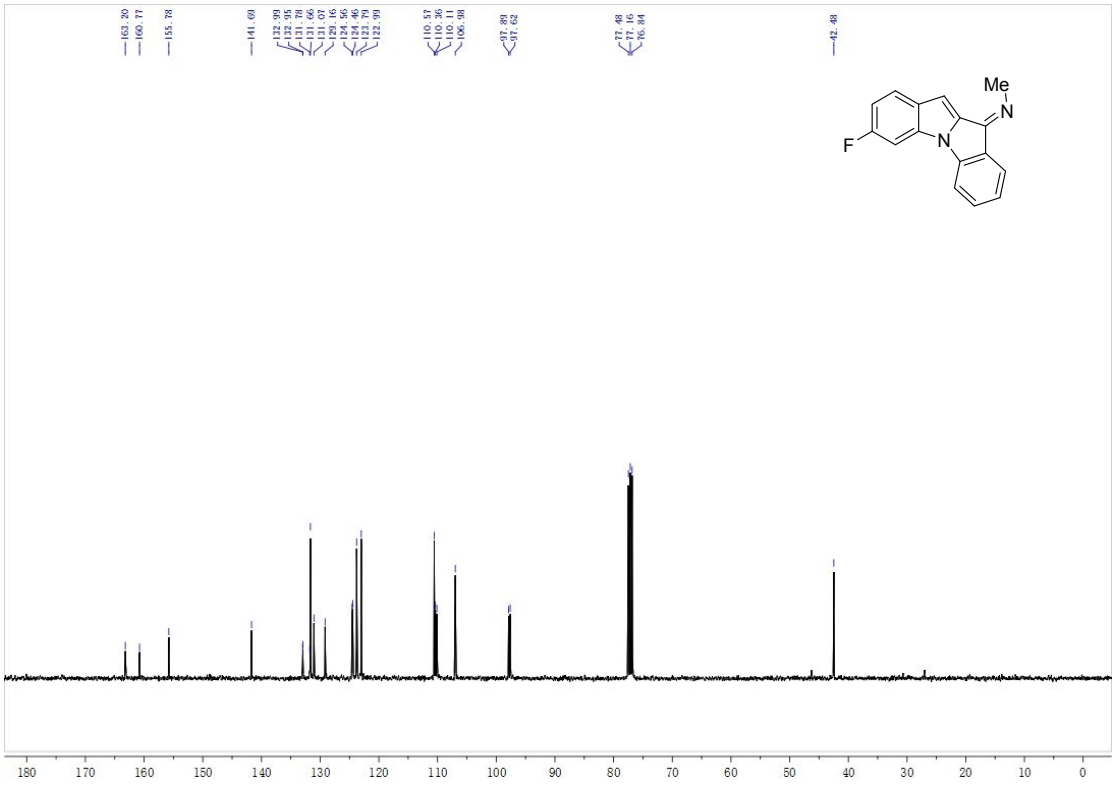


Figure S69. $^{13}C\{^{1}H\}$ NMR for compound **3l**

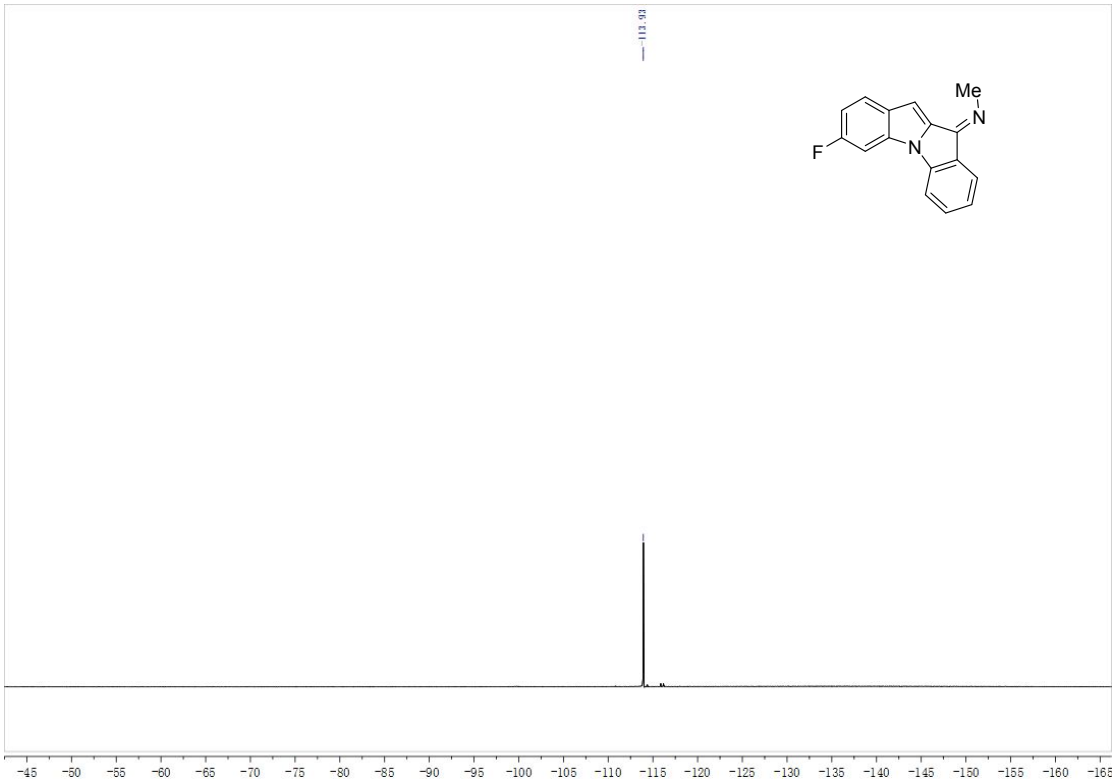


Figure S70. ^{19}F NMR for compound **3l**

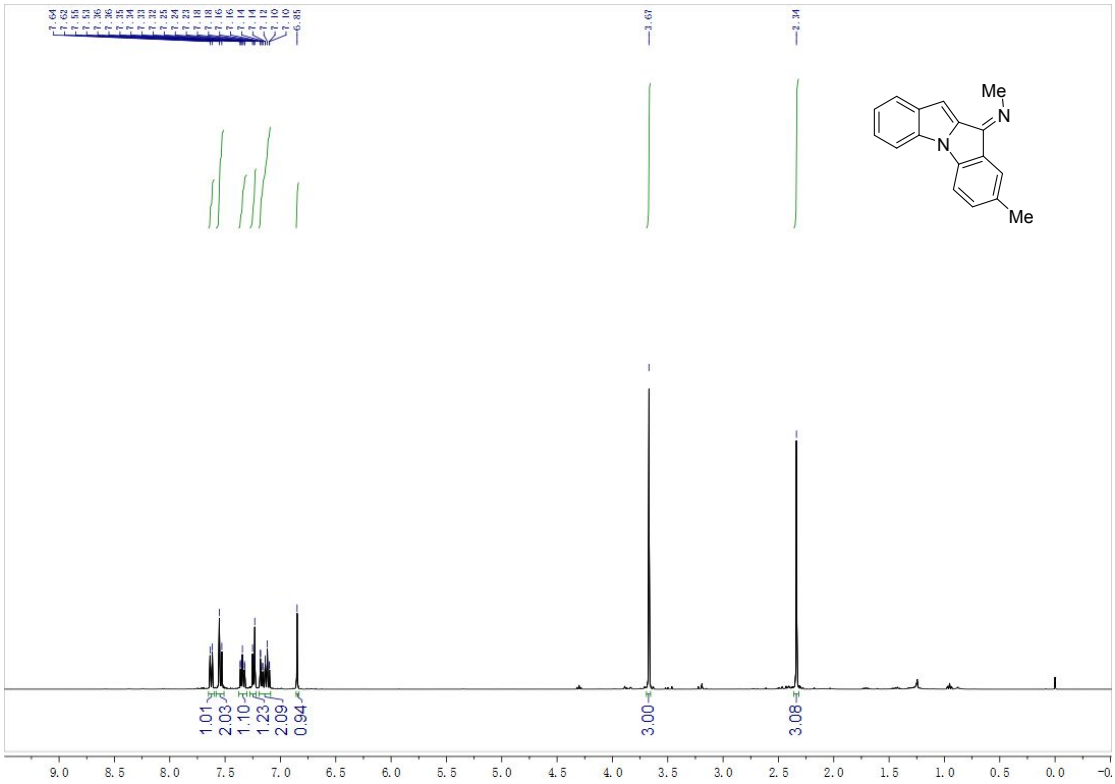


Figure S71. ^{1}H NMR for compound **3m**

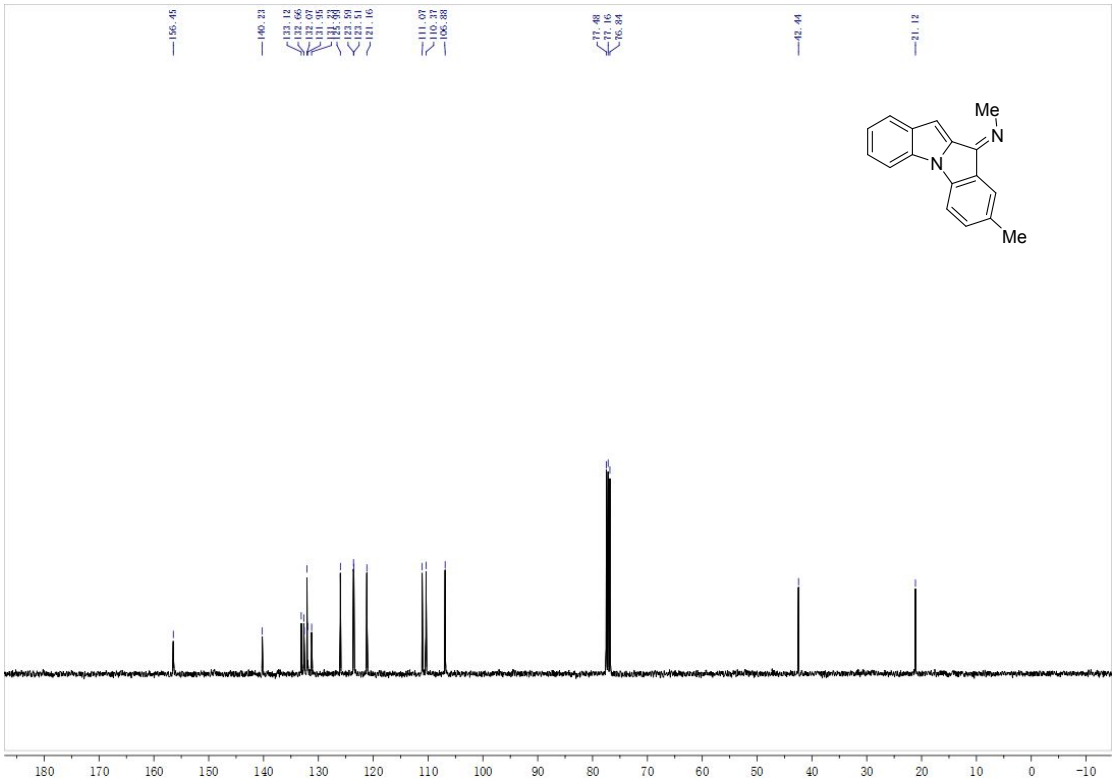


Figure S72. $^{13}C\{^{1}H\}$ NMR for compound **3m**

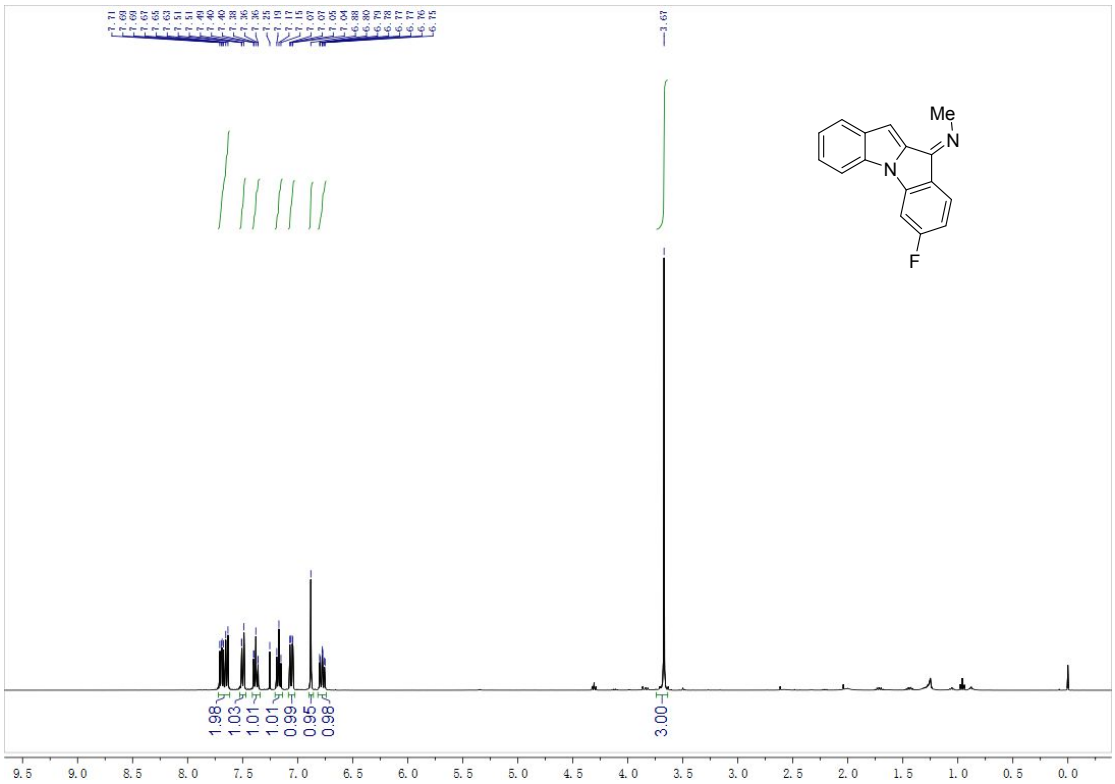


Figure S73. ^{1}H NMR for compound **3n**

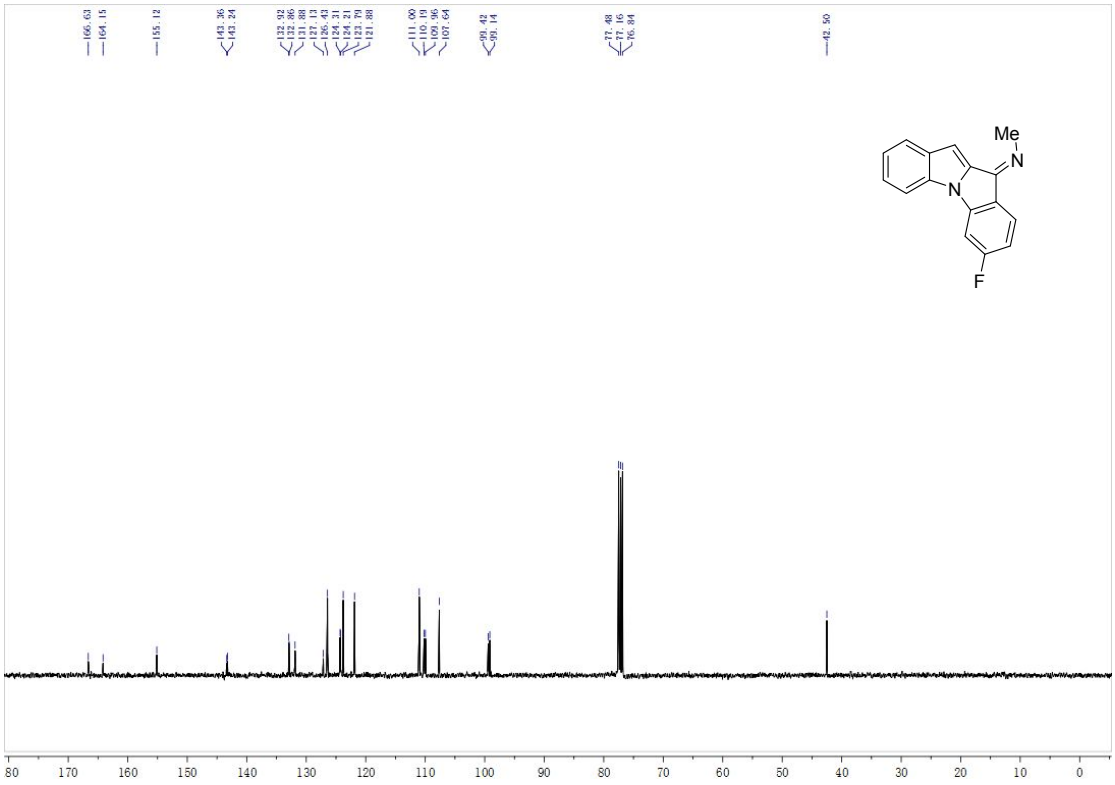


Figure S74. $^{13}C\{^{1}H\}$ NMR for compound **3n**

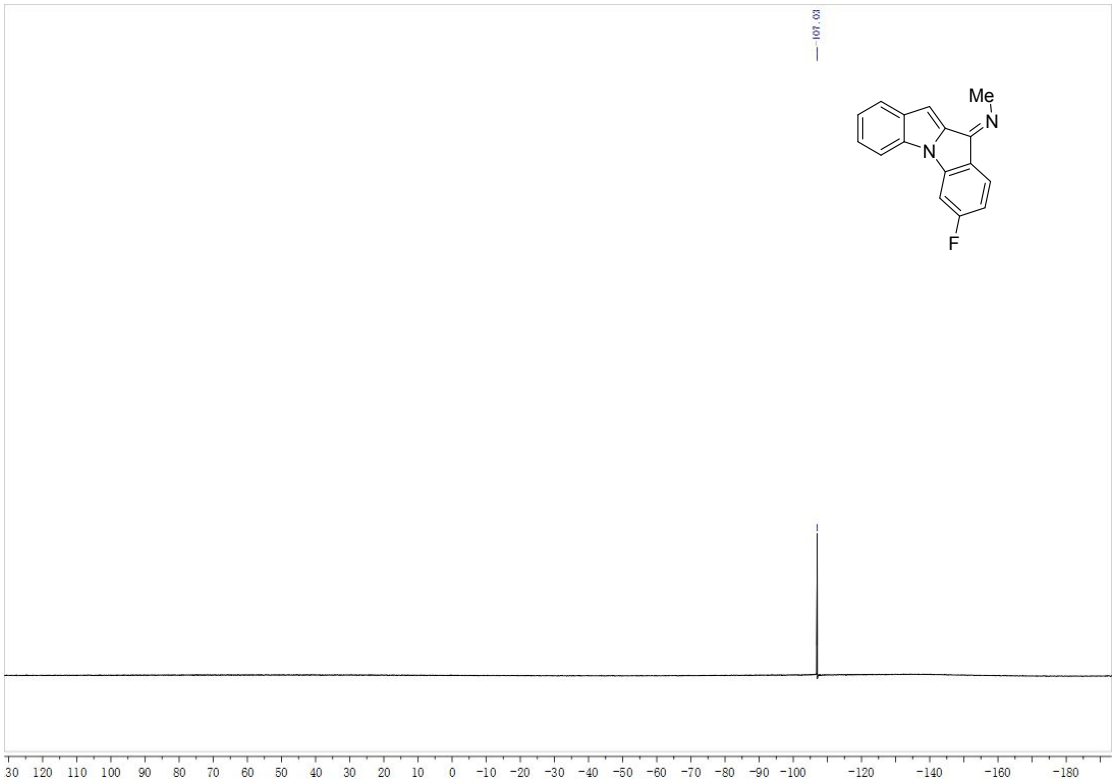


Figure S75. ^{19}F NMR for compound **3n**

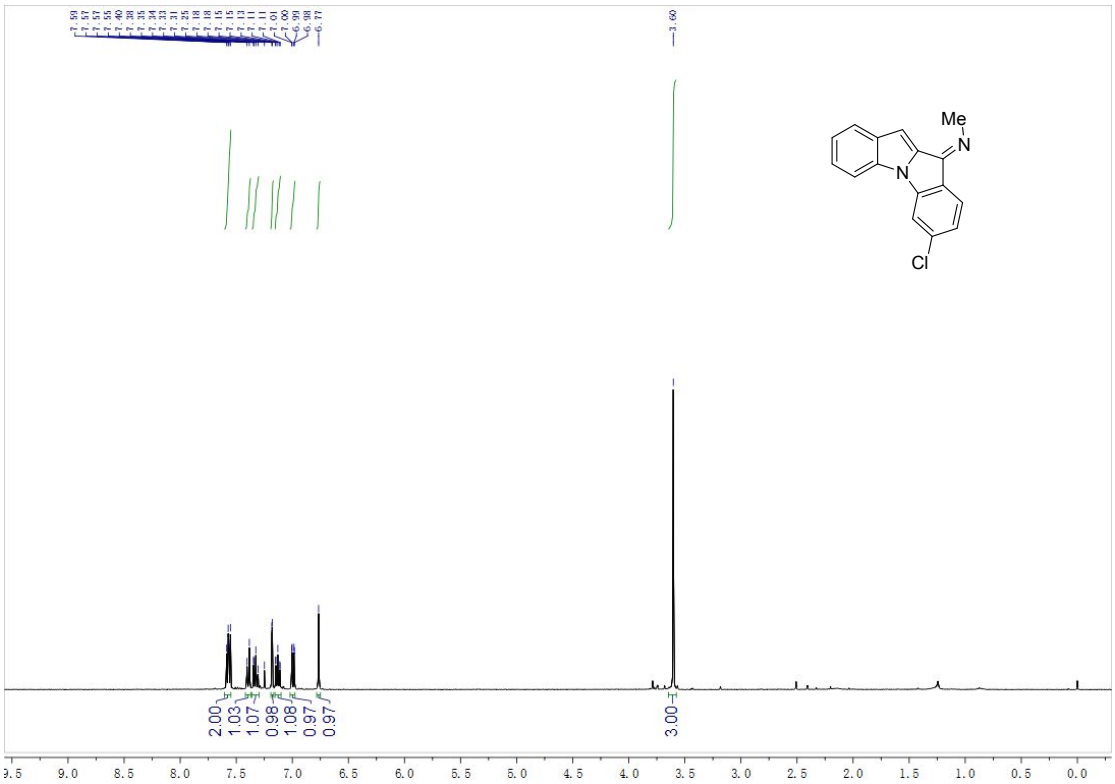


Figure S76. ^{1}H NMR for compound **3o**

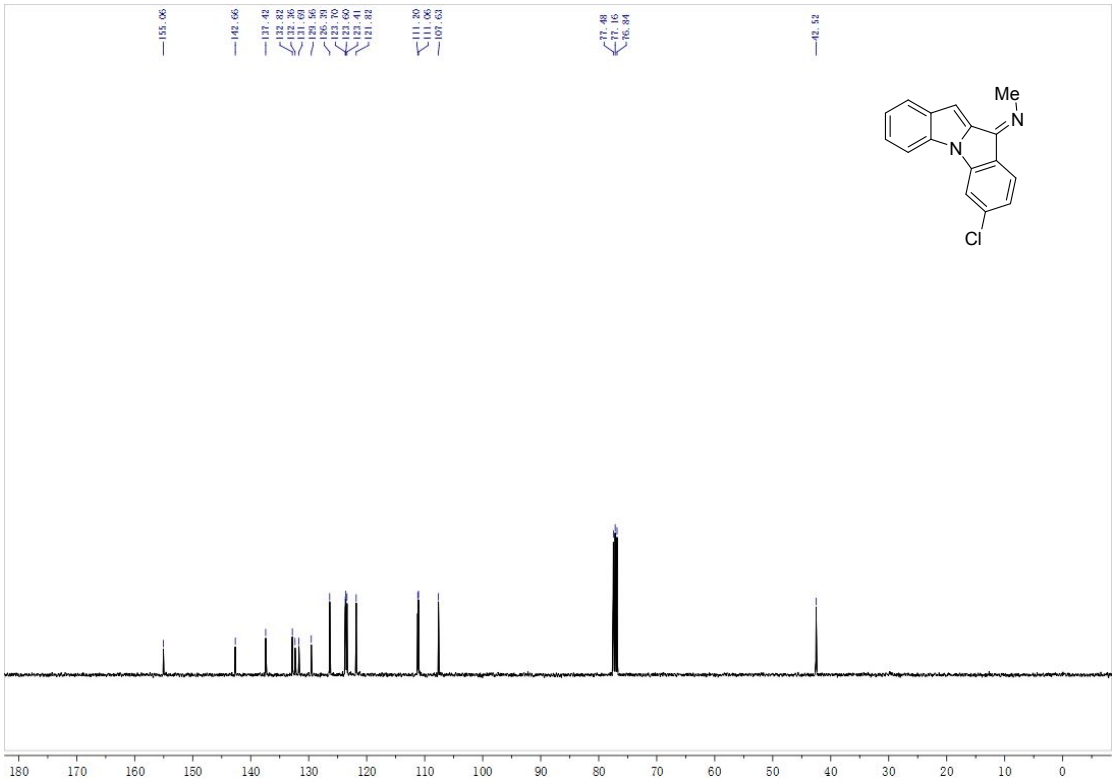


Figure S77. $^{13}C\{^{1}H\}$ NMR for compound **3o**

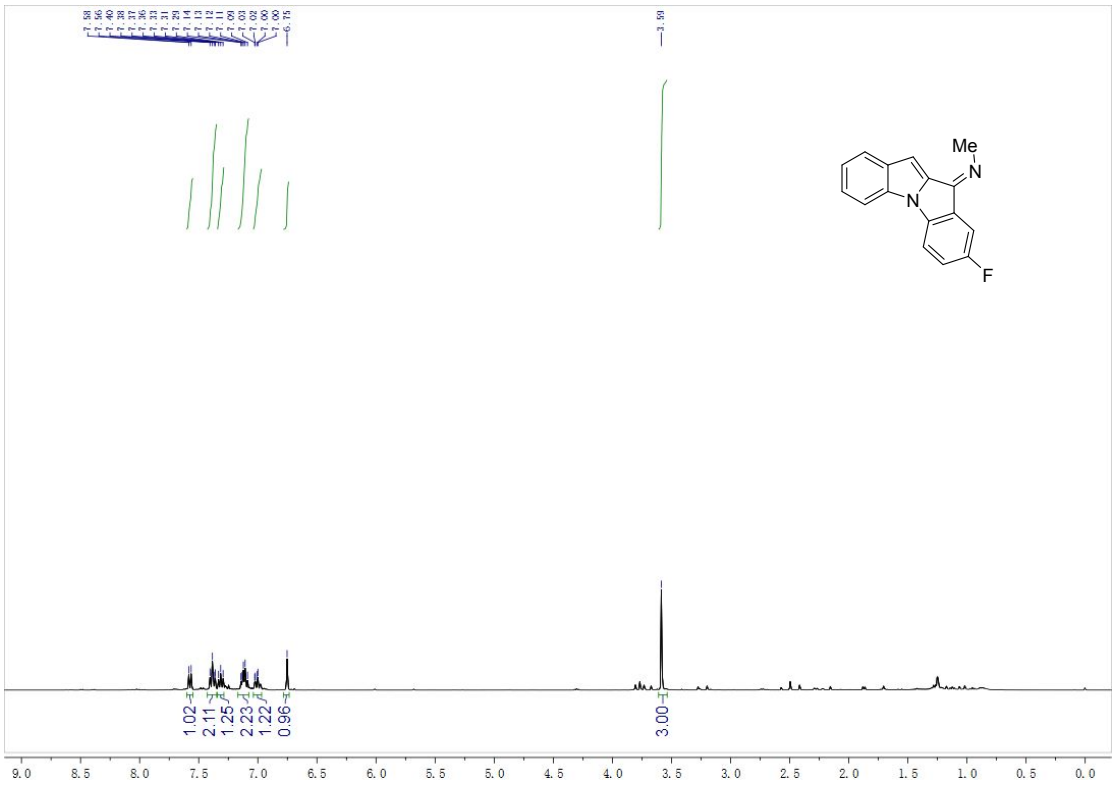


Figure S78. ^{1}H NMR for compound **3p**

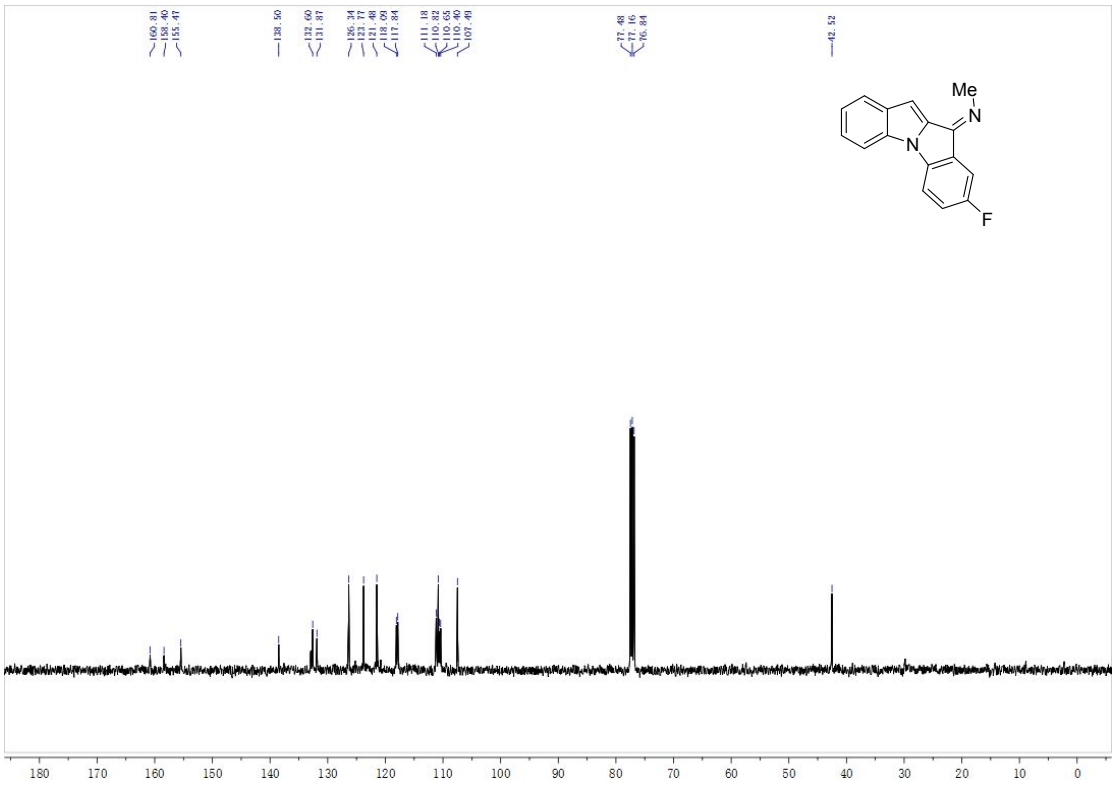


Figure S79. $^{13}C\{^{1}H\}$ NMR for compound **3p**

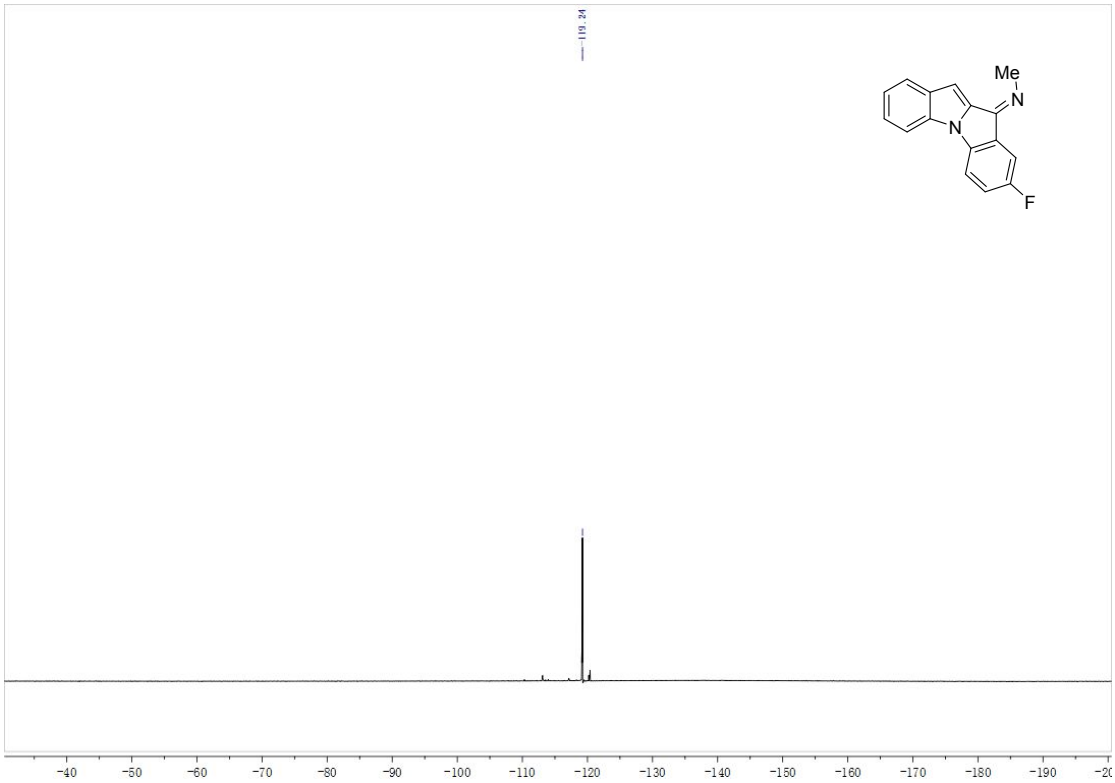


Figure S80. ^{19}F NMR for compound **3p**

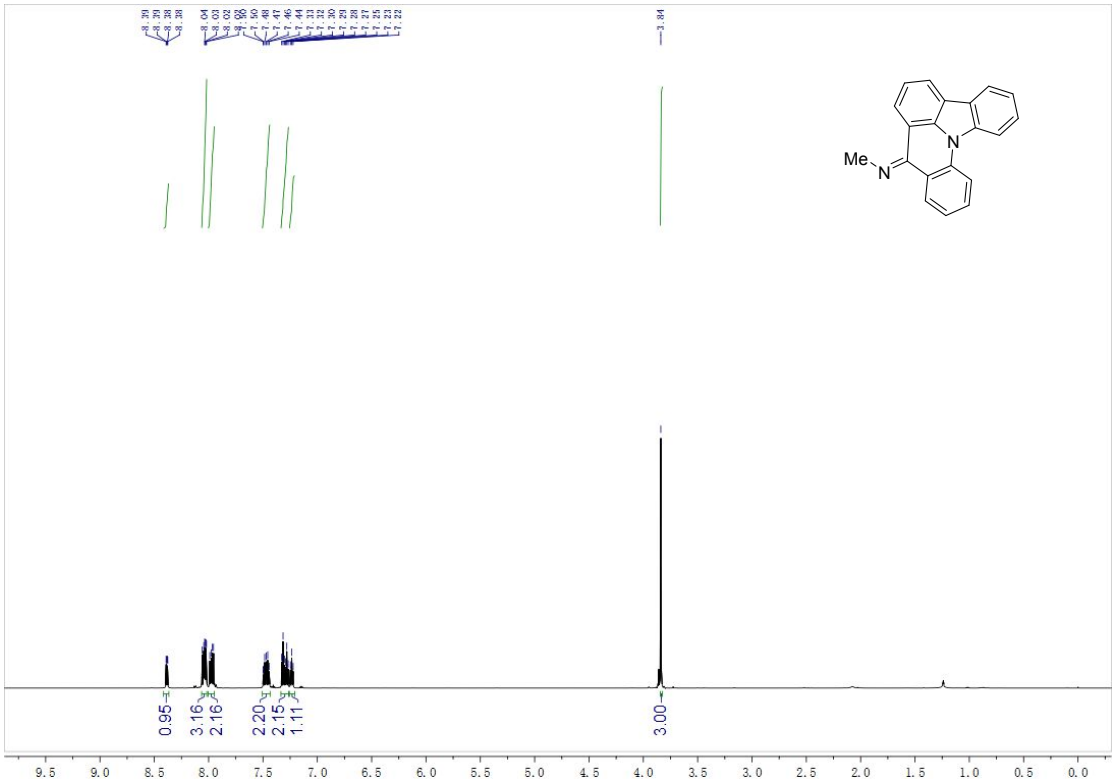


Figure S81. ^{1}H NMR for compound **3q**

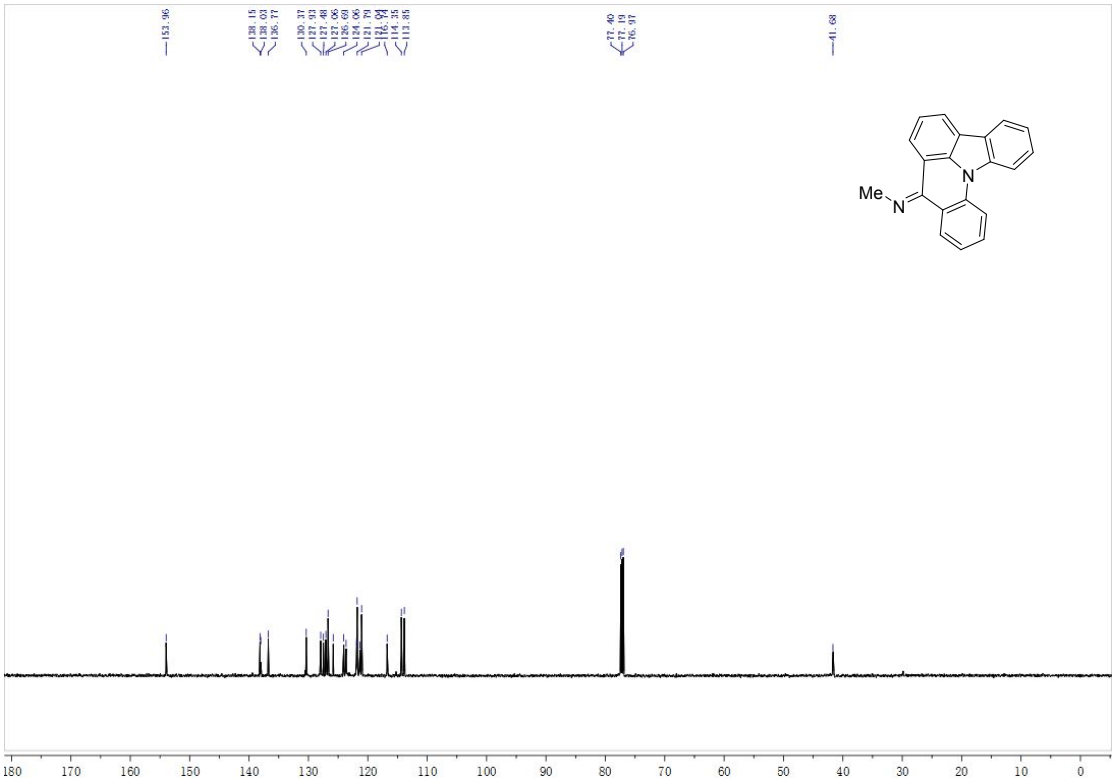


Figure S82. $^{13}C\{^{1}H\}$ NMR for compound **3q**

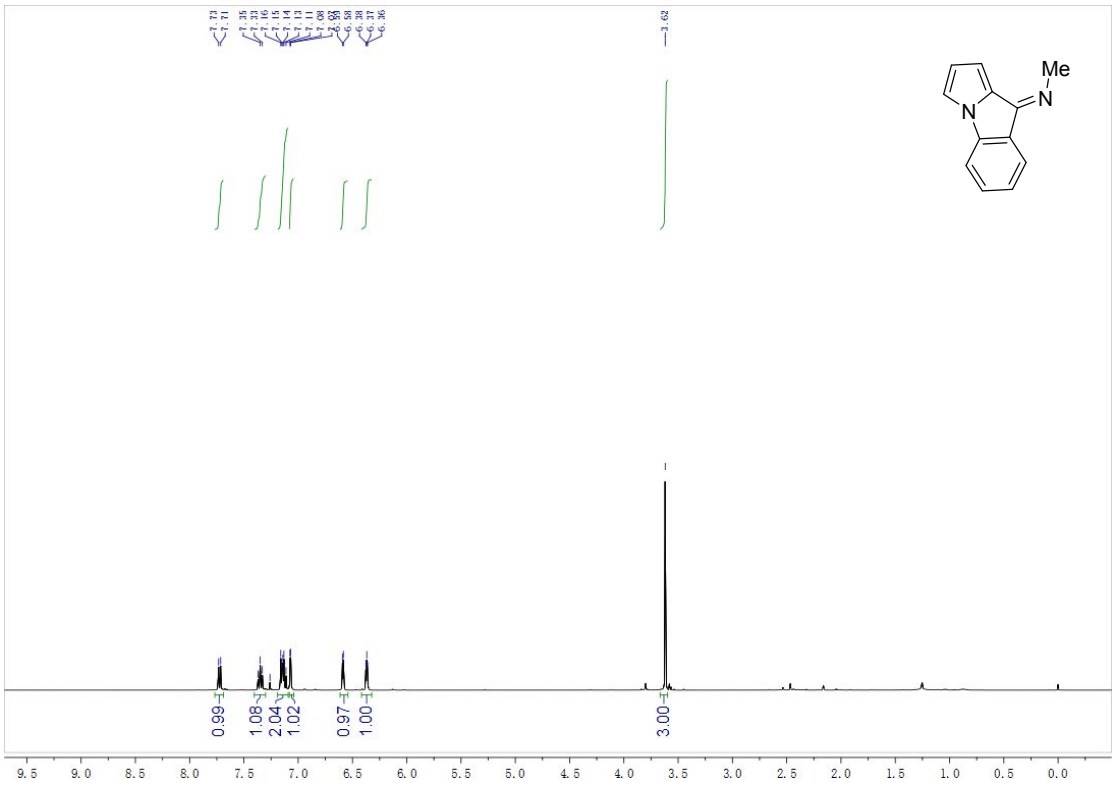


Figure S83. ^{1}H NMR for compound **3r**

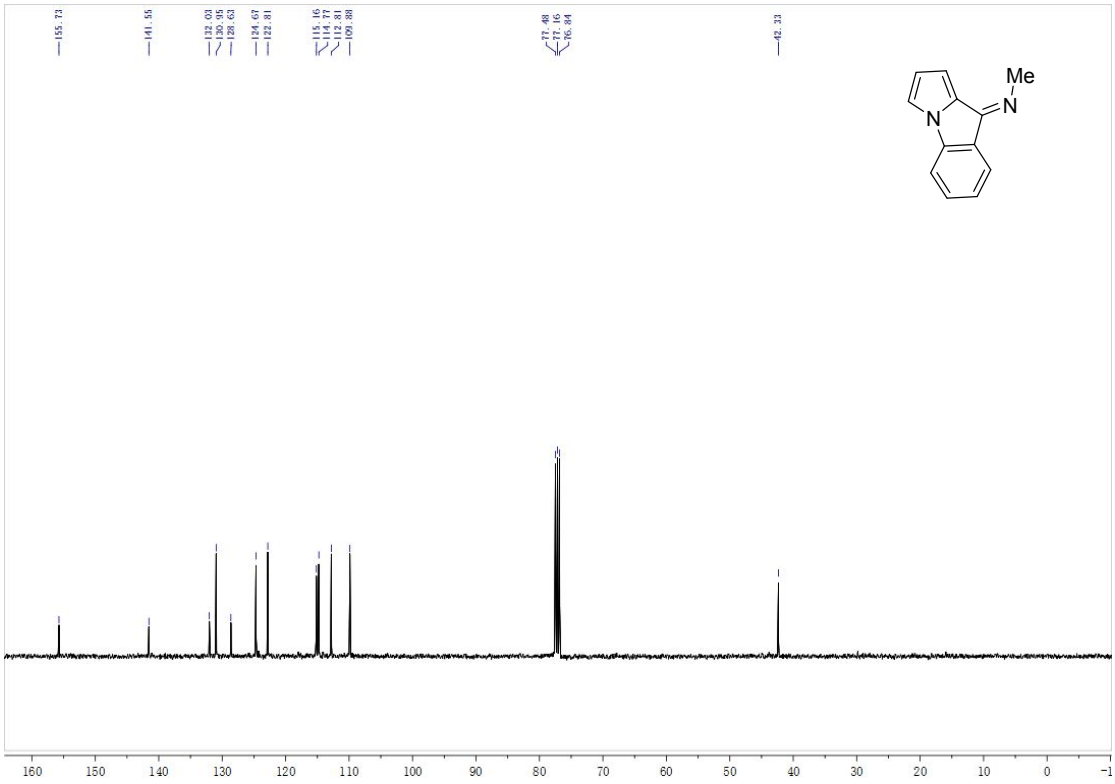


Figure S84. $^{13}C\{^{1}H\}$ NMR for compound **3r**

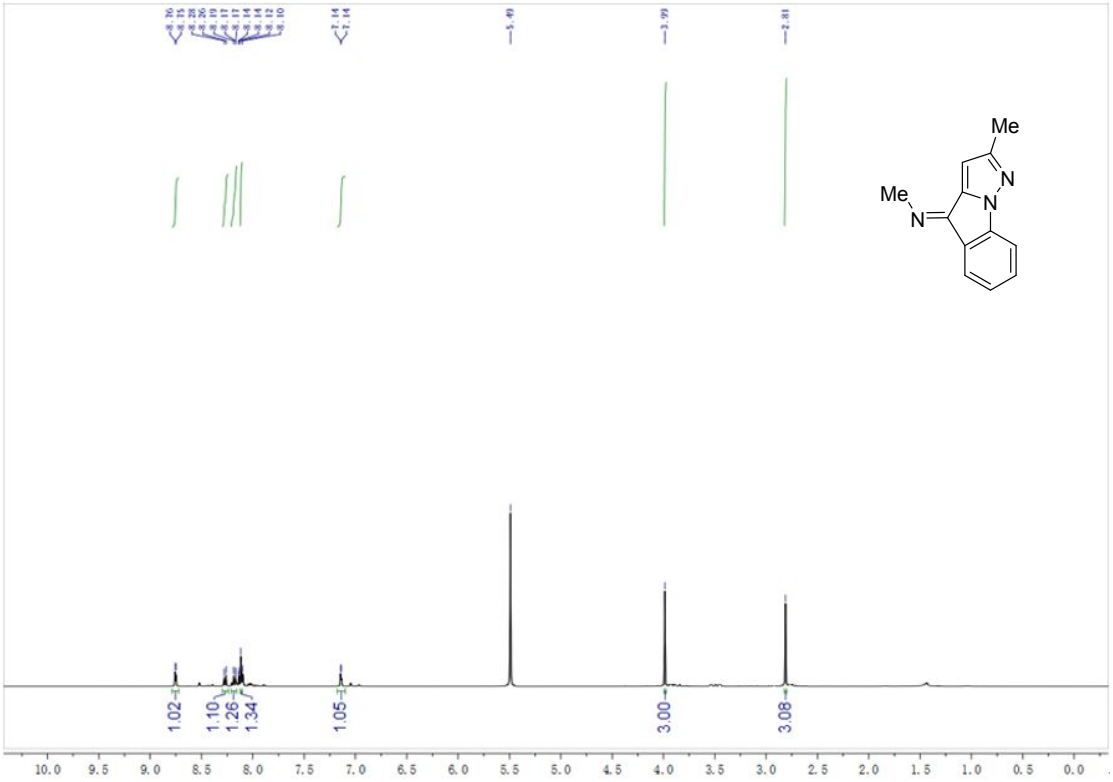


Figure S85. ^{1}H NMR for compound **3s**

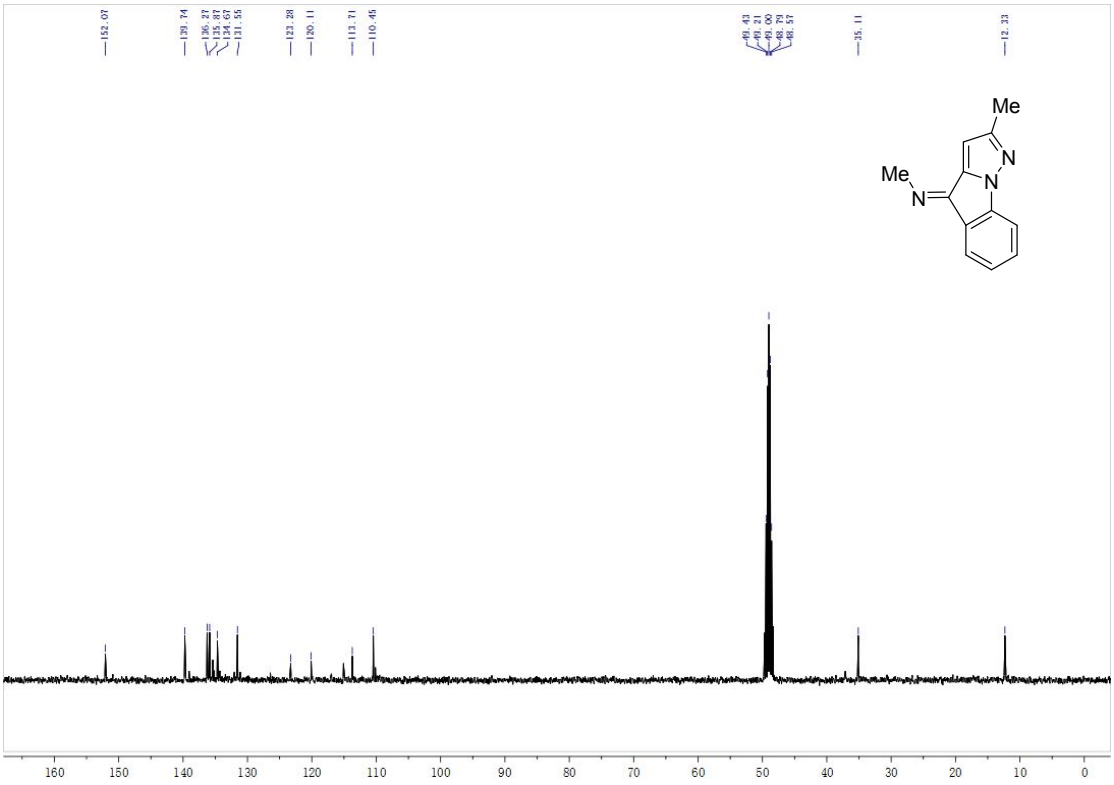


Figure S86. $^{13}C\{^{1}H\}$ NMR for compound **3s**

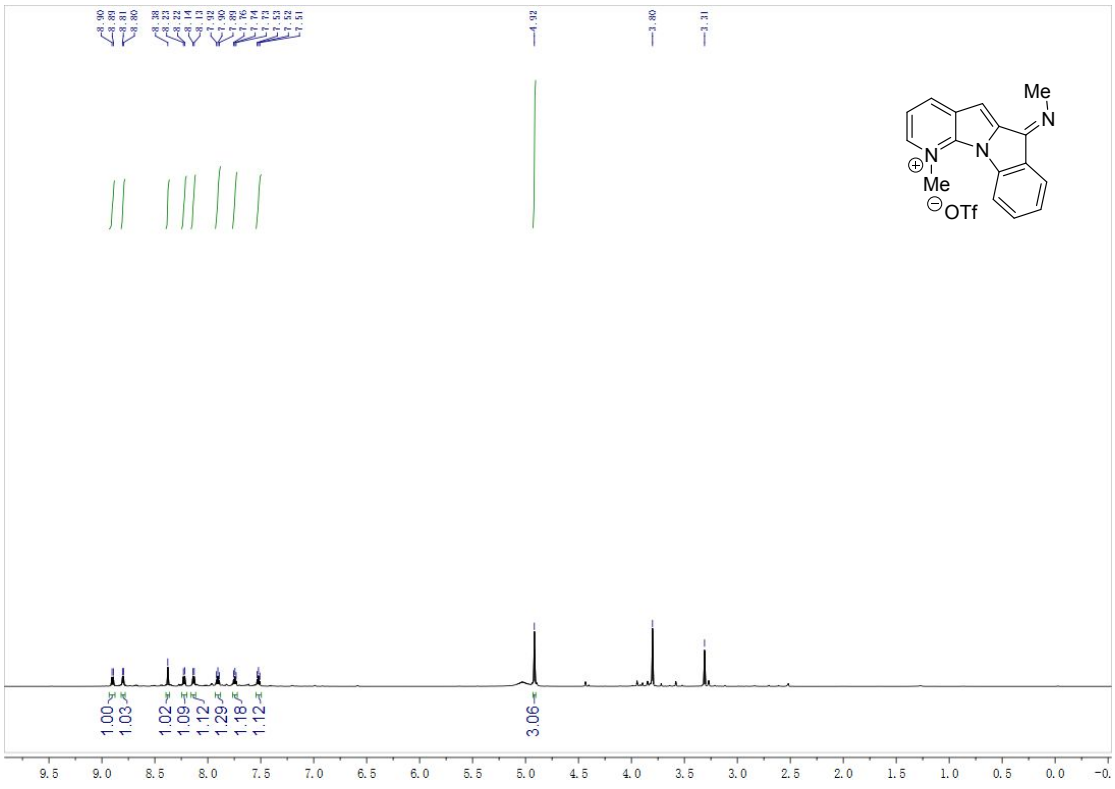


Figure S87. ^{1}H NMR for compound **3t**

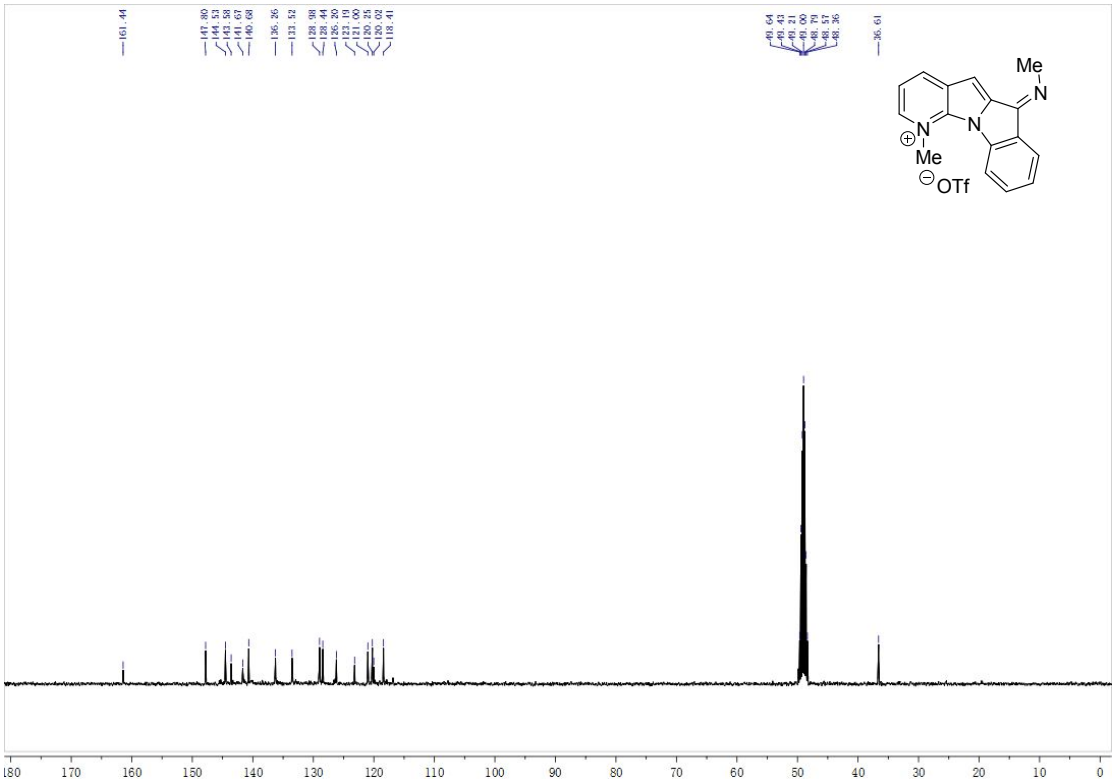


Figure S88. ^{13}C{^{1}H} NMR for compound **3t**

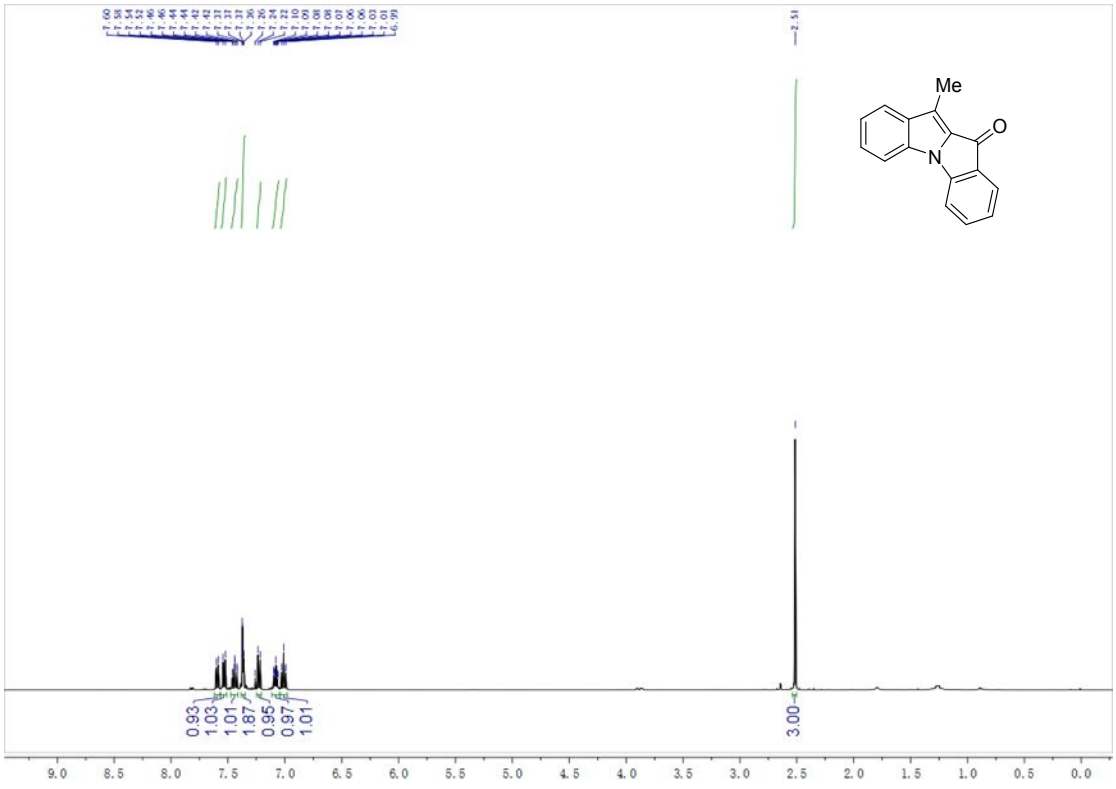


Figure S89. ^{1}H NMR for compound **4a**

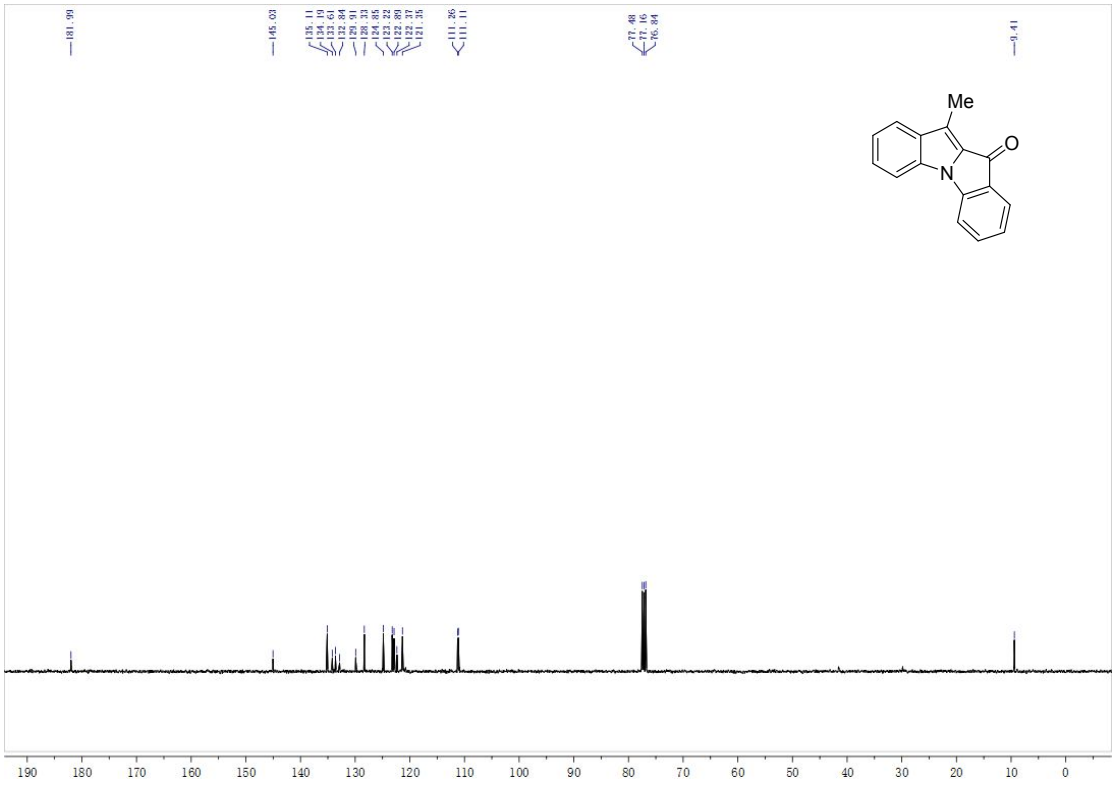


Figure S90. $^{13}C\{^{1}H\}$ NMR for compound **4a**

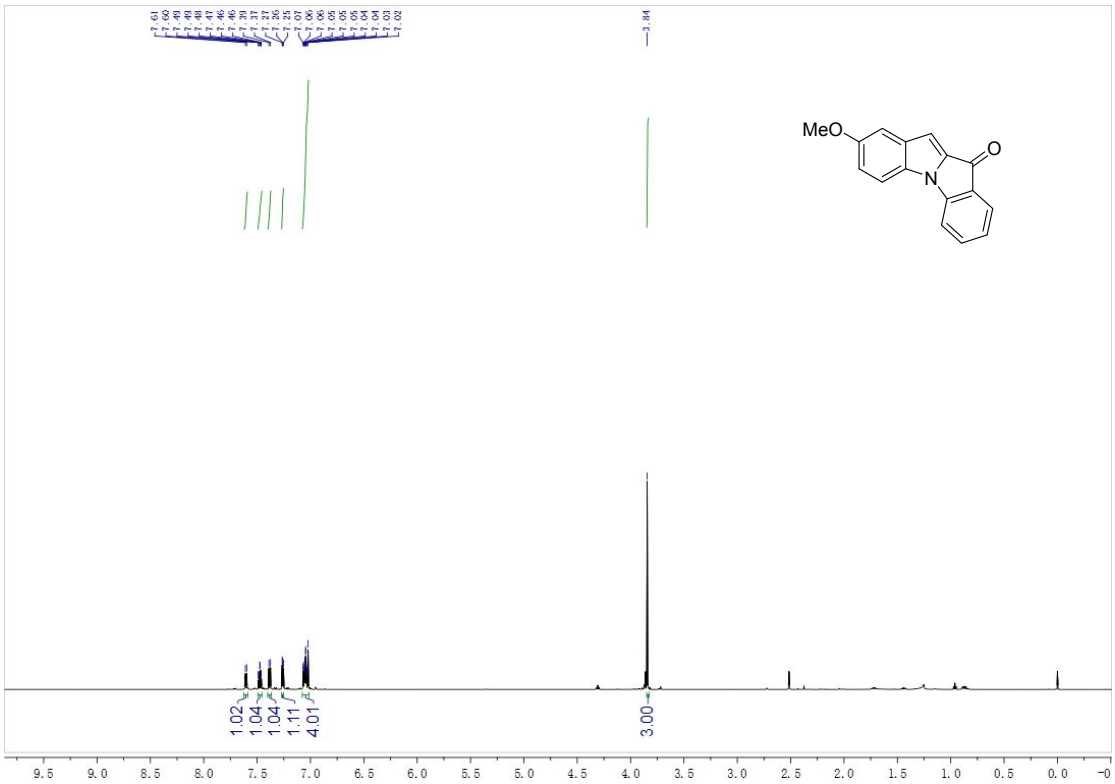


Figure S91. ^{1}H NMR for compound **4f**

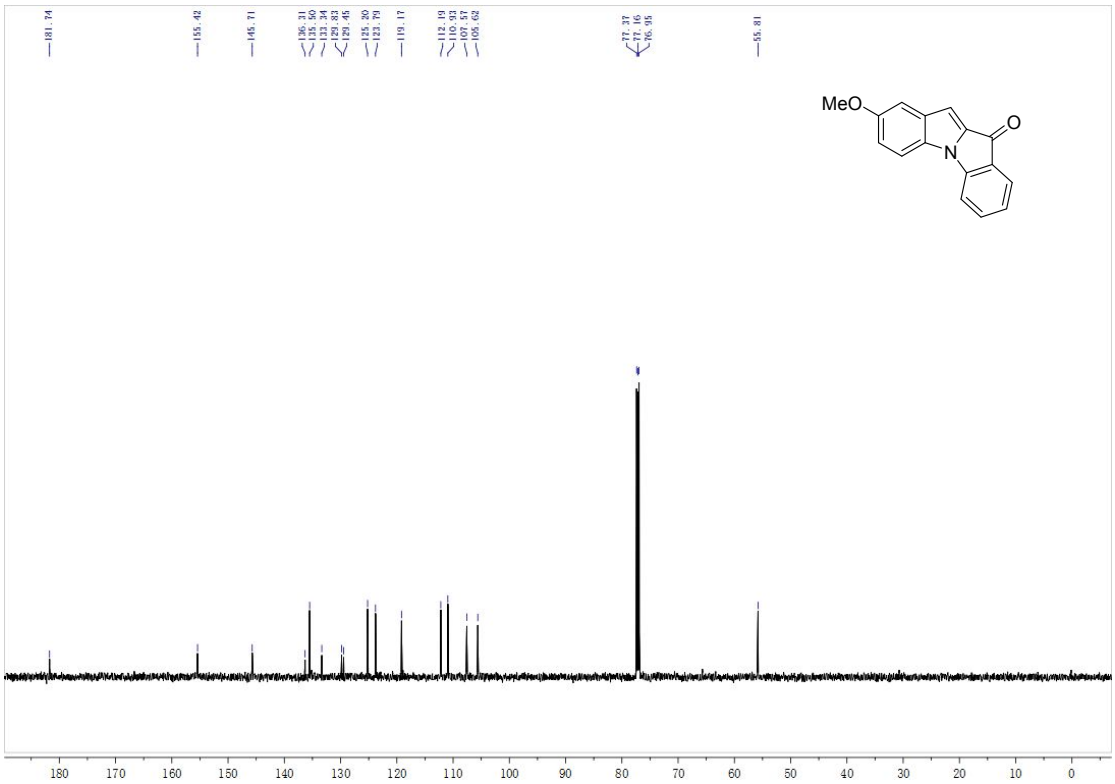


Figure S92. $^{13}C\{^{1}H\}$ NMR for compound **4f**

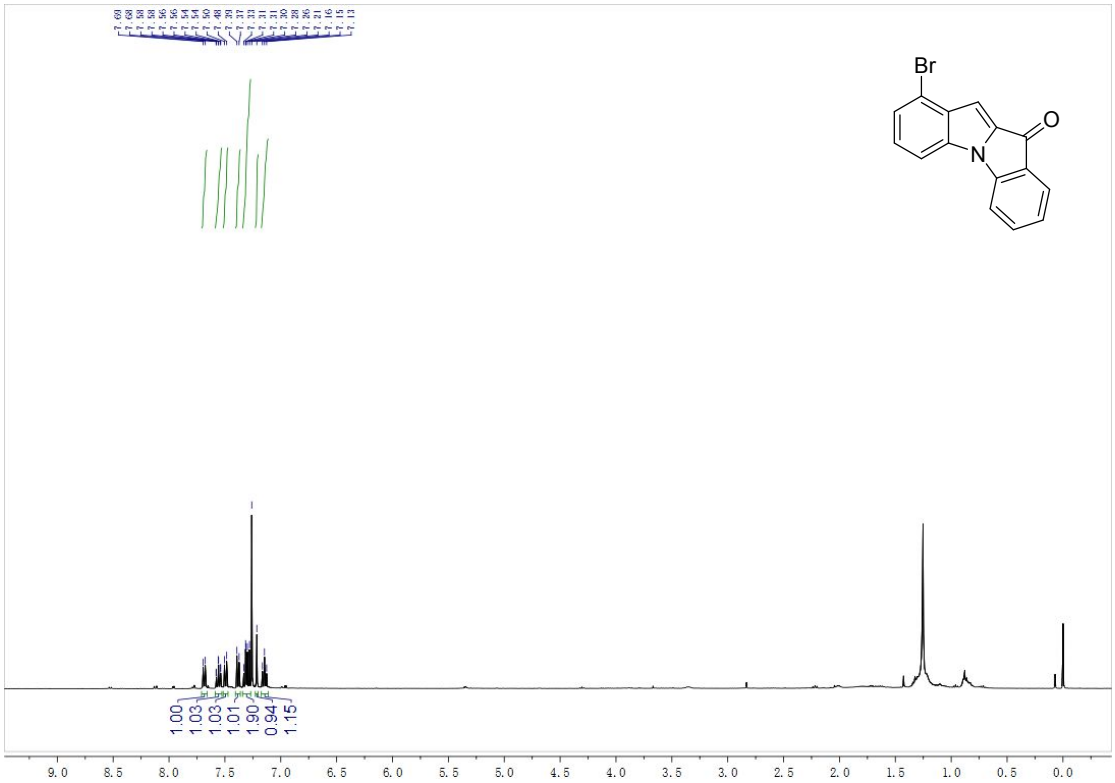


Figure S93. ^{1}H NMR for compound **4h**

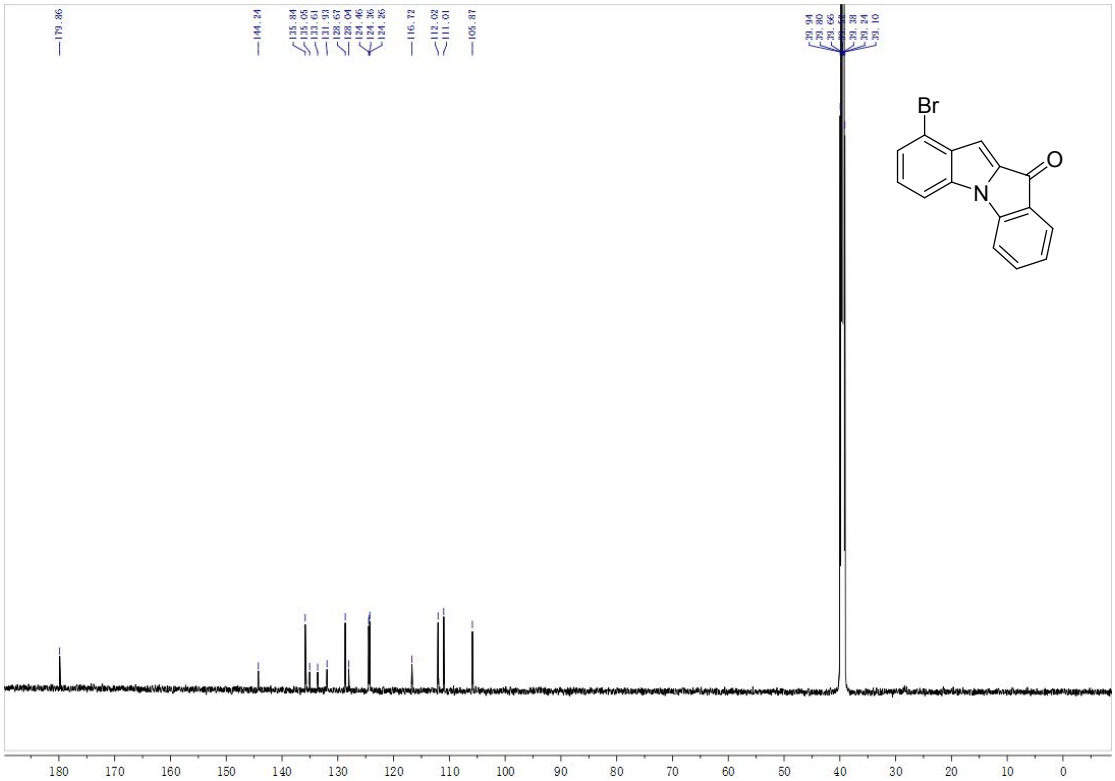


Figure S94. $^{13}C\{^{1}H\}$ NMR for compound **4h**

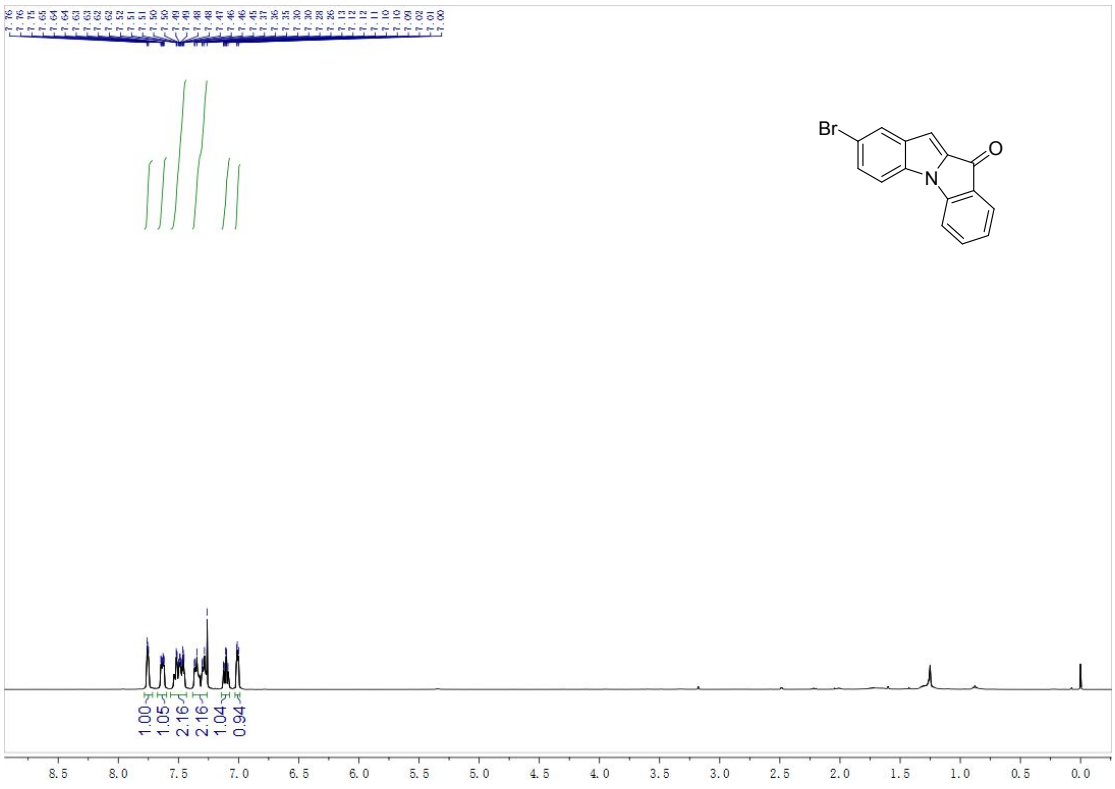


Figure S95. ^{1}H NMR for compound **4i**

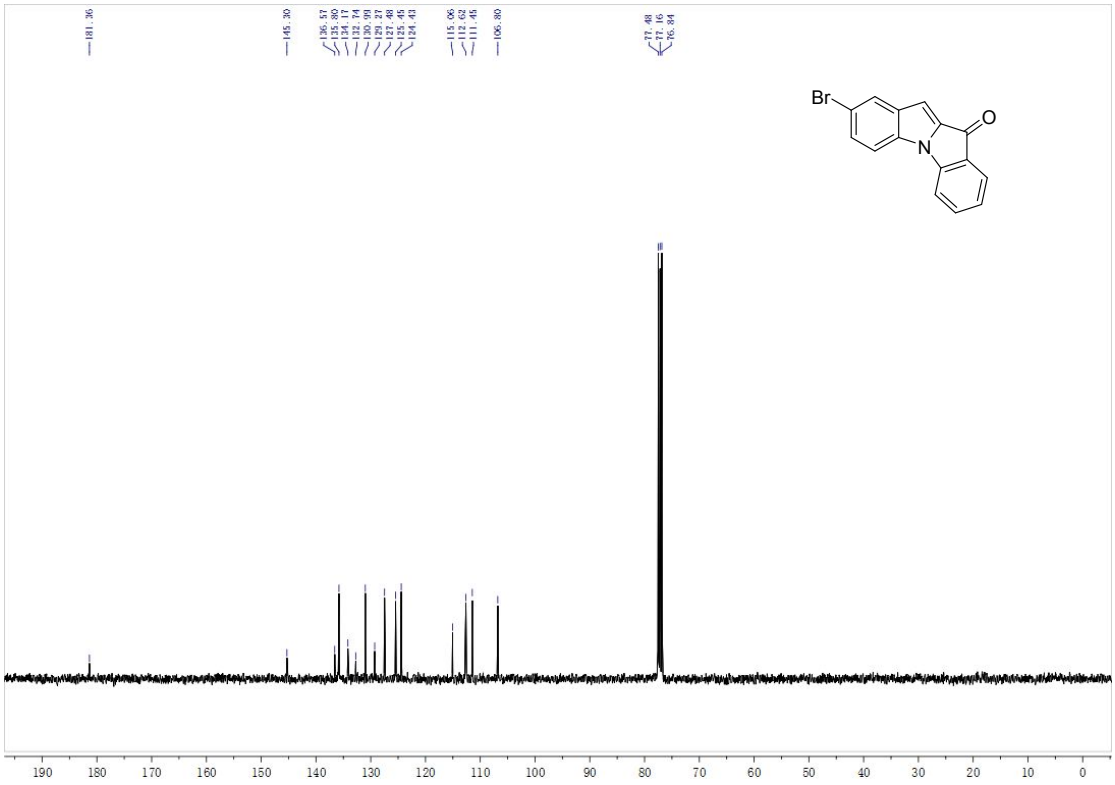


Figure S96. $^{13}C\{^{1}H\}$ NMR for compound **4i**

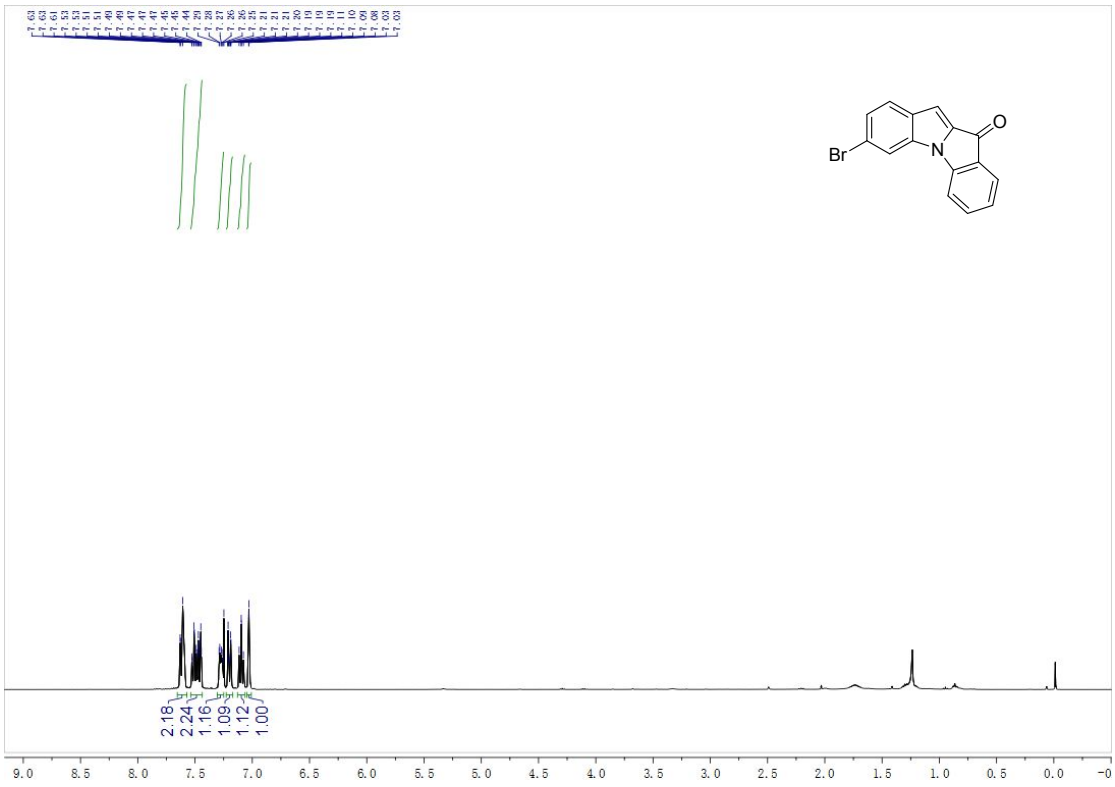


Figure S97. ^{1}H NMR for compound **4j**

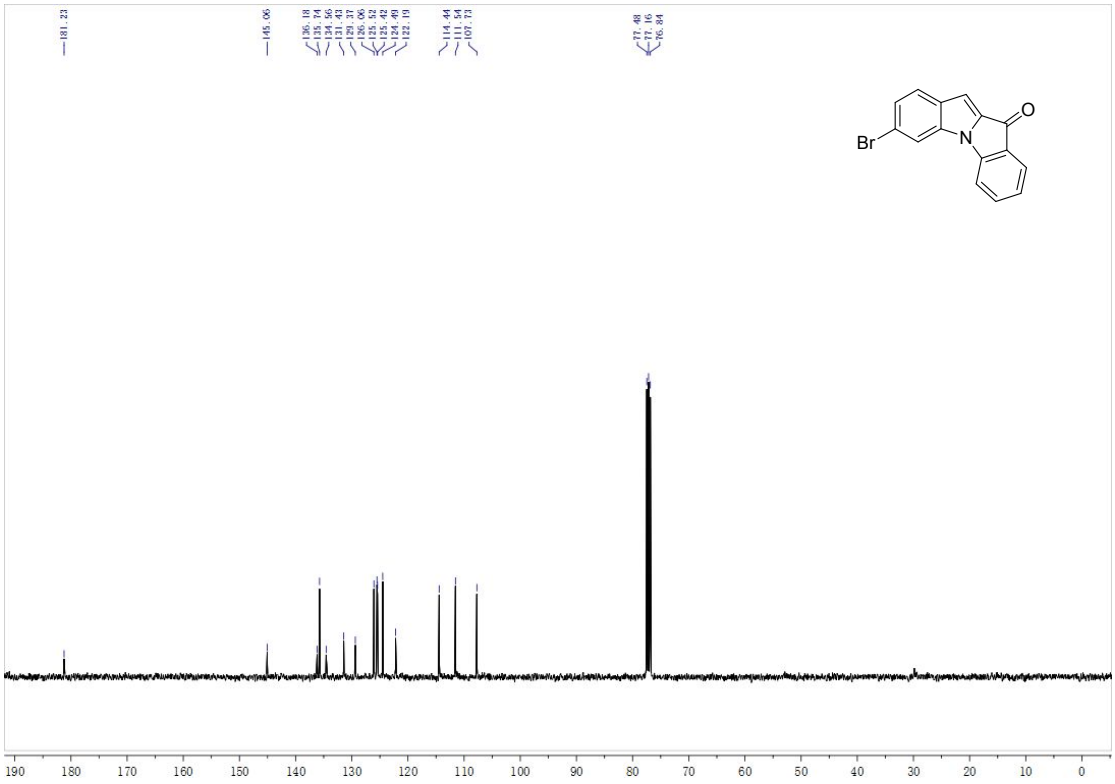


Figure S98. $^{13}C\{^{1}H\}$ NMR for compound **4j**

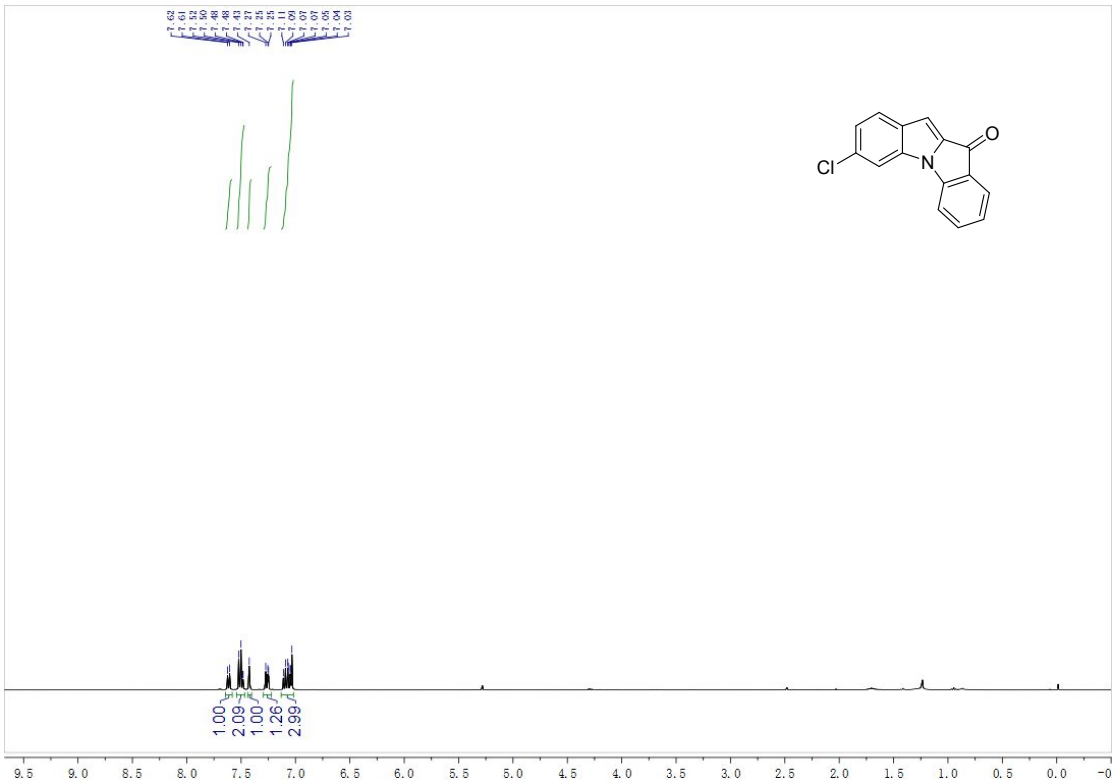


Figure S99. ^{1}H NMR for compound **4k**

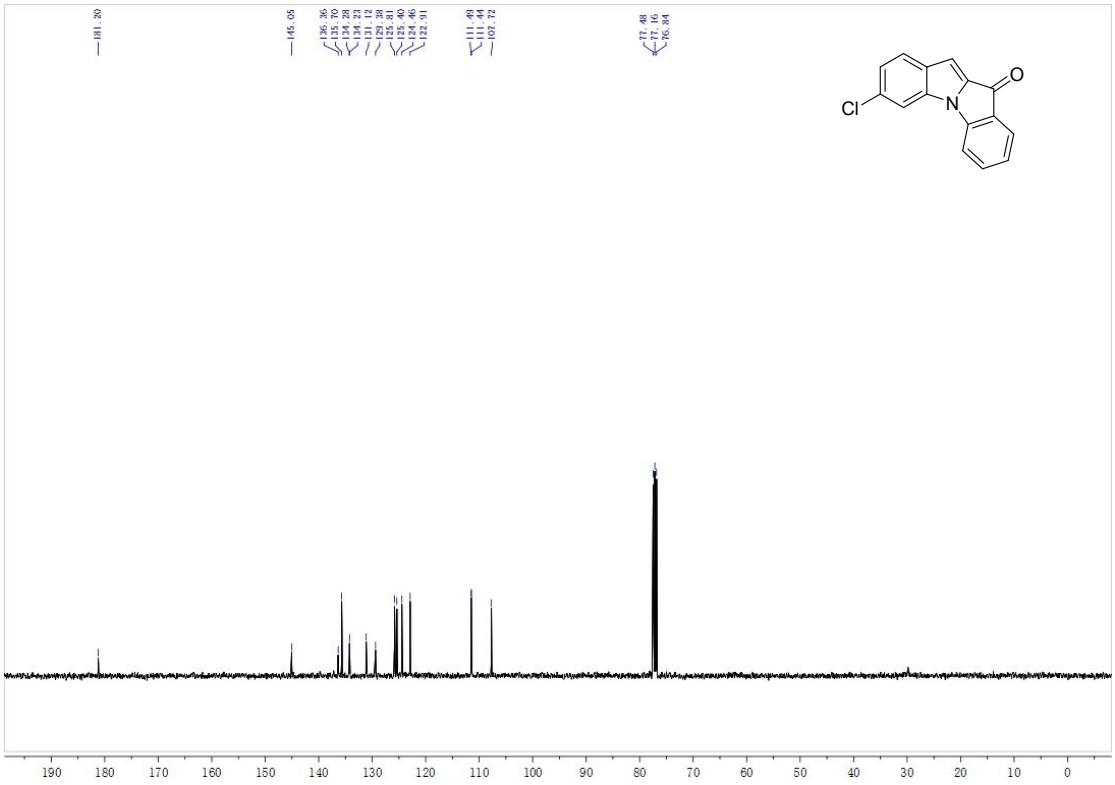


Figure S100. $^{13}C\{^{1}H\}$ NMR for compound **4k**

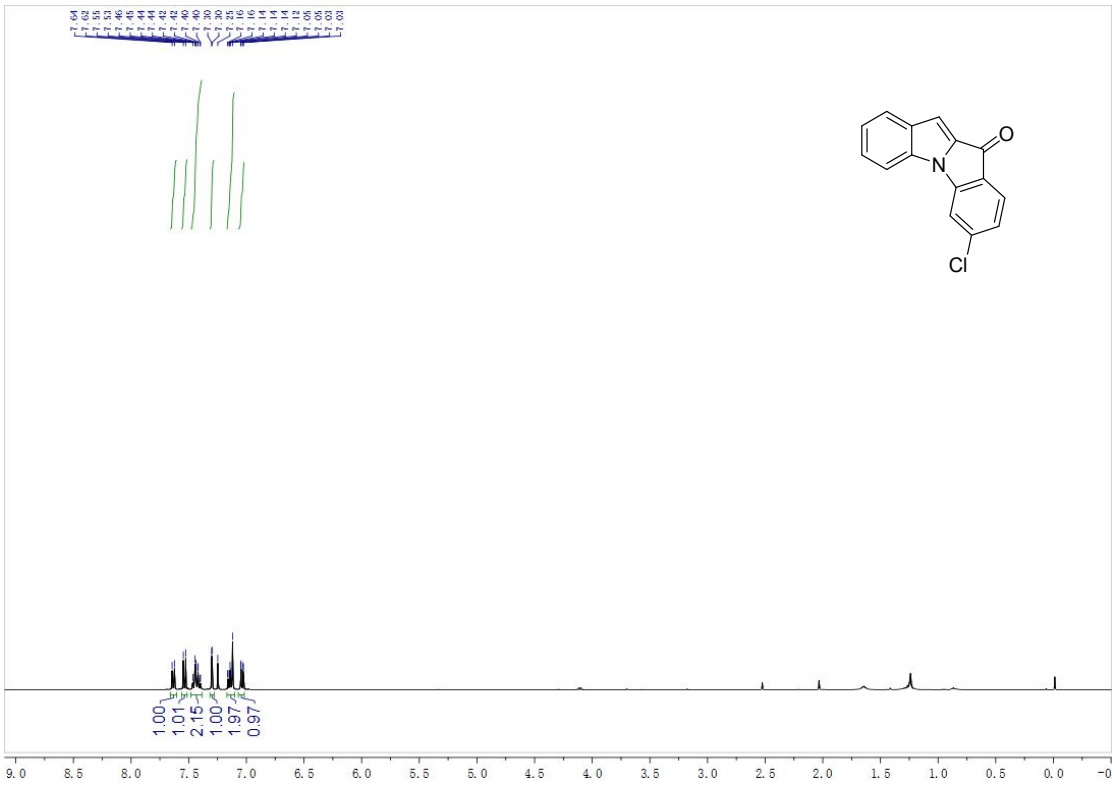


Figure S101. ^{1}H NMR for compound **4o**

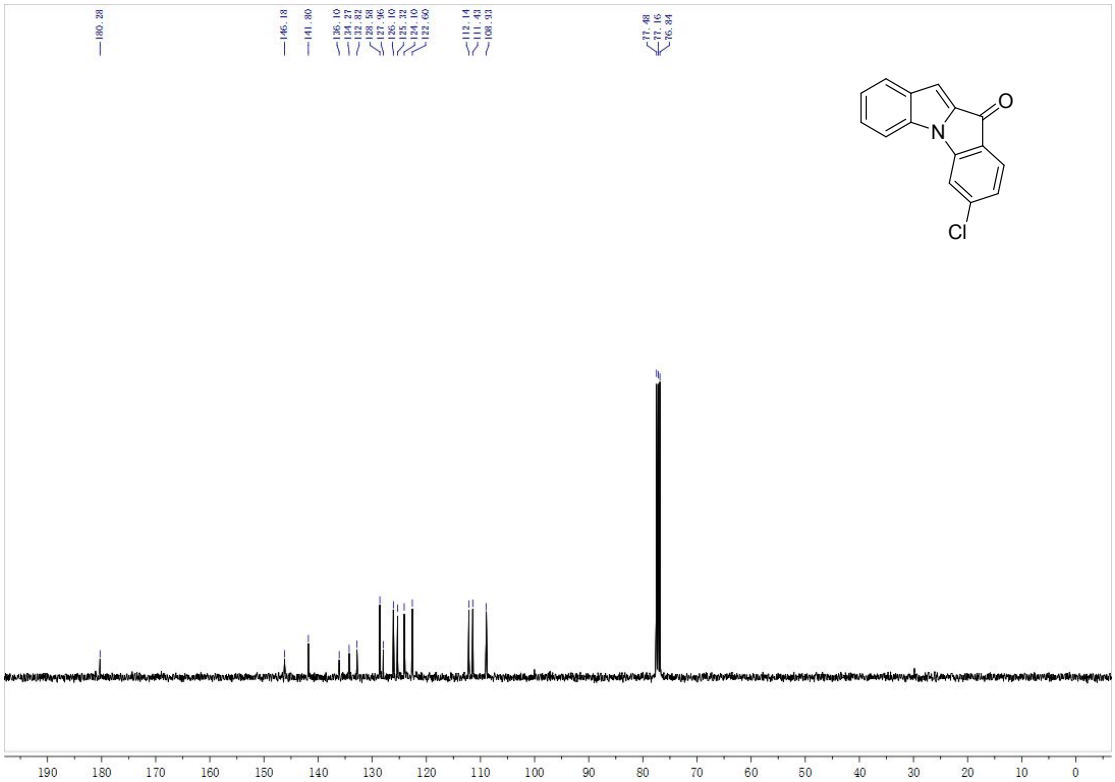


Figure S102. $^{13}C\{^{1}H\}$ NMR for compound **4o**

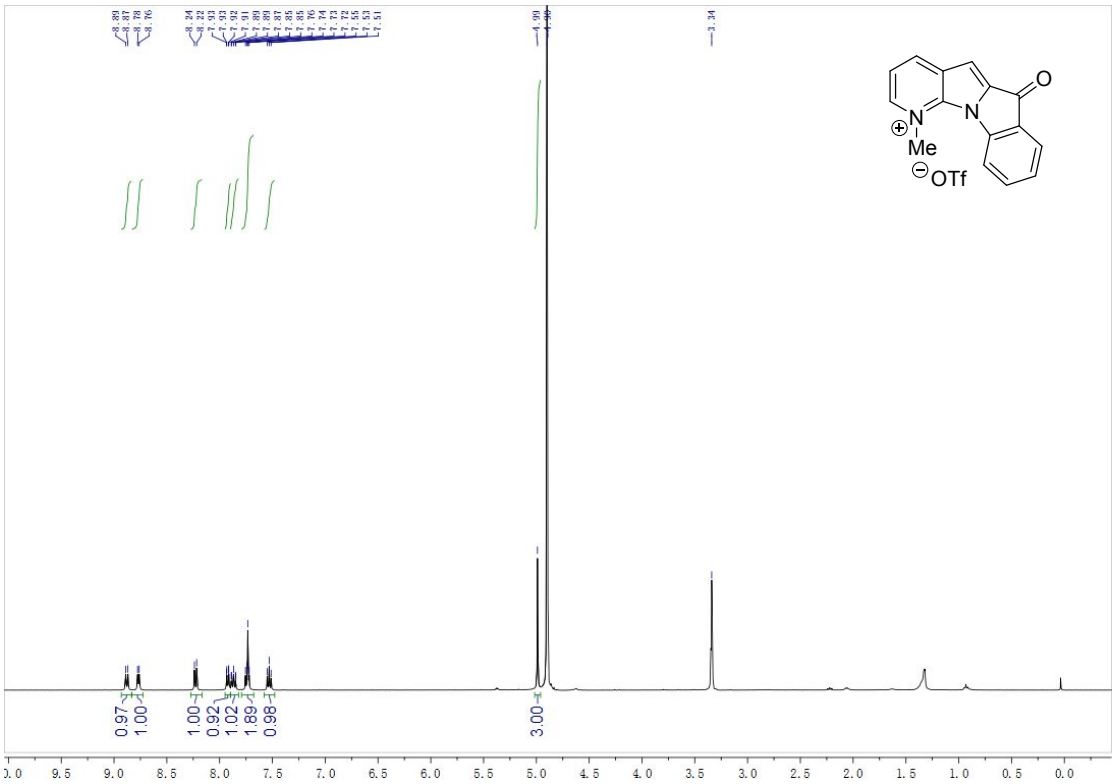


Figure S103. ^{1}H NMR for compound **4t**

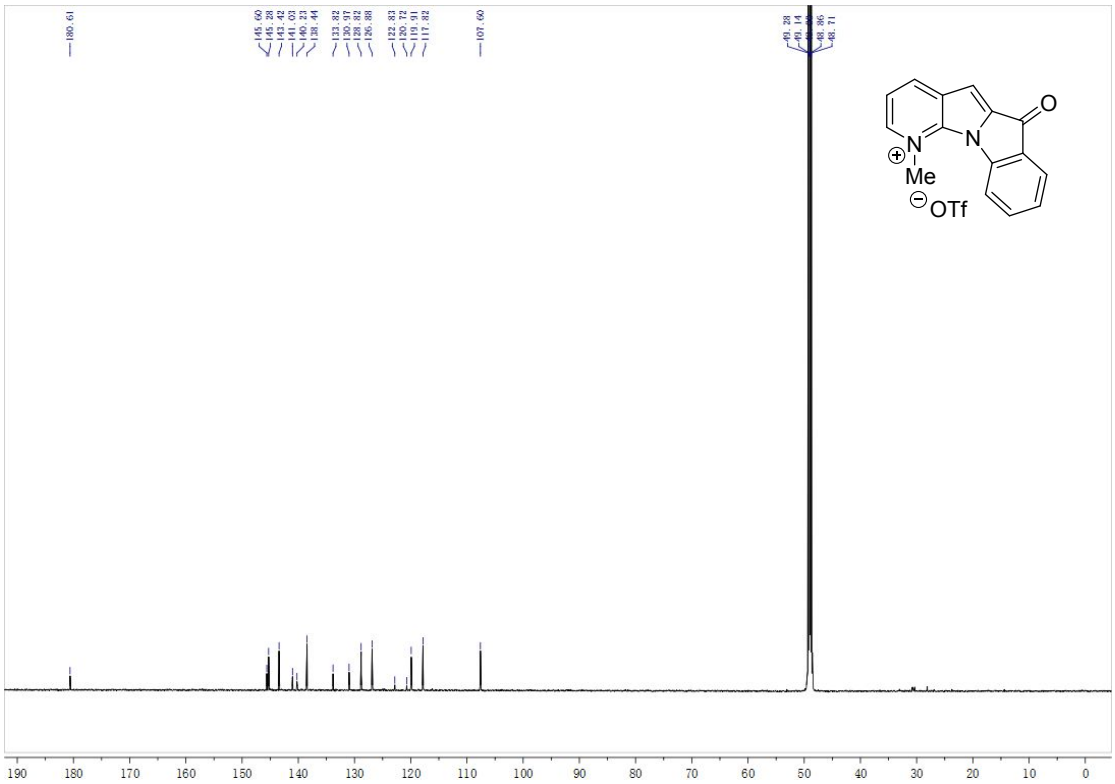


Figure S104. $^{13}C\{^{1}H\}$ NMR for compound **4t**

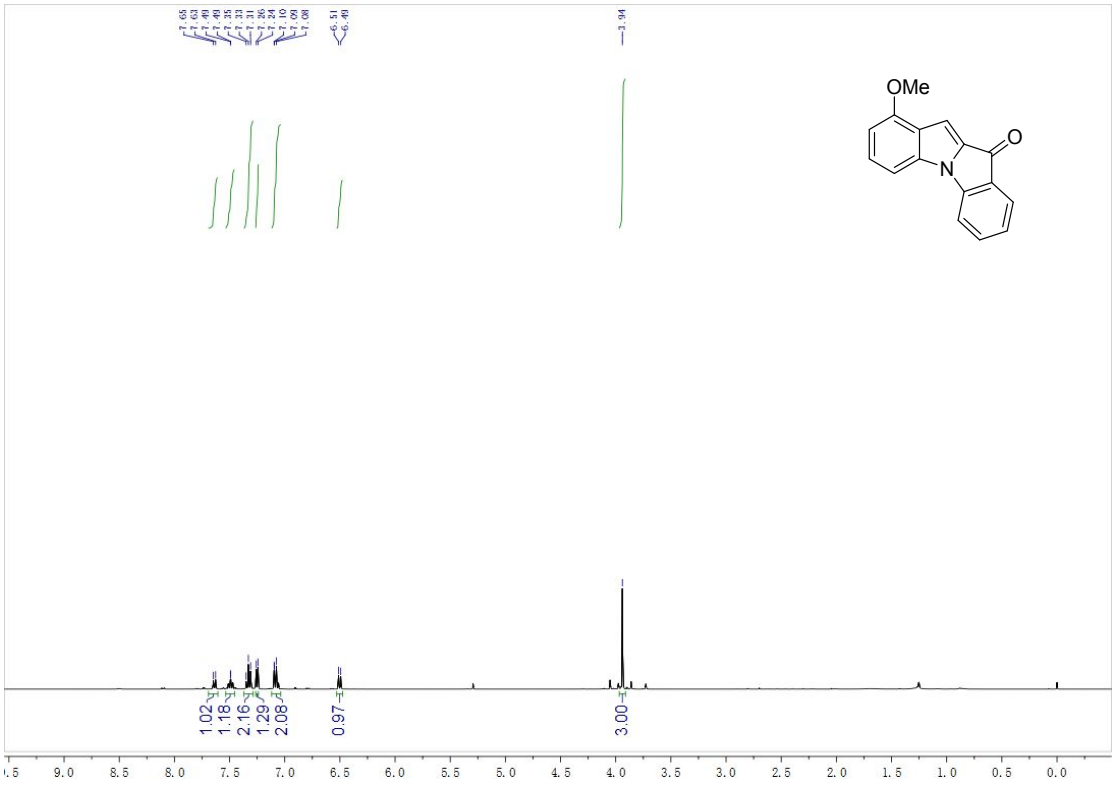


Figure S105. ^{1}H NMR for compound **4u**

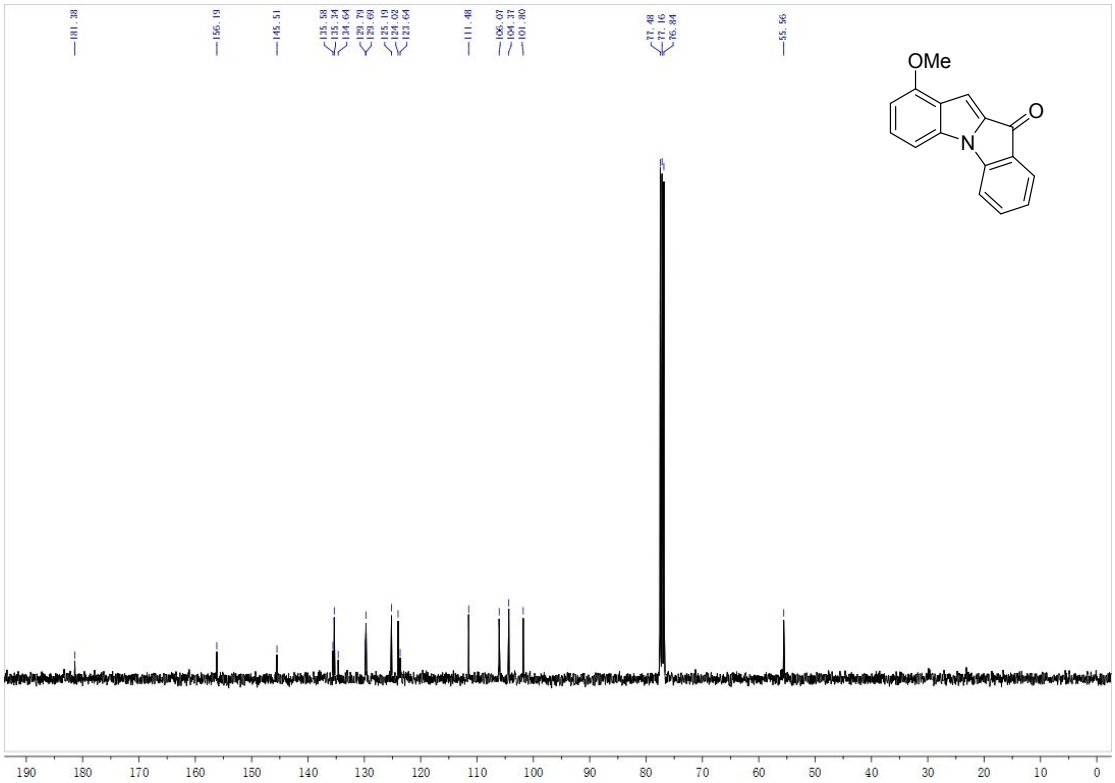


Figure S106. $^{13}C\{^{1}H\}$ NMR for compound **4u**