

## Supporting Information

# Efficient Ternary WORM Memory Devices from Quinoline-based D-A Systems by Varying the Redox Behavior of Ferrocene

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## 1. Materials and methods

5-Bromoisatin, ferrocene, 4-iodoaniline, urea, NaNO<sub>2</sub>, sulphuric acid, hexadecyltrimethylammonium bromide, diethyl ether, copper iodide, trimethylsilyl acetylene, tetrakis(triphenylphosphine)palladium, acetophenone, 4-fluoroacetophenone, 4-acetylanisole, dimethylformamide, tetrahydrofuran, triethylamine were all purchased from commercial sources and used as received unless otherwise mentioned. All the solvents used for analysis were of ACS grade. All the Pd-catalysed reactions were carried out under a nitrogen atmosphere. The reactions were monitored and optimized using thin-layer chromatography and the products were purified by column chromatography with a 100-200 silica mesh. <sup>1</sup>H and <sup>13</sup>C NMR spectra were recorded in Bruker 400 MHz spectrometer utilizing tetramethylsilane (TMS) as the internal standard. High-resolution mass spectra were attained from Thermo Exactive Plus UHPLC-MS. Absorption and emission spectra were recorded using the JASCO UV-NIR spectrophotometer and Perkin-Elmer LS 55 spectrophotometer, respectively. Electrochemical studies were operated in a CHI electrochemical workstation (CHI 6035D). Surface morphology was determined by a VEGA3 TESCAN scanning electron microscope (SEM). DFT and TD-DFT studies were used to attain the theoretical aspects.

### Fabrication and characterization of memory devices

Distilled water, soap solution, acetone, and ethanol were used for sonication for 10 minutes each to thoroughly clean the ITO-covered glass plates. The compounds (**14a-d**) were dissolved in chloroform solution (5 mg mL<sup>-1</sup>) and spin-coated over the ITO-coated glass plate. After that, the thin film was annealed for 20 minutes at 80 °C. The device was then coated with silver contacts via sputtering using a mask of 1 mm dimension. The device was then utilized for the study of memory characteristics. Keithley 4200A semiconductor parameter analyzer was used to perform memory characterizations at ambient conditions.<sup>1</sup>

## 2. Synthesis and analytical data of compounds

Compounds **1a-c**, **2a-c**, and **3a-c** are synthesized as per the reported literature.<sup>2</sup>

**Compound 4:** In a 100 mL round bottom flask, iodoaniline (7.00 g, 0.03 mmol) was taken, and added 15 mL conc. H<sub>2</sub>SO<sub>4</sub> in 30 mL of water and stirred under ice-cold conditions. To this stirring mixture, NaNO<sub>2</sub> (2.20 g, 0.03 mmol) dissolved in water was added. This solution was closed and allowed to stir for 1 hour. To this reaction mixture, urea (48 mg, 0.008 mmol) was added, followed by the addition of ferrocene (3 g, 0.016 mmol), and hexadecyl trimethylammonium bromide (300 mg) in diethyl ether was added dropwise for 1 hour. After 1 hour, the ice-cold condition was removed and stirred the reaction for half an hour at room temperature. The reaction was stopped after completion, and the solvent was removed under reduced pressure. Compound **4** was separated through column chromatography as a yellow solid (30 %). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 7.59 (d, J = 8.4 Hz, 2H), 7.21 (d, J = 8.0 Hz, 2H), 4.61 (s, 2H), 4.33 (s, 2H), 4.03 (s, 5H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 139.28, 137.44, 128.42, 127.98, 126.24, 126.02, 90.68, 85.53, 84.07, 69.79, 69.69, 69.35, 68.99, 66.60, 66.49.

**Compound 5:** Compound **4** (1 g, 0.0025 mmol) was taken in a 100 mL round bottom flask, dissolved in an equal amount of tetrahydrofuran and triethylamine, and purged with nitrogen gas. To this mixture, Pd(PPh<sub>3</sub>)<sub>2</sub>Cl<sub>2</sub> (87.7 mg, 0.0001 mmol) and CuI (23.8 mg, 0.0001 mmol) were added and stirred. Trimethylsilylacetylene (736 mg, 0.0075 mmol) was added to the reaction mixture and stirred for 5 hours at 70 °C while maintaining the nitrogen atmosphere. The product was purified through column chromatography in hexane solvent. The obtained product was dissolved in methanol, and potassium carbonate was added to get compound **5** as a brown solid (70 %). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 7.40 (s, 4H), 4.62- 4.63 (m, 2H), 4.35 – 4.31 (m, 2H), 4.01 (s, 5H), 3.10 (s, 1H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 140.53, 132.21, 125.82, 119.17, 84.06, 69.77, 69.45, 66.60.

### General procedure for Suzuki coupling reaction for compounds **6a-c**

Iodophenylferrocene (100 mg, 0.257 mmol) and the corresponding boronic acid (**3a-c**) are dissolved in tetrahydrofuran (10 mL) in a 100 mL round bottom flask and purged with nitrogen for 15 minutes. 2 M Potassium carbonate in 5 mL of water is then added to the solution followed by the addition of tetrakis(triphenylphosphine) palladium (0.05 eq) under nitrogen atmosphere and stirred the reaction mixture for 12 hours. After the completion of the reaction, the solvent was removed under a vacuum, and the product was separated using column chromatography in a hexane: dichloromethane solvent mixture.

**Compound 6a:** Compounds **4** and **3a** (220.08 mg, 0.565 mmol) underwent the reaction as per the general procedure for Suzuki reaction to get compound **6a** as a red solid (71 %). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 9.05 (s, 1H), 8.45 (s, 1H), 8.29 (d, J = 8.8 Hz, 1H), 8.23 (d, J = 7.6 Hz, 2H), 8.08 (d, J = 8.8 Hz, 1H), 7.72 (d, J = 8.0 Hz, 2H), 7.62 – 7.49 (m, 5H), 4.72 (s, 2H), 4.37 (s, 2H), 4.10 (d, J = 5.2 Hz, 8H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 166.91, 156.39, 148.68, 140.19, 139.30, 138.82, 137.74, 135.32, 130.72, 129.74, 129.36, 129.00, 127.50, 127.44, 126.62, 124.41, 122.66, 120.79, 84.66, 69.72, 69.20, 66.59, 52.81. HRMS (ESI) (m/z): C<sub>33</sub>H<sub>25</sub>FeNO<sub>2</sub>, Calc [M+H]<sup>+</sup>: 524.1307, Observed [M+H]<sup>+</sup>: 524.1306.

**Compound 6b:** Compounds **4** and **3b** (230.24 mg, 0.565 mmol) underwent the reaction as per the general procedure for Suzuki reaction to get compound **6b** as a brown solid (73 %). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.96 (s, 1H), 8.31 (s, 1H), 8.20 – 8.13 (m, 3H), 8.02 – 7.98 (m, 1H), 7.64 (d, J = 8.4 Hz, 2H), 7.53 (d, J = 8.0 Hz, 2H), 7.19 – 7.14 (m, 2H), 4.64 (s, 2H), 4.29 (s, 2H), 4.02 (d, J = 4.8 Hz, 7H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 166.80, 165.27, 162.79, 155.23, 148.61, 140.24, 139.36, 137.67, 135.44, 134.95, 130.61, 129.46, 129.38, 129.29, 127.48, 126.62, 125.04, 124.30, 122.65, 120.37, 115.98, 115.85, 84.62, 69.72, 69.21, 66.59, 52.82. HRMS (ESI) (m/z): C<sub>33</sub>H<sub>24</sub>FFeNO<sub>2</sub>, Calc [M+H]<sup>+</sup>: 542.1213, Observed [M+H]<sup>+</sup>: 524.1213.

**Compound 6c:** Compounds **4** and **3c** (236.89 mg, 0.565 mmol) underwent the reaction as per the general procedure for the Suzuki reaction to get compound **6c** as a reddish brown solid (68 %). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 9.01 (s, 1H), 8.39 (s, 1H), 8.24-8.18 (m, 3H), 8.06-8.03 (m, 1H), 7.70 (d, J = 8.0 Hz, 2H), 7.59 (d, J = 8.4 Hz, 2H), 7.07 (d, J = 8.4 Hz, 2H), 4.71 (s, 2H), 4.36 (s, 2H), 4.08 (s, 8H), 3.90 (s, 3H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 166.98, 161.13, 155.94, 148.67, 139.73, 139.18, 137.82, 135.19, 131.39, 130.48, 129.23, 128.82, 127.45, 126.60, 124.06, 122.66, 120.35, 114.36, 84.69, 69.72, 69.18, 66.58, 55.46, 53.46, 52.77. HRMS (ESI) (m/z): C<sub>34</sub>H<sub>27</sub>FeNO<sub>3</sub>, Calc [M+H]<sup>+</sup>: 554.1413, Observed [M+H]<sup>+</sup>: 554.1418.

#### **General procedure for Sonagashira coupling reaction for compounds 7a-c**

Compound **5** and the corresponding brominated quinoline (**2a-c**) were dissolved in tetrahydrofuran (10 mL) and purged with nitrogen for 15 minutes. Triethylamine (2.5 mL) was then added and again purged with nitrogen for ten more minutes. Finally, Pd(PPh<sub>3</sub>)<sub>4</sub> (0.1 eq) was added under a nitrogen atmosphere and stirred the reaction mixture for 16 hours under reflux conditions. The reaction completion was checked by using thin-layer chromatography, and the solvent was removed under a vacuum after the completion of the reaction. The products

were separated by column chromatography using a hexane: dichloromethane solvent mixture as a pure compound.

**Compound 7a:** Compounds **2a** (100 mg, 0.29 mmol) and **5** (167.61 mg, 0.586 mmol) underwent the reaction as per the general procedure for Sonagashira coupling to get compound **7a** as a brown solid (66 %). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.97 (s, 1H), 8.44 (s, 1H), 8.22 - 7.17 (m, 3H), 7.88 (d, J = 8.8 Hz, 1H), 7.56 - 7.47 (m, 7H), 4.69 (s, 2H), 4.37 (s, 2H), 4.11 (s, 3H), 4.06 (s, 5H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 166.63, 157.02, 148.61, 140.38, 138.53, 135.01, 132.69, 131.83, 130.36, 129.97, 129.02, 128.69, 127.48, 125.91, 123.90, 123.15, 121.00, 120.02, 91.83, 89.67, 84.14, 69.77, 69.46, 66.58, 53.46, 52.92. HRMS (ESI) (m/z): C<sub>35</sub>H<sub>25</sub>FeNO<sub>2</sub>, Calc [M+H]<sup>+</sup>: 548.1307, Observed [M+H]<sup>+</sup>: 548.1307.

**Compound 7b:** Compounds **2b** (100 mg, 0.27 mmol) and **5** (158.44 mg, 0.55 mmol) underwent the reaction as per the general procedure for Sonagashira coupling to get compound **7b** as a red solid (64 %). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.96 (s, 1H), 8.39 (s, 1H), 8.22 (dd, J = 8.0, 5.6 Hz, 2H), 8.16 (d, J = 8.8 Hz, 1H), 7.88 (d, J = 8.8 Hz, 1H), 7.50 (d, J = 6.0 Hz, 4H), 7.23 (d, J = 8.4 Hz, 1H), 4.69 (s, 2H), 4.37 (s, 2H), 4.11 (s, 3H), 4.06 (s, 5H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 166.54, 155.86, 148.55, 140.42, 135.14, 134.67, 132.81, 131.83, 130.27, 129.45, 129.36, 128.68, 125.91, 123.80, 123.21, 120.59, 119.98, 116.12, 115.91, 91.91, 89.60, 84.12, 69.77, 69.47, 66.58, 52.95. HRMS (ESI) (m/z): C<sub>35</sub>H<sub>24</sub>FFeNO<sub>2</sub>, Calc [M+H]<sup>+</sup>: 566.1208, Observed [M+H]<sup>+</sup>: 566.1211.

**Compound 7c:** Compounds **2c** (100 mg, 0.26 mmol) and **5** (153.31 mg, 0.53 mmol) underwent the reaction as per the general procedure for Sonagashira coupling to get compound **7c** as a brownish-yellow solid (60 %). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.86 (s, 1H), 8.31 (s, 1H), 8.11 (d, J = 8.8 Hz, 2H), 8.06 (d, J = 8.8 Hz, 1H), 7.77 (d, J = 8.4 Hz, 1H), 7.44 - 7.39 (m, 4H), 6.98 (d, J = 8.8 Hz, 2H), 4.61 (s, 2H), 4.29 (s, 2H), 4.02 (s, 3H), 3.98 (s, 5H), 3.82 (s, 3H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 166.71, 161.31, 156.54, 148.62, 140.30, 134.86, 132.60, 131.81, 131.08, 130.14, 128.88, 128.70, 125.90, 123.55, 122.63, 120.55, 120.10, 114.38, 91.59, 89.76, 84.16, 69.77, 69.45, 66.58, 55.46, 52.88. HRMS (ESI) (m/z): C<sub>36</sub>H<sub>27</sub>FeNO<sub>3</sub>, Calc [M+H]<sup>+</sup>: 578.1413, Observed [M+H]<sup>+</sup>: 578.1414.

### 3. $^1\text{H}$ , $^{13}\text{C}$ NMR, HRMS Spectra of the synthesized compounds

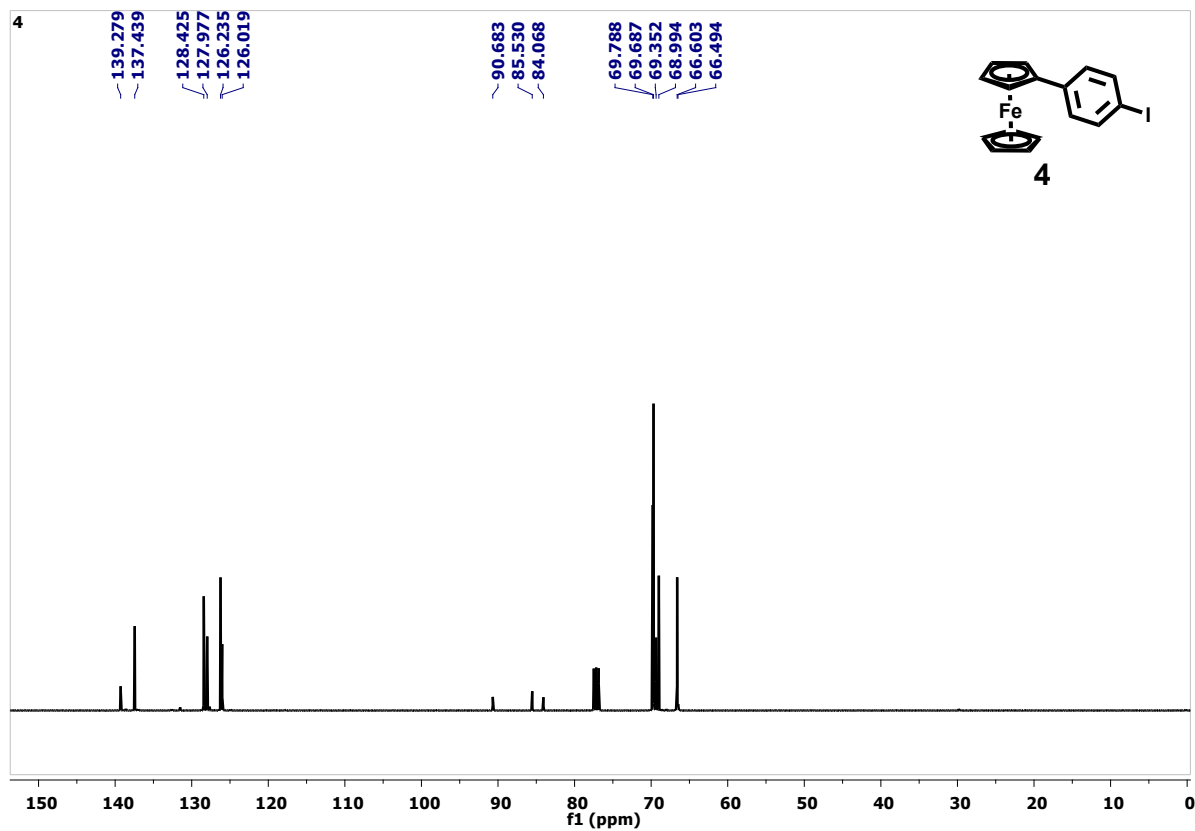
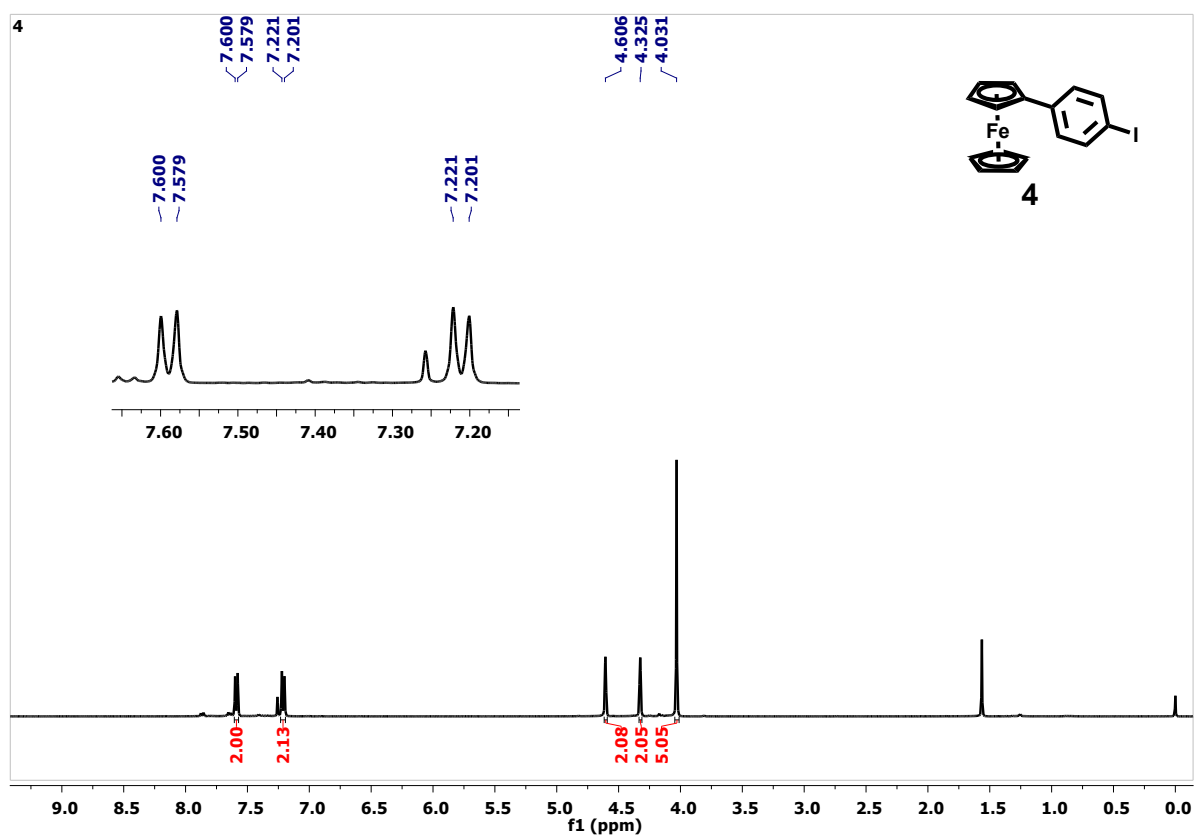


Figure S1:  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of compound 4

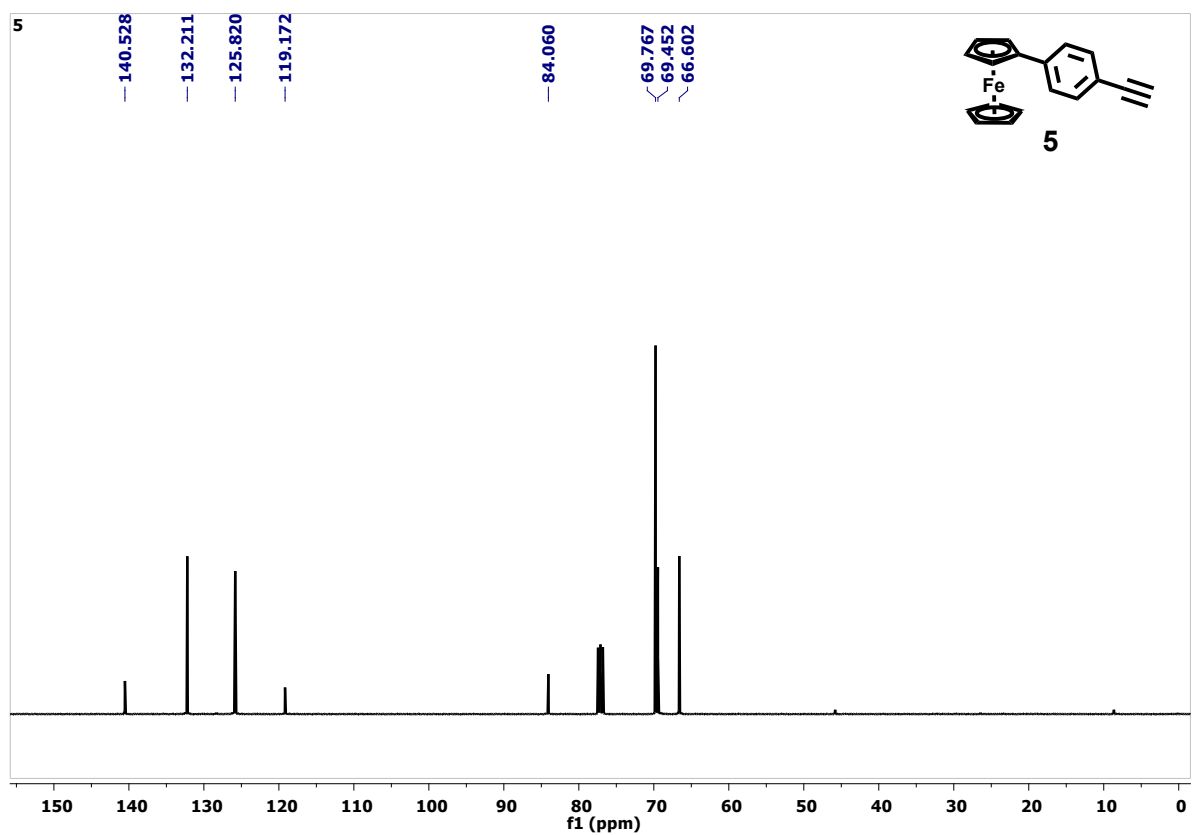
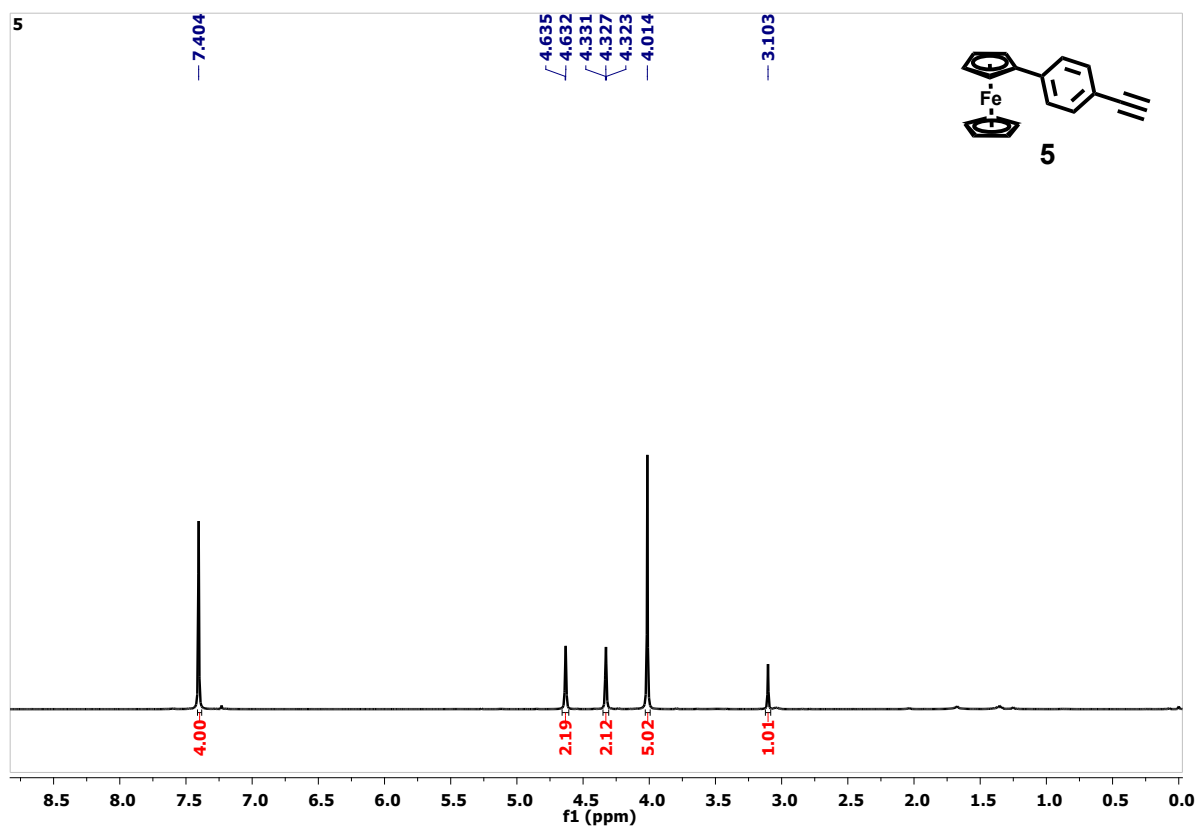
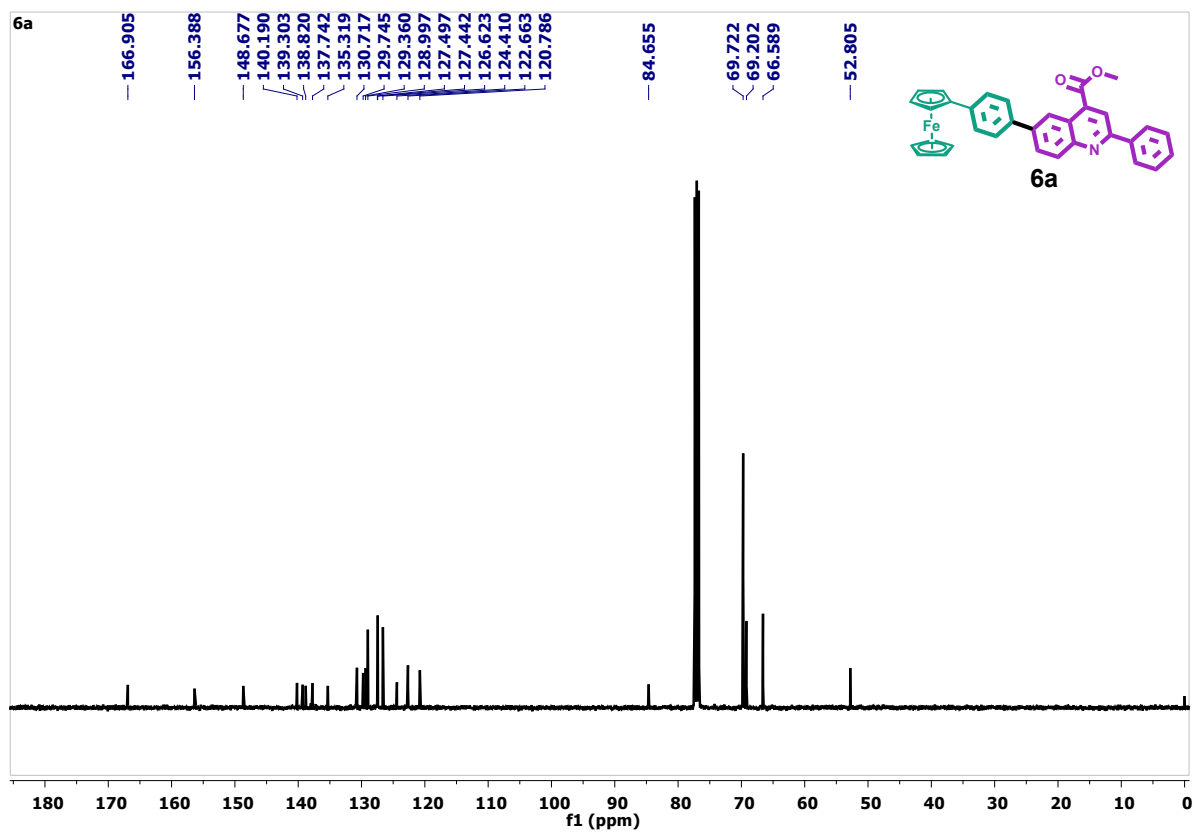
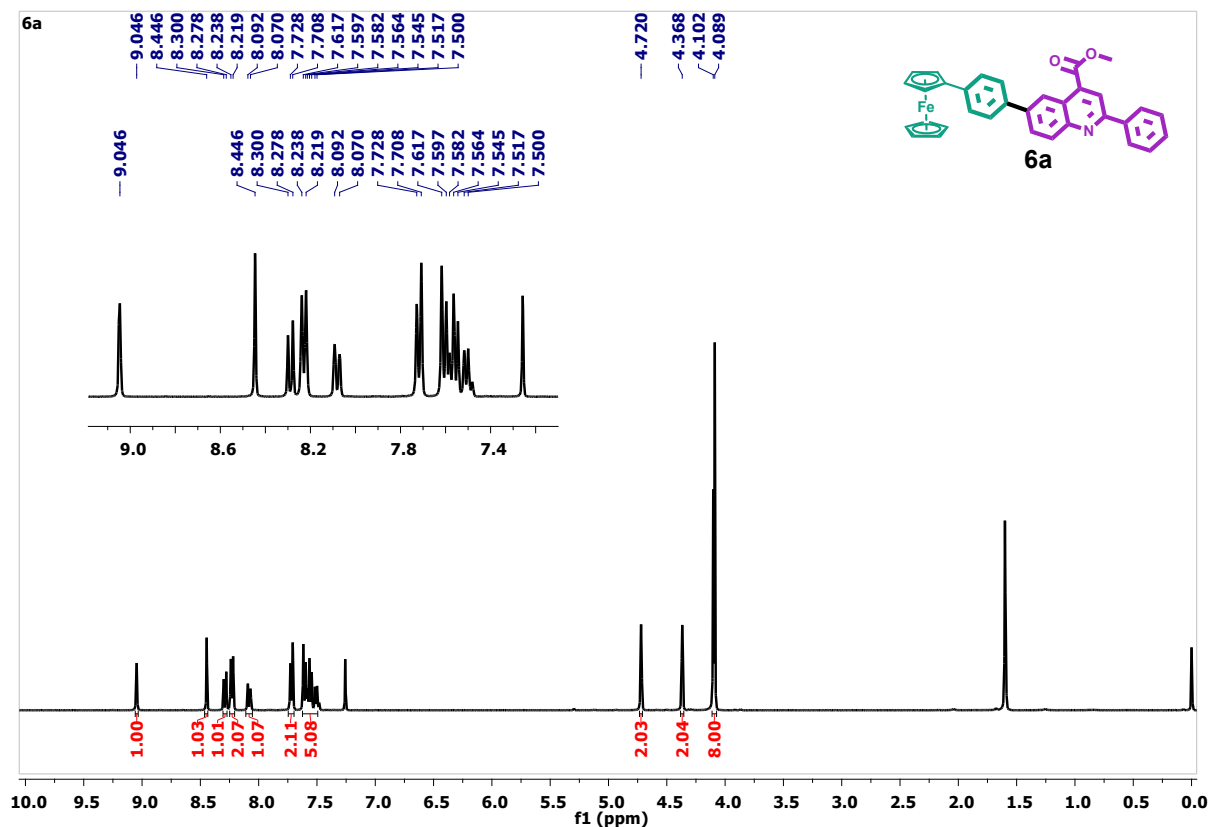


Figure S2:  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of compound 5





**Figure S3:**  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of compound **6a**

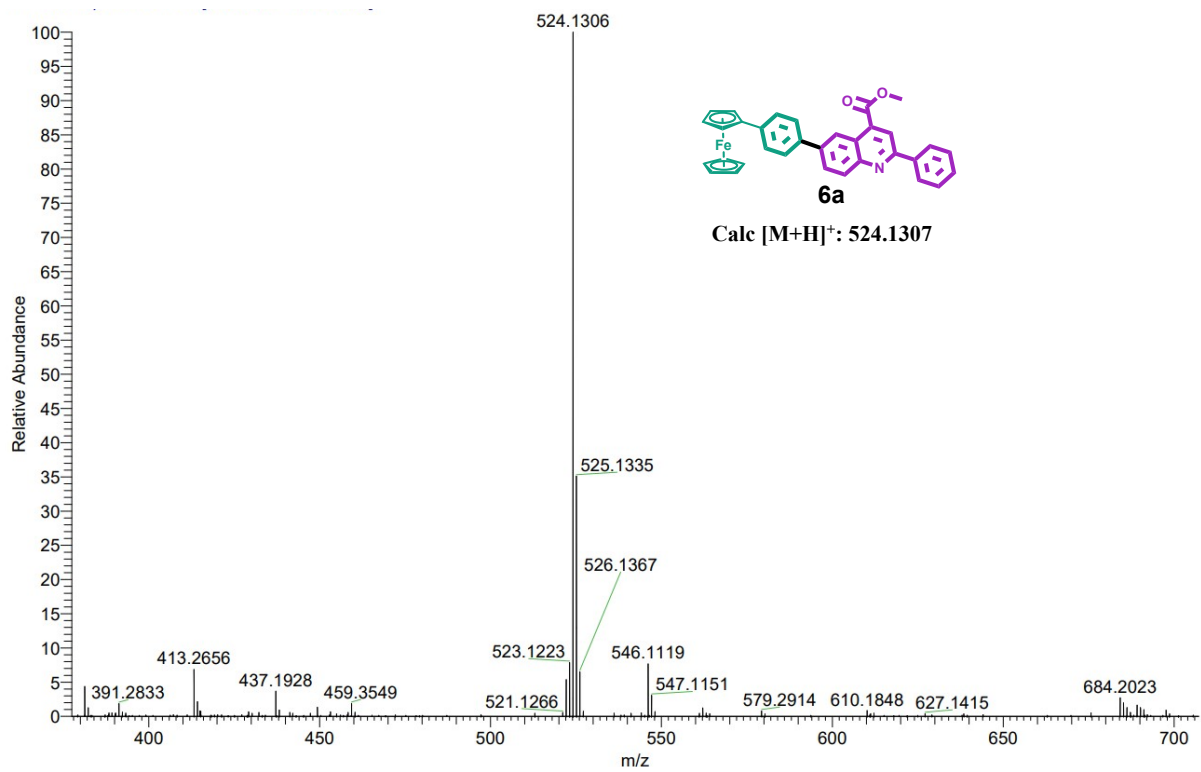
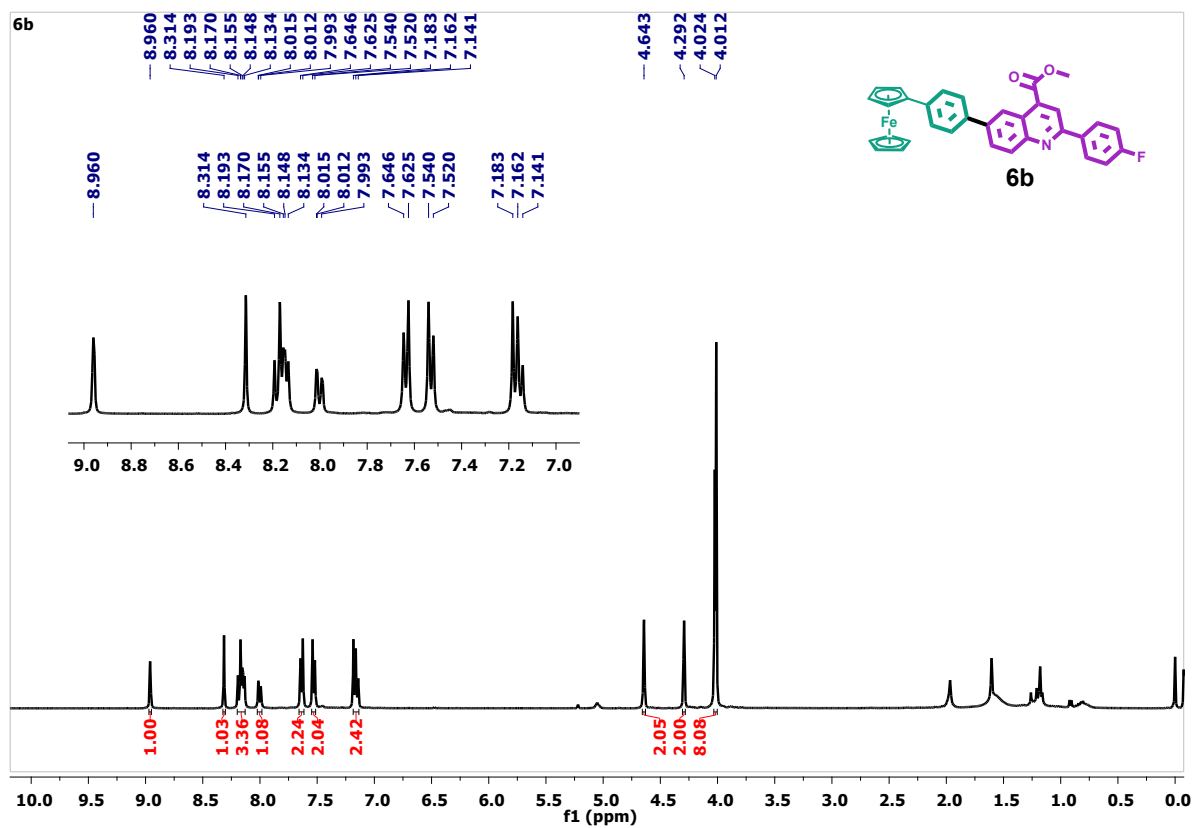
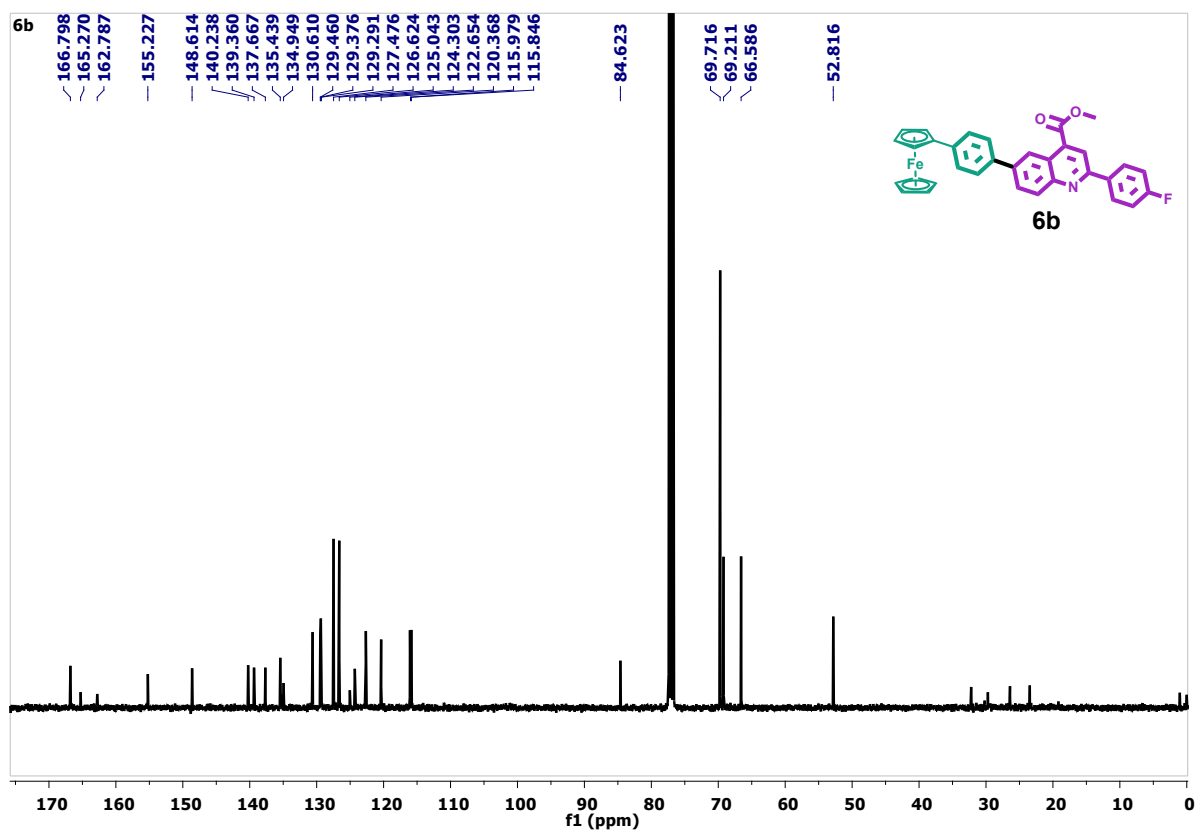
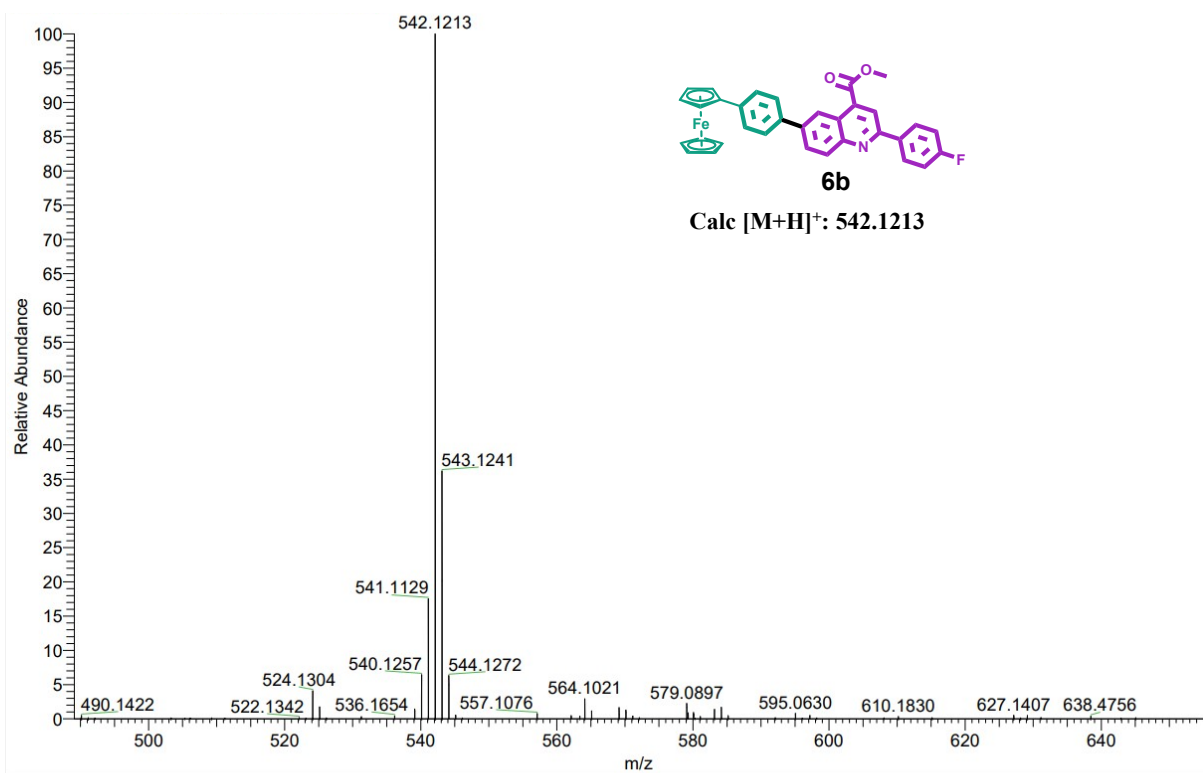


Figure S4: HRMS spectrum of compound **6a**





**Figure S5:**  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of compound **6b**



**Figure S6:** HRMS spectrum of compound **6b**

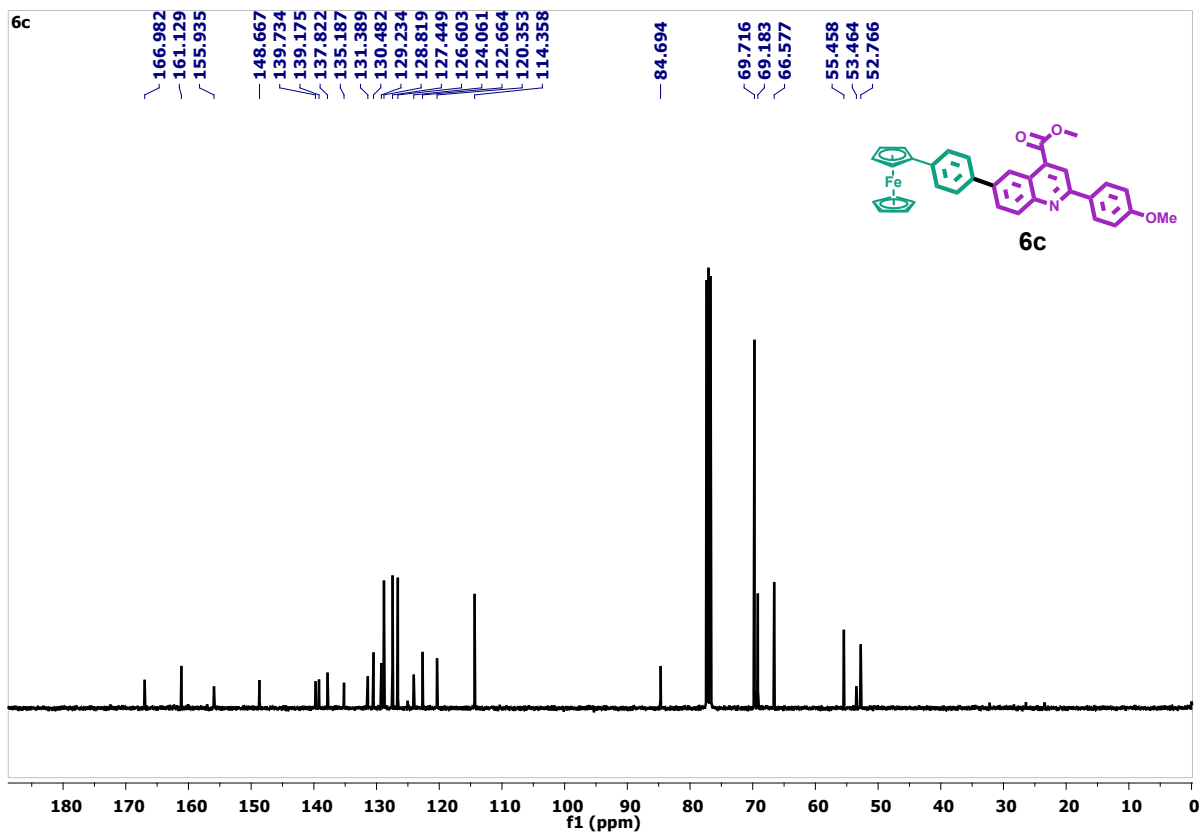
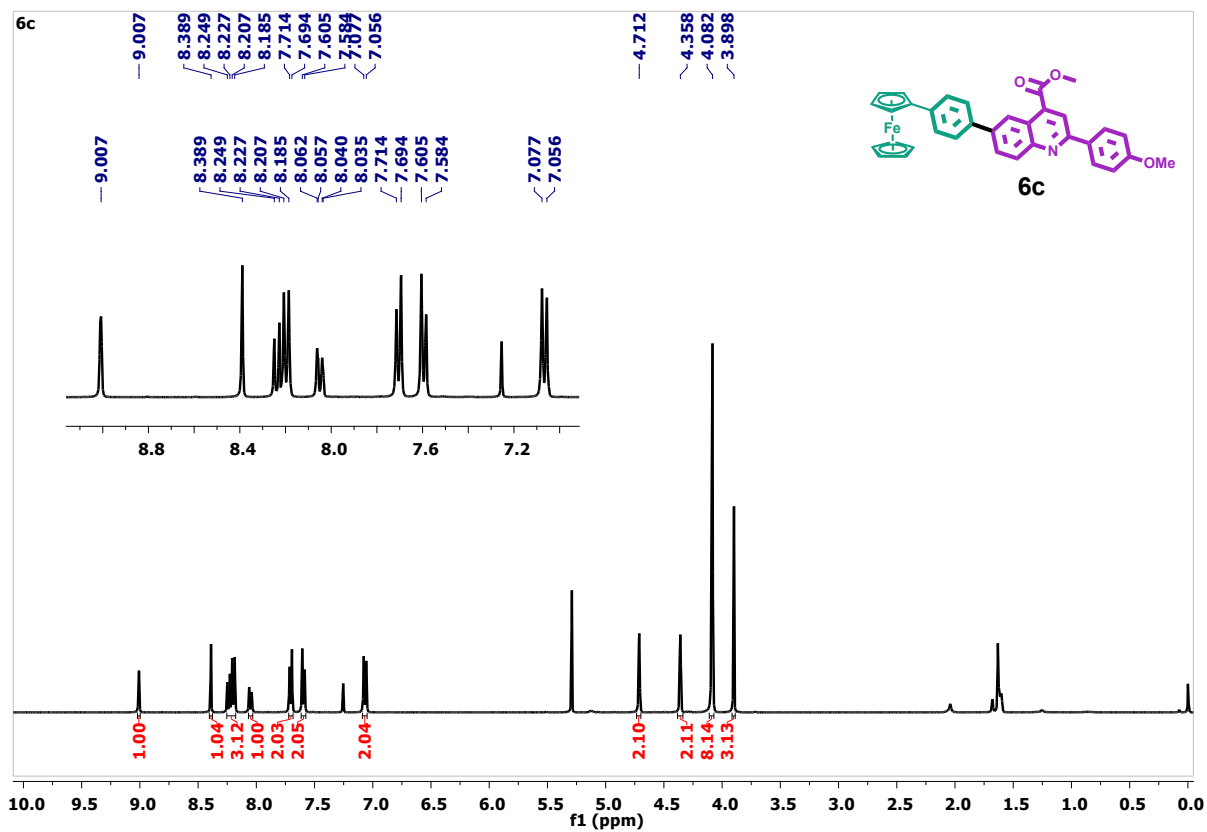


Figure S7:  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of compound **6c**

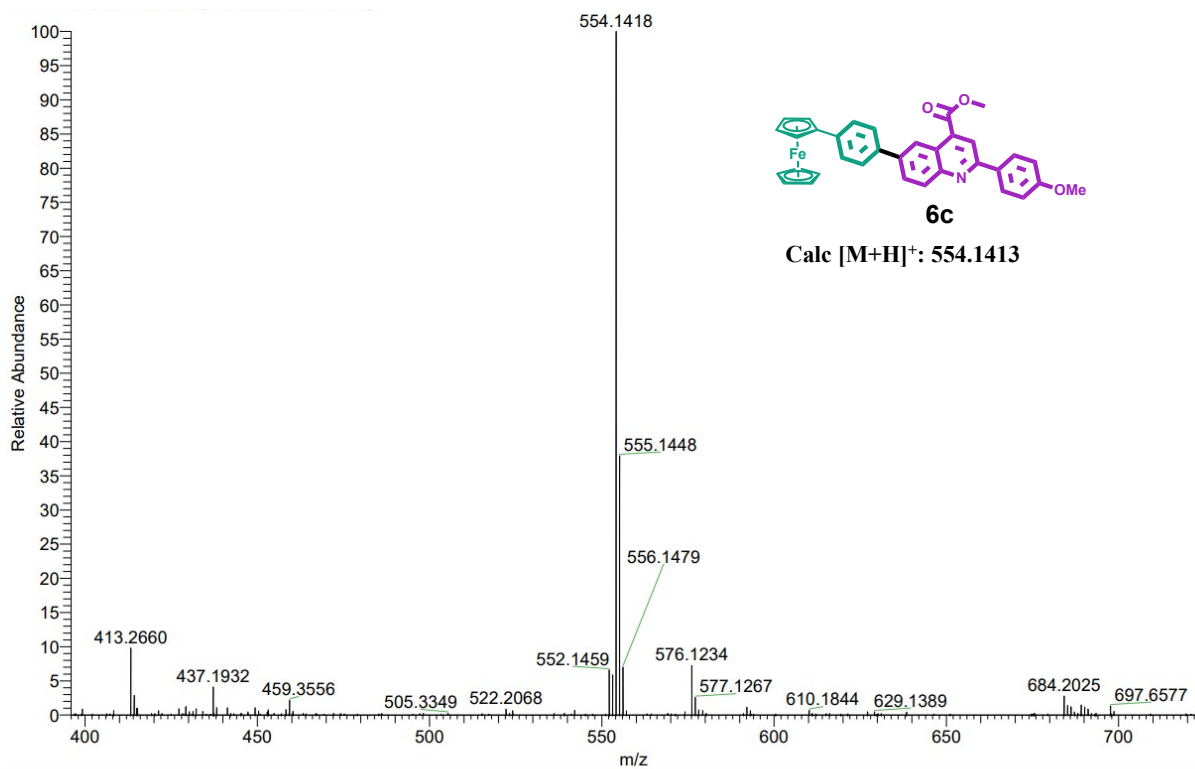
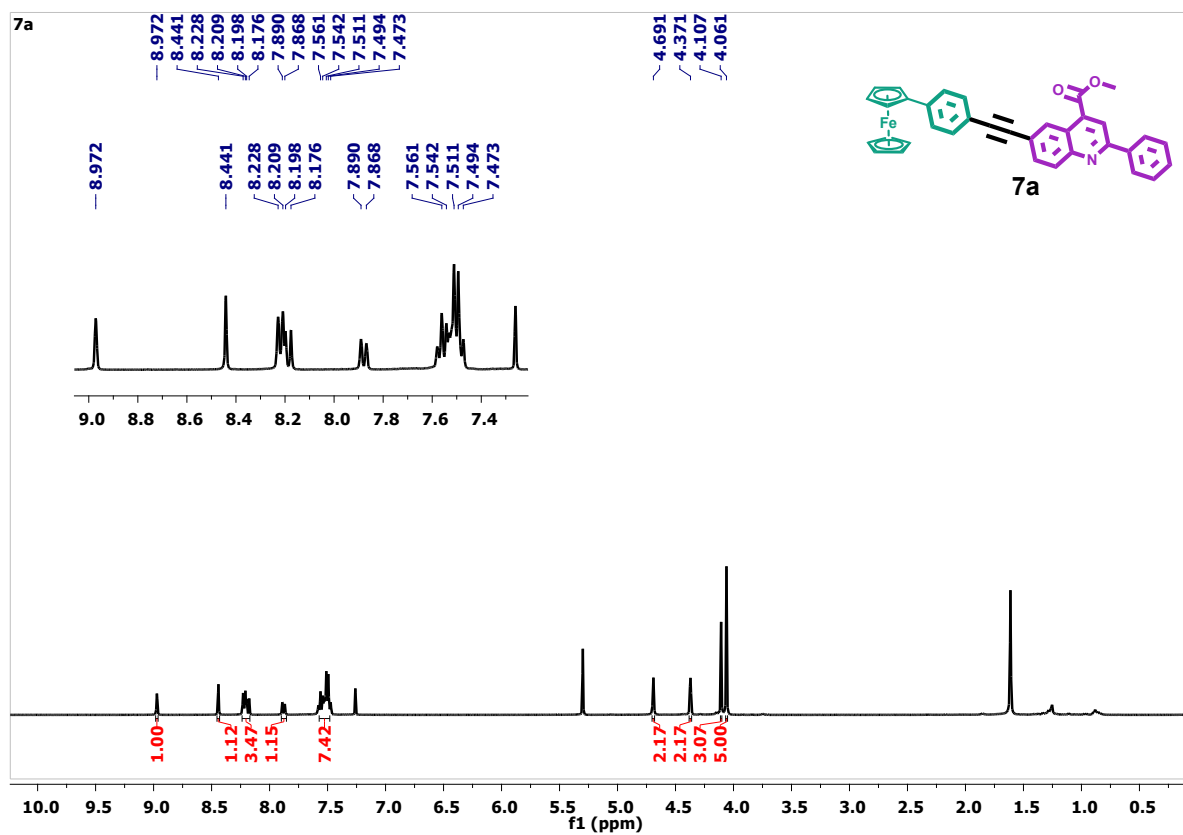
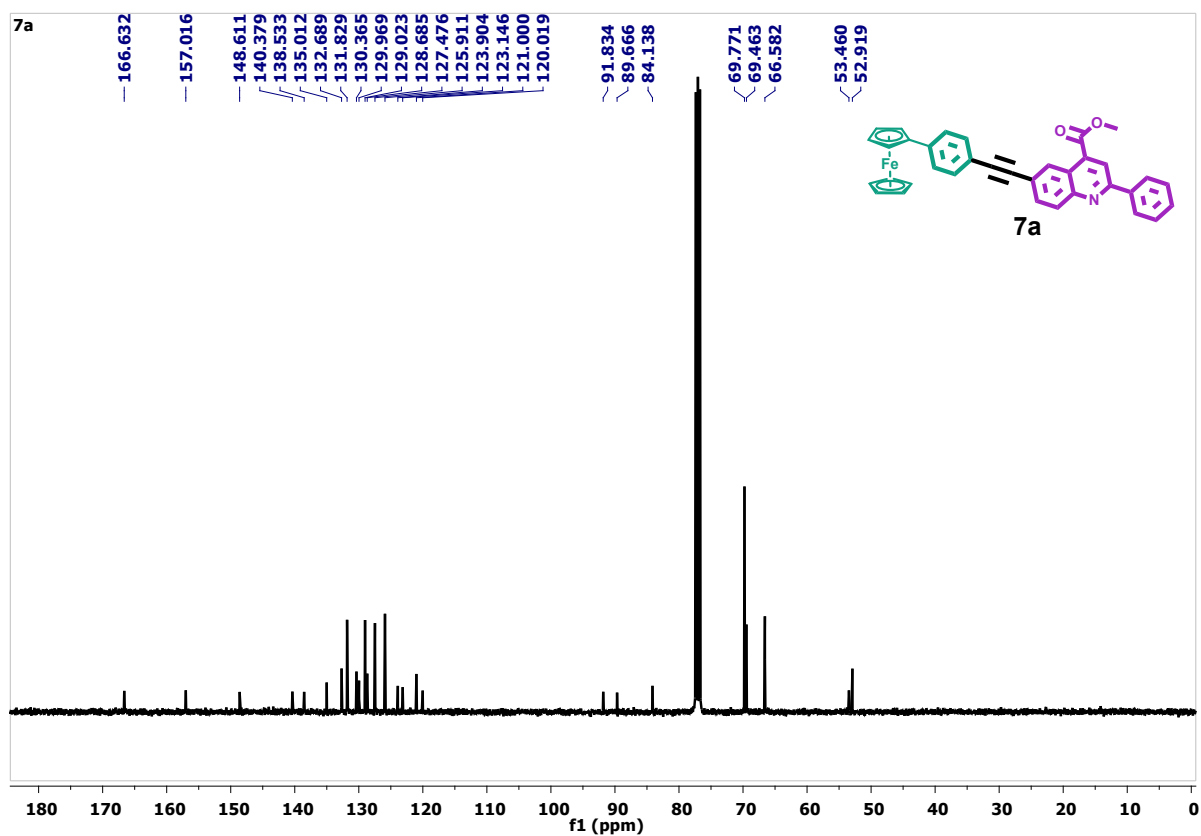
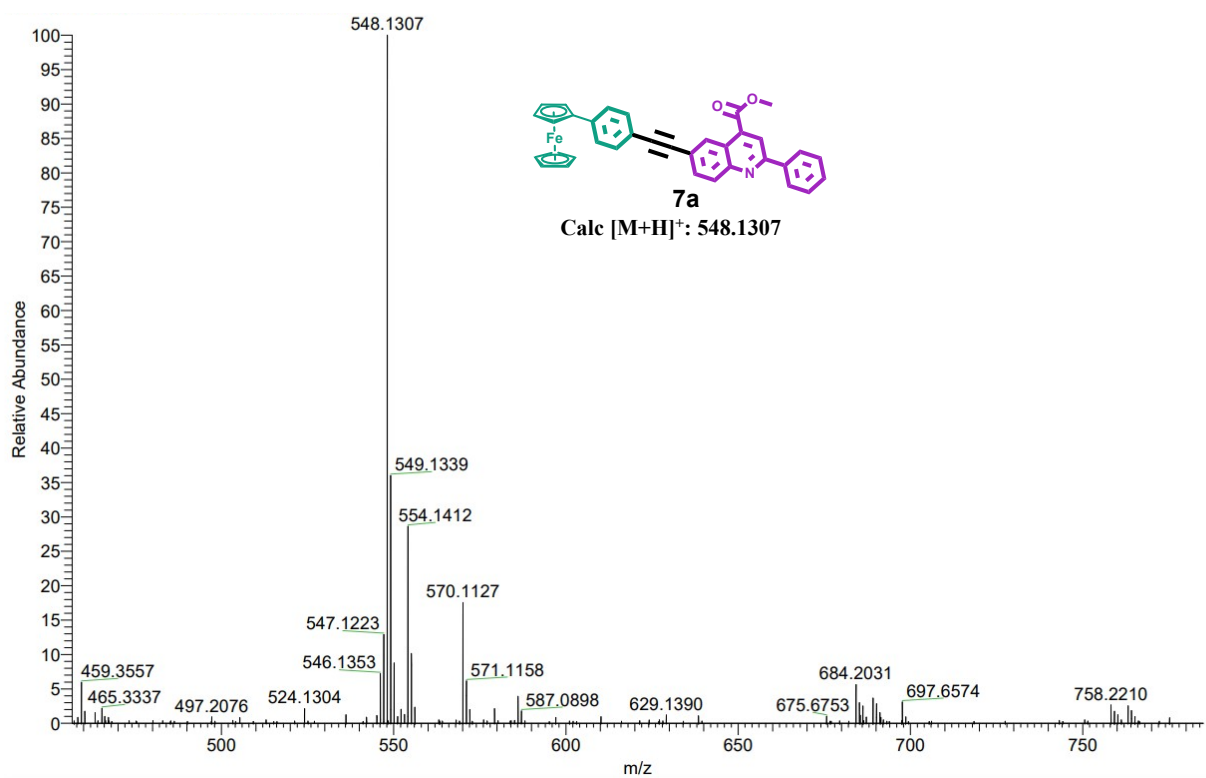


Figure S8: HRMS spectrum of compound **6c**

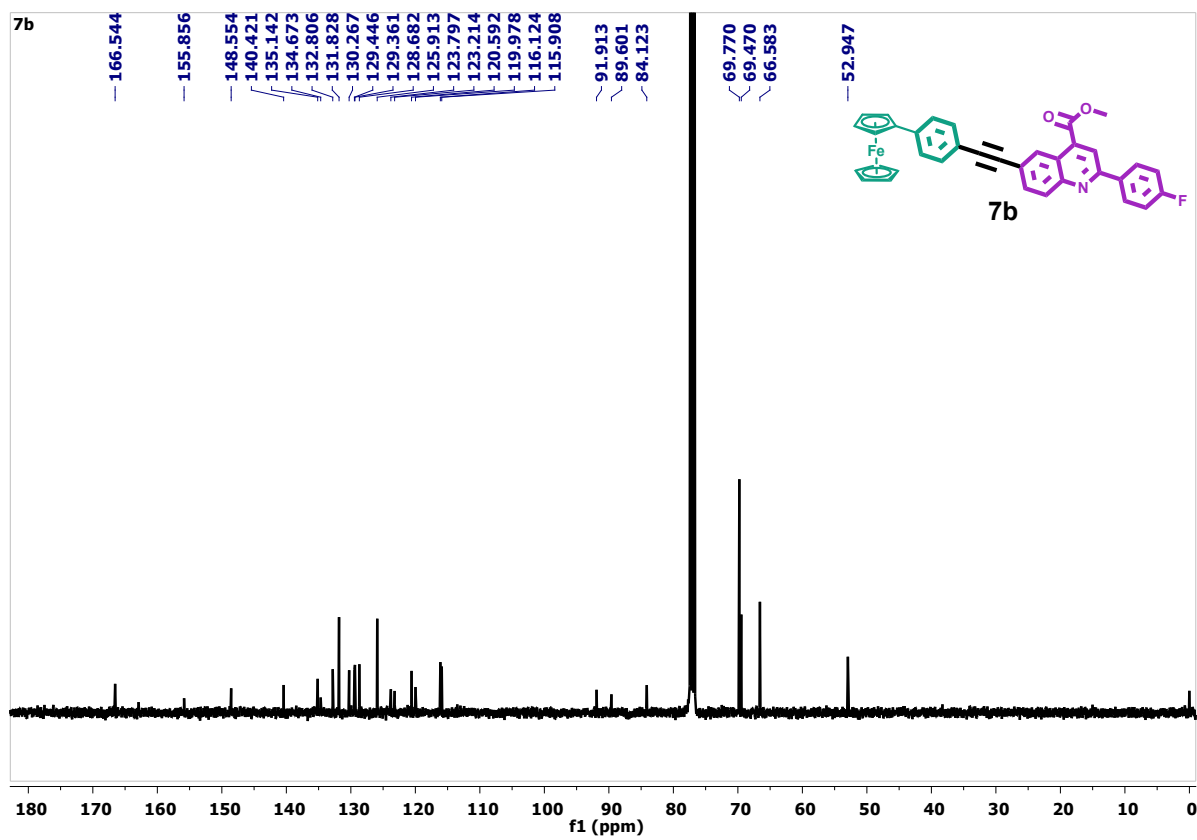
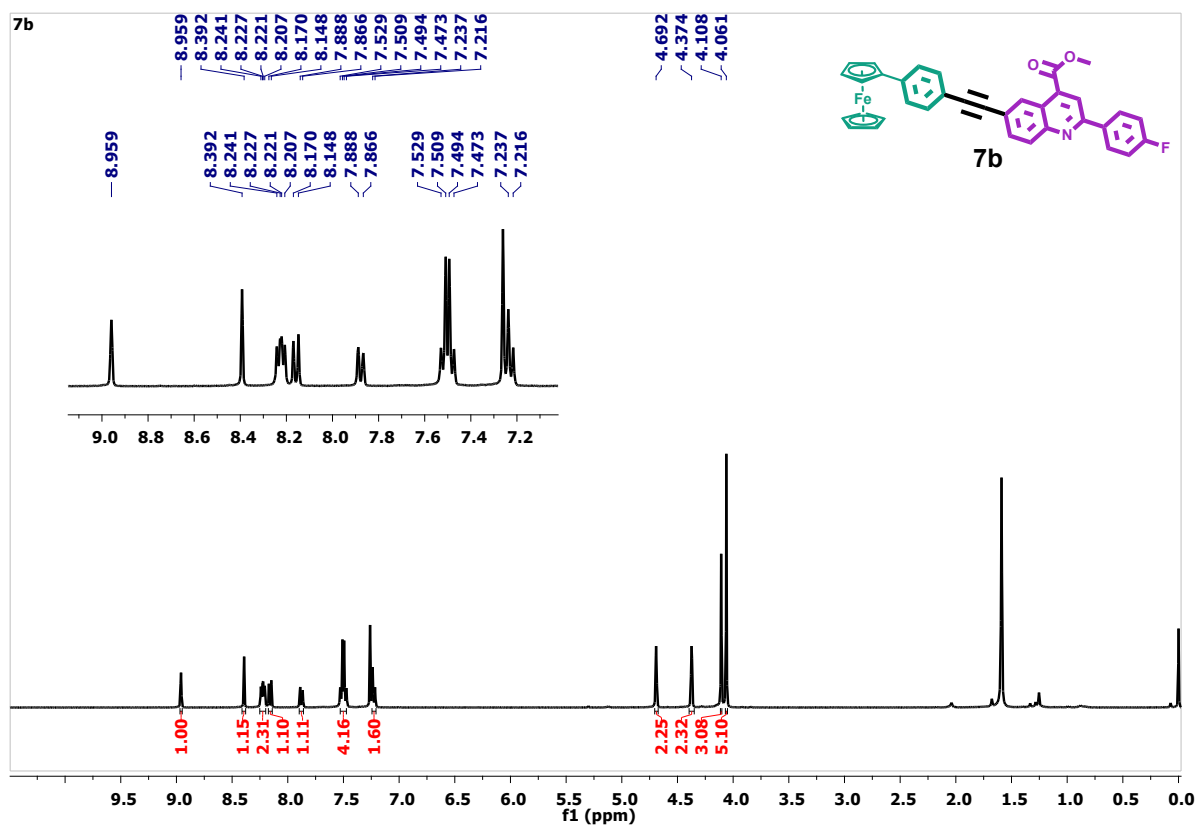




**Figure S9:**  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of compound **7a**



**Figure S10:** HRMS spectrum of compound **7a**



**Figure S11:** <sup>1</sup>H and <sup>13</sup>C NMR spectra of compound **7b**

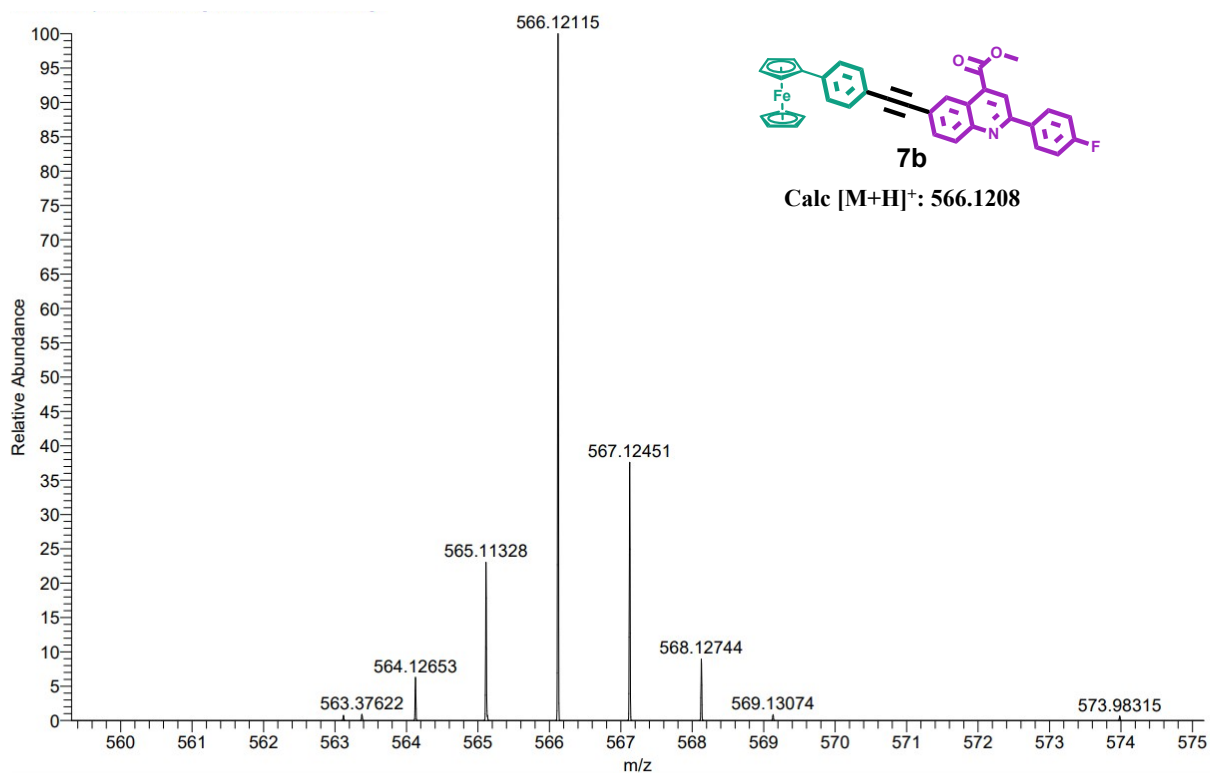
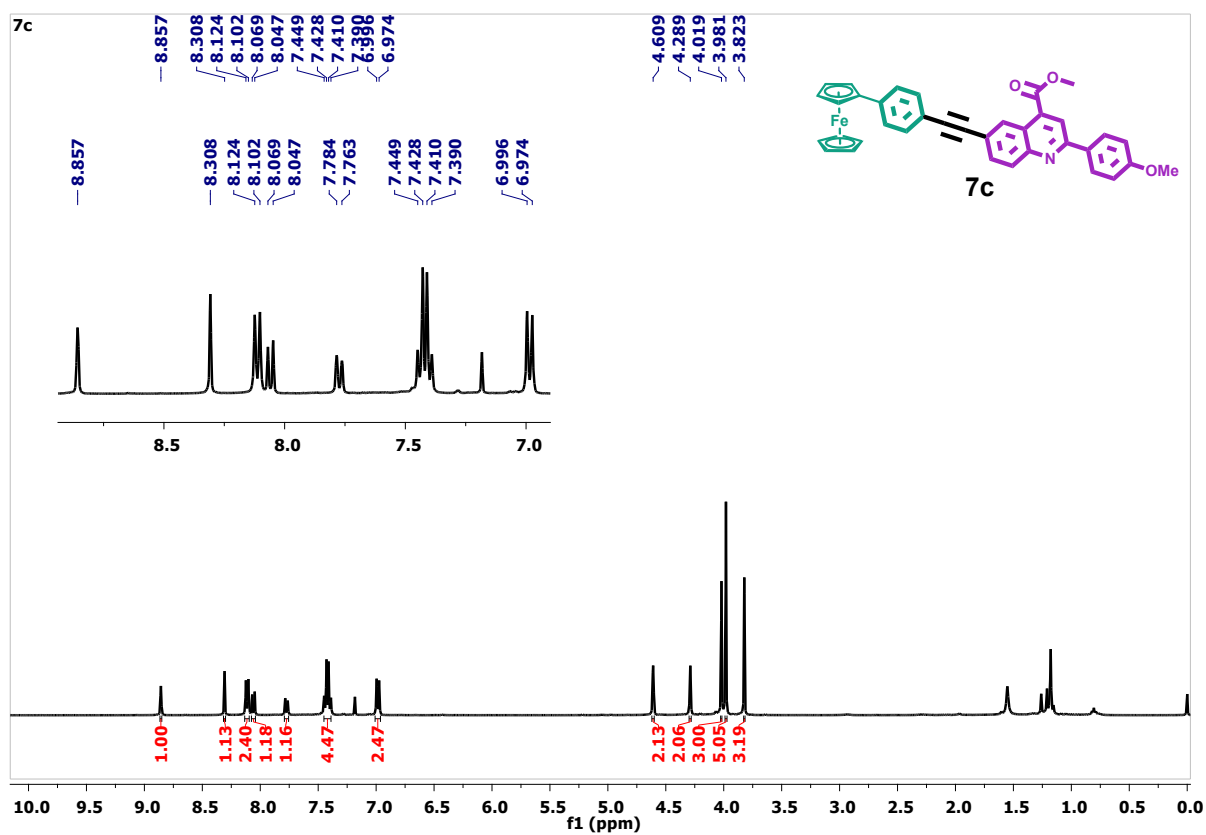
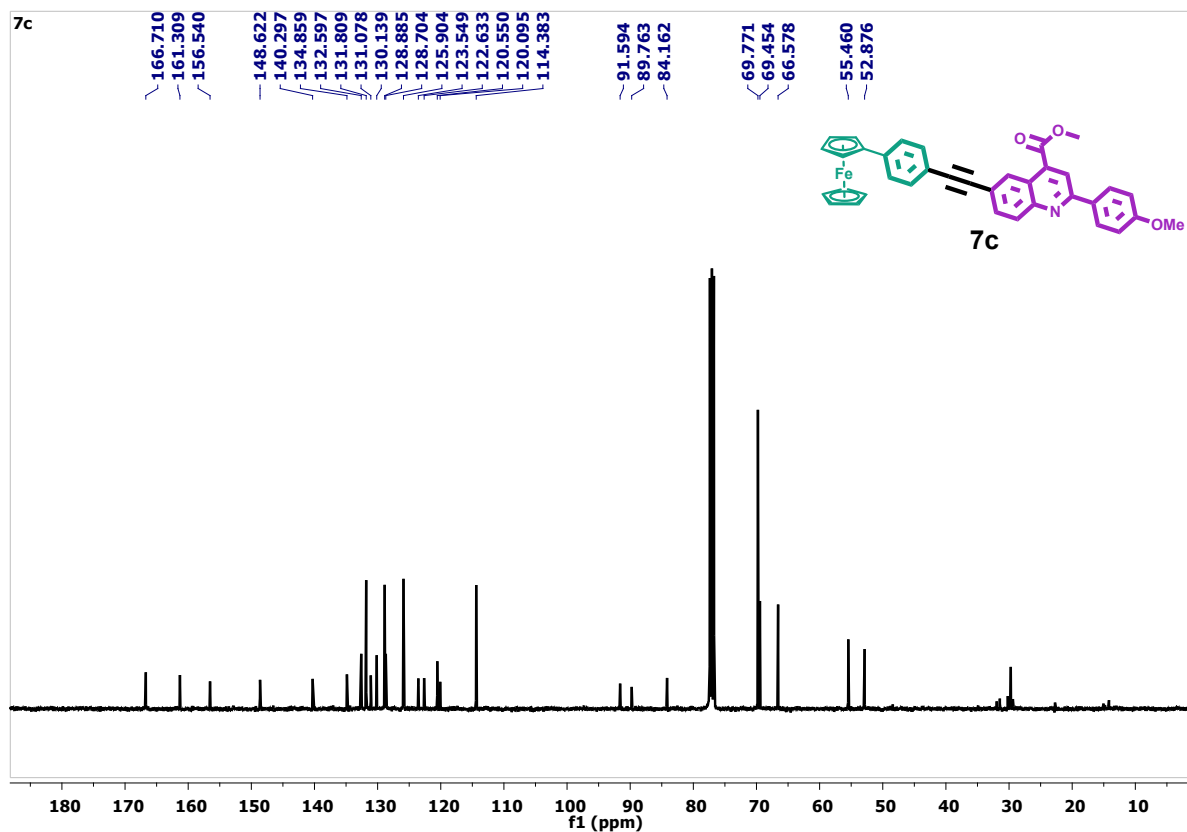


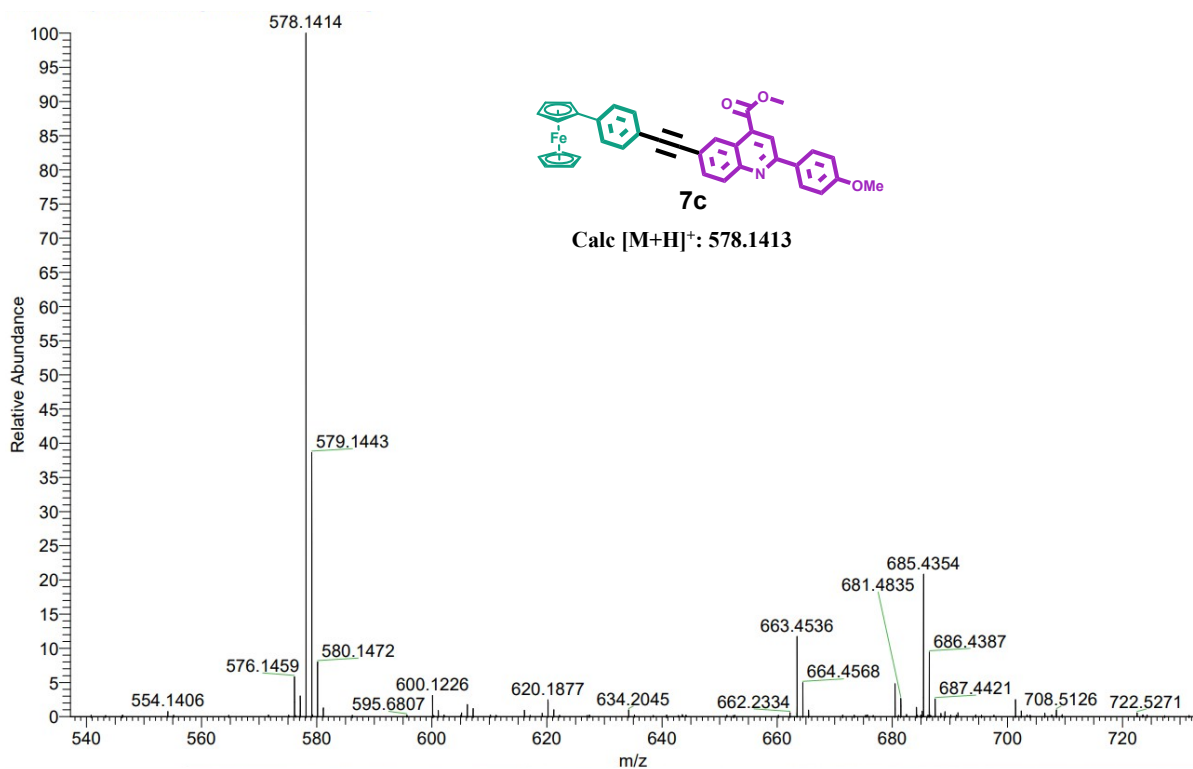
Figure S12: HRMS spectrum of compound **7b**







**Figure S13:**  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of compound **7c**



**Figure S14:** HRMS spectrum of compound **7c**

#### 4. Single crystal analysis of compound 6a

CCDC	2239172
Identification code	<b>6a</b>
Formula	$C_{35}H_{29}Cl_2FeNO_3$
$D_{calc.}/g\text{ cm}^{-3}$	1.487
$m/mm^{-1}$	0.755
Formula Weight	638.34
Colour	yellow
Shape	block-shaped
Size/ $mm^3$	0.16×0.10×0.04
$T/K$	110.00
Crystal System	triclinic
Space Group	$P-1$
$a/\text{Å}$	9.5150(4)
$b/\text{Å}$	12.7854(5)
$c/\text{Å}$	12.8008(5)
$a/^\circ$	80.8430(10)
$b/^\circ$	80.5270(10)
$g/^\circ$	69.0620(10)
$V/\text{Å}^3$	1426.06(10)
$Z$	2
$Z'$	1
Wavelength/ $\text{Å}$	0.71073
Radiation type	$MoK_\alpha$
$Q_{min}/^\circ$	2.594
$Q_{max}/^\circ$	27.530
Measured Refl's.	37506
Indep't Refl's	6546
Refl's $I \geq 2\sigma(I)$	5068
$R_{int}$	0.0645
Parameters	381
Restraints	0
Largest Peak	0.356
Deepest Hole	-0.358
Goof	1.058
$wR_2$ (all data)	0.0939
$wR_2$	0.0911
$R_1$ (all data)	0.0489
$R_1$	0.0369

## 5. Thin-film analysis of the compounds 6a-c and 7a-c

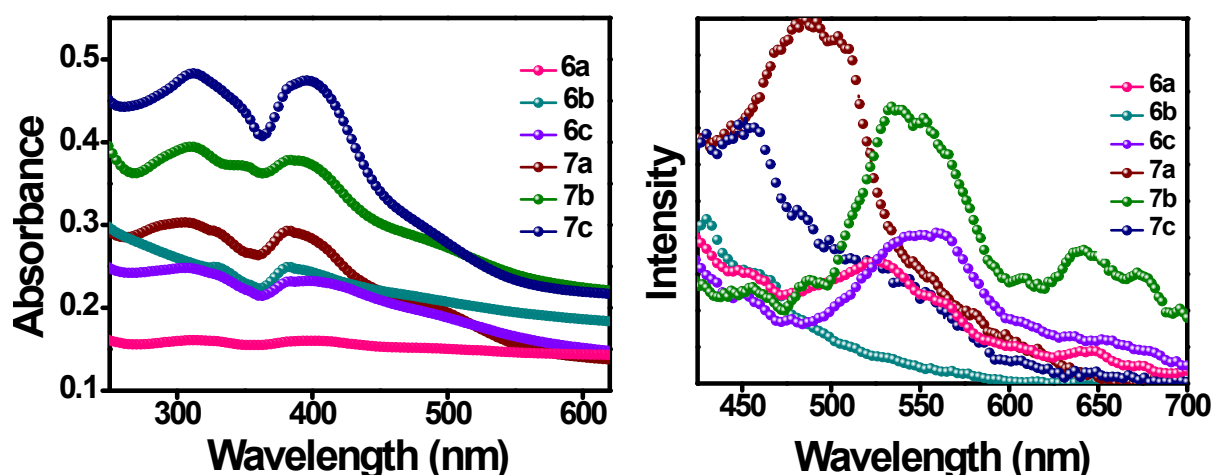


Figure S15: Thin-film absorption and fluorescence spectra of 6a-c and 7a-c

## 6. Electrochemical properties of the compounds 6a-c and 7a-c

Cyclic voltammetry was used to investigate the compounds' electrochemical properties using the conventional three-electrode cell setup in which glassy carbon acts as the working electrode, platinum wire as the counter electrode, and standard calomel as the reference electrode (Figure S19). The experiments were carried out in anhydrous dimethylformamide solvent at room temperature with a scan rate of  $100 \text{ mVs}^{-1}$  where, tetrabutylammonium hexafluorophosphate ( $n\text{-Bu}_4\text{NPF}_6$ , 0.1 M) was used as the supporting electrolyte. Nitrogen was purged into the solution for 10 minutes to prevent oxygen interference with the compounds. Ferrocene/ferrocenium ion ( $\text{Fc}/\text{Fc}^+$ ) redox couple was used for its calibration.<sup>2</sup>

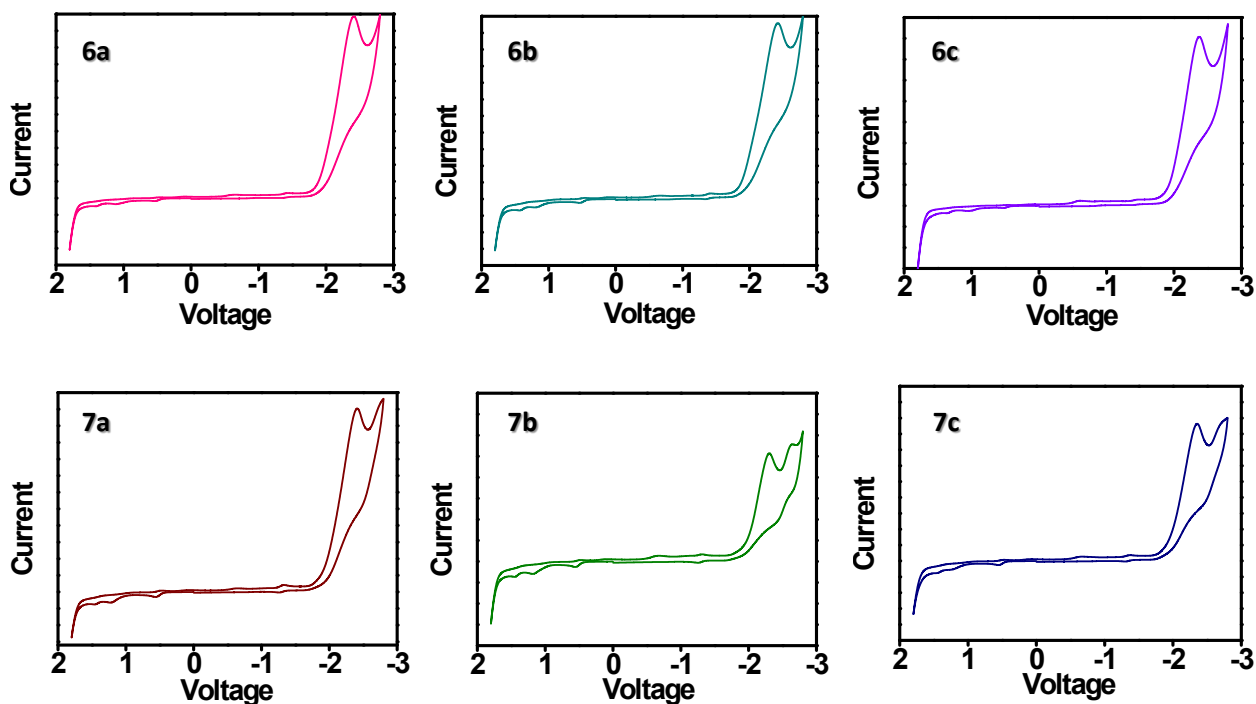


Figure S16: Cyclic voltammograms of the compounds 6a-c and 7a-c

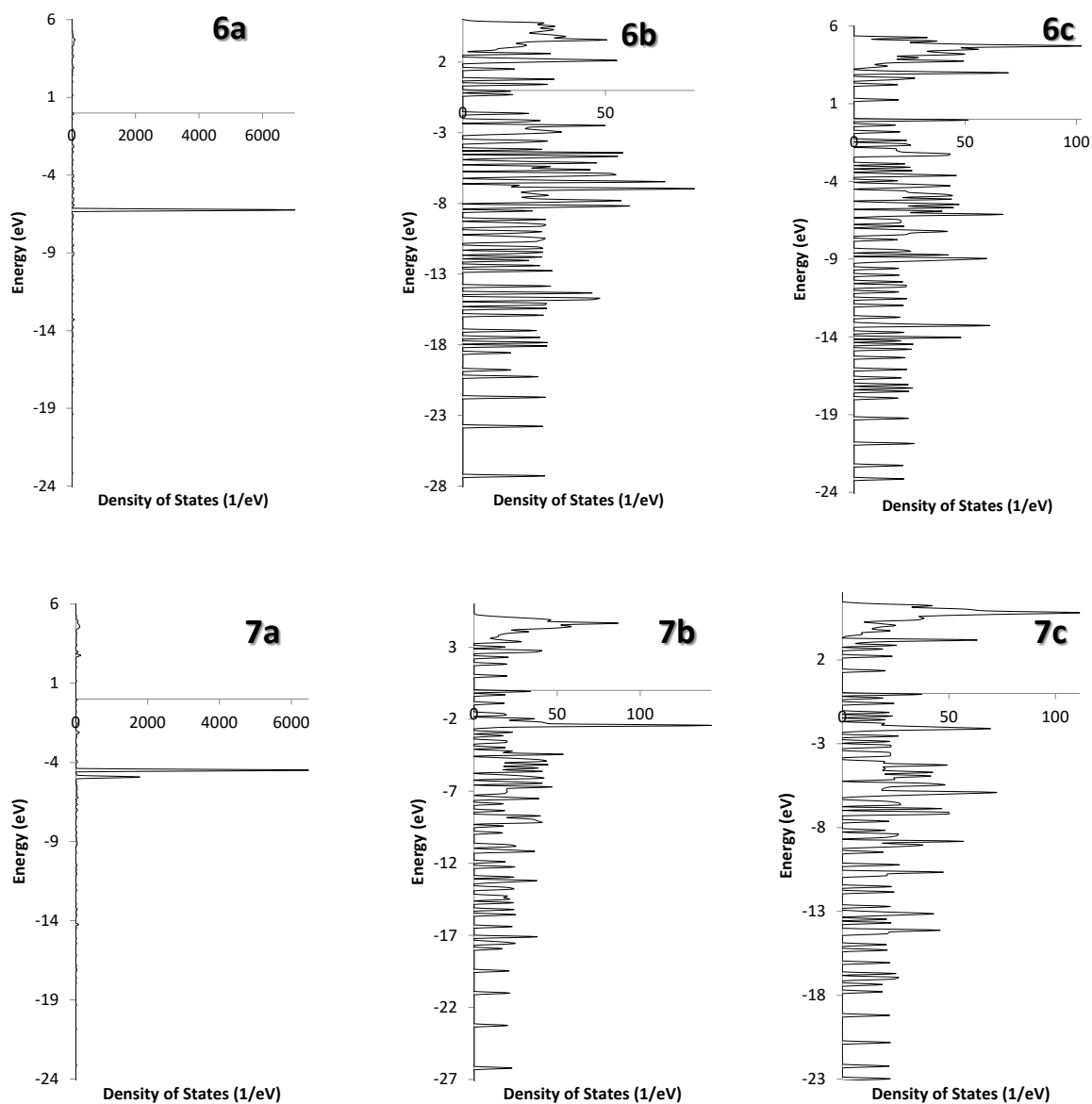
## 7. Computational studies

A series of computational simulations like Molecular Mechanics and semi-empirical methods were used to arrive at the plausible geometries of the structures as it is an essential aspect for packing and predicting electronic properties of the system, some of which are usually non-covalent. The approximate geometrical parameters were then used to compute the optimized structure at the DFT's B3LYP level of theory and TD-DFT for spectral estimation theoretically using Gaussian<sup>3</sup> using the basis set 6-31G (d). The excited states of the series of molecules were calculated by the computational methods and compared with experimentally obtained values (**Table S1**). The TD-DFT (Time-Dependent DFT) theoretical calculations suggest significant spectral insights. The emission bands found in the experimental methods align with the theoretical calculations. Out of the values predicted by the computational methods, only those closest to the experimental ones are chosen with a good frequency factor. The intersystem crossing has been predicted in almost all the systems, and the value of emission spectra agrees with these values.<sup>3</sup>

**Table S1:** The predicted absorption and emission behavior of the targeted compounds

Compound	Absorption wavelength (nm)	Electronic transition	Emission wavelength (nm)	Electronic transition	Dipole moment
<b>6a</b>	368.12	S <sub>1</sub> -S <sub>8</sub>	430.28	S <sub>1</sub> -S <sub>7</sub>	1.524
<b>6b</b>	410.99	S <sub>0</sub> -S <sub>2</sub>	422.53	S <sub>0</sub> -S <sub>3</sub>	2.204
<b>6c</b>	367.09	S <sub>1</sub> -S <sub>7</sub>	429.73	S <sub>0</sub> -S <sub>8</sub>	1.430
<b>7a</b>	427.41	S <sub>0</sub> -S <sub>1</sub>	534.01	S <sub>1</sub> -S <sub>8</sub>	1.479
<b>7b</b>	334.54	S <sub>1</sub> -S <sub>7</sub>	475.36	S <sub>0</sub> -S <sub>1</sub>	2.335
<b>7c</b>	433.10	S <sub>0</sub> -S <sub>7</sub>	471.16	S <sub>0</sub> -S <sub>1</sub>	1.366

The FMOs were visualized using *Gaussview* (reference). The optimized geometry was then used as input geometry for Density of States (DOS) calculations using *VASP (MedeA reference)* software. However, the structures were evaluated involving solvent correction parameters such as GGA-PBE (basis set), and the DOS graphs were subsequently obtained (**Figure S17**). The Fermi Energy level and bandgaps were also computed (**Table S2**). The density and volume of the cells were also computed. All the molecules pertained to the simple *Orthorhombic* system. The optimized geometry of the compounds **6a-c** and **7a-c** are given in **Figure S18**, and their molecular packing obtained through computational methods is presented in **Figure S19**.

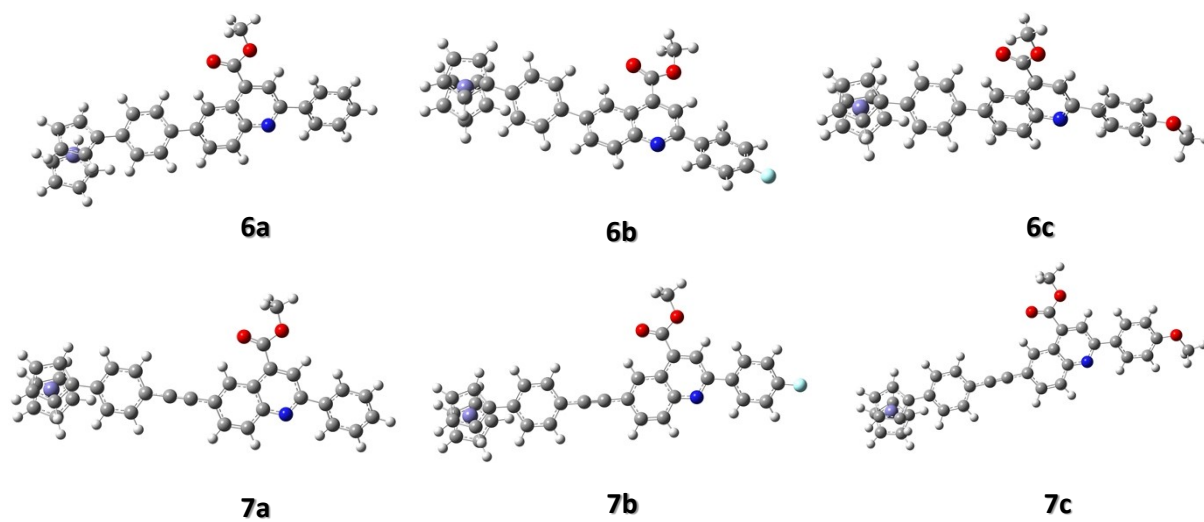


**Figure S17:** The Density of States graphs of the synthesized compounds **6a-c** and **7a-c**

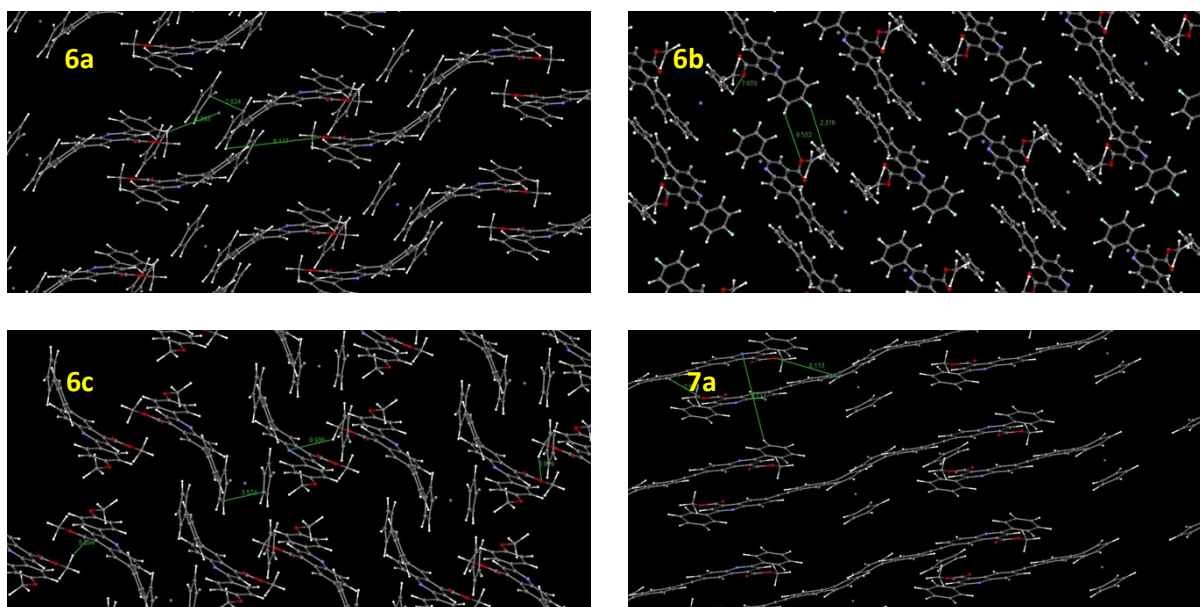
**Table S2:** DOS gap and E-Fermi energy of the molecules **6a-c** and **7a-c**

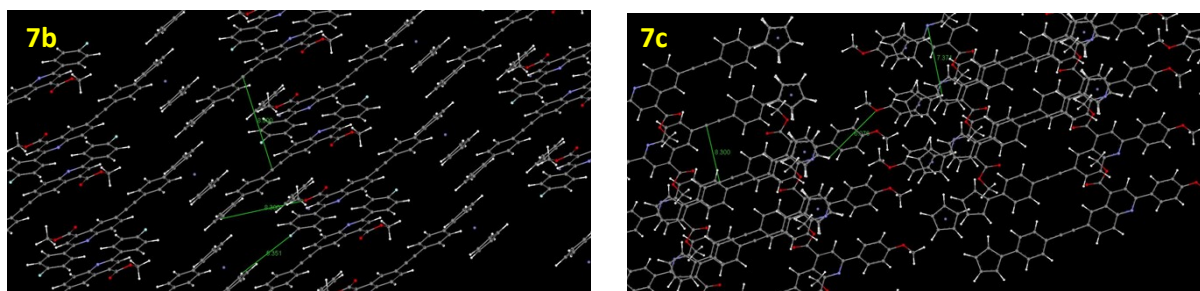
S No	Molecular Formula	Free Energy (eV)	Density (Mg/m <sup>3</sup> )	DOS Gap (eV)	E Fermi (eV)
<b>6a</b>	FeC <sub>33</sub> NO <sub>2</sub> H <sub>25</sub>	-418.44	0.291	2.388	-3.23
<b>6b</b>	FeC <sub>33</sub> NO <sub>2</sub> FH <sub>24</sub>	-397.32	0.351	1.936	-2.42
<b>6c</b>	FeC <sub>34</sub> NO <sub>3</sub> H <sub>27</sub>	-441.25	0.285	2.493	-3.24

<b>7a</b>	$\text{FeC}_{35}\text{NO}_2\text{H}_{25}$	-434.76	0.285	1.082	-3.32
<b>7b</b>	$\text{FeC}_{35}\text{NO}_2\text{FH}_{24}$	-435.05	0.304	1.775	-3.21
<b>7c</b>	$\text{FeC}_{36}\text{NO}_3\text{H}_{27}$	-457.55	0.279	2.596	-3.44



**Figure S18:** Optimized geometry of the compounds **6a-c** and **7a-c**



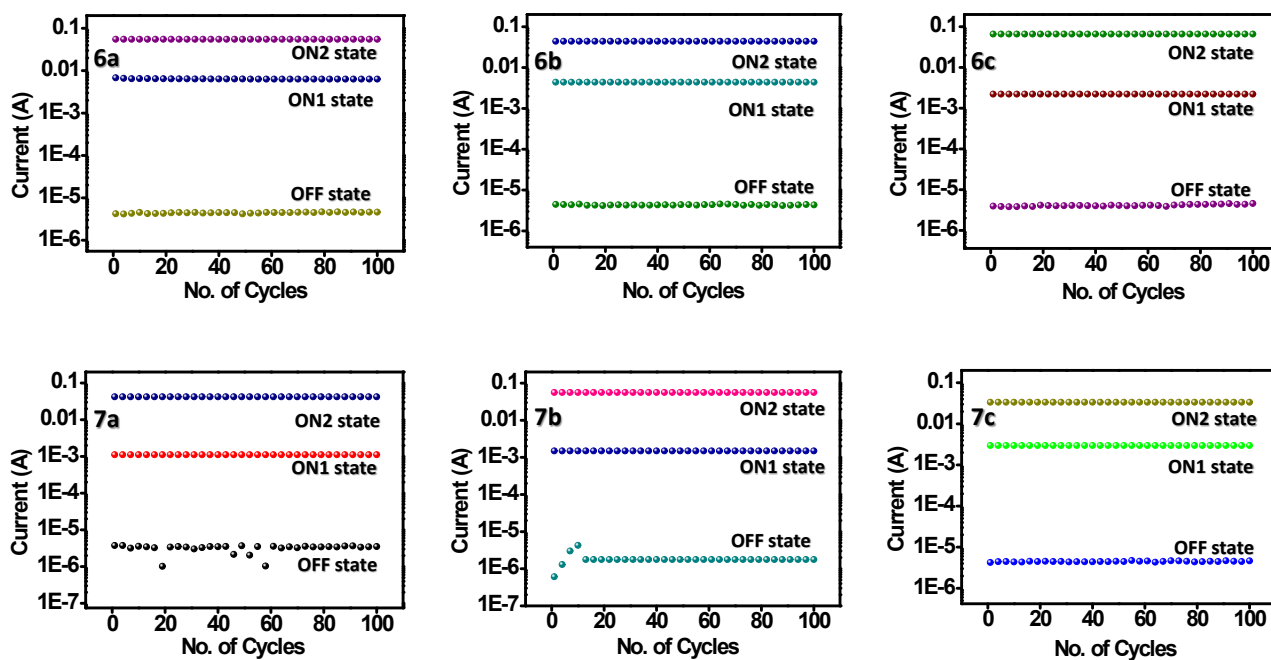


**Figure S19:** Molecular packing of the compounds **6a-c** and **7a-c**

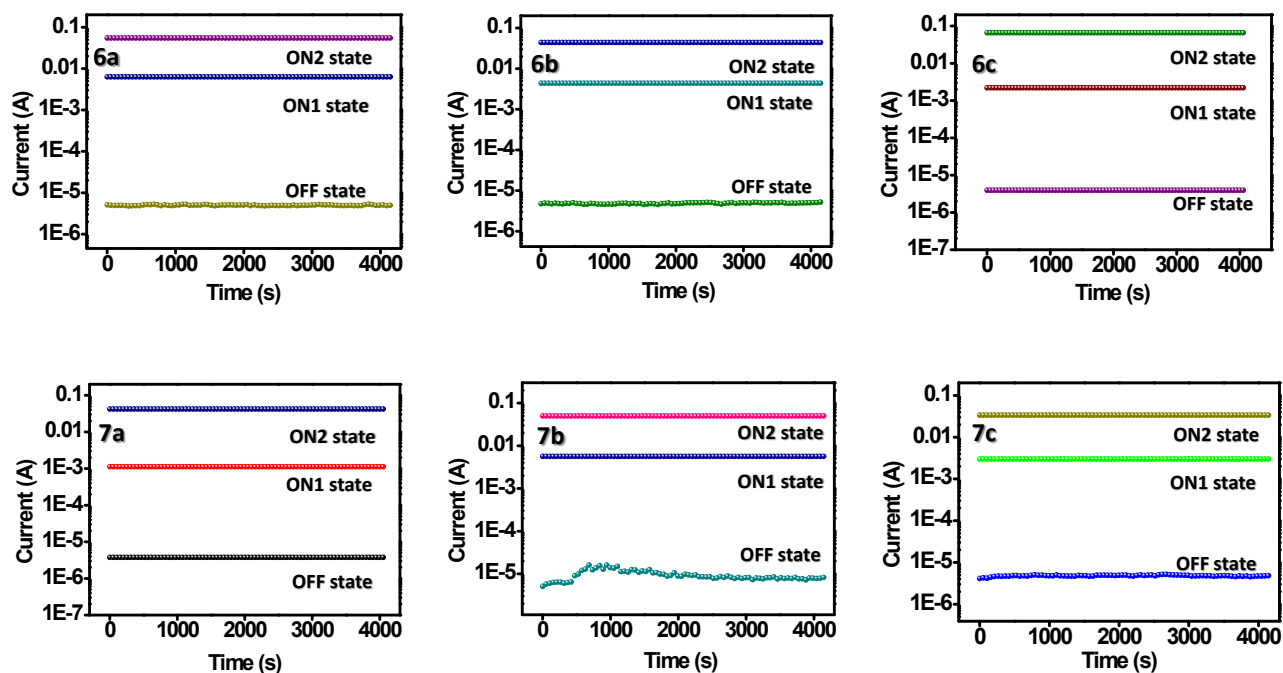
## 8. Memory device characterizations

### Fabrication and characterization of memory devices

Distilled water, soap solution, acetone, and ethanol were used as sonicating agents for 10 minutes each to clean the ITO-covered glass plates thoroughly. The compounds (**14a-d**) were dissolved in chloroform solution ( $5 \text{ mg mL}^{-1}$ ) and spin-coated over the ITO-coated glass plate with a film thickness of around 220 nm. After that, the thin film was annealed for 20 minutes at  $80 \text{ }^\circ\text{C}$ . The device was then coated with silver contacts via sputtering using a mask of 1 mm dimension. The device was then utilized for the study of memory characteristics. Keithley 4200A semiconductor parameter analyzer was used to perform memory characterizations at ambient conditions.



**Figure S20:** Endurance cycle at a constant stress of  $-0.5 \text{ V}$  for 100 cycles for the devices **6a-c** and **7a-c**



**Figure S21:** Retention time at a constant stress of  $-0.5\text{ V}$  for  $4 \times 10^3\text{ s}$  for the devices **6a-c** and **7a-c**

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