Supporting Information for

Mononuclear Fe(III) Schiff Base Antipyrine Complexes for Catalytic Hydrogen Generation

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Materials and Methods

Materials

All experiments were carried out using standard Schlenk techniques under an Ar atmosphere unless otherwise indicated. All reagents were purchased from Acros Organics, Alfa Aesar, Fisher Scientific, or TCI and were used without further purification unless otherwise noted. Tetra-*n*-butylammonium hexafluorophosphate ($TBAPF_6$) was recrystallized from absolute ethanol.

Instrumentation

^1H and ^{13}C NMR spectra were recorded on an Agilent 400MR DD2 spectrometer operating in the pulse Fourier transform mode. Chemical shifts are reported in ppm and referenced to residual solvent. High-resolution mass spectrometry was conducted using positive-ion mode electrospray ionization with an Apollo II ion source on a Bruker 10 Tesla APEX -Qe FTICR-MS. UV/vis experiments were performed with a Cary 60 UV/vis spectrophotometer. GC analysis was recorded on a Shimadzu GC-2014 gas chromatograph equipped with a TCD detector, Ar carrier gas, and a packed column.

X-ray Diffraction

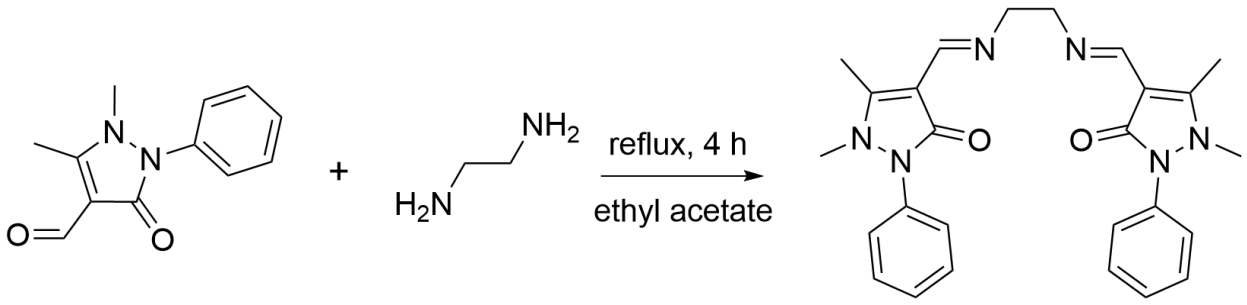
Data collection, structure solution, and structure refinement of **2** were conducted by William W. Brennessel at the University of Rochester's X-ray Crystallographic Facility. A single crystal was placed onto a thin glass optical fiber and mounted onto a Rigaku XtaLAB Synergy-S Dualflex diffractometer equipped with a HyPix-6000HE HPC area detector. Data collection was carried out using a PhotonJet (Cu) X-ray source. The structure was solved using SHELXT and refined using SHELXL.

Reflection contributions from highly disordered solvent were fixed and added to the calculated structure factors using the SQUEEZE routine of program Platon,[1] which determined there to be 599 electrons in 1504 $Å^3$ treated this way. Because the exact identity and amount of solvent were unknown, no solvent was included in the atom list or molecular formula. Thus, all calculated quantities that derive from the molecular formula (e.g., F(000), density, molecular weight, etc.) are known to be inaccurate.[1]

Data collection, structure solution, and structure refinement of **3** and **4** were conducted by Todd M. Reynolds at William & Mary. A single crystal was frozen in Parabar oil on a 50 µm Dual-Thickness MicroMount™ and mounted onto a Bruker D8 Venture diffractometer equipped with a PHOTON III photon counting detector. Data collection was carried out using a IµS 3.0 microfocus (Mo) X-ray source. The structure was solved using SHELXT and refined SHELXL.

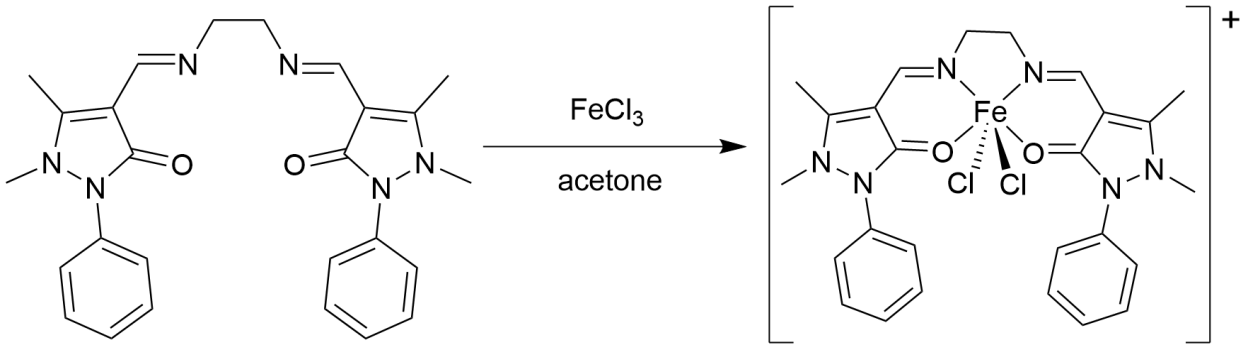
For **3**, reflection contributions from highly disordered solvent were fixed and added to the calculated structure factors using the SQUEEZE routine of program Platon,[1] which determined there to be 39 electrons in 149 $Å^3$ treated this way. Because the exact identity and amount of solvent were unknown, no solvent was included in the atom list or molecular formula. Thus, all calculated quantities that derive from the molecular formula (e.g., F(000), density, molecular weight, etc.) are known to be inaccurate.[1]

Syntheses



Scheme S1. Synthesis of **1**.

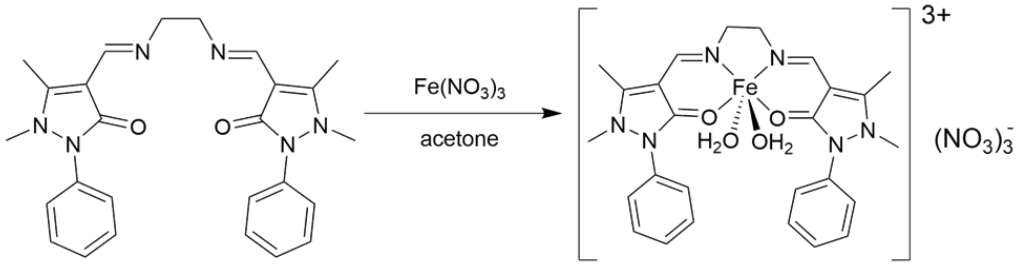
N,N'-bis(4-antipyrylmethylidene)ethylenediamine (1). This compound was synthesized using a modified literature procedure.[2] 4-antipyrine carboxaldehyde (0.5073 g, 2.346 mmol) was dissolved in ethyl acetate and added to a 100 mL round-bottom flask. A solution of ethylenediamine in ethyl acetate (78.3 μL, 1.17 mmol) was added dropwise to the round-bottom flask. The solution was refluxed with stirring for 4 hours. The resulting crystalline solid was obtained by vacuum filtration and rinsed with ethyl acetate. This gave 354 mg of light yellow solid (66% yield). ^{1}H NMR ($CDCl_3$, 400 MHz): δ 2.62 (s, 6 H), 3.18 (s, 6 H), 3.81 (s, 4 H), 7.33 (t, 6 H, J = 8.10 Hz), 7.46 (t, 4 H, J = 7.39 Hz), 8.21 (s, 2 H). ^{13}C NMR ($CDCl_3$, 400 MHz): δ 12.3, 34.4, 62.5, 104.8, 125.2, 126.9, 127.4, 129.0, 129.3, 129.7, 134.6, 154.4, 155.3, 165.3.



Scheme S2. Synthesis of **2.**

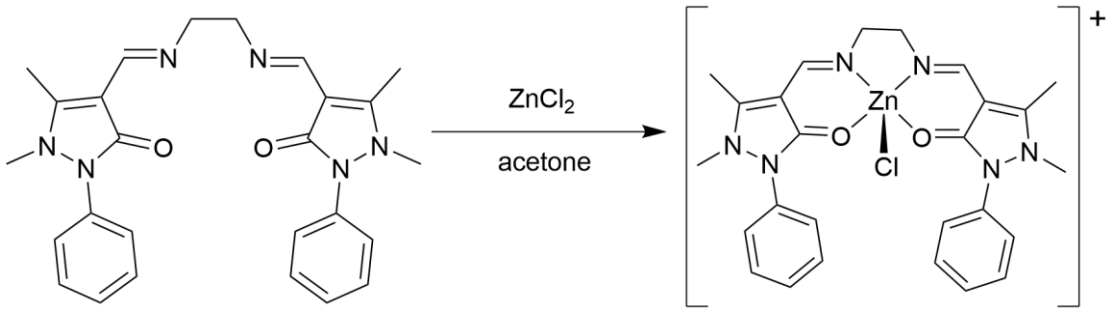
[Iron (III) N,N'- bis(4-antipyrlmethylidene)ethylenediamine dichloride] chloride (2). This complex was synthesized using a modified literature procedure.[3] In a 30 mL screw top vial, **1** (110 mg, 0.241 mmol) was suspended in acetone. $FeCl_3$•$6H_2O$ (67.8 mg, 0.251 mmol) was dissolved in acetone and added dropwise to the screw top vial. Upon metal addition, the solution turned deep red. The vessel was sealed and allowed to reflux while stirring for 1 hour. The solution was cooled and the precipitate was collected using vacuum filtration. The precipitate was rinsed with acetone and dried over vacuum. This resulted in 135.3 mg of **2** as a red solid (96% yield). The complex was purified for electrochemical and photochemical studies using slow diffusion by layering a highly concentrated solution of **2** in methanol with dichloromethane to afford crystalline product. Red needle crystals formed over 24 hours, the mother liquor was decanted, and the crystals were dried and collected. Crystals for structural characterization were made with a slow diffusion of ethyl ether into a highly concentrated solution of **2**. The complex

was characterized using x-ray crystallography and high-resolution mass spectrometry. Crystals for elemental analysis were crushed with a mortar and pestle and dried under vacuum for 24 hours. Samples for elemental analysis were run on a Carlo Erba 1108 CHNS combustion analyzer. Elem. Anal. Calculated for $[C_{26}H_{28}Cl_2FeN_6O_2]Cl$ with 1 H_2O, 0.25 MeOH, and 0.15 CH_2Cl_2: C, 48.23; H, 4.80; N, 12.78; Found: C, 48.29; H, 4.79; N, 12.75. HRMS for $C_{26}H_{28}Cl_2FeN_6O_2^+$: predicted m/z=582.099473, observed m/z=582.099886.



Scheme S3. Synthesis of **3**.

[Diaqua iron (III) N,N'- bis(4-antipyrlmethylidene)ethylenediamine] nitrate (3). This complex was synthesized using a modified literature procedure.[3] In a 100 mL round bottom flask, **1** (200 mg, 0.438 mmol) was suspended in acetone. $Fe(NO_3)_3 \bullet 9H_2O$ (177 mg, 0.438 mmol) was dissolved in acetone and added dropwise to the screw top vial. Upon metal addition, the solution turned deep red. The vessel was sealed and allowed to reflux while stirring for 1 hour. The solution was cooled and the precipitate was collected using vacuum filtration. The precipitate was rinsed with acetone and dried over vacuum. This resulted in 275.9 mg of **3** as a red solid (87% yield). The complex was purified for electrochemical and photochemical studies using slow diffusion by layering a highly concentrated solution of **3** in methanol with toluene. Red needle crystals formed over 24 hours, the mother liquor was decanted, and the crystals were dried and collected. Crystals for structural characterization were made with a slow diffusion of a highly concentrated solution of **3** into toluene. The complex was characterized using x-ray crystallography and high-resolution mass spectrometry. Crystals for elemental analysis were crushed with a mortar and pestle and dried under vacuum for 24 hours. Samples for elemental analysis were run on a Carlo Erba 1108 CHNS combustion analyzer. Elem. Anal. Calculated for $[C_{26}H_{26}N_6O_2Fe(OH_2)_2](NO_3)_3$: C, 45.40; H,4.65; N, 16.15; Found: C,45.54; H,4.56; N, 15.96 HRMS for $C_{26}H_{26}N_6O_2Fe(OH_2)_2^{3+}$: predicted m/z=182.055383, observed m/z=182.055421.



Scheme S4. Synthesis of **4**.

[Zinc (II) N,N'- bis(4-antipyrlmethylidene)ethylenediamine chloride] chloride (4). This complex was synthesized using a modification of the procedure for the synthesis of **2**.[3] **1** (145 mg, 0.318 mmol) was suspended in acetone and heated in a 100 mL round-bottom flask. $ZnCl_2$ (42.9 mg, 0.315 mmol) was dissolved in acetone and added dropwise to the round-bottom flask. No color change was observed upon metal addition. The vessel was allowed to reflux while stirring for 3 hours. The solution was cooled, and a yellow precipitate was collected using vacuum filtration. The precipitate was rinsed with acetone and dried over vacuum. This gave a mass of 147 mg of **4** (84% yield). The complex was purified for structural characterization, electrochemical, and photochemical studies using slow diffusion by layering diethyl ether over a highly concentrated solution of **4** in methanol. Yellow needle crystals and white powder formed over 24 hours. The mother liquor was decanted, and the crystals were dried and collected. The yellow spikes were used for analysis. The complex was characterized by x-ray crystallography, high resolution mass spectroscopy, and NMR. High resolution mass spectrometry for $C_{26}H_{28}N_6O_2ClZn^+$: predicted m/z=555.124825, observed m/z=555.124675. ^{1}H NMR (D_2O, 400 MHz): δ 2.37 (s, 6 H), 3.27 (s, 6H), 3.69 (s, 4 H), 7.26 (d, 4 H, J = 6.91 Hz), 7.46 (d, 4 H, J = 7.65 Hz), 8.40 (s, 2 H). ^{13}C NMR (D_2O, 400 MHz): δ 9.15, 32.7, 55.0, 99.3, 129.0, 130.2, 131.4, 149.7, 158.1, 161.8.

Cyclic Voltammetry General Procedure

All electrochemical experiments were performed under an Ar atmosphere at 23°C and 1 atm using a CH Instruments 620D potentiostat with a CH Instruments 680 amp booster. Cyclic voltammograms (CVs) were taken using a standard three-electrode cell, including a saturated calomel electrode (SCE), platinum auxiliary electrode, and glassy carbon working electrode. The platinum and glassy carbon electrodes were polished prior to each CV using 0.05 μm alumina powder on a cloth-covered polishing pad and rinsed thoroughly with deionized water and acetonitrile. All electrochemical experiments were performed on crystals of each metal complex with recrystallized $TBAPF_6$.

Scan Rate Dependence Study

0.2 g of $TBAPF_6$ (0.1 M), and 0.1 mg of **2** (0.03 mM) were dissolved in 5.0 mL acetonitrile in an electrochemical cell. The resulting solution was degassed with argon. A background CV scan was obtained with no added acid, then 50 μL of 1.1 M trifluoroacetic acid (TFA) (11 mM) was added for each subsequent scan. CVs were taken at a potential range from -0.1 to -1.8 V vs Fc^+/Fc with scan rates ranging from 8-11 mV/s.

Acid Addition Studies

General Procedure. 0.2 g of $TBAPF_6$ (0.1 M), and 0.3 mg of crystalline **1-4** were dissolved in 5.0 mL acetonitrile in an electrochemical cell. The resulting solution was degassed with argon for 12 minutes. A background CV scan was obtained with no added acid, then 20 μL of 0.11 M TFA was added for each subsequent scan. Electrodes were polished between every scan.

pKa Studies. 0.2 g of $TBAPF_6$ (0.1 M), and 0.3 mg of crystalline **1-4** were dissolved in 5.0 mL acetonitrile in an electrochemical cell. The resulting solution was degassed with argon for 10 minutes. A background CV scan was obtained with no added acid, then 20 μL of either 0.11 M

phenol, 0.11 M acetic acid, or 0.11 M salicylic acid was added for each subsequent scan. Electrodes were polished between every scan.

Electrochemical Control Studies

Background Reduction Study. 0.2 g of $TBAPF_6$ (0.1 M), and 0.3 mg of ferrocene (0.1 mM) were dissolved in 5.0 mL acetonitrile in an electrochemical cell. The resulting solution was degassed with argon. A background CV scan was obtained with no added acid, then 20 μL of 0.11 M TFA was added for each subsequent scan. Electrodes were polished between each scan.

0.2 g of $TBAPF_6$ (0.1 M), 0.3 mg of $FeCl_3$ (0.1 mM), and 0.3 mg of ferrocene (0.1 mM) were dissolved in 5.0 mL acetonitrile in an electrochemical cell. The resulting solution was degassed with argon. A background CV scan was obtained with no added acid, then 60 μL of 0.11 M TFA was added for a subsequent scan.

0.2 g of $TBAPF_6$ (0.1 M), 0.3 mg of $ZnCl_2$ (0.1 mM), and 0.3 mg of ferrocene (0.1 mM) were dissolved in 5.0 mL acetonitrile in an electrochemical cell. The resulting solution was degassed with argon. A background CV scan was obtained with no added acid, then 20 μL of 0.11 M TFA was added for each subsequent scan.

Controlled-Potential Coulometry

Controlled-potential coulometry (CPC) was performed in a 500 mL four-neck round bottom flask. 0.3 mg of **2** and 1.937 g of tetrabutylammonium hexafluorophosphate ($TBAPF_6$, 0.1 M) were added to 50 mL of acetonitrile. The flask was capped with two vitreous carbon electrodes separated by VYCOR frits and a silver wire reference electrode, all submerged in the solution. Argon was used to degas the flask for 20 minutes while stirring the solution. A Hamilton gas syringe was used to remove 10 mL of argon from the flask and 10 mL of methane was added as an internal standard. The potential was held at -1.5 V vs. Fc^+/Fc for 3600 seconds while the solution continued stirring. Once the CPC experiment was complete, 0.10 mL of the headspace was removed using a Hamilton gas syringe and injected into a gas chromatograph. The resulting ratio of hydrogen to methane from the chromatograms were used to determine hydrogen produced. See Figure S16.

Hydrogen Evolution Studies

Hydrogen evolution was determined using test tubes with 1:1 ethanol:deionized water solvent, varying amounts of **2 , 3** or **4**, 1.8 mM fluorescein, and 5% triethylamine (TEA, v/v%). Immediately after the addition of TEA, the test tubes were sealed with a septum and copper wire. The test tubes were degassed with argon for 10 minutes in the dark. A Hamilton syringe was used to remove 1.0 mL of argon from the headspace of the test tube and 1.0 mL of methane was added as an internal standard. The test tubes were irradiated with green light (520 nm wavelength) while stirring. After irradiation, 0.10 mL of headspace was removed using a Hamilton gas syringe and injected into a gas chromatograph to determine the ratio of hydrogen to methane to determine hydrogen produced. Natural water samples were obtained from Virginia Beach, VA (saltwater), Hampton, VA (brackish water), and Lake Matoaka in Williamsburg, VA (fresh). All natural water samples were filtered through celite prior to use. Artificial saltwater was prepared by dissolving NaCl in deionized water with concentrations of 0, 20, 35, and 50 ppt. The artificial saltwater replaced deionized water in the 1:1 ethanol:deionized water solvent.

System Optimization Studies

The general procedure for hydrogen evolution studies was followed while varying catalyst or chromophore concentration. Comparison of turnover numbers (TONs) were made between 5, 10, 30, 60, and 180 µL of **2** and 1.0, 1.4, and 1.8 mM fluorescein in ethanol solutions. Studies of the best chromophore and SED pairing were also done by comparing the TONs between fluorescein with TEA, eosin Y with TEA, eosin Y and TEOA, or eosin Y and TEOA at pH 7.

Determination of Overpotential

The overpotential in acetonitrile was calculated according to a literature procedure.[4] E_{ref} for 1 mM TFA in acetonitrile, which is given in the literature to be -0.71 V vs. Fc^+/Fc, can be subtracted from $E_{1/2}$ of **2** in the presence of 1 mM TFA, which is determined to be -1.4 V vs. Fc^+/Fc.[5] Therefore, the overpotential is calculated as such:

$$Overpotential = \left|E_{1/2} - E_{ref}\right|$$

$$= |(-1.4\ V\ vs\ Fc^+/Fc\ ) - (-0.71\ V\ vs\ \ Fc^+/Fc\ )|$$

$$= 700\ mV$$

Calculation of Turnover Frequency (TOF) Using i_{cat}/i_p

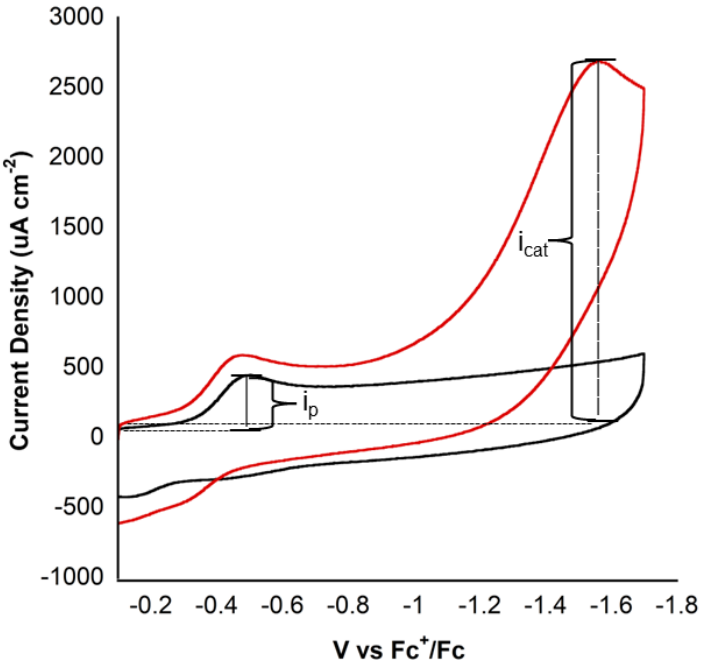


Figure S1. CV of **2** at ν = 10 V/s with no acid (black) and 0.88 (red) mM TFA.

TOF can be calculated using the expression[6-10]:

$$\frac{i_{cat}}{i_p} = \frac{n}{0.4463}\sqrt{\frac{RTk_{obs}}{Fv}}$$

where *n* is the number of electrons catalyzed in the reaction, *F* is Faraday's constant, ν is the scan rate in V/s, T is temperature in K, and R is the gas constant. Finding k_{obs} gives the TOF in s^{-1}.

A sample calculation is included below for **2** using Figure S1.

$$i_p = 449\ \mu A$$

$$i_{cat} = 2690\ \mu A$$

Correction of i_p for dilution[9]:

$$i_p \times \frac{vol_i}{vol_f} = 449\ \mu A \times \frac{5.00\ mL}{5.06\ mL} = 443\ \mu A$$

Calculation of TOF (k_{obs}):

$$k_{obs} = (\frac{i_{cat}}{i_p} \times \frac{0.4463}{n})^2 \left(\frac{Fv}{RT}\right) = (\frac{2690\ \mu A}{443\ \mu A} \times \frac{0.4463}{2})^2 \left(\frac{96485\ \frac{C}{mol} \times 10\frac{V}{s}}{8.314\ \frac{J}{mol\ K} \times 293\ K}\right) = 724\ s^{-1}$$

Calculation of Turnover Number

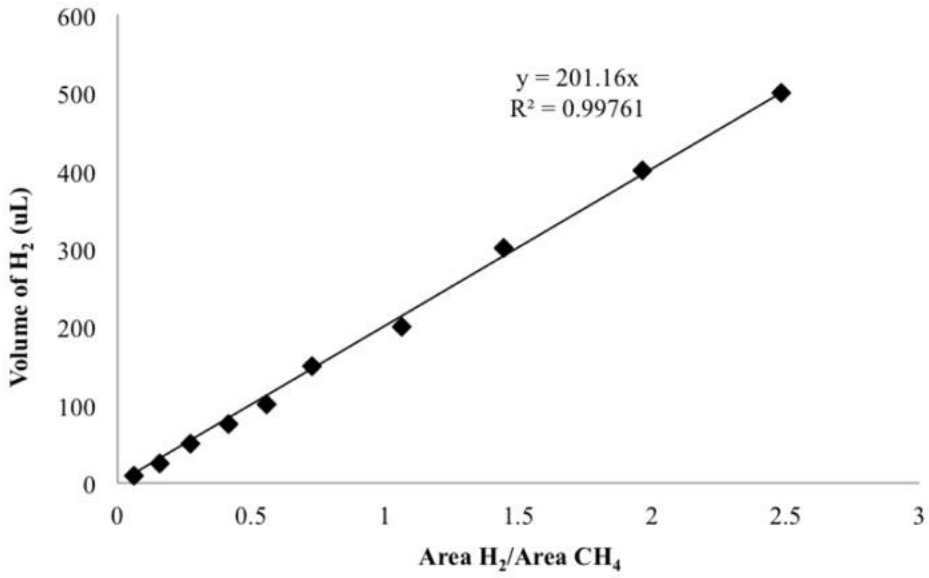


Figure S2. GC Calibration Curve. Calibration curve of H_2 to CH_4 peak areas used for determination of hydrogen generation. The ratio of peak areas was plotted against the volume of H_2 injected into the GC.

TONs is defined as the number of moles of hydrogen generated per mole of catalyst present in the system. We have previously developed a calibration curve relating the ratio of the peak areas of H_2 and CH_4 from our gas chromatography analysis to the volume of hydrogen generated by the system within our reaction vessel. Our calibration curve shows that the volume of hydrogen generated by the system has a linear relationship to the peak area ratio of H_2 to CH_4 with a slope of 201.6, as shown by the relationship[11,12]:

$$\mu L\ H_2 = 201.16(\frac{Area\ H_2}{Area\ CH_4})$$

A sample calculation is included below for 5 mg of catalyst:

$$\mu L\ H_2 = 201.16(\frac{114100.8}{15360.2}) = 1494$$

$$1494\ \mu L\ H_2 \times \frac{1\ L}{1 \times 10^6} \times \frac{1\ mol}{22.4\ L} = 6.67 \times 10^{-5}\ mol\ H_2$$

$$\frac{6.67 \times 10^{-5}\ mol\ H_2}{8.00 \times 10^{-8}\ mol\ catalyst} = 834\ TON$$

Calculation of Quantum Yield

The quantum yield (ϕ_{H2}) was determined by the following calculation proposed by Han et al., where λ is 520 nm, P is power (in W), h is Planck's constant, c is the speed of light, n is the number of photons, t is time (in seconds), k is the average rate of hydrogen production, and q_p is photon flux.[13] The difference between the power of light passing through a blank and through the sample was used to calculate light absorbed by the chromophore using a Nova II Power meter. The average rate of hydrogen production was determined from the amount of hydrogen generated during 24 hours of irradiation.

$$P = \frac{c \times h \times n}{\lambda \times t}$$

$$k = \frac{n}{t(s)}$$

$$q_p = \frac{P \times \lambda}{c \times h}$$

$$\phi_{H_2} = \frac{2k}{q_p}$$

The quantum yield reported was averaged from multiple measurements to account for differences in instrument readings and sample preparation. Sample calculations are shown below for **2**:

$$H_2\ produced: 9.62 \times 10^{-6}\ mol$$

$$k = \frac{(9.62 \times 10^{-6} mol)}{86400\ s} = 1.11 \times 10^{-10}\,\frac{mol}{s}$$

$$P = (1.86 \times 10^{-3}\ W) - (910 \times 10^{-6}\ W) = 9.50 \times 10^{-4}\ W$$

$$q_p = \frac{(P \times \lambda)}{(c \times h)} = \frac{(9.50 \times 10^{-4} W)(520 \times 10^{-9} m)}{(3.00 \times 10^{8}\,\frac{m}{s})(6.626 \times 10^{-34}\,\frac{J}{s})} = 2.49 \times 10^{15}\ s^{-1}$$

$$\frac{q_p}{mol} = \frac{(2.49 \times 10^{15}\ s^{-1})}{(6.02 \times 10^{23}\,\frac{photons}{mol})} = 4.14 \times 10^{-9}\,\frac{mol}{s}$$

$$\phi_{H_2} = \frac{2k}{q_p} = \frac{2(1.11 \times 10^{-10}\,\frac{mol}{s})}{(4.14 \times 10^{-9}\,\frac{mol}{s})} \times 100 = 5.4\%$$

A sample calculation for quantum yield is also included below for **3**:

$$H_2\ produced: 5.19 \times 10^{-6}\ mol$$

$$k = \frac{(5.19 \times 10^{-6}\ mol)}{86400\ s} = 6.00 \times 10^{-11}\,\frac{mol}{s}$$

$$P = (1.86 \times 10^{-3}\ W) - (910 \times 10^{-6}\ W) = 9.50 \times 10^{-4}\ W$$

$$q_p = \frac{(P \times \lambda)}{(c \times h)} = \frac{(9.50 \times 10^{-4} W)(520 \times 10^{-9} m)}{(3.00 \times 10^{8}\,\frac{m}{s})(6.626 \times 10^{-34}\,\frac{J}{s})} = 2.49 \times 10^{15}\ s^{-1}$$

$$\frac{q_p}{mol} = \frac{(2.49 \times 10^{15}\ s^{-1})}{(6.02 \times 10^{23}\,\frac{photons}{mol})} = 4.14 \times 10^{-9}\,\frac{mol}{s}$$

$$\phi_{H_2} = \frac{2k}{q_p} = \frac{2(6.00 \times 10^{-11}\,\frac{mol}{s})}{(4.14 \times 10^{-9}\,\frac{mol}{s})} \times 100 = 2.9\%$$

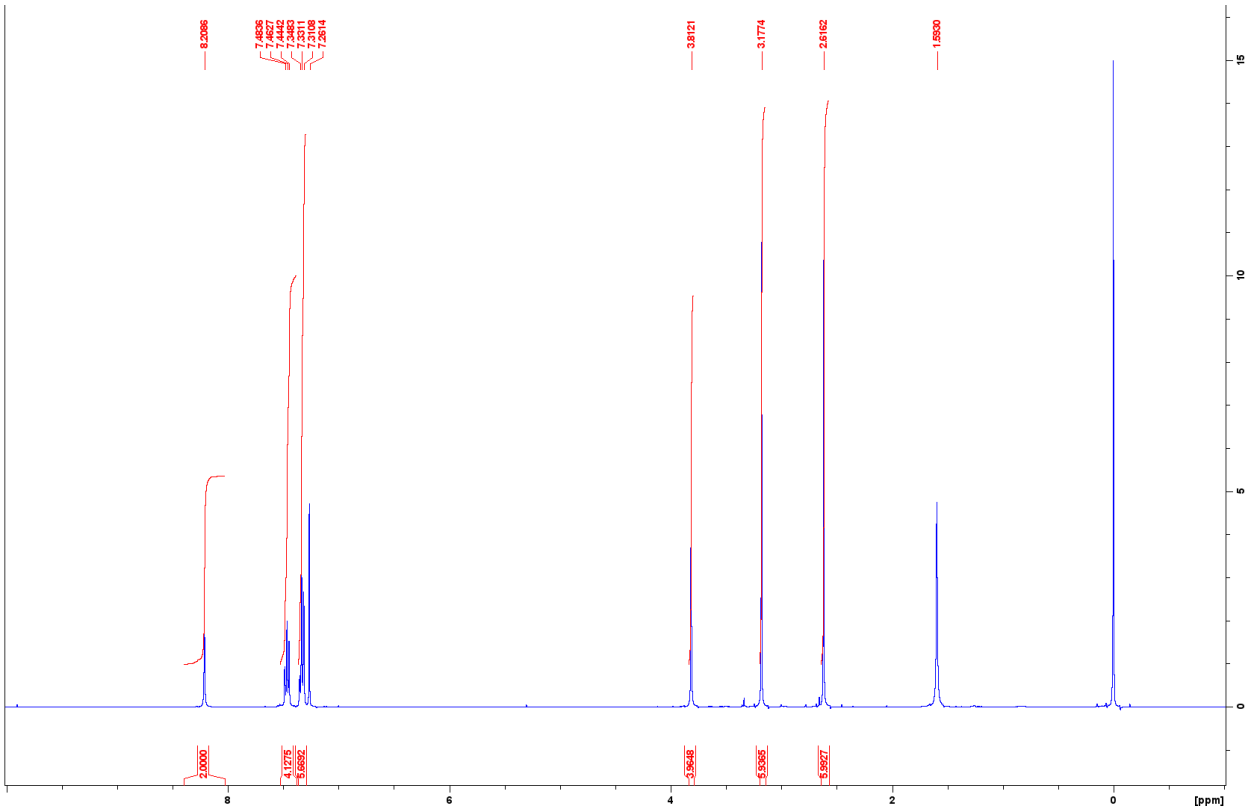


Figure S3. ^{1}H NMR of **1.**

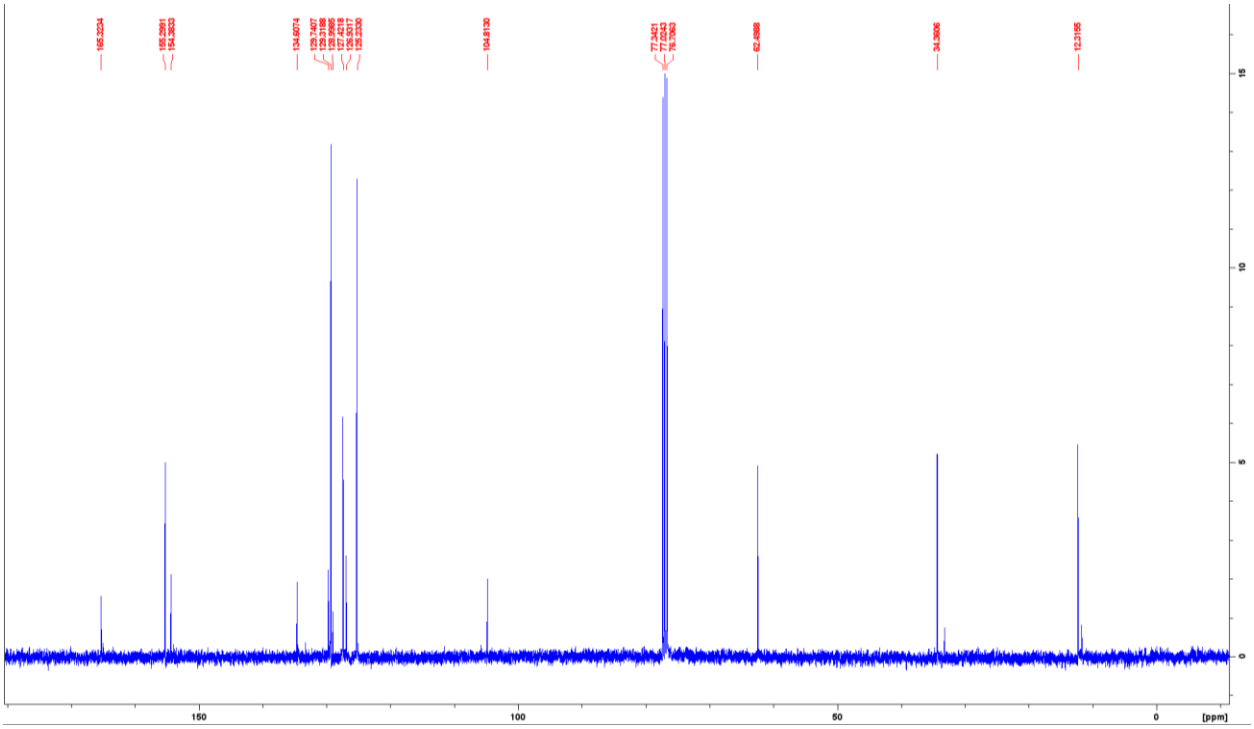


Figure S4. ^{13}C NMR of **1.**

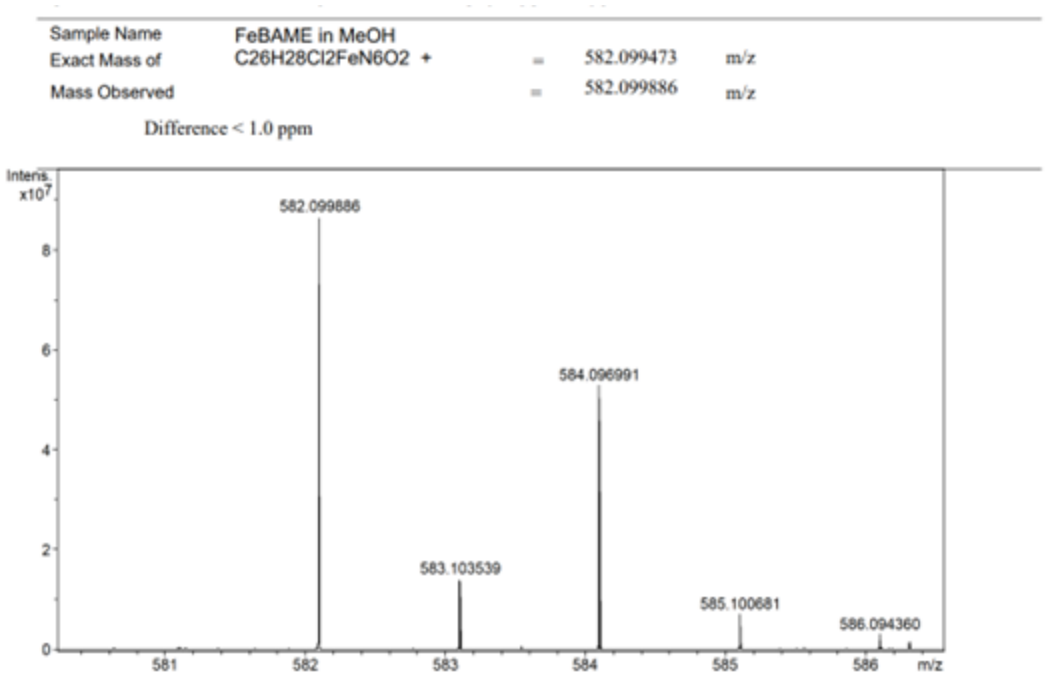


Figure S5. High resolution mass spectrometry results of **2**. The expected molecular ions were observed with a difference of less than 1 ppm.

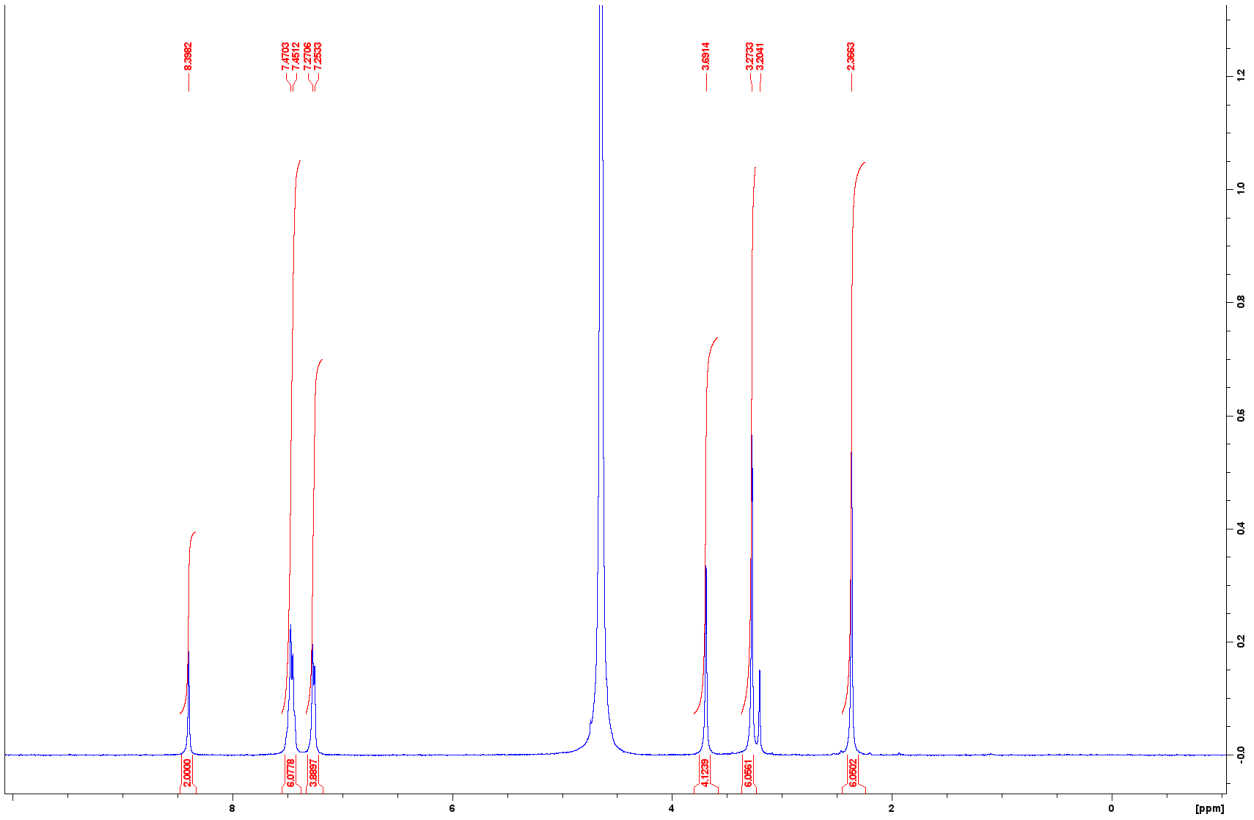


Figure S6. ^{1}H NMR of **4**.

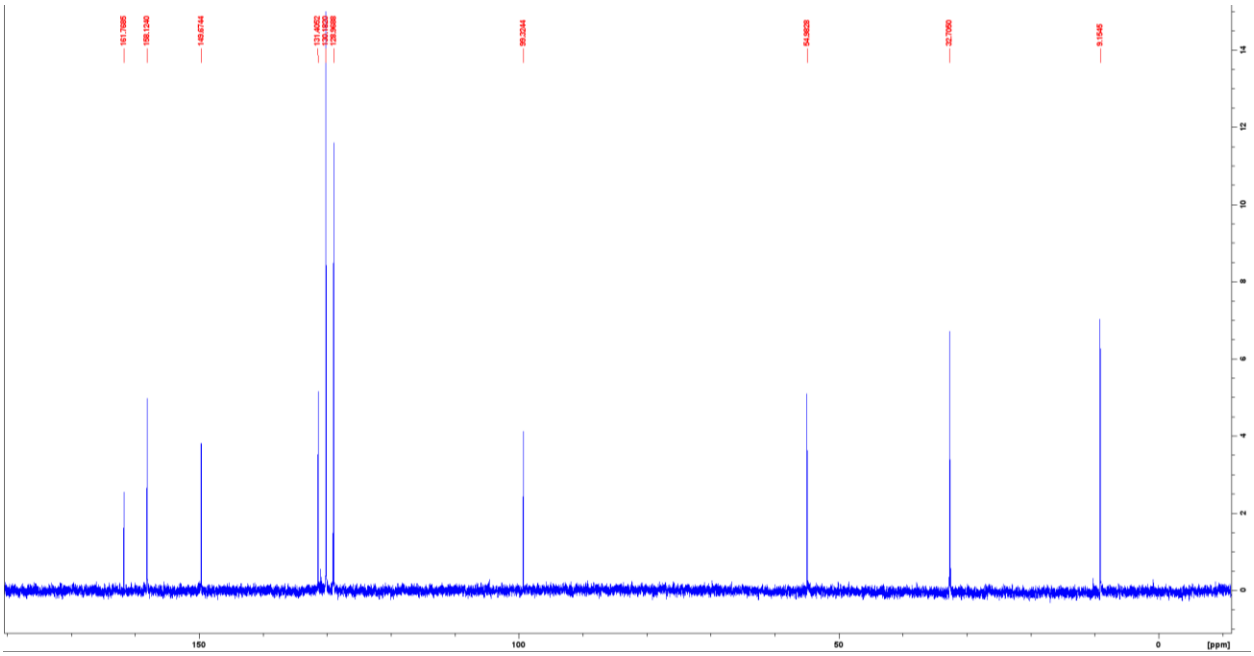


Figure S7. ^{13}C NMR of **4**.

Sample Name	Zn-BAME-spikes in MeOH			
Exact Mass of	C26H28N6O2ClZn +	=	555.124825	m/z
Mass Observed		=	555.124675	m/z

Difference < 1.0 ppm

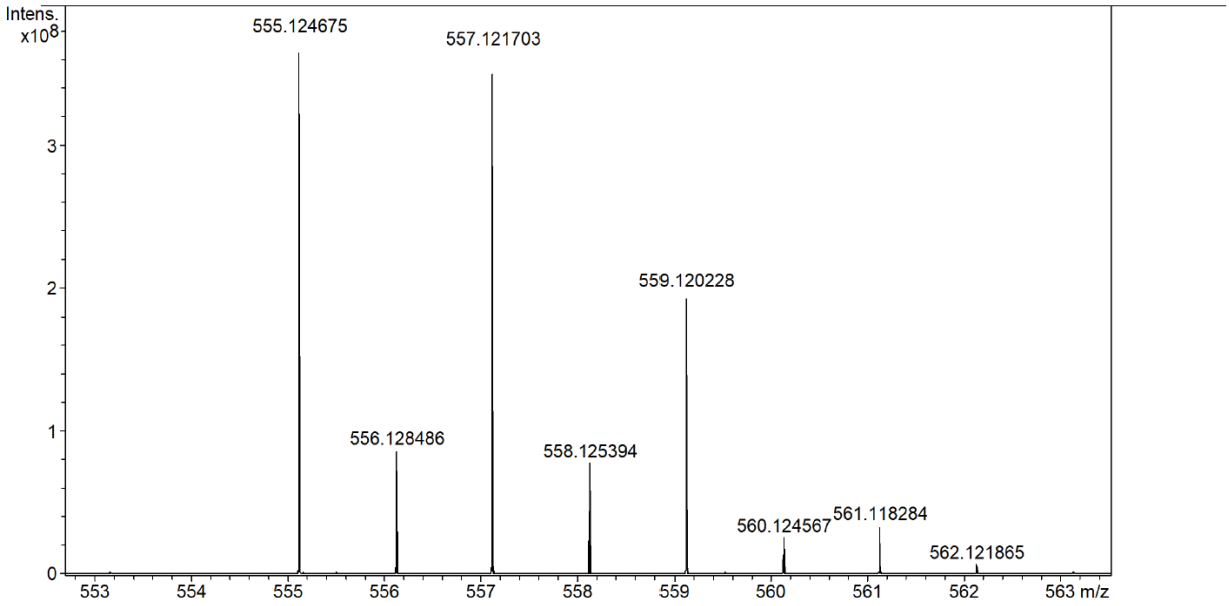


Figure S8. High resolution mass spectrometry results of **4**. The expected molecular ions were observed with a difference of less than 1 ppm.

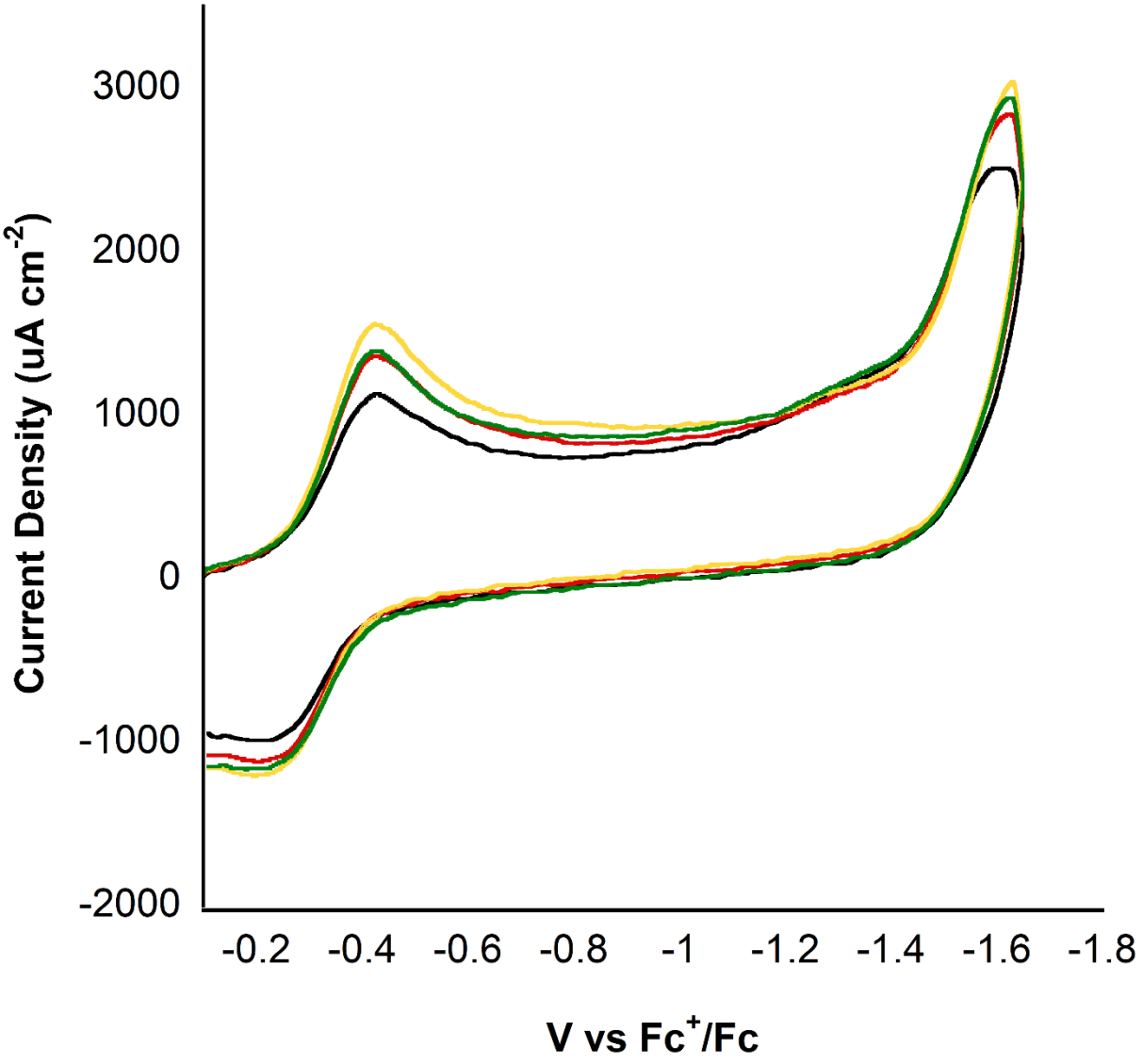


Figure S9. Scan rate study of **2** with scan rates of 8 V/s (black), 9 V/s (red), 10 V/s (yellow), and 11 V/s (green). These CV experiments were run in acetonitrile with 0.1 M $TBAPF_6$.

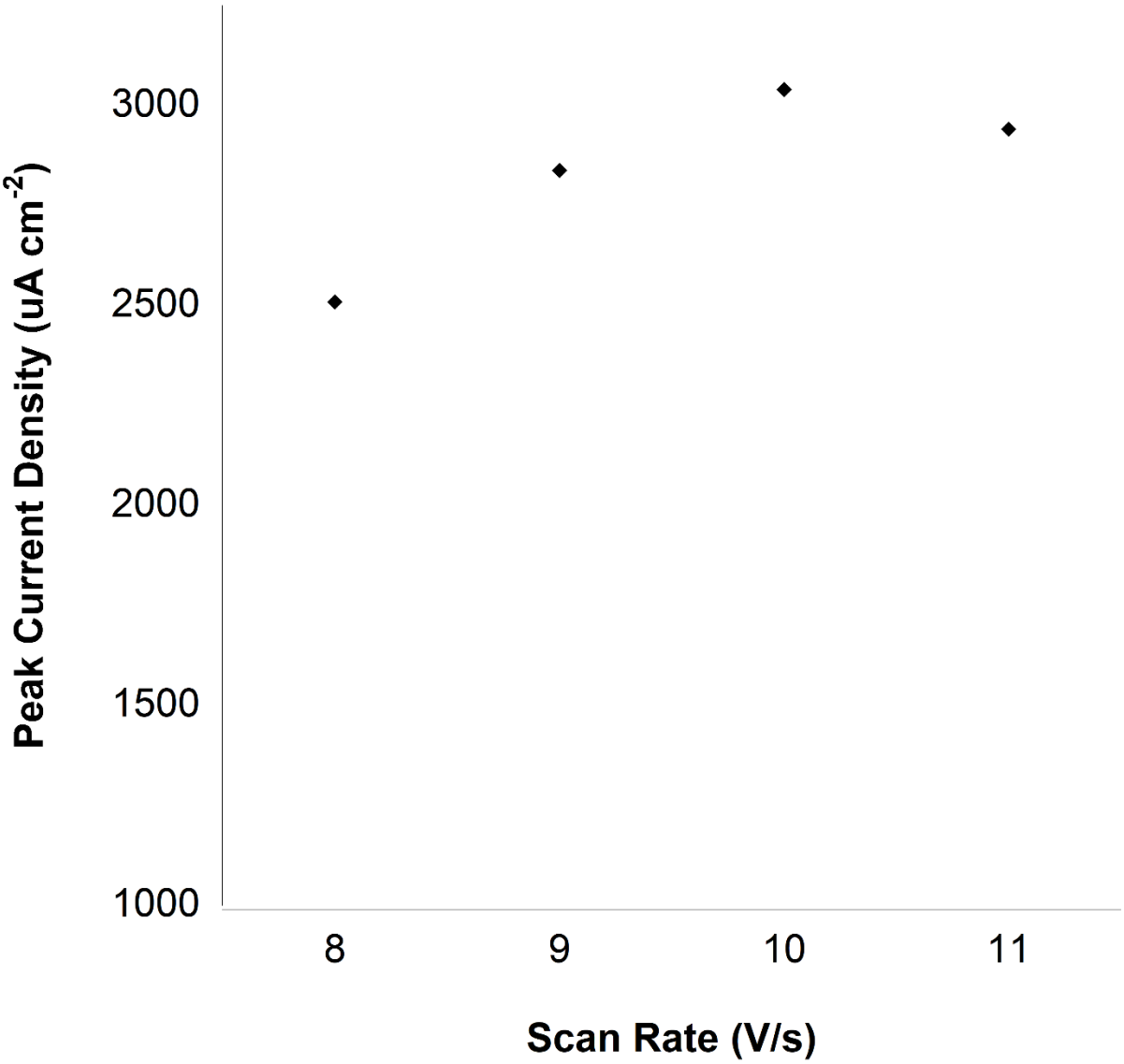


Figure S10. Peak current density (μA cm^{-2}) vs. scan rate (V/s) of **2**.

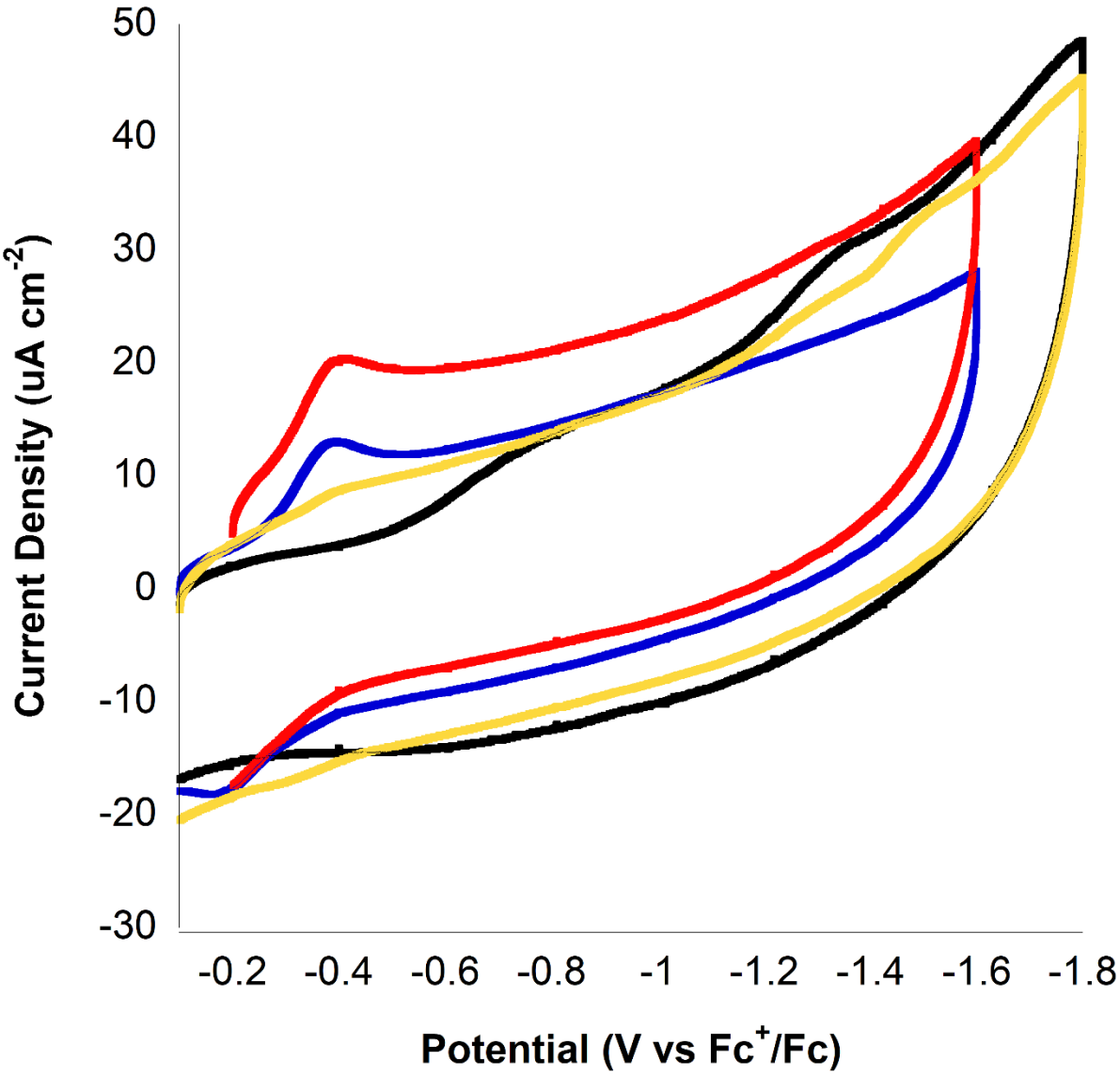


Figure S11. CVs of **1** (black), **2** (blue), **3** (red), and **4** (yellow) with no acid added. These experiments were performed in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM of **1-4** with a scan rate = 200 mV/s. [**2-4**] = 0.1 mM, [**1**] = 0.13 mM

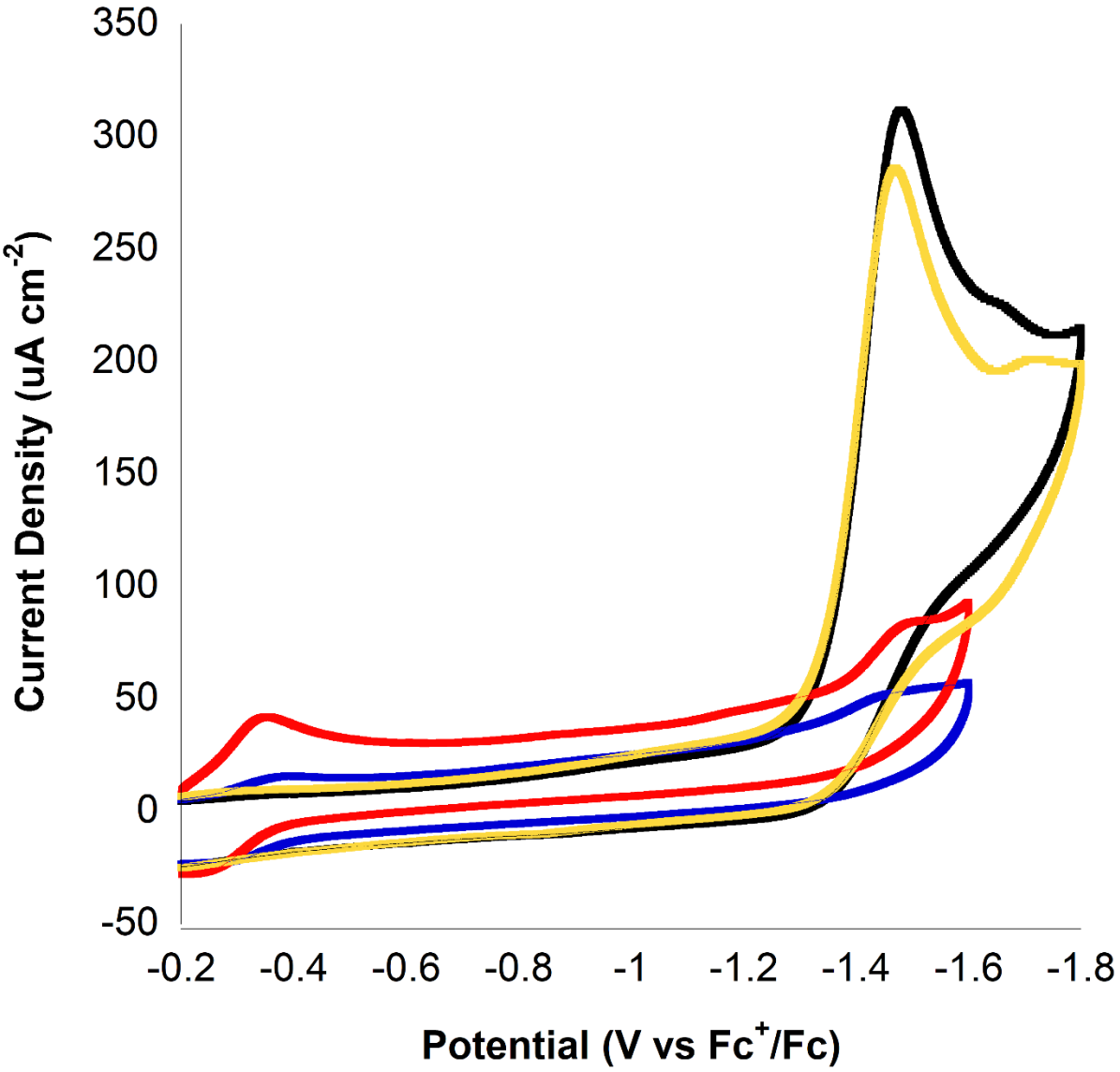


Figure S12. CVs of **1** (black), **2** (blue), **3** (red), and **4** (yellow) with 0.88 mM TFA added. These experiments were performed in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM of **1-4** with a scan rate = 200 mV/s. [**2-4**] = 0.1 mM, [**1**] = 0.13 mM

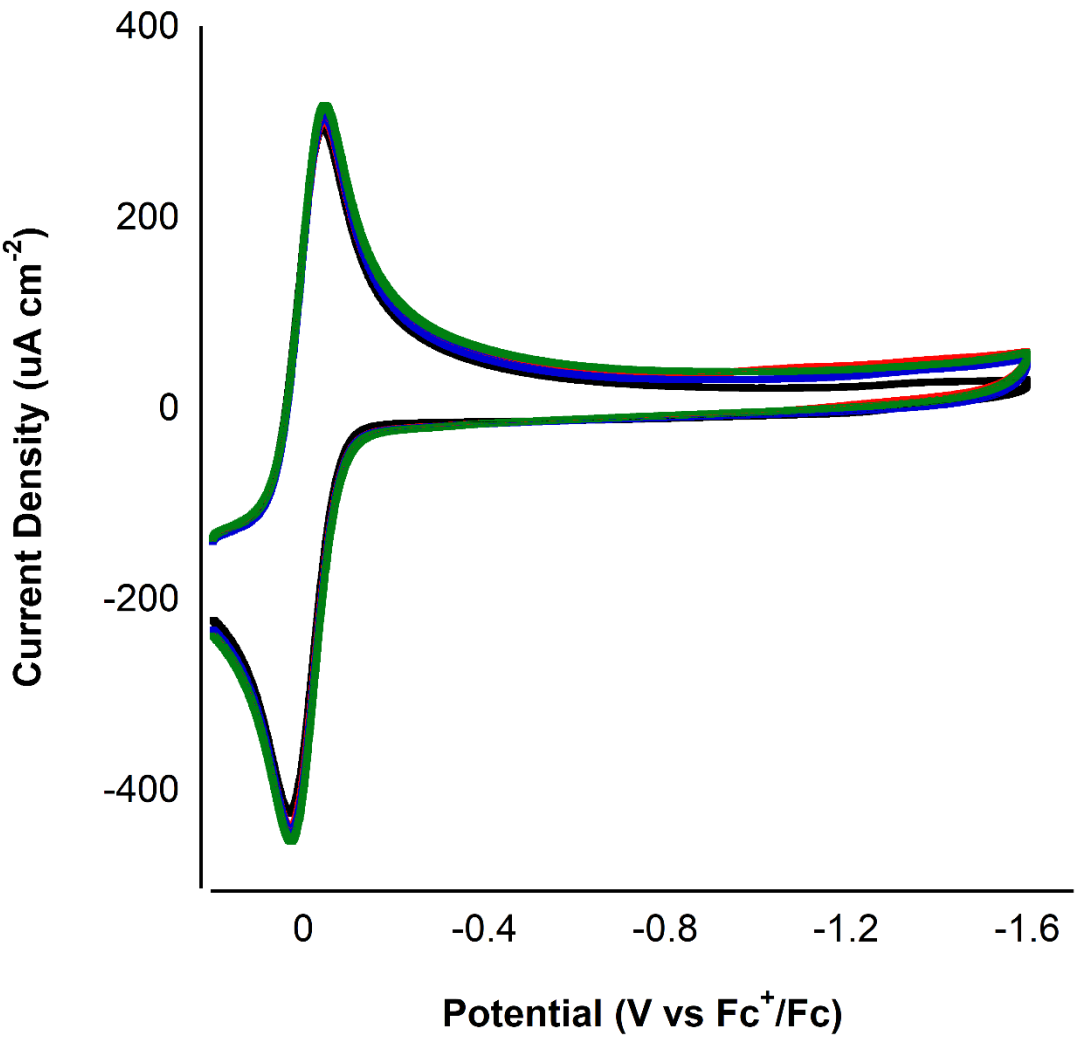


Figure S13. CVs of ferrocene with no acid (black), 0.44 mM (red), 0.66 mM (blue), and 0.88 mM (green) TFA. These experiments were performed in acetonitrile with 0.1 $TBAPF_6$ at a scan rate = 200 mV/s.

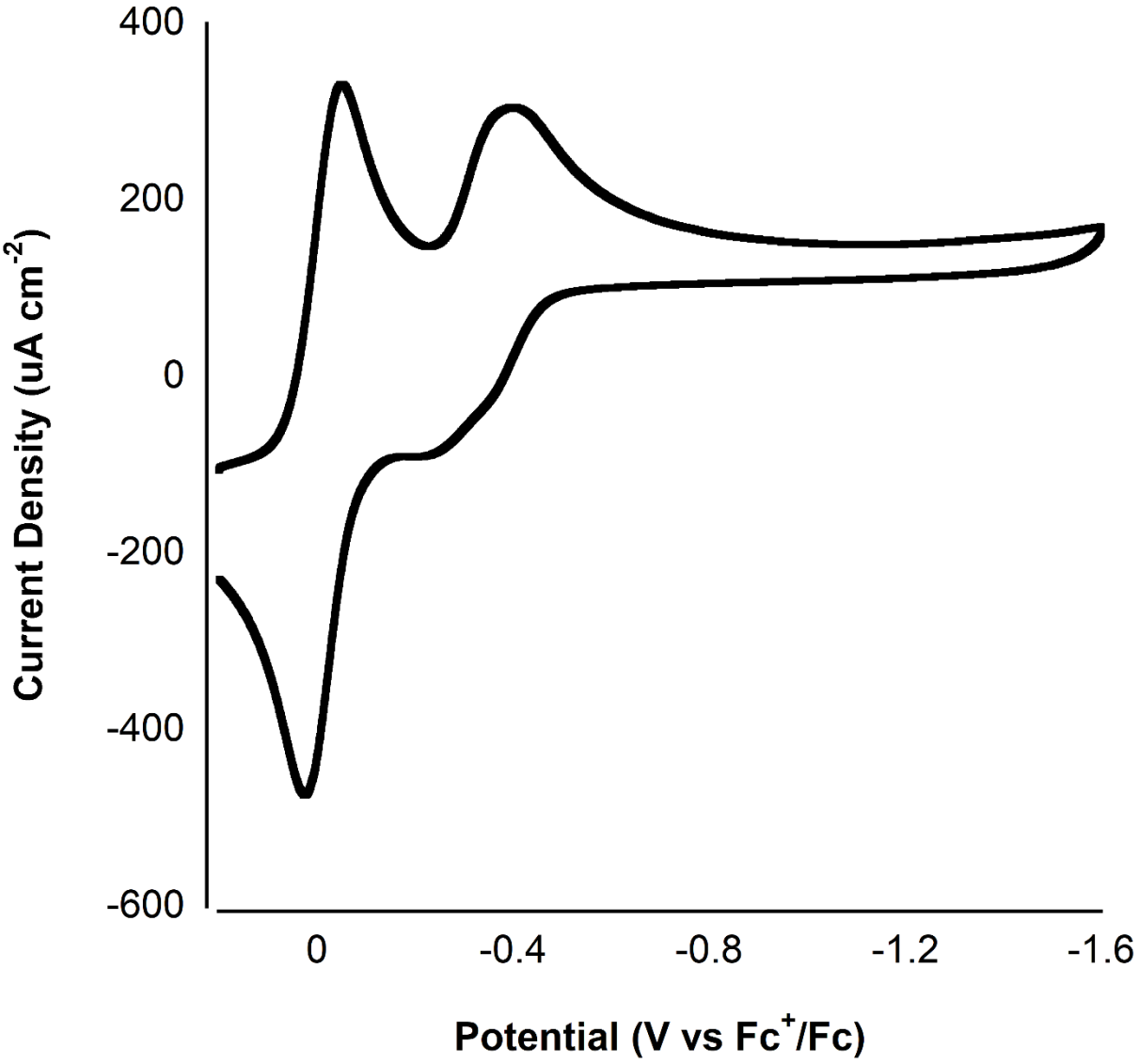


Figure S14. CV of ferrocene and $FeCl_3$ with 0.88 mM TFA. These experiments were performed in acetonitrile with 0.1 M $TBAPF_6$ at a scan rate = 200 mV/s.

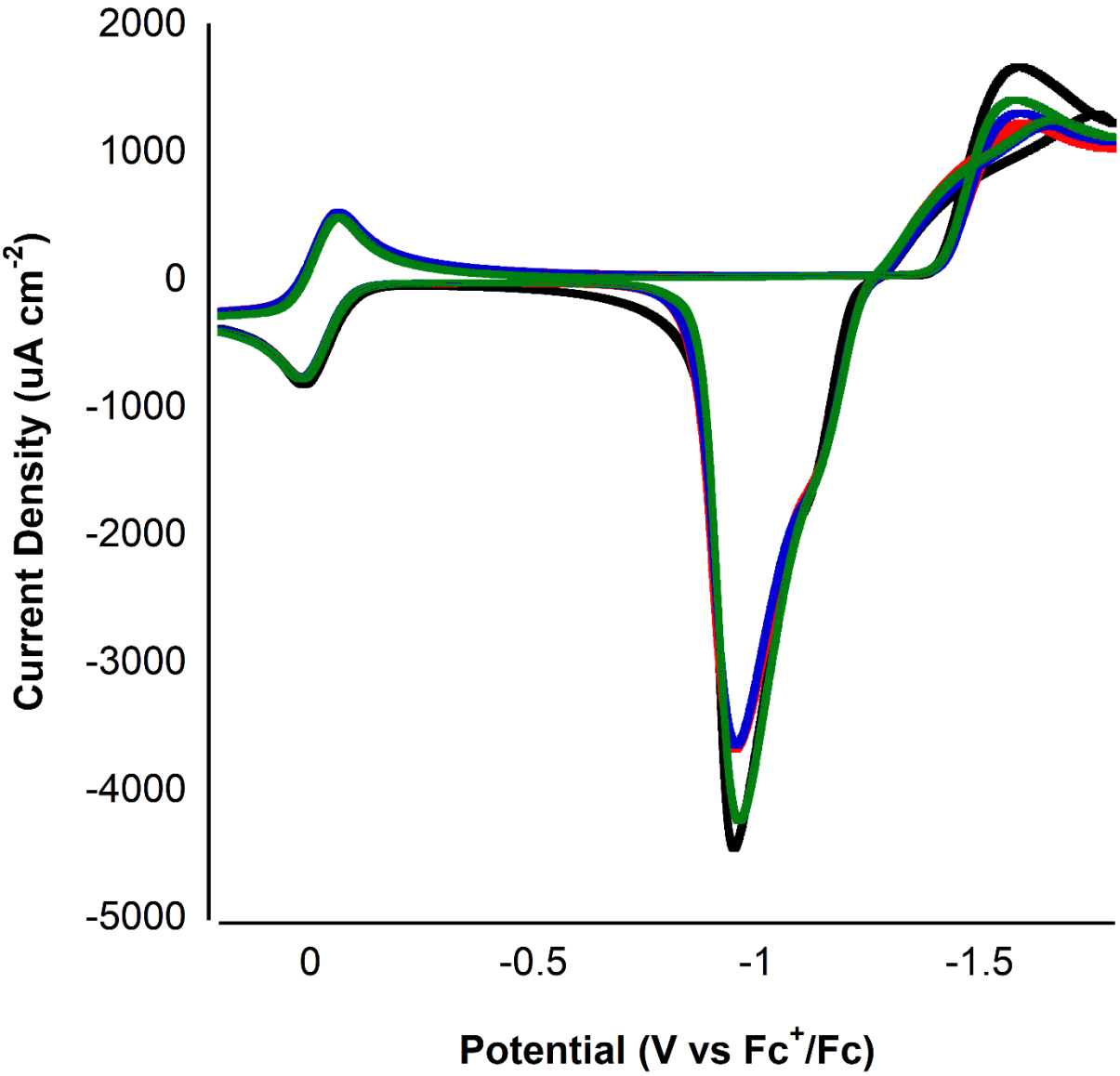


Figure S15. CV of ferrocene and $ZnCl_2$ with no acid (black), 0.44 mM (green), 0.66 mM (red), 0.88 mM (blue) TFA. performed in acetonitrile with 0.1 M $TBAPF_6$ at a scan rate = 200 mV/s.

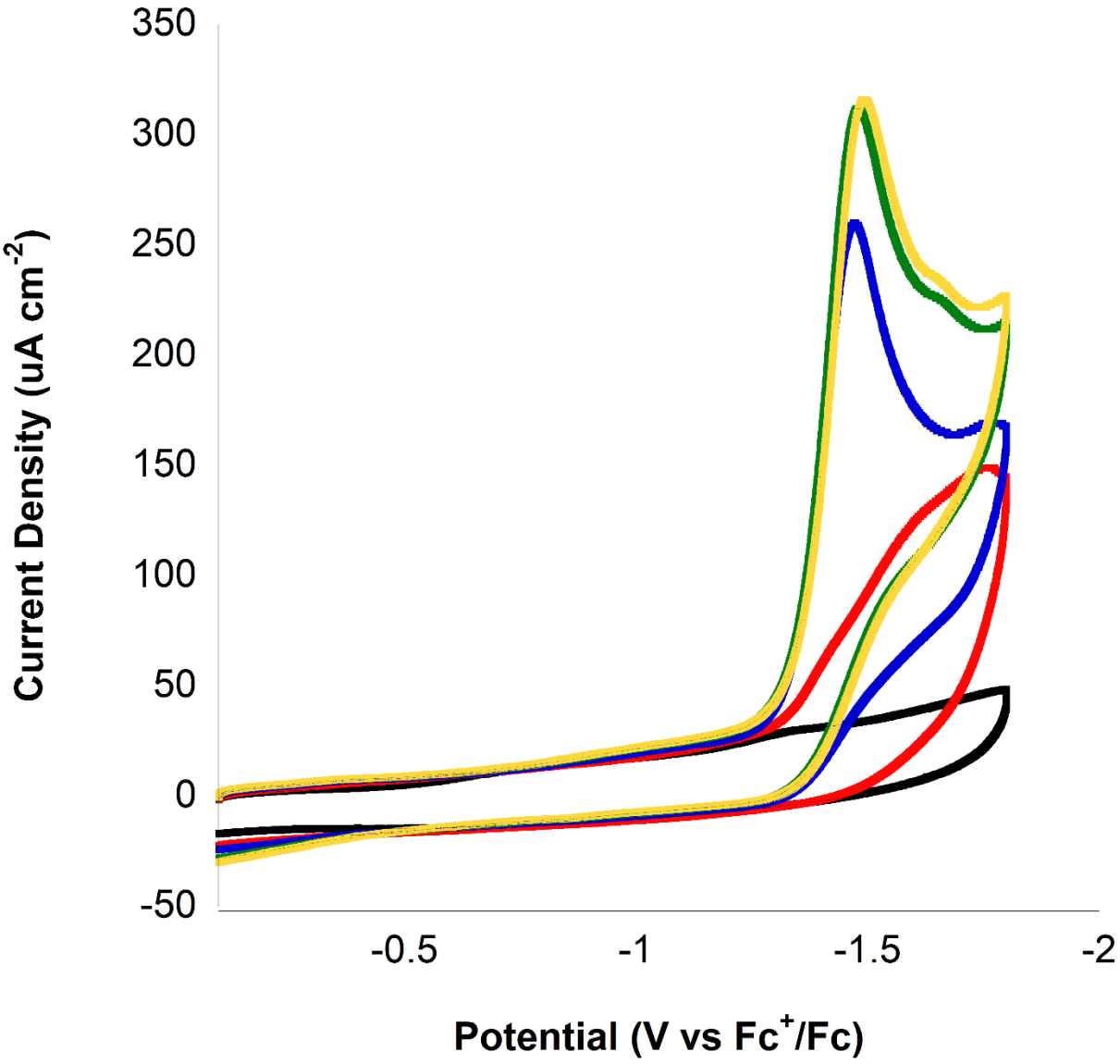


Figure S16. CVs of **1** with no acid (black), 0.44 mM (red), 0.66 mM (blue), 0.88 mM (green), and 1.1 mM (yellow) TFA. These experiments were performed in acetonitrile with 0.1 M $TBAPF_6$ and 0.131 mM **1** at a scan rate = 200 mV/s.

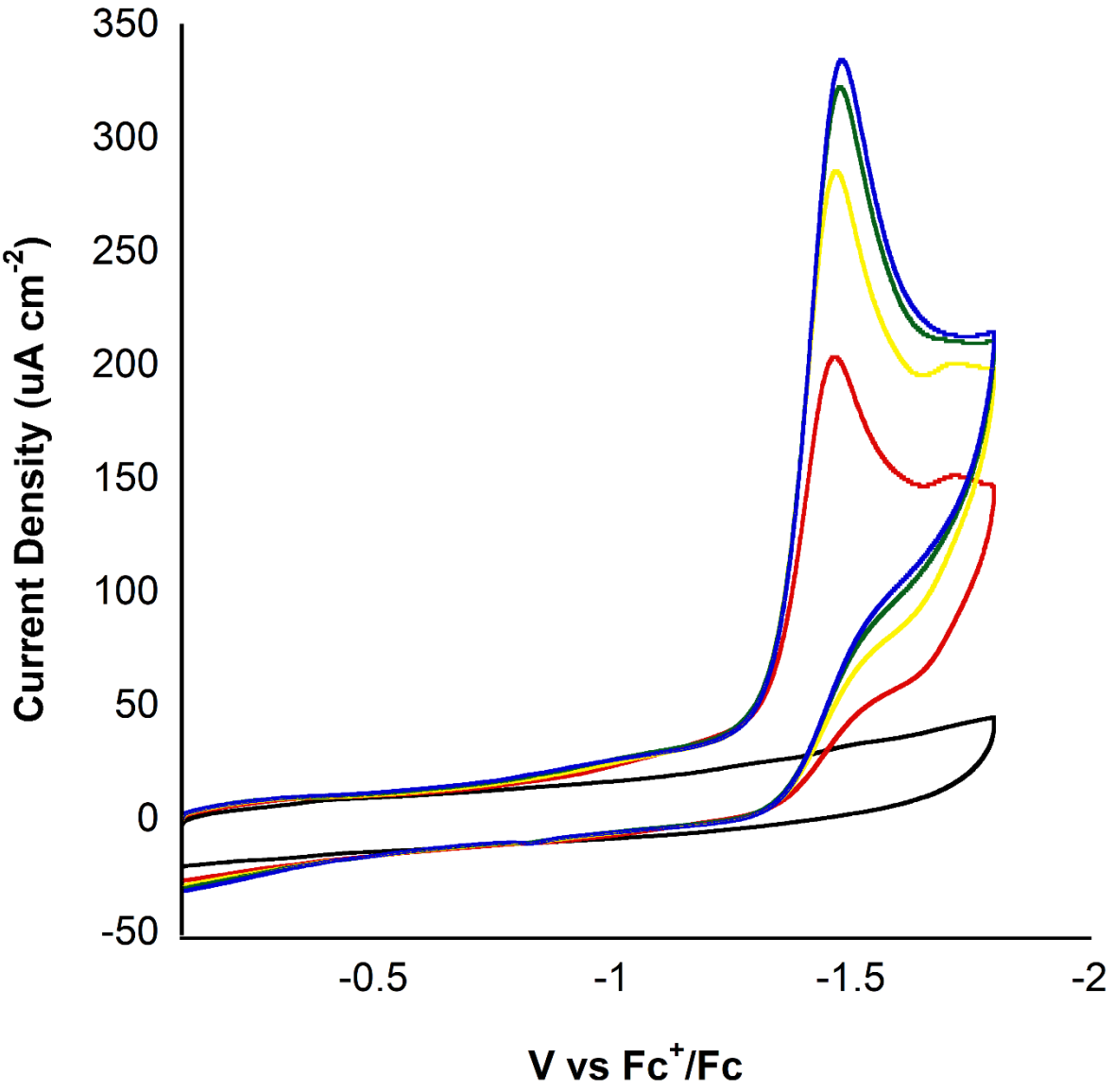


Figure S17. CVs of 0.1 mM **4** with no acid (black), 0.44 mM (red), 0.88 mM (yellow), 1.32 mM (green), and 1.76 mM (blue) TFA in acetonitrile with 0.1 M $TBAPF_6$ and 0.101 mM **4** at a scan rate = 200 mV/s.

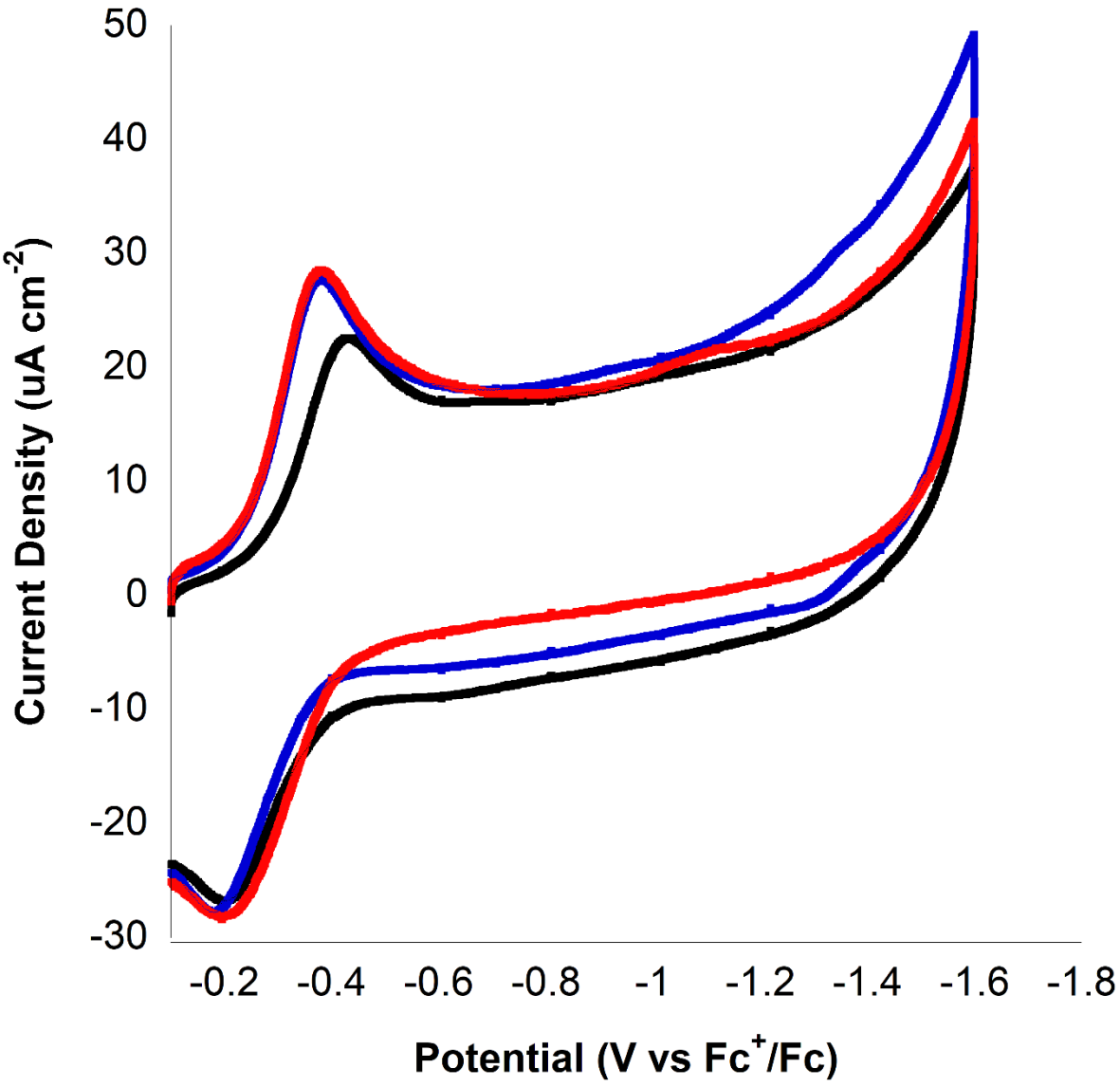


Figure S18. CVs of 0.13 mM **2** with 1.32 mM phenol (black), acetic acid (blue), and salicylic acid (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **2** at a scan rate = 200 mV/s.

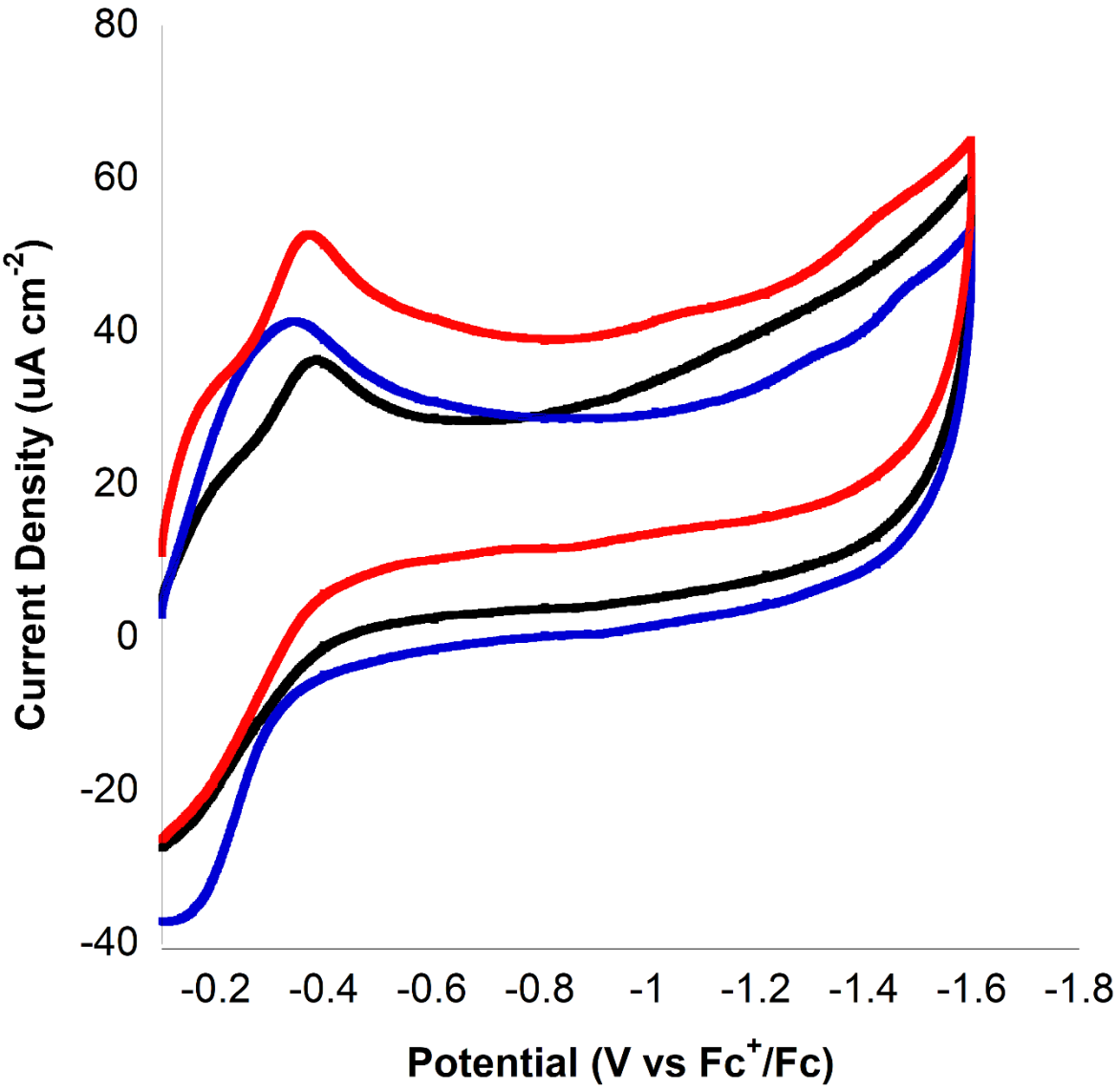


Figure S19. CVs of 0.13 mM **3** with 1.32 mM phenol (black), acetic acid (blue), and salicylic acid (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **3** at a scan rate = 200 mV/s.

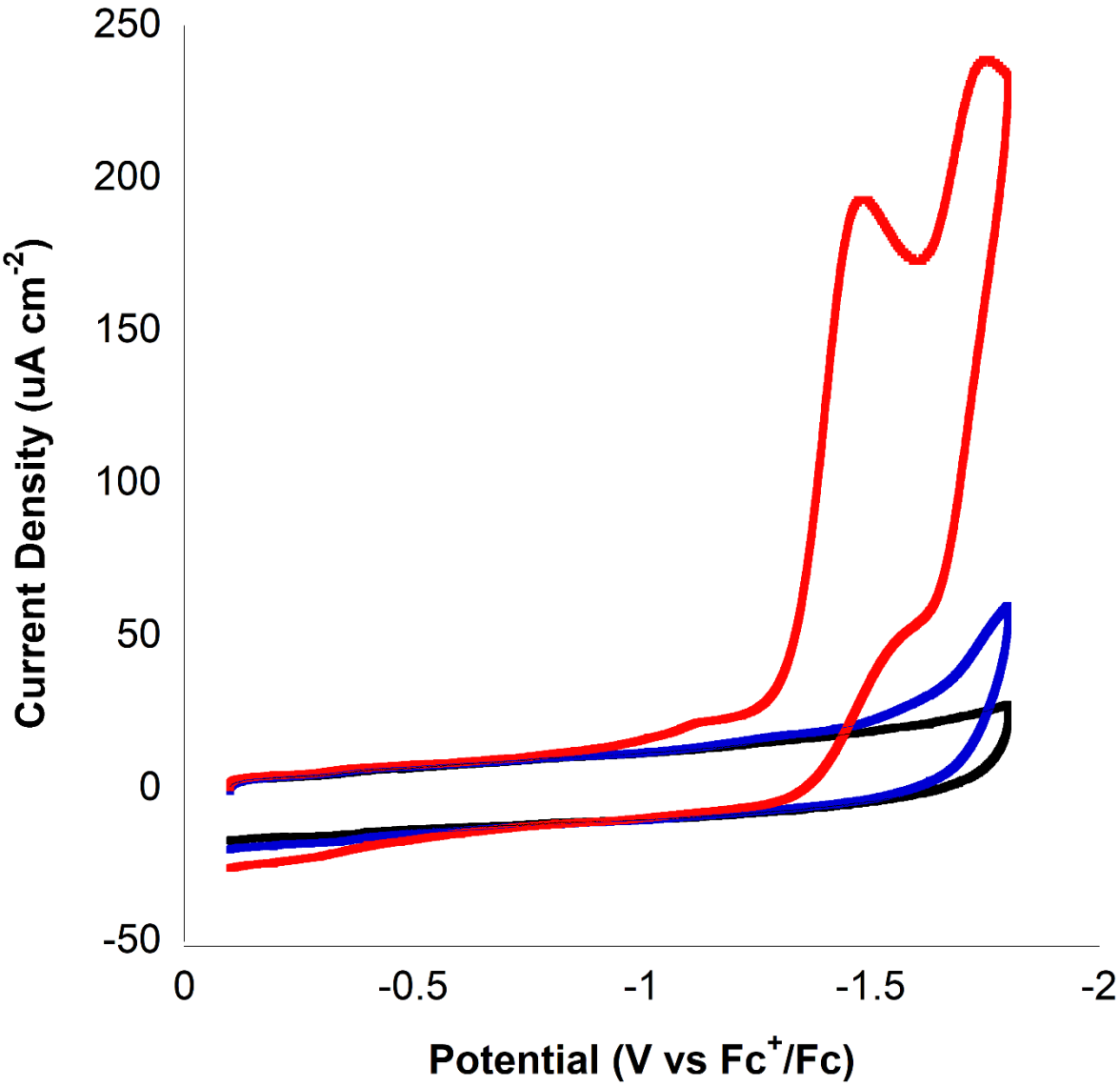


Figure S20. CVs of 0.13 mM **4** with 1.32 mM phenol (black), acetic acid (blue), and salicylic acid (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **4** at a scan rate = 200 mV/s.

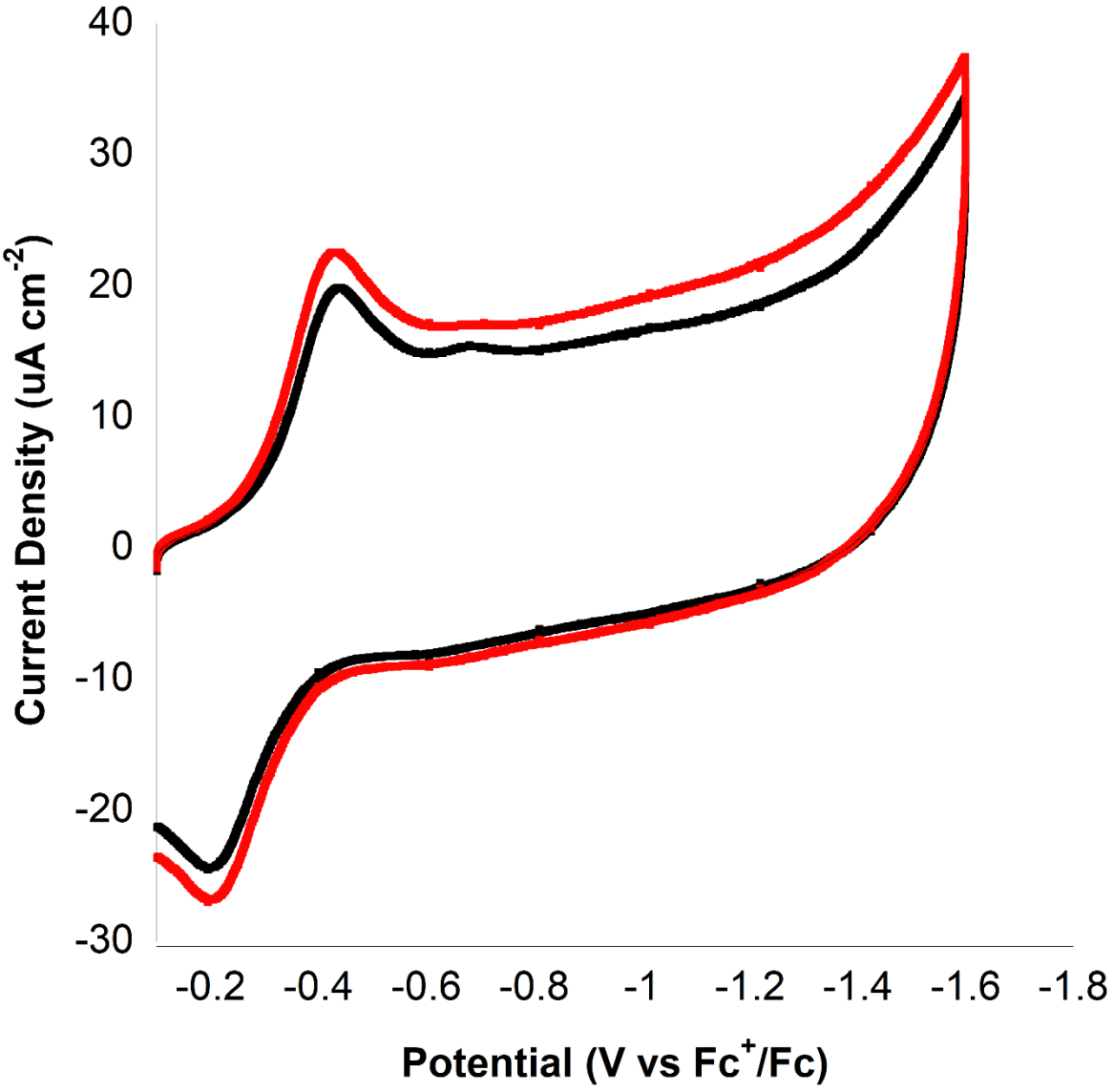


Figure S21. CVs of 0.13 mM **2** with no acid (black) and 1.32 mM phenol (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **2** at a scan rate = 200 mV/s.

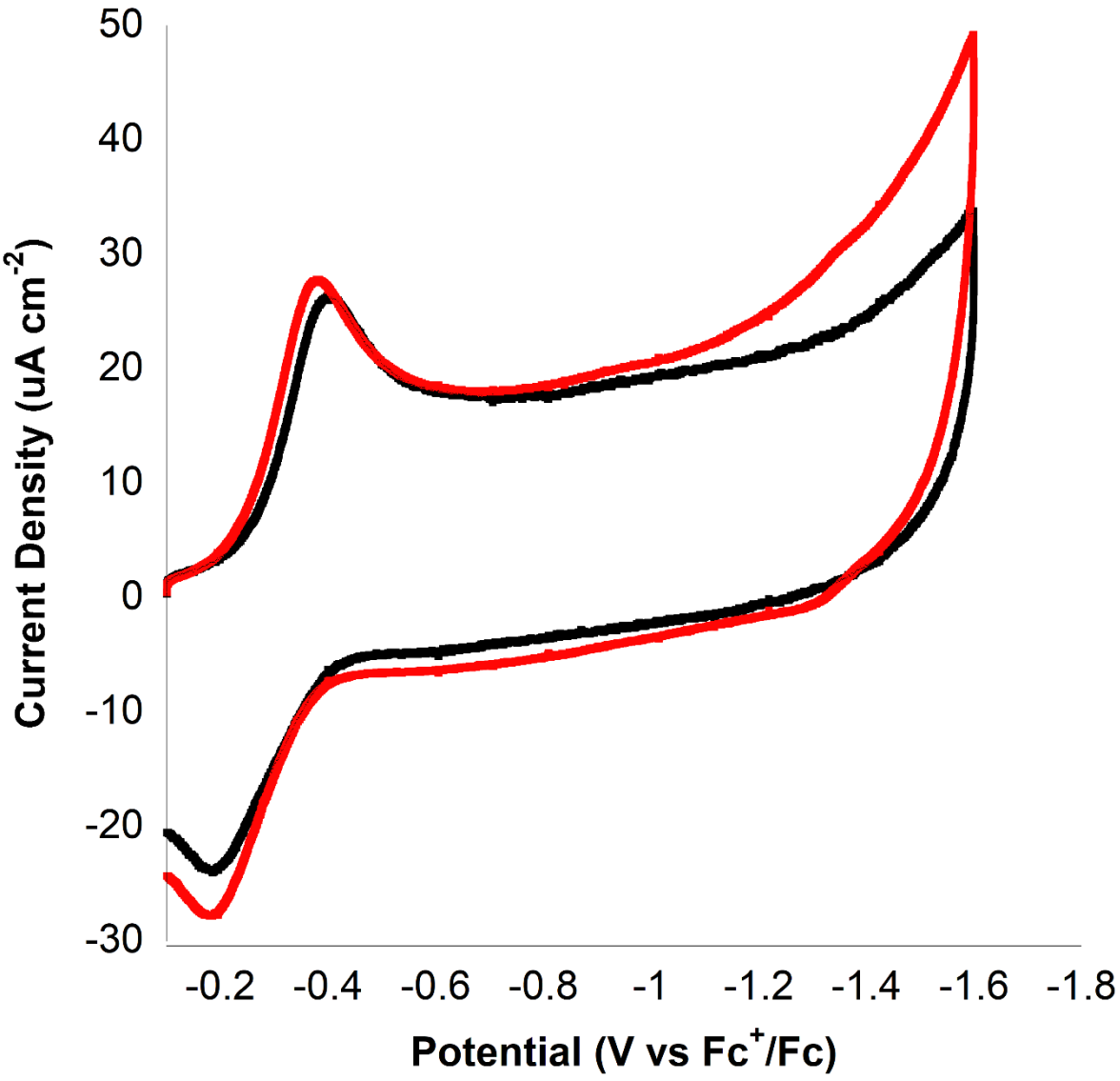


Figure S22. CVs of 0.13 mM **2** with no acid (black) 1.32 mM acetic acid (red) acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **2** at a scan rate = 200 mV/s.

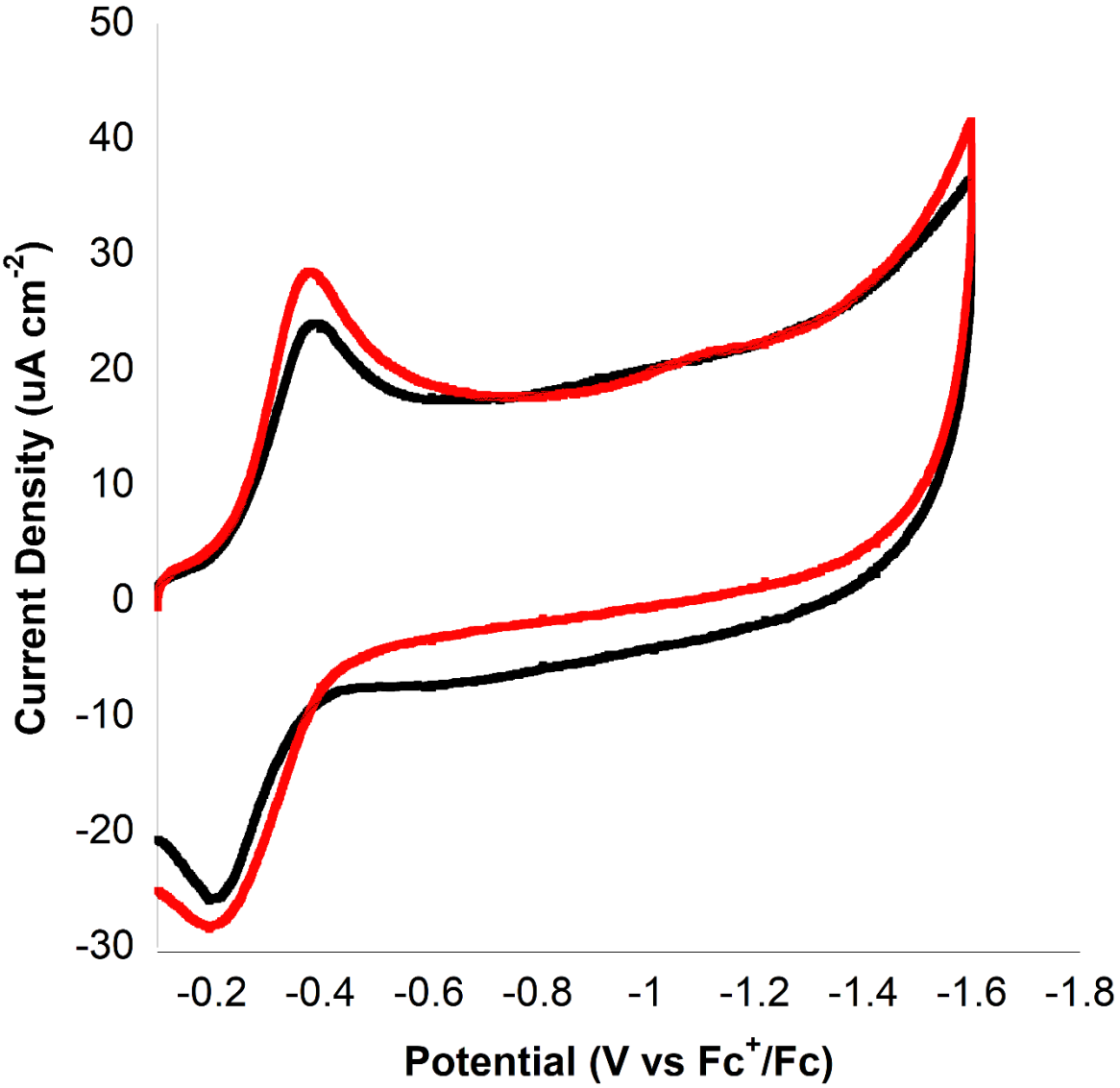


Figure S23. CVs of 0.13 mM **2** with no acid (black) and 1.32 mM salicylic acid (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **2** at a scan rate = 200 mV/s.

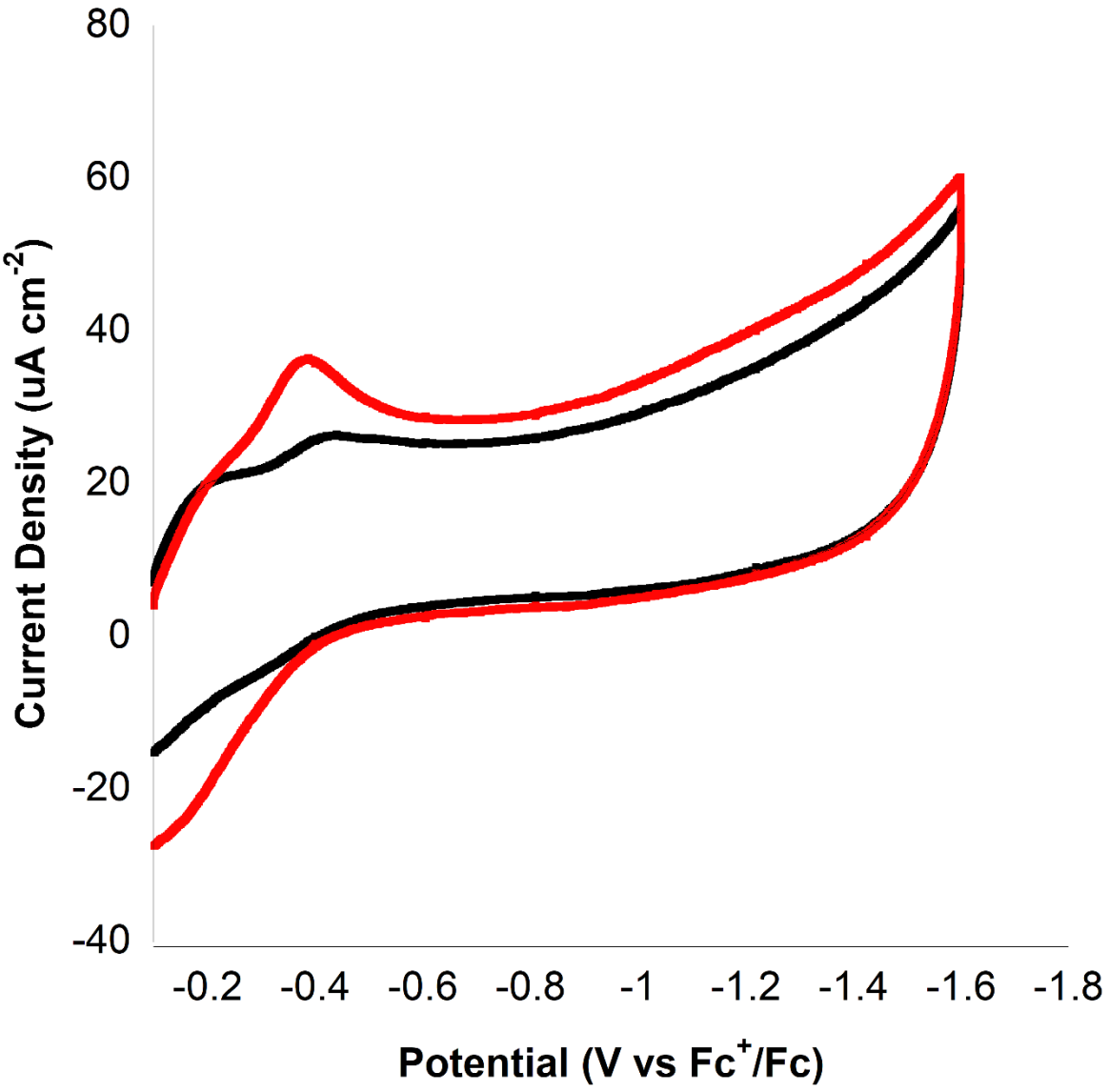


Figure S24. CVs of 0.1 mM **3** with no acid (black) and 1.32 mM phenol (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **3** at a scan rate = 200 mV/s.

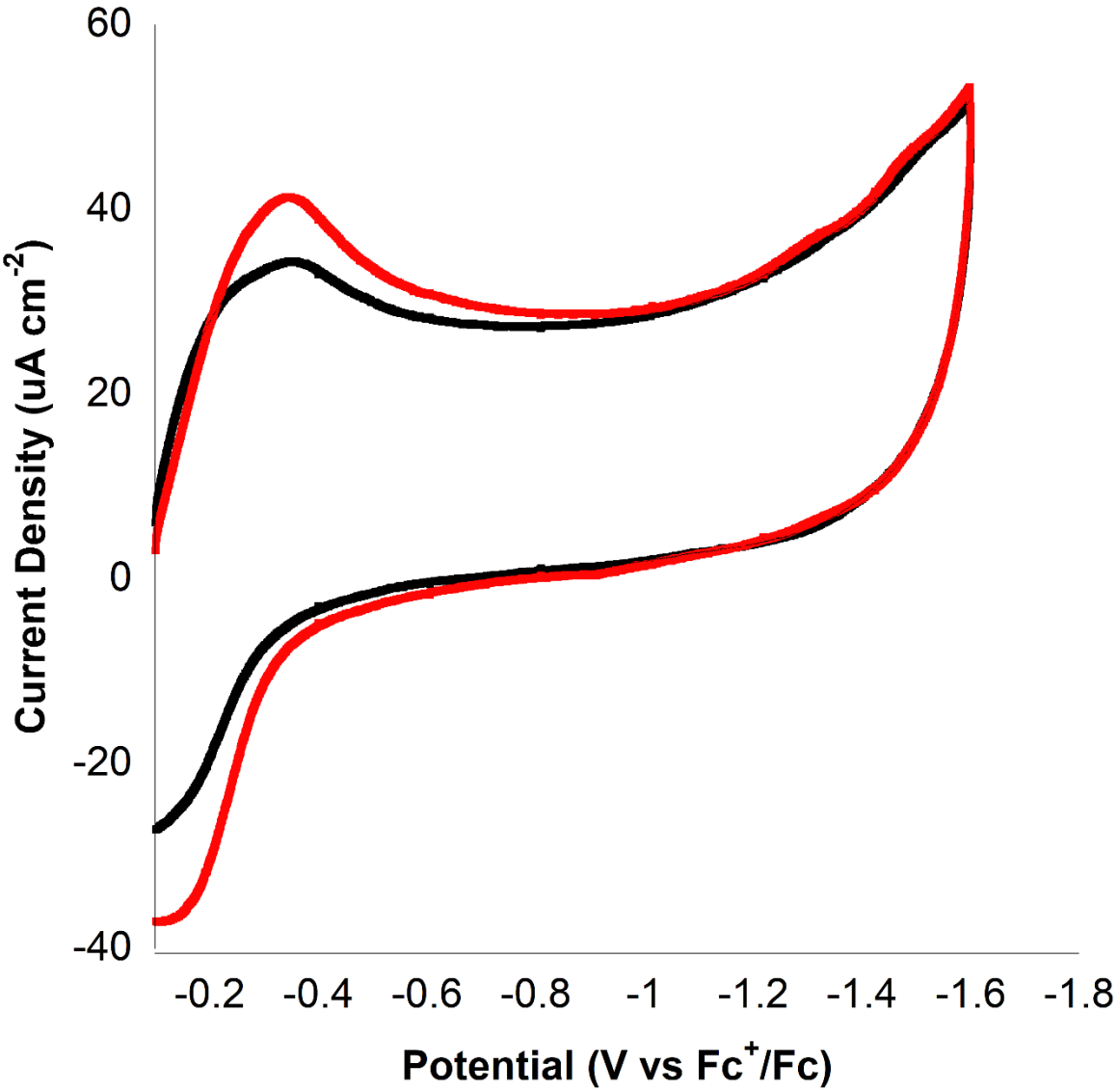


Figure S25. CVs of 0.13 mM **3** with no acid (black) and 1.32 mM acetic acid (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **3** at a scan rate = 200 mV/s.

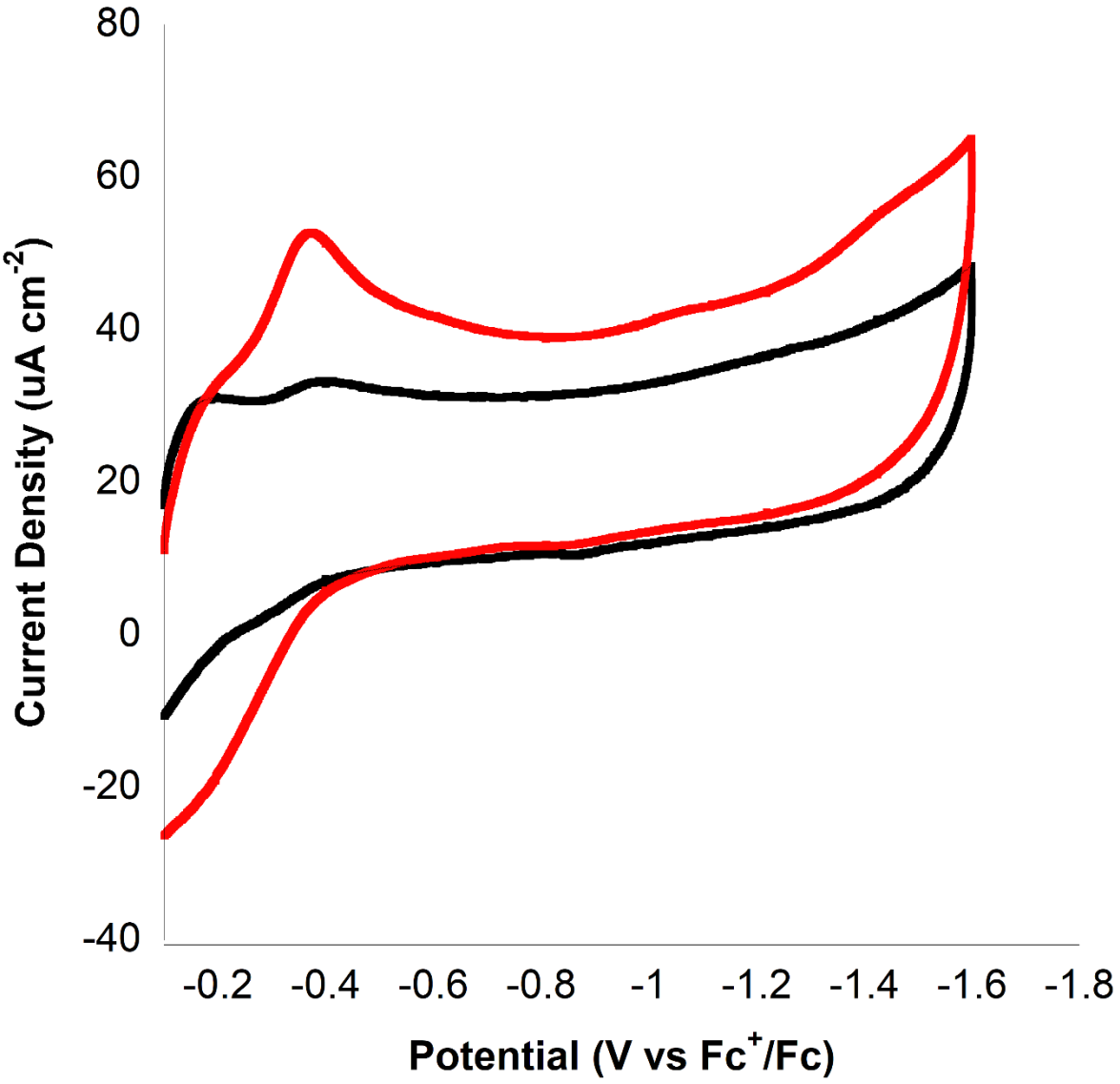


Figure S26. CVs of 0.1 mM **3** with no acid (black) and 1.32 mM salicylic acid (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **3** at a scan rate = 200 mV/s.

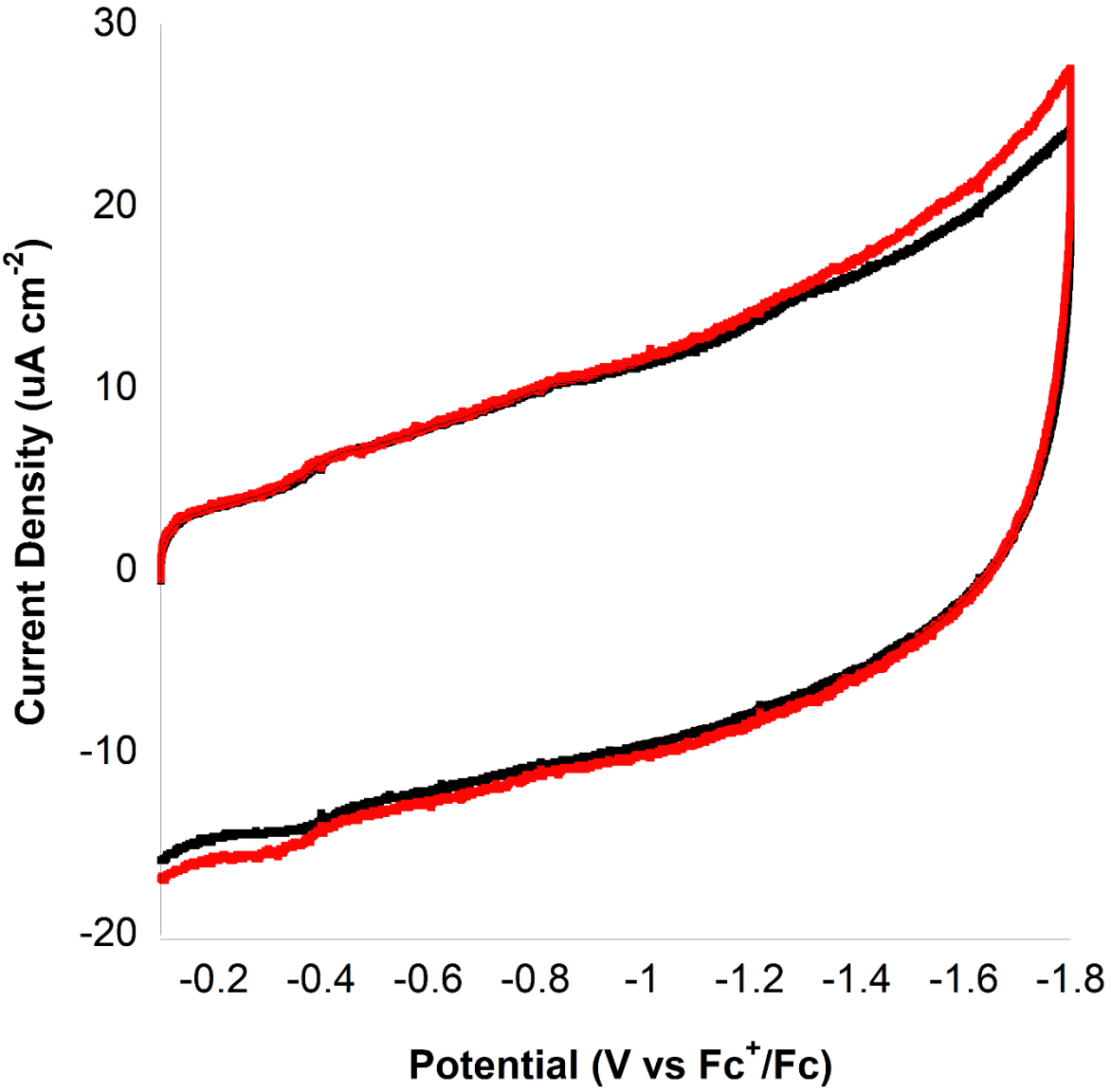


Figure S27. CVs of 0.13 mM **4** with no acid (black) and 1.32 mM phenol (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **4** at a scan rate = 200 mV/s.

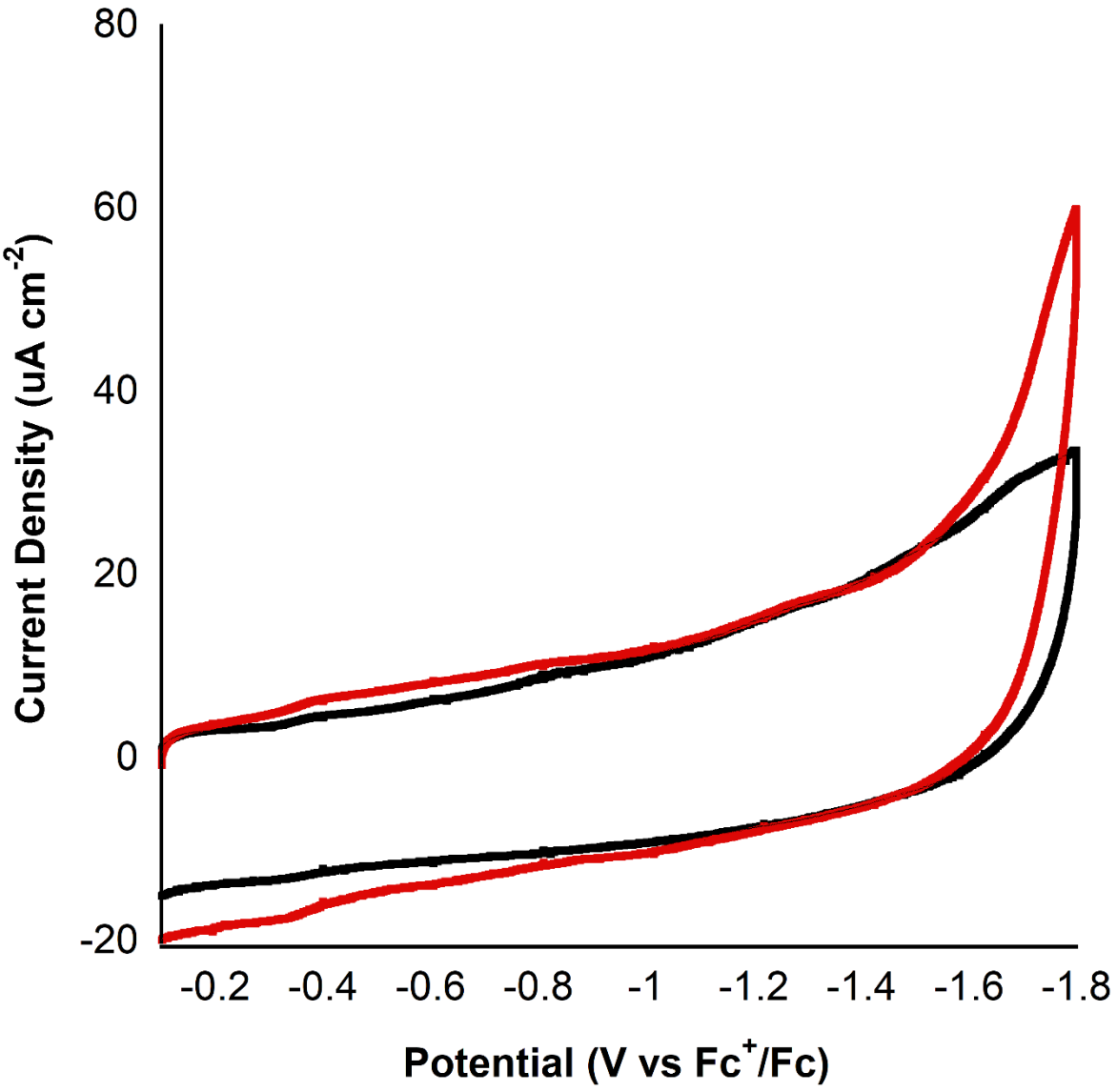


Figure S28. CVs of 0.13 mM **4** with no acid (black) and 1.32 mM acetic acid (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **4** at a scan rate = 200 mV/s.

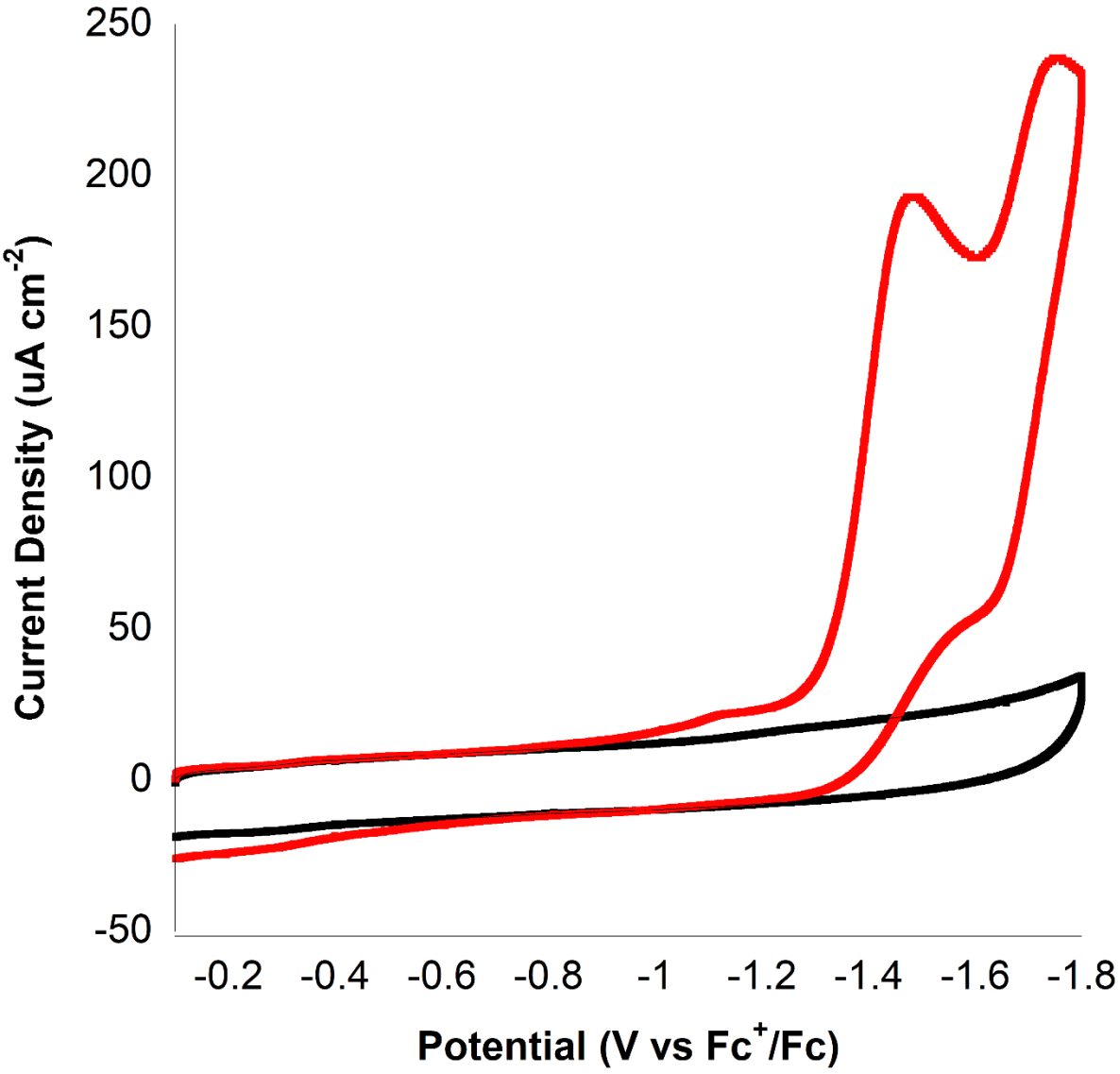


Figure S29. CVs of 0.13 mM **4** with no acid (black) and 1.32 mM salicylic acid (red) in acetonitrile with 0.1 M $TBAPF_6$ and 0.1 mM **4** at a scan rate = 200 mV/s.

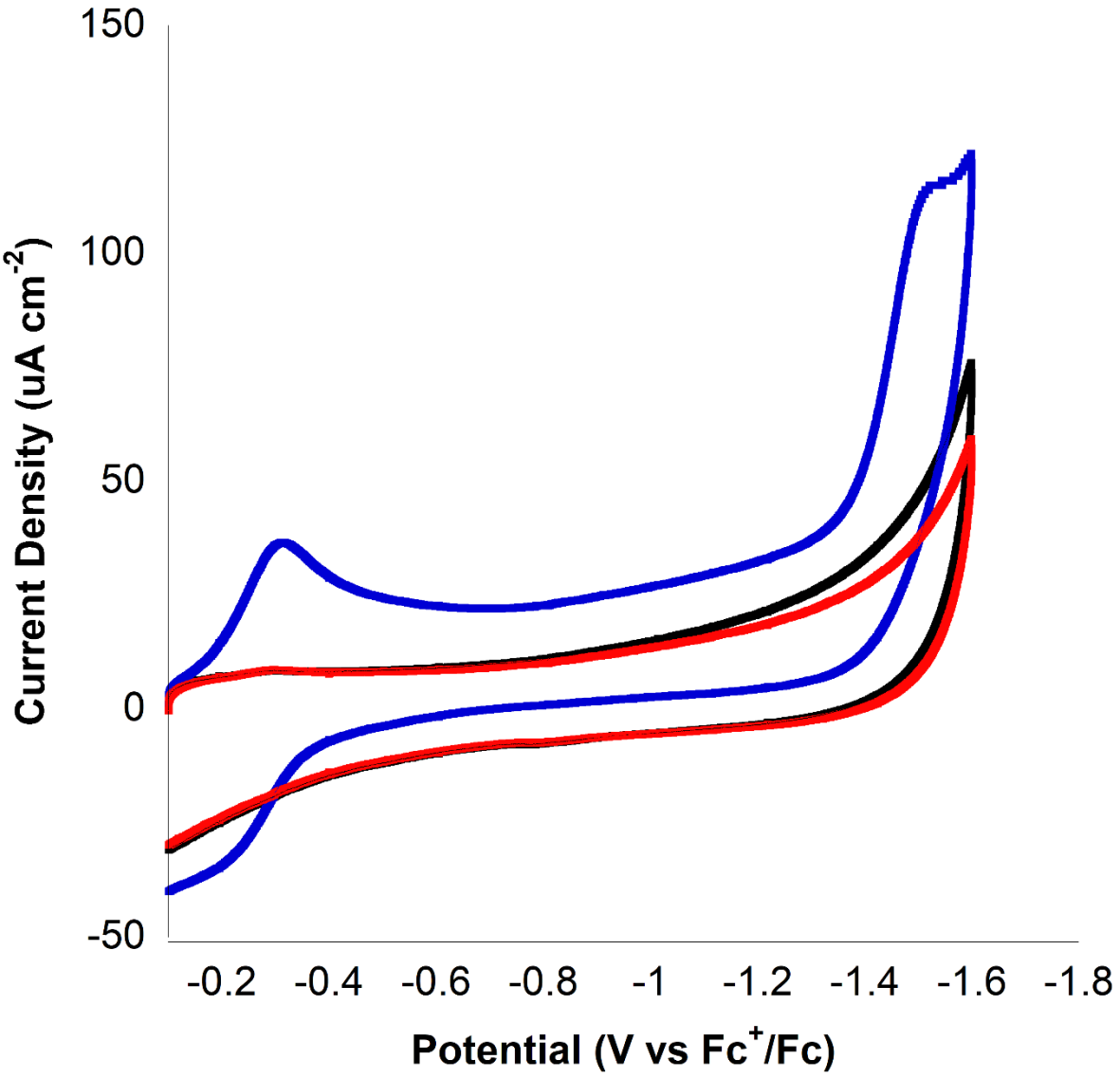


Figure S30. Rinse test for **2** in 2.2 mM TFA with 0.1 M $TBAPF_6$ and 0.1 mM **2** at a scan rate = 200 mV/s. CVs were taken in a blank cell with only TFA and $TBAPF_6$ (black), in a separate cell with an acid addition to **2** (blue), and in the blank cell after lightly rinsing the electrodes from the acid addition with **2** (red) with acetonitrile.

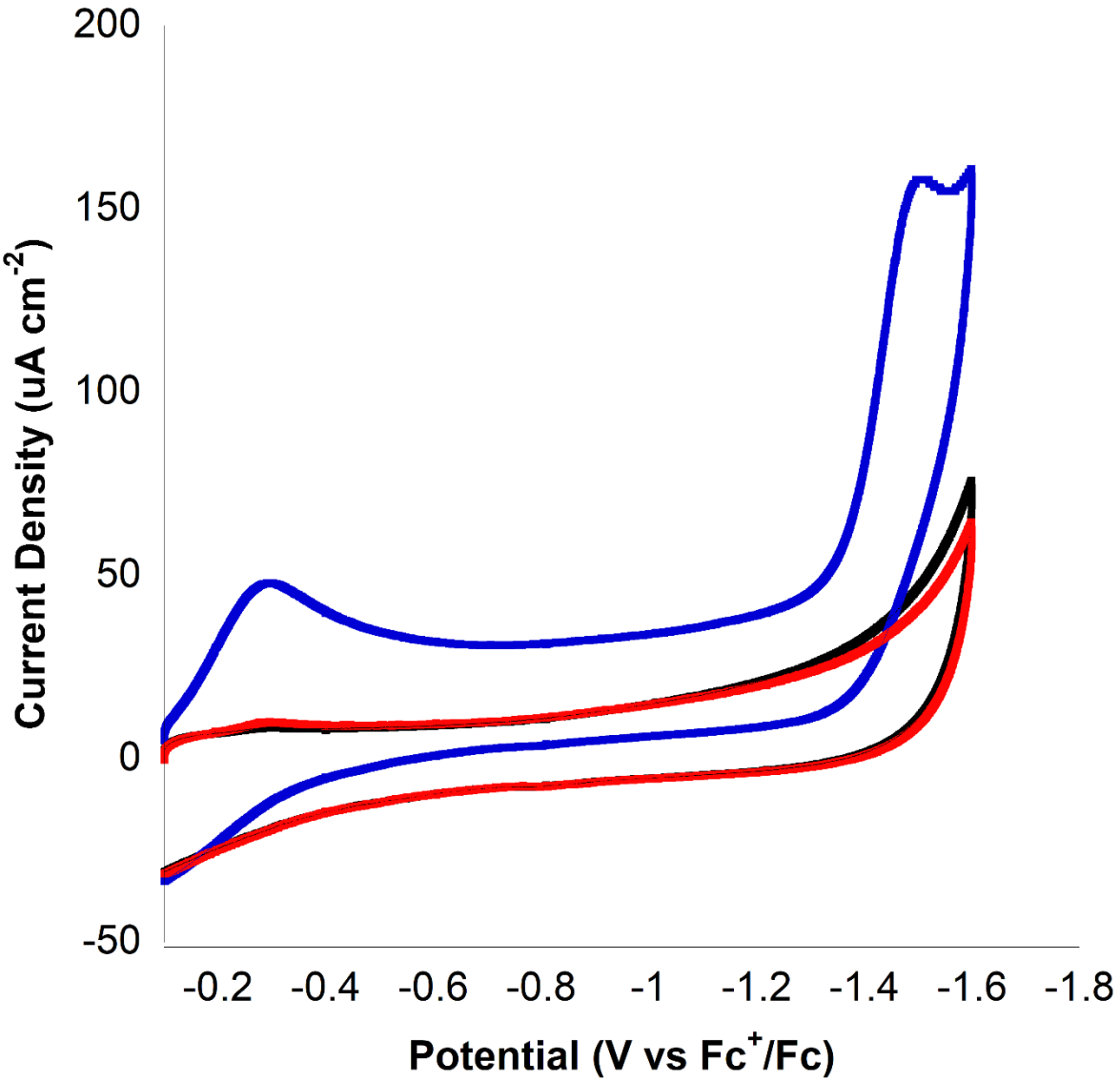


Figure S31. Rinse test for **3** in 2.2 mM TFA with 0.1 M $TBAPF_6$ and 0.1 mM **3** at a scan rate = 200 mV/s. CVs were taken in a blank cell with only TFA and $TBAPF_6$ (black), in a separate cell with an acid addition to **3** (blue), and in the blank cell after lightly rinsing the electrodes from the acid addition with **3** (red) with acetonitrile.

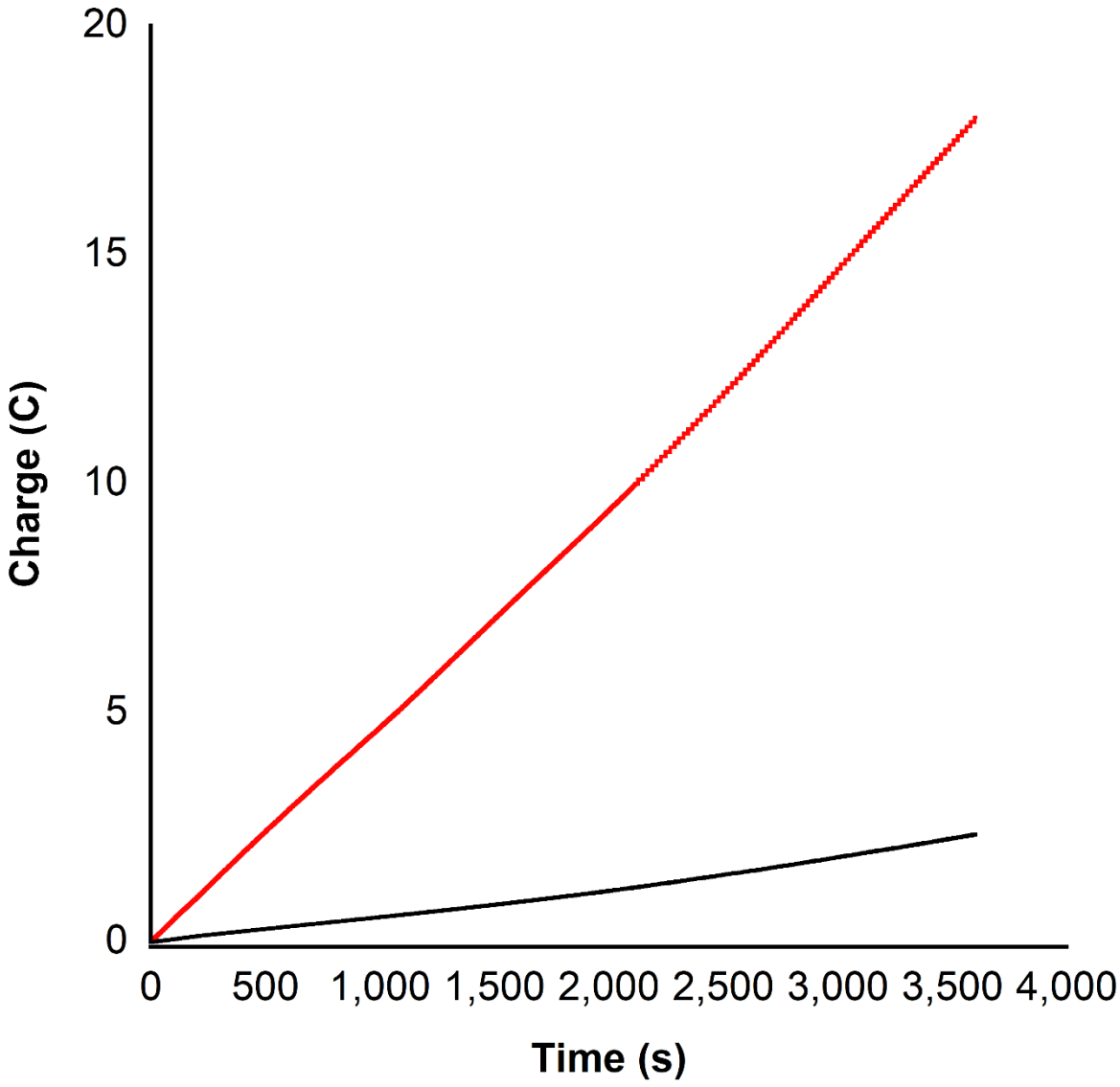


Figure S32. CPC using glassy carbon electrode with no catalyst (black) and with **2** (red). The bulk electrolysis was run in a 0.1 M solution of $TBAPF_6$ in acetonitrile with 65 mM TFA. The potential was held at -1.5 V vs. Fc^+/Fc for 3600 seconds resulting in a charge of 17.9 C. A GC of the headspace gases corresponded to a faradaic yield of 99%.

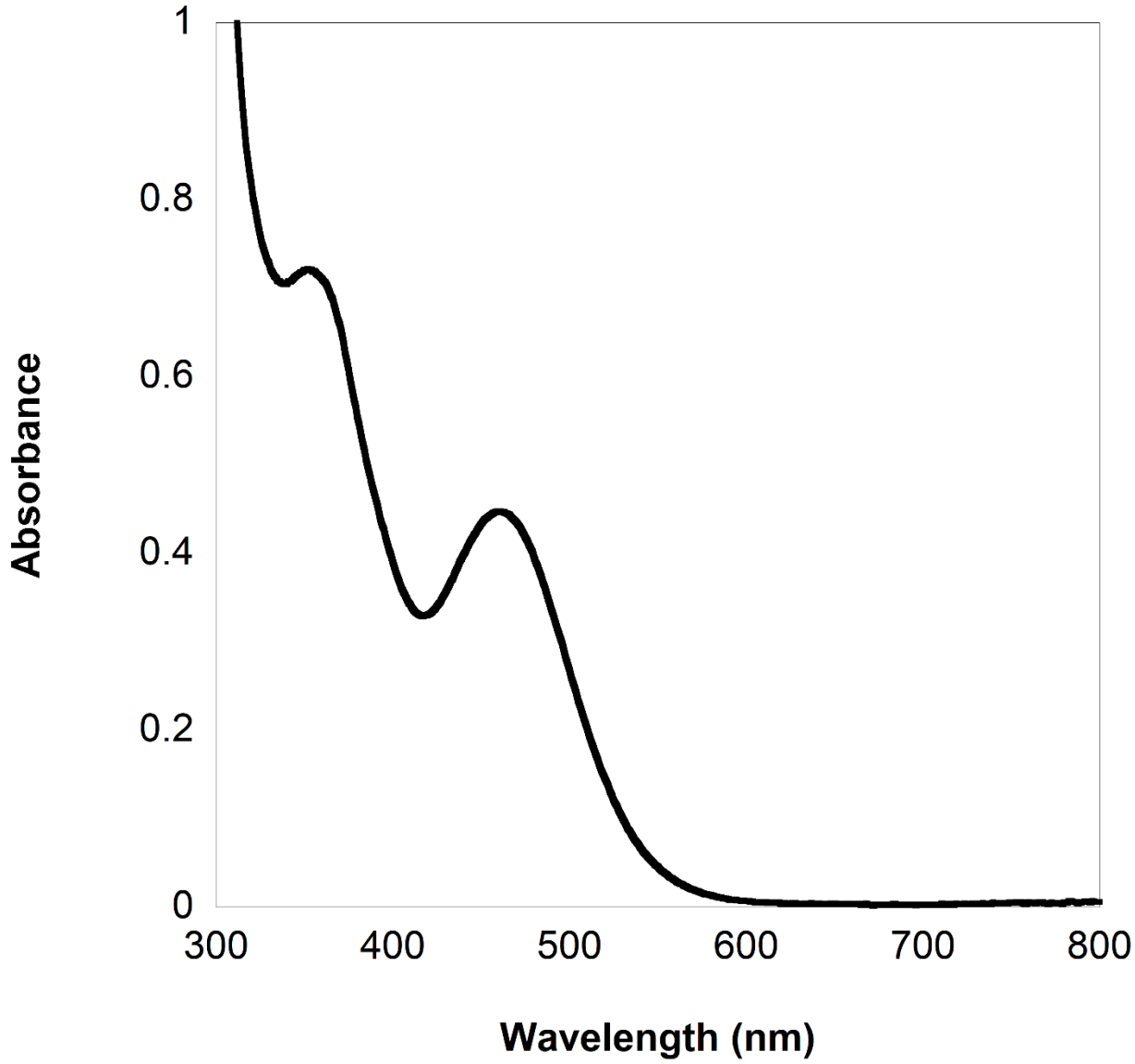


Figure S33. UV/vis spectrum of **2** in DCM.

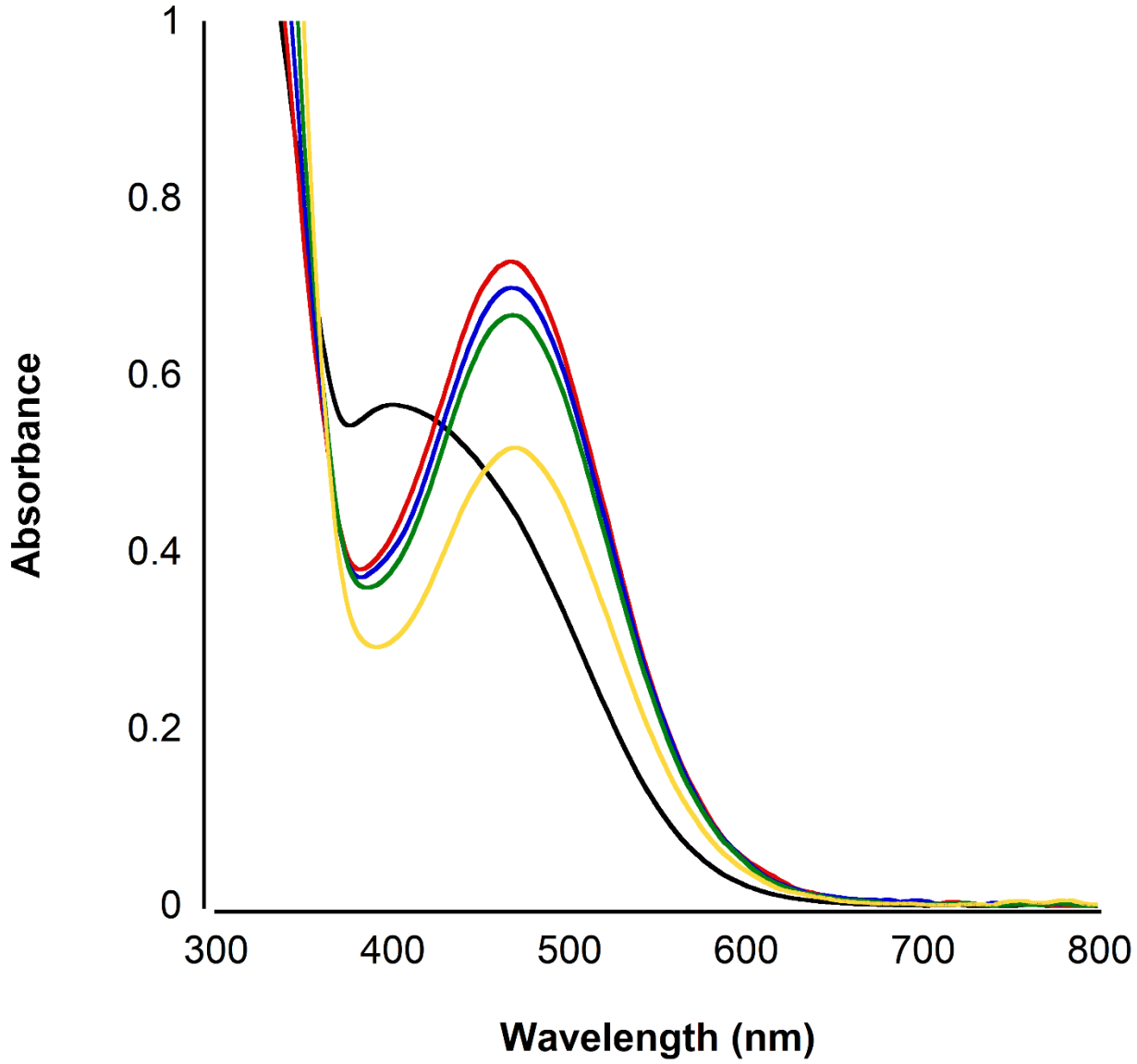


Figure S34. UV/vis spectrum of **2** in 1:1 ethanol:deionized water at pH 7 (black), pH 6 (red), pH 5 (blue), pH 4 (green), and pH 3 (yellow).

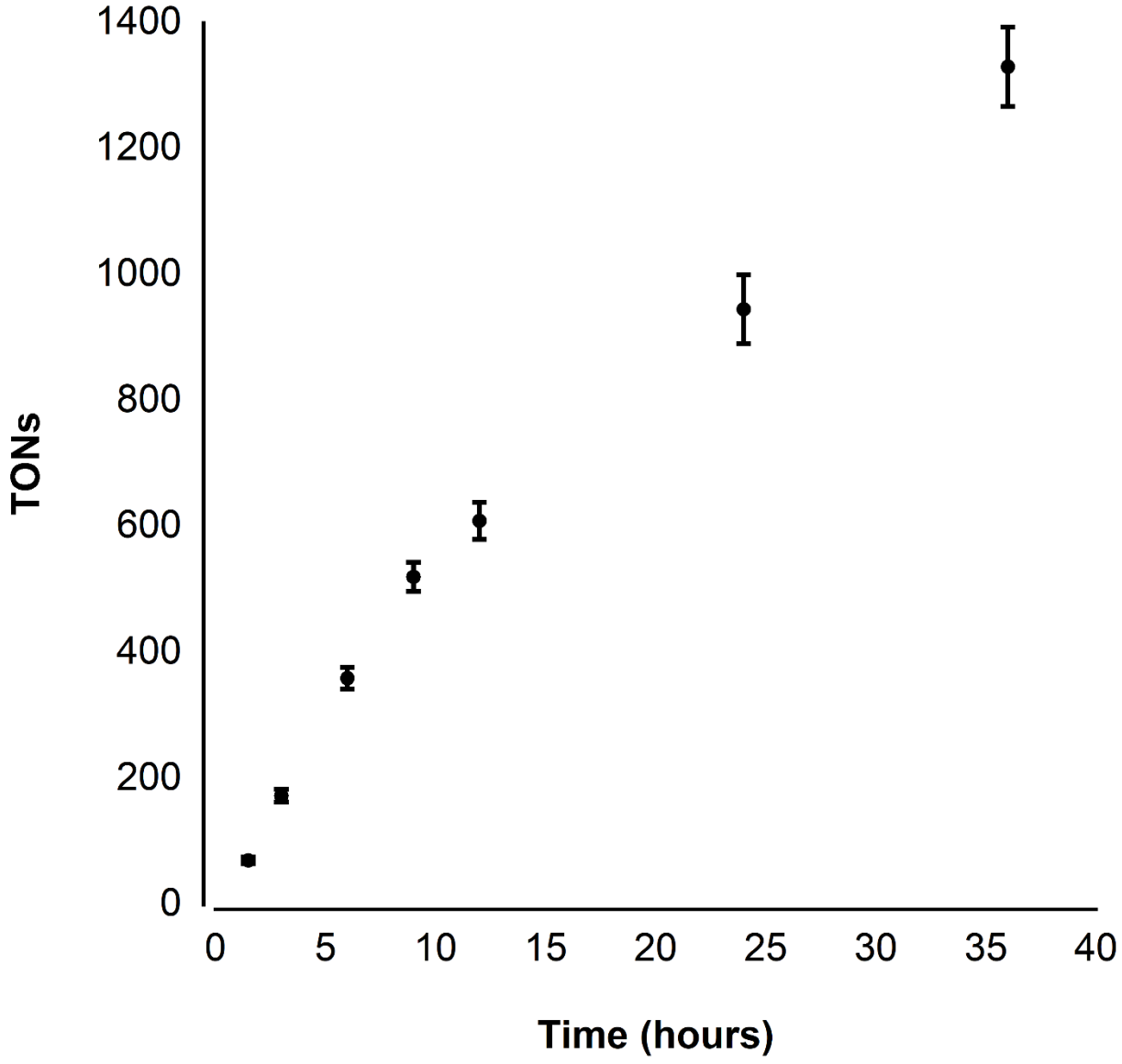


Figure S35. Hydrogen Generation observed for 7.5 μM **3** when paired with 1.8 mM Fl and 5% TEA in 1:1 EtOH:Water.

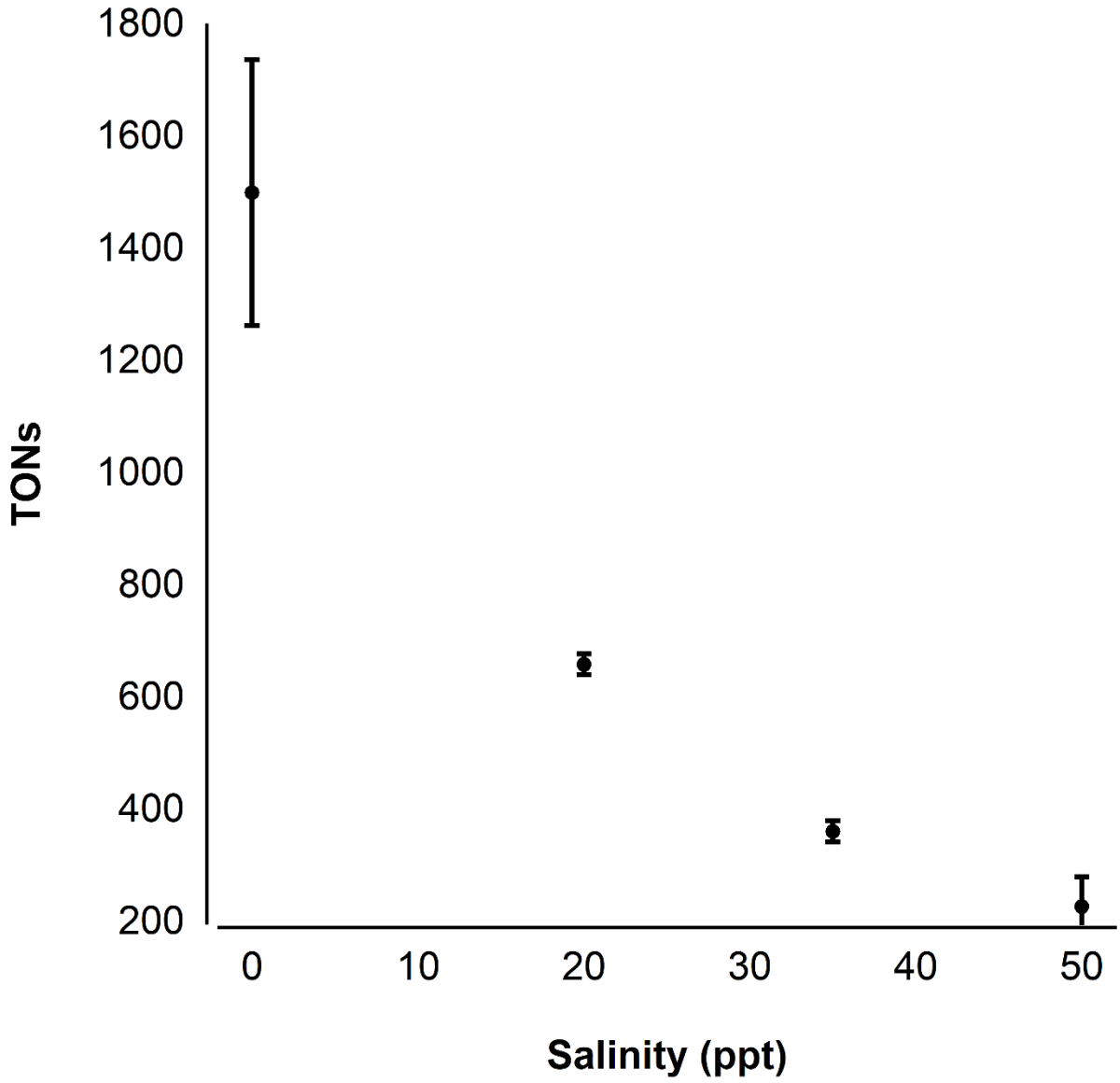


Figure S36. TONs with **2** and 5% TEA in 1:1 ethanol:deionized water after 24 hours with varying water salinity.

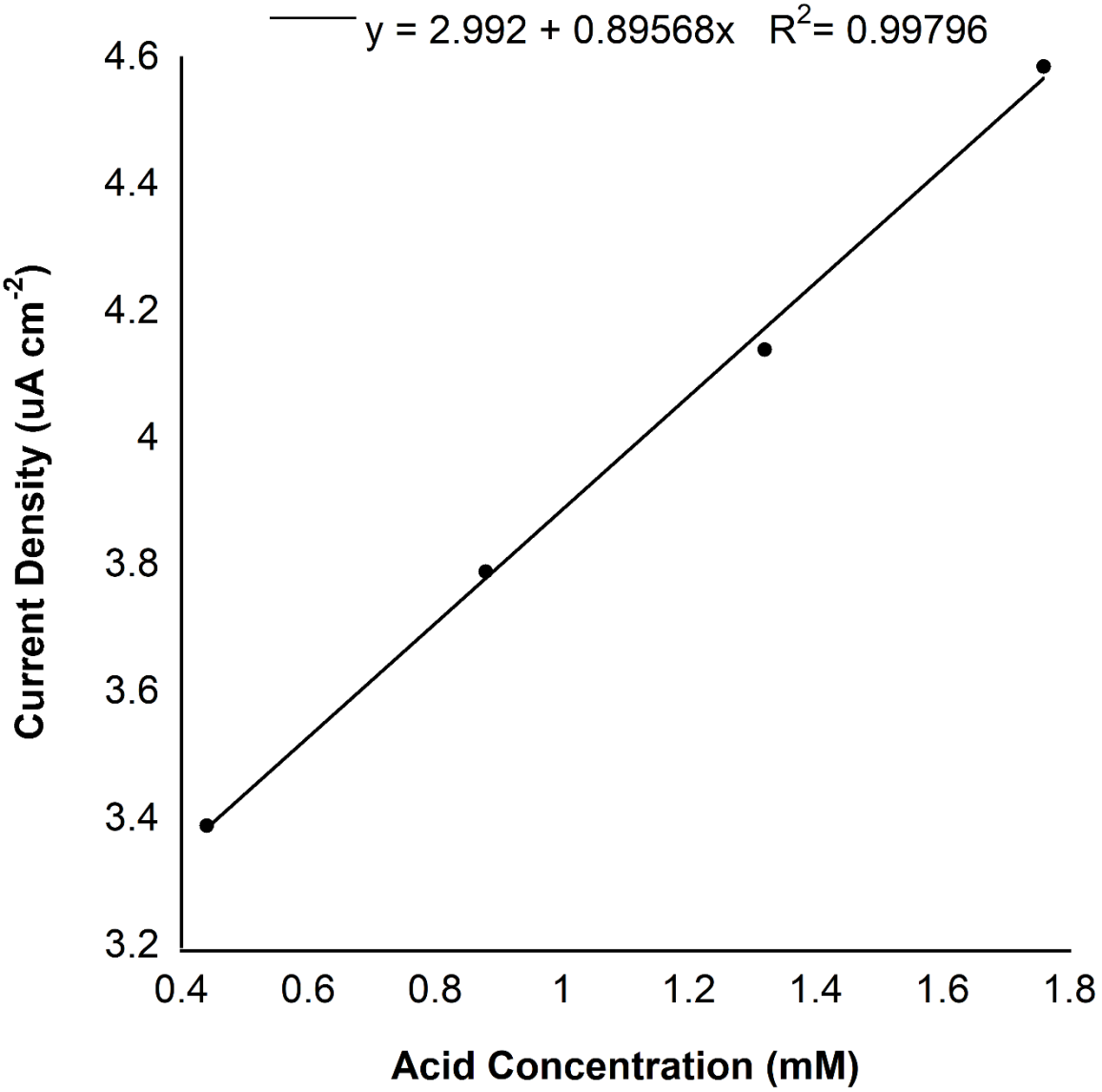


Figure S37. Linear relationship between TFA concentration and current density for **2**.

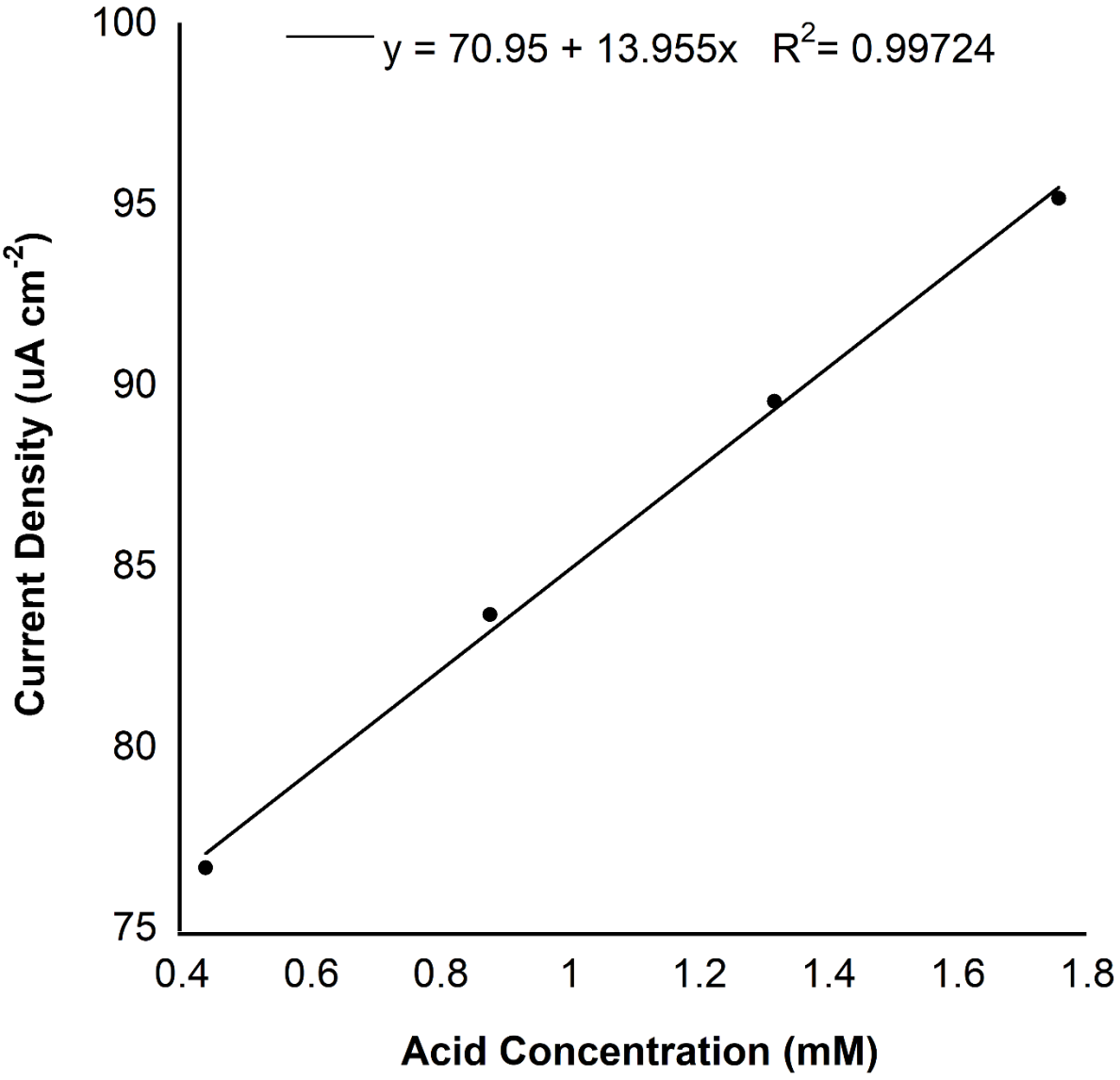


Figure S38. Linear relationship between TFA concentration and current density for **3**.

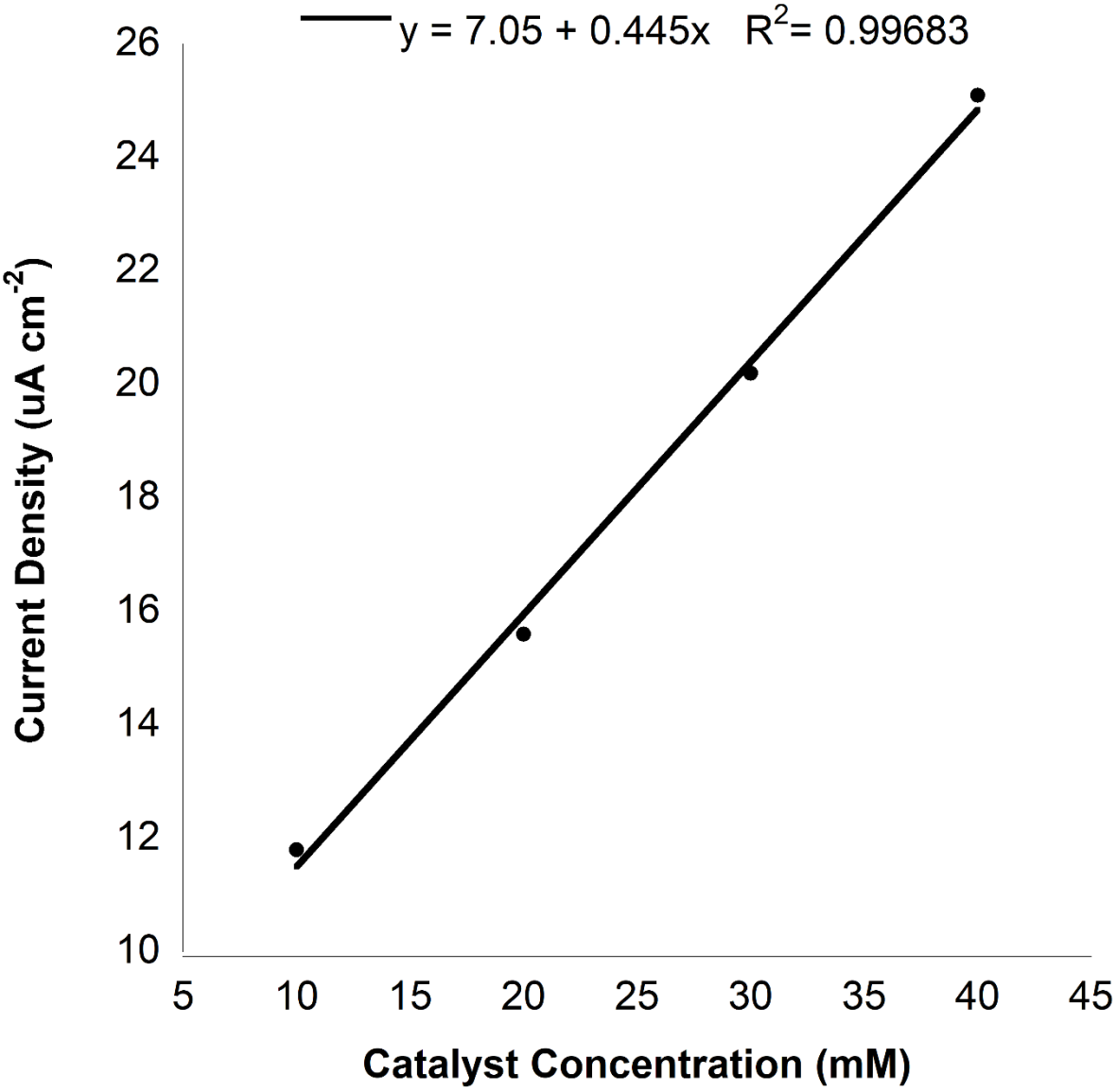


Figure S39. Linear relationship between [**2**] concentration and current density.

Table S1. TON with different [**2**] with 5% TEA in 1:1 ethanol:deionized water after 24 hours.

[Fluorescein] (mM)	[**2**] (M)	H_2 (μL)	TON
1.8	1.25×10^{-6}	56.5	470
1.8	2.50×10^{-6}	156	650
1.8	7.50×10^{-6}	960	1300
1.8	1.50×10^{-5}	243	170

Table S2. TON with different [fluorescein] with 5% TEA in 1:1 ethanol:deionized water after 24 hours.

[Fluorescein] (mM)	[**2**] (M)	H_2 (μL)	TON
1.8	7.50×10^{-6}	1470	2000
1.9	7.50×10^{-6}	953	1300
2.0	7.50×10^{-6}	873	1200

Table S3. Comparison of chromophores and sacrificial electron donors with **2** after 24 hours.

Chromophore/Electron donor	Fl/TEA pH =12.5	EY/TEA pH = 12.5	EY/TEOA pH = 12.5	EY/TEOA pH = 7
TON with 60 nM **2**	208	51	46	0

Table S4. TONs with different [**4**] with 5% TEA in 1:1 ethanol:deionized water after 24 hours.

[Fluorescein] (mM)	[**4**] (M)	H_2 (μL)	TONs
1.8	2.50×10^{-6}	Not Observed	0
1.8	7.50×10^{-6}	Not Observed	0
1.8	1.50×10^{-5}	Not Observed	0

Table S5. Comparison of chromophores and sacrificial electron donors with **4** after 24 hours.

Chromophore/Electron donor	Fl/TEA	Ru(bpy)/AA
TON with 7.5 μM **4**	Not Observed	Not Observed

Table S6. Crystal data and structure refinement for **2**.

Identification code	cwmwm18	
Empirical formula	C26 H28 Cl3 Fe N6 O2	
Formula weight	618.74	
Temperature	100.00(10) K	
Wavelength	1.54184 Å	
Crystal system	orthorhombic	
Space group	$P2_12_12_1$	
Unit cell dimensions	a = 18.0494(2) Å	α = 90°
	b = 26.5169(2) Å	β = 90°
	c = 28.1081(2) Å	γ = 90°
Volume	13452.9(2) Å^3	
Z	16	
Density (calculated)	1.222 Mg/m^3	
Absorption coefficient	6.032 mm^{-1}	
F(000)	5104	
Crystal color, morphology	red, plate	
Crystal size	0.241 x 0.209 x 0.081 mm^3	
Theta range for data collection	3.353 to 81.549°	
Index ranges	$-23 \le h \le 22$, $-26 \le k \le 33$, $-35 \le l \le 35$	
Reflections collected	136133	
Independent reflections	28760 [R(int) = 0.0465]	
Observed reflections	25201	
Completeness to theta = 67.684°	100.0%	
Absorption correction	Multi-scan	
Max. and min. transmission	1.00000 and 0.51928	
Refinement method	Full-matrix least-squares on F^2	

Data / restraints / parameters	28760 / 99 / 1452
Goodness-of-fit on F^2	1.044
Final *R* indices [*I*>2sigma(*I*)]	*R*1 = 0.0493, *wR*2 = 0.1240
R indices (all data)	*R*1 = 0.0577, *wR*2 = 0.1295
Absolute structure parameter	0.052(4)
Largest diff. peak and hole	0.745 and -0.614 e.Å^{-3}

Table S7. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (Å^{2}x 10^3) for **2**. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U_{eq}
Fe1	5098(1)	3692(1)	5390(1)	32(1)
Cl1	4021(1)	3794(1)	5857(1)	43(1)
Cl2	6150(1)	3516(1)	4937(1)	41(1)
O1	4415(2)	3472(1)	4862(1)	34(1)
O2	5266(2)	3045(1)	5760(1)	37(1)
N1	5074(2)	4439(1)	5111(2)	35(1)
N2	5736(2)	4108(1)	5890(2)	36(1)
N3	3049(3)	4071(2)	4216(2)	39(1)
N4	3386(2)	3644(1)	4408(2)	35(1)
N5	6341(3)	2676(2)	6707(2)	40(1)
N6	5848(3)	2581(2)	6338(2)	38(1)
C1	5660(3)	4756(2)	5301(2)	44(1)
C2	5712(3)	4657(2)	5823(2)	40(1)
C3	4598(3)	4629(2)	4825(2)	31(1)
C4	4026(3)	4322(2)	4622(2)	33(1)
C5	3986(3)	3794(2)	4657(2)	33(1)
C6	3438(3)	4478(2)	4344(2)	35(1)
C7	3239(3)	5002(2)	4195(2)	47(1)
C8	2370(3)	4038(2)	3931(2)	50(1)
C9	3086(3)	3146(2)	4362(2)	36(1)
C10	2455(3)	3017(2)	4604(2)	46(1)
C11	2199(4)	2526(2)	4575(2)	55(2)
C12	2579(4)	2173(2)	4314(3)	56(2)

C13	3213(4)	2307(2)	4066(3)	58(2)
C14	3464(3)	2799(2)	4089(2)	48(1)
C15	6122(3)	3924(2)	6228(2)	35(1)
C16	6151(3)	3394(2)	6324(2)	35(1)
C17	5713(3)	3020(2)	6107(2)	32(1)
C18	6535(3)	3157(2)	6698(2)	42(1)
C19	7093(4)	3367(2)	7025(3)	65(2)
C20	6604(4)	2268(2)	7009(2)	63(2)
C21	5413(3)	2126(2)	6331(2)	37(1)
C22	4841(4)	2082(2)	6652(3)	57(2)
C23	4423(4)	1638(3)	6654(3)	75(2)
C24	4591(4)	1261(2)	6335(3)	66(2)
C25	5152(4)	1318(2)	6020(3)	61(2)
C26	5574(3)	1755(2)	6009(2)	44(1)
Fe3	5189(1)	6280(1)	4446(1)	39(1)
Cl5	5887(1)	6166(1)	5147(1)	48(1)
Cl6	4551(1)	6384(1)	3734(1)	60(1)
O5	4268(2)	6111(1)	4807(2)	46(1)
O6	5488(2)	5593(1)	4223(1)	42(1)
N13	5033(3)	7058(1)	4605(2)	41(1)
N14	6139(2)	6615(2)	4135(2)	37(1)
N15	3087(2)	6699(1)	5584(2)	34(1)
N16	3328(2)	6284(2)	5329(2)	38(1)
N17	6650(3)	5156(2)	3343(2)	60(2)
N18	6089(3)	5090(2)	3685(2)	53(1)
C53	5502(3)	7391(2)	4311(2)	50(1)
C54	6255(3)	7140(2)	4265(2)	42(1)
C55	4574(3)	7248(2)	4898(2)	38(1)
C56	4033(3)	6946(2)	5148(2)	36(1)
C57	3926(3)	6422(2)	5069(2)	39(1)
C58	3501(3)	7101(2)	5468(2)	34(1)
C59	3354(3)	7610(2)	5668(2)	42(1)
C60	2458(3)	6656(2)	5907(2)	40(1)
C61	3079(3)	5777(2)	5405(2)	34(1)
C62	3558(3)	5446(2)	5626(2)	43(1)
C63	3347(4)	4950(2)	5675(2)	52(2)

C64	2669(4)	4790(2)	5516(2)	51(2)
C65	2184(3)	5129(2)	5309(2)	49(1)
C66	2393(3)	5634(2)	5250(2)	43(1)
C67	6584(3)	6400(2)	3843(2)	38(1)
C68	6504(3)	5889(2)	3714(2)	40(1)
C69	5982(3)	5540(2)	3902(2)	38(1)
C70	6908(3)	5629(2)	3364(2)	48(1)
C71	7539(4)	5809(2)	3069(3)	62(2)
C72	6762(17)	4779(8)	2958(10)	66(7)
C73	5558(4)	4691(2)	3633(2)	45(1)
C74	5709(4)	4231(2)	3842(2)	49(1)
C75	5213(5)	3837(2)	3761(2)	65(2)
C76	4588(5)	3917(3)	3481(2)	66(2)
C77	4433(5)	4390(3)	3300(2)	67(2)
C78	4936(4)	4778(3)	3371(2)	56(2)
Fe2	4862(1)	8769(1)	5514(1)	40(1)
Cl3	5858(1)	8555(1)	4991(1)	49(1)
Cl4	3897(1)	8923(1)	6034(1)	68(1)
O3	5043(2)	8116(1)	5856(1)	46(1)
O4	4154(2)	8534(1)	5009(1)	43(1)
N7	5660(3)	9153(2)	5930(2)	45(1)
N8	4818(3)	9512(2)	5239(2)	44(1)
N9	6152(3)	7725(2)	6779(2)	41(1)
N10	5621(3)	7641(2)	6425(2)	38(1)
N11	3441(3)	9014(2)	3966(2)	50(1)
N12	3545(3)	8630(2)	4296(2)	47(1)
C27	5817(4)	9672(2)	5789(3)	64(2)
C28	5133(5)	9888(2)	5563(3)	65(2)
C29	6056(3)	8960(2)	6262(2)	46(1)
C30	5996(3)	8442(2)	6399(2)	39(1)
C31	5506(3)	8082(2)	6191(2)	37(1)
C32	6385(4)	8192(2)	6754(2)	48(1)
C33	6968(4)	8398(2)	7074(2)	64(2)
C34	6400(5)	7300(3)	7068(3)	78(2)
C35	5144(3)	7206(2)	6438(2)	37(1)
C36	5353(3)	6783(2)	6195(2)	43(1)

C37	4895(3)	6359(2)	6214(2)	47(1)
C38	4259(3)	6364(2)	6483(2)	42(1)
C39	4038(4)	6800(2)	6716(2)	54(2)
C40	4483(3)	7223(2)	6690(2)	49(1)
C41	4543(3)	9655(2)	4848(2)	45(1)
C42	4152(3)	9316(2)	4547(2)	41(1)
C43	3977(3)	8809(2)	4655(2)	40(1)
C44	3814(3)	9424(2)	4114(2)	47(1)
C45	3859(4)	9890(2)	3812(3)	56(2)
C46	3034(5)	8932(3)	3526(3)	77(2)
C47	3150(3)	8162(2)	4280(2)	39(1)
C48	3476(4)	7751(2)	4070(3)	58(2)
C49	3103(4)	7290(2)	4078(3)	74(2)
C50	2415(4)	7260(3)	4277(3)	70(2)
C51	2084(4)	7681(3)	4470(3)	67(2)
C52	2459(3)	8134(3)	4470(2)	55(2)
Fe4	311(1)	3801(1)	5710(1)	36(1)
Cl7	-411(1)	3632(1)	6370(1)	80(1)
Cl8	1098(1)	3979(1)	5048(1)	64(1)
O7	572(2)	4473(1)	5998(2)	44(1)
O8	-555(2)	4002(1)	5314(2)	47(1)
N19	1233(3)	3445(2)	6025(2)	58(2)
N20	190(3)	3038(1)	5487(2)	39(1)
N21	2010(2)	4929(2)	6683(2)	36(1)
N22	1356(2)	4986(1)	6422(2)	34(1)
N23	-1714(2)	3451(2)	4493(2)	37(1)
N24	-1461(2)	3860(1)	4754(2)	37(1)
C00'	-2095(17)	4409(9)	5310(8)	37(4)
C01'	-2487(12)	4859(7)	5389(9)	48(5)
C02'	-2571(13)	5214(8)	5022(10)	56(5)
C03'	-2275(14)	5128(8)	4578(10)	55(5)
C04'	-1878(15)	4679(8)	4496(7)	37(4)
C99'	-1790(20)	4337(11)	4859(8)	30(4)
C79	1351(5)	2920(2)	5862(4)	85(3)
C80	662(4)	2689(2)	5755(3)	62(2)
C81	1729(3)	3658(2)	6279(2)	49(1)

C82	1705(3)	4175(2)	6391(2)	37(1)
C83	1152(3)	4531(2)	6249(2)	36(1)
C84	2222(3)	4450(2)	6654(2)	35(1)
C85	2918(3)	4280(2)	6875(2)	44(1)
C86	2413(3)	5357(2)	6864(2)	49(1)
C87	888(3)	5421(2)	6501(2)	38(1)
C88	562(4)	5471(2)	6952(2)	49(1)
C89	130(4)	5899(3)	7031(2)	63(2)
C90	51(4)	6264(2)	6681(3)	62(2)
C91	369(4)	6203(2)	6247(2)	54(2)
C92	796(3)	5774(2)	6153(2)	41(1)
C93	-261(3)	2867(2)	5181(2)	40(1)
C94	-785(3)	3187(2)	4945(2)	36(1)
C95	-886(3)	3706(2)	5034(2)	38(1)
C96	-1307(3)	3046(2)	4603(2)	36(1)
C97	-1438(3)	2547(2)	4376(2)	43(1)
C98	-2312(3)	3503(2)	4142(2)	46(1)
C99	-1858(13)	4338(7)	4735(5)	35(3)
C100	-2157(10)	4507(5)	5165(5)	40(3)
C101	-2473(8)	4994(5)	5176(6)	52(4)
C102	-2475(7)	5280(4)	4764(6)	55(4)
C103	-2163(11)	5108(6)	4348(7)	67(4)
C104	-1838(11)	4628(6)	4334(5)	46(3)
Cl10	7410(1)	7367(1)	3132(1)	51(1)
Cl12	2705(1)	2571(1)	6813(1)	57(1)
Cl11	2145(2)	5127(1)	3107(1)	84(1)
Cl9	7205(1)	4835(1)	6878(1)	51(1)
C72'	7057(12)	4688(5)	3177(10)	59(5)

Table S8. Bond lengths [Å] and angles [°] for **2**.

Fe(1)-Cl(1)	2.3606(15)	C(7)-H(7B)	0.9800
Fe(1)-Cl(2)	2.3339(15)	C(7)-H(7C)	0.9800
Fe(1)-O(1)	2.016(3)	C(8)-H(8A)	0.9800
Fe(1)-O(2)	2.030(3)	C(8)-H(8B)	0.9800
Fe(1)-N(1)	2.131(4)	C(8)-H(8C)	0.9800
Fe(1)-N(2)	2.126(4)	C(9)-C(10)	1.371(8)
O(1)-C(5)	1.286(6)	C(9)-C(14)	1.379(8)
O(2)-C(17)	1.269(6)	C(10)-H(10)	0.9500
N(1)-C(1)	1.452(6)	C(10)-C(11)	1.384(7)
N(1)-C(3)	1.279(6)	C(11)-H(11)	0.9500
N(2)-C(2)	1.467(6)	C(11)-C(12)	1.372(10)
N(2)-C(15)	1.275(6)	C(12)-H(12)	0.9500
N(3)-N(4)	1.395(5)	C(12)-C(13)	1.386(10)
N(3)-C(6)	1.336(6)	C(13)-H(13)	0.9500
N(3)-C(8)	1.467(7)	C(13)-C(14)	1.382(8)
N(4)-C(5)	1.350(6)	C(14)-H(14)	0.9500
N(4)-C(9)	1.434(6)	C(15)-H(15)	0.9500
N(5)-N(6)	1.390(6)	C(15)-C(16)	1.432(6)
N(5)-C(18)	1.324(7)	C(16)-C(17)	1.408(7)
N(5)-C(20)	1.456(7)	C(16)-C(18)	1.410(7)
N(6)-C(17)	1.353(6)	C(18)-C(19)	1.472(8)
N(6)-C(21)	1.439(6)	C(19)-H(19A)	0.9800
C(1)-H(1A)	0.9900	C(19)-H(19B)	0.9800
C(1)-H(1B)	0.9900	C(19)-H(19C)	0.9800
C(1)-C(2)	1.495(8)	C(20)-H(20A)	0.9800
C(2)-H(2A)	0.9900	C(20)-H(20B)	0.9800
C(2)-H(2B)	0.9900	C(20)-H(20C)	0.9800
C(3)-H(3)	0.9500	C(21)-C(22)	1.376(9)
C(3)-C(4)	1.433(7)	C(21)-C(26)	1.369(7)
C(4)-C(5)	1.407(6)	C(22)-H(22)	0.9500
C(4)-C(6)	1.381(7)	C(22)-C(23)	1.398(9)
C(6)-C(7)	1.495(7)	C(23)-H(23)	0.9500
C(7)-H(7A)	0.9800	C(23)-C(24)	1.376(11)

C(24)-H(24)	0.9500	C(56)-C(58)	1.377(7)
C(24)-C(25)	1.352(11)	C(58)-C(59)	1.487(7)
C(25)-H(25)	0.9500	C(59)-H(59A)	0.9800
C(25)-C(26)	1.389(8)	C(59)-H(59B)	0.9800
C(26)-H(26)	0.9500	C(59)-H(59C)	0.9800
Fe(3)-Cl(5)	2.3574(18)	C(60)-H(60A)	0.9800
Fe(3)-Cl(6)	2.3263(19)	C(60)-H(60B)	0.9800
Fe(3)-O(5)	1.998(4)	C(60)-H(60C)	0.9800
Fe(3)-O(6)	2.001(3)	C(61)-C(62)	1.380(7)
Fe(3)-N(13)	2.130(4)	C(61)-C(66)	1.367(7)
Fe(3)-N(14)	2.120(4)	C(62)-H(62)	0.9500
O(5)-C(57)	1.268(6)	C(62)-C(63)	1.376(8)
O(6)-C(69)	1.276(6)	C(63)-H(63)	0.9500
N(13)-C(53)	1.475(7)	C(63)-C(64)	1.370(9)
N(13)-C(55)	1.272(7)	C(64)-H(64)	0.9500
N(14)-C(54)	1.453(6)	C(64)-C(65)	1.383(9)
N(14)-C(67)	1.283(7)	C(65)-H(65)	0.9500
N(15)-N(16)	1.384(5)	C(65)-C(66)	1.399(7)
N(15)-C(58)	1.342(6)	C(66)-H(66)	0.9500
N(15)-C(60)	1.459(6)	C(67)-H(67)	0.9500
N(16)-C(57)	1.353(6)	C(67)-C(68)	1.412(7)
N(16)-C(61)	1.433(6)	C(68)-C(69)	1.421(7)
N(17)-N(18)	1.407(7)	C(68)-C(70)	1.407(8)
N(17)-C(70)	1.340(7)	C(70)-C(71)	1.488(8)
N(17)-C(72)	1.486(14)	C(71)-H(71A)	0.9800
N(17)-C(72')	1.514(14)	C(71)-H(71B)	0.9800
N(18)-C(69)	1.353(6)	C(71)-H(71C)	0.9800
N(18)-C(73)	1.437(7)	C(72)-H(72A)	0.9800
C(53)-H(53A)	0.9900	C(72)-H(72B)	0.9800
C(53)-H(53B)	0.9900	C(72)-H(72C)	0.9800
C(53)-C(54)	1.519(8)	C(73)-C(74)	1.380(8)
C(54)-H(54A)	0.9900	C(73)-C(78)	1.364(9)
C(54)-H(54B)	0.9900	C(74)-H(74)	0.9500
C(55)-H(55)	0.9500	C(74)-C(75)	1.396(9)
C(55)-C(56)	1.445(7)	C(75)-H(75)	0.9500
C(56)-C(57)	1.420(6)	C(75)-C(76)	1.392(11)

C(76)-H(76)	0.9500	C(32)-C(33)	1.488(8)
C(76)-C(77)	1.381(11)	C(33)-H(33A)	0.9800
C(77)-H(77)	0.9500	C(33)-H(33B)	0.9800
C(77)-C(78)	1.386(9)	C(33)-H(33C)	0.9800
C(78)-H(78)	0.9500	C(34)-H(34A)	0.9800
Fe(2)-Cl(3)	2.3915(17)	C(34)-H(34B)	0.9800
Fe(2)-Cl(4)	2.310(2)	C(34)-H(34C)	0.9800
Fe(2)-O(3)	2.008(4)	C(35)-C(36)	1.366(7)
Fe(2)-O(4)	2.010(4)	C(35)-C(40)	1.388(8)
Fe(2)-N(7)	2.117(5)	C(36)-H(36)	0.9500
Fe(2)-N(8)	2.117(4)	C(36)-C(37)	1.396(7)
O(3)-C(31)	1.262(6)	C(37)-H(37)	0.9500
O(4)-C(43)	1.273(6)	C(37)-C(38)	1.376(8)
N(7)-C(27)	1.461(7)	C(38)-H(38)	0.9500
N(7)-C(29)	1.280(7)	C(38)-C(39)	1.388(8)
N(8)-C(28)	1.464(7)	C(39)-H(39)	0.9500
N(8)-C(41)	1.264(7)	C(39)-C(40)	1.382(8)
N(9)-N(10)	1.399(6)	C(40)-H(40)	0.9500
N(9)-C(32)	1.309(7)	C(41)-H(41)	0.9500
N(9)-C(34)	1.459(8)	C(41)-C(42)	1.421(8)
N(10)-C(31)	1.357(6)	C(42)-C(43)	1.414(7)
N(10)-C(35)	1.441(6)	C(42)-C(44)	1.393(8)
N(11)-N(12)	1.389(6)	C(44)-C(45)	1.500(8)
N(11)-C(44)	1.346(7)	C(45)-H(45A)	0.9800
N(11)-C(46)	1.453(8)	C(45)-H(45B)	0.9800
N(12)-C(43)	1.362(7)	C(45)-H(45C)	0.9800
N(12)-C(47)	1.434(6)	C(46)-H(46A)	0.9800
C(27)-H(27A)	0.9900	C(46)-H(46B)	0.9800
C(27)-H(27B)	0.9900	C(46)-H(46C)	0.9800
C(27)-C(28)	1.502(10)	C(47)-C(48)	1.372(8)
C(28)-H(28A)	0.9900	C(47)-C(52)	1.358(8)
C(28)-H(28B)	0.9900	C(48)-H(48)	0.9500
C(29)-H(29)	0.9500	C(48)-C(49)	1.395(9)
C(29)-C(30)	1.430(7)	C(49)-H(49)	0.9500
C(30)-C(31)	1.426(7)	C(49)-C(50)	1.365(12)
C(30)-C(32)	1.391(8)	C(50)-H(50)	0.9500

C(50)-C(51)	1.380(11)	C(04')-H(04')	0.9500
C(51)-H(51)	0.9500	C(04')-C(99')	1.374(17)
C(51)-C(52)	1.378(9)	C(79)-H(79A)	0.9900
C(52)-H(52)	0.9500	C(79)-H(79B)	0.9900
Fe(4)-Cl(7)	2.312(2)	C(79)-C(80)	1.418(10)
Fe(4)-Cl(8)	2.3887(18)	C(80)-H(80A)	0.9900
Fe(4)-O(7)	2.014(3)	C(80)-H(80B)	0.9900
Fe(4)-O(8)	1.992(4)	C(81)-H(81)	0.9500
Fe(4)-N(19)	2.109(5)	C(81)-C(82)	1.406(7)
Fe(4)-N(20)	2.129(4)	C(82)-C(83)	1.430(7)
O(7)-C(83)	1.273(6)	C(82)-C(84)	1.397(7)
O(8)-C(95)	1.262(6)	C(84)-C(85)	1.472(8)
N(19)-C(79)	1.482(7)	C(85)-H(85A)	0.9800
N(19)-C(81)	1.276(8)	C(85)-H(85B)	0.9800
N(20)-C(80)	1.468(7)	C(85)-H(85C)	0.9800
N(20)-C(93)	1.269(7)	C(86)-H(86A)	0.9800
N(21)-N(22)	1.398(6)	C(86)-H(86B)	0.9800
N(21)-C(84)	1.329(6)	C(86)-H(86C)	0.9800
N(21)-C(86)	1.442(6)	C(87)-C(88)	1.404(8)
N(22)-C(83)	1.353(6)	C(87)-C(92)	1.364(7)
N(22)-C(87)	1.447(6)	C(88)-H(88)	0.9500
N(23)-N(24)	1.386(6)	C(88)-C(89)	1.395(8)
N(23)-C(96)	1.338(6)	C(89)-H(89)	0.9500
N(23)-C(98)	1.469(7)	C(89)-C(90)	1.387(10)
N(24)-C(99')	1.43(2)	C(90)-H(90)	0.9500
N(24)-C(95)	1.364(6)	C(90)-C(91)	1.359(10)
N(24)-C(99)	1.458(16)	C(91)-H(91)	0.9500
C(00')-H(00')	0.9500	C(91)-C(92)	1.398(8)
C(00')-C(01')	1.404(18)	C(92)-H(92)	0.9500
C(00')-C(99')	1.393(17)	C(93)-H(93)	0.9500
C(01')-H(01')	0.9500	C(93)-C(94)	1.433(7)
C(01')-C(02')	1.403(18)	C(94)-C(95)	1.410(6)
C(02')-H(02')	0.9500	C(94)-C(96)	1.399(7)
C(02')-C(03')	1.38(2)	C(96)-C(97)	1.486(7)
C(03')-H(03')	0.9500	C(97)-H(97A)	0.9800
C(03')-C(04')	1.409(18)	C(97)-H(97B)	0.9800

C(97)-H(97C)	0.9800	C(3)-N(1)-Fe(1)	127.6(3)
C(98)-H(98A)	0.9800	C(3)-N(1)-C(1)	119.4(4)
C(98)-H(98B)	0.9800	C(2)-N(2)-Fe(1)	114.5(3)
C(98)-H(98C)	0.9800	C(15)-N(2)-Fe(1)	126.2(3)
C(99)-C(100)	1.398(13)	C(15)-N(2)-C(2)	119.4(4)
C(99)-C(104)	1.364(14)	N(4)-N(3)-C(8)	121.8(4)
C(100)-H(100)	0.9500	C(6)-N(3)-N(4)	108.7(4)
C(100)-C(101)	1.412(14)	C(6)-N(3)-C(8)	129.5(4)
C(101)-H(101)	0.9500	N(3)-N(4)-C(9)	123.3(4)
C(101)-C(102)	1.383(15)	C(5)-N(4)-N(3)	108.2(4)
C(102)-H(102)	0.9500	C(5)-N(4)-C(9)	128.4(4)
C(102)-C(103)	1.375(16)	N(6)-N(5)-C(20)	120.7(4)
C(103)-H(103)	0.9500	C(18)-N(5)-N(6)	109.3(4)
C(103)-C(104)	1.402(14)	C(18)-N(5)-C(20)	129.9(5)
C(104)-H(104)	0.9500	N(5)-N(6)-C(21)	120.7(4)
C(72')-H(72D)	0.9800	C(17)-N(6)-N(5)	108.5(4)
C(72')-H(72E)	0.9800	C(17)-N(6)-C(21)	128.0(4)
C(72')-H(72F)	0.9800	N(1)-C(1)-H(1A)	110.2
Cl(2)-Fe(1)-Cl(1)	175.04(5)	N(1)-C(1)-H(1B)	110.2
O(1)-Fe(1)-Cl(1)	86.56(11)	N(1)-C(1)-C(2)	107.7(4)
O(1)-Fe(1)-Cl(2)	92.15(11)	H(1A)-C(1)-H(1B)	108.5
O(1)-Fe(1)-O(2)	102.95(13)	C(2)-C(1)-H(1A)	110.2
O(1)-Fe(1)-N(1)	89.16(14)	C(2)-C(1)-H(1B)	110.2
O(1)-Fe(1)-N(2)	165.54(14)	N(2)-C(2)-C(1)	107.6(4)
O(2)-Fe(1)-Cl(1)	86.28(11)	N(2)-C(2)-H(2A)	110.2
O(2)-Fe(1)-Cl(2)	89.34(11)	N(2)-C(2)-H(2B)	110.2
O(2)-Fe(1)-N(1)	167.88(15)	C(1)-C(2)-H(2A)	110.2
O(2)-Fe(1)-N(2)	91.11(15)	C(1)-C(2)-H(2B)	110.2
N(1)-Fe(1)-Cl(1)	94.66(12)	H(2A)-C(2)-H(2B)	108.5
N(1)-Fe(1)-Cl(2)	90.11(12)	N(1)-C(3)-H(3)	119.6
N(2)-Fe(1)-Cl(1)	91.08(13)	N(1)-C(3)-C(4)	120.7(4)
N(2)-Fe(1)-Cl(2)	91.35(13)	C(4)-C(3)-H(3)	119.6
N(2)-Fe(1)-N(1)	76.80(16)	C(5)-C(4)-C(3)	125.0(5)
C(5)-O(1)-Fe(1)	120.3(3)	C(6)-C(4)-C(3)	127.6(4)
C(17)-O(2)-Fe(1)	122.2(3)	C(6)-C(4)-C(5)	107.4(4)
C(1)-N(1)-Fe(1)	112.9(3)	O(1)-C(5)-N(4)	121.3(4)

O(1)-C(5)-C(4)	131.4(5)	N(2)-C(15)-C(16)	122.4(4)
N(4)-C(5)-C(4)	107.3(4)	C(16)-C(15)-H(15)	118.8
N(3)-C(6)-C(4)	108.4(4)	C(17)-C(16)-C(15)	126.2(5)
N(3)-C(6)-C(7)	123.2(5)	C(17)-C(16)-C(18)	106.5(4)
C(4)-C(6)-C(7)	128.3(5)	C(18)-C(16)-C(15)	126.6(5)
C(6)-C(7)-H(7A)	109.5	O(2)-C(17)-N(6)	121.9(4)
C(6)-C(7)-H(7B)	109.5	O(2)-C(17)-C(16)	130.7(4)
C(6)-C(7)-H(7C)	109.5	N(6)-C(17)-C(16)	107.4(4)
H(7A)-C(7)-H(7B)	109.5	N(5)-C(18)-C(16)	108.3(4)
H(7A)-C(7)-H(7C)	109.5	N(5)-C(18)-C(19)	122.3(5)
H(7B)-C(7)-H(7C)	109.5	C(16)-C(18)-C(19)	129.3(5)
N(3)-C(8)-H(8A)	109.5	C(18)-C(19)-H(19A)	109.5
N(3)-C(8)-H(8B)	109.5	C(18)-C(19)-H(19B)	109.5
N(3)-C(8)-H(8C)	109.5	C(18)-C(19)-H(19C)	109.5
H(8A)-C(8)-H(8B)	109.5	H(19A)-C(19)-H(19B)	109.5
H(8A)-C(8)-H(8C)	109.5	H(19A)-C(19)-H(19C)	109.5
H(8B)-C(8)-H(8C)	109.5	H(19B)-C(19)-H(19C)	109.5
C(10)-C(9)-N(4)	119.8(5)	N(5)-C(20)-H(20A)	109.5
C(10)-C(9)-C(14)	121.5(5)	N(5)-C(20)-H(20B)	109.5
C(14)-C(9)-N(4)	118.6(5)	N(5)-C(20)-H(20C)	109.5
C(9)-C(10)-H(10)	120.6	H(20A)-C(20)-H(20B)	109.5
C(9)-C(10)-C(11)	118.8(6)	H(20A)-C(20)-H(20C)	109.5
C(11)-C(10)-H(10)	120.6	H(20B)-C(20)-H(20C)	109.5
C(10)-C(11)-H(11)	119.8	C(22)-C(21)-N(6)	118.2(5)
C(12)-C(11)-C(10)	120.4(6)	C(26)-C(21)-N(6)	119.7(5)
C(12)-C(11)-H(11)	119.8	C(26)-C(21)-C(22)	122.1(5)
C(11)-C(12)-H(12)	119.8	C(21)-C(22)-H(22)	120.7
C(11)-C(12)-C(13)	120.4(5)	C(21)-C(22)-C(23)	118.7(6)
C(13)-C(12)-H(12)	119.8	C(23)-C(22)-H(22)	120.7
C(12)-C(13)-H(13)	120.4	C(22)-C(23)-H(23)	120.3
C(12)-C(13)-C(14)	119.3(6)	C(24)-C(23)-C(22)	119.3(7)
C(14)-C(13)-H(13)	120.4	C(24)-C(23)-H(23)	120.3
C(9)-C(14)-C(13)	119.5(6)	C(23)-C(24)-H(24)	119.6
C(9)-C(14)-H(14)	120.2	C(25)-C(24)-C(23)	120.7(6)
C(13)-C(14)-H(14)	120.2	C(25)-C(24)-H(24)	119.6
N(2)-C(15)-H(15)	118.8	C(24)-C(25)-H(25)	119.4

C(24)-C(25)-C(26)	121.2(6)	C(70)-N(17)-N(18)	109.6(5)
C(26)-C(25)-H(25)	119.4	C(70)-N(17)-C(72)	127.9(9)
C(21)-C(26)-C(25)	118.0(6)	C(70)-N(17)-C(72')	127.6(8)
C(21)-C(26)-H(26)	121.0	N(17)-N(18)-C(73)	120.1(5)
C(25)-C(26)-H(26)	121.0	C(69)-N(18)-N(17)	107.6(4)
Cl(6)-Fe(3)-Cl(5)	177.28(7)	C(69)-N(18)-C(73)	126.9(5)
O(5)-Fe(3)-Cl(5)	89.52(14)	N(13)-C(53)-H(53A)	110.2
O(5)-Fe(3)-Cl(6)	92.95(14)	N(13)-C(53)-H(53B)	110.2
O(5)-Fe(3)-O(6)	100.30(15)	N(13)-C(53)-C(54)	107.3(4)
O(5)-Fe(3)-N(13)	90.08(16)	H(53A)-C(53)-H(53B)	108.5
O(5)-Fe(3)-N(14)	167.49(15)	C(54)-C(53)-H(53A)	110.2
O(6)-Fe(3)-Cl(5)	90.10(12)	C(54)-C(53)-H(53B)	110.2
O(6)-Fe(3)-Cl(6)	88.35(13)	N(14)-C(54)-C(53)	108.3(4)
O(6)-Fe(3)-N(13)	169.56(16)	N(14)-C(54)-H(54A)	110.0
O(6)-Fe(3)-N(14)	91.97(15)	N(14)-C(54)-H(54B)	110.0
N(13)-Fe(3)-Cl(5)	91.10(14)	C(53)-C(54)-H(54A)	110.0
N(13)-Fe(3)-Cl(6)	90.03(14)	C(53)-C(54)-H(54B)	110.0
N(14)-Fe(3)-Cl(5)	88.04(13)	H(54A)-C(54)-H(54B)	108.4
N(14)-Fe(3)-Cl(6)	89.78(14)	N(13)-C(55)-H(55)	118.8
N(14)-Fe(3)-N(13)	77.70(16)	N(13)-C(55)-C(56)	122.4(4)
C(57)-O(5)-Fe(3)	123.7(3)	C(56)-C(55)-H(55)	118.8
C(69)-O(6)-Fe(3)	120.7(3)	C(57)-C(56)-C(55)	123.9(5)
C(53)-N(13)-Fe(3)	112.7(3)	C(58)-C(56)-C(55)	128.5(4)
C(55)-N(13)-Fe(3)	127.2(3)	C(58)-C(56)-C(57)	107.4(4)
C(55)-N(13)-C(53)	120.0(4)	O(5)-C(57)-N(16)	121.8(4)
C(54)-N(14)-Fe(3)	114.4(3)	O(5)-C(57)-C(56)	131.4(5)
C(67)-N(14)-Fe(3)	125.8(3)	N(16)-C(57)-C(56)	106.8(4)
C(67)-N(14)-C(54)	119.7(4)	N(15)-C(58)-C(56)	108.1(4)
N(16)-N(15)-C(60)	120.3(4)	N(15)-C(58)-C(59)	122.0(5)
C(58)-N(15)-N(16)	109.3(4)	C(56)-C(58)-C(59)	129.9(5)
C(58)-N(15)-C(60)	130.4(4)	C(58)-C(59)-H(59A)	109.5
N(15)-N(16)-C(61)	124.8(4)	C(58)-C(59)-H(59B)	109.5
C(57)-N(16)-N(15)	108.4(4)	C(58)-C(59)-H(59C)	109.5
C(57)-N(16)-C(61)	125.7(4)	H(59A)-C(59)-H(59B)	109.5
N(18)-N(17)-C(72)	120.9(8)	H(59A)-C(59)-H(59C)	109.5
N(18)-N(17)-C(72')	117.3(8)	H(59B)-C(59)-H(59C)	109.5

N(15)-C(60)-H(60A)	109.5	C(70)-C(71)-H(71A)	109.5
N(15)-C(60)-H(60B)	109.5	C(70)-C(71)-H(71B)	109.5
N(15)-C(60)-H(60C)	109.5	C(70)-C(71)-H(71C)	109.5
H(60A)-C(60)-H(60B)	109.5	H(71A)-C(71)-H(71B)	109.5
H(60A)-C(60)-H(60C)	109.5	H(71A)-C(71)-H(71C)	109.5
H(60B)-C(60)-H(60C)	109.5	H(71B)-C(71)-H(71C)	109.5
C(62)-C(61)-N(16)	117.8(5)	N(17)-C(72)-H(72A)	109.5
C(66)-C(61)-N(16)	119.9(5)	N(17)-C(72)-H(72B)	109.5
C(66)-C(61)-C(62)	122.3(5)	N(17)-C(72)-H(72C)	109.5
C(61)-C(62)-H(62)	120.7	H(72A)-C(72)-H(72B)	109.5
C(63)-C(62)-C(61)	118.6(6)	H(72A)-C(72)-H(72C)	109.5
C(63)-C(62)-H(62)	120.7	H(72B)-C(72)-H(72C)	109.5
C(62)-C(63)-H(63)	119.6	C(74)-C(73)-N(18)	118.4(6)
C(64)-C(63)-C(62)	120.7(6)	C(78)-C(73)-N(18)	118.6(5)
C(64)-C(63)-H(63)	119.6	C(78)-C(73)-C(74)	122.9(6)
C(63)-C(64)-H(64)	120.0	C(73)-C(74)-H(74)	121.1
C(63)-C(64)-C(65)	120.1(5)	C(73)-C(74)-C(75)	117.8(7)
C(65)-C(64)-H(64)	120.0	C(75)-C(74)-H(74)	121.1
C(64)-C(65)-H(65)	120.0	C(74)-C(75)-H(75)	120.1
C(64)-C(65)-C(66)	120.1(6)	C(76)-C(75)-C(74)	119.8(6)
C(66)-C(65)-H(65)	120.0	C(76)-C(75)-H(75)	120.1
C(61)-C(66)-C(65)	118.2(5)	C(75)-C(76)-H(76)	119.6
C(61)-C(66)-H(66)	120.9	C(77)-C(76)-C(75)	120.8(6)
C(65)-C(66)-H(66)	120.9	C(77)-C(76)-H(76)	119.6
N(14)-C(67)-H(67)	119.1	C(76)-C(77)-H(77)	120.4
N(14)-C(67)-C(68)	121.7(5)	C(76)-C(77)-C(78)	119.3(7)
C(68)-C(67)-H(67)	119.1	C(78)-C(77)-H(77)	120.4
C(67)-C(68)-C(69)	126.8(5)	C(73)-C(78)-C(77)	119.3(6)
C(70)-C(68)-C(67)	126.6(5)	C(73)-C(78)-H(78)	120.3
C(70)-C(68)-C(69)	106.6(4)	C(77)-C(78)-H(78)	120.3
O(6)-C(69)-N(18)	121.0(5)	Cl(4)-Fe(2)-Cl(3)	176.34(7)
O(6)-C(69)-C(68)	130.9(5)	O(3)-Fe(2)-Cl(3)	88.05(13)
N(18)-C(69)-C(68)	108.2(5)	O(3)-Fe(2)-Cl(4)	88.43(14)
N(17)-C(70)-C(68)	108.0(5)	O(3)-Fe(2)-O(4)	100.01(15)
N(17)-C(70)-C(71)	122.9(5)	O(3)-Fe(2)-N(7)	92.21(17)
C(68)-C(70)-C(71)	129.1(5)	O(3)-Fe(2)-N(8)	169.50(17)

O(4)-Fe(2)-Cl(3)	88.31(12)	N(8)-C(28)-H(28A)	109.9
O(4)-Fe(2)-Cl(4)	91.31(13)	N(8)-C(28)-H(28B)	109.9
O(4)-Fe(2)-N(7)	166.62(17)	C(27)-C(28)-H(28A)	109.9
O(4)-Fe(2)-N(8)	90.45(16)	C(27)-C(28)-H(28B)	109.9
N(7)-Fe(2)-Cl(3)	86.68(15)	H(28A)-C(28)-H(28B)	108.3
N(7)-Fe(2)-Cl(4)	94.47(15)	N(7)-C(29)-H(29)	118.7
N(8)-Fe(2)-Cl(3)	91.45(15)	N(7)-C(29)-C(30)	122.5(5)
N(8)-Fe(2)-Cl(4)	92.19(15)	C(30)-C(29)-H(29)	118.7
N(8)-Fe(2)-N(7)	77.29(18)	C(31)-C(30)-C(29)	125.4(5)
C(31)-O(3)-Fe(2)	121.8(3)	C(32)-C(30)-C(29)	127.9(5)
C(43)-O(4)-Fe(2)	122.1(3)	C(32)-C(30)-C(31)	106.7(4)
C(27)-N(7)-Fe(2)	115.8(4)	O(3)-C(31)-N(10)	121.7(4)
C(29)-N(7)-Fe(2)	126.1(4)	O(3)-C(31)-C(30)	131.9(5)
C(29)-N(7)-C(27)	117.8(5)	N(10)-C(31)-C(30)	106.4(4)
C(28)-N(8)-Fe(2)	113.1(4)	N(9)-C(32)-C(30)	109.1(5)
C(41)-N(8)-Fe(2)	127.6(4)	N(9)-C(32)-C(33)	122.9(6)
C(41)-N(8)-C(28)	119.3(4)	C(30)-C(32)-C(33)	128.0(5)
N(10)-N(9)-C(34)	119.0(4)	C(32)-C(33)-H(33A)	109.5
C(32)-N(9)-N(10)	109.4(4)	C(32)-C(33)-H(33B)	109.5
C(32)-N(9)-C(34)	131.3(5)	C(32)-C(33)-H(33C)	109.5
N(9)-N(10)-C(35)	121.2(4)	H(33A)-C(33)-H(33B)	109.5
C(31)-N(10)-N(9)	108.3(4)	H(33A)-C(33)-H(33C)	109.5
C(31)-N(10)-C(35)	127.7(5)	H(33B)-C(33)-H(33C)	109.5
N(12)-N(11)-C(46)	121.8(5)	N(9)-C(34)-H(34A)	109.5
C(44)-N(11)-N(12)	108.5(5)	N(9)-C(34)-H(34B)	109.5
C(44)-N(11)-C(46)	129.6(5)	N(9)-C(34)-H(34C)	109.5
N(11)-N(12)-C(47)	123.2(4)	H(34A)-C(34)-H(34B)	109.5
C(43)-N(12)-N(11)	108.6(4)	H(34A)-C(34)-H(34C)	109.5
C(43)-N(12)-C(47)	127.5(5)	H(34B)-C(34)-H(34C)	109.5
N(7)-C(27)-H(27A)	110.0	C(36)-C(35)-N(10)	118.7(5)
N(7)-C(27)-H(27B)	110.0	C(36)-C(35)-C(40)	121.3(5)
N(7)-C(27)-C(28)	108.3(5)	C(40)-C(35)-N(10)	120.0(5)
H(27A)-C(27)-H(27B)	108.4	C(35)-C(36)-H(36)	120.7
C(28)-C(27)-H(27A)	110.0	C(35)-C(36)-C(37)	118.7(5)
C(28)-C(27)-H(27B)	110.0	C(37)-C(36)-H(36)	120.7
N(8)-C(28)-C(27)	108.9(5)	C(36)-C(37)-H(37)	119.8

C(38)-C(37)-C(36)	120.5(5)	C(52)-C(47)-N(12)	119.5(5)
C(38)-C(37)-H(37)	119.8	C(52)-C(47)-C(48)	121.4(5)
C(37)-C(38)-H(38)	119.8	C(47)-C(48)-H(48)	120.6
C(37)-C(38)-C(39)	120.5(5)	C(47)-C(48)-C(49)	118.8(7)
C(39)-C(38)-H(38)	119.8	C(49)-C(48)-H(48)	120.6
C(38)-C(39)-H(39)	120.5	C(48)-C(49)-H(49)	120.1
C(40)-C(39)-C(38)	119.0(6)	C(50)-C(49)-C(48)	119.8(7)
C(40)-C(39)-H(39)	120.5	C(50)-C(49)-H(49)	120.1
C(35)-C(40)-H(40)	120.0	C(49)-C(50)-H(50)	119.7
C(39)-C(40)-C(35)	119.9(5)	C(49)-C(50)-C(51)	120.6(6)
C(39)-C(40)-H(40)	120.0	C(51)-C(50)-H(50)	119.7
N(8)-C(41)-H(41)	119.2	C(50)-C(51)-H(51)	120.3
N(8)-C(41)-C(42)	121.5(5)	C(52)-C(51)-C(50)	119.5(7)
C(42)-C(41)-H(41)	119.2	C(52)-C(51)-H(51)	120.3
C(43)-C(42)-C(41)	125.7(5)	C(47)-C(52)-C(51)	119.9(6)
C(44)-C(42)-C(41)	127.4(5)	C(47)-C(52)-H(52)	120.0
C(44)-C(42)-C(43)	106.7(5)	C(51)-C(52)-H(52)	120.0
O(4)-C(43)-N(12)	121.6(5)	Cl(7)-Fe(4)-Cl(8)	177.72(9)
O(4)-C(43)-C(42)	131.1(5)	O(7)-Fe(4)-Cl(7)	88.87(13)
N(12)-C(43)-C(42)	107.4(5)	O(7)-Fe(4)-Cl(8)	89.97(13)
N(11)-C(44)-C(42)	108.8(5)	O(7)-Fe(4)-N(19)	92.42(17)
N(11)-C(44)-C(45)	121.1(6)	O(7)-Fe(4)-N(20)	169.51(16)
C(42)-C(44)-C(45)	129.9(5)	O(8)-Fe(4)-Cl(7)	93.34(15)
C(44)-C(45)-H(45A)	109.5	O(8)-Fe(4)-Cl(8)	88.80(14)
C(44)-C(45)-H(45B)	109.5	O(8)-Fe(4)-O(7)	99.91(15)
C(44)-C(45)-H(45C)	109.5	O(8)-Fe(4)-N(19)	166.94(17)
H(45A)-C(45)-H(45B)	109.5	O(8)-Fe(4)-N(20)	90.54(16)
H(45A)-C(45)-H(45C)	109.5	N(19)-Fe(4)-Cl(7)	91.18(19)
H(45B)-C(45)-H(45C)	109.5	N(19)-Fe(4)-Cl(8)	86.91(19)
N(11)-C(46)-H(46A)	109.5	N(19)-Fe(4)-N(20)	77.23(17)
N(11)-C(46)-H(46B)	109.5	N(20)-Fe(4)-Cl(7)	89.68(14)
N(11)-C(46)-H(46C)	109.5	N(20)-Fe(4)-Cl(8)	91.10(14)
H(46A)-C(46)-H(46B)	109.5	C(83)-O(7)-Fe(4)	121.5(3)
H(46A)-C(46)-H(46C)	109.5	C(95)-O(8)-Fe(4)	123.6(3)
H(46B)-C(46)-H(46C)	109.5	C(79)-N(19)-Fe(4)	113.8(4)
C(48)-C(47)-N(12)	119.2(5)	C(81)-N(19)-Fe(4)	126.2(4)

C(81)-N(19)-C(79)	119.3(5)	N(19)-C(79)-H(79A)	109.6
C(80)-N(20)-Fe(4)	113.0(3)	N(19)-C(79)-H(79B)	109.6
C(93)-N(20)-Fe(4)	127.3(3)	H(79A)-C(79)-H(79B)	108.1
C(93)-N(20)-C(80)	119.6(4)	C(80)-C(79)-N(19)	110.2(6)
N(22)-N(21)-C(86)	121.6(4)	C(80)-C(79)-H(79A)	109.6
C(84)-N(21)-N(22)	108.4(4)	C(80)-C(79)-H(79B)	109.6
C(84)-N(21)-C(86)	129.0(5)	N(20)-C(80)-H(80A)	109.6
N(21)-N(22)-C(87)	120.0(4)	N(20)-C(80)-H(80B)	109.6
C(83)-N(22)-N(21)	108.7(4)	C(79)-C(80)-N(20)	110.1(5)
C(83)-N(22)-C(87)	127.4(4)	C(79)-C(80)-H(80A)	109.6
N(24)-N(23)-C(98)	121.6(4)	C(79)-C(80)-H(80B)	109.6
C(96)-N(23)-N(24)	109.0(4)	H(80A)-C(80)-H(80B)	108.2
C(96)-N(23)-C(98)	129.3(4)	N(19)-C(81)-H(81)	118.8
N(23)-N(24)-C(99')	131.4(15)	N(19)-C(81)-C(82)	122.3(5)
N(23)-N(24)-C(99)	119.9(9)	C(82)-C(81)-H(81)	118.8
C(95)-N(24)-N(23)	108.8(4)	C(81)-C(82)-C(83)	127.0(5)
C(95)-N(24)-C(99')	117.7(13)	C(84)-C(82)-C(81)	127.4(5)
C(95)-N(24)-C(99)	131.0(9)	C(84)-C(82)-C(83)	105.6(4)
C(01')-C(00')-H(00')	121.4	O(7)-C(83)-N(22)	122.0(4)
C(99')-C(00')-H(00')	121.4	O(7)-C(83)-C(82)	130.5(4)
C(99')-C(00')-C(01')	117.3(16)	N(22)-C(83)-C(82)	107.5(4)
C(00')-C(01')-H(01')	119.7	N(21)-C(84)-C(82)	109.8(5)
C(02')-C(01')-C(00')	120.5(16)	N(21)-C(84)-C(85)	120.9(5)
C(02')-C(01')-H(01')	119.7	C(82)-C(84)-C(85)	129.3(5)
C(01')-C(02')-H(02')	119.6	C(84)-C(85)-H(85A)	109.5
C(03')-C(02')-C(01')	120.8(16)	C(84)-C(85)-H(85B)	109.5
C(03')-C(02')-H(02')	119.6	C(84)-C(85)-H(85C)	109.5
C(02')-C(03')-H(03')	120.5	H(85A)-C(85)-H(85B)	109.5
C(02')-C(03')-C(04')	119.1(17)	H(85A)-C(85)-H(85C)	109.5
C(04')-C(03')-H(03')	120.5	H(85B)-C(85)-H(85C)	109.5
C(03')-C(04')-H(04')	120.2	N(21)-C(86)-H(86A)	109.5
C(99')-C(04')-C(03')	119.6(17)	N(21)-C(86)-H(86B)	109.5
C(99')-C(04')-H(04')	120.2	N(21)-C(86)-H(86C)	109.5
C(00')-C(99')-N(24)	118.3(18)	H(86A)-C(86)-H(86B)	109.5
C(04')-C(99')-N(24)	118.6(16)	H(86A)-C(86)-H(86C)	109.5
C(04')-C(99')-C(00')	122.7(18)	H(86B)-C(86)-H(86C)	109.5

C(88)-C(87)-N(22)	117.2(5)	N(23)-C(98)-H(98A)	109.5
C(92)-C(87)-N(22)	120.6(5)	N(23)-C(98)-H(98B)	109.5
C(92)-C(87)-C(88)	122.1(5)	N(23)-C(98)-H(98C)	109.5
C(87)-C(88)-H(88)	121.5	H(98A)-C(98)-H(98B)	109.5
C(89)-C(88)-C(87)	117.0(6)	H(98A)-C(98)-H(98C)	109.5
C(89)-C(88)-H(88)	121.5	H(98B)-C(98)-H(98C)	109.5
C(88)-C(89)-H(89)	119.6	C(100)-C(99)-N(24)	115.8(11)
C(90)-C(89)-C(88)	120.9(6)	C(104)-C(99)-N(24)	120.5(12)
C(90)-C(89)-H(89)	119.6	C(104)-C(99)-C(100)	123.0(12)
C(89)-C(90)-H(90)	119.7	C(99)-C(100)-H(100)	121.1
C(91)-C(90)-C(89)	120.7(6)	C(99)-C(100)-C(101)	117.8(12)
C(91)-C(90)-H(90)	119.7	C(101)-C(100)-H(100)	121.1
C(90)-C(91)-H(91)	120.0	C(100)-C(101)-H(101)	120.5
C(90)-C(91)-C(92)	119.9(6)	C(102)-C(101)-C(100)	119.0(11)
C(92)-C(91)-H(91)	120.0	C(102)-C(101)-H(101)	120.5
C(87)-C(92)-C(91)	119.4(6)	C(101)-C(102)-H(102)	119.0
C(87)-C(92)-H(92)	120.3	C(103)-C(102)-C(101)	121.9(11)
C(91)-C(92)-H(92)	120.3	C(103)-C(102)-H(102)	119.0
N(20)-C(93)-H(93)	119.2	C(102)-C(103)-H(103)	120.2
N(20)-C(93)-C(94)	121.6(4)	C(102)-C(103)-C(104)	119.7(13)
C(94)-C(93)-H(93)	119.2	C(104)-C(103)-H(103)	120.2
C(95)-C(94)-C(93)	125.6(5)	C(99)-C(104)-C(103)	118.5(13)
C(96)-C(94)-C(93)	127.2(4)	C(99)-C(104)-H(104)	120.7
C(96)-C(94)-C(95)	107.2(4)	C(103)-C(104)-H(104)	120.7
O(8)-C(95)-N(24)	122.1(4)	N(17)-C(72')-H(72D)	109.5
O(8)-C(95)-C(94)	131.0(5)	N(17)-C(72')-H(72E)	109.5
N(24)-C(95)-C(94)	106.8(4)	N(17)-C(72')-H(72F)	109.5
N(23)-C(96)-C(94)	108.2(4)	H(72D)-C(72')-H(72E)	109.5
N(23)-C(96)-C(97)	121.9(5)	H(72D)-C(72')-H(72F)	109.5
C(94)-C(96)-C(97)	129.8(5)	H(72E)-C(72')-H(72F)	109.5
C(96)-C(97)-H(97A)	109.5		
C(96)-C(97)-H(97B)	109.5		
C(96)-C(97)-H(97C)	109.5		
H(97A)-C(97)-H(97B)	109.5		
H(97A)-C(97)-H(97C)	109.5		
H(97B)-C(97)-H(97C)	109.5		

Table S9. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for **2**. The anisotropic displacement factor exponent takes the form: $-2p^2[\ h^2\ a^{*2}U_{11} + ... + 2\ h\ k\ a^*\ b^*\ U_{12}\ ]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Fe1	37(1)	20(1)	38(1)	1(1)	-4(1)	-1(1)
Cl1	50(1)	29(1)	49(1)	2(1)	9(1)	4(1)
Cl2	41(1)	33(1)	50(1)	0(1)	1(1)	2(1)
O1	36(2)	26(2)	40(2)	1(1)	-6(2)	-2(1)
O2	45(2)	20(1)	46(2)	1(1)	-9(2)	1(1)
N1	37(2)	22(2)	45(2)	6(2)	-5(2)	-2(2)
N2	44(2)	24(2)	40(2)	-1(2)	-7(2)	-2(2)
N3	41(2)	27(2)	47(2)	6(2)	-11(2)	-4(2)
N4	35(2)	24(2)	45(2)	6(2)	-6(2)	-1(2)
N5	50(3)	28(2)	41(2)	3(2)	-18(2)	2(2)
N6	46(3)	26(2)	42(2)	4(2)	-9(2)	-6(2)
C1	52(3)	24(2)	55(3)	6(2)	-12(3)	-14(2)
C2	51(3)	20(2)	48(3)	-1(2)	-4(2)	-9(2)
C3	42(3)	19(2)	33(2)	4(2)	0(2)	-4(2)
C4	35(3)	28(2)	36(2)	0(2)	2(2)	-7(2)
C5	34(2)	25(2)	38(2)	2(2)	2(2)	0(2)
C6	41(3)	25(2)	40(3)	2(2)	0(2)	-2(2)
C7	52(3)	30(3)	58(4)	7(2)	-6(3)	3(2)
C8	44(3)	46(3)	59(4)	9(3)	-19(3)	-8(2)
C9	38(3)	28(2)	43(3)	5(2)	-5(2)	-2(2)
C10	47(3)	39(3)	53(3)	0(2)	9(3)	-9(2)
C11	61(4)	41(3)	64(4)	6(3)	-9(3)	-19(3)
C12	57(4)	30(3)	80(4)	6(3)	-28(3)	-6(3)
C13	53(4)	31(3)	90(5)	-9(3)	-19(3)	5(3)
C14	38(3)	39(3)	68(4)	-6(3)	-4(3)	2(2)
C15	38(3)	29(2)	39(3)	-6(2)	-6(2)	-2(2)
C16	38(3)	26(2)	42(3)	-1(2)	-4(2)	0(2)
C17	37(3)	25(2)	36(2)	-2(2)	-4(2)	5(2)

C18	47(3)	30(2)	49(3)	-1(2)	-13(2)	1(2)
C19	79(5)	33(3)	82(5)	2(3)	-43(4)	-3(3)
C20	87(5)	35(3)	67(4)	7(3)	-46(4)	-3(3)
C21	40(3)	23(2)	49(3)	3(2)	-6(2)	-2(2)
C22	62(4)	34(3)	77(4)	4(3)	9(3)	1(3)
C23	56(4)	55(4)	113(7)	31(4)	9(4)	0(3)
C24	67(4)	35(3)	96(5)	29(3)	-36(4)	-12(3)
C25	91(5)	25(3)	68(4)	5(2)	-32(4)	-7(3)
C26	47(3)	30(2)	55(3)	0(2)	-11(3)	2(2)
Fe3	41(1)	23(1)	52(1)	-1(1)	10(1)	-4(1)
Cl5	58(1)	34(1)	52(1)	-1(1)	5(1)	-6(1)
Cl6	56(1)	57(1)	67(1)	-6(1)	-12(1)	1(1)
O5	47(2)	28(2)	65(2)	-11(2)	21(2)	-10(2)
O6	46(2)	29(2)	50(2)	-7(2)	10(2)	-1(2)
N13	47(3)	21(2)	56(3)	0(2)	11(2)	-4(2)
N14	37(2)	27(2)	47(2)	-2(2)	11(2)	-7(2)
N15	35(2)	24(2)	44(2)	-5(2)	6(2)	-1(2)
N16	33(2)	26(2)	54(3)	-2(2)	8(2)	-6(2)
N17	64(3)	41(3)	74(4)	-17(3)	25(3)	-7(2)
N18	51(3)	34(2)	74(3)	-12(2)	25(3)	-8(2)
C53	59(4)	24(2)	66(4)	0(2)	15(3)	-6(2)
C54	48(3)	29(2)	50(3)	-1(2)	12(2)	-14(2)
C55	43(3)	23(2)	49(3)	-3(2)	4(2)	-1(2)
C56	37(3)	25(2)	45(3)	3(2)	-1(2)	-5(2)
C57	32(2)	27(2)	57(3)	-5(2)	15(2)	-2(2)
C58	35(3)	28(2)	39(3)	-1(2)	-2(2)	-4(2)
C59	44(3)	31(2)	51(3)	-5(2)	10(2)	-5(2)
C60	41(3)	26(2)	53(3)	0(2)	9(2)	-4(2)
C61	34(2)	25(2)	43(3)	-2(2)	1(2)	-2(2)
C62	49(3)	31(2)	49(3)	-5(2)	3(3)	4(2)
C63	69(4)	33(3)	55(3)	0(2)	-2(3)	11(3)
C64	64(4)	31(3)	58(3)	-1(2)	21(3)	-1(2)
C65	42(3)	40(3)	65(4)	-6(3)	3(3)	-12(2)
C66	41(3)	32(2)	57(3)	-1(2)	1(2)	-3(2)
C67	38(3)	33(2)	43(3)	4(2)	7(2)	-4(2)
C68	47(3)	28(2)	47(3)	2(2)	3(2)	-3(2)

C69	40(3)	25(2)	49(3)	-4(2)	3(2)	-1(2)
C70	48(3)	42(3)	54(3)	-7(2)	19(3)	-8(2)
C71	63(4)	54(4)	70(4)	-15(3)	21(4)	-15(3)
C72	82(15)	47(9)	69(12)	-22(9)	42(10)	-9(9)
C73	56(4)	32(3)	48(3)	-2(2)	14(3)	-10(2)
C74	61(4)	32(3)	54(3)	-3(2)	9(3)	2(2)
C75	104(6)	30(3)	61(4)	-2(3)	27(4)	-9(3)
C76	87(5)	61(4)	51(4)	-14(3)	19(4)	-38(4)
C77	80(5)	78(5)	44(3)	0(3)	-11(3)	-35(4)
C78	68(4)	60(4)	40(3)	7(3)	-1(3)	-11(3)
Fe2	45(1)	26(1)	49(1)	-1(1)	-4(1)	-6(1)
Cl3	51(1)	35(1)	62(1)	-3(1)	5(1)	-7(1)
Cl4	67(1)	61(1)	76(1)	-14(1)	17(1)	-6(1)
O3	53(2)	31(2)	55(2)	5(2)	-14(2)	-12(2)
O4	46(2)	32(2)	52(2)	4(2)	-12(2)	-3(2)
N7	56(3)	31(2)	48(3)	-4(2)	-12(2)	-9(2)
N8	55(3)	22(2)	56(3)	3(2)	-9(2)	-2(2)
N9	48(3)	35(2)	40(2)	-2(2)	-9(2)	-6(2)
N10	44(2)	32(2)	39(2)	-4(2)	-3(2)	-7(2)
N11	44(3)	36(2)	69(3)	7(2)	-20(2)	-1(2)
N12	49(3)	27(2)	65(3)	6(2)	-15(2)	-4(2)
C27	90(5)	31(3)	71(4)	0(3)	-16(4)	-25(3)
C28	93(5)	25(2)	77(4)	-1(3)	-28(4)	-11(3)
C29	45(3)	44(3)	48(3)	-9(2)	-3(3)	-7(2)
C30	44(3)	31(2)	43(3)	-4(2)	4(2)	-10(2)
C31	44(3)	32(2)	36(3)	-1(2)	-2(2)	-5(2)
C32	60(4)	40(3)	44(3)	-4(2)	-4(3)	-12(3)
C33	81(5)	46(3)	63(4)	3(3)	-25(4)	-23(3)
C34	104(6)	55(4)	74(5)	12(4)	-47(5)	-23(4)
C35	38(3)	35(2)	39(3)	-2(2)	-1(2)	-2(2)
C36	43(3)	37(3)	49(3)	-8(2)	3(2)	-6(2)
C37	49(3)	33(3)	60(3)	-6(2)	-2(3)	-4(2)
C38	45(3)	33(3)	49(3)	6(2)	-1(2)	-7(2)
C39	55(4)	47(3)	61(4)	-1(3)	12(3)	-7(3)
C40	53(3)	38(3)	57(3)	-2(2)	12(3)	-5(2)
C41	44(3)	23(2)	67(4)	-1(2)	8(3)	0(2)

C42	38(3)	27(2)	58(3)	0(2)	2(2)	2(2)
C43	36(3)	32(2)	51(3)	1(2)	-3(2)	2(2)
C44	40(3)	33(3)	69(4)	1(2)	-3(3)	4(2)
C45	53(4)	39(3)	74(4)	11(3)	-14(3)	-2(3)
C46	94(6)	46(4)	92(6)	15(4)	-42(5)	-15(4)
C47	32(3)	32(2)	52(3)	-1(2)	-7(2)	-2(2)
C48	41(3)	44(3)	90(5)	-15(3)	-7(3)	0(3)
C49	68(5)	34(3)	119(7)	-20(4)	-33(5)	11(3)
C50	72(5)	53(4)	85(5)	13(4)	-41(4)	-24(4)
C51	52(4)	88(5)	61(4)	-5(4)	-4(3)	-26(4)
C52	37(3)	65(4)	61(4)	-20(3)	-1(3)	1(3)
Fe4	41(1)	22(1)	45(1)	-2(1)	-4(1)	1(1)
Cl7	108(2)	54(1)	78(1)	-1(1)	42(1)	-5(1)
Cl8	82(1)	31(1)	80(1)	0(1)	32(1)	0(1)
O7	46(2)	24(2)	63(2)	-7(2)	-12(2)	4(2)
O8	53(2)	29(2)	59(2)	-7(2)	-19(2)	4(2)
N19	65(3)	30(2)	79(4)	-10(2)	-36(3)	11(2)
N20	50(3)	21(2)	47(2)	-1(2)	-12(2)	4(2)
N21	37(2)	27(2)	44(2)	-5(2)	1(2)	0(2)
N22	38(2)	26(2)	38(2)	-2(2)	-2(2)	1(2)
N23	31(2)	30(2)	51(3)	2(2)	-3(2)	0(2)
N24	40(2)	24(2)	48(2)	-4(2)	2(2)	2(2)
C00'	38(8)	36(9)	36(10)	3(7)	-6(8)	-4(7)
C01'	34(7)	35(9)	75(13)	-10(8)	18(10)	-13(7)
C02'	46(7)	31(8)	90(12)	-11(9)	-3(9)	3(7)
C03'	39(9)	31(7)	94(14)	3(10)	-28(11)	7(6)
C04'	52(7)	25(6)	32(9)	-12(7)	-12(8)	-4(5)
C99'	29(7)	25(6)	36(10)	-7(7)	-13(9)	3(5)
C79	95(6)	28(3)	133(7)	-10(4)	-64(6)	21(3)
C80	82(5)	27(3)	79(4)	-5(3)	-35(4)	16(3)
C81	52(3)	32(3)	64(4)	-2(2)	-15(3)	10(2)
C82	42(3)	30(2)	40(3)	-3(2)	1(2)	4(2)
C83	39(3)	28(2)	42(3)	-3(2)	1(2)	-2(2)
C84	39(3)	30(2)	35(2)	0(2)	6(2)	0(2)
C85	45(3)	44(3)	42(3)	-3(2)	2(2)	1(2)
C86	51(3)	33(3)	64(4)	-7(2)	-8(3)	-5(2)

C87	43(3)	23(2)	47(3)	-5(2)	2(2)	-1(2)
C88	62(4)	43(3)	40(3)	-6(2)	6(3)	7(3)
C89	69(5)	61(4)	60(4)	-8(3)	14(3)	23(3)
C90	70(4)	44(3)	72(4)	-21(3)	-8(3)	10(3)
C91	68(4)	28(2)	67(4)	-1(2)	-11(3)	0(3)
C92	49(3)	30(2)	43(3)	-1(2)	-7(2)	-2(2)
C93	51(3)	22(2)	46(3)	-2(2)	-5(2)	-2(2)
C94	44(3)	26(2)	39(3)	5(2)	-1(2)	5(2)
C95	43(3)	25(2)	45(3)	-1(2)	-11(2)	4(2)
C96	35(3)	30(2)	43(3)	0(2)	6(2)	-4(2)
C97	51(3)	29(2)	49(3)	-1(2)	-6(3)	-3(2)
C98	43(3)	41(3)	53(3)	-3(2)	-5(2)	7(2)
C99	36(6)	27(4)	40(7)	-4(5)	-12(6)	1(4)
C100	38(5)	32(6)	51(8)	-2(5)	-2(7)	-4(4)
C101	43(5)	35(7)	79(11)	-14(7)	10(7)	-14(6)
C102	39(6)	33(5)	92(11)	4(6)	-8(7)	7(4)
C103	66(8)	41(5)	95(12)	3(8)	-33(10)	-2(5)
C104	65(6)	36(5)	36(7)	-10(5)	-17(7)	0(4)
Cl10	69(1)	39(1)	47(1)	0(1)	5(1)	-10(1)
Cl12	57(1)	60(1)	54(1)	5(1)	-3(1)	3(1)
Cl11	157(2)	49(1)	47(1)	0(1)	-5(1)	37(1)
Cl9	57(1)	44(1)	53(1)	-2(1)	4(1)	-13(1)
C72'	65(11)	42(7)	71(12)	-12(7)	23(9)	-10(7)

Table S10. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (Å^2 x 10^3) for **2**.

	x	y	z	U(eq)
H1A	5544	5116	5243	53
H1B	6136	4676	5144	53
H2A	6166	4814	5955	48
H2B	5277	4801	5989	48
H3	4624	4977	4747	38
H7A	2750	5089	4322	70
H7B	3229	5021	3847	70
H7C	3609	5238	4319	70
H8A	2403	3748	3716	75
H8B	2310	4347	3744	75
H8C	1942	3996	4142	75
H10	2198	3261	4789	55
H11	1758	2433	4737	66
H12	2406	1835	4303	67
H13	3473	2063	3883	69
H14	3895	2897	3917	58
H15	6400	4147	6423	42
H19A	7590	3277	6913	97
H19B	7016	3228	7344	97
H19C	7045	3735	7035	97
H20A	6183	2114	7175	95
H20B	6955	2402	7243	95
H20C	6852	2012	6813	95
H22	4732	2348	6867	69
H23	4028	1597	6874	90
H24	4310	958	6336	79
H25	5258	1053	5803	73
H26	5963	1795	5784	53
H53A	5554	7725	4465	60
H53B	5277	7439	3993	60

H54A	6553	7312	4018	51
H54B	6526	7161	4571	51
H55	4587	7601	4955	45
H59A	3819	7801	5684	63
H59B	3146	7577	5988	63
H59C	3002	7789	5463	63
H60A	1999	6629	5723	60
H60B	2436	6954	6112	60
H60C	2517	6354	6105	60
H62	4024	5558	5742	51
H63	3675	4716	5821	63
H64	2532	4445	5547	61
H65	1709	5019	5207	59
H66	2066	5870	5107	52
H67	6978	6591	3709	46
H71A	7599	6174	3111	93
H71B	7440	5736	2733	93
H71C	7994	5636	3167	93
H72A	6649	4441	3078	99
H72B	7278	4790	2850	99
H72C	6432	4857	2690	99
H74	6137	4184	4034	59
H75	5301	3514	3897	78
H76	4264	3644	3413	80
H77	3987	4448	3129	81
H78	4847	5102	3238	67
H27A	6234	9680	5561	77
H27B	5956	9874	6072	77
H28A	4767	9976	5811	78
H28B	5260	10198	5384	78
H29	6402	9169	6423	55
H33A	7431	8212	7025	96
H33B	6810	8362	7406	96
H33C	7047	8755	7002	96
H34A	6673	7061	6868	117
H34B	5970	7131	7209	117

H34C	6725	7423	7322	117
H36	5800	6777	6017	52
H37	5025	6065	6040	57
H38	3968	6066	6510	51
H39	3587	6808	6891	65
H40	4336	7526	6844	59
H41	4598	9997	4753	54
H45A	4040	10172	4004	83
H45B	3366	9971	3687	83
H45C	4200	9831	3546	83
H46A	3262	8654	3349	116
H46B	3047	9239	3333	116
H46C	2519	8846	3601	116
H48	3948	7779	3923	70
H49	3327	6998	3944	89
H50	2162	6946	4283	84
H51	1600	7660	4603	80
H52	2235	8426	4603	66
H00'	-2038	4165	5554	44
H01'	-2696	4924	5693	58
H02'	-2836	5517	5081	67
H03'	-2337	5368	4331	65
H04'	-1669	4614	4192	44
H79A	1668	2920	5574	102
H79B	1607	2725	6113	102
H80A	410	2591	6055	75
H80B	746	2379	5566	75
H81	2128	3461	6396	59
H85A	2942	3911	6866	66
H85B	2937	4395	7206	66
H85C	3340	4420	6699	66
H86A	2140	5667	6790	74
H86B	2903	5370	6715	74
H86C	2469	5326	7209	74
H88	632	5224	7193	58
H89	-114	5941	7328	76

H90	-228	6560	6746	74
H91	302	6452	6007	65
H92	1019	5730	5850	49
H93	-251	2517	5106	48
H97A	-1034	2317	4460	65
H97B	-1455	2588	4029	65
H97C	-1909	2409	4488	65
H98A	-2757	3634	4299	69
H98B	-2420	3173	4001	69
H98C	-2157	3738	3892	69
H100	-2149	4300	5442	48
H101	-2681	5123	5461	63
H102	-2698	5604	4769	66
H103	-2168	5314	4071	81
H104	-1608	4508	4052	55
H72D	7160	4471	3451	89
H72E	7525	4787	3027	89
H72F	6751	4505	2947	89

Table S11. Torsion angles [°] for **2**.

Fe1-O1-C5-N4	-160.6(4)	C3-C4-C6-N3	177.9(5)
Fe1-O1-C5-C4	21.9(7)	C3-C4-C6-C7	-1.8(9)
Fe1-O2-C17-N6	-176.8(4)	C5-N4-C9-C10	104.7(6)
Fe1-O2-C17-C16	2.4(8)	C5-N4-C9-C14	-72.2(7)
Fe1-N1-C1-C2	-42.7(5)	C5-C4-C6-N3	0.6(6)
Fe1-N1-C3-C4	-5.2(7)	C5-C4-C6-C7	-179.2(5)
Fe1-N2-C2-C1	-34.2(5)	C6-N3-N4-C5	0.2(6)
Fe1-N2-C15-C16	-2.3(8)	C6-N3-N4-C9	176.4(5)
N1-C1-C2-N2	48.8(6)	C6-C4-C5-O1	177.3(5)
N1-C3-C4-C5	-9.2(8)	C6-C4-C5-N4	-0.5(6)
N1-C3-C4-C6	173.9(5)	C8-N3-N4-C5	-178.8(5)
N2-C15-C16-C17	-7.7(9)	C8-N3-N4-C9	-2.6(8)
N2-C15-C16-C18	-177.3(5)	C8-N3-C6-C4	178.4(5)
N3-N4-C5-O1	-177.8(4)	C8-N3-C6-C7	-1.8(9)
N3-N4-C5-C4	0.2(5)	C9-N4-C5-O1	6.2(8)
N3-N4-C9-C10	-70.7(7)	C9-N4-C5-C4	-175.7(5)
N3-N4-C9-C14	112.4(6)	C9-C10-C11-C12	1.1(10)
N4-N3-C6-C4	-0.5(6)	C10-C9-C14-C13	-1.4(9)
N4-N3-C6-C7	179.3(5)	C10-C11-C12-C13	-1.6(10)
N4-C9-C10-C11	-176.4(5)	C11-C12-C13-C14	0.6(10)
N4-C9-C14-C13	175.5(5)	C12-C13-C14-C9	0.9(10)
N5-N6-C17-O2	-178.4(5)	C14-C9-C10-C11	0.4(9)
N5-N6-C17-C16	2.2(6)	C15-N2-C2-C1	145.6(5)
N5-N6-C21-C22	72.1(7)	C15-C16-C17-O2	8.1(9)
N5-N6-C21-C26	-108.3(6)	C15-C16-C17-N6	-172.6(5)
N6-N5-C18-C16	1.4(7)	C15-C16-C18-N5	171.1(5)
N6-N5-C18-C19	-175.7(6)	C15-C16-C18-C19	-12.0(11)
N6-C21-C22-C23	-178.9(6)	C17-N6-C21-C22	-86.8(7)
N6-C21-C26-C25	178.7(5)	C17-N6-C21-C26	92.8(7)
C1-N1-C3-C4	178.3(5)	C17-C16-C18-N5	-0.1(6)
C2-N2-C15-C16	177.9(5)	C17-C16-C18-C19	176.8(7)
C3-N1-C1-C2	134.3(5)	C18-N5-N6-C17	-2.3(6)
C3-C4-C5-O1	-0.1(9)	C18-N5-N6-C21	-164.9(5)
C3-C4-C5-N4	-177.9(5)	C18-C16-C17-O2	179.4(5)

C18-C16-C17-N6	-1.4(6)	N17-N18-C73-C74	91.3(7)
C20-N5-N6-C17	-178.3(6)	N17-N18-C73-C78	-87.0(7)
C20-N5-N6-C21	19.2(8)	N18-N17-C70-C68	-1.7(8)
C20-N5-C18-C16	176.9(6)	N18-N17-C70-C71	175.9(6)
C20-N5-C18-C19	-0.2(11)	N18-C73-C74-C75	-175.7(5)
C21-N6-C17-O2	-17.5(8)	N18-C73-C78-C77	177.2(6)
C21-N6-C17-C16	163.1(5)	C53-N13-C55-C56	171.5(5)
C21-C22-C23-C24	-0.5(11)	C54-N14-C67-C68	178.5(5)
C22-C21-C26-C25	-1.7(8)	C55-N13-C53-C54	143.3(5)
C22-C23-C24-C25	-0.2(11)	C55-C56-C57-O5	4.0(10)
C23-C24-C25-C26	0.0(10)	C55-C56-C57-N16	-173.8(5)
C24-C25-C26-C21	1.0(9)	C55-C56-C58-N15	175.5(5)
C26-C21-C22-C23	1.5(9)	C55-C56-C58-C59	-3.8(10)
Fe3-O5-C57-N16	-176.2(4)	C57-N16-C61-C62	60.1(8)
Fe3-O5-C57-C56	6.3(9)	C57-N16-C61-C66	-118.5(6)
Fe3-O6-C69-N18	-167.5(4)	C57-C56-C58-N15	0.1(6)
Fe3-O6-C69-C68	14.3(8)	C57-C56-C58-C59	-179.2(5)
Fe3-N13-C53-C54	-39.8(6)	C58-N15-N16-C57	3.3(6)
Fe3-N13-C55-C56	-4.9(8)	C58-N15-N16-C61	171.7(5)
Fe3-N14-C54-C53	-35.5(6)	C58-C56-C57-O5	179.6(6)
Fe3-N14-C67-C68	-3.8(8)	C58-C56-C57-N16	1.8(6)
N13-C53-C54-N14	47.9(6)	C60-N15-N16-C57	-178.2(5)
N13-C55-C56-C57	-4.5(9)	C60-N15-N16-C61	-9.8(7)
N13-C55-C56-C58	-179.2(5)	C60-N15-C58-C56	179.7(5)
N14-C67-C68-C69	-4.5(9)	C60-N15-C58-C59	-1.0(9)
N14-C67-C68-C70	173.1(6)	C61-N16-C57-O5	10.6(9)
N15-N16-C57-O5	178.9(5)	C61-N16-C57-C56	-171.4(5)
N15-N16-C57-C56	-3.1(6)	C61-C62-C63-C64	-1.2(9)
N15-N16-C61-C62	-106.3(6)	C62-C61-C66-C65	-1.8(9)
N15-N16-C61-C66	75.1(7)	C62-C63-C64-C65	-0.9(10)
N16-N15-C58-C56	-2.0(6)	C63-C64-C65-C66	1.8(9)
N16-N15-C58-C59	177.3(5)	C64-C65-C66-C61	-0.4(9)
N16-C61-C62-C63	-176.0(5)	C66-C61-C62-C63	2.7(9)
N16-C61-C66-C65	176.8(5)	C67-N14-C54-C53	142.5(5)
N17-N18-C69-O6	178.3(5)	C67-C68-C69-O6	-1.5(10)
N17-N18-C69-C68	-3.1(7)	C67-C68-C69-N18	-179.9(6)

C67-C68-C70-N17	-178.2(6)	N9-N10-C31-C30	1.7(6)
C67-C68-C70-C71	4.4(11)	N9-N10-C35-C36	-93.7(6)
C69-N18-C73-C74	-118.1(7)	N9-N10-C35-C40	87.1(7)
C69-N18-C73-C78	63.6(9)	N10-N9-C32-C30	2.7(7)
C69-C68-C70-N17	-0.3(7)	N10-N9-C32-C33	-178.3(6)
C69-C68-C70-C71	-177.6(7)	N10-C35-C36-C37	179.1(5)
C70-N17-N18-C69	3.0(8)	N10-C35-C40-C39	-177.8(6)
C70-N17-N18-C73	158.7(6)	N11-N12-C43-O4	179.4(5)
C70-C68-C69-O6	-179.5(6)	N11-N12-C43-C42	-0.2(6)
C70-C68-C69-N18	2.1(7)	N11-N12-C47-C48	96.7(7)
C72-N17-N18-C69	-163.4(19)	N11-N12-C47-C52	-82.8(8)
C72-N17-N18-C73	-8(2)	N12-N11-C44-C42	-1.2(7)
C72-N17-C70-C68	164(2)	N12-N11-C44-C45	175.0(6)
C72-N17-C70-C71	-19(2)	N12-C47-C48-C49	176.9(6)
C73-N18-C69-O6	24.8(10)	N12-C47-C52-C51	-178.0(6)
C73-N18-C69-C68	-156.6(6)	C27-N7-C29-C30	-174.2(6)
C73-C74-C75-C76	-0.4(9)	C28-N8-C41-C42	-172.4(6)
C74-C73-C78-C77	-1.0(10)	C29-N7-C27-C28	-156.9(6)
C74-C75-C76-C77	-3.2(10)	C29-C30-C31-O3	1.4(10)
C75-C76-C77-C78	4.7(11)	C29-C30-C31-N10	-179.5(5)
C76-C77-C78-C73	-2.6(10)	C29-C30-C32-N9	177.8(6)
C78-C73-C74-C75	2.6(9)	C29-C30-C32-C33	-1.2(11)
Fe2-O3-C31-N10	179.2(4)	C31-N10-C35-C36	107.9(6)
Fe2-O3-C31-C30	-1.8(8)	C31-N10-C35-C40	-71.3(7)
Fe2-O4-C43-N12	174.6(4)	C31-C30-C32-N9	-1.5(7)
Fe2-O4-C43-C42	-5.9(8)	C31-C30-C32-C33	179.5(6)
Fe2-N7-C27-C28	28.9(7)	C32-N9-N10-C31	-2.8(6)
Fe2-N7-C29-C30	-0.7(9)	C32-N9-N10-C35	-165.0(5)
Fe2-N8-C28-C27	40.0(7)	C32-C30-C31-O3	-179.3(6)
Fe2-N8-C41-C42	5.6(8)	C32-C30-C31-N10	-0.2(6)
N7-C27-C28-N8	-43.6(8)	C34-N9-N10-C31	-177.1(6)
N7-C29-C30-C31	0.1(9)	C34-N9-N10-C35	20.7(8)
N7-C29-C30-C32	-179.1(6)	C34-N9-C32-C30	176.1(7)
N8-C41-C42-C43	4.9(9)	C34-N9-C32-C33	-4.9(11)
N8-C41-C42-C44	179.0(6)	C35-N10-C31-O3	-18.4(8)
N9-N10-C31-O3	-179.1(5)	C35-N10-C31-C30	162.4(5)

C35-C36-C37-C38	-1.8(9)	Fe4-N19-C81-C82	1.7(10)
C36-C35-C40-C39	3.0(9)	Fe4-N20-C80-C79	-36.4(8)
C36-C37-C38-C39	4.0(9)	Fe4-N20-C93-C94	-1.7(8)
C37-C38-C39-C40	-2.7(10)	N19-C79-C80-N20	43.7(10)
C38-C39-C40-C35	-0.8(10)	N19-C81-C82-C83	0.1(11)
C40-C35-C36-C37	-1.8(9)	N19-C81-C82-C84	-178.3(6)
C41-N8-C28-C27	-141.7(6)	N20-C93-C94-C95	-2.9(9)
C41-C42-C43-O4	-5.0(10)	N20-C93-C94-C96	179.9(5)
C41-C42-C43-N12	174.6(5)	N21-N22-C83-O7	179.8(5)
C41-C42-C44-N11	-173.9(6)	N21-N22-C83-C82	-1.9(6)
C41-C42-C44-C45	10.3(10)	N21-N22-C87-C88	-63.9(6)
C43-N12-C47-C48	-94.3(8)	N21-N22-C87-C92	113.7(6)
C43-N12-C47-C52	86.2(7)	N22-N21-C84-C82	-2.3(6)
C43-C42-C44-N11	1.1(7)	N22-N21-C84-C85	176.8(4)
C43-C42-C44-C45	-174.7(6)	N22-C87-C88-C89	178.0(6)
C44-N11-N12-C43	0.9(7)	N22-C87-C92-C91	-176.9(5)
C44-N11-N12-C47	171.7(5)	N23-N24-C99'-C00'	103(2)
C44-C42-C43-O4	179.9(6)	N23-N24-C99'-C04'	-70(3)
C44-C42-C43-N12	-0.5(6)	N23-N24-C95-O8	-179.2(5)
C46-N11-N12-C43	176.7(6)	N23-N24-C95-C94	-0.3(6)
C46-N11-N12-C47	-12.5(9)	N23-N24-C99-C100	117.7(14)
C46-N11-C44-C42	-176.6(7)	N23-N24-C99-C104	-71(2)
C46-N11-C44-C45	-0.3(11)	N24-N23-C96-C94	0.8(6)
C47-N12-C43-O4	9.1(9)	N24-N23-C96-C97	-179.2(5)
C47-N12-C43-C42	-170.5(5)	N24-C99-C100-C101	173.0(17)
C47-C48-C49-C50	2.1(12)	N24-C99-C104-C103	-173.5(18)
C48-C47-C52-C51	2.5(10)	C00'-C01'-C02'-C03'	0(4)
C48-C49-C50-C51	0.3(12)	C01'-C00'-C99'-N24	-173(3)
C49-C50-C51-C52	-1.3(11)	C01'-C00'-C99'-C04'	-1(4)
C50-C51-C52-C47	0.0(11)	C01'-C02'-C03'-C04'	-1(4)
C52-C47-C48-C49	-3.5(10)	C02'-C03'-C04'-C99'	0(4)
Fe4-O7-C83-N22	176.2(4)	C03'-C04'-C99'-N24	173(3)
Fe4-O7-C83-C82	-1.6(8)	C03'-C04'-C99'-C00'	1(4)
Fe4-O8-C95-N24	-177.5(4)	C99'-N24-C95-O8	-13.8(17)
Fe4-O8-C95-C94	3.9(9)	C99'-N24-C95-C94	165.1(15)
Fe4-N19-C79-C80	-32.2(9)	C99'-C00'-C01'-C02'	1(4)

C79-N19-C81-C82	171.0(7)	C95-C94-C96-N23	-1.0(6)
C80-N20-C93-C94	173.5(6)	C95-C94-C96-C97	179.0(5)
C81-N19-C79-C80	157.2(7)	C96-N23-N24-C99'	-163.1(15)
C81-C82-C83-O7	-0.1(10)	C96-N23-N24-C95	-0.3(6)
C81-C82-C83-N22	-178.1(6)	C96-N23-N24-C99	-174.1(9)
C81-C82-C84-N21	179.8(6)	C96-C94-C95-O8	179.5(6)
C81-C82-C84-C85	0.7(10)	C96-C94-C95-N24	0.8(6)
C83-N22-C87-C88	91.3(7)	C98-N23-N24-C99'	20.0(16)
C83-N22-C87-C92	-91.1(7)	C98-N23-N24-C95	-177.3(5)
C83-C82-C84-N21	1.1(6)	C98-N23-N24-C99	9.0(11)
C83-C82-C84-C85	-177.9(5)	C98-N23-C96-C94	177.5(5)
C84-N21-N22-C83	2.7(6)	C98-N23-C96-C97	-2.6(8)
C84-N21-N22-C87	162.1(4)	C99-N24-C95-O8	-6.4(14)
C84-C82-C83-O7	178.6(5)	C99-N24-C95-C94	172.5(11)
C84-C82-C83-N22	0.5(6)	C99-C100-C101-C102	0(2)
C86-N21-N22-C83	172.3(5)	C100-C99-C104-C103	-3(3)
C86-N21-N22-C87	-28.3(7)	C100-C101-C102-C103	-2(2)
C86-N21-C84-C82	-171.0(5)	C101-C102-C103-C104	1(2)
C86-N21-C84-C85	8.2(8)	C102-C103-C104-C99	2(3)
C87-N22-C83-O7	22.4(8)	C104-C99-C100-C101	2(3)
C87-N22-C83-C82	-159.4(5)	C72'-N17-N18-C69	158.9(14)
C87-C88-C89-C90	-2.0(11)	C72'-N17-N18-C73	-45.4(16)
C88-C87-C92-C91	0.6(8)	C72'-N17-C70-C68	-154.4(16)
C88-C89-C90-C91	2.7(11)	C72'-N17-C70-C71	23.1(19)
C89-C90-C91-C92	-1.6(10)		
C90-C91-C92-C87	-0.1(9)		
C92-C87-C88-C89	0.4(9)		
C93-N20-C80-C79	147.7(7)		
C93-C94-C95-O8	1.9(10)		
C93-C94-C95-N24	-176.9(5)		
C93-C94-C96-N23	176.6(5)		
C93-C94-C96-C97	-3.4(9)		
C95-N24-C99'-C00'	-59(3)		
C95-N24-C99'-C04'	129(2)		
C95-N24-C99-C100	-54(2)		
C95-N24-C99-C104	116.6(15)		

Table S12. Crystal data and structure refinement for **3**.

Identification code	mo_wrm012_0m_a_sq	
Empirical formula	C29.50 H35.50 Fe N9 O13	
Formula weight	780.02	
Temperature	100(2) K	
Wavelength	0.71073 Å	
Crystal system	Triclinic	
Space group	P-1	
Unit cell dimensions	a = 9.8121(5) Å	α= 115.034(2)°.
	b = 14.4735(7) Å	β= 94.913(2)°.
	c = 15.1409(7) Å	γ = 104.992(2)°.
Volume	1834.57(16) $Å^3$	
Z	2	
Density (calculated)	1.412 Mg/m^3	
Absorption coefficient	0.485 mm^{-1}	
F(000)	811	
Crystal size	0.250 x 0.050 x 0.040 mm^3	
Theta range for data collection	2.204 to 26.024°.	
Index ranges	-12<=h<=12, -17<=k<=17, -18<=l<=18	
Reflections collected	75172	
Independent reflections	7168 [R(int) = 0.0267]	
Completeness to theta = 25.242°	99.3 %	
Absorption correction	None	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	7168 / 7 / 523	
Goodness-of-fit on F^2	1.044	
Final R indices [I>2sigma(I)]	R1 = 0.0352, wR2 = 0.0934	
R indices (all data)	R1 = 0.0386, wR2 = 0.0959	
Extinction coefficient	n/a	
Largest diff. peak and hole	0.645 and -0.475 $e.Å^{-3}$	

Table S13. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for **3**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Fe(1)	7502(1)	4890(1)	7984(1)	20(1)
O(1)	6139(1)	3877(1)	6684(1)	22(1)
O(2)	8875(1)	5809(1)	7588(1)	23(1)
O(3)	8876(1)	4037(1)	7808(1)	31(1)
O(4)	6144(1)	5780(1)	8212(1)	24(1)
O(5)	1268(1)	4030(1)	7076(1)	36(1)
O(6)	2447(2)	5646(1)	8253(1)	35(1)
O(7)	3454(1)	4837(1)	7063(1)	29(1)
O(8)	3669(2)	2766(1)	9984(1)	30(1)
O(9)	4452(2)	1850(1)	10614(1)	41(1)
O(10)	4119(2)	1324(1)	9021(1)	33(1)
O(11)	1697(2)	7790(1)	2146(1)	47(1)
O(12A)	2487(17)	8322(5)	3687(5)	267(8)
O(13A)	2147(6)	9437(3)	3209(4)	80(2)
N(9A)	2068(8)	8517(3)	3008(4)	60(2)
O(12B)	1105(7)	8461(5)	3516(4)	83(2)
O(13B)	1471(7)	9391(3)	2716(5)	63(2)
N(9B)	1429(7)	8566(4)	2764(4)	39(1)
N(1)	3875(2)	2666(1)	5765(1)	22(1)
N(2)	2686(2)	2055(1)	5937(1)	29(1)
N(3)	6442(2)	4013(1)	8670(1)	24(1)
N(4)	10930(2)	7189(1)	7863(1)	22(1)
N(5)	12002(2)	7998(1)	8680(1)	24(1)
N(6)	8556(2)	5905(1)	9519(1)	24(1)
N(7)	2391(2)	4848(1)	7477(1)	24(1)
N(8)	4091(2)	1968(1)	9877(1)	26(1)
C(1)	3884(2)	2682(1)	4823(1)	22(1)
C(2)	3814(2)	3593(1)	4753(1)	24(1)
C(3)	3853(2)	3606(2)	3843(1)	30(1)
C(4)	3939(2)	2716(2)	3027(1)	33(1)
C(5)	3984(2)	1814(2)	3110(2)	38(1)

C(6)	3968(2)	1791(2)	4018(1)	33(1)
C(7)	4924(2)	3227(1)	6615(1)	21(1)
C(8)	4384(2)	2934(2)	7331(1)	27(1)
C(9)	2981(2)	2215(2)	6873(1)	34(1)
C(10)	5179(2)	3296(2)	8315(1)	28(1)
C(11)	1897(3)	1713(2)	7312(2)	53(1)
C(12)	1353(2)	1411(2)	5172(2)	35(1)
C(13)	9969(2)	6618(1)	8191(1)	21(1)
C(14)	11700(2)	7966(1)	9510(1)	24(1)
C(15)	10436(2)	7096(1)	9241(1)	22(1)
C(16)	9714(2)	6727(1)	9871(1)	24(1)
C(17)	7229(2)	4341(2)	9679(1)	29(1)
C(18)	7866(2)	5566(2)	10208(1)	28(1)
C(19)	11061(2)	6874(1)	6849(1)	22(1)
C(20)	12260(2)	6600(1)	6550(1)	26(1)
C(21)	12337(2)	6275(2)	5558(2)	32(1)
C(22)	11220(2)	6203(2)	4882(2)	35(1)
C(23)	10022(2)	6457(2)	5190(2)	35(1)
C(24)	9942(2)	6810(2)	6183(1)	29(1)
C(25)	13062(2)	8860(2)	8592(2)	29(1)
C(26)	12591(2)	8801(2)	10515(1)	31(1)
C(31)	10177(3)	10311(2)	1020(3)	65(1)
C(32)	9247(3)	9301(2)	334(3)	65(1)
C(33)	9072(3)	8999(2)	-667(3)	65(1)
C(27)	10296(5)	10549(4)	1985(5)	57(1)

Table S14. Bond lengths [Å] and angles [°] for **3**.

Fe(1)-O(2)	1.9570(12)
Fe(1)-O(1)	1.9763(12)
Fe(1)-O(3)	2.0144(13)
Fe(1)-O(4)	2.0355(12)
Fe(1)-N(3)	2.0947(14)
Fe(1)-N(6)	2.1310(15)
O(1)-C(7)	1.278(2)
O(2)-C(13)	1.279(2)
O(3)-H(1C)	0.924(16)
O(3)-H(1D)	0.913(16)
O(4)-H(1A)	0.920(16)
O(4)-H(1B)	0.914(15)
O(5)-N(7)	1.259(2)
O(6)-N(7)	1.235(2)
O(7)-N(7)	1.2620(19)
O(8)-N(8)	1.278(2)
O(9)-N(8)	1.233(2)
O(10)-N(8)	1.244(2)
O(11)-N(9A)	1.227(5)
O(11)-N(9B)	1.229(5)
O(12A)-N(9A)	1.240(7)
O(13A)-N(9A)	1.213(6)
O(12B)-N(9B)	1.267(7)
O(13B)-N(9B)	1.217(7)
N(1)-C(7)	1.353(2)
N(1)-N(2)	1.388(2)
N(1)-C(1)	1.437(2)
N(2)-C(9)	1.328(2)
N(2)-C(12)	1.455(2)
N(3)-C(10)	1.285(2)
N(3)-C(17)	1.466(2)
N(4)-C(13)	1.357(2)
N(4)-N(5)	1.387(2)
N(4)-C(19)	1.433(2)

N(5)-C(14)	1.331(2)
N(5)-C(25)	1.465(2)
N(6)-C(16)	1.285(2)
N(6)-C(18)	1.473(2)
C(1)-C(6)	1.378(3)
C(1)-C(2)	1.385(2)
C(2)-C(3)	1.390(2)
C(2)-H(2)	0.9500
C(3)-C(4)	1.386(3)
C(3)-H(3)	0.9500
C(4)-C(5)	1.375(3)
C(4)-H(4)	0.9500
C(5)-C(6)	1.390(3)
C(5)-H(5)	0.9500
C(6)-H(6)	0.9500
C(7)-C(8)	1.416(2)
C(8)-C(9)	1.393(3)
C(8)-C(10)	1.426(3)
C(9)-C(11)	1.493(3)
C(10)-H(10)	0.9500
C(11)-H(11A)	0.9800
C(11)-H(11B)	0.9800
C(11)-H(11C)	0.9800
C(12)-H(12A)	0.9800
C(12)-H(12B)	0.9800
C(12)-H(12C)	0.9800
C(13)-C(15)	1.413(2)
C(14)-C(15)	1.401(2)
C(14)-C(26)	1.487(2)
C(15)-C(16)	1.431(2)
C(16)-H(16)	0.9500
C(17)-C(18)	1.522(3)
C(17)-H(17A)	0.9900
C(17)-H(17B)	0.9900
C(18)-H(18A)	0.9900
C(18)-H(18B)	0.9900

C(19)-C(24)	1.381(2)
C(19)-C(20)	1.388(2)
C(20)-C(21)	1.387(3)
C(20)-H(20)	0.9500
C(21)-C(22)	1.386(3)
C(21)-H(21)	0.9500
C(22)-C(23)	1.381(3)
C(22)-H(22)	0.9500
C(23)-C(24)	1.386(3)
C(23)-H(23)	0.9500
C(24)-H(24)	0.9500
C(25)-H(25A)	0.9800
C(25)-H(25B)	0.9800
C(25)-H(25C)	0.9800
C(26)-H(26A)	0.9800
C(26)-H(26B)	0.9800
C(26)-H(26C)	0.9800
C(31)-C(27)	1.338(7)
C(31)-C(32)	1.382(4)
C(31)-C(33)#1	1.401(4)
C(32)-C(33)	1.369(5)
C(32)-H(32)	0.9500
C(33)-H(33)	0.9500
C(27)-H(27A)	0.9800
C(27)-H(27B)	0.9800
C(27)-H(27C)	0.9800

O(2)-Fe(1)-O(1)	100.49(5)
O(2)-Fe(1)-O(3)	86.53(5)
O(1)-Fe(1)-O(3)	95.49(5)
O(2)-Fe(1)-O(4)	93.67(5)
O(1)-Fe(1)-O(4)	86.34(5)
O(3)-Fe(1)-O(4)	178.09(6)
O(2)-Fe(1)-N(3)	166.81(5)
O(1)-Fe(1)-N(3)	91.34(5)
O(3)-Fe(1)-N(3)	86.55(6)

O(4)-Fe(1)-N(3)	92.87(5)
O(2)-Fe(1)-N(6)	90.89(5)
O(1)-Fe(1)-N(6)	167.14(5)
O(3)-Fe(1)-N(6)	91.13(6)
O(4)-Fe(1)-N(6)	86.98(5)
N(3)-Fe(1)-N(6)	78.03(6)
C(7)-O(1)-Fe(1)	122.04(10)
C(13)-O(2)-Fe(1)	124.40(11)
Fe(1)-O(3)-H(1C)	122.2(15)
Fe(1)-O(3)-H(1D)	123.5(15)
H(1C)-O(3)-H(1D)	112(2)
Fe(1)-O(4)-H(1A)	121.6(14)
Fe(1)-O(4)-H(1B)	120.9(14)
H(1A)-O(4)-H(1B)	110.8(19)
O(13A)-N(9A)-O(11)	122.1(5)
O(13A)-N(9A)-O(12A)	119.0(5)
O(11)-N(9A)-O(12A)	118.7(5)
O(13B)-N(9B)-O(11)	126.5(5)
O(13B)-N(9B)-O(12B)	119.6(5)
O(11)-N(9B)-O(12B)	114.0(5)
C(7)-N(1)-N(2)	108.80(13)
C(7)-N(1)-C(1)	128.61(14)
N(2)-N(1)-C(1)	122.55(14)
C(9)-N(2)-N(1)	109.07(15)
C(9)-N(2)-C(12)	129.51(16)
N(1)-N(2)-C(12)	121.34(14)
C(10)-N(3)-C(17)	119.75(15)
C(10)-N(3)-Fe(1)	126.25(12)
C(17)-N(3)-Fe(1)	113.85(11)
C(13)-N(4)-N(5)	108.37(13)
C(13)-N(4)-C(19)	126.68(14)
N(5)-N(4)-C(19)	123.24(13)
C(14)-N(5)-N(4)	109.29(14)
C(14)-N(5)-C(25)	128.19(15)
N(4)-N(5)-C(25)	121.11(14)
C(16)-N(6)-C(18)	119.41(15)

C(16)-N(6)-Fe(1)	126.71(12)
C(18)-N(6)-Fe(1)	113.85(11)
O(6)-N(7)-O(5)	120.78(15)
O(6)-N(7)-O(7)	120.72(15)
O(5)-N(7)-O(7)	118.49(15)
O(9)-N(8)-O(10)	121.77(16)
O(9)-N(8)-O(8)	119.92(15)
O(10)-N(8)-O(8)	118.31(15)
C(6)-C(1)-C(2)	122.04(16)
C(6)-C(1)-N(1)	118.81(16)
C(2)-C(1)-N(1)	119.14(15)
C(1)-C(2)-C(3)	118.47(17)
C(1)-C(2)-H(2)	120.8
C(3)-C(2)-H(2)	120.8
C(4)-C(3)-C(2)	120.11(18)
C(4)-C(3)-H(3)	119.9
C(2)-C(3)-H(3)	119.9
C(5)-C(4)-C(3)	120.38(17)
C(5)-C(4)-H(4)	119.8
C(3)-C(4)-H(4)	119.8
C(4)-C(5)-C(6)	120.40(18)
C(4)-C(5)-H(5)	119.8
C(6)-C(5)-H(5)	119.8
C(1)-C(6)-C(5)	118.58(18)
C(1)-C(6)-H(6)	120.7
C(5)-C(6)-H(6)	120.7
O(1)-C(7)-N(1)	122.77(15)
O(1)-C(7)-C(8)	130.53(16)
N(1)-C(7)-C(8)	106.71(15)
C(9)-C(8)-C(7)	106.94(16)
C(9)-C(8)-C(10)	127.97(16)
C(7)-C(8)-C(10)	125.05(17)
N(2)-C(9)-C(8)	108.44(16)
N(2)-C(9)-C(11)	122.47(18)
C(8)-C(9)-C(11)	129.03(18)
N(3)-C(10)-C(8)	121.86(16)

N(3)-C(10)-H(10)	119.1
C(8)-C(10)-H(10)	119.1
C(9)-C(11)-H(11A)	109.5
C(9)-C(11)-H(11B)	109.5
H(11A)-C(11)-H(11B)	109.5
C(9)-C(11)-H(11C)	109.5
H(11A)-C(11)-H(11C)	109.5
H(11B)-C(11)-H(11C)	109.5
N(2)-C(12)-H(12A)	109.5
N(2)-C(12)-H(12B)	109.5
H(12A)-C(12)-H(12B)	109.5
N(2)-C(12)-H(12C)	109.5
H(12A)-C(12)-H(12C)	109.5
H(12B)-C(12)-H(12C)	109.5
O(2)-C(13)-N(4)	121.69(15)
O(2)-C(13)-C(15)	130.94(15)
N(4)-C(13)-C(15)	107.38(15)
N(5)-C(14)-C(15)	108.39(15)
N(5)-C(14)-C(26)	121.40(16)
C(15)-C(14)-C(26)	130.11(16)
C(14)-C(15)-C(13)	106.52(15)
C(14)-C(15)-C(16)	128.71(16)
C(13)-C(15)-C(16)	124.77(16)
N(6)-C(16)-C(15)	122.09(16)
N(6)-C(16)-H(16)	119.0
C(15)-C(16)-H(16)	119.0
N(3)-C(17)-C(18)	107.63(14)
N(3)-C(17)-H(17A)	110.2
C(18)-C(17)-H(17A)	110.2
N(3)-C(17)-H(17B)	110.2
C(18)-C(17)-H(17B)	110.2
H(17A)-C(17)-H(17B)	108.5
N(6)-C(18)-C(17)	107.99(14)
N(6)-C(18)-H(18A)	110.1
C(17)-C(18)-H(18A)	110.1
N(6)-C(18)-H(18B)	110.1

C(17)-C(18)-H(18B)	110.1
H(18A)-C(18)-H(18B)	108.4
C(24)-C(19)-C(20)	121.87(16)
C(24)-C(19)-N(4)	118.18(15)
C(20)-C(19)-N(4)	119.89(15)
C(21)-C(20)-C(19)	118.44(17)
C(21)-C(20)-H(20)	120.8
C(19)-C(20)-H(20)	120.8
C(22)-C(21)-C(20)	120.29(17)
C(22)-C(21)-H(21)	119.9
C(20)-C(21)-H(21)	119.9
C(23)-C(22)-C(21)	120.29(18)
C(23)-C(22)-H(22)	119.9
C(21)-C(22)-H(22)	119.9
C(22)-C(23)-C(24)	120.27(18)
C(22)-C(23)-H(23)	119.9
C(24)-C(23)-H(23)	119.9
C(19)-C(24)-C(23)	118.80(17)
C(19)-C(24)-H(24)	120.6
C(23)-C(24)-H(24)	120.6
N(5)-C(25)-H(25A)	109.5
N(5)-C(25)-H(25B)	109.5
H(25A)-C(25)-H(25B)	109.5
N(5)-C(25)-H(25C)	109.5
H(25A)-C(25)-H(25C)	109.5
H(25B)-C(25)-H(25C)	109.5
C(14)-C(26)-H(26A)	109.5
C(14)-C(26)-H(26B)	109.5
H(26A)-C(26)-H(26B)	109.5
C(14)-C(26)-H(26C)	109.5
H(26A)-C(26)-H(26C)	109.5
H(26B)-C(26)-H(26C)	109.5
C(27)-C(31)-C(32)	116.4(4)
C(27)-C(31)-C(33)#1	124.9(4)
C(32)-C(31)-C(33)#1	118.7(3)
C(33)-C(32)-C(31)	119.8(3)

C(33)-C(32)-H(32)	120.1
C(31)-C(32)-H(32)	120.1
C(32)-C(33)-C(31)#1	121.6(3)
C(32)-C(33)-H(33)	119.2
C(31)#1-C(33)-H(33)	119.2
C(31)-C(27)-H(27A)	109.5
C(31)-C(27)-H(27B)	109.5
H(27A)-C(27)-H(27B)	109.5
C(31)-C(27)-H(27C)	109.5
H(27A)-C(27)-H(27C)	109.5
H(27B)-C(27)-H(27C)	109.5

Symmetry transformations used to generate equivalent atoms:

#1 -x+2,-y+2,-z

Table S15. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for **3**. The anisotropic displacement factor exponent takes the form: $-2p^2[\ h^2\ a^{*2}U^{11} + ...\ + 2\ h\ k\ a^*\ b^*\ U^{12}\ ]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
Fe(1)	18(1)	27(1)	18(1)	14(1)	6(1)	7(1)
O(1)	22(1)	27(1)	20(1)	14(1)	7(1)	5(1)
O(2)	21(1)	27(1)	20(1)	13(1)	6(1)	4(1)
O(3)	26(1)	40(1)	43(1)	29(1)	16(1)	16(1)
O(4)	22(1)	31(1)	22(1)	13(1)	6(1)	11(1)
O(5)	25(1)	45(1)	32(1)	14(1)	7(1)	7(1)
O(6)	45(1)	40(1)	23(1)	12(1)	13(1)	21(1)
O(7)	24(1)	42(1)	27(1)	16(1)	11(1)	15(1)
O(8)	39(1)	30(1)	26(1)	16(1)	12(1)	16(1)
O(9)	46(1)	58(1)	37(1)	33(1)	13(1)	26(1)
O(10)	38(1)	31(1)	32(1)	13(1)	14(1)	13(1)
O(11)	71(1)	34(1)	30(1)	13(1)	9(1)	13(1)
O(12A)	600(20)	79(4)	73(4)	7(3)	-86(7)	132(8)
O(13A)	105(4)	26(2)	84(3)	12(2)	-17(3)	16(2)
N(9A)	82(4)	26(2)	53(3)	12(2)	-17(3)	8(2)
O(12B)	118(5)	103(4)	66(3)	53(3)	76(3)	53(4)
O(13B)	73(4)	33(2)	86(4)	25(2)	41(3)	16(2)
N(9B)	44(3)	40(3)	37(3)	20(2)	14(2)	13(2)
N(1)	22(1)	25(1)	22(1)	14(1)	7(1)	4(1)
N(2)	24(1)	35(1)	27(1)	19(1)	6(1)	0(1)
N(3)	25(1)	33(1)	22(1)	18(1)	8(1)	10(1)
N(4)	19(1)	25(1)	22(1)	12(1)	4(1)	5(1)
N(5)	19(1)	24(1)	26(1)	11(1)	3(1)	4(1)
N(6)	22(1)	34(1)	20(1)	17(1)	7(1)	11(1)
N(7)	24(1)	36(1)	19(1)	16(1)	6(1)	15(1)
N(8)	24(1)	31(1)	30(1)	19(1)	10(1)	9(1)
C(1)	20(1)	28(1)	19(1)	13(1)	4(1)	4(1)
C(2)	20(1)	29(1)	24(1)	14(1)	4(1)	6(1)
C(3)	24(1)	39(1)	31(1)	24(1)	3(1)	6(1)
C(4)	26(1)	46(1)	22(1)	20(1)	2(1)	-1(1)
C(5)	45(1)	36(1)	23(1)	8(1)	13(1)	5(1)

C(6)	42(1)	28(1)	29(1)	14(1)	14(1)	10(1)
C(7)	24(1)	23(1)	21(1)	13(1)	8(1)	9(1)
C(8)	29(1)	31(1)	26(1)	19(1)	9(1)	6(1)
C(9)	32(1)	40(1)	28(1)	21(1)	7(1)	1(1)
C(10)	30(1)	34(1)	26(1)	21(1)	9(1)	7(1)
C(11)	43(1)	69(2)	40(1)	35(1)	8(1)	-10(1)
C(12)	27(1)	43(1)	29(1)	18(1)	2(1)	-2(1)
C(13)	19(1)	24(1)	23(1)	13(1)	8(1)	9(1)
C(14)	22(1)	28(1)	25(1)	12(1)	6(1)	11(1)
C(15)	20(1)	27(1)	22(1)	12(1)	6(1)	10(1)
C(16)	22(1)	32(1)	20(1)	12(1)	5(1)	12(1)
C(17)	28(1)	41(1)	24(1)	23(1)	6(1)	10(1)
C(18)	27(1)	42(1)	20(1)	19(1)	7(1)	10(1)
C(19)	22(1)	24(1)	24(1)	15(1)	8(1)	5(1)
C(20)	22(1)	31(1)	32(1)	19(1)	8(1)	8(1)
C(21)	29(1)	37(1)	34(1)	19(1)	16(1)	12(1)
C(22)	36(1)	42(1)	26(1)	17(1)	12(1)	8(1)
C(23)	31(1)	46(1)	29(1)	22(1)	3(1)	9(1)
C(24)	23(1)	38(1)	31(1)	21(1)	8(1)	10(1)
C(25)	24(1)	27(1)	34(1)	17(1)	4(1)	2(1)
C(26)	24(1)	33(1)	28(1)	10(1)	4(1)	7(1)
C(31)	35(1)	60(2)	116(3)	46(2)	24(2)	31(1)
C(32)	29(1)	54(2)	127(3)	51(2)	21(2)	19(1)
C(33)	29(1)	44(1)	128(3)	42(2)	16(1)	18(1)
C(27)	38(3)	55(3)	88(4)	34(3)	22(3)	25(2)

Table S16. Hydrogen coordinates (x 10^4) and isotropic displacement parameters ($Å^2$x 10^3) for **3**.

	x	y	z	U(eq)
H(1C)	8600(30)	3350(14)	7751(19)	46
H(1D)	9720(20)	4195(19)	7605(18)	46
H(1A)	5205(18)	5489(17)	7846(15)	36
H(1B)	6230(20)	6326(15)	8830(13)	36
H(2)	3740	4195	5314	29
H(3)	3821	4226	3780	35
H(4)	3968	2729	2407	40
H(5)	4025	1202	2544	46
H(6)	4016	1174	4082	39
H(10)	4763	2998	8719	33
H(11A)	2372	1838	7967	80
H(11B)	1483	936	6867	80
H(11C)	1125	2037	7389	80
H(12A)	570	1195	5477	53
H(12B)	1488	766	4659	53
H(12C)	1104	1839	4865	53
H(16)	10102	7103	10572	28
H(17A)	8010	4018	9643	34
H(17B)	6563	4095	10050	34
H(18A)	7093	5888	10394	33
H(18B)	8590	5811	10826	33
H(20)	13010	6634	7015	32
H(21)	13158	6100	5341	38
H(22)	11279	5979	4203	42
H(23)	9249	6389	4719	42
H(24)	9133	7005	6403	34
H(25A)	13250	9546	9194	43
H(25B)	12679	8916	8004	43
H(25C)	13965	8693	8520	43
H(26A)	12227	8625	11027	46

H(26B)	12532	9508	10632	46
H(26C)	13600	8820	10550	46
H(32)	8732	8819	556	78
H(33)	8426	8306	-1134	78
H(27A)	10802	11323	2403	86
H(27B)	10844	10139	2141	86
H(27C)	9328	10360	2117	86

Table S17. Torsion angles [°] for **3**.

C(7)-N(1)-N(2)-C(9)	-0.8(2)
C(1)-N(1)-N(2)-C(9)	-178.79(16)
C(7)-N(1)-N(2)-C(12)	176.44(17)
C(1)-N(1)-N(2)-C(12)	-1.5(3)
C(13)-N(4)-N(5)-C(14)	-2.13(18)
C(19)-N(4)-N(5)-C(14)	-168.12(15)
C(13)-N(4)-N(5)-C(25)	-169.66(15)
C(19)-N(4)-N(5)-C(25)	24.4(2)
C(7)-N(1)-C(1)-C(6)	109.4(2)
N(2)-N(1)-C(1)-C(6)	-73.0(2)
C(7)-N(1)-C(1)-C(2)	-70.2(2)
N(2)-N(1)-C(1)-C(2)	107.33(19)
C(6)-C(1)-C(2)-C(3)	-1.0(3)
N(1)-C(1)-C(2)-C(3)	178.69(15)
C(1)-C(2)-C(3)-C(4)	0.9(3)
C(2)-C(3)-C(4)-C(5)	0.1(3)
C(3)-C(4)-C(5)-C(6)	-1.2(3)
C(2)-C(1)-C(6)-C(5)	0.0(3)
N(1)-C(1)-C(6)-C(5)	-179.68(17)
C(4)-C(5)-C(6)-C(1)	1.1(3)
Fe(1)-O(1)-C(7)-N(1)	168.19(12)
Fe(1)-O(1)-C(7)-C(8)	-12.1(3)
N(2)-N(1)-C(7)-O(1)	-178.65(15)
C(1)-N(1)-C(7)-O(1)	-0.8(3)
N(2)-N(1)-C(7)-C(8)	1.60(19)
C(1)-N(1)-C(7)-C(8)	179.43(16)
O(1)-C(7)-C(8)-C(9)	178.47(18)
N(1)-C(7)-C(8)-C(9)	-1.8(2)
O(1)-C(7)-C(8)-C(10)	-3.4(3)
N(1)-C(7)-C(8)-C(10)	176.28(17)
N(1)-N(2)-C(9)-C(8)	-0.4(2)
C(12)-N(2)-C(9)-C(8)	-177.32(19)
N(1)-N(2)-C(9)-C(11)	177.1(2)
C(12)-N(2)-C(9)-C(11)	0.1(4)

C(7)-C(8)-C(9)-N(2)	1.3(2)
C(10)-C(8)-C(9)-N(2)	-176.67(19)
C(7)-C(8)-C(9)-C(11)	-175.9(2)
C(10)-C(8)-C(9)-C(11)	6.1(4)
C(17)-N(3)-C(10)-C(8)	-178.68(17)
Fe(1)-N(3)-C(10)-C(8)	6.1(3)
C(9)-C(8)-C(10)-N(3)	-175.8(2)
C(7)-C(8)-C(10)-N(3)	6.5(3)
Fe(1)-O(2)-C(13)-N(4)	176.16(11)
Fe(1)-O(2)-C(13)-C(15)	-3.9(2)
N(5)-N(4)-C(13)-O(2)	-178.96(14)
C(19)-N(4)-C(13)-O(2)	-13.6(3)
N(5)-N(4)-C(13)-C(15)	1.11(17)
C(19)-N(4)-C(13)-C(15)	166.48(15)
N(4)-N(5)-C(14)-C(15)	2.26(19)
C(25)-N(5)-C(14)-C(15)	168.66(16)
N(4)-N(5)-C(14)-C(26)	-174.54(15)
C(25)-N(5)-C(14)-C(26)	-8.1(3)
N(5)-C(14)-C(15)-C(13)	-1.54(19)
C(26)-C(14)-C(15)-C(13)	174.89(17)
N(5)-C(14)-C(15)-C(16)	178.44(16)
C(26)-C(14)-C(15)-C(16)	-5.1(3)
O(2)-C(13)-C(15)-C(14)	-179.69(17)
N(4)-C(13)-C(15)-C(14)	0.23(18)
O(2)-C(13)-C(15)-C(16)	0.3(3)
N(4)-C(13)-C(15)-C(16)	-179.76(15)
C(18)-N(6)-C(16)-C(15)	179.97(15)
Fe(1)-N(6)-C(16)-C(15)	2.3(2)
C(14)-C(15)-C(16)-N(6)	-179.50(17)
C(13)-C(15)-C(16)-N(6)	0.5(3)
C(10)-N(3)-C(17)-C(18)	-135.17(17)
Fe(1)-N(3)-C(17)-C(18)	40.59(17)
C(16)-N(6)-C(18)-C(17)	-145.07(16)
Fe(1)-N(6)-C(18)-C(17)	32.89(17)
N(3)-C(17)-C(18)-N(6)	-46.45(18)
C(13)-N(4)-C(19)-C(24)	67.0(2)

N(5)-N(4)-C(19)-C(24)	-129.65(17)
C(13)-N(4)-C(19)-C(20)	-110.20(19)
N(5)-N(4)-C(19)-C(20)	53.1(2)
C(24)-C(19)-C(20)-C(21)	1.4(3)
N(4)-C(19)-C(20)-C(21)	178.47(16)
C(19)-C(20)-C(21)-C(22)	-1.5(3)
C(20)-C(21)-C(22)-C(23)	0.1(3)
C(21)-C(22)-C(23)-C(24)	1.6(3)
C(20)-C(19)-C(24)-C(23)	0.3(3)
N(4)-C(19)-C(24)-C(23)	-176.88(16)
C(22)-C(23)-C(24)-C(19)	-1.8(3)
C(27)-C(31)-C(32)-C(33)	-179.8(3)
C(33)#1-C(31)-C(32)-C(33)	-0.4(4)
C(31)-C(32)-C(33)-C(31)#1	0.4(4)

Symmetry transformations used to generate equivalent atoms:

#1 -x+2,-y+2,-z

Table S18. Hydrogen bonds for **3** [Å and °].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
O(3)-H(1C)...O(11)#2	0.924(16)	1.676(17)	2.590(2)	169(2)
O(3)-H(1C)...O(12A^a)#2	0.924(16)	2.35(2)	3.025(6)	129(2)
O(3)-H(1C)...N(9A^a)#2	0.924(16)	2.319(18)	3.197(5)	158(2)
O(3)-H(1C)...O(12B^b)#2	0.924(16)	2.62(2)	3.317(6)	132.6(19)
O(3)-H(1C)...N(9B^b)#2	0.924(16)	2.534(19)	3.412(6)	159(2)
O(3)-H(1D)...O(5)#3	0.913(16)	1.803(18)	2.6805(18)	161(2)
O(3)-H(1D)...N(7)#3	0.913(16)	2.603(17)	3.4962(19)	166(2)
O(4)-H(1A)...O(7)	0.920(16)	1.764(16)	2.6822(18)	175(2)
O(4)-H(1A)...N(7)	0.920(16)	2.602(17)	3.4752(19)	158.6(18)
O(4)-H(1B)...O(8)#4	0.914(15)	1.693(16)	2.6029(18)	174(2)
O(4)-H(1B)...N(8)#4	0.914(15)	2.538(17)	3.402(2)	157.9(19)
C(2)-H(2)...O(7)	0.95	2.48	3.288(2)	143.3
C(6)-H(6)...O(13A^a)#5	0.95	2.41	3.041(4)	123.4
C(10)-H(10)...O(8)	0.95	2.37	3.296(2)	163.2
C(10)-H(10)...O(10)	0.95	2.58	3.407(2)	145.2
C(11)-H(11C)...O(12B^b)#6	0.98	2.24	3.004(7)	134.0
C(11)-H(11C)...N(9B^b)#6	0.98	2.39	3.169(7)	136.5
C(12)-H(12A)...O(12B^b)#6	0.98	2.34	3.246(5)	153.2
C(12)-H(12B)...O(13A^a)#5	0.98	2.50	3.459(5)	166.0
C(16)-H(16)...O(11)#7	0.95	2.38	3.292(2)	160.6
C(17)-H(17B)...O(6)#4	0.99	2.50	3.112(2)	119.6
C(18)-H(18A)...O(8)#4	0.99	2.51	3.253(2)	131.5
C(25)-H(25A)...O(10)#8	0.98	2.62	3.198(2)	117.5
C(25)-H(25C)...O(9)#9	0.98	2.43	3.208(2)	136.0
C(26)-H(26A)...O(11)#7	0.98	2.48	3.430(3)	163.0
C(26)-H(26A)...O(13B^b)#7	0.98	2.58	3.428(6)	144.4
C(26)-H(26C)...O(10)#9	0.98	2.36	3.312(2)	163.0

Symmetry transformations used to generate equivalent atoms:

#1 -x+2,-y+2,-z #2 -x+1,-y+1,-z+1 #3 x+1,y,z

#4 -x+1,-y+1,-z+2 #5 x,y-1,z #6 -x,-y+1,-z+1

#7 x+1,y,z+1 #8 x+1,y+1,z #9 -x+2,-y+1,-z+2

Table S19. Crystal data and structure refinement for **4**.

Identification code	WRM007_a	
Empirical formula	C27 H32 Cl2 N6 O3 Zn	
Formula weight	624.85	
Temperature	100(2) K	
Wavelength	0.71073 Å	
Crystal system	Orthorhombic	
Space group	Pbca	
Unit cell dimensions	a = 16.6505(13) Å	α= 90°.
	b = 15.7096(13) Å	β= 90°.
	c = 21.6693(18) Å	γ = 90°.
Volume	5668.1(8) $Å^3$	
Z	8	
Density (calculated)	1.464 Mg/m^3	
Absorption coefficient	1.095 mm^{-1}	
F(000)	2592	
Crystal size	0.403 x 0.222 x 0.162 mm^3	
Theta range for data collection	2.243 to 26.018°.	
Index ranges	-20<=h<=20, -19<=k<=19, -26<=l<=25	
Reflections collected	53808	
Independent reflections	5297 [R(int) = 0.0978]	
Completeness to theta = 25.242°	96.5 %	
Absorption correction	None	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	5297 / 0 / 359	
Goodness-of-fit on F^2	1.285	
Final R indices [I>2sigma(I)]	R1 = 0.0727, wR2 = 0.1514	
R indices (all data)	R1 = 0.0869, wR2 = 0.1585	
Extinction coefficient	0.0042(5)	
Largest diff. peak and hole	0.923 and -0.762 $e.Å^{-3}$	

Table S20. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for **4**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Zn(1)	2837(1)	6333(1)	2626(1)	14(1)
Cl(1)	1771(1)	5518(1)	2342(1)	25(1)
O(1)	3688(2)	5378(2)	2749(1)	20(1)
O(2)	2772(1)	6484(2)	3562(1)	17(1)
N(1)	3474(2)	6729(2)	1818(1)	16(1)
N(2)	5086(2)	4373(2)	1823(1)	16(1)
N(3)	4657(2)	4463(2)	2370(1)	17(1)
N(4)	2493(2)	7620(2)	2513(1)	15(1)
N(5)	2299(2)	8099(2)	4579(2)	16(1)
N(6)	2504(2)	7264(2)	4439(1)	16(1)
C(1)	4167(2)	5165(2)	2328(2)	15(1)
C(2)	4327(2)	5527(2)	1737(2)	14(1)
C(3)	3990(2)	6301(2)	1505(2)	14(1)
C(4)	4921(2)	5038(2)	1458(2)	16(1)
C(5)	5369(2)	5189(2)	875(2)	23(1)
C(6)	5799(2)	3837(2)	1792(2)	22(1)
C(7)	4630(2)	3809(2)	2828(2)	19(1)
C(8)	4447(2)	2979(2)	2658(2)	22(1)
C(9)	4403(2)	2365(3)	3121(2)	28(1)
C(10)	4529(2)	2586(3)	3728(2)	29(1)
C(11)	4717(2)	3412(3)	3891(2)	27(1)
C(12)	4769(2)	4034(3)	3438(2)	23(1)
C(13)	2597(2)	7184(2)	3815(2)	14(1)
C(14)	2450(2)	8012(2)	3565(2)	13(1)
C(15)	2407(2)	8201(2)	2925(2)	15(1)
C(16)	2299(2)	8553(2)	4059(2)	16(1)
C(17)	2166(2)	9490(2)	4063(2)	21(1)
C(18)	2304(3)	8397(2)	5215(2)	24(1)
C(19)	2505(2)	6614(2)	4902(2)	16(1)
C(20)	1784(2)	6342(2)	5148(2)	23(1)
C(21)	1790(3)	5736(3)	5608(2)	33(1)

C(22)	2506(3)	5404(3)	5817(2)	31(1)
C(23)	3222(3)	5671(2)	5561(2)	29(1)
C(24)	3232(2)	6284(2)	5100(2)	21(1)
C(25)	3266(2)	7580(2)	1588(2)	18(1)
C(26)	2462(2)	7839(2)	1864(2)	19(1)
Cl(2)	5548(1)	1867(1)	5132(1)	26(1)
O(3)	4424(2)	2676(2)	6119(2)	46(1)
C(27)	4797(4)	3462(3)	6225(3)	57(2)

Table S21. Bond lengths [Å] and angles [°] for **4**.

Zn(1)-O(2)	2.046(3)
Zn(1)-O(1)	2.081(2)
Zn(1)-N(4)	2.116(3)
Zn(1)-N(1)	2.140(3)
Zn(1)-Cl(1)	2.2718(10)
O(1)-C(1)	1.257(4)
O(2)-C(13)	1.263(4)
N(1)-C(3)	1.283(5)
N(1)-C(25)	1.468(4)
N(2)-C(4)	1.339(5)
N(2)-N(3)	1.391(4)
N(2)-C(6)	1.457(4)
N(3)-C(1)	1.374(5)
N(3)-C(7)	1.430(5)
N(4)-C(15)	1.284(5)
N(4)-C(26)	1.450(5)
N(5)-C(16)	1.334(5)
N(5)-N(6)	1.389(4)
N(5)-C(18)	1.456(5)
N(6)-C(13)	1.366(5)
N(6)-C(19)	1.431(5)
C(1)-C(2)	1.426(5)
C(2)-C(4)	1.390(5)
C(2)-C(3)	1.432(5)
C(4)-C(5)	1.486(5)
C(7)-C(12)	1.388(6)
C(7)-C(8)	1.388(6)
C(8)-C(9)	1.394(6)
C(9)-C(10)	1.376(7)
C(10)-C(11)	1.380(6)
C(11)-C(12)	1.388(6)
C(13)-C(14)	1.430(5)
C(14)-C(16)	1.389(5)
C(14)-C(15)	1.420(5)

C(16)-C(17)	1.489(5)
C(19)-C(20)	1.381(5)
C(19)-C(24)	1.385(5)
C(20)-C(21)	1.378(6)
C(21)-C(22)	1.378(7)
C(22)-C(23)	1.380(7)
C(23)-C(24)	1.388(6)
C(25)-C(26)	1.520(5)
O(3)-C(27)	1.401(6)

O(2)-Zn(1)-O(1)	89.57(10)
O(2)-Zn(1)-N(4)	89.36(11)
O(1)-Zn(1)-N(4)	152.59(11)
O(2)-Zn(1)-N(1)	143.49(11)
O(1)-Zn(1)-N(1)	88.64(10)
N(4)-Zn(1)-N(1)	76.23(11)
O(2)-Zn(1)-Cl(1)	107.03(7)
O(1)-Zn(1)-Cl(1)	99.26(8)
N(4)-Zn(1)-Cl(1)	107.22(9)
N(1)-Zn(1)-Cl(1)	109.25(8)
C(1)-O(1)-Zn(1)	122.1(2)
C(13)-O(2)-Zn(1)	123.0(2)
C(3)-N(1)-C(25)	117.1(3)
C(3)-N(1)-Zn(1)	127.7(2)
C(25)-N(1)-Zn(1)	115.1(2)
C(4)-N(2)-N(3)	108.5(3)
C(4)-N(2)-C(6)	126.2(3)
N(3)-N(2)-C(6)	121.2(3)
C(1)-N(3)-N(2)	109.3(3)
C(1)-N(3)-C(7)	127.2(3)
N(2)-N(3)-C(7)	122.3(3)
C(15)-N(4)-C(26)	120.1(3)
C(15)-N(4)-Zn(1)	129.0(3)
C(26)-N(4)-Zn(1)	110.4(2)
C(16)-N(5)-N(6)	108.7(3)
C(16)-N(5)-C(18)	128.9(3)

N(6)-N(5)-C(18)	120.6(3)
C(13)-N(6)-N(5)	109.3(3)
C(13)-N(6)-C(19)	128.9(3)
N(5)-N(6)-C(19)	121.4(3)
O(1)-C(1)-N(3)	122.8(3)
O(1)-C(1)-C(2)	131.6(3)
N(3)-C(1)-C(2)	105.6(3)
C(4)-C(2)-C(1)	107.7(3)
C(4)-C(2)-C(3)	126.6(3)
C(1)-C(2)-C(3)	125.5(3)
N(1)-C(3)-C(2)	121.4(3)
N(2)-C(4)-C(2)	108.6(3)
N(2)-C(4)-C(5)	121.6(3)
C(2)-C(4)-C(5)	129.7(3)
C(12)-C(7)-C(8)	121.9(4)
C(12)-C(7)-N(3)	118.2(3)
C(8)-C(7)-N(3)	119.9(4)
C(7)-C(8)-C(9)	118.1(4)
C(10)-C(9)-C(8)	120.4(4)
C(9)-C(10)-C(11)	121.0(4)
C(10)-C(11)-C(12)	119.7(4)
C(7)-C(12)-C(11)	118.9(4)
O(2)-C(13)-N(6)	122.4(3)
O(2)-C(13)-C(14)	131.8(3)
N(6)-C(13)-C(14)	105.8(3)
C(16)-C(14)-C(15)	128.0(3)
C(16)-C(14)-C(13)	107.2(3)
C(15)-C(14)-C(13)	124.7(3)
N(4)-C(15)-C(14)	121.7(3)
N(5)-C(16)-C(14)	108.9(3)
N(5)-C(16)-C(17)	121.6(3)
C(14)-C(16)-C(17)	129.5(4)
C(20)-C(19)-C(24)	121.6(4)
C(20)-C(19)-N(6)	119.4(3)
C(24)-C(19)-N(6)	119.0(3)
C(21)-C(20)-C(19)	119.1(4)

C(22)-C(21)-C(20)	120.4(4)
C(21)-C(22)-C(23)	120.0(4)
C(22)-C(23)-C(24)	120.7(4)
C(19)-C(24)-C(23)	118.2(4)
N(1)-C(25)-C(26)	108.6(3)
N(4)-C(26)-C(25)	106.7(3)

Table S22. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for **4**. The anisotropic displacement factor exponent takes the form: $-2p^2[\ h^2\ a^{*2}U^{11} + ...\ + 2\ h\ k\ a^*\ b^*\ U^{12}\ ]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
Zn(1)	17(1)	11(1)	12(1)	2(1)	3(1)	2(1)
Cl(1)	26(1)	20(1)	30(1)	-1(1)	-1(1)	-6(1)
O(1)	23(1)	19(1)	16(1)	4(1)	5(1)	9(1)
O(2)	27(1)	11(1)	14(1)	0(1)	2(1)	4(1)
N(1)	18(1)	14(1)	14(2)	2(1)	1(1)	0(1)
N(2)	14(1)	19(2)	16(2)	-1(1)	2(1)	2(1)
N(3)	20(2)	19(2)	13(2)	1(1)	2(1)	6(1)
N(4)	15(1)	14(2)	16(2)	2(1)	2(1)	2(1)
N(5)	24(2)	9(1)	15(2)	-2(1)	-2(1)	1(1)
N(6)	24(2)	8(1)	14(2)	-1(1)	2(1)	2(1)
C(1)	17(2)	17(2)	12(2)	0(1)	-4(1)	1(1)
C(2)	14(2)	15(2)	14(2)	-3(1)	1(1)	-1(1)
C(3)	17(2)	16(2)	9(2)	-2(1)	1(1)	-4(1)
C(4)	17(2)	16(2)	15(2)	-3(1)	-1(1)	-2(1)
C(5)	26(2)	20(2)	24(2)	-1(2)	8(2)	5(2)
C(6)	18(2)	23(2)	26(2)	-5(2)	0(2)	8(2)
C(7)	14(2)	18(2)	24(2)	5(2)	2(2)	6(1)
C(8)	15(2)	21(2)	31(2)	-1(2)	-2(2)	5(2)
C(9)	15(2)	20(2)	50(3)	7(2)	2(2)	4(2)
C(10)	20(2)	32(2)	36(3)	18(2)	6(2)	7(2)
C(11)	22(2)	39(2)	20(2)	7(2)	2(2)	9(2)
C(12)	20(2)	25(2)	23(2)	0(2)	-1(2)	5(2)
C(13)	14(2)	14(2)	15(2)	0(1)	1(1)	-2(1)
C(14)	11(2)	11(2)	17(2)	2(1)	1(1)	-1(1)
C(15)	13(2)	13(2)	20(2)	4(2)	2(1)	-1(1)
C(16)	14(2)	12(2)	20(2)	-1(2)	-1(1)	-2(1)
C(17)	31(2)	10(2)	21(2)	-2(2)	-2(2)	2(2)
C(18)	42(2)	16(2)	16(2)	-4(2)	-2(2)	4(2)
C(19)	27(2)	11(2)	10(2)	-2(1)	-2(2)	0(2)
C(20)	28(2)	22(2)	20(2)	-3(2)	4(2)	-2(2)
C(21)	49(3)	25(2)	24(3)	0(2)	14(2)	-8(2)

C(22)	67(3)	14(2)	13(2)	0(2)	0(2)	-5(2)
C(23)	47(3)	14(2)	25(2)	-3(2)	-17(2)	4(2)
C(24)	28(2)	12(2)	24(2)	-4(2)	-5(2)	-2(2)
C(25)	24(2)	16(2)	14(2)	2(2)	2(2)	0(1)
C(26)	23(2)	17(2)	16(2)	5(2)	2(2)	6(2)
Cl(2)	37(1)	21(1)	19(1)	0(1)	1(1)	-8(1)
O(3)	44(2)	41(2)	55(3)	-6(2)	11(2)	-1(2)
C(27)	67(4)	38(3)	67(4)	-16(3)	25(3)	-9(3)

Table S23. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (Å^{2}x 10^3) for **4**.

	x	y	z	U(eq)
H(3)	4150	6503	1111	17
H(5A)	5088	5620	629	35
H(5B)	5403	4657	640	35
H(5C)	5911	5391	972	35
H(6A)	5939	3734	1359	34
H(6B)	5692	3293	1997	34
H(6C)	6248	4124	1998	34
H(8)	4354	2834	2239	27
H(9)	4285	1791	3017	34
H(10)	4487	2163	4040	35
H(11)	4809	3554	4311	32
H(12)	4898	4605	3544	27
H(15)	2313	8771	2798	18
H(17A)	2137	9699	3637	31
H(17B)	2612	9770	4277	31
H(17C)	1662	9618	4277	31
H(18A)	2108	8985	5230	37
H(18B)	2853	8374	5378	37
H(18C)	1954	8034	5465	37
H(20)	1291	6570	5003	28
H(21)	1298	5546	5782	39
H(22)	2506	4992	6138	37
H(23)	3713	5432	5701	35
H(24)	3724	6472	4924	25
H(25A)	3230	7573	1133	22
H(25B)	3685	7994	1710	22
H(26A)	2374	8458	1812	23
H(26B)	2019	7530	1657	23
H(3A)	4685	2402	5853	70
H(27A)	5342	3367	6381	86

H(27B)	4488	3785	6530	86
H(27C)	4822	3784	5838	86

Table S24. Torsion angles [°] for **4**.

C(4)-N(2)-N(3)-C(1)	4.5(4)
C(6)-N(2)-N(3)-C(1)	163.2(3)
C(4)-N(2)-N(3)-C(7)	172.5(3)
C(6)-N(2)-N(3)-C(7)	-28.8(5)
C(16)-N(5)-N(6)-C(13)	-2.9(4)
C(18)-N(5)-N(6)-C(13)	-169.0(3)
C(16)-N(5)-N(6)-C(19)	-176.1(3)
C(18)-N(5)-N(6)-C(19)	17.8(5)
Zn(1)-O(1)-C(1)-N(3)	-170.9(2)
Zn(1)-O(1)-C(1)-C(2)	7.9(5)
N(2)-N(3)-C(1)-O(1)	177.2(3)
C(7)-N(3)-C(1)-O(1)	10.0(6)
N(2)-N(3)-C(1)-C(2)	-1.8(4)
C(7)-N(3)-C(1)-C(2)	-169.1(3)
O(1)-C(1)-C(2)-C(4)	179.6(4)
N(3)-C(1)-C(2)-C(4)	-1.4(4)
O(1)-C(1)-C(2)-C(3)	5.1(6)
N(3)-C(1)-C(2)-C(3)	-176.0(3)
C(25)-N(1)-C(3)-C(2)	170.1(3)
Zn(1)-N(1)-C(3)-C(2)	-12.5(5)
C(4)-C(2)-C(3)-N(1)	-176.1(3)
C(1)-C(2)-C(3)-N(1)	-2.5(5)
N(3)-N(2)-C(4)-C(2)	-5.4(4)
C(6)-N(2)-C(4)-C(2)	-162.6(3)
N(3)-N(2)-C(4)-C(5)	171.6(3)
C(6)-N(2)-C(4)-C(5)	14.3(5)
C(1)-C(2)-C(4)-N(2)	4.2(4)
C(3)-C(2)-C(4)-N(2)	178.8(3)
C(1)-C(2)-C(4)-C(5)	-172.4(4)
C(3)-C(2)-C(4)-C(5)	2.1(6)
C(1)-N(3)-C(7)-C(12)	-62.2(5)
N(2)-N(3)-C(7)-C(12)	132.1(4)
C(1)-N(3)-C(7)-C(8)	116.1(4)
N(2)-N(3)-C(7)-C(8)	-49.6(5)

C(12)-C(7)-C(8)-C(9)	0.2(5)
N(3)-C(7)-C(8)-C(9)	-178.1(3)
C(7)-C(8)-C(9)-C(10)	0.7(5)
C(8)-C(9)-C(10)-C(11)	-1.3(6)
C(9)-C(10)-C(11)-C(12)	0.9(6)
C(8)-C(7)-C(12)-C(11)	-0.6(5)
N(3)-C(7)-C(12)-C(11)	177.7(3)
C(10)-C(11)-C(12)-C(7)	0.1(5)
Zn(1)-O(2)-C(13)-N(6)	175.6(2)
Zn(1)-O(2)-C(13)-C(14)	-3.7(5)
N(5)-N(6)-C(13)-O(2)	-178.8(3)
C(19)-N(6)-C(13)-O(2)	-6.3(6)
N(5)-N(6)-C(13)-C(14)	0.6(4)
C(19)-N(6)-C(13)-C(14)	173.1(3)
O(2)-C(13)-C(14)-C(16)	-178.9(3)
N(6)-C(13)-C(14)-C(16)	1.7(4)
O(2)-C(13)-C(14)-C(15)	5.8(6)
N(6)-C(13)-C(14)-C(15)	-173.6(3)
C(26)-N(4)-C(15)-C(14)	-178.7(3)
Zn(1)-N(4)-C(15)-C(14)	-7.7(5)
C(16)-C(14)-C(15)-N(4)	-173.6(3)
C(13)-C(14)-C(15)-N(4)	0.7(5)
N(6)-N(5)-C(16)-C(14)	4.0(4)
C(18)-N(5)-C(16)-C(14)	168.6(3)
N(6)-N(5)-C(16)-C(17)	-175.0(3)
C(18)-N(5)-C(16)-C(17)	-10.4(6)
C(15)-C(14)-C(16)-N(5)	171.6(3)
C(13)-C(14)-C(16)-N(5)	-3.6(4)
C(15)-C(14)-C(16)-C(17)	-9.5(6)
C(13)-C(14)-C(16)-C(17)	175.3(3)
C(13)-N(6)-C(19)-C(20)	-102.3(4)
N(5)-N(6)-C(19)-C(20)	69.4(4)
C(13)-N(6)-C(19)-C(24)	78.8(5)
N(5)-N(6)-C(19)-C(24)	-109.4(4)
C(24)-C(19)-C(20)-C(21)	0.9(6)
N(6)-C(19)-C(20)-C(21)	-177.9(3)

C(19)-C(20)-C(21)-C(22)	-0.2(6)
C(20)-C(21)-C(22)-C(23)	-0.8(6)
C(21)-C(22)-C(23)-C(24)	1.2(6)
C(20)-C(19)-C(24)-C(23)	-0.5(6)
N(6)-C(19)-C(24)-C(23)	178.3(3)
C(22)-C(23)-C(24)-C(19)	-0.5(6)
C(3)-N(1)-C(25)-C(26)	159.5(3)
Zn(1)-N(1)-C(25)-C(26)	-18.2(4)
C(15)-N(4)-C(26)-C(25)	120.1(3)
Zn(1)-N(4)-C(26)-C(25)	-52.4(3)
N(1)-C(25)-C(26)-N(4)	44.8(4)

Table S25. Hydrogen bonds for **4** [Å and °].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
O(3)-H(3A)...Cl(2)	0.84	2.28	3.114(4)	169.6
C(3)-H(3)...Cl(2)#1	0.95	2.80	3.737(4)	169.9
C(5)-H(5A)...Cl(2)#1	0.98	2.77	3.747(4)	173.7
C(5)-H(5B)...Cl(2)#2	0.98	2.65	3.622(4)	173.1
C(6)-H(6A)...Cl(2)#2	0.98	2.89	3.786(4)	151.6
C(6)-H(6C)...Cl(1)#3	0.98	2.76	3.622(4)	147.5
C(8)-H(8)...O(3)#2	0.95	2.56	3.490(6)	167.4
C(17)-H(17A)...O(1)#4	0.98	2.59	3.475(5)	149.6
C(18)-H(18B)...Cl(2)#5	0.98	2.91	3.678(4)	136.3
C(24)-H(24)...Cl(2)#5	0.95	2.88	3.580(4)	131.4
C(27)-H(27B)...Cl(1)#6	0.98	2.95	3.905(6)	165.7

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y+1/2,-z+1/2 #2 x,-y+1/2,z-1/2 #3 x+1/2,y,-z+1/2

#4 -x+1/2,y+1/2,z #5 -x+1,-y+1,-z+1 #6 -x+1/2,-y+1,z+1/2

References

(1) Spek, Anthony L. PLATON SQUEEZE: a tool for the calculation of the disordered solvent contribution to the calculated structure factors. *Acta crystallographica* **2015**, Section C, 71, Pt 1, 9-18.

(2) Joseph, S.; Radhakrishnan, P. K. "Yttrium and Lanthanide Iodide Complexes of N,N′-Bis(4-Antipyrylmethylidene)Ethylenediamine," *Polyhedron* **1999**, 18 (13), 1881–1884.

(3) Raju, K. C.; Radhakrishnan, P. K. "Complexes of Iron(III) with N,N′-bis(4-Antipyrylmethylidene)Ethylenediamine," *Synthesis and Reactivity in Inorganic and Metal-Organic Chemistry* **2003**, 33 (1), 23–33. https://doi.org/10.1081/SIM-120016870.

(4) Hoffert, W. A.; Roberts, J. A. S.; Bullock, R. M.; Helm, M. L. "Production of H2 at Fast Rates Using a Nickel Electrocatalyst in Water–Acetonitrile Solutions," *Chem. Commun.* **2013**, 49 (71), 7767–7769. https://doi.org/10.1039/C3CC43203C. Felton, G. A. N.; Glass, R. S.; Lichtenberger, D. L.; Evans, D. H. "Iron-Only Hydrogenase Mimics. Thermodynamic Aspects of the Use of Electrochemistry to Evaluate Catalytic Efficiency for Hydrogen Generation," *Inorg. Chem.* **2006**, 45 (23), 9181–9184. https://doi.org/10.1021/ic060984e.

(5) Fourmond, V.; Jacques, P.-A.; Fontecave, M.; Artero, V. "H_2 Evolution and Molecular Electrocatalysts: Determination of Overpotentials and Effect of Homoconjugation," *Inorg. Chem.* **2010**, 49 (22), 10338–10347. https://doi.org/10.1021/ic101187v.

(6) Wiese, S.; Kilgore, U. J.; DuBois, D. L.; Bullock, R. M. "[Ni(PMe2NPh2)2](BF4)2 as an Electrocatalyst for H2 Production," *ACS Catal.* **2012**, 2 (5), 720–727. https://doi.org/10.1021/cs300019h.

(7) Nicholson, R. S.; Shain, Irving. "Theory of Stationary Electrode Polarography. Single Scan and Cyclic Methods Applied to Reversible, Irreversible, and Kinetic Systems," *Anal. Chem.* **1964**, 36 (4), 706–723. https://doi.org/10.1021/ac60210a007.

(8) Bard, A. J.; Faulkner, L. R. "Electrochemical Methods: Fundamentals and Applications," 2nd ed.; John Wiley & Sons: New York, 2001.

(9) Saveant, J. M.; Vianello, E. "Potential-Sweep Chronoamperometry: Kinetic Currents for First-Order Chemical Reaction Parallel to Electron-Transfer Process (Catalytic Currents)," *Electrochimica Acta* **1965**, 10 (9), 905–920. https://doi.org/10.1016/0013-4686(65)80003-2.

(10) Savéant, J. M.; Vianello, E. "Potential-Sweep Voltammetry: General Theory of Chemical Polarization," *Electrochimica Acta* **1967**, 12 (6), 629–646. https://doi.org/10.1016/0013-4686(67)85031-X.

(11) Race, N. A.; Zhang, W.; Screen, M. E.; Barden, B. A.; McNamara, W. R. "Iron Polypyridyl Catalysts Assembled on Metal Oxide Semiconductors for Photocatalytic Hydrogen Generation," *Chem. Commun.* **2018**, 54 (26), 3290–3293. https://doi.org/10.1039/C8CC00453F.

(12) Wise, C. F.; Liu, D.; Mayer, K. J.; Crossland, P. M.; Hartley, C. L.; McNamara, W. R. "A Nickel Complex of a Conjugated Bis-Dithiocarbazate Schiff Base for the Photocatalytic Production of Hydrogen," *Dalton Trans.* **2015**, *44* (32), 14265–14271. https://doi.org/10.1039/C5DT02042E.

(13) Han, Z.; Qiu, F.; Eisenberg, R.; Holland, P. L.; Krauss, T. D. "Robust Photogeneration of H2 in Water Using Semiconductor Nanocrystals and a Nickel Catalyst," *Science* **2012**, 338 (6112), 1321–1324. https://doi.org/10.1126/science.1227775.