Supplementary Information for "*Fe[4-(3-Phenylpropyl)pyridine]$_2$[Fe(CN)$_5$NO]: A 2D coordination polymer with thermally-induced spin transition and nature of its asymmetric hysteresis loop*", by K. Scanda, Y. Avila, R. Mojica, A. Cano, M. Gonzalez, J. Rodríguez-Hernández, and E. Reguera

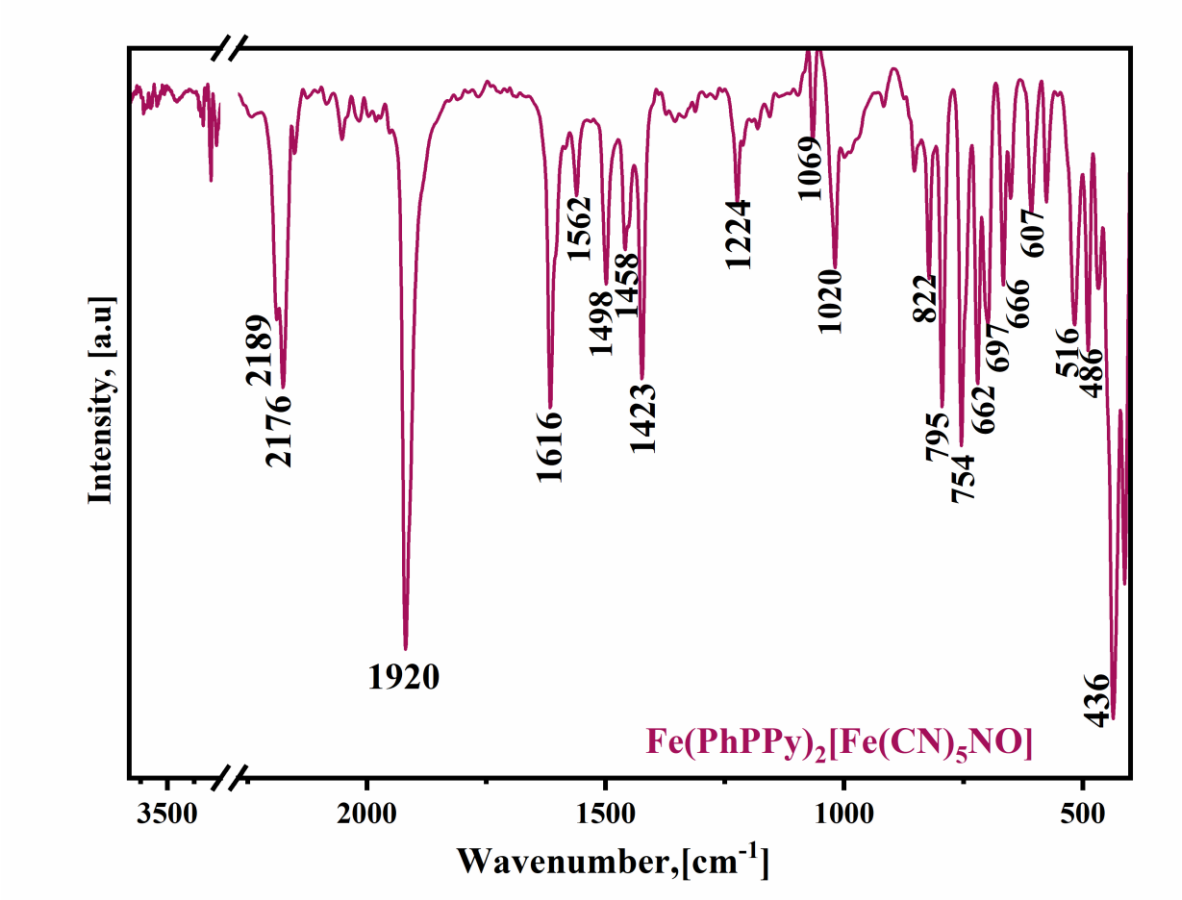

Figure S1: Infrared spectra registered at 80 K for Fe[4-(3-Phenylpropyl)pyridine]$_2$[Fe(CN)$_5$NO].

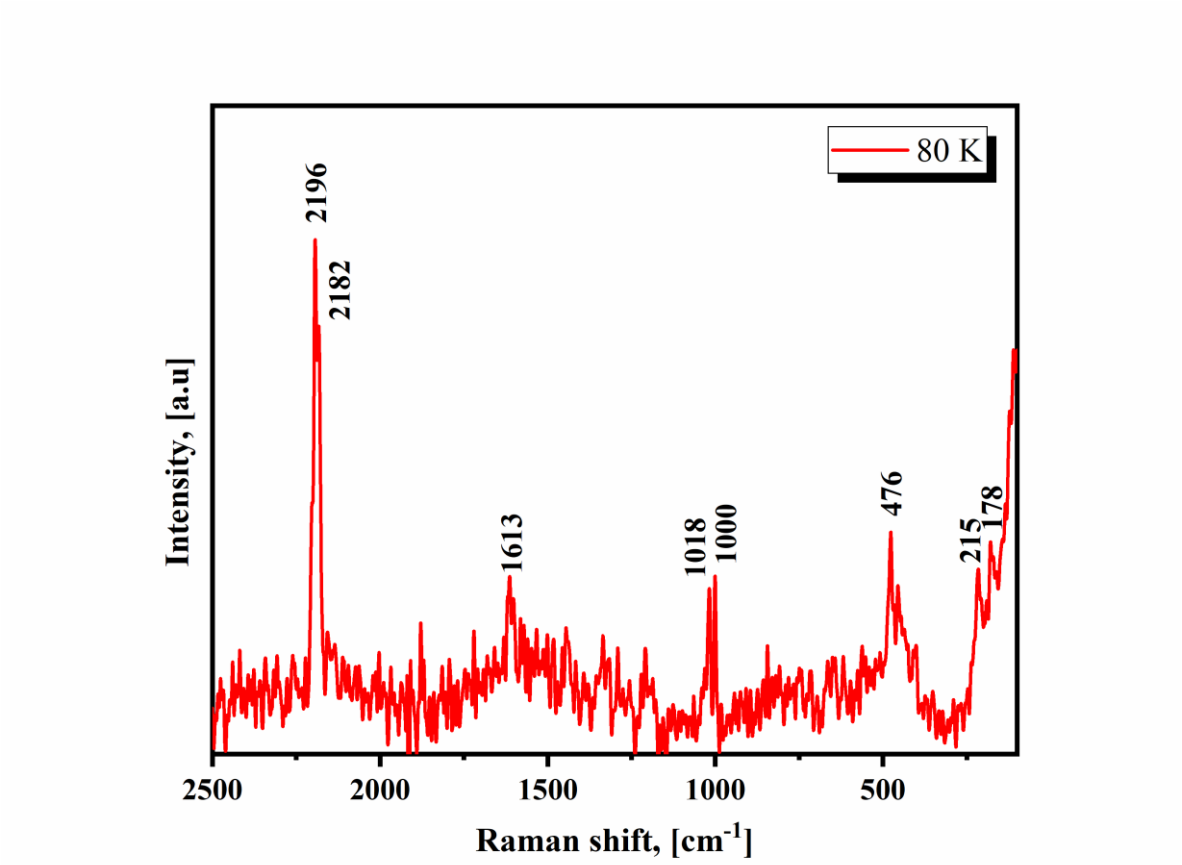

Figure S2: Raman spectra registered at 80 K for Fe[4-(3-Phenylpropyl)pyridine]$_2$[Fe(CN)$_5$NO]

Table S1: Details of data collection, crystal data and structure refinement for Fe[4-(3-Phenylpropyl)pyridine]$_2$[Fe(CN)$_5$NO] material.

	Fe(PhPPy)$_2$[Fe(CN)$_5$NO]	**Fe(PhPPy)$_2$[Fe(CN)$_5$NO]-LS phase calculated**
Data collection		
Diffractometer	D8 Advance (from Bruker)	
Detector	lynx eye	
Wavelength (Å)	CuK$_a$: 1.54183	
2θ range (°)	5.0-60.0	
Step size (°)	0.010591	
Time per step (s)	10	
Unit cell		
Space Group	$P2_12_12$	$P2_12_12$
Cell Parameter (Å)	a= 14.633(2)	a= 14.603(3)
	b= 14.626(5)	b= 14.612(6)
	c= 15.789(5)	c= 15.789(2)

V(Å^3)	3379.4(2)	3368.8(4)
Z	4	4
Refinement		
# of reflections	597	
# *of refined parameters*		
Structural	176	
Profile	46	
R_{exp}	2.81	
R_{wp}	9.93	
R_B	6.64	
S	3.53	

Table S2. Refined atomic positions and thermal (B_{iso}) and occupation (Occ) factors for the materials under study.

Composition	site	x	y	z	Biso	Occ
$Fe(PhPPy)_2[Fe(CN)_5NO]$						
Fei	4c	0.5036(2)	0.2464(3)	0.4706(2)	2.04(2)	1
Fee	4c	0.2542(2)	-0.0018(3)	0.5103(2)	2.15(2)	1
C1	4c	0.4118(5)	0.1532(5)	0.4628(5)	3.3(3)	1
N1	4c	0.3618(4)	0.0943(5)	0.4621(4)	3.3(3)	1
C2	4c	0.5943(3)	0.1506(3)	0.4749(3)	3.3(3)	1
N2	4c	0.6475(6)	0.0949(5)	0.4808(3)	3.3(3)	1
C3	4c	0.5930(4)	0.3432(5)	0.4794(5)	3.3(3)	1
N3	4c	0.6432(3)	0.4014(3)	0.4870(4)	3.3(3)	1
C4	4c	0.4105(5)	0.3388(3)	0.4672(4)	3.3(3)	1
N4	4c	0.3600(3)	0.3970(5)	0.4613(3)	3.3(3)	1
C5	4c	0.5066(5)	0.2498(4)	0.3509(2)	3.3(3)	1
N5	4c	0.5084(3)	0.2517(6)	0.2795(5)	3.3(3)	1
N	4c	0.4988(6)	0.2436(7)	0.5722(6)	3.3(3)	1
O	4c	0.4995(3)	0.2416(4)	0.6438(6)	3.3(3)	1
N100	4c	0.2104(5)	0.0010(3)	0.3493(4)	3.8(3)	1
C101	4c	0.1915(5)	0.0894(6)	0.3319(5)	3.8(3)	1
C102	4c	0.1554(4)	0.1196(5)	0.2557(3)	3.8(3)	1
C103	4c	0.1380(3)	0.0546(4)	0.1943(5)	3.8(3)	1
C104	4c	0.1560(6)	-0.0367(7)	0.2089(5)	3.8(3)	1
C105	4c	0.1921(5)	-0.0599(7)	0.2872(6)	3.8(3)	1
C106	4c	0.0987(6)	0.0837(5)	0.1102(6)	3.8(3)	1
C107	4c	0.1572(6)	0.1518(5)	0.0596(4)	3.8(3)	1
C108	4c	0.1193(4)	0.2502(7)	0.0578(5)	3.8(3)	1
C109	4c	0.0893(5)	0.2846(3)	-0.0267(4)	3.8(3)	1
C110	4c	0.0913(3)	0.3779(4)	-0.0440(5)	3.8(3)	1
C111	4c	0.0633(5)	0.4101(4)	-0.1231(3)	3.8(3)	1
C112	4c	0.0332(5)	0.3489(4)	-0.1848(5)	3.8(3)	1
C113	4c	0.0312(5)	0.2555(5)	-0.1675(3)	3.8(3)	1
C114	4c	0.0593(7)	0.2234(5)	-0.0885(5)	3.8(3)	1
N200	4c	0.3483(4)	0.1089(3)	0.6260(5)	3.8(3)	1
C201	4c	0.3934(6)	0.1468(5)	0.6920(4)	3.8(3)	1
C202	4c	0.3572(4)	0.1553(6)	0.7728(3)	3.8(3)	1
C203	4c	0.2695(6)	0.1228(4)	0.7857(4)	3.8(3)	1
C204	4c	0.2204(3)	0.0835(4)	0.7203(5)	3.8(3)	1
C205	4c	0.2627(3)	0.0782(3)	0.6419(6)	3.8(3)	1
C206	4c	0.2267(5)	0.1304(5)	0.8723(3)	3.8(3)	1
C207	4c	0.2861(3)	0.1771(6)	0.9400(5)	3.8(3)	1
C208	4c	0.2329(5)	0.2403(4)	1.0009(6)	3.8(3)	1
C209	4c	0.2892(4)	0.2899(3)	1.0650(6)	3.8(3)	1
C210	4c	0.3664(7)	0.3387(5)	1.0396(4)	3.8(3)	1
C211	4c	0.4190(4)	0.3851(4)	1.0995(3)	3.8(3)	1
C212	4c	0.3943(5)	0.3827(4)	1.1848(3)	3.8(3)	1
C213	4c	0.3171(7)	0.3339(3)	1.2103(5)	3.8(3)	1
C214	4c	0.2645(5)	0.2875(5)	1.1504(5)	3.8(3)	1

Table S3. Calculated inter-atomic distances (in Å) and bond angles (in °) for the material under study.

Bond distance (Å)	Angles (°)	
$Fe(PhPPy)_2[Fe(CN)_5NO]$		
Fee-N1 = 2.244(1)	N1-Fee-N2 = 81.11(2)	Fee-N100-C101 = 105.20(4)
Fee-N2 = 2.035(1)	N1-Fee-N3 = 96.14(2)	N100-C101-C102 = 124.03(4)
Fee-N3 = 2.190(1)	N1-Fee-N4 = 171.53(2)	C101-C102-C103 = 117.39(7)
Fee-N4 = 2.277(1)	N1-Fee-N100 = 80.38(3)	C102-C103-C104 = 120.88(3)
Fee-N100 = 2.622(2)	N1-Fee-N200 = 60.95(2)	C103-C104-C105 = 117.28(4)
Fee-N200 = 2.803(1)	N2-Fee-N3 = 167.81(2)	C104-C105-N100 = 124.13(7)
Fei-C1 = 1.918(1)	N2-Fee-N4 = 97.41(2)	C105-N100-Fee = 137.86(5)
Fei-C2 = 1.931(1)	N2-Fee-N100 = 87.78(3)	C105-N100-C101 = 116.28(3)
Fei-C3 = 1.933(1)	N2-Fee-N200 = 100.86(2)	C102-C103-C106 = 119.60(6)
Fei-C4 = 1.920(1)	N3-Fee-N4 = 83.55(2)	C104-C103-C106 = 119.52(4)
Fei-C5 = 1.891(1)	N3-Fee-N100 = 80.04(3)	C103-C106-C107 = 115.34(4)
Fei-N = 1.606(1)	N3-Fee-N200 = 87.96(2)	C106-C107-C108 = 114.45(3)
C1-N1 = 1.130(1)	N4-Fee-N100 = 91.24(3)	C107-C108-C109 = 115.90(3)
C2-N2 = 1.131(1)	N4-Fee-N200 = 127.43(2)	C108-C109-C110 = 119.99(4)
C3-N3 = 1.131(1)	N100-Fee-N200 = 137.99(3)	C109-C110-C111 = 120.00(4)
C4-N4 = 1.1311(1)	N-Fei-C1 = 90.88(4)	C110-C111-C112 = 120.07(7)
C5-N5 = 1.128(1)	N-Fei-C2 = 88.65(4)	C111-C112-C113 = 119.93(3)
N-O = 1.131(1)	N-Fei-C3 = 88.65(4)	C112-C113-C114 = 119.94(4)
N100-C101 = 1.350(1)	N-Fei-C4 = 90.85(4)	C113-C114-C109 = 120.16(7)
C101-C102 = 1.386(1)	N-Fei-C5 = 178.82(8)	C114-C109-C108 = 120.12(7)
C102-C103 = 1.381(1)	C5-Fei-C1 = 88.32(4)	C114-C109-C110 = 119.89(3)
C103-C106 = 1.508(1)	C5-Fei-C2 = 92.19(4)	Fee-N200-C201 = 168.20(4)
C103-C104 = 1.380(1)	C5-Fei-C3 = 92.11(4)	N200-C201-C202 = 124.00(7)
C104-C105 = 1.387(1)	C5-Fei-C4 = 88.28(4)	C201-C202-C203 = 117.31(5)
C105-N100 = 1.351(1)	C1-Fei-C2 = 88.16(2)	C202-C203-C204 = 120.98(4)
C106-C107 = 1.537(1)	C1-Fei-C3 = 178.11(3)	C203-C204-C205 = 117.26(7)
C107-C108 = 1.543(1)	C1-Fei-C4 = 90.08(2)	C204-C205-N200 = 124.06(5)
C108-C109 = 1.492(1)	C3-Fei-C2 = 93.66(2)	C205-N200-Fee = 58.32(3)
C109-C110 = 1.392(1)	C3-Fei-C4 = 88.09(2)	C205-N200-C201 = 116.39(4)
C110-C111 = 1.396(1)	C2-Fei-C4 = 178.17(3)	C202-C203-C206 = 119.56(5)
C111-C112 = 1.394(1)	N1-C1-Fei = 174.74(4)	C204-C203-C206 = 119.46(7)
C112-C113 = 1.393(1)	N2-C2-Fei = 177.28(4)	C203-C206-C207 = 115.36(6)
C113-C114 = 1.395(1)	N3-C3-Fei = 177.24(4)	C206-C207-C208 = 114.44(4)
C114-C109 = 1.395(1)	N4-C4-Fei = 174.72(4)	C207-C208-C209 = 115.77(4)
N200-C201 = 1.352(1)	N5-C5-Fei = 179.90(10)	C208-C209-C210 = 120.03(4)
C201-C202 = 1.387(1)	O-N-Fei = 176.97(10)	C209-C210-C211 = 120.04(4)
C202-C203 = 1.384(1)	C1-N1-Fee = 158.19(4)	C210-C211-C212 = 119.95(7)
C203-C206 = 1.508(1)	C2-N2-Fee = 171.03(4)	C211-C212-C213 = 120.08(5)
C203-C204 = 1.383(1)	C3-N3-Fee = 171.35(4)	C212-C213-C214 = 119.97(4)
C204-C205 = 1.386(1)	C4-N4-Fee = 162.27(3)	C213-C214-C209 = 119.96(7)
C205-N200 = 1.354(1)		C214-C209-C208 = 119.97(7)
C206-C207 = 1.538(1)		C214-C209-C210 = 120.00(5)
C207-C208 = 1.544(1)		
C208-C209 = 1.493(1)		
C209-C210 = 1.395(1)		
C210-C211 = 1.395(1)		
C211-C212 = 1.395(1)		
C212-C213 = 1.396(1)		
C213-C214 = 1.395(1)		
C214-C209 = 1.396(1)		

Table S4: Calculated atomic positions and thermal (B_{iso}) and occupation (Occ) factors for the structure of low temperature for the material under study.

Composition	site	x	y	z	Occ
$Fe(PhPPy)_2[Fe(CN)_5NO]$-LS phase calculated at 0K					
Fei	4c	0.50130	0.24530	0.48320	1
Fee	4c	0.26190	0.00820	0.50070	1
C1	4c	0.41290	0.14860	0.46260	1
N1	4c	0.35670	0.09260	0.46240	1
C2	4c	0.59370	0.15310	0.47150	1
N2	4c	0.64960	0.09730	0.47910	1
C3	4c	0.59360	0.33820	0.47940	1
N3	4c	0.64490	0.39860	0.48770	1
C4	4c	0.41140	0.34300	0.46400	1
N4	4c	0.35280	0.39840	0.46330	1
C5	4c	0.50570	0.25050	0.37110	1
N5	4c	0.48610	0.25560	0.30050	1
N	4c	0.50120	0.24650	0.58860	1
O	4c	0.51910	0.27870	0.65410	1
N100	4c	0.20890	0.00440	0.34920	1
C101	4c	0.19040	0.09270	0.32990	1
C102	4c	0.15300	0.12030	0.25410	1
C103	4c	0.12970	0.05160	0.19570	1
C104	4c	0.15330	-0.03650	0.21250	1
C105	4c	0.19470	-0.05910	0.28850	1
C106	4c	0.08500	0.07710	0.11460	1
C107	4c	0.14300	0.14840	0.07100	1
C108	4c	0.10080	0.24740	0.07230	1
C109	4c	0.07820	0.28760	-0.01350	1
C110	4c	0.08790	0.38000	-0.03720	1
C111	4c	0.06540	0.41110	-0.11930	1
C112	4c	0.03690	0.34780	-0.17930	1
C113	4c	0.03330	0.25580	-0.15800	1
C114	4c	0.05550	0.22790	-0.07790	1
N200	4c	0.35260	0.09680	0.62950	1
C201	4c	0.39580	0.13800	0.69430	1
C202	4c	0.35710	0.15720	0.77070	1
C203	4c	0.26950	0.12520	0.78750	1
C204	4c	0.22210	0.08400	0.72160	1
C205	4c	0.26510	0.07460	0.64570	1
C206	4c	0.23250	0.13360	0.87270	1
C207	4c	0.30150	0.17610	0.93740	1
C208	4c	0.25650	0.25150	0.98660	1
C209	4c	0.30150	0.29540	1.06200	1
C210	4c	0.37590	0.34890	1.04090	1
C211	4c	0.42160	0.39850	1.10340	1
C212	4c	0.39440	0.38960	1.18750	1
C213	4c	0.32080	0.33420	1.21130	1
C214	4c	0.27500	0.28760	1.14720	1

Table S5: Calculated inter-atomic distances (in Å) and bond angles (in °) for the structure of low temperature for the material under study.

Bond distance (Å)	Angles (°)	
$Fe(PhPPy)_2[Fe(CN)_5NO]$-LS phase calculated at 0K		
Fee-N1 = 1.950	N1-Fee-N2 = 88.64	Fee-N100-C101 = 104.75
Fee-N2 = 2.040	N1-Fee-N3 = 100.74	N100-C101-C102 = 123.69
Fee-N3 = 2.192	N1-Fee-N4 = 175.27	C101-C102-C103 = 117.24
Fee-N4 = 2.388	N1-Fee-N100 = 86.41	C102-C103-C104 = 119.17
Fee-N100 = 2.515	N1-Fee-N200 = 65.79	C103-C104-C105 = 120.41
Fee-N200 = 2.751	N2-Fee-N3 = 167.64	C104-C105-N100 = 121.25
Fei-C1 = 1.941	N2-Fee-N4 = 88.65	C105-N100-Fee = 137.54
Fei-C2 = 1.916	N2-Fee-N100 = 91.10	C105-N100-C101 = 117.70
Fei-C3 = 1.914	N2-Fee-N200 = 100.03	C102-C103-C106 = 119.55
Fei-C4 = 1.963	N3-Fee-N4 = 81.42	C104-C103-C106 = 121.17
Fei-C5 = 1.773	N3-Fee-N100 = 81.57	C103-C106-C107 = 108.66
Fei-N = 1.664	N3-Fee-N200 = 91.21	C106-C107-C108 = 114.15
C1-N1 = 1.159	N4-Fee-N100 = 89.75	C107-C108-C109 = 115.52
C2-N2 = 1.160	N4-Fee-N200 = 118.55	C108-C109-C110 = 126.03
C3-N3 = 1.165	N100-Fee-N200 = 149.53	C109-C110-C111 = 121.94
C4-N4 = 1.178	N-Fei-C1 = 100.06	C110-C111-C112 = 118.81
C5-N5 = 1.153	N-Fei-C2 = 95.99	C111-C112-C113 = 119.46
N-O = 1.166	N-Fei-C3 = 91.40	C112-C113-C114 = 120.28
N100-C101 = 1.353	N-Fei-C4 = 98.41	C113-C114-C109 = 123.36
C101-C102 = 1.376	N-Fei-C5 = 176.33	C114-C109-C108 = 117.82
C102-C103 = 1.405	C5-Fei-C1 = 83.57	C114-C109-C110 = 115.74
C103-C106 = 1.485	C5-Fei-C2 = 84.74	Fee-N200-C201 = 178.06
C103-C104 = 1.359	C5-Fei-C3 = 84.99	N200-C201-C202 = 124.88
C104-C105 = 1.384	C5-Fei-C4 = 80.73	C201-C202-C203 = 118.86
C105-N100 = 1.350	C1-Fei-C2 = 86.58	C202-C203-C204 = 117.57
C106-C107 = 1.509	C1-Fei-C3 = 168.31	C203-C204-C205 = 118.32
C107-C108 = 1.572	C1-Fei-C4 = 93.36	C204-C205-N200 = 125.65
C108-C109 = 1.513	C3-Fei-C2 = 89.98	C205-N200-Fee = 64.47
C109-C110 = 1.408	C3-Fei-C4 = 87.16	C205-N200-C201 = 114.19
C110-C111 = 1.412	C2-Fei-C4 = 165.38	C202-C203-C206 = 119.39
C111-C112 = 1.388	N1-C1-Fei = 170.15	C204-C203-C206 = 123.02
C112-C113 = 1.387	N2-C2-Fei = 168.53	C203-C206-C207 = 113.47
C113-C114 = 1.368	N3-C3-Fei = 170.61	C206-C207-C208 = 110.36
C114-C109 = 1.380	N4-C4-Fei = 170.76	C207-C208-C209 = 122.14
N200-C201 = 1.344	N5-C5-Fei = 163.50	C208-C209-C210 = 113.25
C201-C202 = 1.361	O-N-Fei = 153.01	C209-C210-C211 = 120.04
C202-C203 = 1.388	C1-N1-Fee = 161.50	C210-C211-C212 = 119.34
C203-C206 = 1.455	C2-N2-Fee = 173.78	C211-C212-C213 = 122.02
C203-C204 = 1.387	C3-N3-Fee = 168.81	C212-C213-C214 = 117.24
C204-C205 = 1.360	C4-N4-Fee = 165.69	C213-C214-C209 = 121.65
C205-N200 = 1.343		C214-C209-C208 = 127.11
C206-C207 = 1.563		C214-C209-C210 = 119.63
C207-C208 = 1.500		
C208-C209 = 1.503		
C209-C210 = 1.379		
C210-C211 = 1.394		
C211-C212 = 1.392		
C212-C213 = 1.397		
C213-C214 = 1.391		
C214-C209 = 1.404		

Table S6. Binding Energies, eV, resulted from the curve fitting of Fe 2p and N 1s spectra recorded at 270 and 114 K.

Fe 2p	**270 K**			**114K**		
	BE, eV	**FWHM, eV**	**At. %**	**BE, eV**	**FWHM, eV**	**At. %**
Fe II-LS	---	---	---	708.3	2.0	15.2
$\Delta 2p_{3/2-1/2}$	---	---	---	13.2	2.2	---
Fe 2+-HS	710.2	2.7	54	708.7	2.0	13.6
$\Delta 2p_{3/2-1/2}$	13.2	2.7	---	13.2	2.1	---
Fe II-NP	711.3	2.8	46	710.6	2.1	71.2
$\Delta 2p_{3/2-1/2}$	13.6	2.7	---	13.2	2.4	---
Satellite	714.4	2.8	---	714.4	2.8	---
$\Delta 2p_{3/2-1/2}$	13.2	2.8	---	13.6	2.8	---

N 1s	270 K			114K		
	BE, eV	FWHM, eV	At. %	BE, eV	FWHM, eV	At. %
CN $L_{inked-HS}$	397.9	1.5	54	397.9	1.5	31.4
CN $_{Unlinked-HS}$	398.5	1.5	13	396.6	1.5	28.3
N $_{Py-HS}$	398.6	1.3	25	398.6	1.5	12.6
N $_{Py-LS}$	---	---	---	400.2	1.5	12.6
NO $_{LS}$	---	---	---	402.2	1.7	3.6
NO $_{HS}$	402.9	1.7	8	402.9	1.7	3.6