Electronic Supplementary Information (ESI) for the article entitled:

"New coordination polymers based on 2-methylimidazole and transition metal nitroprussides containing building blocks: Synthesis, structure and magnetic properties"

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Figure S1: Rietveld refinement of powder XRD pattern for $Mn(2\text{-}MeIm)_2[Fe(CN)_5NO]$. Red points: experimental data. Black line: calculated profile. Blue vertical lines: allowed Bragg positions. Blue line: difference between experimental and calculated profiles.

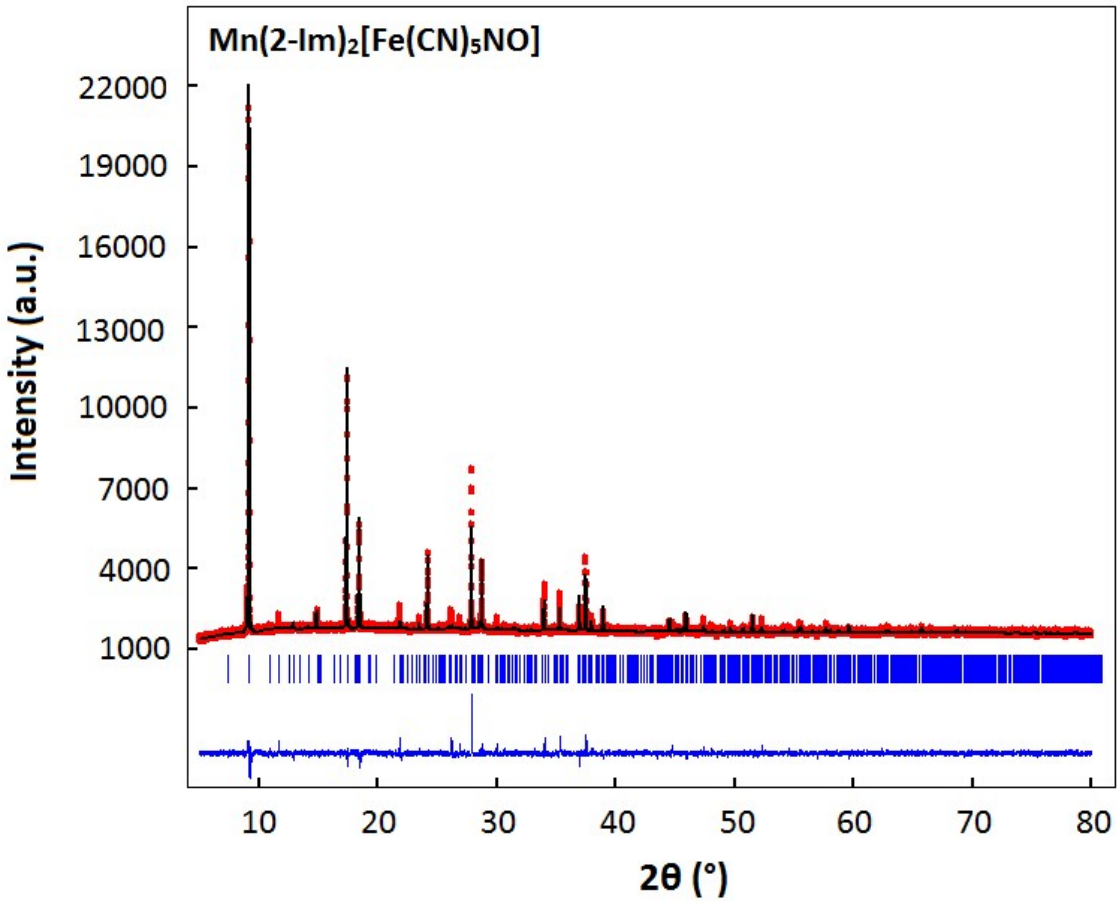

Figure S2: Rietveld refinement of powder XRD pattern for $Fe(2\text{-}MeIm)_2[Fe(CN)_5NO]$. Red points: experimental data. Black line: calculated profile. Blue vertical lines: allowed Bragg positions. Blue line: difference between experimental and calculated profiles.

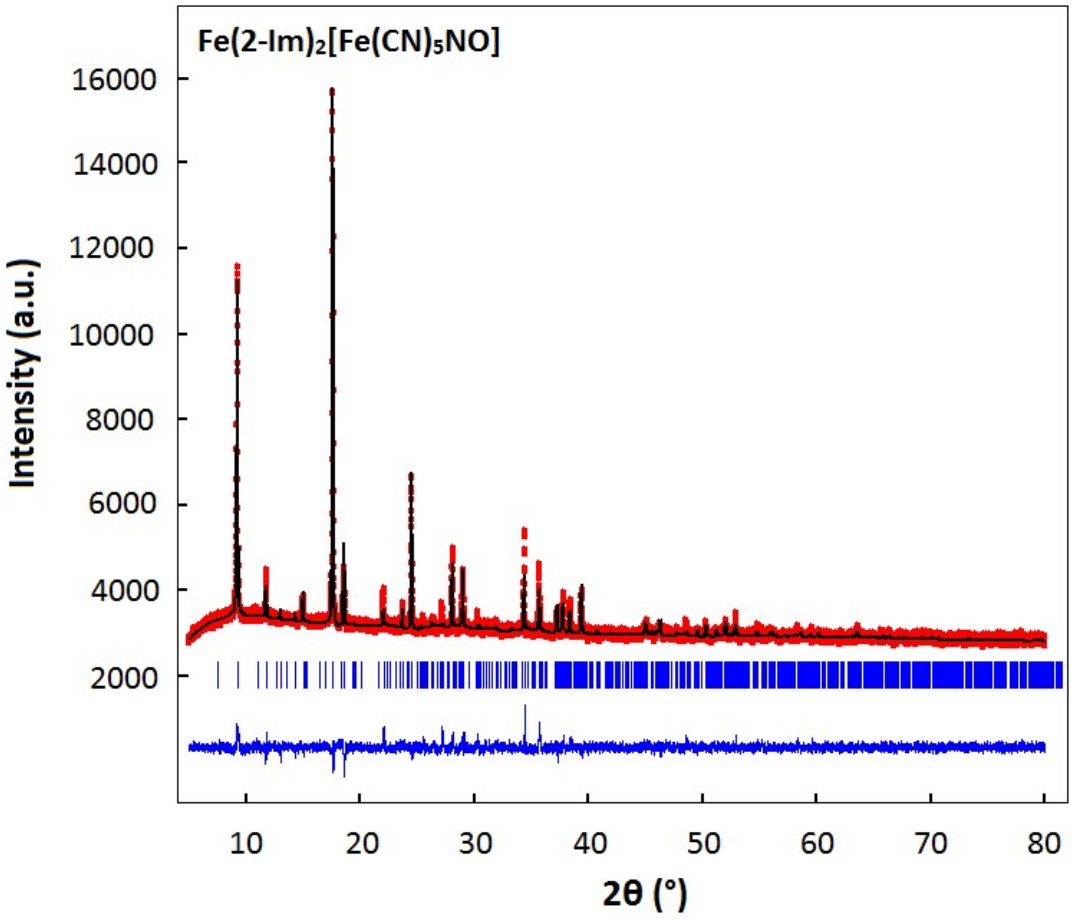

Figure S3: Rietveld refinement of powder XRD pattern for $Co(2\text{-}MeIm)_2[Fe(CN)_5NO]$. Red points: experimental data. Black line: calculated profile. Blue vertical lines: allowed Bragg positions. Blue line: difference between experimental and calculated profiles.

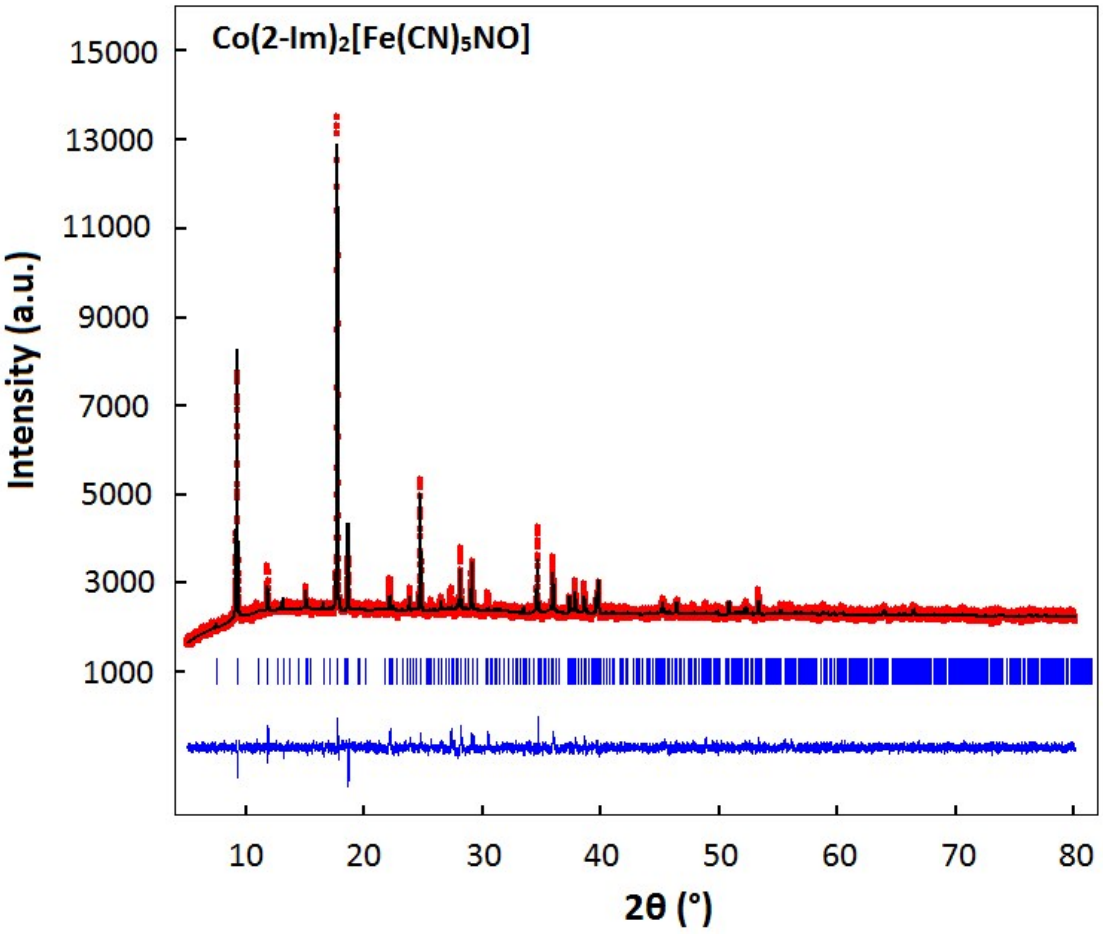

Figure S4: Diffuse reflectance UV-Vis spectra of the solids intercalated T(2-MeIm)$_2$[Fe(CN)$_5$NO]·2H$_2$O (T= Mn, Ni, Co and Fe).

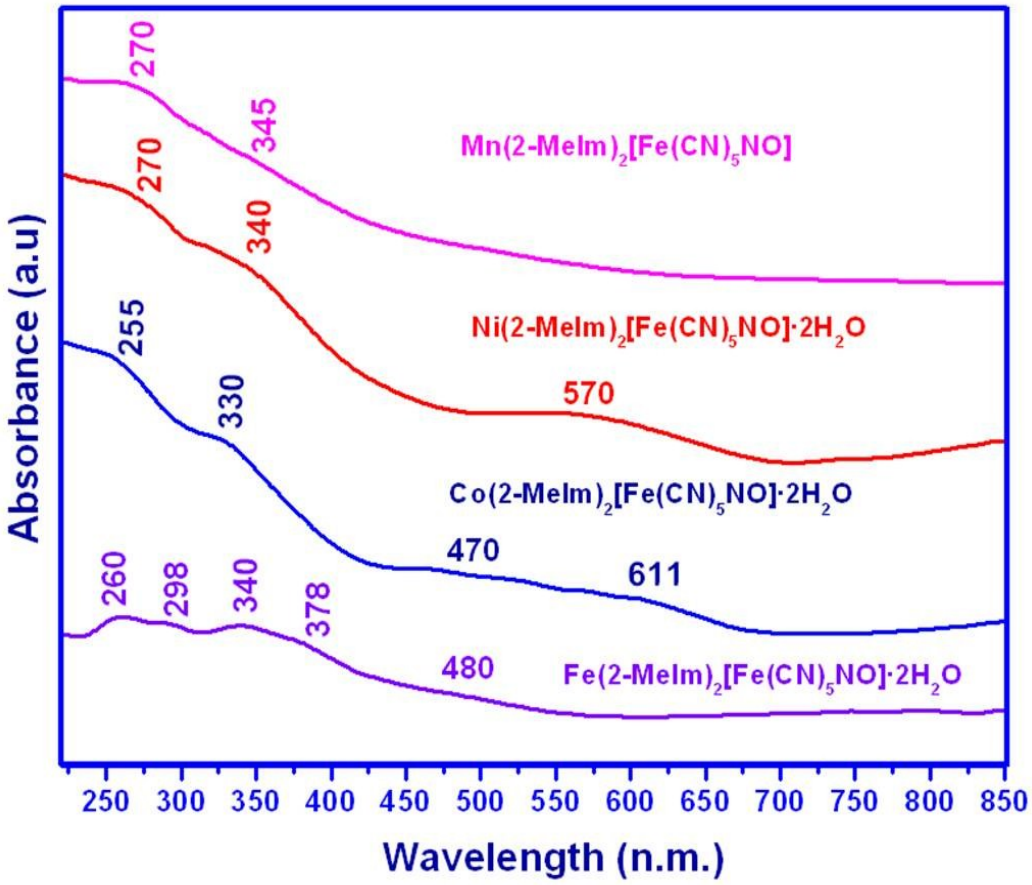

Figure S5: TG and DTA curves for the thermal decomposition of $Mn(2\text{-}MeIm)_2[Fe(CN)_5NO]$ in air.

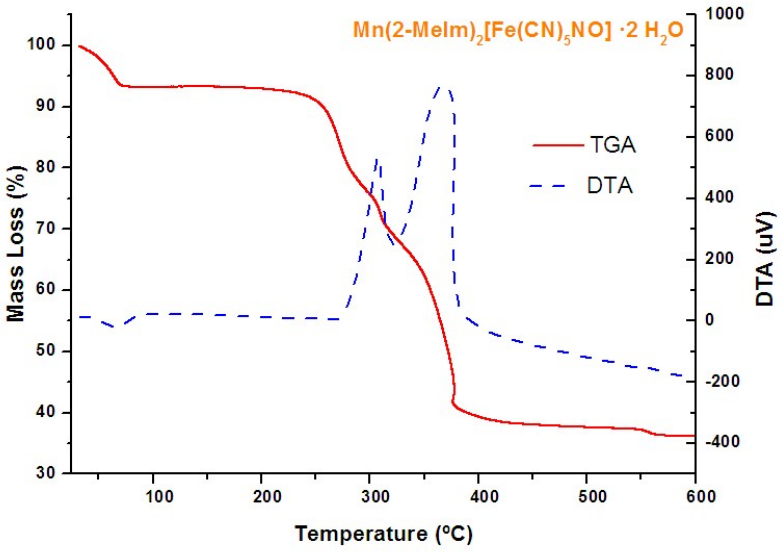

Figure S6: TG and DTA curves for the thermal decomposition of $Mn(2\text{-}MeIm)_2[Fe(CN)_5NO]\cdot 2H_2O$ in air.

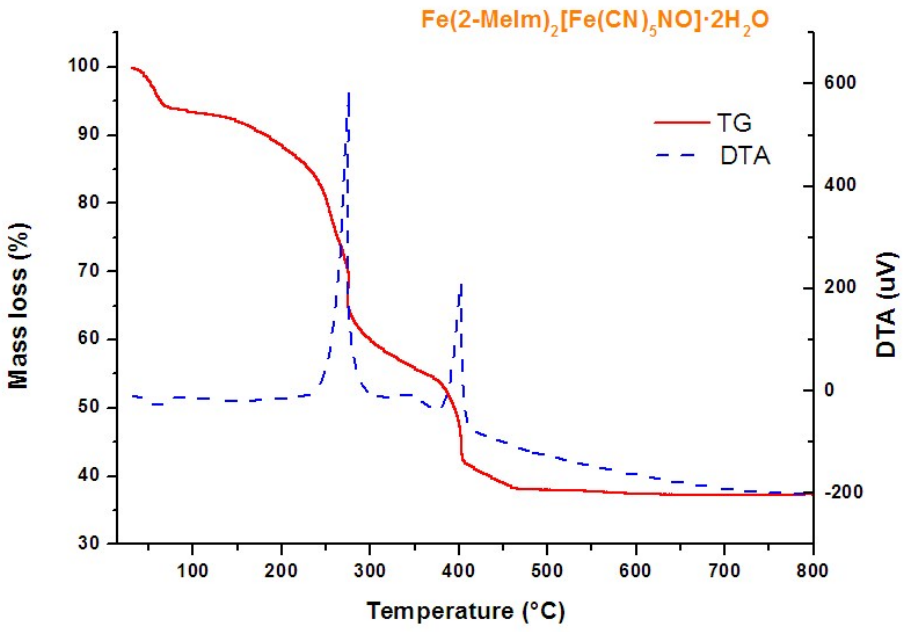

Figure S7: TG and DTA curves for the thermal decomposition of Co(2-MeIm)$_2$[Fe(CN)$_5$NO]·2H$_2$O in air.

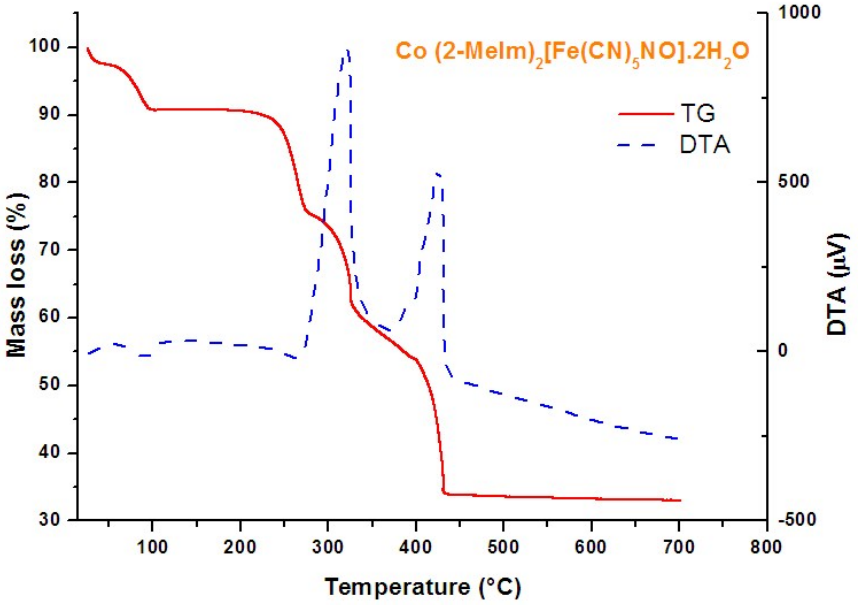

Figure S8: TG and DTA curves for the thermal decomposition of Ni(2-MeIm)$_2$[Fe(CN)$_5$NO]·2H$_2$O in air.

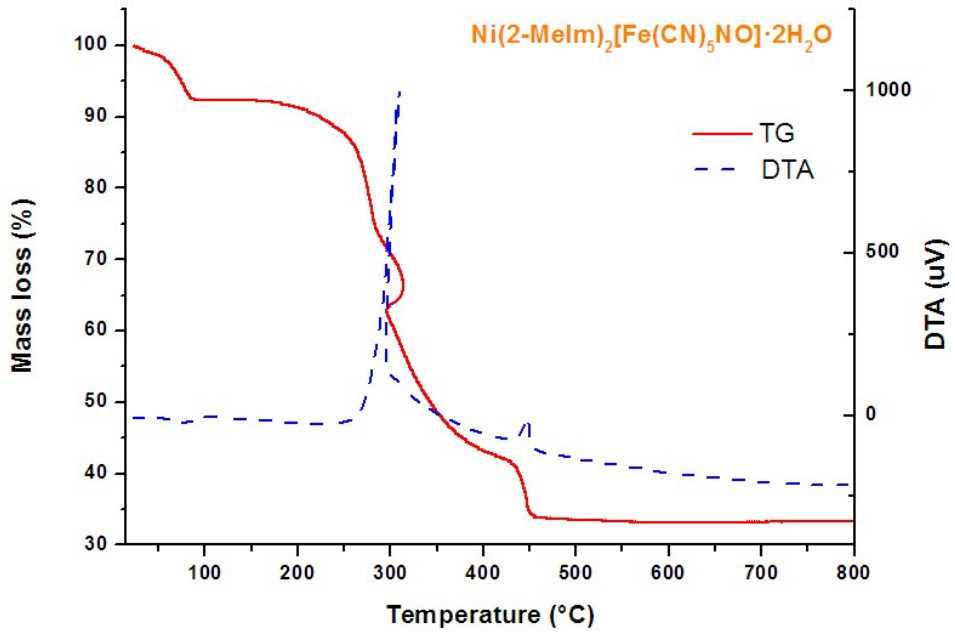

Figure S9: χ^{1} vs *T* plots for all reported complexes at 80 Oe. Full lines correspond to the Curie-Weiss fitting.

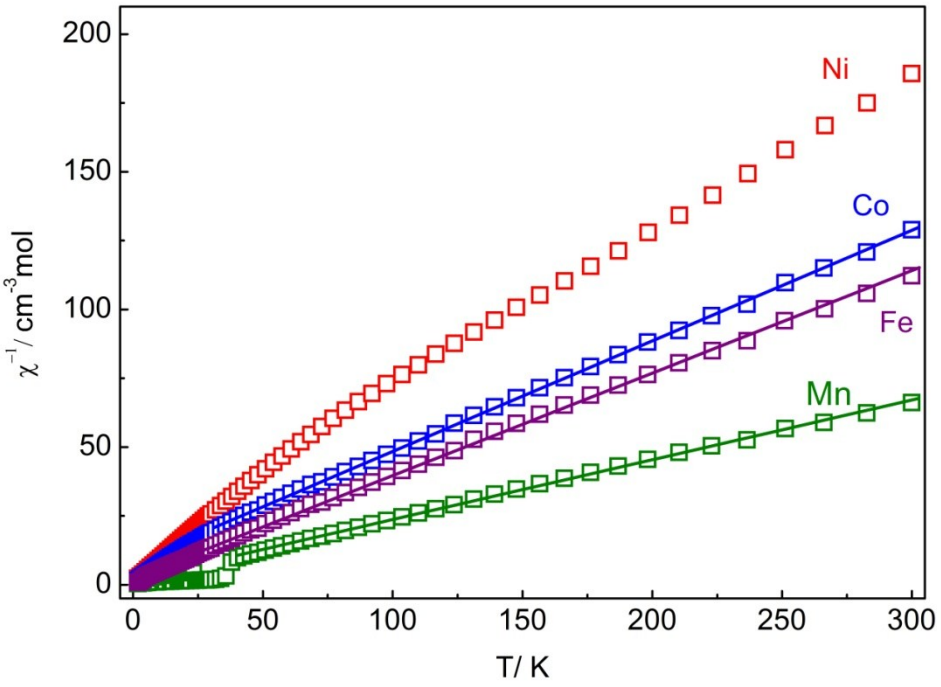

Figure S10: *M* vs *H* plots in the range -70 -70 kOe applied field for all reported complexes at 1.8 K.

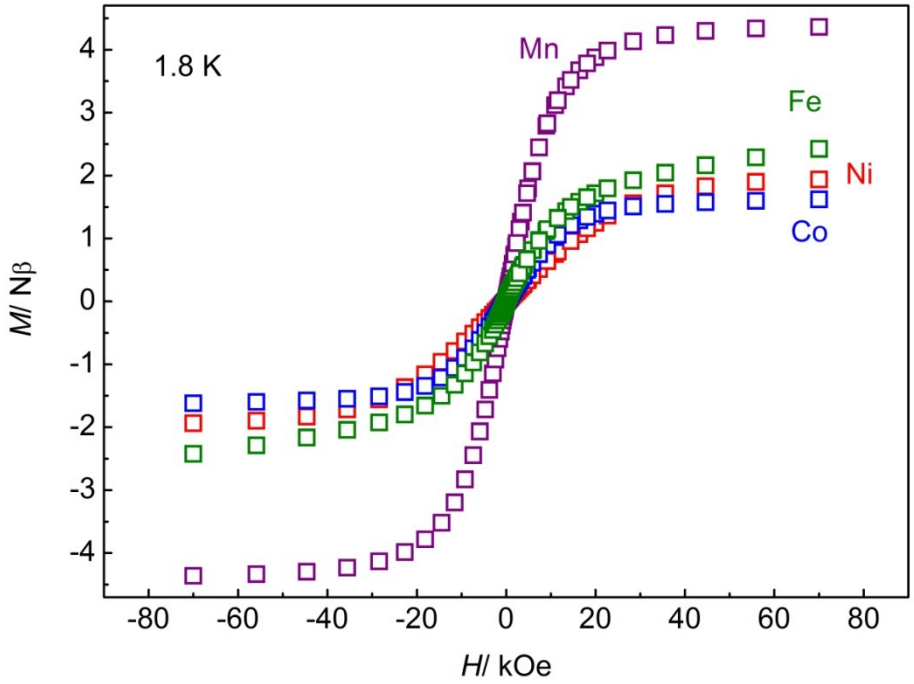

Table S1: Atomic coordinates and equivalent isotropic displacement parameters for $Mn(2\text{-}Im)_2[Fe(CN)_5NO]\cdot 2H_2O$.

Atom	Wyck.	x	y	z	Uiso*/Ueq
Fe	4a	0.0032 (8)	0.9974 (2)	0.2211 (2)	0.036 (1)
Mn	4a	0.0008 (4)	0.7476 (2)	0.7351 (3)	0.036 (1)
C1	4a	0.0006 (9)	0.9042 (5)	0.4006 (8)	0.047 (2)
C2	4a	−0.0061 (6)	1.0904 (5)	0.0431 (8)	0.047 (2)
C3	4a	0.0003 (9)	1.0897 (5)	0.4024 (8)	0.047 (2)
C4	4a	−0.0059 (6)	0.9050 (6)	0.0414 (9)	0.047 (2)
C5	4a	−0.0971 (1)	0.997 (1)	0.233 (1)	0.047 (2)
C6	4a	0.1600 (1)	0.8292 (5)	0.732 (2)	0.047 (2)
C7	4a	0.2319 (1)	0.7210 (5)	0.791 (2)	0.047 (2)
C8	4a	0.1667 (1)	0.6889 (4)	0.791 (2)	0.047 (2)
C9	4a	0.1322 (2)	0.9177 (5)	0.687 (1)	0.047 (2)
C10	4a	−0.1589 (1)	0.8178 (5)	0.732 (2)	0.047 (2)
C11	4a	−0.2275 (2)	0.7025 (6)	0.724 (2)	0.047 (2)
C12	4a	−0.1609 (1)	0.6745 (5)	0.726 (2)	0.047 (2)
C13	4a	−0.1338 (1)	0.9102 (4)	0.740 (2)	0.047 (2)
N1	4a	0.0033 (8)	0.8511 (5)	0.5086 (7)	0.047 (2)
N2	4a	−0.0102 (8)	1.1433 (4)	−0.0677 (7)	0.047 (2)
N3	4a	0.0013 (5)	1.1434 (5)	0.5105 (8)	0.047 (2)
N4	4a	−0.0086 (8)	0.8550 (4)	−0.0740 (6)	0.047 (2)
N5	4a	−0.1570 (1)	0.997 (1)	0.240 (1)	0.047 (2)
N6	4a	0.0895 (1)	0.997 (1)	0.210 (8)	0.047 (2)
N7	4a	0.1200 (1)	0.7564 (8)	0.754 (2)	0.047 (2)
N8	4a	0.2286 (2)	0.8107 (4)	0.753 (2)	0.047 (2)
N9	4a	−0.1164 (1)	0.7465 (9)	0.730 (1)	0.047 (2)
N10	4a	−0.2270 (2)	0.7940 (4)	0.729 (1)	0.047 (2)
O1	4a	0.1483 (9)	0.9976 (9)	0.2031 (7)	0.047 (2)
O2	4a	-0.1254 (1)	1.2070 (3)	0.2023 (8)	0.149 (1)
O3	4a	0.3726 (1)	0.7508 (6)	0.777 (1)	0.149 (1)

Table S2: Selected bond lengths (Å) and bond angle (°) for $Mn(2\text{-}MIm)_2[Fe(CN)_5NO]\cdot 2H_2O$.

Bond distances (Å)			
Fe—C1	1.930 (7)	C4—N4	1.137 (9)
Fe—C2	1.928 (7)	C5—N5	1.149 (3)
Fe—C3	1.929 (7)	N6—O1	1.128 (18)
Fe—C4	1.930 (9)	N7—C6	1.351 (12)
Fe—C5	1.924 (15)	N7—C8	1.385 (11)
Fe—N6	1.656 (16)	C6—N8	1.353 (5)
Mn—N1	2.284 (7)	N8—C7	1.387 (10)
Mn—$N2^{i}$	2.008 (7)	C7—C8	1.341 (4)
Mn—$N3^{ii}$	2.444 (7)	C6—C9	1.478 (10)
Mn—$N4^{iii}$	2.153 (6)	N9—C10	1.352 (13)
Mn—N7	2.293 (8)	N9—C12	1.384 (12)
Mn—N9	2.247 (8)	C10—N10	1.354 (5)
C1—N1	1.130 (9)	N10—C11	1.384 (11)
C2—N2	1.144 (9)	C11—C12	1.345 (6)
C3—N3	1.136 (10)	C10—C13	1.479 (9)
Bond Angles (°)			
Fe—C1—N1	175.6 (8)	Mn—$N2^{i}$—$C2^{i}$	168.2 (8)
Fe—C2—N2	177.1 (7)	Mn—$N3^{ii}$—$C3^{ii}$	174.2 (7)
Fe—C3—N3	177.3 (8)	Mn—$N4^{iii}$—$C4^{iii}$	169.5 (8)
Fe—C4—N4	174.5 (7)	Mn—N7—C6	127.3 (7)
Fe—C5—N5	179.8 (13)	Mn—N7—C8	127.8 (7)
Fe—N6—O1	179 (6)	Mn—N9—C10	126.6 (7)
C1—Fe—C2	173.2 (7)	Mn—N9—C12	128.5 (7)
C1—Fe—C3	93.3 (6)	N1—Mn—$N2^{i}$	95.2 (5)
C1—Fe—C4	86.3 (5)	N1—Mn—$N3^{ii}$	176.9 (5)
C1—Fe—C5	86.6 (8)	N1—Mn—$N4^{iii}$	87.6 (3)
C1—Fe—N6	93.3 (17)	N1—Mn—N7	89.0 (6)
C2—Fe—C3	86.4 (5)	N1—Mn—N9	90.8 (6)
C2—Fe—C4	93.3 (6)	$N2^{i}$—Mn—$N3^{ii}$	87.8 (4)
C2—Fe—C5	86.6 (7)	$N2^{i}$—Mn—$N4^{iii}$	177.1 (5)
C2—Fe—N6	93.5 (15)	$N2^{i}$—Mn—N7	89.6 (6)
C3—Fe—C4	173.2 (7)	$N2^{i}$—Mn—N9	94.2 (6)
C3—Fe—C5	86.7 (8)	$N3^{ii}$—Mn—$N4^{iii}$	89.3 (4)
C3—Fe—N6	93.7 (17)	$N3^{ii}$—Mn—N7	90.4 (5)
C4—Fe—C5	86.5 (8)	$N3^{ii}$—Mn—N9	89.5 (5)
C4—Fe—N6	93.1 (16)	$N4^{iii}$—Mn—N7	90.0 (6)
C5—Fe—N6	179.6 (8)	$N4^{iii}$—Mn—N9	86.1 (6)
Mn—N1—C1	175.6 (7)	N7—Mn—N9	176.2 (3)

Symmetry codes: (i) −x, y−1/2, −z+1/2; (ii) −x, y−1/2, −z+3/2; (iii) x, y, z+1.

Table S3: Hydrogen bond distances (Å) for $Mn(2\text{-}MIm)_2[Fe(CN)_5NO]\cdot 2H_2O$..

D—H···A	**D···A**
O2···$N10^{iV}$	2.835 (4)
O3···N8	2.910 (5)

Symmetry code: (iv) -1/2−x, 2−y,-1/2 +z.

Table S4: Atomic coordinates and equivalent isotropic displacement parameters for Fe(2-Im)$_2$[Fe(CN)$_5$NO]·2H$_2$O.

Atom	Wyck.	x	y	z	Uiso*/Ueq
Fe1	4a	0.0033 (2)	0.9978 (2)	0.2212 (2)	0.037(1)
Fe2	4a	0.0012 (5)	0.7438 (2)	0.7570 (6)	0.037(1)
C1	4a	0.0045 (2)	0.9026 (4)	0.4006 (7)	0.050(3)
C2	4a	−0.006 (1)	1.0918 (4)	0.0410 (8)	0.050(3)
C3	4a	0.0005 (2)	1.0911 (4)	0.4048 (6)	0.050(3)
C4	4a	−0.0020 (1)	0.9037 (4)	0.0406 (8)	0.050(3)
C5	4a	−0.0976 (2)	0.997 (1)	0.233 (1)	0.050(3)
C6	4a	0.1627 (2)	0.8185 (5)	0.754 (2)	0.050(3)
C7	4a	0.2317 (3)	0.7020 (4)	0.754 (2)	0.050(3)
C8	4a	0.1648 (2)	0.6736 (5)	0.754 (2)	0.050(3)
C9	4a	0.1376 (2)	0.9118 (4)	0.750 (2)	0.050(3)
C10	4a	−0.1582 (2)	0.8185 (5)	0.752 (2)	0.050(3)
C11	4a	−0.2273 (3)	0.7021 (5)	0.744 (3)	0.050(3)
C12	4a	−0.1603 (2)	0.6738 (5)	0.746 (2)	0.050(3)
C13	4a	−0.1329 (2)	0.9118 (4)	0.760 (2)	0.050(3)
N1	4a	−0.0004 (8)	0.8508 (4)	0.5137 (7)	0.050(3)
N2	4a	−0.0094 (7)	1.1468 (4)	−0.0654 (5)	0.050(3)
N3	4a	0.002 (1)	1.1447 (4)	0.5165 (7)	0.050(3)
N4	4a	0.0012 (9)	0.8451 (3)	−0.0585 (6)	0.050(3)
N5	4a	−0.1580 (2)	0.997 (1)	0.241 (1)	0.050(3)
N6	4a	0.0903 (2)	0.998 (1)	0.210 (8)	0.050(3)
N7	4a	0.1200 (2)	0.7464 (9)	0.754 (3)	0.050(3)
N8	4a	0.2313 (2)	0.7945 (4)	0.754 (2)	0.050(3)
N9	4a	−0.1155 (2)	0.7465 (9)	0.750 (3)	0.050(3)
N10	4a	−0.2268 (2)	0.7945 (4)	0.749 (2)	0.050(3)
O1	4a	0.1495 (1)	0.9981 (9)	0.2030 (7)	0.050(3)
O2	4a	-0.1261 (5)	1.2075 (2)	0.213 (1)	0.125(6)
O3	4a	0.3690 (4)	0.7505 (5)	0.7616 (2)	0.125(6)

Table S5: Selected bond angle (°) for $Fe(2\text{-}MIm)_2[Fe(CN)_5NO]\cdot 2H_2O$.

Bond distances (Å)			
Fe1—C1	1.930 (6)	C4—N4	1.136 (7)
Fe1—C2	1.929 (7)	C5—N5	1.151 (5)
Fe1—C3	1.931 (6)	N6—O1	1.128 (5)
Fe1—C4	1.926 (6)	N7—C6	1.351 (13)
Fe1—C5	1.922 (5)	N7—C8	1.384 (13)
Fe1—N6	1.657 (6)	C6—N8	1.354 (6)
Fe2—N1	2.383 (7)	N8—C7	1.385 (8)
Fe2—N2^{i}	2.016 (7)	C7—C8	1.342 (7)
Fe2—N3^{ii}	2.214 (7)	C6—C9	1.477 (9)
Fe2—N4^{iii}	2.023 (6)	N9—C10	1.350 (13)
Fe2—N7	2.261 (10)	N9—C12	1.383 (13)
Fe2—N9	2.221 (10)	C10—N10	1.354 (6)
C1—N1	1.133 (8)	N10—C11	1.384 (10)
C2—N2	1.130 (8)	C11—C12	1.344 (7)
C3—N3	1.141 (8)	C10—C13	1.479 (9)
Bond Angles (°)			
Fe1—C1—N1	173.2 (7)	Fe1—N2^{i}—C2^{i}	172.3 (7)
Fe1—C2—N2	178.0 (6)	Fe1—N3^{ii}—C3^{ii}	176.0 (6)
Fe1—C3—N3	176.6 (6)	Fe1—N4^{iii}—C4^{iii}	176.0 (6)
Fe1—C4—N4	172.9 (6)	Fe1—N7—C6	127.9 (9)
Fe1—C5—N5	179.5 (5)	Fe1—N7—C8	127.0 (8)
Fe1—N6—O1	179.8 (5)	Fe1—N9—C10	128.0 (9)
C1—Fe1—C2	175.4 (6)	Fe1—N9—C12	127.0 (8)
C1—Fe1—C3	94.0 (4)	N1—Fe2—N2^{i}	88.6 (4)
C1—Fe1—C4	85.3 (4)	N1—Fe2—N3^{ii}	177.7 (5)
C1—Fe1—C5	88.7 (5)	N1—Fe2—N4^{iii}	89.1 (3)
C1—Fe1—N6	91.3 (15)	N1—Fe2—N7	89.7 (7)
C2—Fe1—C3	86.3 (4)	N1—Fe2—N9	87.6 (7)
C2—Fe1—C4	94.0 (5)	N2^{i}—Fe2—N3^{ii}	91.7 (4)
C2—Fe1—C5	86.7 (7)	N2^{i}—Fe2—N4^{iii}	175.0 (5)
C2—Fe1—N6	93.3 (15)	N2^{i}—Fe2—N7	85.9 (7)
C3—Fe1—C4	175.4 (5)	N2^{i}—Fe2—N9	94.3 (7)
C3—Fe1—C5	86.9 (5)	N3^{ii}—Fe2—N4^{iii}	90.7 (4)
C3—Fe1—N6	93.4 (16)	N3^{ii}—Fe2—N7	92.6 (8)
C4—Fe1—C5	88.5 (5)	N3^{ii}—Fe2—N9	90.1 (5)
C4—Fe1—N6	91.2 (14)	N4^{iii}—Fe2—N7	89.6 (7)
C5—Fe1—N6	179.6 (5)	N4^{iii}—Fe2—N9	90.1 (8)
Fe1—N1—C1	174.4 (7)	N7—Fe2—N9	177.2 (5)

Symmetry codes: (i) −x, y−1/2, −z+1/2; (ii) −x, y−1/2, −z+3/2; (iii) x, y, z+1.

Table S6: Hydrogen bond distances (Å) for $Fe(2\text{-}MIm)_2[Fe(CN)_5NO]\cdot 2H_2O$.

D—H···A	**D···A**
O2···N10^{iV}	2.811 (1)
O3···N8	2.702 (8)

Symmetry code: (iv) -1/2−x, 2−y,-1/2 +z.

Table S7: Atomic coordinates and equivalent isotropic displacement parameters for $Co(2\text{-}Im)_2[Fe(CN)_5NO]\cdot 2H_2O$

Atom	Wyck.	x	y	z	Uiso*/Ueq
Fe	4a	0.0159 (7)	0.9993 (2)	0.249 (1)	0.035 (2)
Co	4a	0.0063 (1)	0.7431 (2)	0.7362 (3)	0.035 (2)
C1	4a	0.0153 (6)	0.9058 (6)	0.4373 (8)	0.046 (3)
C2	4a	0.0048 (6)	1.0922 (6)	0.0642 (8)	0.046 (3)
C3	4a	0.0163 (5)	1.0949 (6)	0.4307 (8)	0.046 (3)
C4	4a	0.0032 (7)	0.9035 (6)	0.0697 (7)	0.046 (3)
C5	4a	−0.0850 (2)	1.000 (1)	0.271 (2)	0.046 (3)
C6	4a	0.1664 (3)	0.8199 (5)	0.762 (2)	0.046 (3)
C7	4a	0.2359 (4)	0.7036 (5)	0.784 (1)	0.046 (3)
C8	4a	0.1693 (3)	0.6744 (5)	0.772 (2)	0.046 (3)
C9	4a	0.1427 (2)	0.9165 (5)	0.754 (3)	0.046 (3)
C10	4a	−0.1480 (2)	0.8254 (5)	0.731 (2)	0.046 (3)
C11	4a	−0.2256 (4)	0.7172 (6)	0.732 (2)	0.046 (3)
C12	4a	−0.1608 (3)	0.6809 (5)	0.733 (2)	0.046 (3)
C13	4a	−0.1179 (2)	0.9207 (4)	0.729 (2)	0.046 (3)
N1	4a	0.0177 (5)	0.8493 (5)	0.5438 (7)	0.046 (3)
N2	4a	−0.0066 (6)	1.1501 (5)	−0.0338 (7)	0.046 (3)
N3	4a	0.0165 (5)	1.1493 (5)	0.5419 (7)	0.046 (3)
N4	4a	−0.0045 (6)	0.8446 (5)	−0.0298 (6)	0.046 (3)
N5	4a	−0.1454 (2)	1.001 (1)	0.2847 (9)	0.046 (3)
N6	4a	0.1027 (2)	0.998 (1)	0.230 (1)	0.046 (3)
N7	4a	0.1241 (2)	0.7470 (7)	0.758 (2)	0.046 (3)
N8	4a	0.2351 (3)	0.7966 (4)	0.7776 (1)	0.046 (3)
N9	4a	−0.1108 (2)	0.7483 (7)	0.732 (2)	0.046 (3)
N10	4a	−0.2182 (3)	0.8096 (4)	0.732 (2)	0.046 (3)
O1	4a	0.1619 (2)	0.9972 (9)	0.2170 (8)	0.046 (3)
O2	4a	-0.1207 (3)	1.2096 (4)	0.1927 (9)	0.155 (8)
O3	4a	0.3732 (1)	0.7513 (4)	0.7650 (8)	0.155 (8)

Table S8: Selected bond lengths (Å) and bond angle (°) for $Co(2\text{-}MIm)_2[Fe(CN)_5NO]\cdot 2H_2O$.

Bond distances (Å)			
Fe—C1	1.941 (9)	C4—N4	1.141 (10)
Fe—C2	1.929 (9)	C5—N5	1.152 (6)
Fe—C3	1.931 (9)	N6—O1	1.129 (5)
Fe—C4	1.937 (9)	N7—C6	1.352 (11)
Fe—C5	1.924 (14)	N7—C8	1.385 (11)
Fe—N6	1.655 (14)	C6—N8	1.355 (8)
Co—N1	2.112 (7)	N8—C7	1.387 (10)
Co—$N2^{i}$	2.008 (7)	C7—C8	1.341 (10)
Co—$N3^{ii}$	2.163 (7)	C6—C9	1.509 (10)
Co—$N4^{iii}$	2.270 (7)	N9—C10	1.349 (11)
Co—N7	2.244 (4)	N9—C12	1.382 (10)
Co—N9	2.226 (4)	C10—N10	1.354 (7)
C1—N1	1.138 (10)	N10—C11	1.384 (11)
C2—N2	1.134 (11)	C11—C12	1.345 (10)
C3—N3	1.138 (10)	C10—C13	1.531 (9)
Bond Angles (°)			
Fe—C1—N1	176.8 (9)	Co—$N2^{i}$—$C2^{i}$	166.8 (8)
Fe—C2—N2	173.4 (9)	Co—$N3^{ii}$—$C3^{ii}$	167.5 (8)
Fe—C3—N3	177.9 (9)	Co—$N4^{iii}$—$C4^{iii}$	164.6 (8)
Fe—C4—N4	177.1 (9)	Co—N7—C6	128.0 (6)
Fe—C5—N5	179.5 (12)	Co—N7—C8	127.1 (6)
Fe—N6—O1	179.9 (13)	Co—N9—C10	123.6 (6)
C1—Fe—C2	173.4 (7)	Co—N9—C12	131.4 (6)
C1—Fe—C3	93.4 (7)	N1—Co—$N2^{i}$	92.5 (5)
C1—Fe—C4	86.2 (5)	N1—Co—$N3^{ii}$	170.6 (6)
C1—Fe—C5	86.6 (8)	N1—Co—$N4^{iii}$	89.7 (4)
C1—Fe—N6	93.1 (8)	N1—Co—N7	85.6 (5)
C2—Fe—C3	86.3 (5)	N1—Co—N9	93.9 (5)
C2—Fe—C4	93.3 (7)	$N2^{i}$—Co—$N3^{ii}$	95.0 (4)
C2—Fe—C5	86.8 (8)	$N2^{i}$—Co—$N4^{iii}$	174.7 (5)
C2—Fe—N6	93.5 (8)	$N2^{i}$—Co—N7	93.8 (6)
C3—Fe—C4	173.1 (7)	$N2^{i}$—Co—N9	91.0 (6)
C3—Fe—C5	86.8 (7)	$N3^{ii}$—Co—$N4^{iii}$	82.4 (4)
C3—Fe—N6	93.5 (8)	$N3^{ii}$—Co—N7	99.5 (5)
C4—Fe—C5	86.2 (8)	$N3^{ii}$—Co—N9	80.3 (5)
C4—Fe—N6	93.5 (9)	$N4^{iii}$—Co—N7	91.2 (5)
C5—Fe—N6	179.6 (7)	$N4^{iii}$—Co—N9	84.1 (5)
Co—N1—C1	171.7 (8)	N7—Co—N9	175.3 (3)

Symmetry codes: (i) −x, y−1/2, −z+1/2; (ii) −x, y−1/2, −z+3/2; (iii) x, y, z+1.

Table S9: Hydrogen bond distances (Å) for $Co(2\text{-}MIm)_2[Fe(CN)_5NO]\cdot 2H_2O$.

D—H···A	**D···A**
O2···$N10^{iV}$	3.087 (8)
O3···N8	2.710 (6)

Symmetry code: (iv) -1/2−x, 2−y,-1/2 +z.

Table S10: Atomic coordinates and equivalent isotropic displacement parameters for Mn(2-Im)$_2$[Fe(CN)$_5$NO]·2H$_2$O.

Atom	Wyck.	x	y	z	Uiso*/Ueq
Fe	4a	0.002 (1)	1.0007 (9)	0.248 (6)	0.036 (7)*
Ni	4a	−0.0083 (1)	0.7506 (9)	0.751 (5)	0.036 (7)*
C1	4a	−0.0006 (5)	0.904 (2)	0.432 (4)	0.057 (1)*
C2	4a	−0.0063 (4)	1.096 (2)	0.067 (4)	0.057 (1)*
C3	4a	0.0035 (5)	1.094 (2)	0.436 (4)	0.057 (1)*
C4	4a	−0.010 (4)	0.906 (2)	0.063 (4)	0.057 (1)*
C5	4a	−0.098 (1)	1.004 (5)	0.268 (9)	0.057 (1)*
C6	4a	0.166 (1)	0.810 (2)	0.762 (1)	0.057 (1)*
C7	4a	0.225 (2)	0.684 (1)	0.768 (8)	0.057 (1)*
C8	4a	0.156 (1)	0.664 (2)	0.764 (9)	0.057 (1)*
C9	4a	0.148 (1)	0.908 (1)	0.756 (9)	0.057 (1)*
C10	4a	−0.163 (1)	0.816 (1)	0.736 (9)	0.057 (1)*
C11	4a	−0.222 (2)	0.689 (2)	0.712 (7)	0.057 (1)*
C12	4a	−0.153 (1)	0.669 (1)	0.716 (8)	0.057 (1)*
C13	4a	−0.145 (1)	0.913 (1)	0.755 (1)	0.057 (1)*
N1	4a	0.000 (3)	0.849 (1)	0.543 (3)	0.057 (1)*
N2	4a	−0.004 (3)	1.151 (1)	−0.047 (3)	0.057 (1)*
N3	4a	0.008 (2)	1.151 (1)	0.544 (3)	0.057 (1)*
N4	4a	−0.014 (2)	0.849 (1)	−0.043 (3)	0.057 (1)*
N5	4a	−0.1589 (9)	1.006 (3)	0.279 (5)	0.057 (1)*
N6	4a	0.0894 (8)	0.997 (3)	0.232 (6)	0.057 (1)*
N7	4a	0.1179 (8)	0.743 (3)	0.761 (8)	0.057 (1)*
N8	4a	0.2326 (9)	0.777 (2)	0.766 (6)	0.057 (1)*
N9	4a	−0.1149 (8)	0.748 (3)	0.730 (7)	0.057 (1)*
N10	4a	−0.229 (1)	0.783 (2)	0.725 (6)	0.057 (1)*
O1	4a	0.1485 (7)	0.995 (2)	0.220 (4)	0.057 (1)*
O2	4a	−0.1237 (8)	1.209 (1)	0.210 (4)	0.071 (3)*
O3	4a	0.3743 (7)	0.752 (3)	0.766 (5)	0.071 (3)*

Table S11: Selected bond lengths (Å) and bond angle (°) for $Ni(2\text{-}MIm)_2[Fe(CN)_5NO]\cdot 2H_2O$.

Bond distances (Å)			
Fe—C1	1.94 (4)	C4—N4	1.13 (3)
Fe—C2	1.91 (4)	C5—N5	1.16 (3)
Fe—C3	1.92 (4)	N6—O1	1.12 (18)
Fe—C4	1.93 (4)	N7—C6	1.35 (4)
Fe—C5	1.91 (3)	N7—C8	1.37 (5)
Fe—N6	1.67 (2)	C6—N8	1.36 (3)
Ni—N1	2.08 (3)	N8—C7	1.38 (3)
Ni—N2^{i}	2.08 (3)	C7—C8	1.34 (4)
Ni—N3^{ii}	2.07 (3)	C6—C9	1.49 (3)
Ni—N4^{iii}	2.07 (3)	N9—C10	1.36 (4)
Ni—N7	2.402 (16)	N9—C12	1.38 (4)
Ni—N9	2.031 (16)	C10—N10	1.35 (3)
C1—N1	1.13 (3)	N10—C11	1.40 (4)
C2—N2	1.15 (3)	C11—C12	1.34 (4)
C3—N3	1.14 (3)	C10—C13	1.48 (2)
Bond Angles (°)			
Fe—C1—N1	177 (4)	Ni—N2^{i}—C2^{i}	176 (4)
Fe—C2—N2	173 (4)	Ni—N3^{ii}—C3^{ii}	175(4)
Fe—C3—N3	176 (4)	Ni—N4^{iii}—C4^{iii}	172 (4)
Fe—C4—N4	177 (4)	Ni—N7—C6	130 (2)
Fe—C5—N5	180(3)	Ni—N7—C8	125 (2)
Fe—N6—O1	179 (2)	Ni—N9—C10	131 (2)
C1—Fe—C2	174 (3)	Ni—N9—C12	123 (2)
C1—Fe—C3	93 (3)	N1—Ni—N2^{i}	89.3 (5)
C1—Fe—C4	86 (2)	N1—Ni—N3^{ii}	175.4 (19)
C1—Fe—C5	87 (3)	N1—Ni—N4^{iii}	91.1 (18)
C1—Fe—N6	93 (2)	N1—Ni—N7	89 (2)
C2—Fe—C3	87 (2)	N1—Ni—N9	92 (2)
C2—Fe—C4	94 (3)	N2^{i}—Ni—N3^{ii}	89.5 (17)
C2—Fe—C5	87 (3)	N2^{i}—Ni—N4^{iii}	176.5 (19)
C2—Fe—N6	93 (2)	N2^{i}—Ni—N7	83 (2)
C3—Fe—C4	174 (3)	N2^{i}—Ni—N9	93 (2)
C3—Fe—C5	87(3)	N3^{ii}—Ni—N4^{iii}	89.9 (15)
C3—Fe—N6	93 (3)	N3^{ii}—Ni—N7	86.7 (19)
C4—Fe—C5	87 (3)	N3^{ii}—Ni—N9	92 (2)
C4—Fe—N6	93 (3)	N4^{iii}—Ni—N7	94 (2)
C5—Fe—N6	179.6 (8)	N4^{iii}—Ni—N9	91 (2)
Ni—N1—C1	175 (4)	N7—Ni—N9	175.5 (13)

Symmetry codes: (i) −x, y−1/2, −z+1/2; (ii) −x, y−1/2, −z+3/2; (iii) x, y, z+1.

Table S12: Hydrogen bond distances (Å) for $Ni(2\text{-}MIm)_2[Fe(CN)_5NO]\cdot 2H_2O$.

D—H···A	**D···A**
O2···N10^{iV}	2.80 (2)
O3···N8	2.71 (2)

Symmetry code: (iv) -1/2−x, 2−y,-1/2 +z.

Table S13: IR and Raman bands and tentative assignments for the intercalated solids $T(2\text{-MeIm})_2[Fe(CN)_5NO]\cdot 2H_2O$ (T= Co, Ni, Mn and Fe).

2-MeIm		**$Co(2\text{-MeIm})_2NP$**		**$Ni(2\text{-MeIm})_2NP$**		**$Mn(2\text{-MeIm})_2NP$**		**$Fe(2\text{-MeIm})_2NP$**		**Assignment[b]**
FT-IR[a]	**Raman**	**FT-IR**	**Raman**	**FT-IR**	**Raman**	**FT-IR**	**Raman**	**FT-IR**	**Raman**	
-	-	3646	-	3643	-	3652	-	3647	-	νH2O
-	-	3567	-	3566		3570		3568	-	νNH
3137 s	-	3150 vw	3154	3153 vw	3258	3147 vw	3147	3150 vw	-	νCH
3110 vs	-	3125 s	3126	3127 s	3130	3123 s	3124	3125 s	-	νCH
2961 vs	2957	2964 vw	-	2965 vw	-	2966 vw	-	2962 vw	-	νaCH_3
2926 vs	2925	2931 w	2931	2931 w	2931	2931w	2931	2930 w	2930	νsCH_3
-	-	-	2192	-	2197	-	2184	-	2186	νCN_{eq}
-	-	2180 vs	2179	2185 vs	2188	2173 vs	-	2174 vs	2179	νCN_{eq}
-	-	2165 w	2163	2165 w	2165	-	2160	2164 w	2161	νCN_{ax}
-	-	1936 vs	-	1937 vs	-	1934 vs	-	1935 vs	-	νNO
-	-	1604	-	1605	-	1605	-	1606	-	δH_2O
1597 vs	-	1569 m	-	1568 m	-	1569 m	-	1566 m		νC-**N**+νC-C
1445 s	1446	1431 w	-	1433 w	-	1433 w	-	1433 w	-	δaCH_3
1411 w	1416	1405 vw	-	1406 vw	1403	1404 vw	-	1405 vw	-	νC-N+νC-C
1373 vs	1370	1362 m	1360	1360 m	1360	1361 m	1358	1361 m	1358	δsCH_3
1303 vs	1303	1290 w	-	1290 w	-	1289 w	-	1291 w	-	νC-**N**+ δC-H +δN-H
1219 s	-	-	-	-	-	-	-	1193	-	νC-**N**+δC-H +δN-H
1155 vs	1163	1151 w	-	1154 w	-	1150 w	-	1149 w	-	νC-**N** + δC-H +νC-C
-	-	1139 s		1141 s		1135 s		1137 s		νC-**N**
1115 vs	1126	1118 m	-	1122 m		1116 m	-	1118 m	-	νC-C + δC-H
1048 w	-	1054 vvw	-	1052 vvw	-	1048 vvw	-	1048 vvw	-	δC-H + δN-H
995 s	993	1016 m	1010	1012 m	1011	1008 m	1008	1010 m	-	ρCH_3
945 s	939	977 vvs	-	976 vvs	-	975 vvs	-	976 vvs	-	νC-**N** +δC-H + ρCH_3
916 s	916	875 s	-	873 s	-	871 s	-	875 s	-	δring
-	840	-	-	-	-	-	-	-	-	γCH
743 vs	-	780 m	-	779 m	-	777 m	-	780 m	-	γCH
682 vs	682	681 s	681	681 s	683	680 s	678	680 s	680	γRing

-	-	667 m	-	667 m	665	667 m	665	667 m	-	δ FeNO
-	-	644 w	644	643 w	643	645 w	641	645 w	642	ν Fe-N
629 s	629	631 vvs	-	631 vvs	-	631 vvs	-	631 vvs	-	γ Ring
-	-	516 s	-	517 s	-	513 s	509	513 s	-	$\delta FeCN_{eq+ax}$
-	479	478 vvs	478	-	479	479 vvs	477	482 vvs	483	γ NH
-	-	446 vs	-	443 vs	-	-	447	441 vs	-	ν Fe-C $_{eq\ A`+A"}$
-	-	427 s	427	426 s	424	421 s	421	424 s	423	νFe-C $_{eq\ A`}$
	269									τ Ring
-	-	-	230	-	249	-	191	-	239	ν M-N (Im)
-	-	-	203	-	217	-	-	-	210	ν M-N(CN)
-	68	-	145	-	153	-	140	-	130	τCH_3

[a] IR intensities; s: strong, m: medium, w: weak, v: very

[b] ν: stretching, δ: bending, γ out-of-plane bending modes; a: anti-symmetric, s: symmetric