Precisely tunable magnetic phase transition temperature, T_C, through formation of molecule alloy in $[Ni_xPt_{1-x}(mnt)_2]^-$-based spin systems (mnt^{2-} = maleonitriledithiolate, x = 0.09-0.91)

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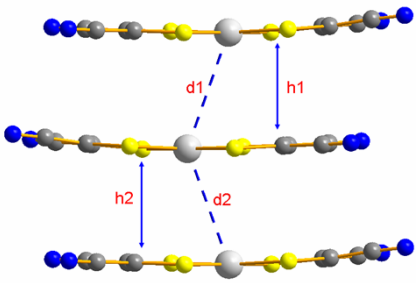
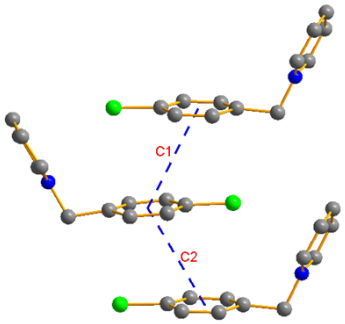
Table S1 Inter-plane distances between the nearest neighbors (h1, h2) and interatomic distances of M…M (d1, d2), S…S (m1, m2, m3, m4) and the centroid-to-centroid distances (c1, c2), within an anionic or cation stack for alloy compounds **[Cl-BzPy][$Ni_xPt_{1-x}(mnt)_2$]** (x = 0.9-0.91) with the parent compounds at 296 K.





Molar fraction	h1	h2	d1	d2	c1	c2
x = 0[2]	3.633	3.602	3.885	3.885	4.168	4.168
x = 0.09	3.643	3.634	3.920	3.930	4.201)	4.201
x = 0.21	3.580	3.616	3.884	3.884	4.152	4.152
x = 0.34	3.616	3.593	3.897	3.897	4.161	4.160
x = 0.42	3.603	3.574	3.892	3.892	4.142	4.142
x = 0.54	3.572	3.607	3.901	3.901	4.145	4.145
x = 0.64	3.611	3.577	3.916	3.916	4.155	4.155
x = 0.73	3.558	3.591	3.904	3.904	4.141	4.141
0.84	3.553	3.585	3.912	3.912	4.139	4.139
x = 0.91	3.548	3.582	3.913	3.913	4.138	4.138
x = 1[4]	3.575	3.539	3.914	3.914	4.133	4.133

Molar fraction	m1	m2	m3	m4	m
x = 0[3]	3.986	4.079	3.883	3.705	3.913
x = 0.09	3.941	3.713	4.068	4.100	3.956
x = 0.21	4.080	3.996	3.698	3.892	3.916
x = 0.34	3.905	3.703	4.019	4.077	3.926
x = 0.42	3.904	3.704	4.020	4.063	3.922
x = 0.54	4.076	4.029	3.712	3.912	3.932

x = 0.64	3.927	3.726	4.050	4.084	3.947
x = 0.73	4.064	4.046	3.714	3.914	3.935
x = 0.84	4.063	4.058	3.728	3.916	3.941
x = 0.91	4.058	4.065	3.729	3.915	3.942
x = 1[1]	4.050	4.069	3.734	3.910	3.941

References

1. X. M. Ren, Q. J. Meng, Y. Song, C. S. Lu, C. J. Hu, X. Y. Chen, *Inorg. Chem.* 2002, **41**, 5686.
2. X. M. Ren, H. Okudera, R. K. Kremer, Y. Song, C. He, Q. J. Meng, P. H. Wu, *Inorg. Chem.* 2004, **43**, 2569.

Figure S1 Variations of interatomic or interplane distances between neighboring anions in a stack (c1, c2, d1, d2, h1 and h2) as x change for [Cl-BzPy][$Ni_xPt_{1-x}(mnt)_2$] (x = 0.09-0.91) with two parent compounds at ambient temperature.

Table S2 Ni and Pt molar fraction measured by Energy Dispersive Spectra (EDS) for [Cl-BzPy][$Ni_xPt_{1-x}(mnt)_2$] (x = 0.09-0.91).

x	0.09	0.21	0.34	0.42	0.54	0.64	0.73	0.84	0.91
Ni%	8.33	17.16	25.77	33.69	42.10	50.24	60.60	70.38	78.12
Pt%	91.67	82.84	74.23	66.31	57.90	49.76	39.40	29.62	21.88

Table S3 Ni and Pt molar fraction measured by Inductively coupled plasma-mass spectra (ICP-MS) for [Cl-BzPy][$Ni_xPt_{1-x}(mnt)_2$] (x = 0.09-0.91).

Compound	Ni	Pt	Ni/Pt[#]	Ni/Pt*	Ni/Pt**
x = 0.09	0.085	2.09	0.1359	0.1111	0.09695
x = 0.21	0.48	5.35	0.2997	0.25	0.26526
x = 0.34	1.22	8.77	0.4647	0.4286	0.50625
x = 0.42	0.59	2.5	0.7884	0.6667	0.7347
x = 0.54	2.11	6.22	1.1333	1	1.15285
x = 0.64	1.03	2.11	1.6308	1.5	1.74033
x = 0.73	2.1	2.45	2.8635	2.3333	2.64977
x = 0.84	4.22	3.06	4.6071	4	5.43625
x = 0.91	5.47	1.93	9.4683	9	10.73985

[#] ICP; *the molar ratio in the starting materials; **the molar ratio from occupation factors

Table S4 Crystal structure of [Cl-BzPy][$Ni_xPt_{1-x}(mnt)_2$] (x = 0.09, 0.54 and 0.91) at 100 K

Compound	x = 0.09	x = 0.54	x = 0.91
Temp. (K)		100	
Wavelength		0.71073	
Formula weight	667.67	601.00	556.34
CCDC no.	893368	893369	893370
Crystal system		Triclinic	
Space group		P-1	
a(Å)	7.1590(2)	7.1472(8)	7.1912(2)
b(Å)	12.2601(4)	12.1875(11)	12.0953(6)
c(Å)	26.4337(11)	26.268(2)	26.0555(12)
α(°)	87.472(3)	87.956(7)	88.532(4)
β(°)	85.016(3)	85.550(8)	86.554(3)
γ(°)	75.650(3)	76.447(9)	77.252(3)
V(Å^3),Z	2238.59(13)/4	2217.3(4)/4	2206.26(16)/4
ρ (g·cm-1)	1.981	1.800	1.675
F(000)	1282	1184	1118

Abs.coeff. (mm-1)	6.286	3.681	1.904
θ Range for data collection (°)	2.99-26.00	2.99-26.00	2.99-26.00
Index ranges	-9<h<8, -14<k<15, -27<l<32	-8<h<8, -15<k<14, -32<l<32	-8<h<8, -14<k<14, -32<l<24
Rint	0.1027	0.1055	
Independent reflect. /restraints/parameters	8775 / 0 / 555	8684 / 12 / 361	8643 / 0 / 560
Refinement method	Full-matrix least-squares on F^2		
Goodness of fit on F2	1.277	1.142	1.121
R1, wR2a [I>2σ(I)]	R1 = 0.1396 wR2 = 0.3277	R1 = 0.2621 wR2 = 0.5928	R1 = 0.2430 wR2 = 0.5535
R1, wR2a [all data]	R1 = 0.1548 wR2 = 0.3329	R1 = 0.2751 wR2 = 0.5969	R1 = 0.2537 wR2 = 0.5572
Residual (e·Å^{-3})	5.225/-11.153	10.100/-9.063	4.569/-5.279

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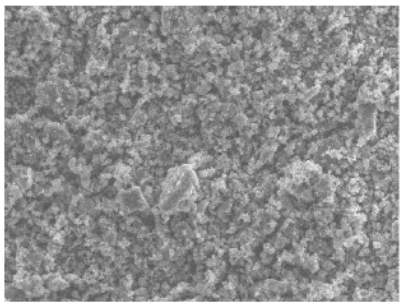

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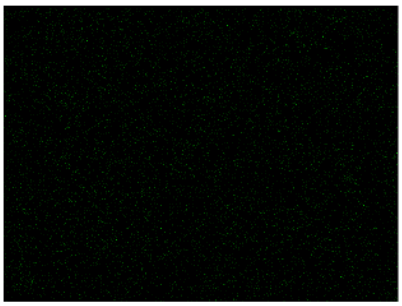
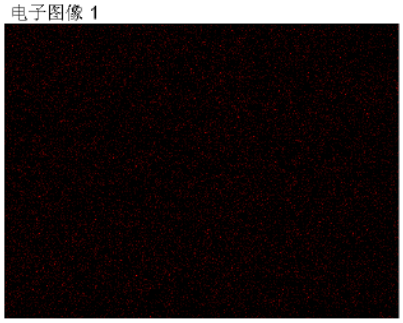


Ni Ka1



Pt La1

01 EDS elemental mapping of Ni, Pt in x = 0.09 sample

EMAX

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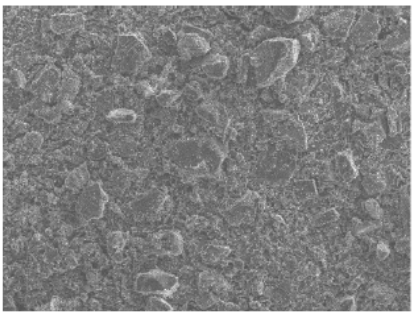
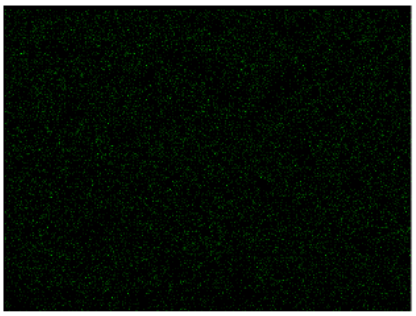
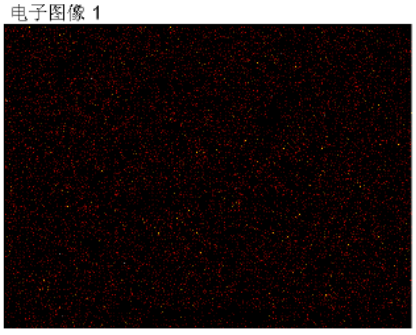


电子图像 1



Ni Ka1



Pt La1

注释：02 EDS elemental mapping of Ni, Pt in x = 0.54 sample

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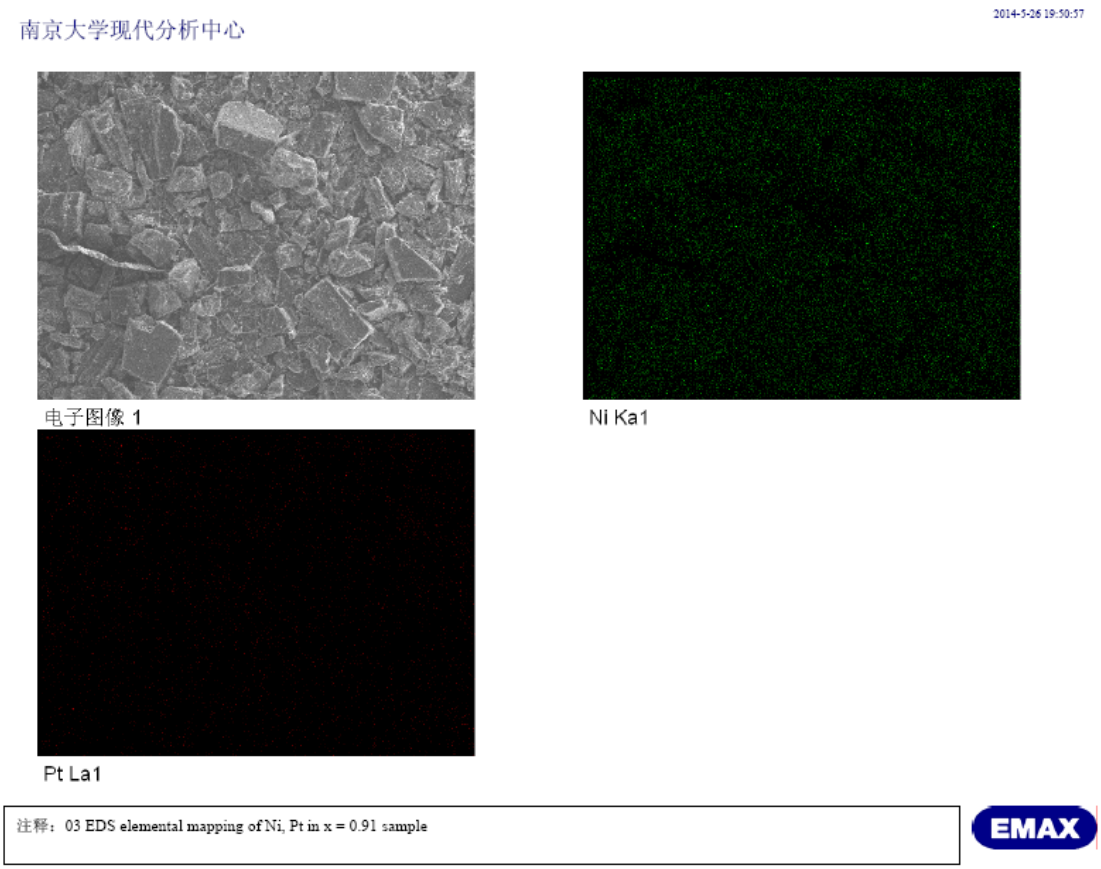


Figure S2 EDS mappings of Ni and Pt in **[Cl-BzPy][$Ni_xPt_{1-x}(mnt)_2$]** (x = 0.09, 0.54 and 0.91).

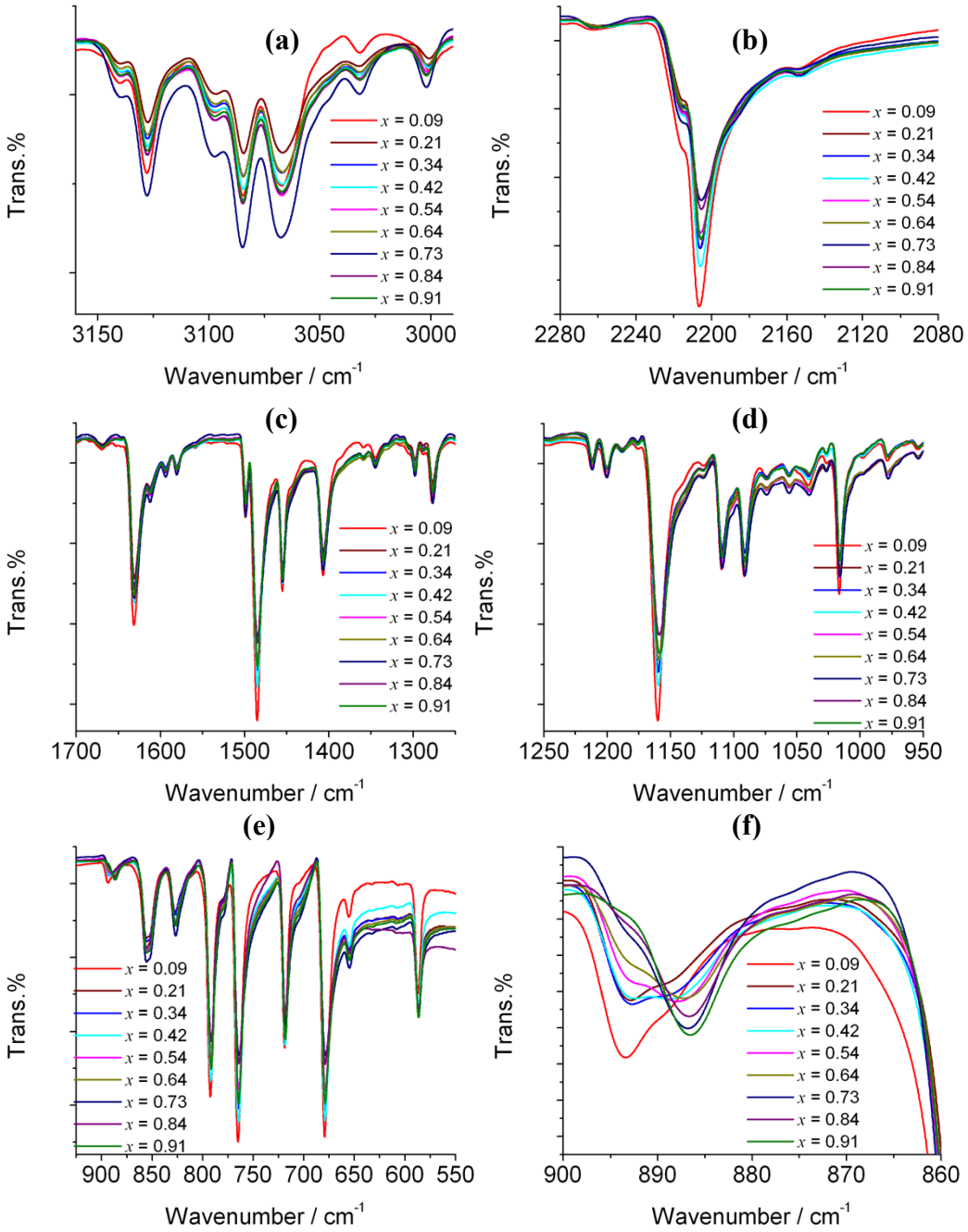


Figure S3 IR spectra for alloy compounds **[Cl-BzPy][$Ni_xPt_{1-x}(mnt)_2$]** (x = 0.09-0.91) in the range 3190-550 cm^{-1}.

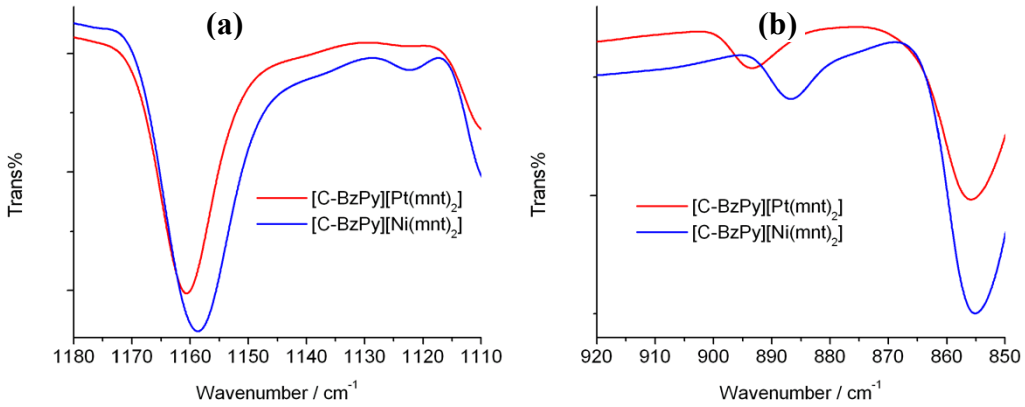
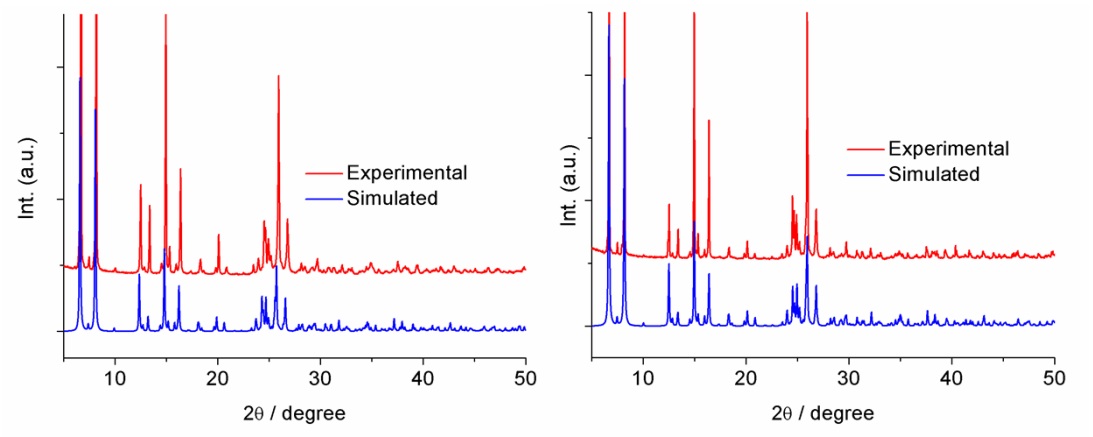


Figure S4 IR spectra of [Cl-BzPy][Ni(mnt)$_2$] and [Cl-BzPy][Pt(mnt)$_2$] (a) the band of $\nu_{C\text{-}S}$+$\nu_{C\text{-}C}$ and $\pi_{C\text{-}CN}$ (b) the band of $\nu_{C\text{-}S}$ from mnt^{2-} ligand.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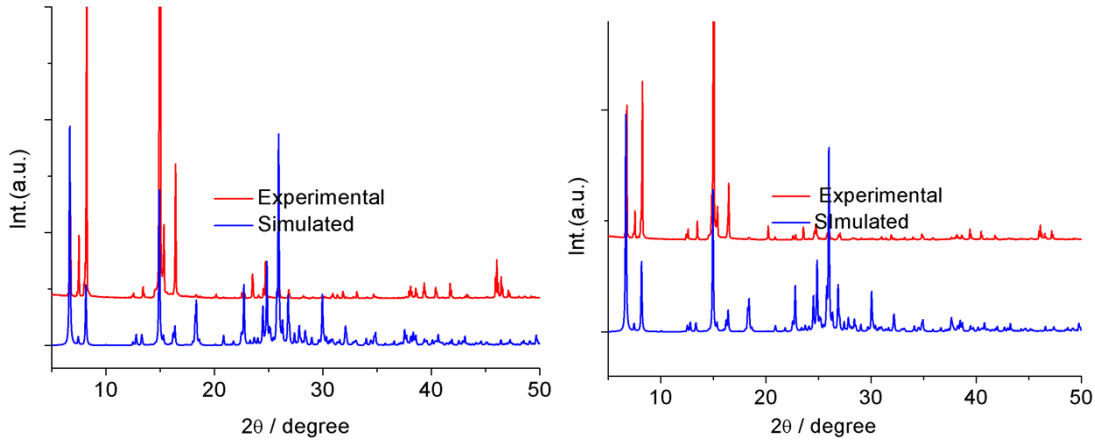
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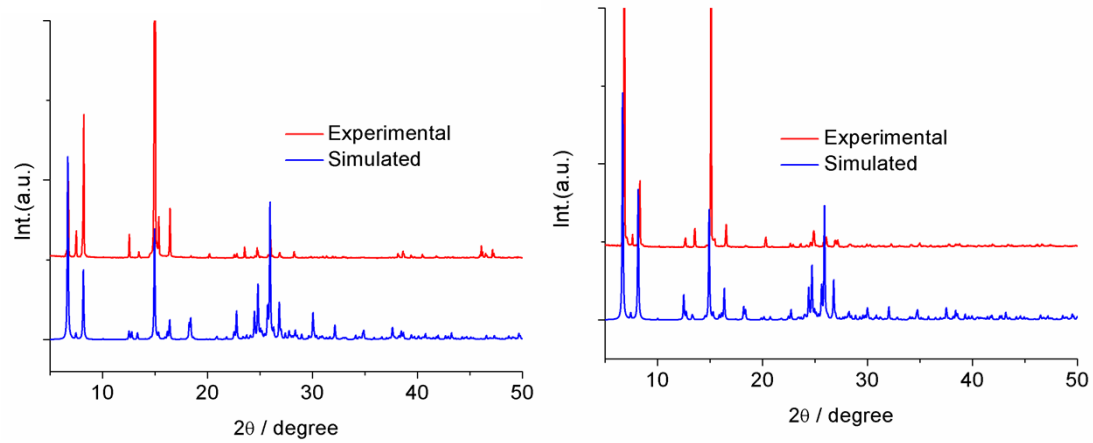
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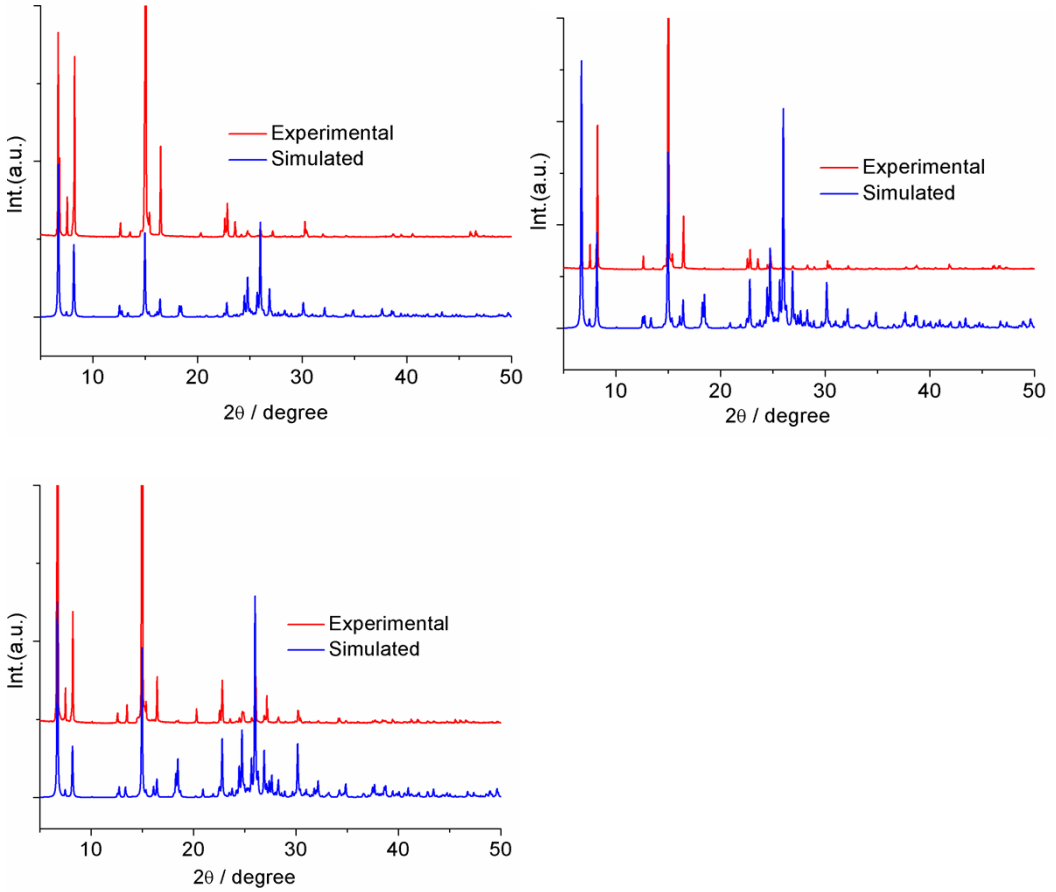

Figure S5 Simulated and Experimental Powder X-ray diffraction profiles for x = (a) 0.09 (b) 0.21 (c) 0.34 (d) 0.42 (e) 0.54 (f) 0.64 (g) 0.73 (h) 0.84 (i) 0.91.

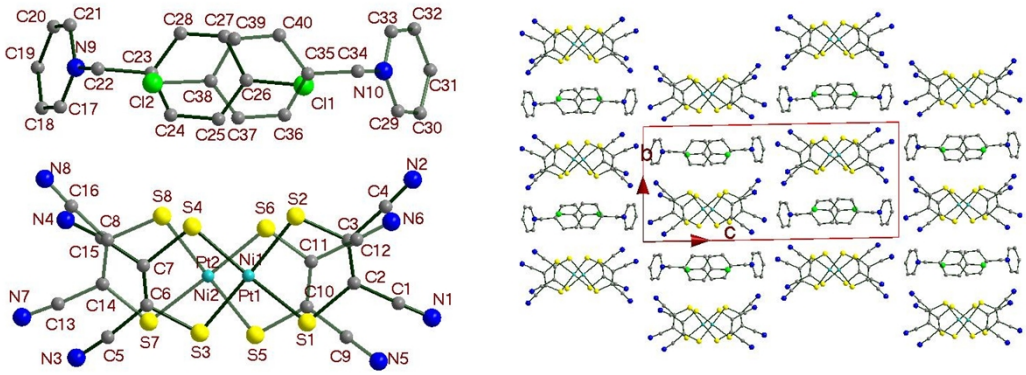

Figure S5 An asymmetric unit with non-H atoms labels and packing structure of **[Cl-BzPy][$Ni_xPt_{1-x}(mnt)_2$]** (x = 0.54) viewed along a-axis at 100 K.