Electronic Supplementary Information (ESI)

for

Magnetic layered perovskites of $[CH_3C(NH_2)_2]_2[M(HCOO)_4]$ ($M = Co^{2+}$ and Ni^{2+}): synthesis, structures and properties

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Table S1. Detailed crystallographic data for **1Co** and **2Ni** at 10 temperatures.

Compound	**1Co**				
T, K	100	140	180	220	260
formula	$C_8H_{18}CoN_4O_8$	$C_8H_{18}CoN_4O_8$	$C_8H_{18}CoN_4O_8$	$C_8H_{18}CoN_4O_8$	$C_8H_{18}CoN_4O_8$
fw	357.19	357.19	357.19	357.19	357.19
crystal system	orthorhombic	orthorhombic	orthorhombic	orthorhombic	orthorhombic
space group	*P ccn*	*P ccn*	*P ccn*	*P ccn*	*P ccn*
a, Å	7.8265(2)	7.8332(1)	7.8422(2)	7.8547(2)	7.8675(2)
b, Å	22.0990(6)	22.0928(5)	22.0907(6)	22.0887(6)	22.0901(6)
c, Å	8.5484(2)	8.5556(2)	8.5634(2)	8.5717(2)	8.5777(2)
α, °	90	90	90	90	90
β, °	90	90	90	90	90
γ, °	90	90	90	90	90
V, Å^3	1478.51(7)	1480.61(5)	1483.52(7)	1487.19(7)	1490.75(7)
Z	4	4	4	4	4
D_c, g/cm^3	1.605	1.602	1.599	1.595	1.592
F(000)	740	740	740	740	740
μ (Cu K_α), mm^{-1}	9.538	9.524	9.506	9.482	9.460
Crystal size, mm^3	0.174×0.156 ×0.093	0.174×0.156 ×0.093	0.174×0.156 ×0.093	0.174×0.156 ×0.093	0.174×0.156 ×0.093
T_{min} and T_{max}	0.316, 0.558	0.317, 0.559	0.314, 0.558	0.319, 0.561	0.320, 0.561
θ_{min}, θ_{max} , °	4.00, 76.62	4.00, 76.39	4.00, 76.60	4.00, 76.45	4.00, 76.41
no. total reflns.	20755	20810	20877	20949	21004
no. uniq. reflns (R_{int})	1539 (0.0379)	1538 (0.0392)	1544 (0.0392)	1544 (0.0395)	1547 (0.0403)
no. obs. [I≥2σ(I)]	1389	1360	1366	1340	1316
no. params	99	99	99	99	99
[a]$R1$, [b]$wR2$ [I≥2σ(I)]	0.0268, 0.0713	0.0258, 0.0680	0.0262, 0.0677	0.0256, 0.0667	0.0257, 0.0681
[a]$R1$, [b]$wR2$ (all data)	0.0304, 0.0740	0.0302, 0.0712	0.0311, 0.0706	0.0309, 0.0704	0.0323, 0.0726
GOF	0.998	1.004	1.003	0.999	1.004
[c]Δρ, e/Å^3	0.271, −0.477	0.256, −0.435	0.253, −0.380	0.230, −0.317	0.228, −0.307
[d]Max. and mean Δ/σ	0.000, 0.000	0.000, 0.000	0.000, 0.000	0.000, 0.000	0.000, 0.000

a. $R1 = \sum||F_o|-|F_c||/\sum|F_o|$; *b*. $wR2 = [\sum w(F_o^2-F_c^2)^2/\sum w(F_o^2)^2]^{1/2}$; *c*. Maximum and minimum residual electron density; *d*. Maximum and mean sigma/shift.

Table S1. continued.

Compound	**1Co**				
T, K	290	320	360	400	440
formula	$C_8H_{18}CoN_4O_8$	$C_8H_{18}CoN_4O_8$	$C_8H_{18}CoN_4O_8$	$C_8H_{18}CoN_4O_8$	$C_8H_{18}CoN_4O_8$
fw	357.19	357.19	357.19	357.19	357.19
crystal system	orthorhombic	orthorhombic	orthorhombic	orthorhombic	orthorhombic
space group	*P ccn*	*P ccn*	*P ccn*	*P ccn*	*P ccn*
a, Å	7.8811(2)	7.8926(2)	7.9102(2)	7.9282(2)	7.9479(3)
b, Å	22.0928(6)	22.0966(7)	22.1031(7)	22.1074(8)	22.1124(11)
c, Å	8.5825(2)	8.5875(2)	8.6003(2)	8.6031(3)	8.6127(3)
α, °	90	90	90	90	90
β, °	90	90	90	90	90
γ, °	90	90	90	90	90
V, Å^3	1494.35(7)	1497.66(7)	1503.68(7)	1507.88(8)	1513.66(11)
Z	4	4	4	4	4
D_c, g/cm^3	1.588	1.584	1.578	1.573	1.567
F(000)	740	740	740	740	740
μ (Cu K_α), mm^{-1}	9.437	9.416	9.378	9.352	9.317
Crystal size, mm^3	0.174×0.156 ×0.093	0.174×0.156 ×0.093	0.174×0.156 ×0.093	0.174×0.156 ×0.093	0.174×0.156 ×0.093
T_{min} and T_{max}	0.318, 0.562	0.321, 0.563	0.323, 0.565	0.324, 0.566	0.326, 0.554
θ_{min}, θ_{max} , °	4.00, 76.51	4.00, 76.47	4.00, 76.61	4.00, 76.89	4.00, 76.48
no. total reflns.	21070	21119	21208	21273	10751
no. uniq. reflns (R_{int})	1552 (0.0415)	1556 (0.0422)	1563 (0.0442)	1566 (0.0457)	1550 (0.0453)
no. obs. [I≥2σ(I)]	1306	1297	1275	1235	1022
no. params	99	99	99	99	99
[a]$R1$, [b]$wR2$ [I≥2σ(I)]	0.0271, 0.0720	0.0270, 0.0696	0.0277, 0.0724	0.0285, 0.0724	0.0426, 0.0993
[a]$R1$, [b]$wR2$ (all data)	0.0342, 0.0770	0.0349, 0.0754	0.0365, 0.0781	0.0392, 0.0795	0.0695, 0.1164
GOF	1.003	1.002	0.999	1.002	1.002
[c]Δρ, e/Å^3	0.219, −0.288	0.212, −0.274	0.212, −0.221	0.196, −0.228	0.386, −0.454
[d]Max. and mean Δ/σ	0.000, 0.000	0.000, 0.000	0.001, 0.000	0.000, 0.000	0.000, 0.000

a. $R1 = \sum||F_o|-|F_c||/\sum|F_o|$; *b*. $wR2 = [\sum w(F_o^2-F_c^2)^2/\sum w(F_o^2)^2]^{1/2}$; *c*. Maximum and minimum residual electron density; *d*. Maximum and mean sigma/shift.

Table S1. continued.

Compound	**2Ni**				
T, K	80	140	180	220	260
formula	$C_8H_{18}NiN_4O_8$	$C_8H_{18}NiN_4O_8$	$C_8H_{18}NiN_4O_8$	$C_8H_{18}NiN_4O_8$	$C_8H_{18}NiN_4O_8$
fw	356.97	356.97	356.97	356.97	356.97
crystal system	orthorhombic	orthorhombic	orthorhombic	orthorhombic	orthorhombic
space group	*P ccn*	*P ccn*	*P ccn*	*P ccn*	*P ccn*
a, Å	7.7921(1)	7.7962(1)	7.8046(1)	7.8166(1)	7.8287(1)
b, Å	22.0548(3)	22.0461(3)	22.0464(3)	22.0478(3)	22.0521(3)
c, Å	8.4600(1)	8.4718(1)	8.4805(1)	8.4882(1)	8.4976(1)
α, °	90	90	90	90	90
β, °	90	90	90	90	90
γ, °	90	90	90	90	90
V, Å^3	1453.88(3)	1456.10(3)	1459.18(3)	1462.85(3)	1467.02(3)
Z	4	4	4	4	4
D_c, g/cm^3	1.631	1.628	1.625	1.621	1.616
F(000)	744	744	744	744	744
μ (Cu K_α), mm^{-1}	2.370	2.367	2.362	2.356	2.349
Crystal size, mm^3	0.154×0.080 ×0.0.049	0.154×0.080 ×0.0.049	0.154×0.080 ×0.0.049	0.154×0.080 ×0.0.049	0.154×0.080 ×0.0.049
T_{min} and T_{max}	0.815, 0.974	0.815, 0.974	0.809, 1.000	0.815, 0.974	0.816, 0.975
θ_{min}, θ_{max} , °	4.01, 76.35	4.01, 76.49	4.01, 76.53	4.01, 76.32	4.01, 76.46
no. total reflns.	20889	20948	21004	21056	21139
no. uniq. reflns (R_{int})	1529 (0.0312)	1532 (0.0317)	1537 (0.0330)	1539 (0.0340)	1543 (0.0347)
no. obs. [I≥2σ(I)]	1422	1397	1399	1374	1380
no. params	99	99	99	99	99
[a]R1, [b]wR2 [I≥2σ(I)]	0.0240, 0.0618	0.0236, 0.0611	0.0255, 0.0671	0.0253, 0.0669	0.0268, 0.0705
[a]R1, [b]wR2 (all data)	0.0263, 0.0635	0.0267, 0.0633	0.0289, 0.0694	0.0290, 0.0700	0.0307, 0.0741
GOF	1.003	1.000	1.002	0.997	1.004
[c]Δρ, e/Å^3	0.293, −0.272	0.249, −0.236	0.247, −0.252	0.235, −0.225	0.241, −0.239
[d]Max. and mean Δ/σ	0.000, 0.000	0.000, 0.000	0.000, 0.000	0.000, 0.000	0.000, 0.000

a. $R1 = \sum||F_o|-|F_c||/\sum|F_o|$; *b*. $wR2 = [\sum w(F_o^2-F_c^2)^2/\sum w(F_o^2)^2]^{1/2}$; *c*. Maximum and minimum residual electron density; *d*. Maximum and mean sigma/shift.

Table S1. continued.

Compound	**2Ni**				
T, K	290	320	360	400	440
formula	$C_8H_{18}NiN_4O_8$	$C_8H_{18}NiN_4O_8$	$C_8H_{18}NiN_4O_8$	$C_8H_{18}NiN_4O_8$	$C_8H_{18}NiN_4O_8$
fw	356.97	356.97	356.97	356.97	356.97
crystal system	orthorhombic	orthorhombic	orthorhombic	orthorhombic	orthorhombic
space group	*P ccn*	*P ccn*	*P ccn*	*P ccn*	*P ccn*
a, Å	7.8376(2)	7.8494(2)	7.8655(2)	7.8802(2)	7.8983(2)
b, Å	22.0576(4)	22.0641(4)	22.0691(4)	22.0783(4)	22.0866(4)
c, Å	8.5034(2)	8.5078(2)	8.5179(2)	8.5264(2)	8.5326(2)
α, °	90	90	90	90	90
β, °	90	90	90	90	90
γ, °	90	90	90	90	90
V, Å^3	1470.06(6)	1473.47(6)	1478.58(6)	1483.43(6)	1488.48(6)
Z	4	4	4	4	4
D_c, g/cm^3	1.613	1.609	1.604	1.598	1.593
F(000)	744	744	744	744	744
μ (Cu K_α), mm^{-1}	2.344	2.339	2.331	2.323	2.315
Crystal size, mm^3	0.154×0.080 ×0.0.049	0.154×0.080 ×0.0.049	0.154×0.080 ×0.0.049	0.154×0.080 ×0.0.049	0.154×0.080 ×0.0.049
T_{min} and T_{max}	0.817, 0.975	0.818, 0.976	0.818, 0.976	0.819, 0.976	0.810, 0.976
θ_{min}, θ_{max} , °	4.01, 76.53	4.01, 76.61	4.01, 76.39	4.00, 76.53	4.00, 76.63
no. total reflns.	21185	21231	21283	21351	21422
no. uniq. reflns (R_{int})	1545 (0.0363)	1550 (0.0373)	1551 (0.0376)	1545 (0.0390)	1561 (0.0435)
no. obs. [I≥2σ(I)]	1378	1335	1321	1313	1270
no. params	99	99	99	99	99
[a]$R1$, [b]$wR2$ [I≥2σ(I)]	0.0278, 0.0738	0.0282, 0.0748	0.0289, 0.0764	0.0298, 0.0777	0.0320, 0.0817
[a]$R1$, [b]$wR2$ (all data)	0.0319, 0.0775	0.0340, 0.0796	0.0352, 0.0822	0.0365, 0.0840	0.0403, 0.0883
GOF	1.004	1.003	1.003	1.003	0.996
[c]Δρ, e/Å^3	0.244, −0.258	0.238, −0.235	0.229, −0.215	0.268, −0.185	0.260, −0.233
[d]Max. and mean Δ/σ	0.000, 0.000	0.000, 0.000	0.000, 0.000	0.000, 0.000	0.000, 0.000

a. $R1 = \sum||F_o|-|F_c||/\sum|F_o|$; *b*. $wR2 = [\sum w(F_o^2-F_c^2)^2/\sum w(F_o^2)^2]^{1/2}$; *c*. Maximum and minimum residual electron density; *d*. Maximum and mean sigma/shift.

Table S2. Selected bond distances (Å) and bond angles (°), the geometries of the N−H···O hydrogen bonds (N···O distances, Å and N−H···O angles, °) between the cation and the anionic framework, and the M···M distances (Å) for the two compounds under the 10 temperatures.

1Co

T, K	100	140	180	220	260
M−O	2.103(1) ×2	2.103(1) ×2	2.104(1) ×2	2.104(1) ×2	2.104(1) ×2
	2.097(1) ×2	2.099(1) ×2	2.101(1) ×2	2.103(1) ×2	2.105(1) ×2
	2.097(1) ×2	2.097(1) ×2	2.098(1) ×2	2.099(1) ×2	2.101(1) ×2
O−C of *anti-anti*	1.252(2)	1.252(2)	1.251(2)	1.249(2)	1.247(2)
formate	1.261(2)	1.261(2)	1.259(2)	1.258(2)	1.256(2)
O−C of *single*	1.257(2)	1.253(2)	1.248(2)	1.244(2)	1.237(2)
anti formate	1.242(2)	1.241(2)	1.239(2)	1.237(2)	1.235(2)
C−N	1.304(2), 1.316(2)	1.303(2), 1.315(2)	1.302(2), 1.313(2)	1.301(2), 1.310(2)	1.298(2), 1.309(3)
C−C	1.496(2)	1.492(2)	1.492(2)	1.488(2)	1.486(3)
cis- O−M−O	85.34(4) − 94.66(4)	85.29(4) − 94.71(4)	85.24(5) − 94.76(5)	85.20(5) − 94.80(5)	85.20(5) − 94.80(4)
trans- O−M−O	180.00	180.00	180.00	180.00	180.00
M−O−C	119.6(1) − 129.3(1)	119.8(1) − 129.2(1)	120.0(1) − 129.3(1)	120.3(1) − 129.4(1)	120.6(1) − 129.6(1)
O−C−O	124.7(1), 125.3(2)	124.7(1), 125.5(2)	124.7(1), 125.6(2)	124.9(2), 125.7(2)	125.0(2), 126.1(2)
N/C−C−N/C	118.9(2) − 121.9(2)	119.0(2) − 122.0(2)	119.0(2) − 121.9(2)	118.9(2) − 121.8(2)	119.0(2) − 121.8(2)
N···O	2.860(2) − 2.879(2)	2.863(2) − 2.880(2)	2.868(2) − 2.884(2)	2.873(2) − 2.891(2)	2.876(2) − 2.901(2)
N−H···O	169.9 − 175.1	169.5 − 174.7	169.2 − 174.3	168.9 − 174.2	169.0 − 174.0
intralayer M···M	5.795	5.800	5.806	5.813	5.820
interlayer M···M	11.722	11.720	11.721	11.722	11.725

T, K	290	320	360	400	440
M−O	2.105(1) ×2	2.104(1) ×2	2.106(1) ×2	2.107(1) ×2	2.106(2) ×2
	2.107(1) ×2	2.110(1) ×2	2.111(1) ×2	2.112(1) ×2	2.108(2) ×2
	2.101(1) ×2	2.103(1) ×2	2.105(1) ×2	2.106(2) ×2	2.106(3) ×2
O−C of *anti-anti*	1.247(2)	1.244(2)	1.241(2)	1.239(2)	1.233(4)
formate	1.253(2)	1.254(2)	1.252(2)	1.251(2)	1.250(4)
O−C of *single*	1.236(2)	1.232(2)	1.228(3)	1.220(3)	1.219(5)
anti formate	1.232(3)	1.231(3)	1.227(3)	1.227(3)	1.226(5)
C−N	1.296(3), 1.307(3)	1.294(3), 1.307(3)	1.295(3), 1.304(3)	1.292(3), 1.303(3)	1.291(5), 1.299(5)
C−C	1.485(3)	1.484(3)	1.483(3)	1.483(4)	1.478(6)
cis- O−M−O	85.14(5) − 94.86(5)	85.09(6) − 94.91(6)	85.05(6) − 94.95(6)	85.09(6) − 94.91(6)	85.1(1) − 94.9(1)
trans- O−M−O	180.00	180.00	180.00	180.00	180.00
M−O−C	120.9(1) − 129.7(1)	121.1(1) − 129.9(1)	121.5(1) − 130.1(2)	121.8(1) − 130.4(2)	122.5(2) − 130.8(3)
O−C−O	125.1(2), 126.1(2)	125.1(2), 126.4(2)	125.4(2), 126.6(2)	125.4(2), 126.9(2)	126.3(3), 127.2(4)
N/C−C−N/C	119.0(2) − 121.9(2)	118.8(2) − 121.9 (2)	118.9(2) − 121.9 (2)	118.8(2) − 122.1(2)	119.0(4) − 121.6(4)
N···O	2.882(3) − 2.906(2)	2.882(3) − 2.911(2)	2.887(3) − 2.924(2)	2.886(3) − 2.933(2)	2.887(5) − 2.954(4)
N−H···O	168.8 − 173.9	168.7 − 173.9	168.7 − 173.8	168.1 − 173.5	168.8 − 173.3
intralayer M···M	5.826	5.832	5.842	5.850	5.860
interlayer M···M	11.728	11.732	11.738	11.743	11.749

Table S2. continued.

2Ni

T, K	80	140	180	220	260
M−O	2.0558(9) ×2	2.0571(9) ×2	2.0585(9) ×2	2.0598(9) ×2	2.0610(9) ×2
	2.0686(9) ×2	2.0699(9) ×2	2.073(1) ×2	2.075(1) ×2	2.077(1) ×2
	2.0637(9) ×2	2.0644(9) ×2	2.065(1) ×2	2.067(1) ×2	2.069(1) ×2
O−C of *anti-anti*	1.254(2)	1.251(2)	1.251(2)	1.249(2)	1.247(2)
formate	1.262(2)	1.261(2)	1.259(2)	1.259(2)	1.258(2)
O−C of *single*	1.256(2)	1.251(2)	1.247(2)	1.244(2)	1.237(2)
anti formate	1.246(2)	1.243(2)	1.240(2)	1.237(2)	1.237(2)
C−N	1.309(2), 1.315(2)	1.306(2), 1.313(2)	1.303(2), 1.312(2)	1.302(2), 1.311(2)	1.300(2), 1.308(2)
C−C	1.494(2)	1.490(2)	1.490(2)	1.488(2)	1.486(2)
cis- O−M−O	85.02(4) − 94.98(4)	84.90(4) − 95.10(4)	84.90(4) − 95.10(4)	84.85(4) − 95.15(4)	84.86(5) − 95.14(5)
trans- O−M−O	180.00	180.00	180.00	180.00	180.00
M−O−C	119.8 (1) − 129.0(1)	120.2(1) − 129.0(1)	120.3(1) − 129.1(1)	120.6(1) − 129.2(1)	120.9(1) − 129.3(1)
O−C−O	124.2(1), 125.3(1)	124.3(1), 125.4(1)	124.4(1), 125.7(1)	124.4(1), 125.7(2)	124.7(1), 125.9(2)
N/C−C−N/C	118.8(1) − 121.8(1)	118.8(1) − 121.8(1)	118.8(1) − 121.9(1)	118.8(1) − 121.9(1)	118.8(2) − 121.9(2)
N···O	2.861(2) − 2.876(2)	2.868(2) − 2.876(2)	2.872(2) − 2.884(2)	2.876(2) − 2.892(2)	2.879(2) − 2.900(2)
N−H···O	170.1 − 175.1	170.5 − 174.5	170.0 − 174.3	170.0 − 174.1	169.7 − 174.0
intralayer M···M	5.751	5.757	5.763	5.770	5.777
interlayer M···M	11.695	11.692	11.694	11.696	11.700

T, K	290	320	360	400	440
M−O	2.062(1) ×2	2.063(1) ×2	2.066(1) ×2	2.066(1) ×2	2.068(1) ×2
	2.078(1) ×2	2.078(1) ×2	2.081(1) ×2	2.084(1) ×2	2.084(1) ×2
	2.069(1) ×2	2.070(1) ×2	2.072(1) ×2	2.072(1) ×2	2.074(1) ×2
O−C of *anti-anti*	1.246(2)	1.244(2)	1.243(2)	1.242(2)	1.240(2)
formate	1.257(2)	1.256(2)	1.254(2)	1.251(2)	1.252(2)
O−C of *single*	1.235(2)	1.232(2)	1.227(2)	1.223(2)	1.219(3)
anti formate	1.235(2)	1.232(2)	1.231(2)	1.229(3)	1.225(3)
C−N	1.298(2), 1.307(2)	1.296(2), 1.308(3)	1.295(2), 1.306(3)	1.293(3), 1.305(3)	1.292(3), 1.302(3)
C−C	1.486(3)	1.483(3)	1.483(3)	1.483(3)	1.480(3)
cis- O−M−O	84.83(5) − 95.17(5)	84.82(5) − 95.18(5)	84.81(5) − 95.19(5)	84.79(6) − 95.21(6)	84.83(6) − 95.17(6)
trans- O−M−O	180.00	180.00	180.00	180.00	180.00
M−O−C	121.10(9) − 129.5(1)	121.3(1) − 129.6(1)	121.6(1) − 129.8(1)	121.9(1) − 130.0(1)	122.2(1) − 130.3(2)
O−C−O	124.7(1), 126.1(2)	124.9(2), 126.2(2)	124.8(2), 126.6(2)	125.0(2), 126.8(2)	125.2(2), 127.0(2)
N/C−C−N/C	118.7(2) − 122.0(2)	118.8(2) − 121.9(2)	118.7(2) − 121.9(2)	118.7(2) − 121.9(2)	118.8(2) − 121.9(2)
N···O	2.881(2) − 2.906(2)	2.880(2) − 2.913(2)	2.885(3) − 2.922(2)	2.886(3) − 2.932(2)	2.888(3) − 2.943(2)
N−H···O	169.5 − 173.8	169.6 − 173.7	169.4 − 173.6	169.4 − 173.6	169.5 − 173.6
intralayer M···M	5.782	5.788	5.797	5.805	5.814
interlayer M···M	11.704	11.709	11.714	11.721	11.728

Table S3. IR absorption bands (cm^{-1}) and their assignments for **1Co** and **2Ni** (s: strong, m: medium, w: weak, vw: very weak, sh: shoulder).

Assignment	**1Co**	**2Ni**
N−H, stretching	3269m, 3154m, 3110m, 3048sh	3265m, 3153m, 3104m, 3048sh
$2\nu_4$	2925sh	2925sh
C−H stretching, ν_1	2879m, 2830m	2884m, 2833m
$2\nu_5$	2714w	2712w
N−H bending	1722m	1725m
COO, stretching, antisym., ν_4	1589s	1585s
ν(NCN)	1527m	1528m
$\delta(CH_3)$	1428w	1428w
COO deformation, antisym., ν_5	1392s	1389s
COO stretching, sym., ν_2	1361s	1365s
ν(CCN), antisym.	1162w	1163w
C−H, out of plane deformation, ν_6	1075vw, 1062vw	1076vw, 1061vw
ν(CCN), sym.	877sh,w	877sh,w
COO deformation, sym., ν_3	805m, 785m	811m, 788m
$\rho_t(NH_2)$	761m	762m

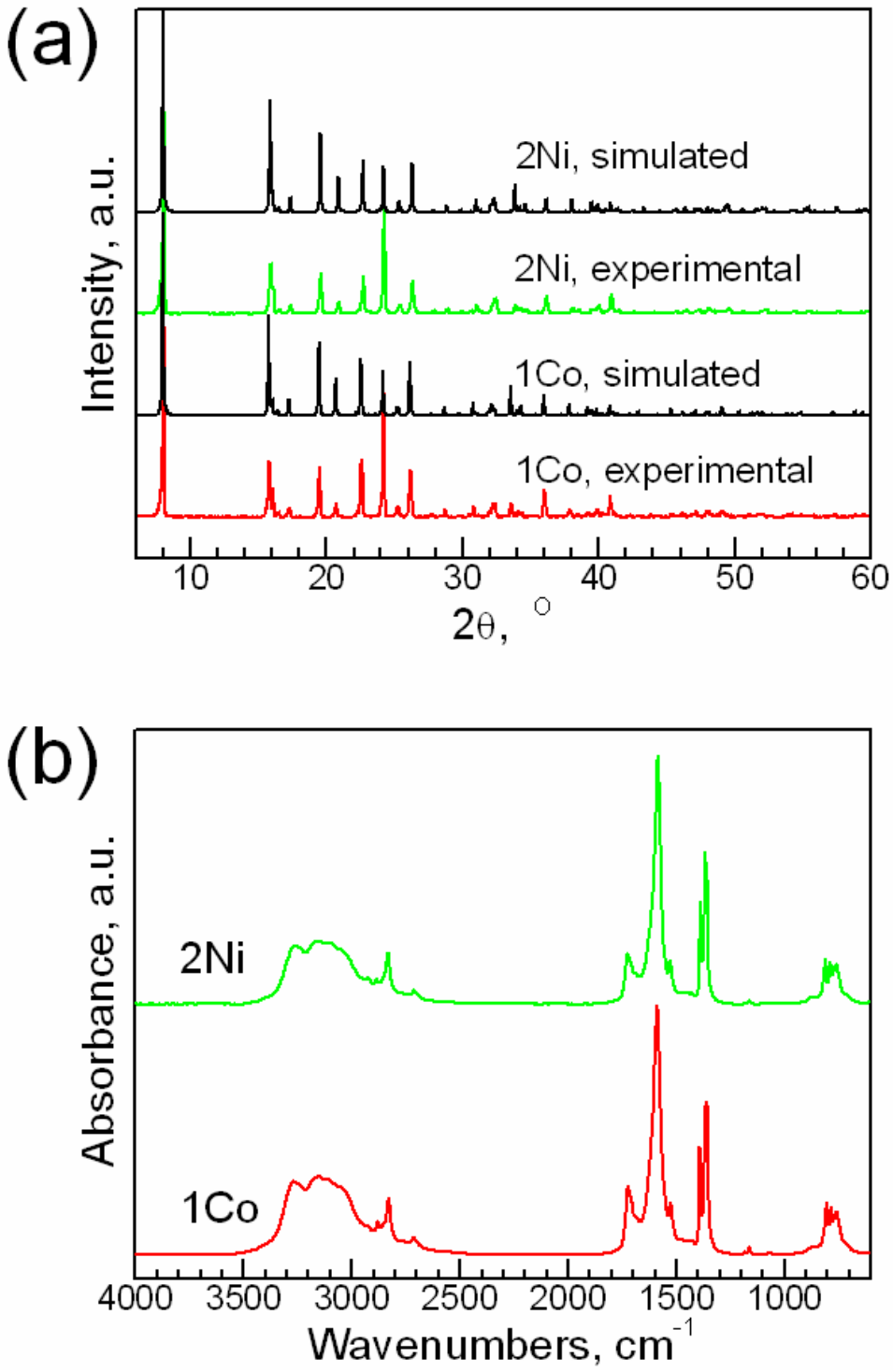

Fig. S1. (a) The PXRD patterns for the as-prepared bulk samples of the two compounds and the simulated one based on their single crystal structures at room temperature. (b) IR spectra of the two compounds.

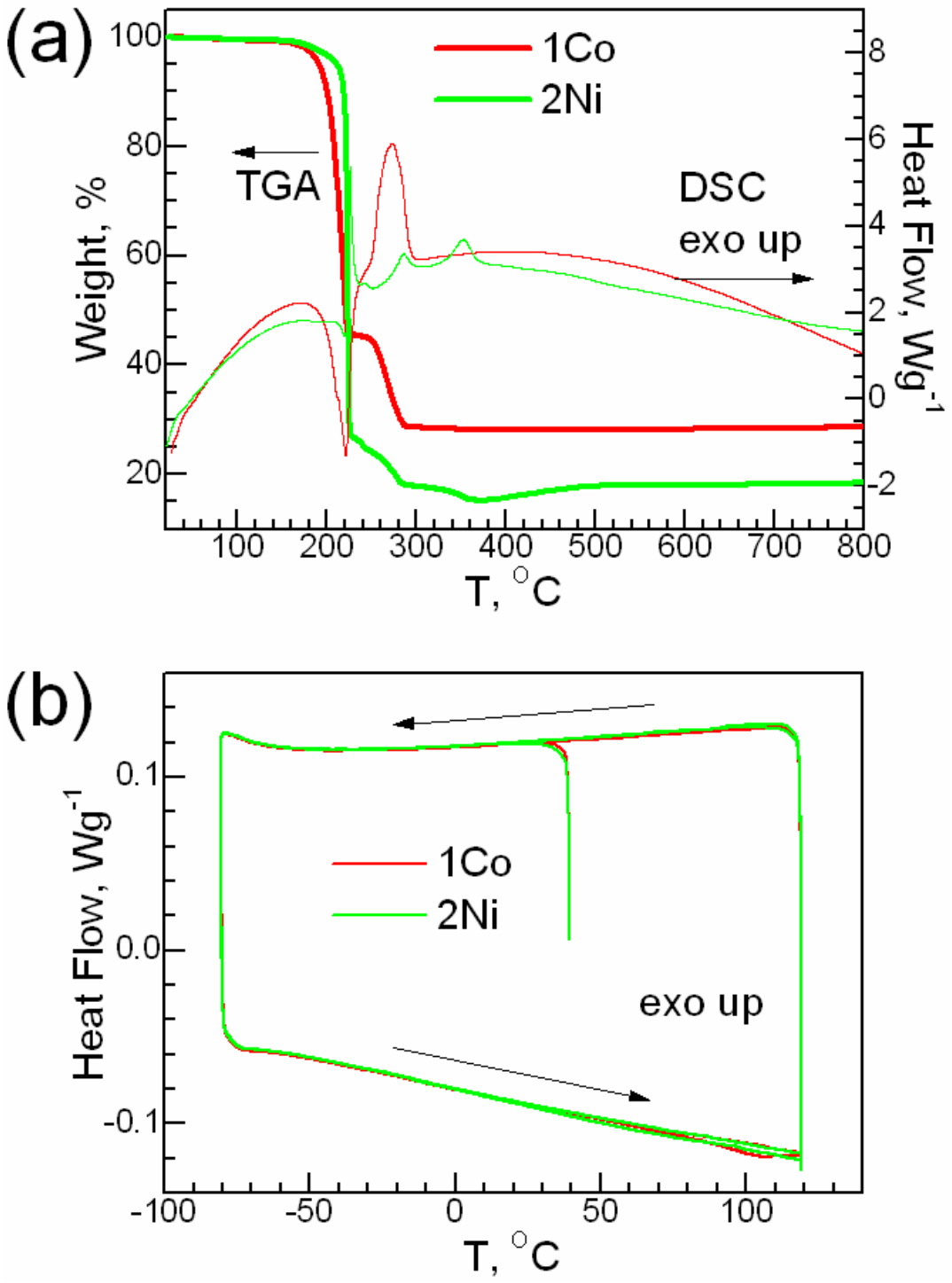

Fig. S2. (a) The combined TGA-DSC runs and (b) the DSC traces (two cycles) for the two compounds.

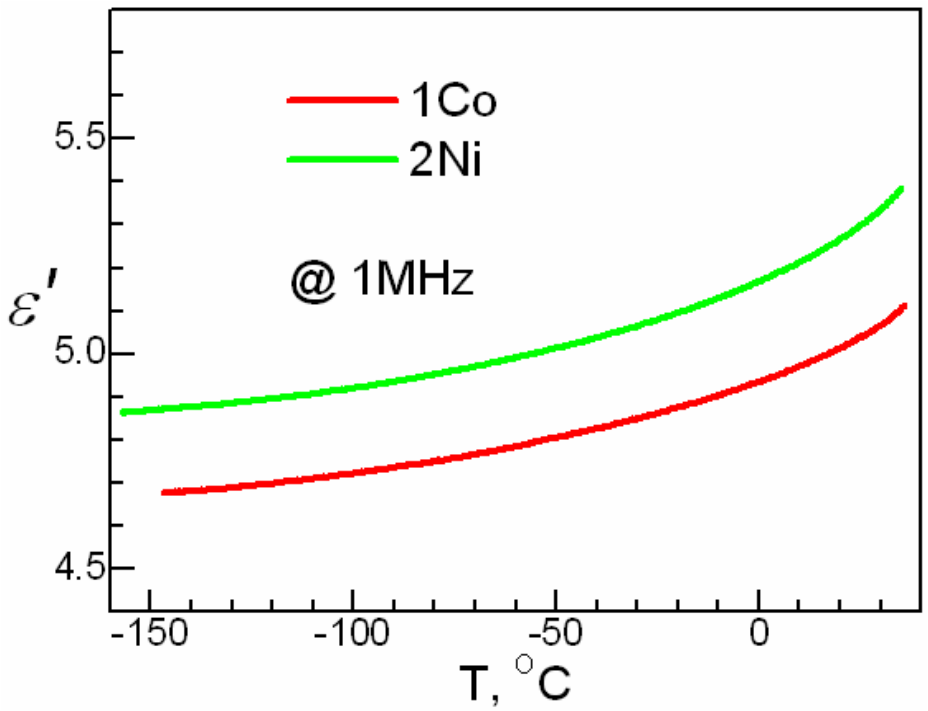

Fig. S3. The temperature-dependent traces of the dielectric constants (*ε' vs T*, at 1 MHz) on cooling for the two compounds.

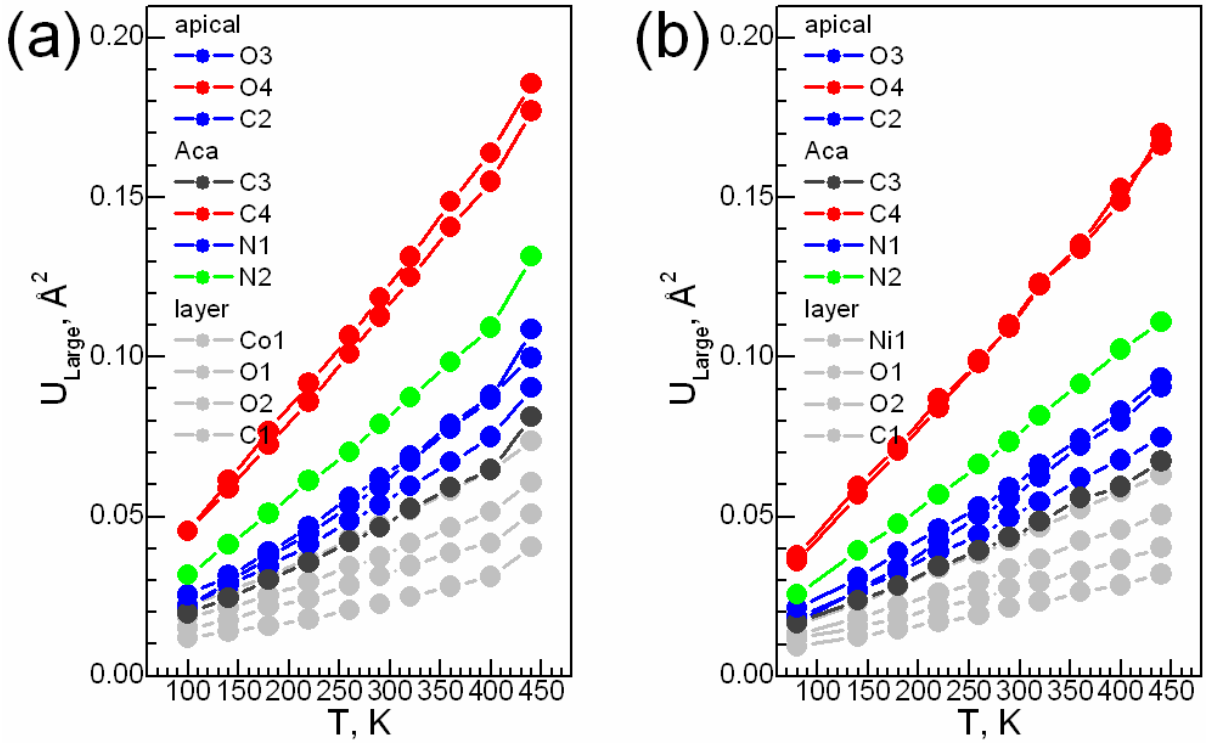

Fig. S4. The temperature-dependent plots of the largest principal mean square atomic displacements U of the non-H atoms in the two structures, (a) **1Co** and (b) **2Ni**.

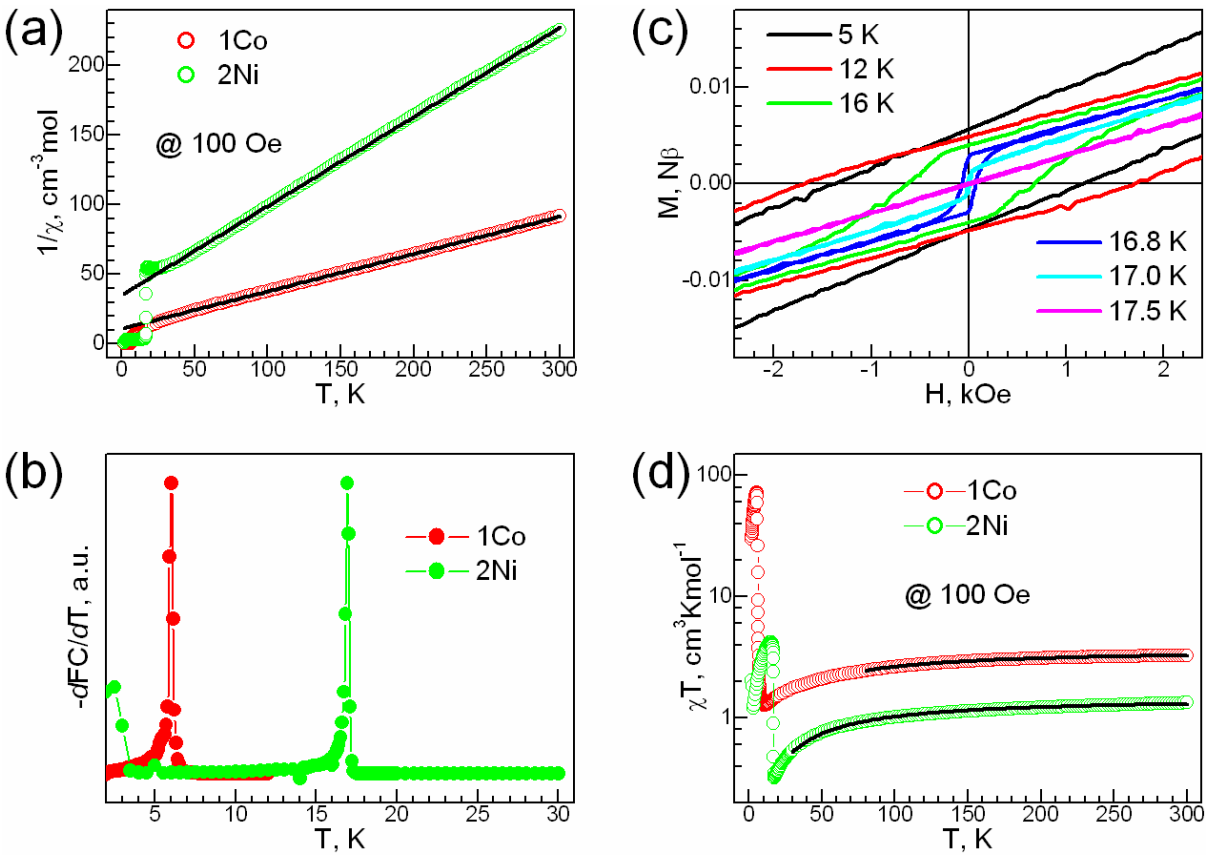

Fig. S5. Additional magnetic data for **1Co** and (b) **2Ni**. (a) The $1/\chi$ *vs T* plots at 100 Oe, with the Curie-Weiss fittings in black lines. (b) The normalized $-d\mathrm{FC}/dT$ *vs T* traces under 10 Oe. (c) The hysteresis loops in low field region for **2Ni** under several temperatures above 5 K. (d) Plots of χT *vs T* under 100 Oe, with the solid lines the best fit using Lines' quadratic-layer antiferromagnet model (see text).