Supplementary information

3.3. NMR Spectra Analyses

The ^{1}H-NMR spectrum shows (Figure 1) six peaks, corresponded to the cyclohexyl ring protons of the primary ammonium cation. These peaks at (δ 2.90, 1.83, 1.66, 1.51, 1.24-1.17, and 1.09 ppm) were in agreement with those reported for the different proton environments in the cyclohexyl ring (Table 1) [1] . Due to the fast proton exchange with the deuterons of the solvent (H/D exchange), the peak of ammonium protons was not detected.

The ^{13}C-NMR spectrum, in deuterated methanol, exhibited four peaks, corresponded to four different C-atom electronic environments of the cyclohexylammonium cation, at δ 51.45, 31.85, 25.82, and 25.29 ppm. The ^{13}C-NMR analysis confirmed the absence of isothiocyanate carbon peak, as shown in Figure 2 and Table 2. The absence of isothiocyanate carbon peak was reported for many organic compounds in the Sigma-Aldrich spectral database. This phenomenon was due to the quadrupolar broadening effect by ^{14}N [2] .

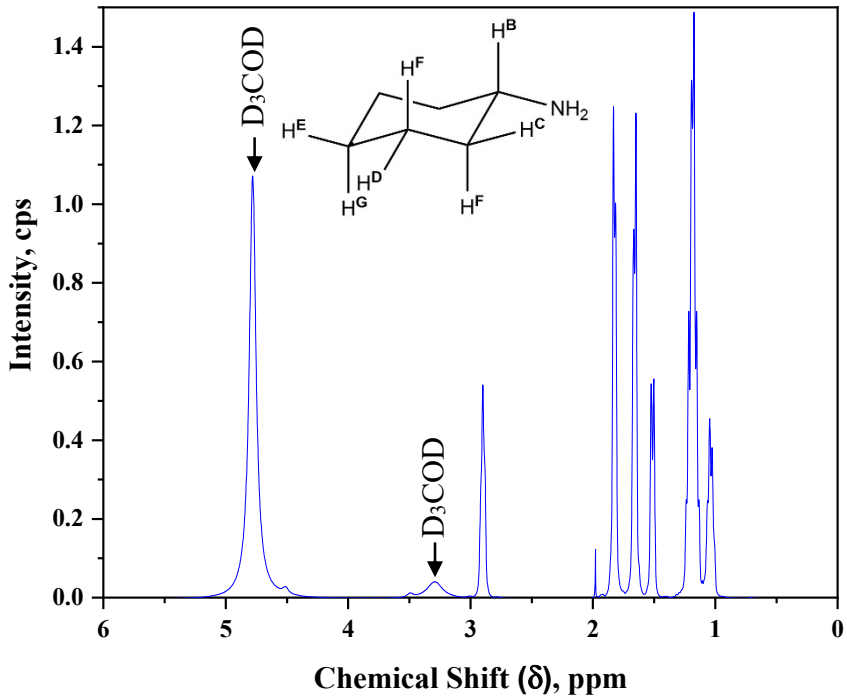

Figure S1. ^{1}H-NMR spectrum of $(C_6H_{11}NH_3)_4[Ni(NCS)_6]\cdot 2H_2O$ in methanol-d^4.

Table S1. ^{1}H-NMR of $(C_6H_{11}NH_3)_4[Ni(NCS)_6]\cdot 2H_2O$ at room temperature.

Hydrogen Atom Label	Chemical Shift (δ), ppm	
	Sample*	Reference# [1]
B	2.90	3.16
C	1.83	1.99
D	1.66	1.80
E	1.51	1.65
F	1.24–1.17	1.41–1.28
G	1.09	1.18

*in D_3COH; #in D_2O

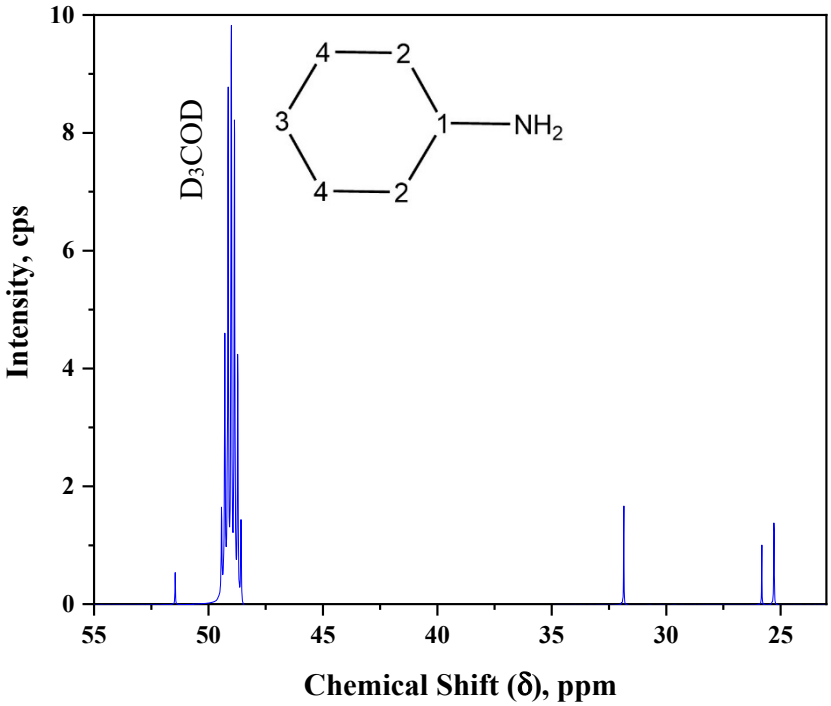

Figure S2. ^{13}C-NMR spectrum of $(C_6H_{11}NH_3)_4[Ni(NCS)_6]\cdot 2H_2O$ in methanol-d^4.

Table S2. ^{13}C-NMR of $(C_6H_{11}NH_3)_4[Ni(NCS)_6].2H_2O$ at room temperature.

Carbon Atom Label	Chemical Shift (δ), ppm	
	Sample*	Reference# [1]
1	51.45	51.25
2	31.85	31.16
3	25.82	25.14
4	25.29	24.65

*in D_3COH; #in D_2O

X-ray single crystal refinement

Table S3. Atomic coordinates and isotropic displacement parameters (in Å ^{2}) at 293K

Atom	**Wyck.**	**x**	**y**	**z**	**U**
Ni1	2*b*	1/2	1/2	1/2	
N1	4*e*	0.53658	0.30880	0.51275	
N2	4*e*	0.44986	0.46839	0.62033	
N3	4*e*	0.37397	0.45802	0.39675	
N4	4*e*	0.33841	1.18836	0.46720	
N5	4*e*	0.36148	0.72276	0.64339	
S1	4*e*	0.59085	0.08699	0.62352	
S2	4*e*	0.40879	0.28846	0.74309	
S3	4*e*	0.21257	0.50285	0.24277	
O1	4*e*	0.39125	0.99001	0.63085	
C1	4*e*	0.56000	0.21699	0.55840	
C2	4*e*	0.43268	0.39489	0.67139	
C3	4*e*	0.30703	0.47913	0.33269	
C4	4*e*	0.24090	1.17699	0.43440	
C5	4*e*	0.19746	1.21988	0.49624	
C6	4*e*	0.09706	1.19802	0.46481	
C7	4*e*	0.05372	1.15666	0.36515	
C8	4*e*	0.09770	1.11294	0.30616	
C9	4*e*	0.19851	1.13251	0.33639	
C10	4*e*	0.26264	0.71089	0.59908	
C11	4*e*	0.23139	0.75207	0.48904	
C12	4*e*	0.13048	0.74104	0.44354	
C13	4*e*	0.09716	0.61471	0.45988	
C14	4*e*	0.13086	0.57442	0.56773	
C15	4*e*	0.23172	0.58255	0.61018	
H1n5	4*e*	0.37943	0.69372	0.70553	0.0587
H2n5	4*e*	0.38645	0.67992	0.60662	0.0587
H3n5	4*e*	0.37654	0.80161	0.64400	0.0587
H1n4	4*e*	0.35932	1.13799	0.43107	0.0734
H2n4	4*e*	0.35285	1.26556	0.45860	0.0734
H3n4	4*e*	0.36110	1.16831	0.53146	0.0734
H1c4	4*e*	0.20817	1.11173	0.45445	0.1503
H1c5	4*e*	0.20996	1.30787	0.50928	0.1967
H2c5	4*e*	0.22591	1.18801	0.56405	0.1967
H1c6	4*e*	0.06836	1.27295	0.47727	0.1997
H2c6	4*e*	0.08452	1.14103	0.51205	0.1997
H1c8	4*e*	0.06925	1.14181	0.23742	0.2346
H2c8	4*e*	0.08404	1.02534	0.29243	0.2346
H1c9	4*e*	0.22642	1.05551	0.32666	0.1774
H2c9	4*e*	0.21122	1.18753	0.28803	0.1774
H1c10	4*e*	0.23645	0.76536	0.63666	0.0688
H1c11	4*e*	0.25886	0.70019	0.45109	0.1027
H2c11	4*e*	0.24888	0.83789	0.48482	0.1027
H1c12	4*e*	0.11204	0.75865	0.37167	0.1589
H2c12	4*e*	0.10293	0.80423	0.47274	0.1589
H1c13	4*e*	0.03284	0.61509	0.43624	0.1251
H2c13	4*e*	0.11406	0.55386	0.41845	0.1251
H1c14	4*e*	0.11239	0.48934	0.57299	0.1335
H2c14	4*e*	0.10539	0.62686	0.60735	0.1335
H1c15	4*e*	0.25180	0.55963	0.68111	0.1114
H2c15	4*e*	0.25727	0.52417	0.57473	0.1114
H1c7	4*e*	0.01143	1.21922	0.32857	0.2707
H2c7	4*e*	0.00692	1.09885	0.36461	0.2707
H2o1	4*e*	0.44573	1.00695	0.66388	0.0871
H1o1	4*e*	0.36703	0.97653	0.67659	0.0871

Table S4. Atomic coordinates and isotropic displacement parameters (in Å ^{2}) at 120K

Atom	Wyck.	Occ.	x	y	z	U
Ni1	2*b*	1	1/2	1/2	1/2	0.01465(9)
N1	4*e*	1	0.53600(8)	0.30622(10)	0.51117(9)	0.0191(4)
N2	4*e*	1	0.44970(9)	0.46714(12)	0.62102(9)	0.0250(4)
N3	4*e*	1	0.37322(8)	0.45878(12)	0.39791(10)	0.0253(4)
N4	4*e*	1	0.33545(8)	1.19450(12)	0.47487(11)	0.0284(4)
N5	4*e*	1	0.36212(7)	0.72287(10)	0.64726(9)	0.0199(4)
S1	4*e*	1	0.59313(2)	0.07899(3)	0.62162(2)	0.01988(11)
S2	4*e*	1	0.40741(2)	0.28245(3)	0.74631(3)	0.02125(12)
S3	4*e*	1	0.20928(3)	0.50369(4)	0.24497(4)	0.04281(17)
O1	4*e*	1	0.39426(8)	0.98799(10)	0.63285(9)	0.0313(4)
C1	4*e*	1	0.55992(8)	0.21187(11)	0.55620(9)	0.0145(4)
C2	4*e*	1	0.43231(8)	0.39104(12)	0.67429(9)	0.0170(4)
C3	4*e*	1	0.30523(11)	0.47915(13)	0.33384(12)	0.0234(5)
C4	4*e*	0.5	0.23204(19)	1.2154(3)	0.4312(3)	0.0259(11)
C4'	4*e*	0.5	0.24060(19)	1.1530(4)	0.4557(2)	0.0257(10)
C5'	4*e*	0.5	0.1916(3)	1.2443(5)	0.4987(4)	0.0384(15)
C5	4*e*	0.5	0.1953(4)	1.1721(7)	0.5137(4)	0.064(2)
C6	4*e*	1	0.09225(12)	1.1987(3)	0.47528(17)	0.0694(10)
C7	4*e*	0.5	0.0478(2)	1.1914(5)	0.3590(3)	0.0490(15)
C7'	4*e*	0.5	0.0508(3)	1.1148(5)	0.3812(5)	0.074(2)
C8'	4*e*	0.5	0.0918(4)	1.1604(9)	0.2976(5)	0.093(4)
C8	4*e*	0.5	0.0980(5)	1.0980(5)	0.3174(5)	0.0477(19)
C9'	4*e*	0.5	0.1994(7)	1.1588(10)	0.3317(7)	0.041(2)
C9	4*e*	0.5	0.1916(8)	1.1228(10)	0.3441(8)	0.042(2)
C10	4*e*	1	0.26203(9)	0.71033(13)	0.60192(11)	0.0217(4)
C11	4*e*	1	0.23233(10)	0.75137(15)	0.49020(12)	0.0299(5)
C12	4*e*	1	0.13001(11)	0.74018(19)	0.44497(15)	0.0455(7)
C13	4*e*	1	0.09806(11)	0.60670(19)	0.45719(14)	0.0431(6)
C14	4*e*	1	0.13082(12)	0.56365(19)	0.56836(15)	0.0468(7)
C15	4*e*	1	0.23306(11)	0.57456(16)	0.61224(13)	0.0381(6)
H1o1	4*e*	1	0.4499(8)	1.008(3)	0.642(2)	0.087(10)
H2o1	4*e*	1	0.3907(19)	1.029(2)	0.6848(14)	0.082(8)
H1n5	4*e*	1	0.37896	0.69519	0.71019	0.0238
H2n5	4*e*	1	0.38822	0.67796	0.61143	0.0238
H3n5	4*e*	1	0.37733	0.80274	0.64652	0.0238
H1n4	4*e*	1	0.34860	1.11795	0.45938	0.0340
H2n4	4*e*	1	0.36278	1.25057	0.44846	0.0340
H3n4	4*e*	1	0.35287	1.20326	0.54123	0.0340
H1c4	4*e*	0.5	0.21922	1.30250	0.40957	0.0311
H1c4'	4*e*	0.5	0.23641	1.07113	0.48542	0.0309
H1c5'	4*e*	0.5	0.19229	1.32682	0.46834	0.0461
H2c5'	4*e*	0.5	0.21978	1.24981	0.57154	0.0461
H1c5	4*e*	0.5	0.22273	1.22046	0.57507	0.0771
H2c5	4*e*	0.5	0.20559	1.08212	0.52494	0.0771
H1c6	4*e*	1	0.08049	1.28074	0.49955	0.0833
H2c6	4*e*	1	0.06244	1.14087	0.50825	0.0833
H1c8	4*e*	0.5	0.07235	1.09457	0.24409	0.0573
H2c8	4*e*	0.5	0.08875	1.01364	0.33973	0.0573
H1c8'	4*e*	0.5	0.07035	1.24477	0.27529	0.1119
H2c8'	4*e*	0.5	0.06798	1.10954	0.23692	0.1119
H1c9	4*e*	0.5	0.22413	1.04360	0.35781	0.0503
H2c9	4*e*	0.5	0.20580	1.15023	0.28429	0.0503
H1c9'	4*e*	0.5	0.23946	1.09170	0.32618	0.0488
H2c9'	4*e*	0.5	0.19476	1.22309	0.28040	0.0488
H1c10	4*e*	1	0.23370	0.76516	0.63866	0.0260
H1c11	4*e*	1	0.26032	0.69749	0.45247	0.0359
H2c11	4*e*	1	0.25006	0.83839	0.48551	0.0359
H1c12	4*e*	1	0.11173	0.76219	0.37351	0.0546
H2c12	4*e*	1	0.10201	0.80005	0.47834	0.0546
H1c13	4*e*	1	0.03345	0.60427	0.43235	0.0517
H2c13	4*e*	1	0.11941	0.54840	0.41610	0.0517
H1c14	4*e*	1	0.11314	0.47655	0.57282	0.0562
H2c14	4*e*	1	0.10346	0.61552	0.60799	0.0562
H1c15	4*e*	1	0.25214	0.55074	0.68335	0.0457
H2c15	4*e*	1	0.26049	0.51804	0.57572	0.0457
H1c7	4*e*	0.5	0.05027	1.27383	0.32927	0.0588
H2c7	4*e*	0.5	-0.01345	1.16367	0.34394	0.0588
H1c7'	4*e*	0.5	-0.01324	1.12706	0.35715	0.0887
H2c7'	4*e*	0.5	0.06706	1.02723	0.39828	0.0887

Table S5. Anisotropic displacement parameters (in Å^2) at 293K

Atom	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Ni1	0.04375	0.03138	0.03823	0.00450	0.01873	0.00683
N1	0.05768	0.04025	0.05012	0.00675	0.02450	0.00790
N2	0.06643	0.05153	0.05325	0.00899	0.03359	0.01107
N3	0.05296	0.04922	0.06000	0.00451	0.01466	0.00189
N4	0.04927	0.05470	0.07881	-0.00038	0.02028	0.00128
N5	0.04917	0.05030	0.04775	-0.00478	0.01661	-0.00049
S1	0.07258	0.03955	0.04570	0.01425	0.01996	0.00998
S2	0.07717	0.04223	0.05446	-0.00630	0.03547	0.00205
S3	0.07123	0.11976	0.07325	0.01187	0.00197	0.03321
O1	0.07463	0.06672	0.07349	-0.00157	0.02056	-0.01157
C1	0.03862	0.03418	0.03629	0.00263	0.01558	-0.00028
C2	0.04512	0.03994	0.03624	0.00343	0.01724	-0.00328
C3	0.05808	0.04623	0.04936	0.00309	0.02277	0.00552
C4	0.04996	0.20088	0.12823	-0.04102	0.03414	-0.06576
C5	0.06077	0.31053	0.11834	-0.00547	0.02673	-0.10671
C6	0.06245	0.30535	0.13970	-0.00691	0.04444	-0.05991
C7	0.04619	0.39358	0.20980	0.00888	0.00585	-0.11205
C8	0.07329	0.33452	0.14005	-0.00151	-0.01634	-0.12579
C9	0.07301	0.25834	0.09231	0.03564	0.00082	-0.08158
C10	0.04580	0.06477	0.06298	-0.00233	0.01997	-0.00275
C11	0.06125	0.10332	0.07385	-0.01622	-0.00217	0.03207
C12	0.06657	0.13789	0.14890	-0.01129	-0.02324	0.05605
C13	0.05201	0.14106	0.10521	-0.02726	0.00671	0.00609
C14	0.06553	0.14926	0.11060	-0.04374	0.01805	0.02163
C15	0.07005	0.09866	0.09322	-0.03101	0.00496	0.03051

Table S6. Anisotropic displacement parameters (in Å^2) at 120K

Atom	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Ni1	0.01924(14)	0.01153(13)	0.01501(14)	0.00183(8)	0.00802(10)	0.00277(7)
N1	0.0216(6)	0.0160(5)	0.0214(6)	0.0018(4)	0.0093(5)	0.0014(4)
N2	0.0330(7)	0.0237(6)	0.0240(6)	0.0063(5)	0.0170(5)	0.0053(5)
N3	0.0245(6)	0.0209(6)	0.0282(6)	0.0018(5)	0.0051(5)	-0.0029(5)
N4	0.0212(6)	0.0233(6)	0.0421(8)	-0.0033(5)	0.0123(6)	-0.0041(5)
N5	0.0207(6)	0.0214(5)	0.0174(5)	-0.0019(4)	0.0059(4)	-0.0013(4)
S1	0.02611(18)	0.01576(16)	0.01744(17)	0.00481(12)	0.00641(13)	0.00433(11)
S2	0.02963(19)	0.01675(16)	0.02051(18)	-0.00238(12)	0.01238(15)	0.00097(11)
S3	0.0323(2)	0.0514(3)	0.0358(3)	0.00615(18)	-0.0019(2)	0.01708(18)
O1	0.0298(6)	0.0307(6)	0.0337(7)	-0.0031(4)	0.0103(5)	-0.0114(4)
C1	0.0151(6)	0.0148(6)	0.0150(6)	-0.0010(4)	0.0068(5)	-0.0018(4)
C2	0.0194(6)	0.0174(6)	0.0150(6)	0.0036(5)	0.0067(5)	-0.0019(5)
C3	0.0291(7)	0.0182(6)	0.0253(7)	0.0014(5)	0.0118(6)	0.0013(5)
C4	0.0181(14)	0.0147(14)	0.047(2)	0.0034(11)	0.0126(13)	0.0110(13)
C4'	0.0185(14)	0.0255(16)	0.0286(16)	-0.0021(13)	0.0008(12)	0.0011(13)
C5'	0.0222(18)	0.065(3)	0.027(2)	-0.005(2)	0.0068(17)	-0.020(2)
C5	0.0205(19)	0.145(7)	0.028(2)	0.003(4)	0.0089(16)	0.007(4)
C6	0.0208(9)	0.142(2)	0.0493(12)	-0.0085(11)	0.0164(9)	-0.0142(13)
C7	0.0198(17)	0.069(3)	0.050(2)	0.004(2)	-0.0001(16)	-0.011(2)
C7'	0.0208(19)	0.036(2)	0.152(6)	-0.0041(18)	0.009(3)	-0.021(3)
C8'	0.025(2)	0.207(10)	0.034(3)	0.019(5)	-0.010(2)	-0.038(5)
C8	0.045(3)	0.044(2)	0.040(3)	-0.008(2)	-0.007(2)	-0.013(2)
C9'	0.020(2)	0.081(6)	0.017(3)	0.001(3)	0.001(2)	-0.012(3)
C9	0.035(4)	0.067(5)	0.027(3)	0.014(3)	0.014(2)	-0.003(3)
C10	0.0165(6)	0.0260(7)	0.0239(7)	-0.0025(5)	0.0085(5)	-0.0028(5)
C11	0.0215(7)	0.0330(8)	0.0301(8)	-0.0041(6)	0.0008(6)	0.0105(6)
C12	0.0232(8)	0.0510(11)	0.0505(12)	-0.0008(7)	-0.0051(8)	0.0123(9)
C13	0.0206(8)	0.0597(11)	0.0432(10)	-0.0144(7)	0.0016(7)	0.0014(8)
C14	0.0297(9)	0.0623(13)	0.0461(11)	-0.0225(8)	0.0087(8)	0.0085(9)
C15	0.0324(8)	0.0407(9)	0.0343(9)	-0.0164(7)	0.0008(7)	0.0117(7)

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[1] "Spectral Database for Organic Compounds SDBS," https://sdbs.db.aist.go.jp/sdbs/cgi-bin/direct_frame_top.cgi (accessed on 30/01/2022).

[2] R. Glaser, R. Hillebrand, W. Wycoff, C. Camasta, and K. S. Gates, "Near-silence of isothiocyanate carbon in 13C NMR spectra: a case study of allyl isothiocyanate," *The Journal of organic chemistry,* vol. 80, no. 9, pp. 4360-4369, 2015. doi.org/10.1021/acs.joc.5b00080