S1. Molar heat capacity of $[Co(NH_3)_6](ClO_4)_3$. M=459.47 g mol^{-1}.

T / K	C_p / J K^{-1} mol^{-1}	T / K	C_p / J K^{-1} mol^{-1}	T / K	C_p / J K^{-1} mol^{-1}
14.753	14.93	65.563	195.44	92.785	270.30
16.330	19.50	66.774	198.69	93.555	272.52
17.892	24.59	67.261	199.58	93.890	273.80
19.460	30.10	67.987	201.65	94.059	274.94
21.003	35.81	69.057	203.95	95.603	281.60
22.515	41.70	69.202	204.78	95.671	282.26
24.011	47.56	70.419	207.83	97.455	293.61
25.491	53.23	70.845	208.34	97.641	296.05
26.979	59.55	71.638	210.78	99.682	283.49
28.456	66.06	72.627	212.56	101.736	274.05
29.915	72.31	72.858	213.64	103.796	273.99
31.353	78.25	74.082	216.43	105.856	276.24
32.770	83.89	74.403	216.79	107.916	280.79
34.166	89.89	75.307	219.34	109.975	287.39
35.540	95.68	76.176	220.96	112.034	287.95
36.895	101.09	76.535	222.37	114.105	285.37
38.233	106.12	77.765	225.43	116.183	287.72
39.556	110.79	77.946	225.45	118.267	290.15
40.866	115.38	78.998	228.25	120.356	292.68
42.163	119.99	79.715	229.35	122.450	295.39
43.449	124.65	80.235	231.14	124.551	298.08
44.724	129.49	81.474	233.80	126.659	300.76
45.989	134.15	81.483	234.11	128.774	303.04
47.246	138.83	82.717	237.43	130.897	305.76
48.494	143.60	83.251	237.93	133.027	308.61
49.735	148.09	83.964	240.38	135.164	311.10
50.970	152.42	85.021	242.74	137.310	313.66
52.199	156.65	85.214	243.51	139.464	316.31
53.424	160.78	86.467	247.11	141.627	319.21
54.644	164.65	86.791	247.47	143.799	321.71
55.862	168.14	87.349	248.71	145.979	324.35
57.079	171.71	87.724	250.44	148.170	326.89
58.293	175.14	88.563	252.67	150.369	329.76
59.506	178.69	88.984	254.57	152.578	332.41
60.719	182.25	89.430	255.37	154.797	335.22
61.930	185.40	90.248	259.04	157.025	337.93
63.141	188.71	90.337	259.07	159.264	340.42
63.167	189.05	91.498	264.35	161.513	343.10
64.351	192.22	91.515	264.96	163.771	345.94
65.453	194.88	92.113	267.08	166.041	348.41

S1. continued.

T / K	C_p / J K^{-1} mol^{-1}	T / K	C_p / J K^{-1} mol^{-1}	T / K	C_p / J K^{-1} mol^{-1}
168.321	351.21	282.302	490.34	333.956	1591.37
170.612	354.44	285.650	495.51	334.168	1653.04
172.913	357.28	289.014	500.52	334.554	564.74
175.225	359.65	292.395	505.45	334.815	562.87
177.548	362.31	295.793	509.86	334.994	556.19
179.883	365.33	296.143	510.46	335.650	551.63
182.229	367.93	299.207	515.33	336.283	554.31
184.585	370.95	299.654	516.04	336.546	554.05
186.954	374.01	303.180	521.90	337.502	554.78
189.334	376.71	306.723	527.25	338.417	556.53
191.725	379.41	307.819	531.10	338.517	557.13
194.127	382.09	310.005	532.86	339.593	557.10
196.621	384.95	310.283	534.56	340.548	557.23
199.309	389.18	312.185	537.82	340.726	556.48
202.116	392.50	313.858	540.87	341.921	558.31
204.944	395.97	314.358	541.84	342.674	557.91
207.793	399.66	316.527	546.15	343.292	558.86
210.662	403.11	317.453	547.62	344.796	559.82
213.553	406.02	318.690	550.57	344.812	560.47
216.464	409.25	320.850	555.40	346.631	560.07
219.396	412.62	320.940	555.82	346.916	563.04
222.349	416.28	322.703	560.99	349.032	563.49
225.322	419.68	324.000	562.79	349.347	563.25
228.317	423.78	324.248	564.77		
231.331	427.25	325.786	570.75		
234.367	430.48	326.441	571.93		
237.422	434.49	327.321	578.02		
240.498	438.64	328.268	578.53		
243.593	441.27	328.852	589.04		
246.710	444.76	329.791	589.95		
249.848	448.58	330.381	603.80		
253.006	452.31	331.300	618.92		
256.185	456.27	331.359	626.31		
259.382	460.47	331.793	629.25		
262.599	464.20	332.224	646.72		
265.836	468.72	332.651	678.39		
269.092	473.52	332.776	699.74		
272.366	477.74	333.070	741.40		
275.659	481.72	333.473	943.41		
278.972	485.89	333.835	1468.89		

S2. Molar heat capacity of $[Co(NH_3)_6](ClO_4)_3$ after annealed at 98.3 K for 10 h.

T / K	C_p / J K^{-1} mol^{-1}	T / K	C_p / J K^{-1} mol^{-1}	T / K	C_p / J K^{-1} mol^{-1}
9.777	4.86	59.482	176.24	108.645	286.86
10.723	6.34	60.171	178.36	108.989	288.41
11.635	7.98	60.848	180.28	109.333	290.77
12.523	9.74	61.514	182.04	109.675	293.44
13.392	11.70	62.507	184.78	110.016	296.93
14.245	13.70	63.842	188.53	110.354	300.07
15.085	15.92	65.196	191.98	110.689	304.79
16.227	19.29	66.567	195.55	111.021	313.38
17.639	23.76	67.955	199.51	111.350	315.48
19.009	28.45	69.359	202.42	111.678	315.83
20.360	33.30	70.778	205.71	112.005	318.36
21.765	38.63	72.212	208.96	112.331	318.12
23.181	44.10	73.660	212.50	112.661	307.21
24.587	49.54	75.122	215.98	112.994	298.64
25.972	54.81	76.598	219.16	113.331	289.49
27.329	60.77	78.085	223.03	113.669	286.89
28.656	66.55	79.584	226.44	114.072	286.14
29.974	71.96	81.096	230.04	114.603	286.05
31.264	77.37	82.620	233.84	115.260	286.75
32.508	82.34	84.156	237.55	116.042	287.92
34.018	88.74	85.703	241.57	116.946	288.96
35.740	95.93	87.263	245.53	118.222	290.42
37.408	102.44	88.834	249.72	119.816	292.51
38.932	107.79	90.416	254.69	121.426	294.42
40.332	112.58	92.008	260.76	123.052	296.45
41.780	117.67	93.610	266.20		
43.265	122.92	95.223	271.33		
44.778	128.51	96.846	277.25		
46.313	134.24	98.477	284.72		
47.863	141.09	100.122	279.24		
49.425	146.29	101.784	275.84		
51.000	150.84	103.463	274.38		
52.584	156.05	105.155	275.86		
54.018	160.46	106.183	277.21		
55.050	163.51	106.538	278.56		
55.829	165.56	106.891	278.74		
56.590	167.98	107.244	280.28		
57.335	169.97	107.595	281.87		
58.064	172.37	107.946	282.01		
58.780	174.30	108.296	284.15		

S3. Molar heat capacity of $[Co(NH_3)_6](ClO_4)_3$ after annealed at 90.9 K for 30 h.

T / K	C_p / J K^{-1} mol^{-1}	T / K	C_p / J K^{-1} mol^{-1}	T / K	C_p / J K^{-1} mol^{-1}
13.412	12.06	79.683	224.28	107.017	279.03
14.928	15.80	81.043	227.45	107.506	280.25
16.464	20.20	82.378	230.69	107.992	280.31
18.002	25.12	83.689	233.69	108.476	282.25
19.524	30.40	84.978	236.76	108.958	283.95
21.005	35.83	86.246	239.90	109.437	285.53
22.530	41.65	87.495	242.80	109.911	290.53
24.148	47.91	88.726	245.89	110.378	314.03
25.794	54.14	89.939	249.31	110.830	332.23
27.434	61.19	90.825	252.05	111.270	358.51
29.007	67.94	91.390	253.97	111.698	413.83
30.390	73.61	91.951	255.52	112.130	375.85
31.712	78.93	92.510	256.99	112.571	348.05
33.155	84.69	93.065	258.34	113.025	308.93
34.679	91.21	93.616	260.52	113.490	291.97
36.256	97.74	94.164	262.10	113.957	288.08
37.689	102.88	94.710	263.55	114.425	287.99
39.061	107.47	95.252	265.26	114.891	287.36
40.590	112.67	95.791	266.87	115.420	287.94
42.209	118.42	96.328	267.96	116.201	288.63
43.897	124.44	96.860	270.35	117.481	289.94
45.636	130.87	97.390	272.90	119.080	291.84
47.412	139.20	97.916	277.18		
49.066	144.66	98.442	277.26		
50.631	148.39	98.963	277.92		
52.280	153.19	99.479	278.13		
53.999	158.39	99.994	278.36		
55.778	163.49	100.507	278.42		
57.609	168.70	101.018	278.49		
59.485	174.00	101.527	277.09		
61.401	179.44	102.035	275.38		
63.353	184.99	102.541	275.07		
65.307	190.28	103.045	274.62		
67.239	195.28	103.548	274.53		
69.150	199.65	104.049	275.18		
71.046	203.87	104.548	275.23		
72.929	208.31	105.045	276.00		
74.802	212.77	105.541	277.09		
76.666	217.09	106.035	277.13		
78.297	221.05	106.527	278.16		