

## Heat capacities and thermodynamic functions for beryl, $\text{Be}_3\text{Al}_2\text{Si}_6\text{O}_{18}$ , phenakite, $\text{Be}_2\text{SiO}_4$ , euclase, $\text{BeAlSiO}_4(\text{OH})$ , bertrandite, $\text{Be}_4\text{Si}_2\text{O}_7(\text{OH})_2$ , and chrysoberyl, $\text{BeAl}_2\text{O}_4$

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### ABSTRACT

The heat capacities of beryl, phenakite, euclase, and bertrandite have been measured between about 5 and 800 K by combined quasi-adiabatic cryogenic calorimetry and differential scanning calorimetry. The heat capacities of chrysoberyl have been measured from 340 to 800 K. The resulting data have been combined with solution and phase-equilibrium experimental data and simultaneously fit using the program PHAS20 to provide an internally consistent set of thermodynamic properties for several important beryllium phases. The experimental heat capacities and tables of derived thermodynamic properties are presented in this report.

The derived thermodynamic properties at 1 bar and 298.15 K for the stoichiometric beryllium phases beryl, phenakite, euclase, and bertrandite are entropies of  $346.7 \pm 4.7$ ,  $63.37 \pm 0.27$ ,  $89.09 \pm 0.40$ , and  $172.1 \pm 0.77$  J/(mol·K), respectively, and Gibbs free energies of formation (elements) of  $-8500.36 \pm 6.39$ ,  $-2028.39 \pm 3.78$ ,  $-2370.17 \pm 3.04$ , and  $-4300.62 \pm 5.45$  kJ/mol, respectively, and  $-2176.16 \pm 3.18$  kJ/mol for chrysoberyl. The coefficients  $c_1$  to  $c_5$  of the heat-capacity functions are as follows:

phase	$c_1$	$c_2$	$c_3 \times 10^5$	$c_4$	$c_5 \times 10^{-6}$	valid range
beryl	1625.842	-0.425 206	12.0318	-20 180.94	6.825 44	200–1800 K
phenakite	428.492	-0.099 582	1.9886	-5 670.47	2.082 6	200–1800 K
euclase	532.920	-0.150 729	4.1223	-6 726.30	2.197 6	200–1800 K
bertrandite	825.336	-0.099 651	...	-10 570.31	3.662 17	200–1400 K
chrysoberyl	362.701	-0.083 527	2.2482	-4 033.69	-6.797 6	200–1800 K

where  $C_p^o = c_1 + c_2T + c_3T^2 + c_4T^{-0.5} + c_5T^{-2}$  and  $T$  is in kelvins.

### INTRODUCTION

Recent advances in technology (Webster and London, 1979) have improved the mechanical properties of beryllium products at both room temperature and at elevated temperatures and have led to a renewed expansion in the use of beryllium in, and perhaps beyond, the traditional areas of weapons, space, optical, nuclear, and guidance systems; alloy-property modification; and, in military aircraft, disc brakes. The renewed interest in beryllium applications has rekindled an interest in the geochemistry and thermodynamic properties of beryllium-bearing phases. We report here the heat capacities and derived thermodynamic functions for beryl, phenakite, euclase, bertrandite, and chrysoberyl. A companion paper (Barton, 1986) presents an evaluation of solution and

phase-equilibrium experimental data, and a discussion of the petrology of selected beryllium phases.

All economic beryllium deposits are postmagmatic and related to acidic igneous rocks, generally biotite granites, but rarely high-silica and alkalic granites (Beus, 1966). The deposits are genetically related to late stages of pegmatitic processes or to various stages of hydrothermal processes (Jahns, 1955). Historically, beryllium production was limited to handpicking large beryl crystals from beryl-bearing granitic pegmatites. More recently, beryllium has been produced from hydrothermally altered deposits such as Iron Mountain, New Mexico (Jahns, 1944), and Spor Mountain, Thomas Range, Utah (Staatz, 1963). The pegmatitic process and related hydrothermal processes have been reviewed by Jahns (1955) and by Jahns and Burnham (1969).

Table 1. Experimental low-temperature heat capacities of beryl,  $\text{Be}_3\text{Al}_2\text{Si}_6\text{O}_{18} \cdot 0.36\text{H}_2\text{O}$

Temp.	Heat capacity	Temp.	Heat capacity	Temp.	Heat capacity
K	J/(mol·K)	K	J/(mol·K)	K	J/(mol·K)
<b>Series 1</b>		<b>Series 2</b>		<b>Series 3</b>	
299.16	442.2	169.80	262.6	4.95	0.9516
303.41	447.5	175.47	272.3	5.33	0.8519
308.37	452.6	181.11	281.9	5.77	0.7386
313.36	457.7	186.75	291.6	6.21	0.7534
318.54	462.9	192.37	300.8	6.72	0.8776
			197.98	309.8	7.40
				0.9827	
<b>Series 2</b>		203.58	318.4	8.20	1.026
		209.17	326.6	9.10	1.214
54.89	42.60	214.73	334.8	10.11	1.410
59.92	51.45	220.29	343.0	11.23	1.544
64.67	59.99	225.84	351.2	12.48	1.746
69.96	69.95	231.39	359.0	13.85	1.992
76.05	81.98	236.95	367.1	15.36	2.291
82.07	94.07	242.50	374.3	17.01	2.699
88.13	106.2	248.04	381.7	18.82	3.242
94.19	118.4	253.58	388.8	20.84	3.983
100.21	130.4	259.10	395.8	23.09	4.935
106.19	142.4	264.63	402.8	25.59	6.259
112.12	154.2	270.16	409.4	28.38	8.087
118.00	166.0	275.68	415.5	31.50	10.56
123.85	177.6	281.19	422.4	34.99	13.81
129.57	188.9	286.69	428.8	38.89	18.10
135.46	200.2	292.16	435.1	43.28	23.87
141.24	211.2	297.62	440.8	48.19	30.89
146.99	222.0	303.07	446.3	53.69	40.01
152.72	232.4	308.49	451.9		
<b>Series 4</b>					
		317.36	460.5	124.24	28.28
		322.54	466.3	130.11	30.79
		327.52	472.8	135.94	33.25
		332.52	477.5	141.75	35.73
		337.70	480.7	147.54	38.22
		342.86	486.0	153.31	40.65

The generalized geochemistry of the beryllium phases studied in this report may be summarized as follows. Beryl is the most abundant beryllium mineral and is most commonly found in granite pegmatites and granites. Beryl is also commonly found in greisens and quartz veins and occasionally in skarns and other metamorphic rocks. Chrysoberyl is the second most abundant beryllium mineral; however, the formation of chrysoberyl appears to require either a local enrichment of aluminum or a reduced silica activity. Chrysoberyl is most abundant in fluoritized skarns and is only occasionally found in pegmatites. Bertrandite is one of the few beryllium minerals that has a relatively wide occurrence and is found in most deposits of beryllium minerals. Bertrandite is present in commercial quantities in hydrothermally altered tuffs in the western part of the Thomas Range, Utah (Staatz, 1963). In pegmatites, bertrandite forms in the late stages, commonly forming as a hydrothermal alteration product of beryl or other beryllium phases often with euclase, muscovite, or bavenite coprecipitating as the sink for aluminum released in the alteration process. Euclase is relatively rare and is generally found in lower-temperature late-stage hydrothermal vein deposits. Phenakite forms in a wider range of deposits than euclase, but phenakite formation appears to be restricted to depositional environments with moderate to low aluminum activity. A more complete discussion of the petrology of these minerals may be found

Table 2. Experimental low-temperature heat capacities of phenakite,  $\text{Be}_2\text{SiO}_4$

Temp.	Heat capacity	Temp.	Heat capacity	Temp.	Heat capacity
K	J/(mol·K)	K	J/(mol·K)	K	J/(mol·K)
<b>Series 1</b>		<b>Series 1</b>		<b>Series 4</b>	
8.07	0.0130	193.17	57.42	315.49	100.7
9.15	0.0179	198.80	59.72	318.94	101.7
10.18	0.0190	204.42	62.01	324.05	103.2
11.38	0.0215	210.03	64.23	328.96	104.6
12.74	0.0327	215.63	66.48	333.86	106.0
14.19	0.0497	221.22	68.64	338.88	107.4
15.79	0.0685	226.81	70.80	343.90	108.6
17.55	0.0885			348.92	109.8
19.50	0.1217	<b>Series 2</b>		353.93	110.9
21.66	0.1635			358.91	112.0
24.05	0.2273	231.06	72.44	363.88	113.1
26.72	0.3244	236.45	74.49	368.83	114.5
29.71	0.4805	241.57	76.34	373.78	115.6
33.04	0.7246	246.74	78.23	378.71	116.5
36.77	1.062	252.17	80.15		
45.61	1.910	257.58	82.07	<b>Series 5</b>	
50.85	2.661	262.99	84.00		
56.67	3.835	273.76	87.83	297.13	95.31
62.91	5.417	279.17	89.55	302.42	97.26
69.28	7.334	284.53	91.30		
75.65	9.515	289.84	93.07	<b>Series 6</b>	
81.95	11.79	295.15	94.75		
88.15	14.08	300.43	96.29	28.15	0.4006
94.30	16.38	305.68	97.83	29.34	0.4595
100.39	18.67	310.89	99.47	30.19	0.5099
106.42	21.00	316.08	101.0	31.11	0.5805
112.41	23.38			32.24	0.6653
118.34	25.80	<b>Series 3</b>		33.34	0.7557
124.24	28.28			34.46	0.8515
130.11	30.79	265.51	84.80	35.59	0.9558
135.94	33.25	266.54	85.14	36.72	1.066
141.75	35.73	267.47	85.76	37.84	1.177
147.54	38.22	268.40	86.02	40.08	1.407
153.31	40.65	269.37	86.51	41.21	1.545
159.05	43.07	270.35	87.00	42.32	1.679
164.78	45.50	271.32	87.00	43.43	1.787
170.49	47.94	272.29	86.99	45.65	1.907
176.18	50.30	273.26	87.21	46.77	2.049
181.86	52.67	274.23	87.61	47.88	2.191
187.52	55.05			48.99	2.356
				50.10	2.528
				51.20	2.712
				52.31	2.963
				53.40	3.109
				54.51	3.372

in Barton (1986), Beus (1966) and Mulligan (1968) discussed various beryllium deposits.

Kelley (1939) measured the heat capacities of bromellite and phenakite between about 54 and 294 K. Chase et al. (1974) reported that Kelley's data for bromellite deviate by +50% at 56 K, +8% at 100 K, and +1% at 200 K from more recent measurements based upon improved technology. Similar systematic errors can be expected for the heat-capacity data for phenakite reported by Kelley.

This study was motivated by the need to provide a consistent set of thermodynamic functions for important beryllium minerals to aid in the development of geochemical models for beryllium ore genesis and to help in the development of extraction procedures for beryllium obtained from new source materials. To this end, low-temperature heat-capacity measurements of beryl, phenakite, euclase, and bertrandite were combined with high-temperature heat capacities for these and other beryllium-bearing phases and with phase-equilibrium data and were simultaneously evaluated.



Table 5. Molar thermodynamic properties of beryl,  
 $\text{Be}_3\text{Al}_2\text{Si}_6\text{O}_{18} \cdot 0.36\text{H}_2\text{O}$ , to 340 K

Temp. T Kelvin	Heat capacity $C_p^o$	Entropy $S_T^o - S_0^o$	Enthalpy function $(H_T^o - H_0^o)/T$	Gibbs energy function $-(G_T^o - G_0^o)/T$
$J/(mol \cdot K)$				
5	0.140	0.046	0.034	0.012
10	0.873	0.327	0.240	0.087
15	2.091	0.899	0.644	0.255
20	3.660	1.704	1.191	0.513
25	5.921	2.745	1.894	0.851
30	9.318	4.107	2.832	1.275
35	13.82	5.868	4.068	1.801
40	19.48	8.069	5.627	2.442
45	26.22	10.75	7.534	3.212
50	33.84	13.89	9.774	4.119
60	51.57	21.61	15.24	6.365
70	70.08	30.93	21.74	9.192
80	89.87	41.57	29.01	12.56
90	110.0	53.32	36.89	16.43
100	130.0	65.94	45.20	20.74
110	150.0	79.27	53.82	25.45
120	169.9	93.18	62.66	30.51
130	189.6	107.6	71.68	35.88
140	208.8	122.3	80.79	41.53
150	227.4	137.4	89.95	47.41
160	245.4	152.6	99.11	53.51
170	263.0	168.0	108.2	59.79
180	280.1	183.5	117.3	66.24
190	296.9	199.1	126.3	72.82
200	312.8	214.8	135.3	79.53
210	328.0	230.4	144.1	86.34
220	342.7	246.0	152.8	93.25
230	357.1	261.6	161.3	100.2
240	371.0	277.1	169.8	107.3
250	384.2	292.5	178.1	114.4
260	396.9	307.8	186.3	121.5
270	409.1	323.0	194.3	128.7
280	421.0	338.1	202.2	135.9
290	432.4	353.1	209.9	143.1
300	443.4	367.9	217.5	150.4

Table 6. Molar thermodynamic properties of phenakite,  
 $\text{Be}_2\text{SiO}_4$ , to 370 K

Temp. T Kelvin	Heat capacity $C_p^o$	Entropy $S_T^o - S_0^o$	Enthalpy function $(H_T^o - H_0^o)/T$	Gibbs energy function $-(G_T^o - G_0^o)/T$
$J/(mol \cdot K)$				
5	0.002	0.001	0.001	0.000
10	0.017	0.006	0.004	0.002
15	0.058	0.019	0.014	0.005
20	0.137	0.046	0.034	0.012
25	0.258	0.087	0.065	0.023
30	0.500	0.154	0.115	0.038
35	0.900	0.259	0.197	0.062
40	1.409	0.412	0.316	0.096
45	1.880	0.606	0.465	0.141
50	2.513	0.834	0.635	0.199
60	4.643	1.467	1.113	0.354
70	7.577	2.394	1.818	0.575
80	11.05	3.628	2.751	0.877
90	14.74	5.142	3.877	1.264
100	18.53	6.890	5.152	1.738
110	22.45	8.839	6.546	2.293
120	26.52	10.97	8.039	2.926
130	30.71	13.25	9.621	3.631
140	34.96	15.68	11.28	4.405
150	39.23	18.24	13.00	5.241
160	43.48	20.91	14.77	6.137
170	47.72	23.67	16.59	7.087
180	51.92	26.52	18.43	8.087
190	56.09	29.44	20.31	9.133
200	60.19	32.42	22.20	10.22
210	64.22	35.45	24.10	11.35
220	68.16	38.53	26.02	12.52
230	72.02	41.65	27.93	13.72
240	75.77	44.79	29.85	14.95
250	79.43	47.96	31.76	16.20
260	82.99	51.15	33.66	17.49
270	86.46	54.34	35.55	18.79
280	89.82	57.55	37.43	20.12
290	93.07	60.76	39.29	21.46
300	96.20	63.97	41.14	22.83
310	99.20	67.17	42.96	24.21
320	102.1	70.37	44.77	25.60
330	104.8	73.55	46.55	27.00
340	107.5	76.72	48.30	28.42
350	110.0	79.87	50.03	29.84
360	112.4	83.00	51.72	31.28
370	114.8	86.12	53.40	32.72
373.15	87.53	55.35	36.15	19.21
298.15	95.63	63.37	40.80	22.57

weighed 33.4966 g, corrected for buoyancy. The scanning-calorimetric sample was 26.178 mg.

Bertrandite [ $\text{Be}_4\text{Si}_2\text{O}_7(\text{OH})_2$ ] was obtained from the Field Museum of Natural History (FMNH 6969). The source location listed was Albany, Maine. Microprobe analyses of the sample detected only silicon. A few of the crystals were cloudy in appearance. Inspection of some of these crystals revealed the presence of a claylike phase. The cell constants for the sample are  $a = 0.87135(4)$  nm,  $b = 1.52677(14)$  nm, and  $c = 0.4583(3)$  nm, and the molar volume is  $91.50(1)\text{ cm}^3$ . Weight-loss studies indicated that the sample contained 7.4(6) wt%  $\text{H}_2\text{O}$ . The low-temperature heat-capacity sample weighed 22.4678 g, corrected for buoyancy. The scanning-calorimetric sample was 25.006 mg.

The chrysoberyl ( $\text{BeAl}_2\text{O}_4$ ) sample was a tabular crystal fragment. The pale-green grain was opaque and contained 3.1 wt%  $\text{Fe}_2\text{O}_3$  (Fe by microprobe). The scanning-calorimetric sample was 20.429 mg. The cell constants for the

sample are  $a = 0.54801(3)$  nm,  $b = 0.94119(7)$  nm, and  $c = 0.44288(3)$  nm, and the molar volume is  $34.32\text{ cm}^3$ .

Low-temperature heat capacities were measured using the intermittent heating method under quasi-adiabatic conditions. The cryostat has been described by Robie and Hemingway (1972), the provisional low-temperature scale by Robie et al. (1978), and the electrical measurement system by Hemingway et al. (1984). The samples were sealed in the calorimeter under a small pressure of pure helium gas (about 5 kPa).

High-temperature heat capacities were determined by differential scanning calorimetry (DSC) following the procedures outlined by Hemingway et al. (1981). The samples were enclosed in unsealed gold pans.

The formula weights were based upon the 1975 values for the atomic weights (Commission on Atomic Weights, 1976). The formula weights are 544.74, 537.505, 110.107, 145.084, 238.230, and 126.973 g, respectively, for beryl

Table 7. Molar thermodynamic properties of euclase,  
BeAlSiO<sub>4</sub>(OH), to 380 K

Temp. T Kelvin	Heat capacity $C_p^{\circ}$	Entropy $S_T^{\circ} - S_0^{\circ}$	Enthalpy function $(H_T^{\circ} - H_0^{\circ})/T$	Gibbs energy function $-(G_T^{\circ} - G_0^{\circ})/T$
	$J/(mol \cdot K)$			
5	0.002	0.001	0.000	0.000
10	0.017	0.005	0.004	0.001
15	0.069	0.020	0.015	0.005
20	0.197	0.055	0.043	0.012
25	0.387	0.119	0.092	0.027
30	0.767	0.219	0.169	0.050
35	1.361	0.379	0.294	0.085
40	2.282	0.618	0.482	0.135
45	3.296	0.945	0.738	0.206
50	4.525	1.351	1.051	0.300
60	8.001	2.478	1.915	0.563
70	11.80	3.984	3.043	0.940
80	16.61	5.867	4.433	1.434
90	21.75	8.118	6.069	2.049
100	27.13	10.69	7.905	2.782
110	32.78	13.54	9.907	3.628
120	38.60	16.64	12.06	4.581
130	44.48	19.96	14.32	5.635
140	50.33	23.47	16.69	6.782
150	56.12	27.14	19.12	8.016
160	61.85	30.94	21.61	9.330
170	67.54	34.86	24.15	10.72
180	73.11	38.88	26.71	12.17
190	78.49	42.98	29.30	13.68
200	83.69	47.14	31.89	15.25
210	88.75	51.35	34.48	16.87
220	93.68	55.59	37.06	18.53
230	98.48	59.86	39.62	20.24
240	103.1	64.15	42.17	21.98
250	107.6	68.45	44.70	23.75
260	111.9	72.76	47.20	25.55
270	116.0	77.06	49.68	27.38
280	119.9	81.35	52.12	29.23
290	123.7	85.62	54.52	31.10
300	127.4	89.88	56.89	32.99
310	130.9	94.11	59.22	34.89
320	134.3	98.32	61.51	36.81
330	137.5	102.5	63.77	38.74
340	140.7	106.7	65.98	40.67
350	143.7	110.8	68.16	42.62
360	146.6	114.9	70.30	44.57
370	149.3	118.9	72.40	46.52
380	151.9	122.9	74.45	48.48
273.15	117.2	78.41	50.45	27.96
298.15	126.7	89.09	56.45	32.64

of the composition Be<sub>3</sub>Al<sub>2</sub>Si<sub>6</sub>O<sub>18</sub>·0.36H<sub>2</sub>O, stoichiometric beryl, phenakite, euclase, bertrandite, and chrysoberyl.

#### LOW-TEMPERATURE HEAT CAPACITIES AND THERMODYNAMIC FUNCTIONS

Heat-capacity measurements for beryl, phenakite, euclase, and bertrandite are listed in the chronological order of measurement (series) in Tables 1–4. The results are corrected for curvature (e.g., Robie and Hemingway, 1972). The data were smoothed using cubic spline routines and were graphically extrapolated to zero kelvin using the experimental and smoothed values for temperatures less than 30 K plotted in the form of  $C_p^{\circ}/T$  vs.  $T^2$ . Smoothed values of the heat capacities and derived thermodynamic functions are listed in Tables 5–8.

The entropy changes,  $S_T^{\circ} - S_0^{\circ}$ , at 298.15 K are  $365.2 \pm 0.7$ ,  $63.37 \pm 0.13$ ,  $89.09 \pm 0.18$ , and  $172.1 \pm 0.34$  J/(mol·K),

Table 8. Molar thermodynamic properties of bertrandite,  
Be<sub>4</sub>Si<sub>2</sub>O<sub>7</sub>(OH)<sub>2</sub>, to 370 K

Temp. T Kelvin	Heat capacity $C_p^{\circ}$	Entropy $S_T^{\circ} - S_0^{\circ}$	Enthalpy function $(H_T^{\circ} - H_0^{\circ})/T$	Gibbs energy function $-(G_T^{\circ} - G_0^{\circ})/T$
	$J/(mol \cdot K)$			
5	0.021	0.007	0.005	0.002
10	0.164	0.055	0.041	0.014
15	0.517	0.179	0.133	0.046
20	1.198	0.409	0.303	0.105
25	2.300	0.791	0.590	0.202
30	3.860	1.337	0.994	0.343
35	5.845	2.079	1.543	0.536
40	8.488	3.025	2.240	0.785
45	11.39	4.188	3.091	1.097
50	15.02	5.567	4.094	1.473
60	24.62	9.151	6.712	2.438
70	31.67	13.50	9.800	3.703
80	39.22	18.20	12.98	5.218
90	48.31	23.34	16.40	6.942
100	57.54	28.91	20.05	8.857
110	66.98	34.83	23.89	10.95
120	76.34	41.06	27.87	13.19
130	85.82	47.55	31.96	15.59
140	95.20	54.25	36.14	18.11
150	104.5	61.14	40.39	20.74
160	113.7	68.17	44.69	23.49
170	122.9	75.34	49.02	26.33
180	132.0	82.63	53.38	29.25
190	140.9	90.01	57.75	32.26
200	149.6	97.46	62.13	35.33
210	158.0	105.0	66.49	38.47
220	166.3	112.5	70.84	41.66
230	174.4	120.1	75.17	44.90
240	182.5	127.7	79.47	48.19
250	190.9	135.3	83.76	51.53
260	200.7	143.0	88.06	54.89
270	209.8	150.7	92.42	58.30
280	213.3	158.4	96.68	61.74
290	218.9	166.0	100.8	65.20
300	225.6	173.5	104.8	68.69
310	232.1	181.0	108.8	72.19
320	238.2	188.5	112.8	75.71
330	243.8	195.9	116.7	79.24
340	249.5	203.3	120.5	82.78
350	255.1	210.6	124.3	86.33
360	260.7	217.9	128.0	89.88
370	266.0	225.1	131.6	93.44
380	273.15	211.1	153.2	93.78
273.15	298.15	224.4	172.1	104.1
298.15				68.04

K), respectively, for beryl (Be<sub>3</sub>Al<sub>2</sub>Si<sub>6</sub>O<sub>18</sub>·0.36H<sub>2</sub>O), phenakite, euclase, and bertrandite. Our value for the entropy of phenakite is about 1.5% lower than the value reported by Kelley (1939). Our heat capacities for phenakite are roughly equivalent to those reported by Kelley (1939) at the higher temperatures, but at lower temperatures the values show significant deviations. Kelley's (1939) heat capacities are 13% larger near 55 K. Similar discrepancies were noted by Chase et al. (1974) for BeO (Kelley, 1939) and were expected in this study.

#### HIGH-TEMPERATURE HEAT CAPACITIES AND THERMODYNAMIC FUNCTIONS

High-temperature heat-capacity measurements for beryl (Be<sub>3</sub>Al<sub>2</sub>Si<sub>6</sub>O<sub>18</sub>·0.36H<sub>2</sub>O), anhydrous beryl powder, phenakite, euclase, bertrandite, and chrysoberyl were made between 340 and about 800 K. The experimental values are listed in Tables 9–14. Each scan represents one con-



Table 11. Experimental high-temperature heat capacities of phenakite,  $\text{Be}_2\text{SiO}_4$ 

Temp.	Heat capacity	Temp.	Heat capacity	Temp.	Heat capacity
K	J/(mol·K)	K	J/(mol·K)	K	J/(mol·K)
Scan 1	1	Scan 2	2	Scan 4	4
340.0	107.2	410.8	125.0	580.5	149.1
350.0	109.4	420.8	127.2	590.5	150.1
360.0	111.5	430.7	128.8	600.5	150.8
370.0	113.6	440.7	130.2	610.5	151.8
380.0	115.5	450.7	131.1	620.4	152.5
390.0	117.6	460.7	132.2	630.4	152.9
400.0	119.5	470.7	135.5	640.4	153.2
410.0	121.7	480.7	137.5	650.4	154.4
420.0	123.8	490.7	138.6	660.4	155.1
430.0	125.9	500.6	139.6	670.4	156.0
440.0	127.9			680.4	154.9
450.0	129.8	Scan 3	690.3	159.3	
460.0	131.3		700.3	159.3	
470.0	132.8	460.7	133.2		
480.0	135.1	470.7	134.9	Scan 5	
490.0	137.5	480.7	136.8		
500.0	139.0	490.7	138.1	690.3	157.7
				700.3	158.1
Scan 2	510.6	139.9	710.3	159.3	
	520.6	142.1	720.3	159.5	
330.9	104.4	530.6	143.2	730.3	160.7
340.9	106.4	540.6	144.5	740.3	161.4
350.9	109.3	550.6	145.4	750.2	161.9
360.9	111.8	560.5	146.1	760.2	162.3
370.8	114.6	570.5	146.8	770.2	162.4
380.8	117.5	580.5	147.7	780.2	163.8
390.8	120.4	590.5	148.7	790.2	164.3
400.8	122.6	600.5	149.5	800.2	164.9

ancillary data from Robie et al. (1979) to provide the smoothed values of the heat capacities and derived thermodynamic functions listed in Tables 16–20 for the temperature interval 298.15 to 1800 K (1200 K for bertrandite).

## DISCUSSION

The entropy correction for the  $\text{H}_2\text{O}$  in beryl may be estimated by assuming that the  $\text{H}_2\text{O}$  in beryl is similar in bonding characteristics to that in analcime or clinoptilolite. Under this assumption we calculate the entropy contribution of 1 mol of  $\text{H}_2\text{O}$  to be 55 J/(mol·K) at 298.15 K from the data of Johnson et al. (1982) for analcime and dehydrated analcime or 57 J/(mol·K) calculated from the data of Hemingway and Robie (1984) for clinoptilolite. The calculated value for the entropy of anhydrous beryl at 298.15 K is  $345.0 \pm 5$  J/(mol·K) based upon this model and is in excellent agreement with the value of  $346.7 \pm 4.7$  J/(mol·K) predicted from the multiple regression analysis (Barton, 1986).

The heat capacity of beryl is unexpectedly high at low temperatures (less than 30 K) for a compound with such a low mean atomic weight. We can calculate the Debye temperatures for two beryls, goshenite as 795.6 K and aquamarine as 799.1 K, from the elastic constant measurements of Yoon and Newnham (1973). The heat capacity of material with a Debye temperature of approximately 800 K can be represented reasonably well to about 16 K by a Debye function. From such a function we would

Table 12. Experimental high-temperature heat capacities of euclase,  $\text{BeAlSiO}_4(\text{OH})$ 

Temp.	Heat capacity	Temp.	Heat capacity	Temp.	Heat capacity
K	J/(mol·K)	K	J/(mol·K)	K	J/(mol·K)
Scan 1	1	Scan 2	2	Scan 5	10
343.5	140.9	522.4	177.4	690.9	196.8
363.3	146.6	542.2	180.5	710.8	198.2
383.1	151.9	562.0	183.4	730.6	199.8
403.0	156.8	581.9	186.3	749.5	201.8
422.8	161.3	600.7	188.5		
442.6	165.4	621.6	190.8	Scan 11	
462.4	169.4	641.4	192.3		
482.3	173.1	661.3	193.5	690.9	196.8
501.1	176.1	681.1	195.7	710.8	198.3
		700.0	197.8	730.6	199.9
				749.5	202.2
Scan 2		Scan 6		Scan 12	
470.7	167.7				
480.7	171.0	621.6	191.6		
490.7	173.2	641.4	192.4	740.5	200.9
500.6	175.3	661.3	193.9	760.4	202.8
510.6	176.5	681.1	196.3	780.2	203.8
520.6	178.1	700.0	198.5	799.1	205.0
530.6	179.3				
540.6	180.3	Scan 7		Scan 13	
550.6	181.7				
560.5	183.0	590.5	187.0	740.5	201.5
570.5	184.0	600.5	188.5	760.4	202.7
580.5	185.0	610.4	189.7	780.2	204.4
590.5	186.7	620.4	190.8	799.1	206.1
600.5	187.2	630.4	191.8		
		640.4	192.9	Scan 14	
Scan 3	650.4	194.0			
	660.4	194.2	770.2	205.0	
472.8	172.4	670.3	195.0	780.2	204.2
492.6	175.5	690.3	196.3	790.2	203.5
512.4	178.3				
532.3	181.7	Scan 8		Scan 15	
551.1	184.3				
	571.8	185.0	770.2	203.3	
Scan 4	591.6	187.3	780.2	204.0	
	611.4	189.5	790.2	201.9	
472.8	171.9	631.3	191.3	800.2	203.6
492.6	174.9	650.1	193.5		
512.4	177.9			Scan 16	
532.3	181.1	Scan 9			
551.1	183.7				
	571.8	185.8	858.8	207.3	
	591.6	187.5	868.6	210.4	
	611.4	190.3			
	631.3	191.9			
	650.1	193.3			

estimate a heat capacity for beryl that is only about 19% of the measured heat capacity at 16 K. The heat capacity of 0.36 mol of ice is similarly about 20% of the measured heat capacity at 16 K. Finally, a Schottky contribution to the heat capacity, arising from  $\text{Cr}^{3+}$  ions in solid solution, may be expected at very low temperatures. However, it is unlikely that the contribution would be 60% of the heat capacity observed at 16 K. We have no explanation for the deviation of our measured heat capacities from our theoretical estimates.

Small numbers of fluid inclusions in the phenakite and bertrandite samples produced small anomalies in the heat capacities near 269 K. The bertrandite sample produced a larger anomalous heat capacity and, therefore, contained a greater quantity of fluid, generally associated with a claylike phase. The smoothed values of the thermodynamic properties of these phases have been corrected for

Table 13. Experimental high-temperature heat capacities of bertrandite,  $\text{Be}_4\text{Si}_2\text{O}_7(\text{OH})_2$ 

Temp.	Heat capacity	Temp.	Heat capacity	Temp.	Heat capacity
K	J/(mol·K)	K	J/(mol·K)	K	J/(mol·K)
Scan 1	250.7	Scan 3	251.9	Scan 5	
343.5	340.8	472.8	309.3		
363.3	261.6	350.8	258.0	340.0	115.7
383.1	271.9	360.8	263.1	350.0	117.8
403.0	280.3	370.8	268.3	360.0	119.9
422.8	288.3	380.8	272.9	370.0	122.0
442.6	295.7	390.8	276.1	380.0	123.7
462.4	303.3	400.7	280.6	390.0	125.8
482.3	310.4	410.7	284.9	400.0	127.4
501.1	316.5	420.7	289.0	410.0	129.4
		430.7	293.2	420.0	131.3
Scan 2	440.7	296.3	480.6	430.0	133.0
		490.6	312.9	440.0	134.8
		500.6	315.6	450.0	136.8
450.7	300.2	Scan 4	520.6	460.0	138.2
460.7	303.4		530.6	470.0	139.9
470.6	307.3	472.8	310.0	480.0	141.9
480.6	310.4	492.6	315.5	490.0	143.8
490.6	314.0	512.4	321.2	500.0	145.4
500.6	317.6	532.3	328.1		
		551.1	333.5	Scan 2	520.6
		580.5	335.7		147.4
		590.5	339.0		530.6
		600.5	342.4	340.9	116.9

the small contribution to the heat capacities of the  $\text{H}_2\text{O}$  in the inclusions.

The high-temperature heat capacities were obtained despite significant experimental problems. At temperatures greater than about 470 K, small quantities of volatiles were emitted from some of the samples during the differential-scanning-calorimetric measurements. The excess heat required for diffusion of the volatiles to the sample surface and for vaporization from the surface resulted in a large scatter in calculated heat capacities and, in some cases, the calculation of erroneous heat capacities from the DSC scans (Hemingway and Kirby, unpub. data, 1985). Experimental results obtained for temperatures greater than 800 K were of poor quality and were not reported. Therefore, it is estimated that the uncertainties in the high-temperature heat capacities may average  $\pm 2\%$ .

Use of the Haas-Fisher equation for high-temperature  $C_p$  extrapolations recently has been criticized, in particular with respect to equation form. It should be noted that

Table 14. Experimental high-temperature heat capacities of chrysoberyl,  $\text{BeAl}_2\text{O}_4$ 

Temp.	Heat capacity	Temp.	Heat capacity	Temp.	Heat capacity
K	J/(mol·K)	K	J/(mol·K)	K	J/(mol·K)
Scan 1	420.8	Scan 2	133.0	Scan 3	580.5
340.0	115.7	430.7	134.7	440.7	155.1
350.0	117.8	360.7	136.3	590.5	155.8
360.0	119.9	370.7	138.1	600.5	155.6
370.0	122.0	380.7	139.4	610.5	157.2
380.0	123.7	390.7	141.3	620.4	157.2
390.0	125.8	400.7	142.0	630.4	159.0
400.0	127.4	410.7	144.4	640.4	158.9
410.0	129.4	420.6	145.5	650.4	160.0
420.0	131.3	430.0	146.3	660.4	160.6
430.0	133.0	440.0	148.6	Scan 5	
440.0	134.8	450.0	148.6		
450.0	136.8	460.0	149.9	670.4	161.3
460.0	138.2	470.0	141.3	680.4	161.9
470.0	139.9	480.0	142.2	690.3	163.5
480.0	141.9	490.0	143.9	700.3	164.4
490.0	143.8	500.0	145.0		
500.0	145.4	510.6	146.3	710.3	164.4
		510.6	147.4	720.3	163.3
		520.6	147.4	730.3	164.8
		530.6	148.6	740.3	165.8
		540.6	149.9	750.2	166.8
		550.6	151.1	760.2	167.1
		560.5	152.2	770.2	167.2
		570.5	153.5	780.2	167.0
		580.5	154.2	790.2	168.2
		590.5	155.4	800.2	168.2
		600.5	156.5		
		131.2			

the properties of equations used to fit experimental results, the function of the Haas-Fisher equation, are different from the properties of equations developed to extrapolate or estimate values beyond the known universe of experimental results. Such differences and the consequences of ignoring such differences are discussed in mathematics courses and are beyond the scope of this paper. Well-behaved equations, that is, equations that do not quickly change slope beyond the end points of a data set (e.g., the Meyer-Kelley equation) do not guarantee reliable extrapolations of experimental data. It is sufficient to note that the problems discussed by several authors lie in the mis-

Table 15. Heat capacity equations for selected beryllium minerals

Phase	$c_1$	$c_2$	$c_3$	$c_4$	$c_5$
Beryl	1625.842	-0.425206	$1.20318 \times 10^{-4}$	-20180.94	$6.82544 \times 10^6$
Phenakite	428.492	-0.099582	$1.9886 \times 10^{-5}$	-5670.47	$2.0826 \times 10^6$
Euclase	532.920	-0.150729	$4.1223 \times 10^{-5}$	-6726.30	$2.1976 \times 10^6$
Bertrandite	825.336	-0.099651		-10570.31	$3.66217 \times 10^6$
Chrysoberyl	362.701	-0.083527	$2.2482 \times 10^{-5}$	-4033.69	$-6.7976 \times 10^4$

The form of the equation is

$$C_p^\circ = c_1 + c_2 T + c_3 T^2 + c_4 T^{-0.5} + c_5 T^{-2}$$

Table 16. Molar thermodynamic properties of beryl,  $\text{Be}_3\text{Al}_2\text{Si}_6\text{O}_{18}$ , to 1800 K

Temp. T kelvin	Heat capacity $C_p^{\circ}$	Entropy $S_T^{\circ} - S_0^{\circ}$	Enthalpy function $(H_T^{\circ} - H_{298}^{\circ})/T$	Gibbs energy function $-(G_T^{\circ} - H_{298}^{\circ})/T$	Formation from the elements		
					Gibbs Enthalpy free energy kJ/mol	log $K_f$	
298.15	417.8	346.7	0.000	346.7	-9006.56 7.10 6.39	-8500.36 1489.23	
Uncertainty		4.7					
300	419.8	349.3	2.583	346.7	-9006.36 -8497.07	1479.47	
400	508.6	483.1	118.75	364.3	-9014.86 -8331.73	1088.01	
500	568.1	603.4	203.05	400.4	-9008.66 -8156.06	852.057	
600	609.1	710.8	267.51	443.3	-9004.80 -8999.40	-7985.81 -7816.41	
700	638.3	807.0	318.51	488.5	-8996.47 -8993.19	653.267	
800	659.8	893.7	359.90	533.9	-8993.19 -8986.63	499.349	
900	676.3	972.5	394.18	578.3	-8986.63 -7480.05	434.130	
1000	689.6	1044	423.08	621.3	-9001.30 -7311.03	381.889	
1100	700.9	1111	447.83	662.9	-8994.00 -8986.47	339.202	
1200	711.0	1172	469.35	702.8	-8986.47 -6974.69	303.600	
1300	720.7	1229	488.31	741.1	-8978.73 -8970.74	273.510	
1400	730.5	1283	505.26	777.9	-8970.74 -8962.43	247.765	
1500	740.7	1334	520.61	813.3	-8962.43 -6474.86	225.474	
1600	751.7	1382	534.71	847.4	-8997.73 -9290.46	205.927	
1700	763.6	1428	547.81	880.2	-9290.46 -9277.44	188.582	
1800	776.7	1472	560.16	911.8	-9277.44 -5952.32	172.732	
Transitions in the reference state elements							
Aluminum....		melting point	933.25 K				
Beryllium....		alpha - beta	1527 K				
		melting point	1560 K				
Silicon.....		melting point	1685 K				

Table 17. Molar thermodynamic properties of phenakite,  $\text{Be}_2\text{SiO}_4$ , to 1800 K

Temp. T kelvin	Heat capacity $C_p^{\circ}$	Entropy $S_T^{\circ} - S_0^{\circ}$	Enthalpy function $(H_T^{\circ} - H_{298}^{\circ})/T$	Gibbs energy function $-(G_T^{\circ} - H_{298}^{\circ})/T$	Formation from the elements		
					Gibbs Enthalpy free energy kJ/mol	log $K_f$	
298.15	95.60	63.37	0.000	63.37	-2143.12 3.78 3.78	-2028.39 355.365	
Uncertainty		0.27					
300	96.16	63.96	0.591	63.37	-2143.08 -2143.91	-2027.62 -1989.03	
400	121.3	95.31	27.835	67.47	-2143.91 -2143.47	259.740 203.749	
500	138.4	124.3	48.351	75.99	-2143.47 -2134.48	-1950.33 -1760.20	
600	150.2	150.7	64.402	86.28	-2142.59 -2140.59	-1912.15 -1873.55	
700	158.5	174.5	77.282	97.21	-2140.59 -2128.66	-1835.55 -1648.70	
800	164.3	196.1	87.816	108.2	-2128.66 -2136.58	-1835.55 -1797.76	
900	168.5	215.7	96.563	119.1	-2136.58 -2134.48	-1797.76 -1760.20	
1000	171.6	233.6	103.92	129.7	-2134.48 -2125.55	-1760.20 -1575.11	
1100	173.8	250.0	110.17	139.9	-2132.44 -2130.49	-1722.82 -1685.67	
1200	175.4	265.2	115.54	149.7	-2130.49 -2128.66	-1685.67 -1648.70	
1300	176.6	279.3	120.20	159.1	-2128.66 -2127.02	-1648.70 -1611.96	
1400	177.6	292.5	124.26	168.2	-2127.02 -2125.55	-1611.96 -1575.11	
1500	178.4	304.7	127.84	176.9	-2125.55 -2200.14	-1575.11 -1457.51	
1600	179.1	316.3	131.02	185.2	-2153.61 -2202.27	-1537.69 -1498.66	
1700	179.9	327.1	133.88	193.3	-2202.27 -2200.14	-1498.66 -1457.51	
1800	180.7	337.5	136.45	201.0	-2200.14 -1457.51	-1457.51 -12.296	
Transitions in the reference state elements							
Beryllium....		alpha - beta	1527 K				
		melting point	1560 K				
Silicon.....		melting point	1685 K				

Transitions in the reference state elements

Beryllium.... alpha - beta 1527 K  
 melting point 1560 K  
 Silicon..... melting point 1685 K

Table 18. Molar thermodynamic properties of euclase,  $\text{BeAlSiO}_4(\text{OH})$ , to 1800 K

Temp. T kelvin	Heat capacity $C_p^{\circ}$	Entropy $S_T^{\circ} - S_0^{\circ}$	Enthalpy function $(H_T^{\circ} - H_{298}^{\circ})/T$	Gibbs energy function $-(G_T^{\circ} - G_{298}^{\circ})/T$	Formation from the elements		
					Gibbs Enthalpy free energy kJ/mol		$\log K_f$
					-	-	
298.15	126.8	89.09	0.000	89.09	-2532.91	-2370.17	415.243
Uncertainty	0.40				3.05	3.04	
300	127.5	89.88	0.784	89.09	-2532.86	-2369.10	412.497
400	156.6	130.8	36.363	94.48	-2533.97	-2314.34	302.221
500	175.8	168.0	62.469	105.5	-2533.40	-2259.50	236.048
600	188.8	201.3	82.514	118.8	-2531.89	-2204.84	191.948
700	197.9	231.1	98.384	132.7	-2529.87	-2150.49	160.471
800	204.3	258.0	111.25	146.7	-2527.59	-2096.45	136.884
900	209.2	282.3	121.87	160.5	-2525.26	-2042.64	118.552
1000	212.9	304.6	130.79	173.8	-2533.63	-1988.45	103.866
1100	216.0	325.0	138.40	186.6	-2531.08	-1934.01	91.838
1200	218.8	343.9	144.99	198.9	-2529.22	-1880.55	81.858
1300	221.4	361.5	150.76	210.8	-2525.85	-1825.86	73.364
1400	224.0	378.1	155.90	222.1	-2523.17	-1772.26	66.124
1500	226.9	393.6	160.54	233.1	-2520.43	-1718.64	59.848
1600	230.0	408.3	164.78	243.6	-2532.27	-1664.77	54.349
1700	233.4	422.4	168.71	253.7	-2579.46	-1610.34	49.480
1800	237.3	435.8	172.42	263.4	-2575.55	-1553.32	45.076
Transitions in the reference state elements							
Aluminum.....	melting point	933.25 K					
Beryllium....	alpha - beta	1527 K					
	melting point	1560 K					
Silicon.....	melting point	1685 K					

application of the Haas-Fisher equation and not in the form of the equation, and that responsibility for developing and justifying algorithms to extrapolate values beyond the experimentally determined data set always resides with those who need to make the estimate. An expanded discussion of the petrology and a discussion of solution and phase-equilibrium data for the beryllium

minerals may be found in a companion paper by Barton (1986).

#### ACKNOWLEDGMENTS

We thank our U.S. Geological Survey colleague Harvey Belkin for examining our sample of bertrandite and bringing to our attention the presence of the claylike impurity. This work was

Table 19. Molar thermodynamic properties of bertrandite,  $\text{Be}_4\text{Si}_2\text{O}_7(\text{OH})_2$ , to 1800 K

Temp. T kelvin	Heat capacity $C_p^{\circ}$	Entropy $S_T^{\circ} - S_0^{\circ}$	Enthalpy function $(H_T^{\circ} - H_{298}^{\circ})/T$	Gibbs energy function $-(G_T^{\circ} - G_{298}^{\circ})/T$	Formation from the elements		
					Gibbs Enthalpy free energy kJ/mol		$\log K_f$
					-	-	
298.15	224.7	172.1	0.000	172.1	-4580.50	-4300.62	753.451
Uncertainty	0.8				5.47	5.46	
300	225.9	173.5	1.389	172.1	-4580.38	-4300.50	748.783
400	279.8	246.3	64.671	181.7	-4580.80	-4204.85	549.097
500	317.4	313.1	111.68	201.4	-4578.19	-4111.16	429.490
600	344.2	373.5	148.33	225.1	-4573.67	-4018.13	349.809
700	363.5	428.0	177.76	250.3	-4567.89	-3925.96	292.958
800	377.6	477.6	201.91	275.6	-4561.30	-3834.74	250.383
900	387.8	522.7	222.03	300.6	-4554.26	-3744.31	217.314
1000	395.1	563.9	239.00	324.9	-4547.02	-3654.65	190.899
1100	400.0	601.8	253.43	348.4	-4539.82	-3565.77	169.324
1200	403.2	636.8	265.79	371.0	-4532.82	-3477.58	151.375

Table 20. Molar thermodynamic properties of chrysoberyl,  $\text{BeAl}_2\text{O}_4$ , to 1800 K

Temp. kelvin	Heat capacity $C_p^{\circ}$	Entropy $S_T^{\circ} - S_0^{\circ}$	Enthalpy function $(H_T^{\circ} - H_{298}^{\circ})/T$	Gibbs energy function $-(G_T^{\circ} - G_{298}^{\circ})/T$	Formation from the elements		
					Gibbs Enthalpy free energy kJ/mol	log $K_f$	
					-----	-----	
298.15	105.4	66.25	0.000	66.25	-2298.49	-2176.16	381.254
Uncertainty		0.30			3.18	3.18	
300	106.0	66.90	0.652	66.25	-2298.45	-2175.35	378.762
400	130.8	101.1	30.366	70.75	-2299.35	-2133.28	278.578
500	145.9	132.0	52.073	79.97	-2298.92	-2092.91	218.645
600	155.8	159.6	68.590	90.99	-2297.86	-2051.84	178.628
700	162.7	184.1	81.568	102.6	-2296.53	-2010.90	150.055
800	167.5	206.2	92.025	114.2	-2295.19	-1970.21	128.642
900	171.2	226.2	100.63	125.5	-2293.98	-1929.70	111.997
1000	174.0	244.3	107.83	136.5	-2314.35	-1887.66	98.601
1100	176.3	261.0	113.96	147.1	-2312.96	-1845.05	87.614
1200	178.4	276.5	119.24	157.2	-2311.53	-1802.67	78.468
1300	180.2	290.8	123.86	167.0	-2310.07	-1760.25	70.728
1400	182.0	304.2	127.95	176.3	-2308.58	-1717.99	64.099
1500	183.8	316.9	131.61	185.3	-2307.07	-1675.99	58.363
1600	185.7	328.8	134.93	193.9	-2320.19	-1633.50	53.328
1700	187.8	340.1	137.98	202.1	-2318.21	-1590.63	48.874
1800	190.1	350.9	140.82	210.1	-2316.04	-1547.85	44.917
<i>Transitions in the reference state elements</i>							
Aluminum....	melting point		933.25 K				
Beryllium....	alpha - beta		1527 K				
	melting point		1560 K				

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