

Heat capacities of the alkali feldspars between 350 and 1000 K from differential scanning calorimetry, the thermodynamic functions of the alkali feldspars from 298.15 to 1400 K, and the reaction quartz + jadeite = analbite

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Abstract

The heat capacities, C_p° , of low albite, analbite, microcline, and high sanidine have been measured between 350 and 1000 K by differential scanning calorimetry. Our data for the alkali feldspars were combined with low-temperature heat-capacity and high-temperature heat-content data taken from the literature to derive the following equations:

$$\begin{aligned} C_p^\circ (\text{albite}) &= 583.9 - 0.09285T + 2.272 \times 10^{-5}T^2 - 6424T^{-1/2} \\ &\quad + 1.678 \times 10^6T^{-2} (\pm 0.6 \text{ percent } 298\text{--}1400 \text{ K}) \\ C_p^\circ (\text{analbite}) &= 671.4 - 0.1467T + 3.659 \times 10^{-5}T^2 - 7974T^{-1/2} \\ &\quad + 3.174 \times 10^6T^{-2} (\pm 0.3 \text{ percent } 298\text{--}1400 \text{ K}) \\ C_p^\circ (\text{microcline}) &= 759.5 - 0.2171T + 6.433 \times 10^{-5}T^2 - 9527T^{-1/2} \\ &\quad + 4.764 \times 10^6T^{-2} (\pm 0.6 \text{ percent } 298\text{--}1400 \text{ K}) \\ C_p^\circ (\text{high sanidine}) &= 693.4 - 0.1717T + 4.919 \times 10^{-5}T^2 - 8305T^{-1/2} \\ &\quad + 3.462 \times 10^6T^{-2} (\pm 0.5 \text{ percent } 298\text{--}1400 \text{ K}) \end{aligned}$$

where T is in kelvins and C_p is in units of $J/(\text{mol} \cdot \text{K})$. Smoothed values of the thermodynamic properties of these feldspar phases derived from the experimental data given in this report are listed in Robie *et al.* (1979).

Evaluation of the results of several phase equilibrium studies of the reaction quartz + jadeite = analbite yields -3029.870 ± 4.200 and -2851.300 ± 4.200 kJ/mol for the enthalpy and Gibbs free energy of formation, respectively, of jadeite at 298.15 K and 1 bar.

The enthalpy of the low albite–analbite transition determined from calorimetry using Amelia albite for low albite underestimates the magnitude of the enthalpy of the transition because Amelia albite is partially disordered (*e.g.*, Holm and Kleppa, 1968).

Introduction

The alkali feldspars are among the most important of the rock forming silicates. Although accurate heat-capacity, C_p° , and entropy, S° , data for the range 15 to 370 K exists for these minerals (Openshaw *et al.*, 1976), values of C_p° for temperatures above 400 K are based on a limited number of heat-content measurements at somewhat widely separated temperatures. The derived values for C_p° consequently have a large uncertainty.

The existing high-temperature heat-capacity values for the alkali feldspars are based on the heat-content data of White (1919) who determined $H_T^\circ - H_{273}^\circ$ for "microcline" and albite at 6 temperatures between 273 and 1373 K, on the heat-content data of Kelley *et al.* (1953) who measured $H_T^\circ - H_{298}^\circ$ for

low albite at 200 K intervals to a maximum temperature of 1273 K, and upon a single value each for $H_{971}^\circ - H_{298}^\circ$ for low albite and analbite obtained by transposed drop calorimetry (Holm and Kleppa, 1968).

The purpose of this study was to improve our knowledge of the high-temperature heat capacities of the alkali feldspars, low albite, analbite, microcline, and sanidine, and to apply the data for low albite and analbite to the phase equilibria studies of the reaction quartz + jadeite = analbite in order to resolve the difference of opinion regarding the magnitude of the Al/Si configurational entropy of analbite (Holm and Kleppa, 1968; Ribbe *et al.*, 1969; Ulbrick and Waldbaum, 1976; and, Kerrick and Darken, 1975), and to improve the value for the enthalpy of formation of jadeite.

Materials, apparatus, and experimental results

Our feldspar samples were all portions of the materials used for low-temperature heat-capacity measurements by Openshaw *et al.*, (1976) who give a complete description of the chemical and physical constants of these materials. The sample weights, determined by means of a Mettler M-5A microbalance and corrected for buoyancy, were 35.30, 31.98, 33.04, and 34.05 mg for low albite, analbite, microcline, and high sanidine, respectively. All samples were encapsulated in gold pans.

The high-temperature heat capacities were measured using a Perkin-Elmer DSC-2 differential scanning calorimeter similar to that described by O'Neill and Fyans (1971). The measurements were made at a heating rate of 10 K/min and a range setting (sensitivity) of 20.9 mJ/sec. The heat capacities were determined using the method outlined by O'Neill (1966) which utilizes synthetic sapphire as a C_p° reference standard. Our reference standard disk of synthetic sapphire weighed 30.66 mg. The heat-capacity values of Ditmars and Douglas (1971) for Standard Reference Material 720, synthetic sap-

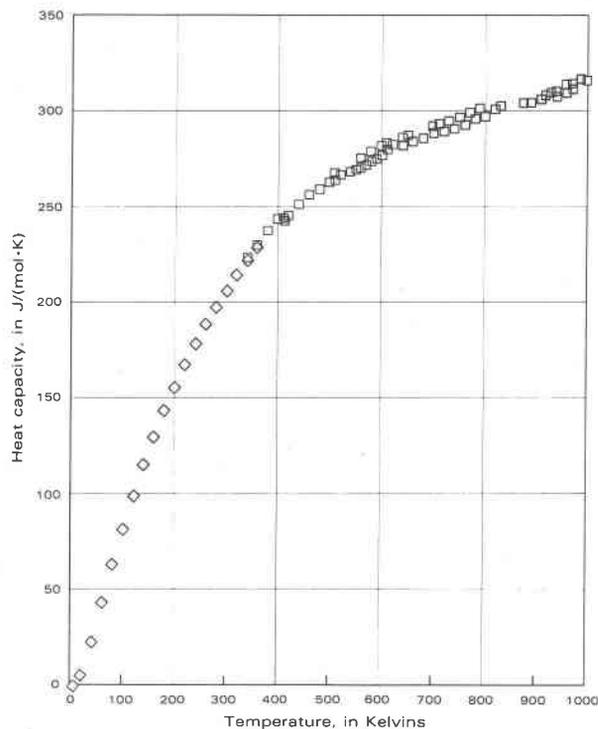


Fig. 1. Molar Heat capacity of low albite between 0 and 1000 K. The diamonds represent the least squares fit to the experimental data of Openshaw *et al.* (1976). The squares represent the experimental heat capacity values obtained by differential scanning calorimetry.

Table 1. Experimental heat capacities of 262.225 g (1 mole) of low albite, $\text{NaAlSi}_3\text{O}_8$. The equation represents the least squares fit to the experimental data.

Temp. K	Heat capacity J/(mol·K)	Temp. K	Heat capacity J/(mol·K)	Temp. K	Heat capacity J/(mol·K)
339.8	223.3	609.8	279.8	730.1	294.9
359.8	229.8	614.8	281.6	750.1	296.5
379.8	237.4	619.8	282.6	770.2	299.0
399.8	242.9	659.8	275.4	780.2	299.7
409.8	244.2	579.8	278.9	770.2	296.6
419.8	245.2	599.9	281.4	780.2	296.2
409.8	243.0	609.9	282.6	800.3	297.1
419.8	245.5	619.9	282.2	820.3	300.8
439.8	250.9	639.9	286.1	830.3	302.4
459.8	256.2	649.9	287.3	874.8	304.0
479.8	258.7	639.9	281.5	889.6	304.3
499.8	262.4	649.9	283.7	909.3	306.3
509.8	263.8	659.9	283.8	919.1	308.3
519.8	266.2	679.9	285.6	909.2	306.0
509.8	267.4	700.0	288.2	919.1	307.5
514.8	266.9	710.0	289.3	938.8	307.6
519.8	266.9	720.0	289.3	948.6	309.4
529.8	267.9	710.0	289.8	928.9	309.9
539.8	268.7	720.0	291.0	938.8	310.5
549.8	269.5	740.0	290.6	958.5	309.4
559.8	270.3	760.0	292.8	968.3	311.8
569.8	271.8	780.0	297.9	958.5	313.9
579.8	273.3	790.1	301.1	968.3	314.1
589.8	275.0	700.0	291.9	988.1	316.5
599.8	276.4	710.1	292.5	997.8	316.0

$$C_p^\circ = 583.9 - 0.09285 T + 2.272 \times 10^{-5} T^2 - 6424 T^{-0.5} + 1.678 \times 10^6 T^{-2}$$

(average deviation 0.6 percent, 298 to 1400 K)

phire, were used as the reference values. The temperature calibration of the calorimeter was checked by measuring the transition temperature of several inorganic compounds in the thermal standard sets, NBS-ICTA Standard Reference Materials 758 and 759 (McAdie *et al.*, 1972), specifically, indium, quartz, and potassium chromate.

Our experimental values for the molar heat capacities are listed in Tables 1-4 and are shown graphically in Figures 1-4. For the sake of clarity, some of the experimental C_p° values have been omitted from Figures 1-4. The gram-formula weights were calculated using the 1975 atomic weights (Commission on Atomic Weights, 1976), and the calorimetric unit conversion was made using 1 cal = 4.1840 J. None of the experimental C_p° measurements showed any evidence of the low albite-analbite or the microcline-high sanidine transitions.

Thermodynamic functions of low albite, analbite, microcline, and high sanidine

Our experimental C_p° measurements for the four alkali feldspars between 350 K and 1000 K were

Table 2. Experimental heat capacities of 262.225 g (1 mole) of analbite, $\text{NaAlSi}_3\text{O}_8$. The equation represents the least squares fit to the experimental data.

Temp.	Heat capacity	Temp.	Heat capacity	Temp.	Heat capacity
K	J/(mol·K)	K	J/(mol·K)	K	J/(mol·K)
339.8	221.1	609.8	282.4	730.1	294.6
359.8	227.9	614.8	282.7	750.1	295.1
379.8	234.9	619.8	283.0	770.2	296.8
399.8	241.7	559.8	274.0	780.2	297.1
409.8	243.4	579.8	276.7	770.2	297.7
419.8	245.5	599.9	278.8	780.2	300.1
409.8	242.1	609.9	280.4	800.2	300.3
419.8	245.0	619.9	280.1	820.3	301.7
439.8	249.8	639.9	284.0	830.3	303.1
459.8	254.1	649.9	286.3	874.8	305.0
479.8	258.8	639.9	283.4	889.6	304.8
499.8	262.7	649.9	286.1	909.3	305.7
509.8	264.1	659.9	286.2	919.1	308.2
519.8	266.3	679.9	287.5	909.3	308.2
509.8	266.9	700.0	290.1	919.1	309.3
514.8	267.6	710.0	291.3	938.8	312.2
519.8	268.6	720.0	291.2	948.6	313.2
529.8	269.3	710.0	291.6	928.9	311.0
539.8	270.8	720.0	294.2	938.8	310.3
549.8	272.7	740.0	295.5	958.5	311.3
559.8	274.1	760.0	297.4	968.3	311.1
569.8	275.4	780.0	299.4	958.5	309.8
579.8	277.1	790.1	301.9	968.3	310.2
589.8	278.8	700.0	292.2	988.1	311.9
599.8	279.9	710.1	293.6	997.8	310.4

$$C_p^{\circ} = 671.4 - 0.1467 T + 3.659 \times 10^{-5} T^2 - 7974 T^{-0.5} + 3.174 \times 10^6 T^{-2}$$

(average deviation 0.3 percent, 298 to 1400 K)

combined with the low-temperature heat capacities of Openshaw *et al.* (1976) and the heat-content data of Kelley (1960), and were fit by least squares to an equation of the form suggested by Haas and Fisher (1976). The equations were constrained to join smoothly with the C_p° values between 300 and 370 K obtained by accurate cryogenic adiabatic calorimetry.

Our final C_p° equations and the average deviation of the measured C_p° values from the derived equations are given in Table 1-4. The thermodynamic functions C_p° , $(H_T^{\circ} - H_{298}^{\circ})/T$, S_T° , and $-(G_T^{\circ} - H_{298}^{\circ})/T$ calculated from these equations are listed at 50 K intervals in Table 5 for analbite and at 100 K intervals in Robie *et al.* (1979) for low albite, microcline, and high sanidine. The errors in the derived thermodynamic functions are estimated to be ± 0.7 percent in C_p° and $(H_T^{\circ} - H_{298}^{\circ})/T$, and ± 0.4 percent in S_T° and $(G_T^{\circ} - H_{298}^{\circ})/T$ between 298 and 1000 K.

In Table 5 we have accepted the entropy for analbite reported by Openshaw *et al.* (1976). Openshaw *et al.* assumed analbite to be completely

disordered and accepted the value for S_0° , the zero point entropy, of analbite of $18.7 \text{ J}/(\text{mol} \cdot \text{K})$ [*i.e.*, $-4R(0.75 \ln 0.75 + 0.25 \ln 0.25)$]. Table 6 was calculated from our high temperature C_p° measurements on analbite and the $S_{298}^{\circ} - S_0^{\circ}$ data of Openshaw but assuming that $S_0^{\circ} = 12.6 \text{ J}/(\text{mol} \cdot \text{K})$, that is, assuming the aluminum avoidance principle applies to the structure of analbite (Kerrick and Darken, 1975 and Mazo, 1977). These models will be applied to calculations in a latter section of this paper.

Our heat capacity measurements for low albite may be compared directly with the values of Openshaw *et al.* (1976) and indirectly with the heat-content data given by White (1919) and by Kelley *et al.* (1953), if we first integrate our C_p° equation between 298.15 and T , where T is the temperature of the heat-content observation. The average deviation from the heat-content data of Kelley *et al.* is less than 0.5 percent.

It should be noted here that Openshaw *et al.* (1976) presented evidence for a transition in microcline at temperatures below 300 K. Because of the thermal hysteresis observed in the reported heat-capacity measurements, it is impossible to evaluate the magnitude of the microcline transition. Open-

Table 3. Experimental heat capacities of 278.333 g (1 mole) of microcline, $\text{KA1Si}_3\text{O}_8$. The equation represents the least squares fit to the experimental data.

Temp.	Heat capacity	Temp.	Heat capacity	Temp.	Heat capacity
K	J/(mol·K)	K	J/(mol·K)	K	J/(mol·K)
339.8	222.5	579.8	272.6	770.2	297.0
359.8	228.8	599.9	275.8	780.2	297.6
379.8	235.6	609.9	276.7	770.2	296.9
399.8	240.7	619.9	277.9	780.2	296.2
409.8	243.7	639.9	281.3	800.3	298.4
419.8	246.2	649.9	283.4	820.3	301.0
409.8	241.6	639.9	280.9	830.3	302.5
419.8	241.7	649.9	280.7	874.8	304.5
439.8	248.3	659.9	280.9	889.6	305.5
459.8	253.4	679.9	284.7	909.3	305.8
479.8	255.9	700.0	287.1	919.1	306.1
499.8	259.3	710.0	287.6	909.2	303.8
509.8	261.1	720.0	288.6	919.1	304.0
519.8	259.8	710.0	288.0	938.8	305.5
509.8	264.2	720.0	289.1	948.6	309.4
519.8	265.8	740.0	292.1	928.9	305.1
539.8	268.6	760.0	291.5	938.8	304.4
559.8	272.1	780.0	294.3	958.5	306.1
579.8	274.5	790.1	295.3	968.3	307.8
599.8	276.6	700.0	288.6	958.5	310.7
609.8	278.5	710.1	289.3	968.3	310.0
619.8	280.3	730.1	291.5	988.1	313.8
559.8	270.2	750.1	294.5	997.8	313.1

$$C_p^{\circ} = 759.5 - 0.2171 T + 6.433 \times 10^{-5} T^2 - 9527 T^{-0.5} + 4.764 \times 10^6 T^{-2}$$

(average deviation 0.6 percent, 298 to 1400 K)

Table 4. Experimental heat capacities of 278.333 g (1 mole) of sanidine, KAISi_3O_8 . The equation represents the least squares fit to the experimental data.

Temp. K	Heat capacity J/(mol·K)	Temp. K	Heat capacity J/(mol·K)	Temp. K	Heat capacity J/(mol·K)
339.8	220.1	579.8	277.7	770.2	295.9
359.8	227.1	599.9	281.8	780.2	297.1
379.8	233.7	609.9	282.0	770.2	295.4
399.8	240.1	619.9	283.0	780.2	297.1
409.8	243.5	639.9	286.7	800.2	297.0
419.8	246.7	649.9	287.6	820.3	299.4
409.8	243.9	639.9	281.7	830.3	299.7
419.8	246.3	649.9	281.9	874.8	305.3
439.8	250.2	659.9	282.6	889.6	305.6
459.8	256.1	679.9	285.6	909.3	306.5
479.8	258.5	700.0	288.0	919.1	308.7
499.8	263.2	710.0	288.9	909.3	308.6
509.8	268.6	720.0	290.3	919.1	308.9
519.8	267.0	710.0	289.0	938.8	308.5
509.8	262.3	720.0	289.5	948.6	310.1
519.8	263.4	740.0	291.3	928.9	310.9
539.8	267.9	760.0	292.1	938.8	309.7
559.8	271.8	780.0	295.8	958.5	311.7
579.8	275.4	790.1	297.0	968.3	312.3
599.8	280.1	700.0	291.1	958.5	309.5
609.8	280.6	710.1	292.8	968.3	309.7
619.8	282.8	730.1	295.0	988.1	311.9
559.8	274.7	750.1	295.7	997.8	312.0

$$C_p^\circ = 693.4 - 0.1717 T + 4.919 \times 10^{-5} T^2 - 8305 T^{-0.5} + 3.462 \times 10^6 T^{-2}$$

(average deviation 0.5 percent, 298 to 1400 K)

shaw *et al.* (1979) have shown that the molar volume of microcline shows a similar thermal hysteresis. Wyncke *et al.* (1981) examined the far infrared spectra of both natural microcline and the ion exchanged Amelia albite sample studied by Openshaw *et al.* and placed the transition temperature at 245 ± 5 K when the temperature of the sample is lowered.

Openshaw *et al.* (1976) obtained the entropy of microcline from a smoothed heat capacity curve fitted to the experimental data below 250 K, the experimental heat-capacity values for the stable microcline phase above 300 K, and an empirical set of values which smoothly connect the two experimental data sets. This procedure provides a first approximation to the entropy of microcline at 298.15 K and 1 bar. In order to improve upon this value, a new set of heat capacity data obtained at a considerably lower heating rate will be required. Wyncke *et al.* (1981) have not observed the hysteresis effect in natural microcline containing some albite lamellae. Therefore, it may be possible to evaluate this transition using a natural microcline sample.

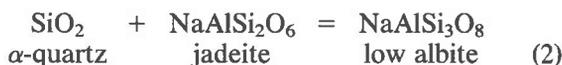
The albite breakdown reaction and the thermodynamic properties of jadeite

Birch and LeCompte (1960), Newton and Smith (1967), Bell and Roseboom (1969), Johannes, *et al.* (1971), Huang and Wyllie (1975) and Holland (1980) have studied the equilibrium represented by equation 1



at temperatures between 770 and 1650 K and at pressures between 14 and 33.5 kbar. Hlabse and Kleppa (1968) pointed out that the actual reaction studied by Birch and LeCompte and most probably by Newton and Smith involved analbite or partially disordered albite, not low albite.

The enthalpy change, ΔH , for reaction 2



at 298.15 K has been determined by Kracek, Neuvonen and Burley (1951) using HF-solution calorimetry.

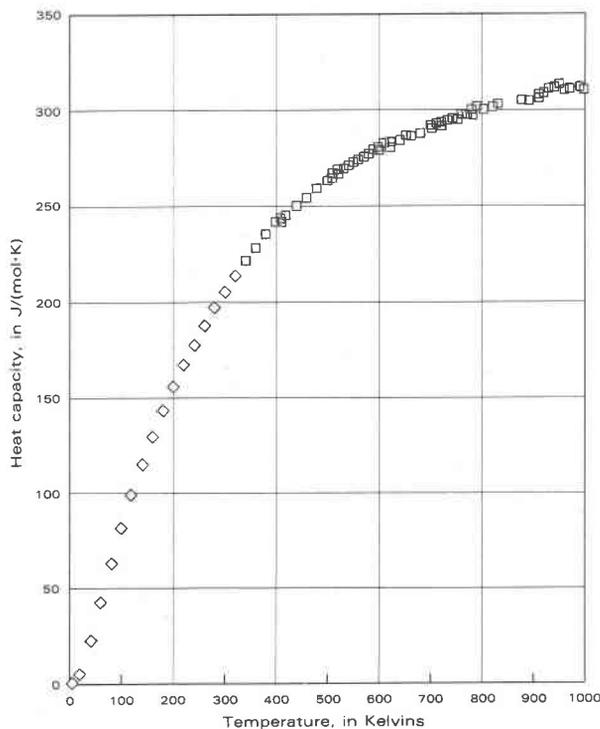


Fig. 2. Molar heat capacity of analbite between 0 and 1000 K. The diamonds represent the least squares fit to the experimental data of Openshaw *et al.* (1976). The squares represent the experimental heat capacity values obtained by differential scanning calorimetry.

etry and by Hlabse and Kleppa (1968) using the molten salt technique. The results of these two investigations are in poor agreement. Six values for ΔH_{298} of reaction 2 which range from $+3.34 \pm 1.00$ to -6.40 ± 1.55 kJ may be calculated from the data given by Kracek *et al.*, whereas Hlabse and Kleppa obtain -1.13 ± 1.38 kJ for reaction 2 at 964 K. The uncertainties associated with Hlabse and Kleppa's results have been recalculated to correspond to the convention regarding the uncertainty interval adopted by Kracek *et al.* and to standard thermochemical procedures.¹

The data of Kracek *et al.* (1951) may be improved somewhat by using the interpretation of Hemingway and Robie (1977) that one of the heats of solution reported by Kracek *et al.* for the two albite samples was in error and by using the heat of solution of quartz reported by Bennington, Ferrante, and Stuve (1978) and supported by the results and interpretation of Hemingway and Robie. These data will yield values of $+3.64 \pm 1.0$, $+1.09 \pm 1.71$, and -2.97 ± 1.13 kJ for the enthalpy of reaction 2 based upon jadeite from Japan ground in an agate mortar, jadeite from Burma, and jadeite from Japan ground in a mullite mortar, respectively, and albite from Varuträsk, Sweden.

Kracek and Neuvonen (1952), Holm and Kleppa (1968), Waldbaum and Robie (1971), Hovis (1971), and Newton *et al.* (1980) have determined the enthalpy of the transformation low albite \rightarrow analbite. Except for the data of Holm and Kleppa, the reported enthalpies of transformation are in reasonable agreement. The data of Holm and Kleppa must be viewed as a qualitative result (Anderson and Kleppa, 1969). The value of $+10.88 \pm 1.25$ kJ/mol

¹The standard uncertainty normally adopted in thermochemical investigations (Rossini and Deming, 1939) is twice the standard deviation of the mean (sdm) *i.e.*, $2[\sum(x_i - \bar{x})^2/n(n-1)]^{1/2}$. This corresponds to a probability of 96 percent that the correct value is within the interval $(\bar{x} - 2 \text{ sdm})$ to $(\bar{x} + 2 \text{ sdm})$. Uncertainties reported by Hlabse and Kleppa are apparently calculated from the expression $[\sum(\bar{x}_i - x_i)^2/n]^{1/2}$ which is actually the standard deviation for an individual observation, not for the mean value obtained for *n* observations. (See for example, Meyer (1975)). Recalculation of the uncertainties reported by Hlabse and Kleppa is done for the convenience of the reader. That is, a comparison of results is simplified if the data are all presented in the same form. It should be noted that, in general, there are not a sufficient number of enthalpy of solution measurements to provide a valid statistical set (*e.g.*, Meyer).

We have also reported the average deviation of our heat capacity data from the least squares fit to the data because this is the form in which the data have traditionally been present (*e.g.*, Kelley, 1960).

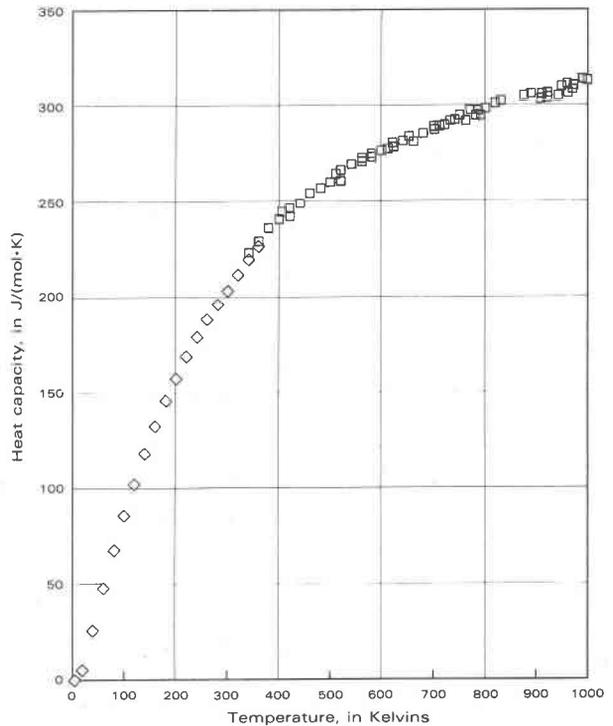


Fig. 3. Molar heat capacity of microcline between 0 and 1000 K. The diamonds represent the least squares fit to the experimental data of Openshaw *et al.* (1976). The squares represent the experimental heat capacity values obtained by differential scanning calorimetry.

Table 5. Molar thermodynamic properties of analbite assuming $S_0^* = 18.7$ J/mol · K

Temp. T Kelvin	Heat Capacity C_p^*	Entropy S_T^* J/(mol·K)	Enthalpy Function $(H_T^* - H_{298}^*)/T$	Gibbs energy Function $-(G_T^* - H_{298}^*)/T$
298.15	204.8	226.4	0.0	226.4
350	224.2	260.8	31.8	229.0
400	239.7	291.8	56.9	234.9
450	252.6	320.8	77.9	242.9
500	263.3	348.0	95.9	252.0
550	272.3	373.5	111.6	261.9
600	279.8	397.5	125.3	272.2
650	286.3	420.2	137.4	282.7
700	291.7	441.6	148.3	293.3
750	296.4	461.9	158.0	303.9
800	300.5	481.2	166.8	314.4
850	304.0	499.5	174.7	324.7
900	307.1	516.9	182.0	334.9
950	309.9	533.6	188.7	345.0
1000	312.3	549.6	194.8	354.8
1050	314.5	564.9	200.4	364.4
1100	316.5	579.6	205.7	373.9
1150	318.3	593.7	210.5	383.1
1200	320.1	607.2	215.1	392.2
1250	321.7	620.3	219.3	401.1
1300	323.2	633.0	223.3	409.7
1350	324.8	645.2	227.0	418.2
1400	326.2	657.1	230.5	426.5

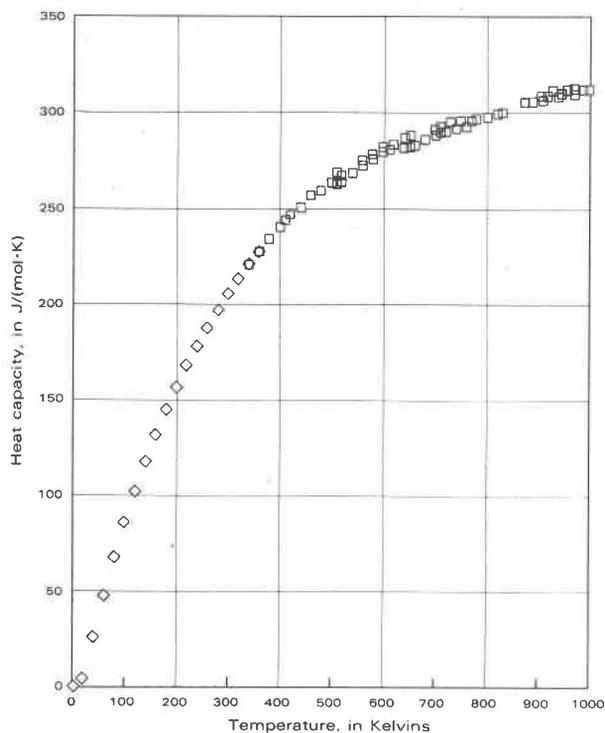


Fig. 4. Molar heat capacity of sanidine between 0 and 1000 K. The diamonds represent the least squares fit to the experimental data of Openshaw *et al.* (1976). The squares represent the experimental heat capacity values obtained by differential scanning calorimetry.

reported by Waldbaum and Robie was adopted for our calculations.

By combining the data given above, we calculate values ranging from 14.52 ± 1.58 to 7.91 ± 1.68 kJ for reaction 1 from HF solution calorimetry and 11.98 ± 1.86 kJ from molten salt calorimetry all at 298.15 and 1 bar with analbite as the Na-feldspar. The results presented above will be discussed in greater detail in a later section of this paper.

We wish to use the equilibrium data to see if we can improve the calorimetric values of ΔH for the albite-jadeite reaction and to examine the validity of the aluminum avoidance principle (Lowenstein, 1954) as it has been applied to Na-feldspars by Kerrick and Darken (1975) and Mazo (1977). We shall consider two models. For model 1 we assume that the zero-point entropy of analbite is 18.7 J/(mol · K), and in model 2 we will use 12.6 J/(mol · K) for S_0° of analbite.

If we assume that the phase-equilibria data presented by Holland (1980), Huang and Wyllie (1975), Hays and Bell (1973), Johannes *et al.* (1971), Newton and Smith (1967), and Birch and LeComte

(1960) represent, in fact, phase reversal information for reaction 1, then we may construct curves that fit these reaction data and that have the appropriate slopes for the entropy models 1 and 2 cited above. The two curves are shown in Figure 5 together with selected phase-equilibria data. The enthalpies of reaction at 298.15 K given in Table 7 are calculated from these curves using the third law method (equation 3)

$$\Delta G_{P,T} = 0 = \Delta H_{298}^\circ + T\Delta[(G_T^\circ - H_{298}^\circ)/T] - (P - 1)\Delta V_T^\circ \quad (3)$$

described by Lewis and Randall (1961), Robie (1965) and by Krupka, *et al.* (1979) and using the molar volumes at high temperatures given by Cameron *et al.* (1973) and by Yoder and Weir (1951) for jadeite; by Stewart and von Limbach (1967), and Grundy and Brown (1969) for analbite, and Skinner (1966) for quartz. The estimated equilibrium pressures have been rounded to the nearest 0.1 kbar and no estimate of the effect of pressure on the molar volume has been made.

We have given rather little weight to the single data point of Bell and Roseboom (1969) at 33.5 kbar because of the unknown, and unapplied, friction correction and because it would require extrapolating the C_p data for jadeite about 500 K.

Table 6. Molar thermodynamic properties of analbite assuming $S_0^\circ = 12.6$ J/mol · K

Temp. T Kelvin	Heat Capacity C_p°	Entropy S_T° J/(mol · K)	Enthalpy Function $(H_T^\circ - H_{298}^\circ)/T$	Gibbs energy Function $-(G_T^\circ - H_{298}^\circ)/T$
298.15	204.8	220.3	0.0	220.3
350	224.2	254.7	31.8	222.9
400	239.7	285.7	56.9	228.8
450	252.6	314.7	77.9	236.8
500	263.3	341.9	95.9	245.9
550	272.3	367.4	111.6	255.8
600	279.8	391.4	125.3	266.1
650	286.3	414.1	137.4	276.6
700	291.7	435.5	148.3	287.2
750	296.4	455.8	158.0	297.8
800	300.5	475.1	166.8	308.3
850	304.0	493.4	174.7	318.6
900	307.1	510.8	182.0	328.8
950	309.9	527.5	188.7	338.9
1000	312.3	543.5	194.8	348.7
1050	314.5	558.8	200.4	358.3
1100	316.5	573.5	205.7	367.8
1150	318.3	587.6	210.5	377.0
1200	320.1	601.1	215.1	386.1
1250	321.7	614.2	219.3	395.0
1300	323.2	626.9	223.3	403.6
1350	324.8	639.1	227.0	412.1
1400	326.2	651.0	230.5	420.4

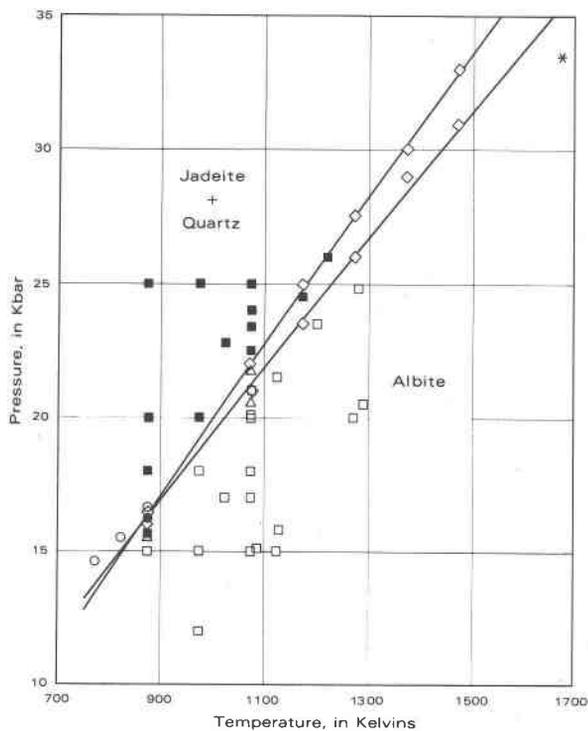


Fig. 5. Experimental data and calculated equilibrium curves for the reaction quartz + jadeite = analbite. The filled circle is the "consensus" value of Johannes and others (1971); squares are the data of Birch and Le Comte (1960); triangles are the data of Huang and Wyllie (1975); asterisk is the data of Bell and Roseboom (1969); open circles are the data of Newton and Smith (1967); diamonds are the data of Holland (1980). Above 900K, the upper solid line is the calculated curve based upon model 1, S_0° analbite = 18.7 J/(mol · K), and the lower solid line is the calculated curve based on model 2, S_0° analbite = 12.6 J/(mol · K).

Using the tabulated values for $(G_T^{\circ} - H_{298}^{\circ})/T$ for analbite from Tables 5 and 6 and the Gibbs energy functions for jadeite and quartz adopted by Robie *et al.* (1979) we obtain +16.33 kJ for ΔH_{298}° of reaction 1 assuming model 1 for S_0° of analbite. For model 2 we calculate +11.01 kJ for ΔH_{298}° of reaction 1. In combining the calorimetric and equilibrium data it must be recalled that a major contribution to the uncertainty is the ± 1.3 J/(mol · K) associated with the entropy of jadeite caused by the extrapolation of the C_p° data from 50 to 0 K. This corresponds to ± 1135 J at 873 K. The largest part of the uncertainty in the entropy could be eliminated by measurements of the heat capacity of jadeite in the temperature range 5–50 K.

Using the value of ΔH_{298}° (reaction) calculated on the basis of model 1, together with the values of the enthalpy of formation at 298.15 K, $\Delta H_{f,298}^{\circ}$, for

quartz and analbite adopted by Robie *et al.* (1979), we obtain -3029.87 ± 4.2 kJ/mol for jadeite. Combining this value with the entropy of jadeite and the requisite and entropies of the elements from Robie *et al.*, we get -2851.30 ± 4.20 kJ/mol for the Gibbs free energy of formation, $\Delta G_{f,298}^{\circ}$, of jadeite. Similarly, using model 2 we obtain -3024.55 ± 4.20 kJ/mol and -2845.98 ± 4.20 kJ/mol for the enthalpy and Gibbs free energy of formation, respectively, for jadeite. The values for the molar volumes at 873 K used in our calculations are 23.72 ± 0.01 , 61.34 ± 0.40 , and 101.86 ± 0.12 cm³ for β -quartz, jadeite and analbite, respectively, and 106.4 ± 0.2 , 352.6 ± 1.3 , 507.6 ± 0.5 , and 501.5 ± 0.5 J/(mol · K) for the entropies of β -quartz, jadeite, analbite (model 1), and analbite (model 2), respectively. Alpha quartz is the stable phase under the conditions of the phase equilibria experiments cited above, however, no correction was estimated for the difference in heat capacity, entropy, or volume between α - and β -quartz as the differences are small and of the order of the uncertainty in the estimate.

The striking feature seen in Figure 5 is that the curve constructed for model 1 (complete Al/Si disorder) parallels the first occurrence of jadeite plus quartz in the reaction data of Birch and LeComte (1960) whereas the curve constructed for model 2 (aluminum avoidance) is parallel to the first occurrence of albite in the Birch and LeComte data, with both curves having 873 K and about 16.5 kbar as a common point, which is consistent with the value given by Johannes *et al.* (1971) as a "consensus" value representing the mean of the results from six laboratories. Holland (1980) has shown that at the higher temperatures and pressures in his study, the original albite broke down to jadeite and quartz prior to the adjustment of the system to the desired run temperature and pressure. Consequently, the first occurrence of albite in these runs represents conditions similar to those of Birch and LeComte, but at higher temperatures and pressures. Because the slope of the curve consistent with model 1 represents essentially the maximum slope which can be constructed from this data set, whereas that consistent with model 2 similarly represents the minimum slope, an average curve constructed through the experimental data must have a slope greater than that predicted by model 2. Consequently, the available equilibrium data do not unambiguously differentiate between the two models. We point out, however, that if the true equilibrium curve has a slope greater than that calculated

Table 7. Thermodynamic calculation of the equilibrium pressure for the reaction jadeite + quartz = analbite at temperatures between 700 and 1300 K for models 1 ($S_0^\circ = 18.7 \text{ J/mol} \cdot \text{K}$) and 2 ($S_0^\circ = 12.6 \text{ J/mol} \cdot \text{K}$) of the zero point entropy of analbite.

Temp.	ΔV_T°	Model 1				Model 2		
		ΔH_{298}°	P	$\Delta[(C_T^\circ - H_{298}^\circ)/T]$	ΔH_{298}°	P	$\Delta[(C_T^\circ - H_{298}^\circ)/T]$	
K	J/bar	J	kbar	J/(mol·K)	J	kbar	J/(mol·K)	
700	1.713	16330	11.3	-50.98	11010	11.9	-44.88	
800	1.703	16330	14.3	-50.86	11010	14.2	-44.76	
900	1.686	16330	17.3	-50.60	11010	17.2	-44.50	
1000	1.707	16330	20.0	-50.44	11010	19.5	-44.34	
1100	1.726	16330	22.6	-50.23	11010	21.7	-44.13	
1200	1.739	16330	25.2	-50.09	11010	24.0	-43.99	
1300	1.761	16330	27.6	-49.89	11010	26.1	-43.79	

using model 2 this would be sufficient to exclude the aluminum avoidance model (model 2).

Holland (1980) has recently examined reaction 1 using a similar data set to that used in this study. Holland's calculated values of the enthalpies of formation of jadeite and high albite and the entropy of high albite differ from the values derived earlier in this paper. It is important to examine the sources of these differences.

Holland (1980) calculated the enthalpy of formation of jadeite from the molten salt calorimetric study of reaction 1 reported by Hlabse and Kleppa (1968) and the ancillary thermodynamic data for quartz and low albite taken from Robie *et al.* (originally printed in 1978, revised 1979). Holland then calculated the enthalpy of formation and the entropy of analbite from the enthalpy of formation of jadeite, his phase equilibria results, and the necessary ancillary thermodynamic data from Robie *et al.* (1979), thermal expansion data from Skinner (1966) and compressibility data from Birch (1966). Finally, Holland calculated an entropy and enthalpy of disorder of high albite.

Our procedure followed the reverse thermodynamic cycle, calculating the enthalpy of formation of jadeite from our selected value for the enthalpy of formation and entropy chosen for analbite. The uncertainties involved in proceeding through the thermodynamic cycle in either direction are equivalent, but the choices in interpretation have different consequences.

The initial step in each study was the somewhat arbitrary choice of a specific value for the initial reaction considered in the cycle from the several

values available in the literature. Holland (1980) accepted the enthalpy of reaction 2 as given by Hlabse and Kleppa (1968) as -0.15 kJ . As noted earlier, Kracek *et al.* (1951) have reported several values for the enthalpy of solution of jadeite and low albite from which the enthalpy of reaction 2 may be calculated.

Six values for reaction 2 may be calculated from the two values for the enthalpy of solution of the two low albite samples, from the enthalpy of solution of jadeite from Burma, and from the two different values for the enthalpy of solution of jadeite from Japan prepared in mortars of different composition. Three of these values may be eliminated, as discussed in an earlier section, on the basis of the apparently erroneous enthalpy of solution reported for Amelia albite (*e.g.*, Hemingway and Robie, 1977). However, sufficient data are not available to discriminate between the remaining values.

Hlabse and Kleppa (1968) have reported two enthalpy values for reaction 2. Hlabse and Kleppa attempted to make a correction for the impurities in their natural samples through a comparison of the enthalpy of solution of glass samples prepared from these materials. This procedure is questionable on several grounds (*e.g.*, a partial crystallization of the glass in the calorimeter prior to that portion of the sample undergoing dissolution or different coordination of the components in the jadeite + quartz glass than in the crystalline form to which it is compared). However, the presence of impurities within a sample can have a major effect upon the observed enthalpies of solution. The two values for

the enthalpy of reaction 2 at 298.1 K are -0.15 ± 1.02 (uncorrected) and 1.10 ± 1.22 (corrected) kJ.

Once Holland (1980) had accepted a value for the enthalpy of formation of jadeite, he was able to calculate values for the enthalpy of formation and entropy of high albite based upon his study of reaction 1 by phase equilibria. Holland obtained an entropy of disorder of $14.2 + 2.4 \text{ J}(\text{mol} \cdot \text{K})^{-1}$ which is larger than that representative of a configuration of Al and Si obeying the aluminum avoidance principle, but considerably smaller than the entropy associated with complete Al/Si disorder. The enthalpy of disorder (calculated with respect to low albite from the derived enthalpy of the reaction and based upon the selected enthalpy of formation of jadeite) was also considered less than the value accepted by Holland as the best value for the enthalpy of the reaction low albite \rightarrow analbite, (that value obtained from R.C. Newton and T.V. Charlú through personal communication).

The published values for the enthalpy of the reaction low albite \rightarrow analbite are in poor agreement when taken at face value. Kracek and Neuvonen (1952) have reported values of 9.20 and 10.17 kJ for samples prepared from Amelia albite and Varuträsk, Sweden, albite, respectively; Waldbaum and Robie (1971) and Hovis (1971) have reported 10.88 kJ, and Thompson *et al.* (1974) have given 11.00 kJ for samples prepared from Amelia, Va. albite. Each of the studies was based upon the difference in the enthalpies of solution of the original low albite and a heat-treated portion of the same source material in aqueous hydrofluoric acid of about 20 percent HF concentration by weight at temperatures below 355 K.

Holm and Kleppa (1968) found 13.87 kJ and Newton and Charlú (unpub. data cited by Holland, 1980) obtained 14.22 kJ for the enthalpy of the reaction low albite \rightarrow analbite at 298.15 K from molten salt solution calorimetry at about 970 K, where the results were extrapolated to 298.15 K using the data reported in this paper. Newton *et al.* (1980) have recently given the value 11.97 kJ as the preferred enthalpy of the reaction low albite \rightarrow analbite. Newton *et al.* were unable to determine the source of the discrepancy between this value and the value which they had given to Holland (see the discussion by Newton *et al.*).

The enthalpy of solution of the Amelia low albite sample given by Kracek and Neuvonen (1952, first reported by Kracek *et al.*, 1951) has been shown to

be inconsistent with other results on Amelia albite (Hemingway and Robie, 1977). Furthermore, Kracek and Neuvonen failed to describe the heat treatment applied to the low albite samples. Hence, the results of Kracek and Neuvonen must be viewed as qualitative.

The enthalpy of solution of low albite and analbite reported by Thompson *et al.* (1974) are 1 percent too large as a consequence of an error in the design of the energy measurement circuit (B.S. Hemingway, 1978, personal communication). Correcting each enthalpy of solution reported by Thompson *et al.* yields a value for the enthalpy of the reaction low albite \rightarrow analbite of 10.88 kJ, in exact agreement with the values reported by Waldbaum and Robie (1971) and Hovis (1971). Waldbaum (1966) reported 11.016 kJ for the enthalpy of reaction of low albite \rightarrow analbite at 298.15 K. However, corrections to Waldbaum's computer programs used for raw data reduction (B.S. Hemingway and R.A. Robie, personal communication, 1968) resulted in the corrected value of 10.88 kJ reported by Waldbaum and Robie (1971) for the same raw data set.

Portions of the samples of low albite and analbite used by Openshaw *et al.* (1976) and in this study for the heat-capacity measurements were used by Hovis (1971) for the measurement of the enthalpy of the reaction low albite \rightarrow analbite. Thompson *et al.* (1974) determined the enthalpy of the reaction using a portion of the analbite sample used by Hovis and a portion of the low albite sample used by Waldbaum and Robie (1971). Thus the samples used to determine the enthalpy of reaction by aqueous HF calorimetry have been intercompared.

The enthalpy of the reaction low albite \rightarrow analbite determined by Newton *et al.* (1980) from molten salt calorimetry at 970 K is midway between the earlier result reported by Holm and Kleppa (1968), also from molten salt calorimetry at about 970 K, and the results from low-temperature HF solution calorimetry.

Holm and Kleppa (1968) estimated the heat content ($H_{971} - H_{298}$) of low albite and analbite by the method of transposed drop calorimetry. Using their results, the enthalpy of the reaction low albite \rightarrow analbite is 10.92 kJ at 298.15 K and the results of Newton *et al.* (1980) become 8.66 kJ at 298.15 K. The heat contents determined by Holm and Kleppa for low albite and analbite are smaller (0.6 percent) and larger (0.9 percent), respectively, than the values calculated from the experimental heat capac-

ities reported in this study. The values of the heat content obtained by Holm and Kleppa must be considered qualitative (Hlabse and Kleppa, 1968).

The uncertainties in the enthalpies of the reaction low albite \rightarrow analbite range from ± 1.1 to ± 1.7 kJ. An additional ± 0.1 and ± 0.2 kJ arise from the heat-content values calculated from the heat capacities reported in this study and from the values of Holm and Kleppa, respectively. Thus the uncertainties in the reaction enthalpies of the data set obtained from molten salt calorimetry and from aqueous hydrofluoric solution calorimetry are of equal magnitude and nearly overlap.

Holland (1980) has attributed all of the observed enthalpy of the reaction low albite \rightarrow analbite determined by the molten salt calorimetric technique to an enthalpy of disorder. Helgeson *et al.* (1978) suggested that the two step enthalpy change observed by Holm and Kleppa (1968) represented a disorder enthalpy and an enthalpy of a displacive inversion. The interpretation of Helgeson *et al.* would appear incorrect in light of the analysis by Smith (1974) of the data relevant to the temperature of the monoclinic-triclinic inversion. Simply stated, the analbite samples used in each of these investigations were equilibrated at temperatures of 1318 K or higher for more than 668 hours, which should have produced nearly monoclinic topochemistry (see Smith, 1974, for a discussion of the difference between topologic and topochemical symmetry and the importance of topochemical symmetry to the monoclinic-triclinic inversion in high albite) in each sample. These analbite samples should invert when heated to temperatures near 1253K (*e.g.*, Smith). The molten salt calorimetric solvent was maintained at about 970 K, well below the inversion temperature, that is, within the analbite field not the monalbite field. No evidence was found in the experimental heat-capacity data to suggest that analbite underwent partial or full inversion in the temperature range of 350 to 1000 K during the time interval required to measure the heat capacity with the differential scanning calorimeter. Holland found no curvature in this calculated equilibrium curve for reaction 1 and concluded that albite does not undergo Al/Si ordering in the experimental runs, also contradicting the interpretation of Helgeson, *et al.* (1978).

Winter *et al.* (1979) have shown that the synthesis procedures used by Holm and Kleppa (1968), Waldbaum and Robie (1971), Hovis (1971), and Thompson *et al.* (1974) should not produce complete Al/Si

disorder in the analbite samples as suggested by R.C. Newton (cited by Holland, 1980). The energy differences associated with the slight deviations from monoclinic topochemistry must be small as suggested by Winter *et al.*

Holland (1980) suggests that the sample prepared by Newton and Charlu at 1473 K and 20 kbar should represent maximum Al/Si disordering. It is not clear, however, that this statement is true. Winter *et al.* (1979) have shown that true monoclinic topochemistry was developed only in a sample which was held near the melting point at 1 bar (sample exhibited a glassy rind). If the analbite sample crystallized from albite glass by Newton and Charlu was anhydrous, then crystallization occurred nearly 100 degrees below the anhydrous melting curve for albite. Winter *et al.* suggest that a 20 K difference at 1 bar is significant for heat treated low albite samples. At 20 kbar, a difference of 100 degrees could significantly effect the degree to which the analbite approached monoclinic topochemistry. If Newton and Charlu synthesized analbite in the presence of a small amount of water the melting curve would be significantly depressed (*e.g.*, Eggler and Kadik, 1979); however this would greatly increase the risk of the synthetic analbite containing a small glass fraction which in turn would yield erroneous values for the enthalpy of solution (about 600 J for each 1 percent of glass, *e.g.*, Hlabse and Kleppa, 1968) and consequently an overestimate of the enthalpy of the reaction low albite \rightarrow analbite. Therefore, it cannot be assumed *a priori* that the sample prepared by Newton and Charlu at 20kbar represents the best analbite sample and that the thermodynamic data based upon that sample is better than that derived from other synthetic analbites, a conclusion also reached by Newton *et al.* (1980). An analysis of the type performed by Winter *et al.* (1979) is needed for the sample prepared by Newton and Charlu and for the samples used in the HF calorimetric studies.

Of greater importance in this discussion of the literature data for the entropy and enthalpy of disorder of analbite are two observations. First, Holm and Kleppa (1968) have shown that Amelia albite is not fully ordered. From the site occupancy data given by Bragg and Claringbull (1965) for Amelia albite, Holm and Kleppa calculated an entropy of disorder of $18.45 \text{ J}(\text{mol} \cdot \text{K})^{-1}$ for analbite and $3.39 \text{ J}(\text{mol} \cdot \text{K})^{-1}$ Amelia albite. From the site occupancy data of Harlow and Brown (1980) we obtain $2.5 \text{ J}(\text{mol} \cdot \text{K})^{-1}$ for Amelia albite. Second,

Waldbaum and Robie (1971) have shown that the enthalpy of solution of Amelia albite differs by about 2.2 kJ mol^{-1} from the enthalpy of solution of a sodium exchanged Amelia microcline. These results taken together indicate that the heat treated analbite sample closely approaches complete Al/Si disorder. Errors to be assigned to the enthalpy of the reaction low albite \rightarrow analbite as determined by calorimetry arise from the use of partially disordered Amelia albite. As Amelia albite was used as the reference low albite in all of these studies, including the study of Newton and Charlu, major energy differences cannot be expected to arise from differences between the degree of Al/Si disorder reached in the samples used in these studies, unless they arise from differences in the original Amelia albite. If the difference between the enthalpy of solution of Amelia albite and the sodium-exchanged Amelia microcline represents the difference between partially disordered and ordered low albite, then 2.2 kJ should be added to the enthalpies calculated for reaction 1 from the calorimetric studies. This would change the observed range of calorimetric values for reaction 1 to 16.7 ± 1.7 to $10.1 \pm 1.7 \text{ kJ}$. These values bracket the results obtained earlier from the phase equilibrium data.

We wish to note at this point that we have determined the enthalpy of formation of jadeite from the thermodynamic parameters for analbite. These values were determined independent of the results for low albite and therefore are not subject to errors in the thermochemical data for low albite. The calorimetric data, however, is referenced to low albite and therefore would be subject to a systematic error in the data for low albite.

In the final analysis, the major difference between this study and that of Holland (1980) lies in the interpretation of phase equilibria experiments with regard to the Al/Si disorder in the analbite. Although a curve representing an entropy of disorder of $14.2 \text{ J}(\text{mol} \cdot \text{K})^{-1}$ may be constructed through the experimental data given by Holland, no structural or chemical constraint could be called upon to limit the disorder reached by the Al and Si to a constant value (over the 600 K temperature range studied) between the fully disordered model and that which would be applicable to the chemical constraint of aluminum avoidance. Furthermore, experience would lead us to expect the degree of disorder to increase with increasing temperature (e.g., Smith, 1974) unless there is some constraint upon the disorder. Consequently, we feel it would

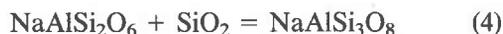
be extremely fortuitous to find the same degree of partial disorder developed over a 600 K interval without some external constraint and therefore we reject the interpretation made by Holland.

Only two constraints could fix the Al/Si disorder over the temperature range investigated by Holland (1980). These are represented by the aluminum avoidance model and by the model which represents complete Al/Si disorder. As noted earlier, a curve consistent with either model can be constructed through the experimental phase equilibrium data for this system (see Figure 5). Therefore, the phase equilibrium data cannot discriminate between the two models.

Noting site occupancy calculations such as those of Winter *et al.* (1979) and the observation that a disorder model compatible with the principle of aluminum avoidance would require a curve having the minimum acceptable slope through the phase equilibrium data given in Figure 5 has led us to conclude that the analbite which reacts to form jadeite and quartz is totally disordered.

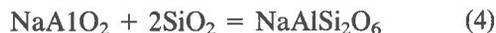
Subsequent to the completion of this study, Navrotsky *et al.* (1980) published data for the enthalpy of solution of NaAlO_2 , quartz, Amelia albite, and jadeite in $2\text{PbO} \cdot \text{B}_2\text{O}_3$ at 985 K. These data when combined with the appropriate ancillary thermodynamic data may be used to calculate the enthalpy of formation of jadeite at 298.15 K.

From reaction 2



we obtain $\Delta H_r = 3.30 \text{ kJ}$ at 985 K. Combining this value with the enthalpy of formation of quartz and Amelia albite at 985 K as given in Robie *et al.* (1979) we obtain -3034.5 kJ/mol for the enthalpy of formation of jadeite at 985 K and $-3029.2 \pm 3.7 \text{ kJ/mol}$ for the same quantity at 298.15 K (using the necessary heat-content data given in Robie *et al.*).

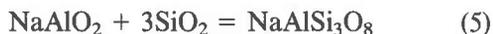
Following the same procedure, the enthalpy of formation of jadeite at 298.15 K derived from reaction 4



was $-3029.5 \pm 1.6 \text{ kJ/mol}$ (based upon the data for NaAlO_2 given in the JANAF thermochemical tables, Stull and Prophet, 1971).

We may make a further comparison through reaction 5 and note that the enthalpy of formation of Amelia albite at 298.15 K derived from the enthalpies of solution of quartz, Amelia albite, and NaAlO_2 given by Navrotsky *et al.* (1980) and the enthalpies

of formation of NaAlO_2 and quartz cited above was -3935.2 ± 1.5 kJ/mol, a value which is identical with the value -3935.1 ± 3.4 kJ/mol given by Robie *et al.* (1979) for the same phase.



Although these values are essentially identical to the value of the enthalpy of formation of jadeite at 298.15 reported here (-3029.9 ± 4.2 kJ/mol) and are identical with our earlier value (Robie *et al.*, 1979), of -3029.4 ± 4.2 kJ/mol provided without a detailed derivation, but determined through the procedures listed in this paper (without the data of Holland, 1980), the agreement may be fortuitous considering the assumptions which are necessary in order to derive these sets of values.

Conclusions

Heat-capacity measurements obtained by differential scanning calorimetry on samples of low albite, analbite, microcline, and sanidine improve our knowledge of the thermodynamic properties of these phases between 298.15 K and 1000 K. Smoothed values of the revised thermodynamic properties of these phases are given by Robie *et al.* (1979).

Our measurements of the thermodynamic properties of low albite and analbite have been combined with the phase-equilibrium data of Holland (1980), Huang and Wyllie (1975), Johannes *et al.* (1971), Bell and Roseboom (1969), and Birch and LeCompte (1960) in order to derive an improved value of the enthalpy of formation of jadeite, -3029.87 ± 4.20 kJ/mol.

We agree with the conclusion reached by Holland (1980) that albite does not undergo differential Al/Si ordering in the phase-equilibrium experiments in the temperature range of 873 to 1473 K. We disagree with Holland's calculated entropy of disorder for high albite of $14.2 \text{ J}(\text{mol} \cdot \text{K})^{-1}$ and conclude that fully disordered analbite appears to be the equilibrium phase which breaks down to form jadeite and quartz. The albite phase which grows at the expense of jadeite and quartz appears to closely approximate analbite obeying the chemical constraints of the aluminum avoidance principle. The consequence of this interpretation is, of course, that the phase equilibria data do not represent equilibrium reversals for a single phase.

Finally, because Amelia albite has been used as the reference low albite phase and Holm and Kleppa (1968) and Harlow and Brown (1980) have

shown that Amelia albite is partially disordered, the current values for the enthalpy of the low albite-analbite transition determined calorimetrically from the difference in the enthalpies of solution of Amelia albite and heat treated Amelia albite must be viewed as a minimum value.

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*Manuscript received, April 27, 1981;
accepted for publication, July 27, 1981.*