

## THE THERMOCHEMISTRY OF JADEITE

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## ABSTRACT

The enthalpy changes associated with the mineralogical reactions

- (1) Albite = Jadeite + Quartz
- (2) Albite + Nepheline = 2 Jadeite

have been redetermined at 964°K by oxide melt solution calorimetry. For (1) we find  $\Delta H^{\circ}_{964} = +0.27 \pm 0.50$  kcal/mole; for (2)  $\Delta H^{\circ}_{964} = -6.85 \pm 0.25$  kcal/mole. When these results are combined with available heat capacity and entropy data they may be compared with the results of the room temperature thermochemical work of Kracek, Neuvonen and Burley and with high temperature-high pressure equilibrium studies. In order to achieve agreement with equilibrium work above 700°C it is essential to take into account the enthalpy and entropy changes associated with the low→high albite transformation.

## INTRODUCTION

During the past fifteen to twenty years there has been much interest in the thermodynamic properties and stability relations of jadeite. The geological interest in this mineral is centered on two important mineralogical reactions which are of special significance in petrology and geophysics:

- (1) albite ( $\text{NaAlSi}_3\text{O}_8$ ) = jadeite ( $\text{NaAlSi}_2\text{O}_6$ ) + quartz ( $\text{SiO}_2$ )
- (2) albite ( $\text{NaAlSi}_3\text{O}_8$ ) + nepheline ( $\text{NaAlSiO}_4$ ) = 2 jadeite ( $2\text{NaAlSi}_2\text{O}_6$ )

Various thermodynamic aspects of one or both of these reactions were the subject of experimental work by Yoder (1950a), by Yoder and Weir (1951), and by Kelley, Todd, Orr, King and Bonnickson (1953). As a result of this work information is available on molar volume, thermal expansion, and compressibility of jadeite as well as on low and high temperature heat capacities, entropies, etc. At about the same time Kracek, Neuvonen and Burley (1951) determined the standard enthalpy changes associated with these transformations at 25°C by means of aqueous hydrofluoric acid calorimetry. These data provided the basis for the first calculation of the *P-T* stability field of jadeite by Adams (1953).

More recently these mineralogical reactions have been studied directly by the high pressure-high temperature equilibrium approach, notably reaction (2) by Robertson, Birch and MacDonald (1957), reaction (1) by Birch and LeComte (1960) and reaction (1) again by Newton and Smith (1967). The work of Birch *et al.* was carried out in the temperature range 700–1100°C, and at pressures from 15–25 kbars. By means of the Clapeyron equation the authors extrapolated their results to lower temperatures

and pressures for comparison with thermochemical data and with field geology.

Field geological observations suggest that jadeite may be formed during metamorphism according to reaction (1) at pressures of the order of 5–10 kbars and temperatures of 150–300°C (see *e.g.* Essene, Fyfe and Turner, 1965). This is 2–4 kbars less than the pressures indicated by the extrapolation of Birch and LeComte. This discrepancy, and similar discrepancies for other mineralogical reactions, had led to the postulation of a corresponding “tectonic over pressure”, *i.e.* the existence of rock-forming pressures over and above those arising from the actual burial of sediments (Coleman and Lee, 1962). This provided the background for the recent reinvestigation of reaction (1) by Newton and Smith at temperatures from 500 to 600°C, *i.e.* at somewhat lower temperatures than those used by Birch and LeComte. The new extrapolation based on Newton and Smith’s data implies about 2 kbars lower equilibrium pressures at 200°C than the work of Birch and LeComte. Newton and Smith also show that solid solution formation through the replacement of  $\text{Al}^{3+}$  by  $\text{Fe}^{3+}$  will only lower the equilibrium pressure to a very limited extent. Thus the discrepancy between field geological and laboratory observations is reduced but not completely eliminated.

The interest of the present authors in the jadeite problem was aroused by the very significant experimental uncertainties associated with the reported calorimetric data for reactions (1) and (2). Thus, Kracek, Neuvonen and Burley studied two different samples of (low) albite; one from Amelia County, Virginia, and the other from Varuträsk, Sweden. These two samples gave widely different enthalpies of solution in aqueous HF,  $-149.79 \pm 0.11$  kcal and  $-148.12 \pm 0.21$  kcal/mole, respectively. Similarly in the case of jadeite from Japan, Kracek *et al.* found rather different enthalpies of solution for a sample ground in an agate mortar ( $-114.35 \pm 0.09$ ) and for the same sample ground in mullite ( $-115.93 \pm 0.17$ ). A sample of jadeite from Burma gave an intermediate value,  $-114.96 \pm 0.35$  kcal/mole. These results serve to illustrate some of the pitfalls associated with thermochemical work on minerals, and to our knowledge have never been fully explained. As a result the user of Kracek’s thermochemical data has a wide range of values from which to choose. This is well demonstrated, *e.g.*, in the tabulation prepared by Birch and LeComte.

The present investigation was initiated to improve the thermochemical data for reactions (1) and (2). For this purpose we have made use of oxide melt solution calorimetry at 691°C, as originally developed by Yokokawa and Kleppa (1964). Our results will be compared with those of Kracek *et al.*

From the new thermochemical values, combined with the available entropy and heat capacity data, we have recalculated the equilibrium lines in the  $p$ - $T$  plane as done originally by Adams. On the basis of our calculated diagram we propose a possible new interpretation of the remaining discrepancy between field geology and experimental mineralogy in the case of reaction (1).

#### EXPERIMENTAL

*Materials.* The sample of albite used in the present work was from Amelia County, Virginia (Smithsonian #118026). According to information provided by the museum the analysis of this mineral is as follows:

SiO <sub>2</sub>	67.84	CaO	0.00
Al <sub>2</sub> O <sub>3</sub>	19.65	Na <sub>2</sub> O	11.07
Fe <sub>2</sub> O <sub>3</sub>	0.03	K <sub>2</sub> O	0.29
FeO	0.02	H <sub>2</sub> O <sup>+</sup>	0.56
MgO	0.04	H <sub>2</sub> O <sup>-</sup>	0.03

The jadeite was a gift from Dr. R. G. Coleman (54-RGC-58). It consisted of a powder prepared from jadeite from the Clear Creek area of the New Idria Peak District, California. A microprobe analysis of another sample of jadeite from the same area but from a different locality is reported by Prewitt and Burnham (1966). This analysis indicates that the best approximation to the chemical formula of this jadeite is (Na<sub>0.93</sub>Ca<sub>0.02</sub>)(Al<sub>0.99</sub>Mg<sub>0.01</sub>)(Si<sub>1.99</sub>Fe<sub>0.01</sub>)O<sub>6</sub>. The sample of quartz was a pure, powdered, crystalline sample from Brazil of the type used in the oscillator plate industry. It had been ground in a tungsten carbide ball mill, with no detectable W or C contamination in spectrographic analysis. It was a gift of Dr. D. R. Waldbaum, and according to an analysis furnished by him it contains less than 0.003 percent B and less than 0.001 percent of all other detected impurities (Al, Ca, Ba, Cu). The nepheline was a synthetic powdered sample purchased from Tem-Pres Research, Inc., State College, Pa. According to a statement from the manufacturer

"The nepheline was prepared from a gel of 2 molar NaNO<sub>3</sub> (reagent grade), 2 molar AlNO<sub>3</sub> (reagent grade) and ammonia stabilized Ludox (tech-grade). The gel was dried and denitrated in a platinum crucible and fired at 1150°C. The product was ground and refired in platinum at 1215°C, ground and refired at 1250°C. The material gave a good x-ray pattern for nepheline optically  $n = 1.532$  with good crystallization and a single phase."

We checked the X-ray diffraction pattern of this sample and found the following values of  $2\theta$  for the six strongest lines: 20.50°, 21.26°, 23.16°, 27.34°, 29.85°, 31.09°, (Cu K $\alpha$  radiation). These values are very comparable to those for high temperature synthetic nepheline reported by Donnay, Schairer and Donnay (1959). Before use in the calorimeter the samples were, if necessary, ground in an agate mortar, passed through a 200-mesh sieve, washed with dilute hydrochloric and sulfuric acid and dried at 400°C.

*Apparatus and Procedures.* All calorimetric experiments were performed in the twin differential microcalorimeter for work up to 800° previously used by Yokokawa and Kleppa (1964). The procedures were analogous to those of Navrotsky and Kleppa (1968). All experiments were carried out at  $691 \pm 1^\circ\text{C}$ . Calibration of the calorimeter was by the gold-drop method, based on the heat content equation for pure gold given by Kelley (1960).

The calorimetric solvent was prepared from reagent grade lead (II) oxide and boric acid in the ratio 2 PbO:B<sub>2</sub>O<sub>3</sub>. In each experiment a small powdered sample of mineral or

glass (0.2-1.0 millimole) was dissolved in about 30 g of oxide melt ( $\sim 180$  millimole of oxides). In each case the calorimetric solution experiment was complete within one hour or less.

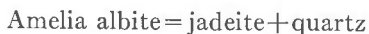
It is well known that a significant uncertainty in thermochemical work on natural minerals frequently arises from the presence of even small amounts of impurities. In the present work this problem was encountered in particular with respect to jadeite.

### RESULTS AND DISCUSSION

In order to assess the magnitude of the possible error arising from the impurities in the jadeite sample we carried out a separate series of measurements:

Two glasses of the albite composition were prepared (a) from our sample of Amelia albite and (b) from the stoichiometric mixture of jadeite+quartz. Both samples were prepared from finely divided powders in closed platinum capsules. After the sample had been melted and held at about  $1350^{\circ}\text{C}$  for two hours it was quenched to room temperature, again ground to a fine powder and then remelted at the same temperature. This procedure was repeated three times, after which the enthalpies of solution in lead borate of the two glasses were determined. The index of refraction of the Amelia albite glass was  $1.490 \pm 0.005$ , that of (jadeite+quartz) glass was  $1.495 \pm 0.005$ . The results of the enthalpy of solution measurements are recorded in Table 1. They show an enthalpy of solution of Amelia albite glass of  $5.05 \pm 0.19$  kcal/mole, while the corresponding value for (jadeite+quartz) glass was  $4.75 \pm 0.30$  kcal/mole.

The difference between these two values is comparable in magnitude to the errors associated with the two determinations. While this might provide a justification for neglecting the effect entirely, we consider it established beyond doubt that the effect is real, and we have applied the difference as a correction in calculating the enthalpy change for reaction (1). This correction was applied as follows: Let the corrected enthalpy change for the process



be  $\Delta H$ . This quantity is obtained from enthalpy of solution terms related to each of the three part processes:

$$\begin{aligned} \text{Amelia albite} &= \text{glass (A a)}; \Delta H_a \\ \text{glass (A a)} &= \text{glass (J + Q)}; \Delta H_b \\ \text{glass (J + Q)} &= \text{jadeite} + \text{quartz}; \Delta H_c \\ \hline \Delta H &= \Delta H_a + \Delta H_b + \Delta H_c \end{aligned}$$

From the quoted enthalpy of solution data we have  $\Delta H_b = 0.30 \pm 0.36$  kcal. In view of the small magnitude and large uncertainty in this correction we made no similar measurements and corrections for reaction (2).

Apart from the data on the enthalpies of solution of the two glasses of

TABLE 1. EXPERIMENTAL ENTHALPIES OF SOLUTION OF SUBSTANCES  
USED IN THE PRESENT WORK.

(Data in kcal/mole) <sup>a</sup>		Mean
Amelia albite glass	+4.78, +5.04, +5.10, +5.26, +5.23, +4.89;	+5.05 ± 0.19
(Jadeite+quartz) glass	+4.98, +4.69, +4.43, +4.73, +4.70, +4.90, +4.54, +4.77, +5.10, +4.36, +4.47, +5.43, +4.62;	+4.75 ± 0.30
Quartz	-1.10, -0.98, -1.08, -0.93, -1.01, -1.13, -0.98, -0.98, -1.02, -1.00, -0.97, -1.05;	-1.02 ± 0.05
Jadeite	+20.77, +20.52, +20.44, +20.54, +20.79, +20.41;	+20.58 ± 0.16
Albite	+20.11, +19.43, +20.20, +19.75, +20.40, +20.26, +20.15, +19.91, +20.59, +20.14, +20.18, +20.47;	+20.13 ± 0.32
Nepheline	+14.10, +14.11, +14.38, +14.27, +14.04;	+14.18 ± 0.14

<sup>a</sup> Quoted uncertainties are standard deviations from the mean.

the albite composition referred to above, we record in Table 1 the results of all the calorimetric solution experiments performed in the course of the present work. From these results we have calculated for reactions (1) and (2) the standard enthalpy changes at 964°K,  $\Delta H^{\circ}_{964}$ . These are recorded in column 1, Table 2. The quoted experimental limits of error for  $\Delta H^{\circ}_{964}$  represent the square root of the sum of the squares of the standard deviation of the various averages which were used in the calculations. Roughly one half of the relatively large uncertainty given for reaction (1) ( $\pm 0.50$  kcal/mole) represents the uncertainty which arises from the small difference between the two enthalpies of solution for the glasses of the albite composition.

From the enthalpy values listed in column 1, we have also calculated  $\Delta H^{\circ}_{298}$ , making use of the reported high temperature heat content data for albite, jadeite, nepheline and quartz quoted by Kelley (1960). The values of  $\Delta H^{\circ}_{298}$  are listed in column 2. For comparison we give in each case the same quantity calculated from the results of Kracek, Neuvonen

TABLE 2. THERMODYNAMIC DATA IN KCAL/MOLE FOR REACTIONS INVOLVING JADEITE<sup>a</sup>

Investigators (method)	$\Delta H^{\circ}_{964}$	$\Delta H^{\circ}_{298}$	$\Delta G^{\circ}_{298}$
a) Low albite = jadeite + quartz ( $T\Delta S^{\circ}_{298} = -2.51$ )			
Present work (calorimetry)	+0.27 ± 0.50	-0.66 (± 0.5)	+1.85
Kracek, Neuvonen & Burley (calorimetry)		-0.61 ± 0.9 <sup>b</sup>	+1.90 <sup>c</sup>
Birch & LeComte (extrapolated)			+2.70 <sup>d</sup>
Newton & Smith (extrapolated)			+1.90 <sup>d</sup>
b) Low albite + nepheline = 2 jadeite ( $T\Delta S^{\circ}_{298} = -4.81$ )			
	$\Delta H^{\circ}_{964}$	$\Delta H^{\circ}_{298}$	$\Delta G^{\circ}_{298}$
Present work (calorimetry)	-6.85 ± 0.25 <sup>e</sup>	-7.04 (± 0.3)	-2.23
Kracek, Neuvonen & Burley (calorimetry)		-6.19 ± 1.3 <sup>b</sup>	-1.38
Robertson, Birch & MacDonald (extrapolated)			+1.20 <sup>d</sup>

<sup>a</sup> Entropy data from Robie (1966)).

<sup>b</sup> Calculated from average values for two different samples each of jadeite and albite.

<sup>c</sup> In evaluating  $\Delta G^{\circ}$  for this reaction from the data of Kracek et al. Adams made use of a different value of  $T\Delta S^{\circ}_{298}$  (-2.21 Kcal rather than -2.51). This explains the lower equilibrium pressures calculated by him and shown in Fig. 1.

<sup>d</sup> Calculated from the reported extrapolated  $P$ - $T$  curve, neglecting compressibility corrections.

<sup>e</sup> Quoted limits of error make no allowance for possible effects of impurities or non-stoichiometry.

and Burley. In considering the latter we have for each mineral adopted the mean value of the various enthalpies of solution listed by the authors. Thus we have not made any biased selection from their data so as to achieve better agreement with our own work or with the results of equilibrium investigations. This accounts for the rather large experimental uncertainties attributed to the data of Kracek *et al.* When the limits of error in Kracek's study and in our own are taken into account, we consider the agreement to be very satisfactory. Thus we find in the present work for reaction (1)  $\Delta H^{\circ}_{298} = -0.66 (\pm 0.5)$  kcal. while the earlier result is  $-0.61 (\pm 0.9)$ . Similarly for reaction (2) the present work gives  $\Delta H^{\circ}_{298} = -7.04 (\pm 0.3)$  while the study of Kracek *et al.* yields  $-6.19 (\pm 1.3)$ .

For all the minerals considered in this investigation standard entropy data at 298°K are reported in the literature (*e.g.* Robie (1966)). This al-

lows us to calculate  $\Delta G^{\circ}_{298}$  both from the present results and from the data of Kracek *et al.* The values calculated in this manner are given in column 3, Table 2. For comparison we list in table also the corresponding values of  $\Delta G^{\circ}_{298}$  calculated from the extrapolation to 298°K of the high temperature-high pressure equilibrium data of Birch *et al.* and from the recent work of Newton and Smith.

*P-T Stability Field of Jadeite.* As an aid in the present discussion we have in Figure 1 assembled the available high pressure-high temperature equilibrium data for reactions (1) and (2) for comparison with curves calculated from the present work. This figure incorporates the information contained in Table II. The necessary volume data used in calculating the equilibrium curves were taken from the compilation by Robie.<sup>1</sup>

We note first that Table 2 and Figure 1 both show that for reaction (1), albite  $\rightarrow$  jadeite + quartz, there is now excellent agreement between the thermochemical data and the recent equilibrium study of Newton and Smith. While the limits of error of the thermochemical data still leave room for improvement, this agreement is very gratifying.

At first sight an examination of the data listed in Table 2 would seem to imply that a discrepancy exists between the earlier equilibrium work of Birch *et al.* and the thermochemical data. This apparent discrepancy arises from the fact that Birch *et al.* completely neglected the disordering entropy of albite in extrapolating their data to lower temperatures. For reaction (1) this point was noted already by Newton and Smith. It in large measure explains the 2 kbar pressure difference at 200°C between Birch and LeComte on the one hand and Newton and Smith on the other. A similar situation exists in the case of reaction (2). Thus we see from Figure 1 that the extrapolated equilibrium pressure of Robertson, Birch and MacDonald at 200°C is about 3.5 kbars higher than that calculated from our own data (and by Adams from the data of Kracek *et*

<sup>1</sup> From the work of Yoder (1950b) we know that the inversion temperature of quartz rises from 848°K at zero pressure to about 1100°K at 10 kb. Thus it is apparent that the modification of quartz which is of interest in the present work is always low quartz. Since standard thermodynamic data for low quartz are available only at temperatures below 848°, this introduces some additional uncertainty into our calculation of the P-T curves for reaction (1) above this temperature. According to Kelley (1960) the enthalpy change for the low-high quartz inversion is 290 cal/mole while the entropy change is 0.34 cal/deg mole. In our calculation we have made the rather arbitrary assumption that these enthalpy and entropy changes do not change with temperature above 848°K. We accordingly adopted Kelley's enthalpy and entropy values for high quartz after subtracting the above values for the inversion process. In the final calculations no corrections were made for compressibility and thermal expansion effects since preliminary calculations had indicated that these corrections were much smaller than the uncertainty in our measurements.

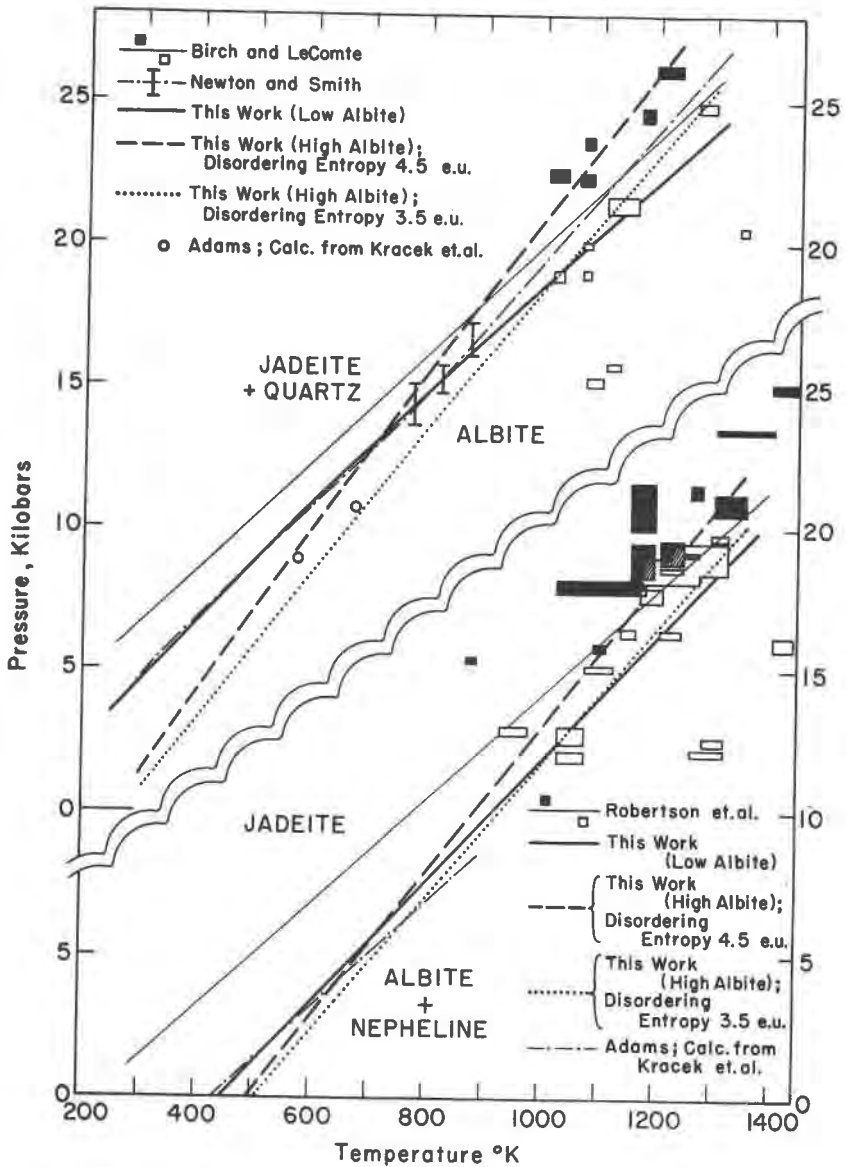


FIG. 1. *P-T* equilibrium curves for the reactions albite=jadeite+quartz, and 2 jadeite=albite+nepheline. Solid and open squares reflect phases observed by Birch and LeComte and by Robertson *et al.*



*al.*). For this reaction an additional entropy effect may arise from the presence of Al-Si disorder in nepheline.

The present discussion illustrates that the disordering of albite and similar phases must be taken into account when high temperature equilibrium data are extrapolated to lower temperatures. Clearly, the effect must also be considered when room temperature thermodynamic data are used to calculate equilibrium curves above the low→high albite "inversion" temperature. Fortunately, the necessary thermodynamic data for this transformation, while still somewhat uncertain, are now available. According to the recent calorimetric study of Holm and Kleppa (1968) the enthalpy change associated with the low albite→high albite transformation ranges from about 2.6 kcal/mole at room temperature to about 3.4 kcal/mole at 700°C. While the thermodynamic character (order) of this transformation is not known, both thermodynamic and kinetic considerations suggest that the transformation under equilibrium conditions would occur in the temperature range 500° to 700°C.

The curves given in Figure 1 have been calculated taking these heat and entropy changes in albite into account. Since there is some uncertainty about the disordering entropy (Holm and Kleppa, 1968), we have drawn separate curves in Figure 1 for the case when the entropy change in albite is assumed to be 3.5 cal/deg mole, and when it is 4.5 cal/deg mole. These two values roughly correspond to transformation temperatures low→high albite of 725°K and 1000°K, respectively. Since the volume change associated with this transformation is small, it will not vary much with pressure. For reaction (2) we have neglected the disordering entropy in nepheline. If this were taken into account, it would tend to raise the transformation pressures somewhat, particularly at higher temperatures. Thus the agreement between our calculated curves and the data of Robertson, Birch and MacDonald may in part be fortuitous. However, it is perhaps significant that the calculated curve for an assumed disordering entropy of 4.5 cal/degree mole seems to fit the high temperature equilibrium data slightly better in the case of reaction (2) than in the case of reaction (1).

Note that our calculated  $p$ - $T$  curve for reaction (1) is in excellent agreement with the experimental points of Newton and Smith. This is as expected, since the experiments of these authors were carried out at 500–600°C, *i.e.* at temperatures where the free energy difference between low and high albite certainly is small. On the other hand, as we go on to compare our calculated  $p$ - $T$  curves with the experimental points of Birch and LeComte, we find a different situation. Since their measurements were carried out from 700 to 1050°C, it is essential to make allowance for the low albite→high albite transformation in order to achieve agreement be-

tween the thermochemically calculated curve and experiment. From Figure 1 it would seem that the curve based on an entropy change of 3.5 cal/deg mole fit the experimental data somewhat better than the curve based on 4.5 cal/deg mole.

Finally, we want to draw attention to the fact that below about 500°C the metastable parts of the transformation curves high albite→jadeite + quartz involve lower pressures than the corresponding equilibrium curve for low albite.<sup>1</sup> In fact these curves for metastable equilibrium extrapolate to near zero pressures at room temperatures.

Now it is well known from laboratory investigations that it is extremely difficult to synthesize ordered low albite. Thus, it was shown by MacKenzie (1957) that synthetic albite always forms in the high temperature disordered form even under conditions where low albite is stable. Only during the last year has the synthesis of low albite been achieved in the laboratory, in the presence of Na<sub>2</sub>Si<sub>2</sub>O<sub>5</sub> at 200°C and 10 kilobars water pressure (Martin, 1967).

These observations, along with our own Figure 1, suggest the possibility that the metamorphic formation of jadeite according to reaction (1) at low temperatures may be facilitated by high albite acting as an intermediate. A clearer understanding of the proposed mechanism may perhaps be obtained through examination of Figure 2, which gives a *G-T* diagram for a pressure of about 7 kbars. According to our calculations (jadeite+quartz) is stable at this pressure below about 140°C, while low albite is stable above this temperature. However, (jadeite+quartz) is stable with respect to *high* albite at all temperatures below about 220°C. High albite as a metastable intermediate may give rise to two alternative reactions: It may either transform directly to the stable low albite form (by creating long range Al-Si order in the feldspar structure) or it may transform to jadeite+quartz. If the kinetics of reversion of jadeite + quartz to low albite is slow, it is possible that some of the jadeite will be preserved.

It is well known (Christie, 1962) that Al-Si disordered, thermally metastable potassium feldspars exist in nature, and that they have existed for long times. In view of this it is not at all unreasonable to assume that thermally disordered albites may have existed for times which are long enough so that the synthesis of jadeite could occur.

<sup>1</sup> The reader may be concerned to find that at low temperatures the sequence of equilibrium curves with increasing pressure, represent disordering entropies of 3.5, 4.5 and 0 cal/deg. mole, respectively (rather than 4.5, 3.5 and 0). This sequence of curves reflects the fact that we have adopted the enthalpy change for this transformation given by Holm and Kleppa (1968) and have let the (assumed) transformation temperature low→high albite vary with the entropy, according to the relation  $\Delta G = \Delta H - T\Delta S = 0$ . The transformation temperatures indicated in Figure 1 were obtained from this relation.

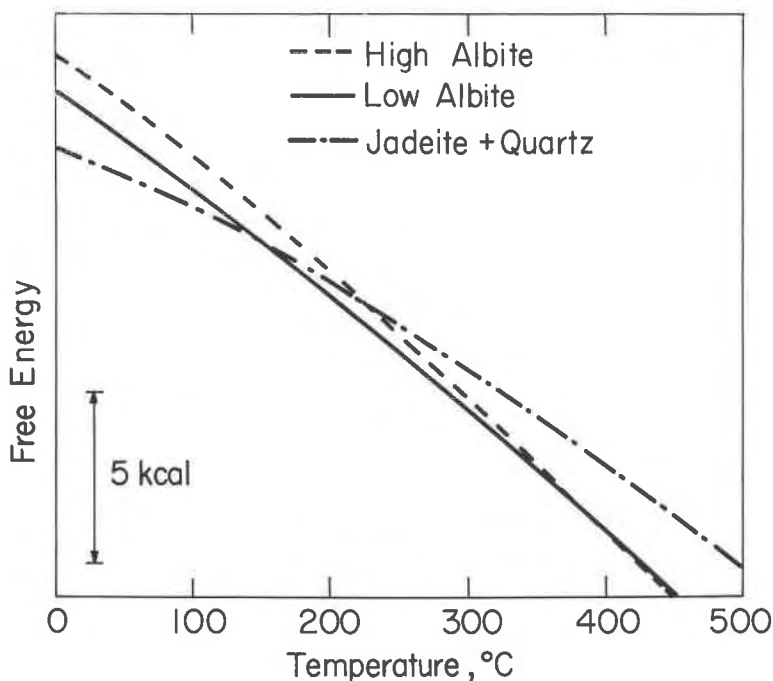


FIG. 2.  $G$ - $T$  diagram for low albite, high albite and (jadeite+quartz) valid at approximately 7 kbars. Note that in the temperature range 140–220° (jadeite+quartz) is stable with respect to high albite, but unstable with respect to low albite.

At this point we want to stress that it is not essential that the postulated intermediate should be a fully disordered high albite. Recently Holm and Kleppa (1968) have demonstrated that the presence of even a relatively modest degree of Al-Si disorder in *low* albite will have a significant influence on its thermodynamic properties, and accordingly on the location of the equilibrium curve. Therefore, we believe that it is now no longer necessary to invoke "tectonic over-pressure" in order to account for the metamorphic formation of jadeite from albite according to reaction (1).

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