

THERMAL EXPANSION OF LOW AND HIGH ALBITE¹

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ABSTRACT

X-ray powder diffraction data obtained on a heating stage determined the unit-cell parameters of low albite to 1127°C and high albite to above 1000°C. Low albite remained triclinic at all temperatures studied. Contamination of high albite by potassium from the heating stage seriously affected the results, and the only monoclinic phases observed at high temperatures proved to be potassium-bearing anorthoclases when studied at room temperature. After correction for compositional effects, the best estimate of the triclinic-monoclinic inversion in NaAlSi₃O₈ is 1100° ± 50°C. It is uncertain if monalbite occurs before melting takes place.

Good agreement is shown with earlier data obtained by dilatometry by others on low albite, though the present results give axial and angular details previously unavailable, as well as new data for high albite. Various expressions of the volume thermal expansion of each polymorph are presented. The percent volume expansions of both polymorphs are almost identical, but the coefficients of thermal expansion are not. The effect of temperature on the cell angles of both polymorphs is the same as that caused by solid solution with potassium feldspar. The effects of temperature on the position of the rhombic section and on the obliquity are calculated.

INTRODUCTION

Measurement of the thermal expansion of triclinic crystals by X-ray powder diffractometry has been greatly facilitated recently by the development of heating stages for diffractometers and of computer programs that index the diffraction pattern and, by least-squares methods, refine the unit-cell dimensions and calculate the associated standard error. It is possible with this technique to measure the thermal expansion and to obtain results with a precision and accuracy comparable to superior dilatometer studies even when working on crystals that pose the most general problems of symmetry changes, high pseudosymmetry, incomplete diffraction patterns, and irreversible phenomena.

Knowledge of the symmetry and unit-cell parameters of the various sodium feldspar polymorphs at high temperatures gives details on the polymorphism of this compound and on the crystal chemistry of alkali feldspars. Incidental results of high temperature X-ray measurements are uniquely indexed powder-diffraction patterns, and an evaluation of thermal effects on the position of the rhombic section and on obliquity, both of which reflect the tendency toward complex twinning. Changes in the size and orientation of the ellipsoid of dilation in crystals of low symmetry could be measured.

Throughout the paper, the name *low albite* will be applied to triclinic

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$\text{NaAlSi}_3\text{O}_8$ with dimensions at room temperature as given in Table 1. *High albite* will be used for the triclinic polymorph of $\text{NaAlSi}_3\text{O}_8$ with dimensions at room temperature as given in Table 4. As will be shown below, this polymorph may become monoclinic when heated close to its melting point. If it does become monoclinic before melting, the name *monalbite* would be appropriate for a monoclinic substance with $\text{NaAlSi}_3\text{O}_8$ composition. The supposed polymorphs of $\text{NaAlSi}_3\text{O}_8$ called "analbite" and "monalbite" by Schneider (1957), Brown (1960), and Robinson

TABLE 1. CELL DIMENSIONS OF LOW ALBITE AT ROOM TEMPERATURE

a Å	b Å	c Å	α	β	γ	Volume ¹		Remarks
						Å ³	cm ³ /mol	
8.140	12.792	7.161	94° 17.4'	116° 37.8'	87° 40.8'	664.7	100.079	Amelia, Virginia
.002	.004	.002	1.5'	1.0'	1.2'			Quartz internal standard
8.138	12.786	7.163	94° 15.9'	116° 35.5'	87° 43.2'	664.64	100.070	Silicon internal standard
.002	.003	.002	1.6'	1.4'	1.4'	.21	.032	
8.136	12.786	7.159	94° 17.3'	116° 33.7'	87° 39.1'	664.3	100.019	Position of heating stage
.005	.009	.003	3.2'	2.0'	3.0'			calibrated vs MgO, Pt.
8.138	12.788	7.161	94° 16.9'	116° 35.7'	87° 41.0'	664.55	100.057	Average of above three de-
.002	.003	.001	0.6'	1.4'	1.3'	.2	.030	terminations.
8.144	12.787	7.160	94° 15.6'	116° 34.8'	87° 40.2'	665.1	100.139	Amelia (Smith, 1956, p. 54)
8.138	12.789	7.156	94° 20.0'	116° 34.0'	87° 39.0'	664.2	100.004	Ramona, California (Ferguson, Traill, Taylor, 1958)
8.135	12.788	7.154	94° 13.8'	116° 31.2'	87° 42.6'	664.0	99.974	Kodarma, India (Cole, Sorum, and Taylor, 1951)

¹ Avogadro's number was taken as 6.02252×10^{23} (National Bureau of Standards, 1963) throughout this paper.

(1961) have been shown to be potassium-bearing by W. L. Brown (written communication, 1965).

In this paper, we present rather complete data for the thermal expansion of low albite and sufficient data to estimate the thermal expansion of high albite to about 1000°. In our investigation no evidence was found for a polymorph of $\text{NaAlSi}_3\text{O}_8$ with a higher structural state than that of high albite.

PREVIOUS STUDIES OF ALBITE POLYMORPHS AT HIGH TEMPERATURES

Polymorphism and symmetry changes. Since Foerstner (1884) discovered that sodium-rich alkali feldspars from volcanic rocks undergo a reversible symmetry change from triclinic to monoclinic on heating, many papers have appeared that discuss the polymorphism and symmetry changes of alkali feldspars, particularly albite. MacKenzie and Smith (1961, p. 53–56) summarized much of the research on the polymorphism of albite

published in the 10 years prior to the date of their paper; the discussions following their paper (p. 64–68) describe some continuing problems to which this paper is especially pertinent. An excellent summary of polymorphism in plagioclase, including albite, is given by Gay (1962).

The transformation of low albite to high albite is known to involve changing the arrangement of aluminum and silicon atoms in the tetrahedral framework, and the new arrangement (and intermediate stages) can be quenched from high temperatures. The change of symmetry from triclinic to monoclinic that occurs in sodium-rich alkali feldspars involves only minor angular adjustments within the framework, and is not quenchable. It is important to distinguish these two kinds of transformation. Unless the aluminum-silicon distribution in a triclinic feldspar is such that it can fulfill the requirements of monoclinic symmetry, no symmetry change can occur until the distribution changes. Therefore, probably only high albite with its random aluminum-silicon distribution can possibly become monoclinic on heating.

Both optical and X-ray methods have been used at high temperatures to study the transformations. Rinne (1914) used an optical method to study the variation in the cleavage angle $(001) \wedge (010)$ ($=\alpha^*$) of low albite on heating to 600°C. The first X-ray investigation directly comparable to ours was by Davis (*in* Tuttle and Bowen, 1950, p. 577), who heated high albite to 1050°C using a heating stage on a diffractometer. He did not find clear evidence that high albite became monoclinic, and no cell dimensions were measured. Tuttle and Bowen (*op. cit.*) could not record any differential heat effects on heating high albite through the interval 100°–1000°C, and none would be expected in light of the present investigation.

MacKenzie (1952) measured the relative separation of the (111) and $(\bar{1}\bar{1}\bar{1})$ reflections of high albite and contiguous feldspar compositions as a function of temperature, also using a heating stage for the diffractometer. These reflections gradually converge, on heating, and merge to a single reflection when the crystal becomes monoclinic.¹ In practice the reflections are not resolved when the obliquity is small, so a projection from a series of temperatures is required to estimate the temperature of the symmetry change, as demonstrated in Figure 1 with data from the present investigation. MacKenzie's study of a series of albite specimens with different known thermal histories revealed that the estimated temperature of the symmetry change of high and intermediate albite may vary by at least 200°C from 950° to above 1150°C, being lower for

¹ It must be recognized that crystals that are dimensionally monoclinic may retain triclinic intensity distributions that are only resolvable by study of single crystals.

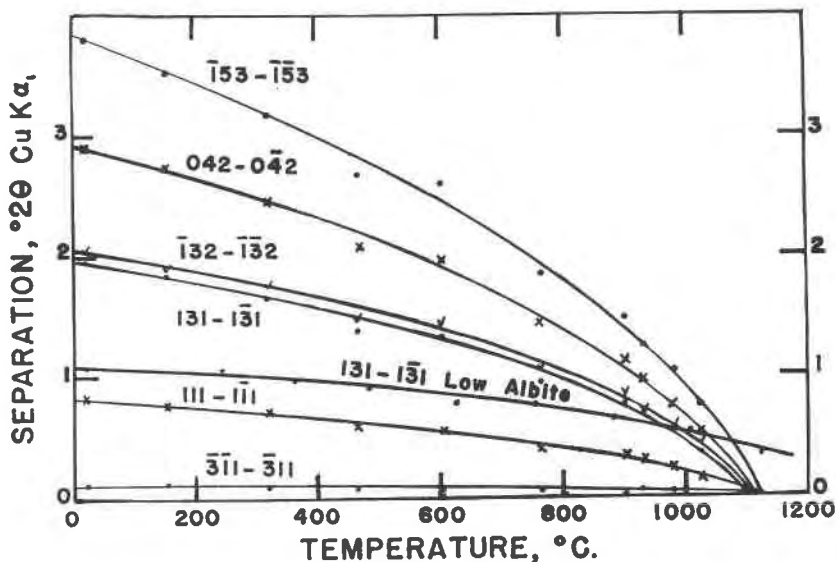


FIG. 1. Angular separation of symmetrically related pairs of diffraction lines of high albite as a function of temperature. A selection of pairs is shown to illustrate the great range of possible separations below 2θ ($\text{CuK}\alpha$) = 60° . The points plotted were calculated from the data in Table 5; the two pairs with the greatest separations were not observed on diffractometer patterns. Data are shown only for material with Or < 3 percent. Projection of each curve toward zero separation should afford a common point which would be an estimate of the inversion temperature. For this sample, the inversion temperature appears to be $1100^\circ\text{C} \pm 50^\circ\text{C}$. However, the marked curvature of the curves, even where potassium exchange is not a problem, makes projections uncertain, and thus accounts for the large uncertainty in the estimate. The separation of the $1\bar{3}1$ and $13\bar{1}$ lines of low albite calculated from the data in Table 2 is shown for comparative purposes. Inspection of Figures 6 and 7 indicates that at the temperature at which the angular separation of $1\bar{3}1$ and $13\bar{1}$ is the same for both high and low albite ($\sim 950^\circ\text{C}$), the dimensions of their unit cells are quite different.

specimens synthesized at higher temperatures. The temperature of the symmetry change is also lowered by heating for long periods of time above 1030°C . This suggests the higher the structural state, the lower the inversion temperature.¹ Optical studies of the disappearance of triclinic twins in natural feldspars by MacKenzie (1952) and Laves (1952) yielded essentially the same curve for the inversion temperature as a function of the Or content, and a long extrapolation indicated that the inversion temperature for $\text{NaAlSi}_3\text{O}_8$ composition was close to its melt-

¹ MacKenzie assumed that no potassium exchanged for sodium during the investigation. We suggest that it is possible that the lowering of the inversion temperature and its dependence on heating time, as well as the melting observed at 1075°C (MacKenzie, 1952, p. 323) could all result from potassium exchange for sodium in the samples.

ing temperature. Baskin (1956, Table 3) produced crystals of albite with structures intermediate between high and low albite by dry heat treatment of low albite. She demonstrated that there is a series of polymorphs intermediate between low and high albite, and that all the unit-cell parameters changed regularly in the polymorphous series. MacKenzie (1957) synthesized many intermediate polymorphs of albite hydrothermally, and recognized that the unquenchable (displacive) symmetry change "must involve some change other than the order-disorder change postulated for the gradation from low-temperature to high-temperature albite" (p. 510).

Stewart (1960a, 1960b) has duplicated many of MacKenzie's experiments, and has investigated the effects of additional components on the rate of producing intermediate polymorphs.

Studies by Schneider (1957), Brown (1960), and Robinson (1961) were concerned with further changes that take place in high albite heated for prolonged times, but it is now known that these changes resulted from alkali exchange between ceramics and the heated feldspars (W. L. Brown, written communication, 1965).

Thermal expansion. A useful compilation of earlier data on the thermal expansion of feldspar is given by Saucier and Saplevitch (1962). Kozu and Ueda (1933) and Rosenholtz and Smith (*in* Yoder and Weir, 1951) have determined the volume thermal expansion of low albite by dilatometer measurements.

Kuellmer (1961, p. 115) has reported the positions from room temperature to 750°C of the $\bar{2}01$ line of natural low albite and of the high albite prepared from it by heating. Our results for the movements of these lines in nearly pure high and low albite are virtually identical with Kuellmer's. The difference between the positions of $(\bar{2}01)$ for these two polymorphs at room temperature and higher temperatures is more directly related to the fact that the polymorphs differ crystallographically than to compositional differences as suggested by Kuellmer.

Brown (1962, footnote on p. 362) reported that the angles of the unit cell of low albite changed on heating toward those of maximum microcline at room temperature, the values of α^* and γ^* at 1000° being 87° 39' and 91° 08', respectively. Our results are in excellent agreement with this (see Fig. 3).

Kayode (1964) studied a number of low and high plagioclases by a method similar to that used in this paper. His results are in preparation for publication elsewhere.

Stewart and von Limbach (1964) gave preliminary values for the thermal expansion of low and high albite which are superseded by those which appear below.

MATERIALS STUDIED

A large ground sample (<150 mesh) of albite from Amelia, Virginia, was used as the source of low albite. The sample contained about 1 percent each of quartz, muscovite, and clayey alteration products. Partial analysis by J. J. Fahey showed CaO 0.25 percent, K₂O 0.27 percent, and Na₂O 10.90, 10.88 percent. Recalculation of these oxides to 100 percent of feldspar components yields Ab_{97.6}An_{1.3}Or_{1.7}, in excellent agreement with the results of chemical analyses of Amelia albite tabulated by Deer, Howie, and Zussman (1963, table 13). Careful optical examination revealed only very rare plagioclase grains with γ greater than 1.540, again in excellent agreement with the analysis and other optical measurements on Amelia albite. The cell dimensions of our sample (Table 1) are typical of those in the literature for low albite.

The high albite measured was synthesized hydrothermally in sealed metal tubes from charges consisting of 100 mg. of carefully prepared glass of NaAlSi₃O₈ composition plus 10 mg. of distilled water at 925°C, 3500 psi for 3 hours. Optical examination of the product revealed no glass or crystalline phases other than high albite. The separation of the 131 and 1 $\bar{3}$ 1 reflections was $1.98 \pm .02^\circ 2\theta$ with CuK α radiation. The dimensions are given in Table 4 and are identical with those for high albite found by other investigators within the limits of error of the various methods.

DATA COLLECTION AND REFINEMENT

The cell dimensions were determined from averaged powder-diffraction data collected in triplicate on a diffractometer at room temperature using NaCl, or quartz or silicon calibrated against the same NaCl, as internal standards. The value for the cell edge ($a=5.64119 \text{ \AA}$ at 26°C) and thermal expansion of NaCl were taken from Table 2.5.2 of the International Tables for X-ray Crystallography (1962). The value of CuK α radiation used was 1.54180 \AA . The data were refined with a digital computer by alternate use of fixed and variable indexing options of the crystallographic indexing program developed by Evans, Appleman, and Handwerker (1963).

Data at elevated temperatures were collected using a heating stage for the X-ray diffractometer (Skinner, Stewart, and Morgenstern, 1962) calibrated by the method described in Stewart, Walker, Wright, and Fahey (1966). The heating stage was brought to operating temperature in approximately one-half hour. After about another half hour temperature regulation was within 1 percent of the set point, and data collection began. Approximately three hours at the assigned temperature was required to make three traverses across the 2θ range from 20° to 58°. The controller was then set for the next temperature and the procedure was repeated.

The data were collected in a sequence of temperatures designed so that irreversible changes that might have occurred could be detected. As many uniquely indexed lines as possible within the range 20° to $56^\circ 2\theta$ ($\text{CuK}\alpha$) were used in the final cycle of fixed indexing.¹ In general, the group of strong lines at $\sim 28^\circ 2\theta$ was not used because they were too strong to be individually resolved at the instrument settings used. Feldspar lines which were interfered with by lines of mullite from the sample holder (the $\bar{2}21$ line of high albite, for example), or platinum sublimed from the furnace windings, were omitted from consideration.

Both high albite and low albite show strong pseudosymmetry such that $0kl$ reflections (with $k \neq 2l$) interfere with $0, 2l, k/2$ —i.e., 022 and 041 , or $0\bar{6}2$ and $0\bar{4}3$. Thus some lines were not usable for the refinements because they could not be unambiguously indexed. Dr. Paul H. Ribbe (written communication, September 20, 1963) generously made available to us some of his intensity data for three-dimensional refinements of albites which helped resolve several ambiguities in the indexing. In addition, when line positions for a number of temperatures are compared as in Figure 2, it is possible to identify and index various lines that happen to coincide at room temperature by their relative shifts at higher temperatures.

One mount of low albite and two mounts of high albite were examined. After heating to 1010°C the sample of low albite was unchanged as determined by either optical or X-ray examination. After 1127°C the sample was found to be fritted and partially melted. Twinning had largely disappeared, veinlets of glass could be seen in the grains, the γ index had decreased to less than 1.537, and two poorly resolved sets of 131 – $\bar{1}31$ reflections could be seen on the powder-diffraction pattern, indicating partial transformation to high albite.

The high albite samples showed measurable differences after heating and, being very fine grained and partially plated with metallic platinum, tended to be unsuitable for detailed optical examination. Some grains showed highly twinned areas like those found in microcline. Irreversible changes were noted on the X-ray powder diffraction patterns of samples which were heated above 900°C . After the sample had been heated to 902°C , the 131 – $\bar{1}31$ separation at room temperature had decreased to 1.94 ± 0.03 ; it had decreased to 1.63 ± 0.04 after heating to 1062°C . Shifts in the positions of other lines were also noted. These changes result

¹ A document listing the computer input (fixed Miller index and associated observed 2θ) and the final cycle of refinement for each temperature studied has been deposited as Document No. 9299 with the American Documentation Institute, Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington, D. C. Copies may be received by citing the document number, and remitting in advance \$8.75 for photoprints or \$3.00 for 35 mm. microfilm.

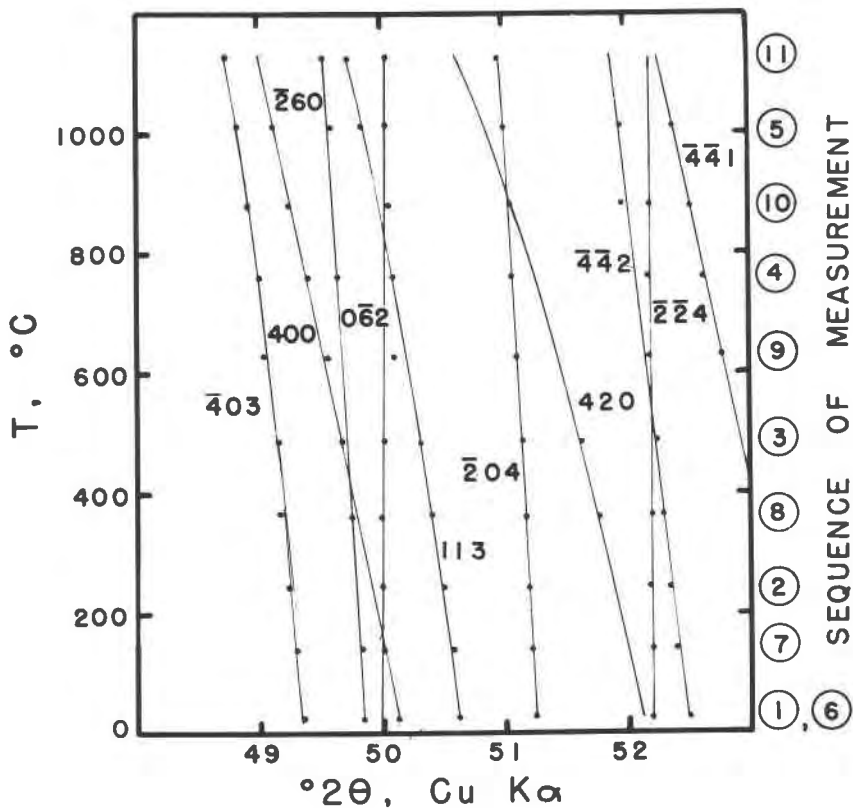


FIG. 2. Plot of the positions of certain powder-diffraction lines of low albite from room temperature to 1127°C. Dots indicate lines measured; line positions at temperatures where no line was measurable are controlled by the position calculated for that temperature from the data in Table 2. Weak lines such as (420) at 488°C are measurable only when remote from other lines. Interference between lines is clearly shown by (062) and (113) at 605°C; this line was discarded in the final cycle of refinement, as were several other measurements shown.

The data were collected in the sequence shown in the right margin. Reversibility is shown by the smooth fit of the data for all temperatures.

from K-exchange from the "mullite" ceramic sample holder, which contained potassium.¹ The estimated composition of the potassium-bearing solid solution at each temperature is given in Table 5. Such feldspars are

¹ After the data were collected, H. J. Rose, Jr., of the U. S. Geological Survey, determined by X-ray fluorescence methods that the commercial "mullite" used contained 0.8 percent K₂O, 0.2 percent Na₂O, and 0.05 percent CaO. Presumably the alkalis are present in the vitric binder between the mullite grains, and as the amount of binder is small, it must be quite rich in K₂O. Although alkali diffusion and exchange took place rapidly with high albite at temperatures above 900°C, the phenomenon was not detectable in low albite until a temperature over 1100°C was attained.

anorthoclase. (They have also been called analbite by Laves, 1960). The composition was estimated by comparison of the individual unit-cell parameters and unit-cell volume with the appropriate curves for the solid-solution series from high albite toward high sanidine as determined by Orville (written communication, 1965) by the same measurement and refinement procedures used in this investigation. As a check on such estimates, the sample (9.5 mg.) heated to 1062°C was analyzed for potassium by X-ray fluorescence methods by H. J. Rose, Jr., who found 2.1 percent K_2O (equivalent to Or 12.4%) compared to the 1.9 percent K_2O that would be present in anorthoclase estimated dimensionally to contain 10.4 percent Or. The accuracy of the X-ray fluorescence analysis for such a small sample is 5 percent of the amount present, and that of the estimate from the dimensions probably 10 percent of the amount present for low concentrations, so the methods agree well within the limits of error. The compositions for samples not at room temperature were estimated by correcting for thermal expansion by analogy to the thermal expansion of low albite. The known sequence of exposure to high temperatures provided additional control on the probable composition of the samples. Only a short interval for data collection at high temperature was permissible before appreciable potassium exchange occurred, and only the first five measurements on mount A were acceptable (Or < 3%), necessitating a second mount. The first three measurements on mount B were satisfactory and have been combined with the acceptable measurements on mount A to calculate the thermal expansion.

RESULTS

Cell dimensions of low albite. The cell dimensions and indexed X-ray diffraction patterns of low and high albite given by Smith (1956) were used as starting values for our refinements. The results of our refinements of the cell dimensions of our sample of Amelia albite are compared in Table 1 with those of other workers for other samples of albite. The agreement is within a part in one thousand, which is satisfactory. Our indexing of the portion of the powder-diffraction pattern that we measured is in good agreement with Smith's below $37.4^\circ 2\theta$ $CuK\alpha$ with the exception of the line at ~ 24.31 , which we indexed as $\bar{1}30$ rather than $\bar{1}\bar{3}1$. Substantial agreement on all strong lines continues to 52° , but our patterns had some weak lines not found by Smith (and *vice versa*), and we observed small differences in the 2θ values of some lines that resulted in different assignments of indexes for Smith's 310 (our $\bar{2}40$), 042 (our $\bar{1}\bar{5}1$), 202 (our 401), 421 (our $\bar{2}\bar{2}\bar{2}$), and 043 (our $0\bar{6}\bar{2}$). These differences are not serious. We indexed patterns only to about 58° .

The average of our three determinations of the dimensions of our sample is quite close to the dimensions obtained in the measurement with

a silicon internal standard. That particular measurement was therefore chosen for inclusion in Table 2 as representative of the dimensions at 26°C. Table 2 is a tabulation of the unit-cell parameters of low albite at a number of elevated temperatures, and at room temperature after exposure to high temperatures. No irreversible changes can be measured until a temperature of 1127°C was reached, at which temperature the sample underwent partial melting and inversion toward high albite, and possible potassium exchange. The reciprocal cell angles α^* and γ^* of low albite vary with increasing temperature toward the values obtained for maximum microcline studied at room temperature (Fig. 3).

Thermal expansion of low albite. The volume expansion of low albite was measured with a dilatometer by Kozu and Ueda in 1933. They measured the change in dimensions of a single crystal of albite from Alp Rischuna that had been polished into a perfect cube with the a axis normal to one face, and with the perpendicular to (010) normal to another face. In dilatometry of triclinic crystals only the volume expansion derived from the three mutually perpendicular linear expansions has significance, because in the triclinic system the triaxial ellipsoid of thermal dilation is not constrained by symmetry to any fixed orientation relative to the crystallographic axes, and the principal axes of linear thermal expansion are different from the crystallographic axes of albite. A similar dilatometer measurement of the volume expansion of low albite from Amelia was performed by Rosenholtz and Smith (*in* Yoder and Weir, 1951, p. 684). The present X-ray method of measuring thermal expansion removes the restriction that large single crystals be available, and in addition yields the true crystallographic parameters.

Our X-ray results for the volume percent expansion of low albite from Amelia are compared with Kozu and Ueda's and Rosenholtz and Smith's dilatometer results on Figure 4. The agreement is within a few percent. Some systematic differences may exist, but they may result from differences in the smoothed functions fitted to the observations.

Low albite is triclinic at all temperatures, and can be superheated until partial melting takes place before the inversion to higher-temperature polymorphs has proceeded significantly. The percent axial expansion is greatest along the a axis, being about three times that of the b axis, and about ten times that of the c axis.

The change of molar volume with increase of absolute temperature was fitted by the method of least squares with a digital computer to the equation (Skinner, Clark, and Appleman, 1961, p. 665):

$$V = \frac{A}{T} + B + CT + DT^2$$

TABLE 2. CELL DIMENSIONS OF LOW ALBITE AT HIGH TEMPERATURE

Sequence of Measurement	T°C	a, Å	b, Å	c, Å	α	β	γ	Volume Å ³	a*	b*	c*	α^*	β^*	γ^*	V*	No. of lines used	Standard error, % ²
(1)	26	8.138 .002	12.786 .003	7.163 .002	94°15.9' 1.6	116°35.5' 1.4	87°43.2' 1.4	664.64 0.21	.13743	.078426	.15646	86°22.4'	63°29.4'	90°25.4'	.0015048	24	.0179
(6)	26 ¹	8.141 .007	12.790 .009	7.157 .004	94 13.5 3.5	116 35.8 2.9	87 46.3 3.1	664.51 0.50	.13738	.078398	.15657	86 23.3	63 29.2	90 22.7	.0015049	21	.0339
(12)	26 ²	8.139 .008	12.816 .010	7.159 .003	93 56.1 2.9	116 19.6 2.5	88 1.7 3.6	667.75 0.56	.13708	.078210	.15613	86 35.0	63 44.5	90 15.3	.0014976	18	.0322
(7)	141	8.154 .006	12.794 .005	7.161 .003	94 11.9 2.6	116 32.3 2.4	87 42.8 2.5	666.60 0.43	.13708	.078374	.15638	86 26.8	63 33.0	90 27.7	.0015001	26	.0321
(2)	245	8.165 .004	12.802 .005	7.158 .002	94 12.3 2.3	116 29.9 1.7	87 38.5 2.4	667.78 0.34	.13685	.078329	.15640	86 28.5	63 35.6	90 32.5	.0014975	40	.0362
(8)	364	8.180 .002	12.804 .004	7.158 .001	94 2.0 2.0	116 21.8 1.1	87 40.1 1.6	679.05 0.22	.13647	.078298	.15618	86 39.2	63 43.6	90 36.4	.0014924	32	.0204
(3)	488	8.189 .002	12.818 .003	7.159 .002	93 57.6 1.8	116 19.8 1.2	87 36.3 1.5	671.81 0.33	.13625	.078211	.15611	86 45.9	63 45.7	90 42.9	.0014885	33	.0236
(9)	628	8.209 .003	12.829 .004	7.165 .002	93 43.1 2.3	116 17.4 1.6	87 38.0 2.3	675.06 0.28	.13588	.078121	.15588	87 1.2	63 47.8	90 48.3	.0014813	29	.0256
(4)	759	8.227 .003	12.841 .004	7.165 .002	93 36.9 2.3	116 14.1 1.5	87 33.9 2.0	677.59 0.25	.13552	.078038	.15578	87 10.1	63 51.3	90 56.1	.0014758	29	.0215
(10)	883	8.246 .003	12.850 .004	7.170 .002	93 27.3 2.3	116 12.6 1.7	87 37.3 2.0	680.32 0.41	.13518	.077973	.15562	87 19.2	63 52.5	90 57.2	.0014699	26	.0213
(5)	1010	8.264 .004	12.862 .005	7.176 .003	93 18.2 3.0	116 8.5 1.9	87 33.8 2.5	683.43 0.34	.13483	.077892	.15538	87 30.9	63 56.6	91 5.7	.0014632	26	.0321
(11)	1127	8.278 .006	12.863 .009	7.180 .003	92 46.8 4.0	116 2.7 2.2	87 43.1 3.3	685.93 0.47	.13448	.077848	.15512	88 1.2	64 1.4	91 11.0	.0014579	16	.0207
	26	0.000	0.000	0.000				0.000									
	26 ¹	0.036	0.023	-0.070				0.000									
	26 ²	0.012	0.027	-0.042				0.497									
	141	0.197	0.055	-0.014				0.316									
	245	0.332	0.117	-0.056				0.497									
	364	0.516	0.133	-0.056				0.843									
	488	0.627	0.242	-0.042				1.099									
	628	0.872	0.328	0.042				1.595									
	759	1.094	0.422	0.042				2.378									
	883	1.327	0.493	0.112				2.844									
	1010	1.548	0.587	0.195				3.221									
	1127	1.720	0.594	0.251													

AXIAL AND VOLUME EXPANSION IN PERCENT

¹ After heating to 1010°C.
² After heating to 1127°C.

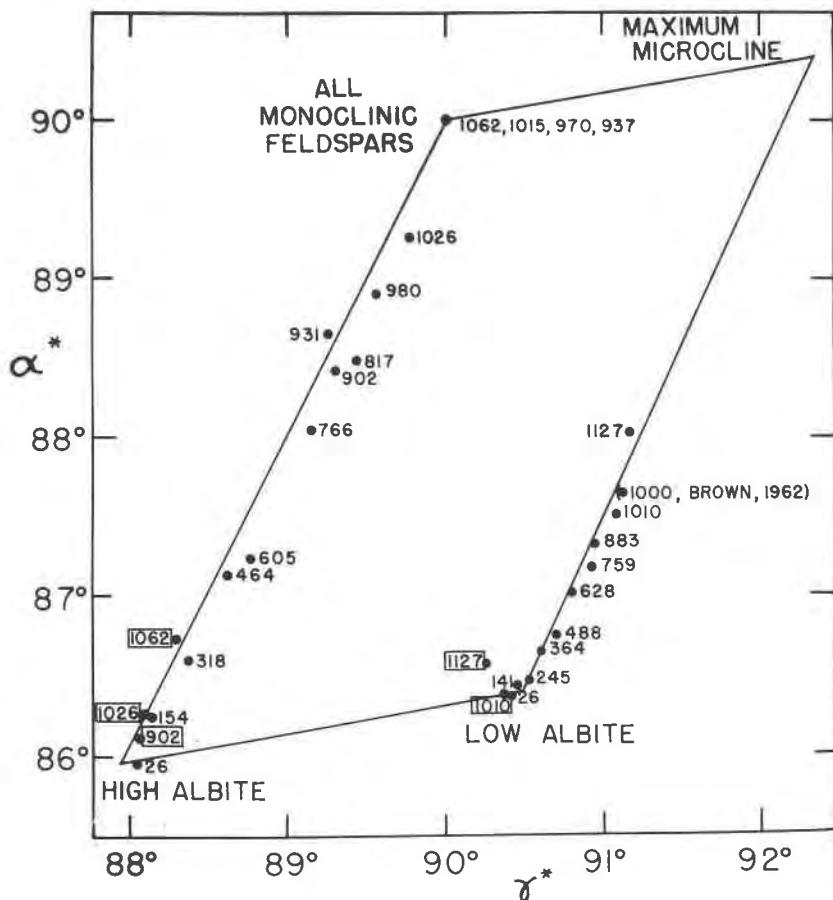


FIG. 3. A plot of α^* and γ^* of low and high albite on heating to the temperatures shown by each point. Measurements made after cooling the sample to room temperature are indicated by boxes drawn around the highest temperature to which the sample was heated. The values for the corners of the parallelogram are from MacKenzie and Smith, 1962.

The effect of heating on these cell angles is the same as that caused by the substitution of potassium for sodium in a solid solution series with the same degree of order of aluminum and silicon. It is supposed that the alkali coordination polyhedron is enlarged by the thermal vibration of the sodium atom, and that the framework "unpuckers" as the temperature increases. Brown earlier reported (1962, p. 362 footnote) that the angles of low albite varied on heating toward those of maximum microcline.

The value of the molar volume at room temperature was given three times unit-weight. The value of A is $-7.8821 \text{ cm}^3 \text{ }^\circ\text{K}/\text{mole}$, $B = 99.5300 \text{ cm}^3/\text{mole}$, $C = 1.6422 \times 10^{-3} \text{ cm}^3/\text{mole } ^\circ\text{K}$, and $D = 7.5321 \times 10^{-7} \text{ cm}^3/\text{mole } (^\circ\text{K})^2$. The standard deviation of the data is $0.0368 \text{ cm}^3/\text{mole}$. The calculated molar volume from the least squares equation, the two expres-

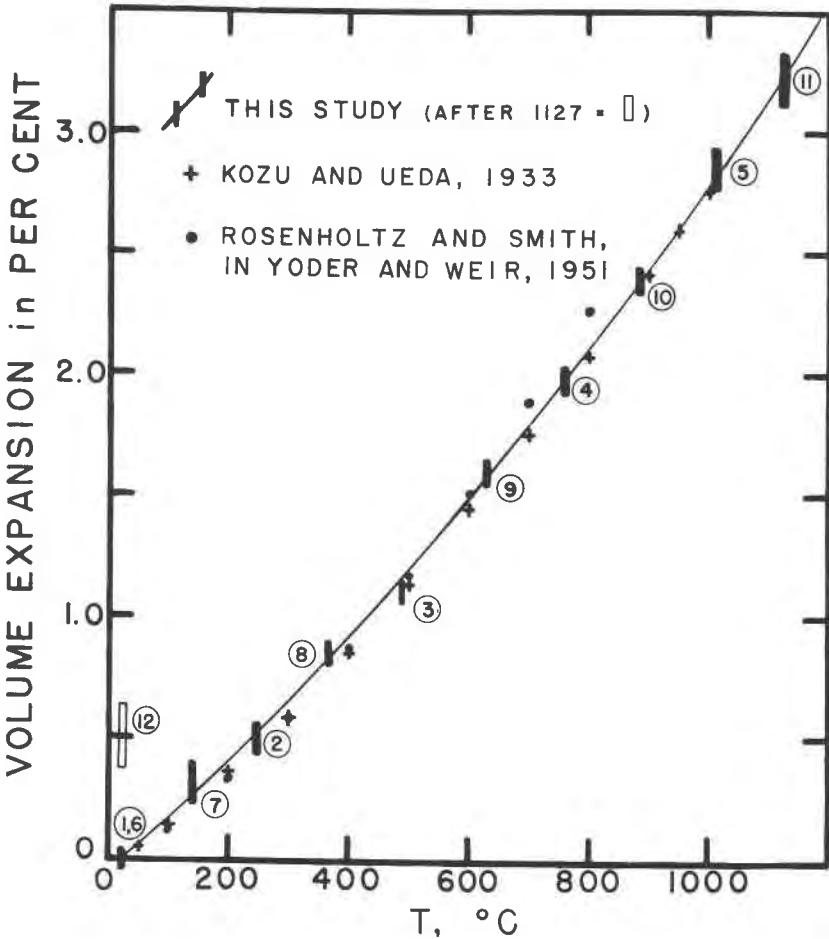


FIG. 4. Volume expansion of low albite as determined by x-ray and dilatometer methods. The curve drawn was fitted by the method of least squares to data from Table 2. The sequence of data collection is shown by numbered circles, demonstrating reversibility. Excellent agreement between methods is shown. Values obtained by Kayode (1965, personal communication) are higher than those shown here.

Because the volume expansion of high albite is so similar to that of low albite (Table 3), it has not been illustrated to avoid confusion.

sions of the thermal expansion $[(dV/dT)_p]$, the derivative of the least squares equation, and α_V , this derivative divided by the volume], and the percent volume change from room temperature are given in Table 3. A graph of α_V is shown on Figure 5.

Cell dimensions of high albite. Cell dimensions for high albite at room

TABLE 3. CALCULATED MOLAR VOLUMES, THERMAL EXPANSIONS, AND PERCENT VOLUME EXPANSIONS OF ALBITE POLYMORPHS

Low Albite						High Albite				
T°C	V cm ³ /mol	$(\partial V/\partial T)_p$ cm ³ / mol °C ×10 ⁻³	αV °C ⁻¹ ×10 ⁻⁵	$\Delta V, \%$		T°C	V cm ³ /mol	$(\partial V/\partial T)_p$ cm ³ / mol °C ×10 ⁻³	αV °C ⁻¹ ×10 ⁻⁵	ΔV_{26} in %
				This study, 26°C	Kozu & Ueda (1933) 20°C					
0	100.006	2.159	2.15	-0.06		0	100.429	-.396	-.40	+0.01%
26	100.062	2.181	2.17	0.00	0.00	26	100.422	.358	.36	0.00
100	100.226	2.261	2.25	0.16	0.14	100	100.482	1.737	1.74	0.06
200	100.459	2.390	2.38	0.40	0.36	200	100.671	2.721	2.72	0.25
300	100.705	2.529	2.51	0.64	0.58	300	100.926	3.280	3.28	0.50
400	100.965	2.613	2.65	0.90	0.85	400	101.219	3.645	3.64	0.79
500	101.260	2.820	2.78	1.20	1.13	500	101.534	3.909	3.91	1.11
600	101.529	2.968	2.92	1.47	1.44	600	101.865	4.126	4.13	1.44
700	101.833	3.116	3.06	1.77	1.75	700	102.206	4.286	4.29	1.78
800	102.152	3.266	3.20	2.09	2.07	800	102.556	4.434	4.43	2.13
900	102.486	3.415	3.33	2.42	2.41	900	102.913	4.565	4.57	2.48
1000	102.835	3.565	3.47	2.77	2.75	1000	103.274	4.686	4.69	2.84
1100	103.199	3.715	3.60	3.14	—	1100	103.639	4.798	4.80	3.20
1200	103.578	3.865	3.73	3.51	—					

temperature are given in Table 4. New refinements of the data reported by Donnay and Donnay (1952) and J. V. Smith (1956) are included, along with other sets of cell dimensions. Our synthetic sample of high

TABLE 4. CELL DIMENSIONS OF HIGH ALBITE AT ROOM TEMPERATURE

a Å	b Å	c Å	α	β	γ	Volume		Remarks
						Å ³	cm ³ /mol	
8.156	12.877	7.103	93° 31.7'	116° 20.1'	90° 11.3'	666.9	100.410	Synthesized at 925°C, 3500 psi, 3 hrs; $\Delta 2\theta_{\text{calc}}$, (131-131) = 1.977°
8.159	12.865	7.104	93° 34.3'	116° 19.9'	90° 11.9'	666.6	100.365	
8.165	12.869	7.111	93° 32.2'	116° 25.4'	90° 10.8'	667.4	100.486	
8.160	12.870	7.106	93° 32.7'	116° 21.8'	90° 11.3'	666.98	100.423	Average of above three determinations, this study.
.005	.007	.005	1.6'	3.6'	0.5'	.40	.060	
8.165	12.872	7.111	93° 27.0'	116° 25.8'	90° 16.8'	668.03	100.581	Smith (1956, p. 54).
8.161	12.872	7.110	93° 28.4'	116° 25.6'	90° 15.3'	667.2	100.456	Heated Amelia; Smith's 1956 data refined by new program.
.001	.002	.001	1.0'	0.7'	0.9'	±.2	.030	
8.171	12.872	7.108	93° 28.2'	116° 23.4'	90° 19.8'	668.02	100.579	Hydrothermal (Smith, 1956).
8.160	12.871	7.110	93° 32.5'	116° 21.7'	90° 14.1'	667.4	100.486	Data of Donnay and Donnay (1952, p. 129) refined by new program after adding 0.025° 2 θ to each observation as suggested by Smith (1956, p. 537).
.003	.002	.002	2.1'	1.6'	1.9'	.3	.045	
8.149	12.880	7.106	93° 22.0'	116° 18.0'	90° 17.0'	666.7	100.380	Heated Amelia (Ferguson, Traill, and Taylor, 1958).

albite is identical within the errors of measurement with samples prepared by heating Amelia albite.

Our indexing of the portion of the powder-diffraction pattern of high albite which we measured agrees very well with that given by Smith (1956, p. 50–51). Our patterns in general are not as complete as his, and we indexed the line at about $42.60^\circ 2\theta$ as $2\bar{4}1$ rather than 003, and at

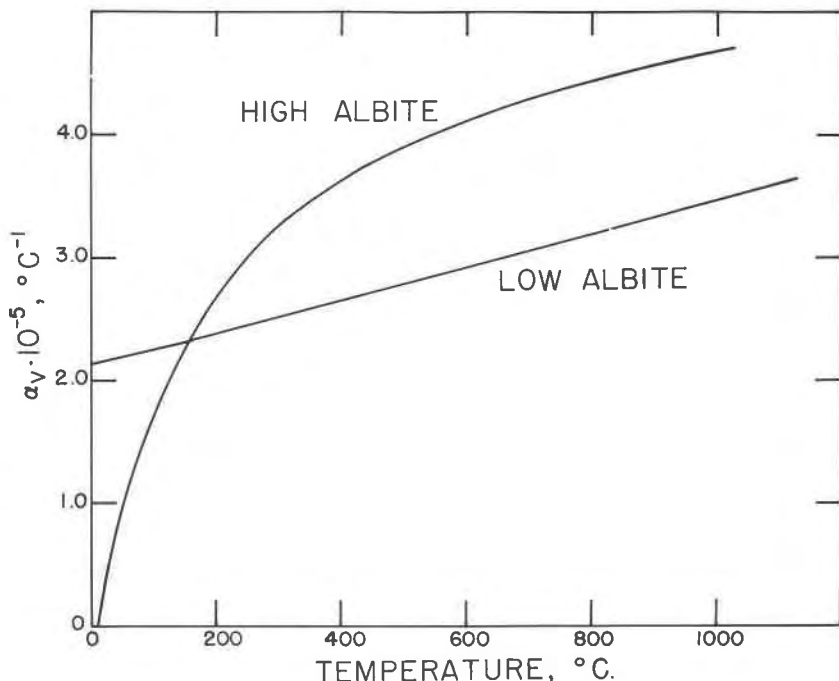


FIG. 5. Graph of the thermal expansion, α_v , of low albite, and high albite with Or < 3 percent, from data given in Table 3.

about 48.50° as $2\bar{6}0$ rather than 222. The line at 45.5° is misprinted in Smith's table as $1\bar{1}3$; it should be $1\bar{3}3$.

Table 5A is a tabulation of cell dimensions of high albite at elevated temperatures, and at lower temperatures and room temperature after exposure to temperatures above 900°C , where irreversible changes due to potassium exchange for sodium first becomes measureable. The sample, though not of pure $\text{NaAlSi}_3\text{O}_8$ composition, was triclinic at 1026°C . Pairs with large separations, such as the $1\bar{3}2$ and $1\bar{3}2$ reflections, were clearly resolved, even though pairs such $111\text{--}\bar{1}\bar{1}\bar{1}$, $130\text{--}\bar{1}\bar{3}0$, etc., were not resolvable. Because of the simultaneous variation in composition during

TABLE 5B. AXIAL AND VOLUME EXPANSIONS OF HIGH ALBITE SAMPLES

Seq. of Meas.	Est. Or cont.	T, °C	a	b percent	c	Volume percent
A1	0	26	0.000	0.000	0.000	0.000
A7	3	26 ¹	0.159	-0.047	0.141	0.193
B6	8	26 ²	0.490	0.249	0.155	0.868
A13	10	26 ³	0.564	0.202	0.169	1.063
A2	0	154	0.123	0.054	-0.028	0.118
A3	0	318	0.233	0.155	0.042	0.523
A6	3	464 ¹	0.502	0.256	0.141	1.078
A4	0	605	0.723	0.326	0.239	1.452
A8	3	766 ¹	1.017	0.489	0.352	2.232
B5	8	817 ²	1.054	0.622	0.450	2.592
A5	3	902	1.115	0.583	0.352	2.472
B1	0	931	1.103	0.614	0.478	2.517
A12	10	937 ³	1.936	0.846	0.675	4.182
A11	9	970 ³	2.132	0.824	0.507	4.136
B2	2	980	1.250	0.598	0.464	2.726
A10	9	1015 ³	2.132	0.839	0.591	4.181
B3	3	1026	1.385	0.660	0.492	2.951
A9	5	1062	1.863	0.723	0.591	3.806

¹ After heating to 902°C.

² After heating to 1026°C.

³ After heating to 1062°C, Or = 12.4% by X-ray fluorescence.

measurements at high temperature, it was not possible to identify the exact temperature of the symmetry change, or even to establish whether it would occur before melting would take place. Various methods of extrapolating the available data were tried without yielding compelling values for the temperature of the symmetry change. One example is given as Figure 1, where a temperature of $1100^{\circ}\text{C} \pm 50^{\circ}\text{C}$ was estimated for our sample. Most other extrapolations gave comparable results when allowance of about -30°C per 1% Or was made for the compositional change measured after cooling the samples to room temperature. The value for the compositional effect was taken from MacKenzie (1952, Fig. 3).

A. A. Kayode (personal communication, 1965) reported that his sample of high albite, prepared by long heating of Amelia albite, was still triclinic at 1100°C . In a very careful X-ray study of the superheating of platy low albite (cleavelandite, $\text{Ab}_{98.5}\text{Or}_{0.5}\text{An}_{1.0}$) from Portland, Connecticut, and of the high albite produced from it by heating at $1050^{\circ}\text{C} \pm 5^{\circ}\text{C}$ for 8 weeks in an electric muffle furnace, Dietz (1965), p. 52-53 reported "However, there is little doubt that the high modification of cleavelandite albite transforms to the monoclinic above its own melting point, and somewhere in the range of 1150°C ." Dietz used chromium radiation which yields a larger separation of the $111\text{-}\bar{1}\bar{1}1$ peaks, and hence requires a shorter extrapolation along the temperature axis to zero separation.

Thermal expansion of high albite. There were no earlier measurements of the thermal expansion of high albite when the experimental part of this study was completed, and the problem of alkali exchange with ceramics had not been recognized as such. In view of present knowledge, only data for experiments in which potassium exchange was less than or equivalent to 3 percent Or have been used. The remaining data are applicable to 1026°C. The molar volume at 26°C was given three times unit weight, and a least-squares curve was fitted to the thermal expansion equation given above. The constants are

$$\begin{aligned} A &= 327.1717 \text{ cm}^3 \text{ }^\circ\text{K/mole,} \\ B &= 98.2150 \text{ cm}^3/\text{mole,} \\ C &= 3.7053 \times 10^{-3} \text{ cm}^3/\text{mole }^\circ\text{K,} \\ D &= 5.2459 \times 10^{-7} \text{ cm}^3/\text{mole } (^\circ\text{K})^2, \end{aligned}$$

with a standard deviation of 0.0354 cm³/mole. The molar volumes at various temperatures calculated from this equation are given in Table 3 along with the various expressions for the thermal expansion. Caution must be used in accepting these values; a study in which alkali contamination is strictly avoided is needed. The work of Kayode (1964) may be helpful, if it can be established that alkali transfer was avoided.

Comparison of properties of low and high albite. Graphs of the variations of cell-edge parameters and cell volume of low albite and high albite are given in Figures 6 and 7. The graphs demonstrate the regular and reversible variation in the dimensions of high and low albite until alkali exchange becomes a problem at high temperature. The reversibility of the thermal expansion data up to at least 900°C for both high and low albite show that the structures of these polymorphs are not easily disturbed. Although it is true that sluggish transformations are desirable in order to preserve a record of the geologic history of a sample, it is also true that it is the sluggishness of aluminum-silicon interdiffusion that generates so many complex and confusing structural states in albite. It may well be that when our knowledge of factors that influence these reactions is more complete their existence will be a more obvious boon than it is at present.

The graphs demonstrate the subparallel variation with temperature of the various parameters of high and low albite and make it reasonable to suppose that all the parameters of intermediate albites will lie between the values for high and low albite at corresponding temperatures.

The percent volume thermal expansions of low and high albite are almost identical, as can be seen from Table 3. Figure 5 shows that the ther-

mal expansion, α_V , differs, but the temperature coefficient of α_V has essentially the same value in high and low albite at temperatures above 600°C.

The agreement between the percent volume expansion of low albite determined by dilatometer and X-ray methods would at first seem surprising because higher order terms involving changes of the cell angles can not be evaluated in dilatometry. The angular changes are small in low albite, and the departure of the faces of the initial cube from mutually perpendicular for the wavelengths of visible light would be small also.

As was noted for calcic labradorite (Stewart, Walker, Wright, and Fahey, 1966) the sum of the three nonorthogonal linear expansions of both high and low albite sum to less than the true volume expansion (compare Yoder and Weir, 1951, p. 684). The orthogonal axes of the ellipsoid of thermal dilation cannot be coincident with all of the crystallographic axes of triclinic crystals, and are not constrained by symmetry to bear any simple relationship to them. The axis of greatest thermal dilation is closest to the a axial direction in both high and low albite.

The cell angles and edges of low and high albite vary in the same way with increase of temperature as with replacement of Na atoms by K atoms in the respective solid solution series at room temperature. This suggests that the effect of temperature is to increase the vibration of the sodium atoms sufficiently so that the alkali coordination polyhedron is enlarged by an amount comparable to that found in solid solutions containing approximately 35 percent Or. Equivalent temperature increments cause greater angular changes in high albite than in low albite (Fig. 3), so aluminum-silicon disorder in the framework appears to favor easier distortion of the tetrahedral linkage. It was noted above that alkali exchange was more rapid in high albite than low albite, and presumably the rate of diffusion is greater in the more easily deformed disordered structure. Dietz (1965) found that low albite superheats to a much greater extent than does high albite, which also indicates the greater intrinsic strength of the ordered tetrahedral framework.

Variation of obliquity, ϕ , and orientation of rhombic section, σ . The unit-cell parameters have been used to calculate the variation with temperature of ϕ , the obliquity of albite twins (Donnay, 1940; Smith, 1958; Stewart and others, 1966) and σ , the angle between the a -axis and the trace of the rhombic section on $\{010\}$. A summary of earlier data on σ is given by Smith (1958); the topic is discussed also by Brown (1962, p. 362-364), Smith (1962), Kayode (1964), and Barth and Thoresen (1965).

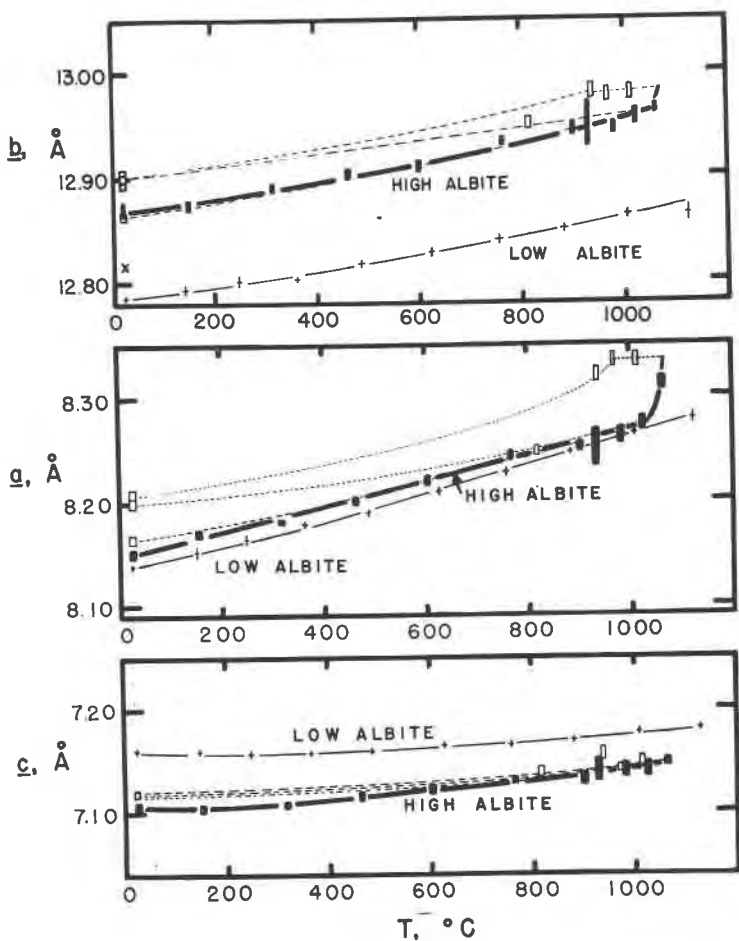


FIG. 6. Variations of axial parameters δ , of high and low albite with temperature, showing reversible and irreversible effects. Symbols are elongated \pm one standard error to indicate relative quality of the measurements.

High albite that has changed composition to become more potassic than Or_3 during heating is marked by squares. Presumed cooling paths are shown by dashed lines.

The smaller the obliquity, ϕ , the easier twinning becomes, except, of course, albite twinning is not possible at $\phi = 0$. As shown in Table 6, ϕ decreased linearly with temperature, but the change was only about 1.3° in 1100°C (a linear decrease of about 0.9° in 1100°C was observed in calcic labradorite by Stewart and others, 1966). The variation of ϕ in high albite was linear over the interval from 25° to 900°C in which compositional changes were negligible; the variation was almost two and a half times

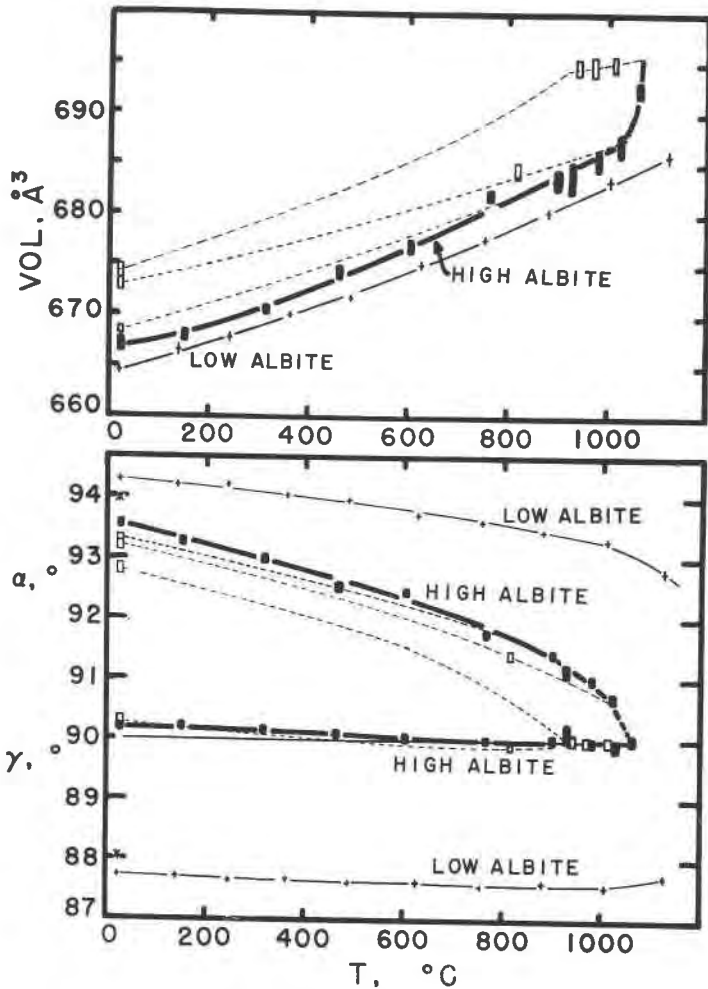


FIG. 7. Variations of unit cell angles (except β) and volume of high and low albite with temperature, showing reversible and irreversible effects. β differs little in high and low albite, and varies little with temperature (Tables 2 and 5). Values of γ for high albite at room temperature are equivalent for all cooling paths observed.

Low albite at 26°C after heating to 1010°C is changed less than one standard error from the parameters of the starting material. After heating to 1127°C α and γ are as shown by x's; a and c are not changed by their standard errors from the starting material, and the cell volume is 667.75 Å³.

greater than that observed in low albite. Projection of the lines to $\phi=0$ (monoclinic symmetry) indicates that high albite will melt before becoming monoclinic.

TABLE 6. EFFECT OF HEATING LOW AND HIGH ALBITE ON THE OBLIQUITY, ϕ , AND THE ANGLE BETWEEN THE a -AXIS AND THE TRACE OF THE RHOMBIC SECTION ON $\{010\}$, σ

Low albite																		
$T^{\circ}\text{C}$	26	26 ¹	26 ²	141	245	364	488	628	759	883	1010	1127						
ϕ	4°17.5'	4°14.5'	3°55.5'	4°13.5'	4°15.0'	4° 4.5'	4° 0.5'	3°48.0'	3°44.0'	3°35.0'	3°28.5'	3°1.0'						
σ	32°14.7'	31°42.2'	30° 0.9'	32°47.5'	33°48.8'	34°53.6'	36°32.3'	38°29.0'	40°43.3'	41°36.1'	44°28.0'	49°4.1'						
High albite																		
$T^{\circ}\text{C}$	26	26 ³	26 ⁴	26 ⁶	154	318	464 ³	605	766 ³	817 ⁴	902	931	937 ⁵	970 ⁵	980	1015 ⁵	1026	1062
ϕ	-4° 3.0'	-3°50.5'	-3°44.5'	-4°10.5'	-3°49.0'	-3°24.5'	-2°52.5'	-2°46.0'	-1°57.0'	-1°32.0'	-1°36.0'	-1°23.0'	0° 0'	0° 0'	-1° 5.0'	0° 0'	-0° 46'	0° 0'
σ	-2°41.5'	-3°58.7'	-4°24.4'	-5°11.3'	-3°30.1'	-2°24.8'	-2°31.6'	-0°32.7'	0°12.1'	4° 7.3'	-0°28.5'	-6°54.7'	0° 0'	0° 0'	2°34.9'	0° 0'	9°0.4'	0° 0'

¹ After heating to 1010°C.

² After heating to 1127°C.

³ After heating to 902°C.

⁴ After heating to 1026°C.

⁵ After heating to 1062°C.

The position of the rhombic section, the composition plane of pericline twins, is strongly temperature-dependent in both high and low albite, and especially in the latter (see Table 6). The orientation of the rhombic section in low albite changes ever more rapidly with increasing temperature and may well be similar in high albite, except that as the cell angles approach 90° at high temperatures very slight errors in their determination produce very large errors in the calculated value of σ , so that resolution is lost. In addition, with compositional change our sample tends toward monoclinic symmetry and this strongly affects the observed scatter of σ . The permanent change in σ on cooling is that expected by comparison with σ calculated by MacKenzie (1956, table 1) for synthetic sodic anorthoclases.

The small variation of ϕ of low albite with temperature makes it improbable that any correlation of the frequency of twinning with temperature can be established, and, indeed, the low albite of metamorphic rocks occurs either as single crystals or very coarse albite twins throughout the temperature range of its occurrence.

The closest natural equivalents of high albite formed under equilibrium conditions are the potassium- and calcium-bearing anorthoclases, some of which are monoclinic at magmatic temperatures (Laves, 1952; Smith and MacKenzie, 1958). Only feldspars with $Or < 15$ are triclinic at magmatic temperatures and might be expected to reveal a temperature dependence in their frequency of twinning, but such compositions are very rare in nature. More potassic samples are highly twinned after both the albite and pericline laws on cooling (Laves, 1950; MacKenzie, 1956), and were quite likely monoclinic at the time of formation.

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