

A review of the expansion behavior of alkali feldspars: coupled variations in cell parameters and possible phase transitions

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Abstract

The literature on feldspar unit-cell parameters and phase transitions is reviewed, and the data are used to examine systematics of alkali feldspar expansion as a function of composition, temperature and pressure. Several different trends in expansion behavior are identified, and these trends may be linked by phase transitions. Compositional expansion can be divided into two main trends (I and II), possibly separated by a phase transition like that occurring in microcline at low temperature. Trend I is characterized by a coupled *b*–*c* expansion, whereas in Trend II, *b* and *c* are uncoupled. Trend I is divided into two parts (IA and IB) at the displacive triclinic–monoclinic transition in disordered feldspars or at the analogous changes in ordered AlSi_3 feldspars. Thermal expansion may or may not follow Trend I, depending on the size of the cavity cation (*M*). The trends and phase transitions are analyzed in terms of the available structure refinements. It is thought that the expansion is dominated by the framework and not by expansion of the short *M*–*O* bonds.

The thermodynamic nature of the transitions is discussed. The monoclinic–triclinic symmetry change is an almost pure second-order transition, whereas the microcline transition involves hysteresis and may be a smeared (partially martensitic ?) transition.

Introduction

There is considerable interest in the response of mineral structures to changes in composition, temperature and pressure (Hazen, 1976a,b; 1977; Hazen and Prewitt, 1977a,b; Taylor, 1983). The present article discusses the expansion behavior of the alkali feldspars from published unit cell data as a function of pressure (*P*), temperature

(*T*), and composition (*X*) for both the framework and large cavity cations. The unit cell parameters show certain systematic variations, which may be associated with distinct types of expansion behavior, or expansion trends, in different regions of *P*–*T*–*X* space.

Furthermore, the feldspars show several types of phase transitions which are either diffusion-controlled and slow (diffusive) or independent of diffusion and rapid (displacive). One type of diffusionless transition that has received much attention is the displacive monoclinic–triclinic transition observed in disordered alkali feldspars. A second type of diffusionless transition occurs in anorthite and involves small movements, mainly of the *Ca* atoms,

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producing antiphase domains. Both of these transitions are rapid and unquenchable (for references see Smith, 1974; also Adlhart et al., 1980; Harlow, 1982). A third type of transition has been described for an ion-exchanged microcline (Openshaw et al., 1979 a,b; Wynnke et al., 1981). This is also considered to be diffusionless, as it occurs rapidly on cooling below room temperature in a feldspar with complete Al,Si order, and it may involve hysteresis on reheating. Thompson and Hovis (1979) suggested that the phase transition in microcline is of the same type as the displacive monoclinic–triclinic transition, but we believe it to be an entirely new type of transition in feldspars and its possible nature is discussed below.

Taylor (1972) showed that the thermal expansion of certain framework silicates, including leucite, quartz and cristobalite, occurs by untwisting of the partially collapsed tetrahedral framework towards a more symmetrical “ideal fully expanded state”. A discontinuity occurs in the expansion rate when this state is reached. Leucite shows two discontinuities in its expansion behavior, the first occurring at the tetragonal-cubic inversion and the second when the structure attains the “ideal fully expanded state” (Taylor and Henderson, 1968; Taylor, 1972). As feldspars are much less symmetrical than leucite, quartz or cristobalite, fully expanded *planes* or *directions* and limiting values to *cell angles* might be expected rather than a unique fully expanded state. There should be a change in expansion behavior once a fully expanded direction or plane is attained and this could show up as abrupt changes on plots of unit cell parameters or in the orientations of the expansion ellipsoids. Whether or not a given feldspar has achieved a fully expanded direction or plane depends on the nature and degree of order of the tetrahedrally coordinated cations, the size of the cavity cation, and the temperature and pressure at which the cell parameters are measured. We present below an interpretation of the response of the feldspar framework to changes in temperature, pressure, and composition that distinguishes several major types of expansion characterized by the coupling and uncoupling of cell parameters.

Expansion of alkali feldspars as a function of composition, temperature and pressure

Compositional expansion of alkali feldspars

In this section we consider the effect on a given feldspar framework of changing the large cavity cation. To cover as large a range of cell parameters as possible we have used data from the literature for feldspars with AlSi_3 , GaSi_3 , AlGe_3 , GaGe_3 and FeGe_3 frameworks and with Na, K and Rb cavity cations. The Al_2Si_2 -framework with Ca, Sr, Pb and Ba cavity cations is only briefly discussed. End-member feldspars are discussed first and the AlSi_3 solid-solution series are considered in more detail in a following subsection. References to the original

data are given in the figures. We show here that the covariation of certain cell parameters may be used to characterize different stages in feldspar expansion.

End-member feldspars. Figures 1a and 1b are plots of b vs. c for feldspar end members. For all alkali feldspar frameworks, the K end member has larger a , b and c parameters and volume and smaller β than the Na end member. The percentage changes (a , +5 to 6%, b , +1 to 2%, c , +0.6 to 1.2%, β , -0.2 to -0.5%) and the b - c slopes for all frameworks are essentially the same, suggesting that the same expansion mechanism is operating in each case. The Rb end members have larger a parameters and volumes, larger β and slightly larger c parameters than the K end members. The behavior of b on substituting Rb for K is more complex and is summarized below:

Framework type	behavior of b (K \rightarrow Rb)
disordered AlSi_3	increases $\sim 0.11\%$
disordered AlGe_3	increases $\sim 0.09\%$
disordered FeGe_3	remains constant
disordered GaGe_3	decreases $\sim 0.02\%$
ordered AlSi_3	decreases $\sim 0.04\%$
disordered GaSi_3	decreases $\sim 0.08\%$
ordered GaSi_3	decreases $\sim 0.13\%$

It is of interest that, in general, the Rb-feldspars are not collinear with their Na and K-analogues on these plots. This suggests that K \rightarrow Na and Rb \rightarrow K substitutions in alkali feldspars result in different types of expansion of the feldspar framework. K for Na substitution causes an increase in both b and c , whereas in Rb \rightarrow K substitution, b remains nearly constant relative to an increase in c , with the exception of disordered AlSi_3 and AlGe_3 feldspars. Here we refer to the former type of expansion as b - c coupled (Trend I), whereas the latter with no clear covariation in the parameters as being b - c uncoupled (Trend II). This uncoupling seems to occur generally with K as the cavity cation, but is also a function of the framework chemistry and ordering state. Similar relationships are also shown for Ca, Sr, Pb and BaAl_2Si_2 -feldspars (Fig. 1a) and perhaps also for alkaline-earth Al_2Ge_2 -feldspars (Nager, 1974), but unfortunately no cell-parameter data exist for PbAl_2Ge_2 -feldspar.

The corresponding angular relationships in triclinic feldspars are described briefly. Substitution of K for Na in AlSi_3 and GaSi_3 feldspars (Fig. 2) produces an increase in α^* and no change in γ . In contrast, substitution of Rb for K in microcline or in ordered KGaSi_3O_8 produces no change in α^* and an increase in γ . These latter changes are quite different from those produced by Na-substitution but are similar to those produced by Al,Si disordering. Thus α^* (or α) and γ show one type of behavior in K for Na substitution and a different one in Rb for K substitution in ordered Al or Ga silicate feldspars.

AlSi_3 solid-solution series. The general features of the

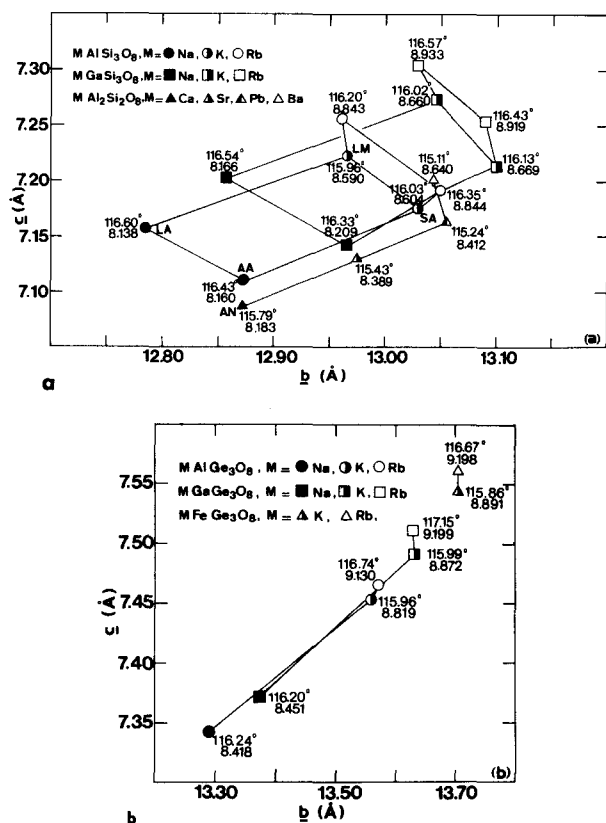


Fig. 1. Plot of the b and c parameters of end-member feldspars. For each end-member the values of β and a are given. (a) Al and Ga silicate feldspars. LA—low albite, AA—analbite, LM—low microcline, SA—sanidine, AN—anorthite. Lattice parameter data from Smith (1974, v. 1, Table 7-1, p. 219) except for disordered $\text{RbAlSi}_3\text{O}_8$ (Henderson, 1978), disordered $\text{RbGaSi}_3\text{O}_8$ (Henderson, unpublished data), ordered $\text{RbAlSi}_3\text{O}_8$ (McMillan et al., 1980) and ordered $\text{RbGaSi}_3\text{O}_8$ (Pentinghaus, 1977, pers. comm.). Ordered (upper left) and disordered (lower right) pairs have nearly the same values of β and a . a increases with ionic radius, whereas β decreases from Na to K and increases from K to Rb end-members. The slopes of lines joining Na and K end-members for both ordered and disordered frameworks are the same as those joining Ca, Sr and Pb feldspars. Abrupt changes of slope occur between most K-Rb end-members and between Pb-Ba end-members. (b) Al, Ga and Fe disordered germanate feldspars. Lattice parameter data from Smith (op. cit.). The slopes joining Na and K germanate feldspars are the same as for silicate feldspars (scale for c twice that for b). There are again abrupt changes of slope between certain K and Rb feldspars.

expansion behavior of the feldspar framework described above are confirmed by the AlSi_3 solid solutions. Bruno and Pentinghaus (1974) plotted room-temperature cell parameters for various feldspar frameworks against the ionic radii of the cavity cations and showed that the relationships for cell volumes are almost linear, whereas those for a , b and c are not collinear and were interpreted

as showing changes in slope. Similar changes in slope were found for these parameters by McMillan et al. (1980).

The b and c parameters for the high albite-sanidine exchange series of Orville (1967) are shown in Figure 3a and points for intermediate compositions lie on or very close to the line joining the end members. This is also the case for other high albite-sanidine series (Donnay and Donnay, 1952; Luth and Querol Suñé, 1970; Hovis, 1977; not shown in Fig. 3a) and for an orthoclase series (Fig. 3a). This correlation is well known and supports the coupling of b and c as described in the previous subsection. The four points close together at the top right of the K-Na disordered series cover the compositional range Or_{84-100} (mol. %) and, as already suggested by the end-member data, b and c appear to be approaching a limit close to pure sanidine. This is clearly seen in plots of b and c against composition (Orville, 1967, his Fig. 3). The covariation of b and c parameters in the above series of feldspars is not affected by the presence of the monoclinic-triclinic transition (Fig. 3a, arrows). This is also true for end-member and intermediate Al_2Si_2 feldspars with Ca (triclinic) and Sr (monoclinic) as cavity cations (Nager, 1974; Bambauer and Nager, 1981).

In ordered alkali AlSi_3 feldspars at room temperature the partial substitution of K for Na in albite-rich compositions produces a coupled change of b and c similar to that in disordered feldspars (Fig. 3a); the relationships in the K-rich compositions are more complex (Hovis and Peckins, 1978; Fig. 3a). However, the substitution of Rb and K in microcline produces changes quite different from those in sanidine but similar to those in Ga-Si and Ga-Ge-feldspars. c shows substantial increase (Fig. 1a, 3a), whereas b is slightly erratic for intermediate compositions (Fig. 3a; McMillan et al., 1980), but tends to show very little change.

Examination of other plots reveals further details of the expansion behavior. Figure 3b shows a plot of b versus a for the same AlSi_3 feldspar series. Initially, b and a increase linearly showing coupled behavior. The triclinic-monoclinic inversion points in the two disordered series are again shown as arrows. Beyond these arrows the curvature of the lines on the b - a plot becomes marked with increase in average cation size. An analogous change in slope in the ordered series occurs near $b = 12.9\text{Å}$, at a composition near Or_{40} close to the first break in slope of b versus cation radius as pointed out by McMillan et al. (1980, their Fig. 2). As a continues to increase at the same rate with increasing cation size (McMillan et al., 1980, their Fig. 2), this curvature must be due to a gradually decreasing expansion along b , which nevertheless remains coupled with c . Finally, for Rb substitution in the K end members, the a parameter increases rapidly in relation to b , especially in the more ordered series. We find this to be general for the expansion behavior of feldspars in which b and c are uncoupled.

The detail of the cell angle relationships for the Na-K

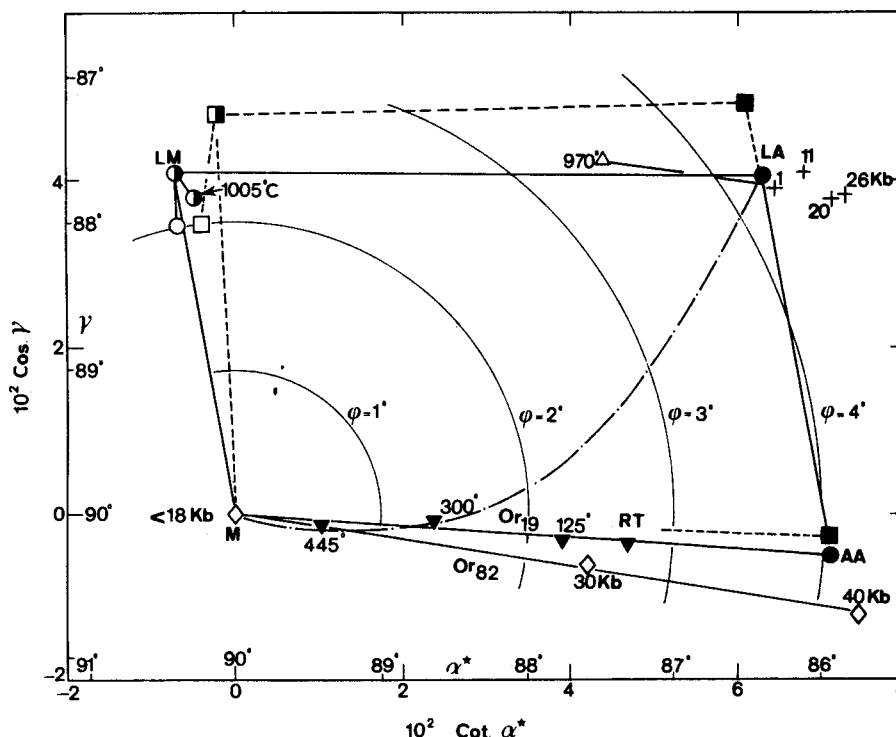


Fig. 2. Plot of $\cos \gamma$ against $\cot \alpha^*$ for Al and Ga silicate feldspars. Symbols in part as in Fig. 1a, M—monoclinic, RT—room temperature. Lattice angles taken or calculated from data in Smith (1974) and from sources in caption to Fig. 1a with, in addition, microcline at 1005°C (Openshaw et al., 1979 a), Δ low albite at 970°C (Winter et al., 1977), + low albite at 1–26 kbar (Hazen and Prewitt, 1977 a), ∇ Or₁₉ at room and high temperatures (Henderson, 1979), \diamond Or₈₂ at high pressure (Hazen, 1976a). Dash-dot curved line for equilibrated albites from Thompson and Hovis (1979) after Grundy and Brown (1969). Circles of equal obliquity, ϕ , in degrees are also given.

solid solution series can be seen in Figure 7 of Thompson and Hovis (1978)—the points lie on nearly straight lines parallel to the $\cot \alpha^*$ axis (cf. Fig. 2). For clarity, only the end point for Rb substitution in microcline is plotted in Figure 2, the intermediate compositions plotting on the line joining K to Rb.

The situation is quite different in $\text{AlSi}_3\text{--Al}_2\text{Si}_2$ solid solutions. On substituting anorthite for albite in the plagioclase series, points for intermediate compositions follow a straight line from LA to AA on Figure 1a up to about An₆₀. Thereafter, b appears to be fully expanded and the points lie on a vertical line joining AA to AN. A similar change in behavior does not exist for the angles, as points for the plagioclases lie on the line joining LA to AA (Fig. 2) and its extension to AN (just off Fig. 2 at 7.13 and -2.20).

The two trends (I and II) obtained from end-member feldspars are confirmed by the solid solution series and can be further refined. Trend I may be divided into two sub-trends IA and IB (corresponding to expansion modes A and B of McMillan et al., 1980). Trend IA represents the initial expansion on substitution of K for Na up to the triclinic/monoclinic symmetry change. Trend IB operates

for feldspars with higher K and is related to IA by the removal of a b - c expansion component. This effect was also noted by Harlow (1982). The b - c decoupling relating Trends I and II generally occurs when the average cavity cation size is at or just smaller than that of K.

Thermal expansion of feldspars

Figures 2 and 4a show the α^* - γ and b - c covariations with temperature for several Na-rich AlSi_3 feldspars. The thermal expansion is parallel to Trend I observed for compositional expansion, suggesting a similar expansion mode. In addition, a increases and β decreases with increasing T , as for K-substitution. The effect of increasing temperature on the cell parameters of high albite and analbite is very similar to substitution of K for Na. Over the temperature range studied, a rise of 1000°C has an effect similar to the substitution of about 35% Or (Grundy and Brown, 1969).

Similar behavior is shown by two Na-rich synthetic feldspars (Or₁₉ and Or₃₈—Figs. 2, 4a and b) studied by Henderson (1979). The a parameters of these samples show substantial increases with increasing T , whereas β for Or₁₉ decreases and may show a discontinuity. The

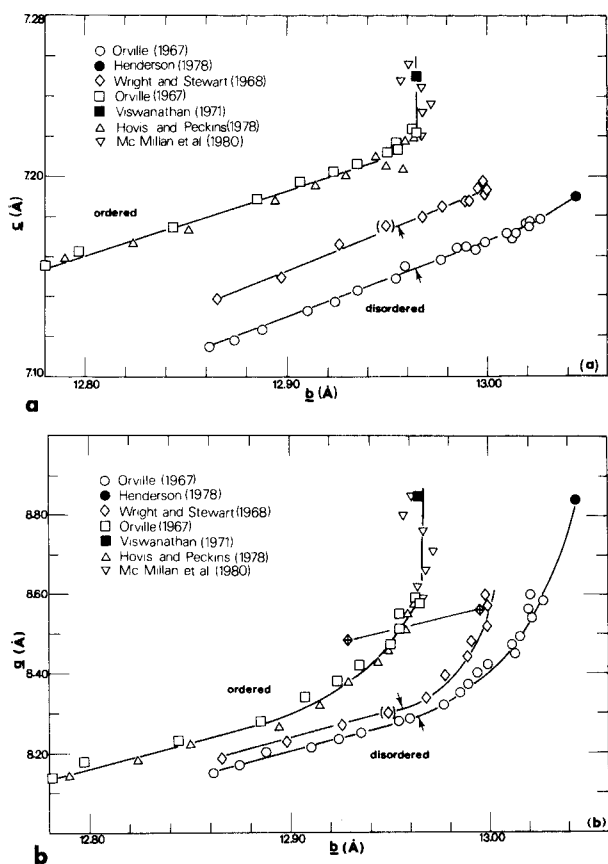


Fig. 3. Plots for the Na-K-Rb solid solution series in the AlSi_3 framework at room temperature. The arrows show the triclinic/monoclinic transition. (a) Plot of b against c for the disordered and ordered series, as well as for an orthoclase ion-exchange series (Wright and Stewart, 1968). (b) Plot of b against a for the same series. The variation is linear up to the phase transition or its analogue in the ordered series (Trend IA) and then steepens considerably as b then increases much less rapidly relative to a and ionic radius (Trend IB). The effect of an increase of pressure of 10 kbar on the parameters of orthoclase, Φ , is shown (Bridgman, 1928).

effect of T on the b and c parameters is again very similar to the substitution of further K for Na, but is smaller the higher the Or content (Fig. 4a and b). The triclinic-monoclinic inversion in Or_{19} (Fig. 2) has no effect on the coupling of b and c , though again the expansion rates of b and c clearly decrease above the inversion temperature (Fig. 1 in Henderson, 1979).

The thermal expansion behavior of K and Rb-rich feldspars is more complex though in all cases a increases and β decreases with T . In sanidine (Henderson, 1979) b and c decrease very slightly with T and plot close to the high albite-sanidine join, but trending towards high albite (Fig. 4b). The data of Ohashi and Finger (1974; 1975) for Laacher See sanidine at 25, 400 and 800°C show the same

behavior. On increasing temperature, microcline (Fig. 4b and 5) shows a larger decrease of b than sanidine and virtually no change in c . In the same way as for b and c , the changes in α^* and γ (Fig. 2) are intermediate between Trends I and II. The thermal expansion ellipsoids of microcline (Openshaw et al., 1979a) and sanidine (Henderson, 1979) are very different from those of Ab-rich feldspars (Willaime et al., 1974). With increasing T the b and c parameters of Rb-Al and Rb-Ga silicate feldspars (Henderson, 1978 and unpublished data), are also uncoupled. For both these phases the trends with increasing T are very different from those of Na-feldspars and are similar to those for K-rich feldspars though the effect is greater (Fig. 4b).

It appears that the thermal expansion of Na-rich feldspars parallels the compositional expansion along Trend I, up to a certain limit. For feldspars which have reached or nearly reached the compositional expansion limit of Trend I, a different thermal expansion trend is initiated. This we have called Trend III, to differentiate it from that observed in compositional expansion along Trend II. The relations between trends for microcline are illustrated in Figures 2 and 5.

Effect of pressure on feldspar cell parameters

Cell parameters or compressibilities have been measured at high pressure for a few feldspars: results are summarized in Figures 2, 3b and 4a. For all feldspars, pressure has the opposite effect to that of K for Na substitution— a , b , c , and V decrease, and β increases. Data are shown for low albite (Hazen and Prewitt, 1977a), orthoclase (Bridgman, 1928) and two natural sanidines (Or_{67} and Or_{82} , Hazen, 1976a). Bridgman (1928) gave only fractional changes in a , b , and c and these were applied to the lattice parameters of an orthoclase from Wright and Stewart (1968) in Figures 3b and 4a. In all cases both for b - c and α^* - γ , it may be observed that the cell parameters decrease nearly parallel to Trend I, suggesting a compression mode opposite to that of Trend I expansion. This is confirmed as a decreases (Fig. 3b) and β increases with P . The effect of pressure on low albite produces a compression opposite to that of K substitution, b and a being coupled. The points for the compression of two sanidines (Hazen, 1976a) follow either the sanidine curve (Or_{67}) or the orthoclase curve (Or_{82}); for both, the compressibility of b is linear up to 40 kbar, that of a decreases with pressure. The data from Bridgman (1928) shown in Figure 3b appear to indicate linear compression parallel to Trend IA, though they may be less precise. Due to lack of data, it is not possible at present to analyze the compression behavior of feldspars beyond Trend I.

Summary of expansion trends of the alkali feldspars

Trend I. This trend is characterized by a coupled b - c expansion. No lower limit has yet been found to this trend

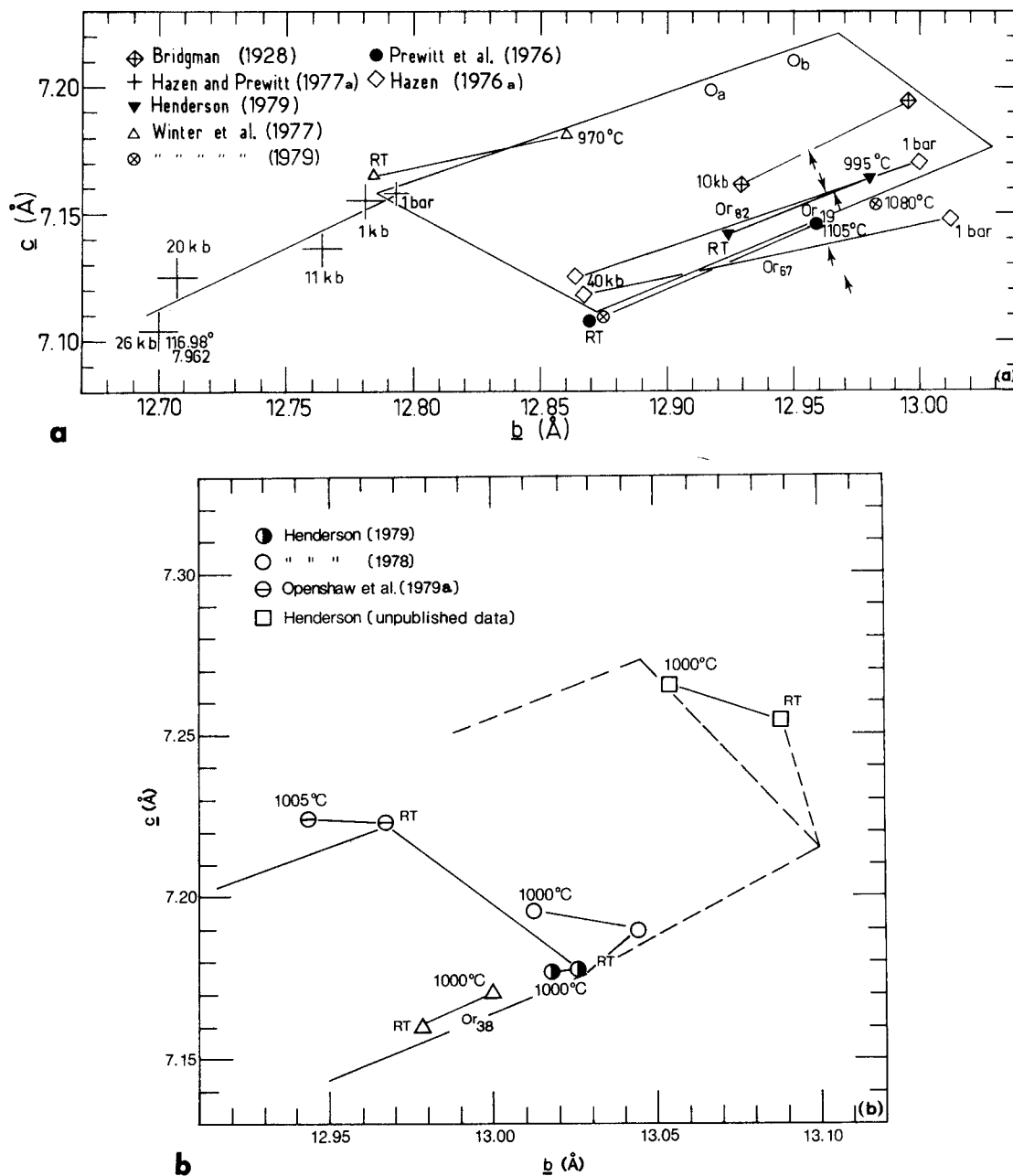


Fig. 4. Plots of *b* against *c* for AlSi₃ feldspars showing effects of temperature or pressure. (a) Effect of *P* on Na-K feldspars and of *T* on Na-rich feldspars. Arrows show the triclinic-monoclinic transitions for the different feldspars. In addition, the lowest arrow is for the Ca-Sr series from Bambauer and Nager (1981) and the highest for the orthoclase series (Fig. 3b). a and b represent the positions of the analogous changes in the ordered series at Or₄₀ and Or₇₀. Low albite (+, Δ), high albite (●, ⊗), Or₁₉ (▼), sanidine Or₆₇ and Or₈₂ (◇), orthoclase (⊕); RT room temperature. (b) The effect of *T* on Or₃₈ and on K and Rb feldspars. Or₃₈ follows Trend I with increasing *T*, whereas Or₁₀₀ follows an inverse trend. For microcline, and Rb feldspars, Trend III is more variable. Or₃₈ (Δ), sanidine Or₁₀₀ (●), microcline (⊖), disordered RbAlSi₃O₈ (○), disordered RbGaSi₃O₈ (□); RT—room temperature.

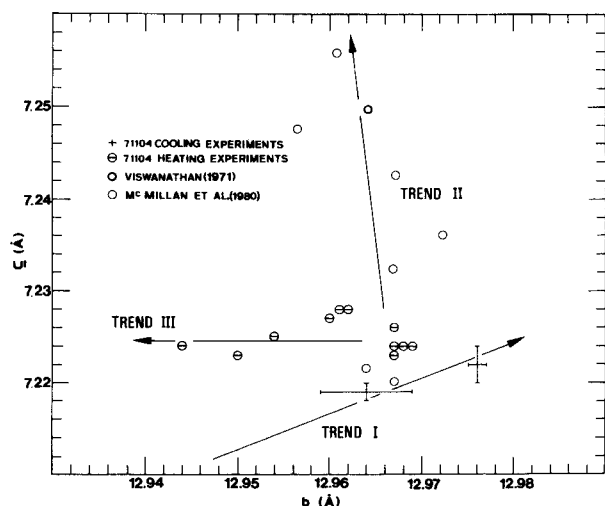


Fig. 5. Plot of b against c for ordered K-Rb-AlSi₃ feldspars showing the three Trends: I Na-K substitution or cooling (+ (Openshaw *et al.*, 1979 a); II K-Rb substitution (O) III heating of microcline Θ (Openshaw *et al.*, 1979 a). See text for detailed explanation.

with pressure. The boron feldspar, reedmergnerite, also lies nearly on the same trend with much smaller b and c values.

This trend can be divided into two parts, IA and IB, separated by the triclinic-monoclinic transition or its analogue in ordered feldspars, the latter occurring at room temperature at a composition near Or₄₀ (McMillan *et al.*, 1980; Hovis, 1980). Up to this transition or its analogue, γ remains unchanged whereas α^* , a , b , c and the volume increase linearly and β decreases with increasing cation size, temperature or reduction in pressure (Vogel *et al.*, 1973; McMillan *et al.*, 1980). Beyond the transition, a continues to increase at the same rate relative to the average cation radius, whereas b and c increase less rapidly, but remain coupled throughout the whole of Trend I. We consider that the triclinic-monoclinic inversion or its analogue in ordered AlSi₃ feldspars marks the removal of one expansion component in the b - c plane. Though the transition is very important in the development of microscopic textures, in Al,Si ordering, and for the thermodynamic properties, it does not eliminate b - c coupling, which occurs throughout Trend I.

Trend II. This trend characterizes the compositional expansion for all the frameworks studied after the limit for the coupled b - c expansion of Trend IB is reached. The trend involves an increased rate in expansion relative to ionic radius along a and in the volume, coupled with a small increase in c and β . Little or no change is observed in b , which has reached a fully extended state. In triclinic feldspars, γ increases and α^* remains unchanged. This trend appears to be more variable than Trend I judging from Figures 1a and b. For ordered AlSi₃ and disordered

GaGe₃ and FeGe₃ frameworks, the trend is almost vertical on a b - c plot. For GaSi₃ frameworks the trend on a b - c plot has a negative slope due to the decreases in b on substituting Rb for K. Trend II is further distinguished from Trend I in that to a variable extent it dimensionally resembles changes produced by variations in T-site order (Fig. 1 and 2). This should be compared to the changes in substitution of anorthite in low albite which on Figure 1a follow a line from AL to AA up to about An₆₀, then straight down to An as b has apparently reached a limit for Al→Si substitution.

Trend III. The thermal expansion of K and Rb feldspars along Trend III involves a large expansion of a , a decrease of β and b , whereas c remains constant or increases. The trend is quite variable and may depend on the exact nature of Trend II and the room-temperature position of the feldspar with regard to Trend II. Compare sanidine, microcline, RbAlSi₃O₈ and RbGaSi₃O₈ on Figures 1a and 4b. For sanidine which has not reached its limit of compositional expansion for Trend I, an increase of temperature produces a decrease in both b and c (a trend similar to an inverse I). For microcline which has reached a Trend I compositional limit, an increase in temperature produces no change in c but a decrease in b (Fig. 5). For disordered RbGaSi₃O₈ which is well beyond the limit of Trend I, b decreases and c increases on raising T .

Analogous variables. Hazen (1977) compared the effects of composition, temperature and pressure on various simple structures and concluded that they were structurally analogous variables. This concept was applied to feldspars by Hazen (1976a; 1977) and Hazen and Prewitt (1977a). Our study shows clearly that composition, temperature and reduction in pressure are only analogous variables within the limits of Trend I. At and beyond Trend I, this relationship no longer holds true as is clearly demonstrated by microcline, the thermal expansion of which follows Trend III and the compositional expansion of which follows Trend II. Levien *et al.* (1980) also suggested that the atomic changes which occur in quartz with increasing temperature are not the reverse of those with increasing pressure. The concept of analogous variables is therefore of limited use and should be applied with caution (see Taylor, 1983).

Discussion

Structural aspects of feldspar expansion and transitions

Megaw (1974) demonstrated that many changes in the lattice parameters of feldspars at room temperature in response to changes in M-cation composition and size can be understood qualitatively as being due to tilts and rotations of regular rigid TO₄ tetrahedra. Furthermore, structural studies of high and low-albite at temperatures up to about 1000°C showed that T-O distances change

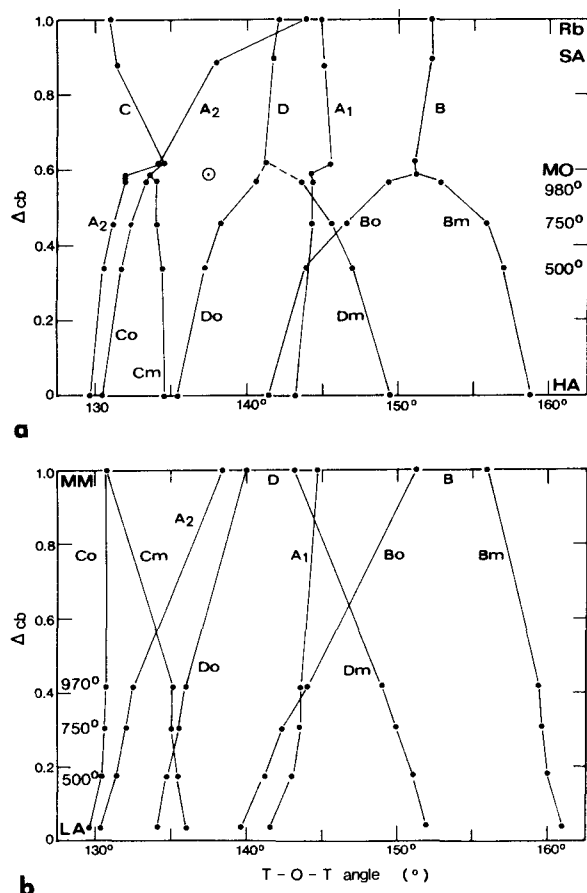


Fig. 6. Variation of T-O-T angles as a function of the positions of the feldspar on a b - c plot expressed as the fractional distance ($\Delta c/b$) along Trend I from analbite or low albite. (a) Data for disordered feldspars (upper figure) for high albite at room temperature (HA) and high temperature (500, 750 and 980°C) and monalbite at 980 and 1060°C (MO) from Winter et al. (1979) and for sanidine (SA) and RbAlSi₃O₈ (Rb) at room temperature from Smith (1974). One value (circled) for O_D for monalbite at 980° does not lie near the others and may be a misprint. (b) Data for ordered series (lower figure) for low albite at room temperature (LA) and high temperatures (500, 750 and 970°C) from Winter et al. (1977) and for microcline at room temperature (MM) average values from Smith (1974) and Dal Negro et al. (1978).

little with temperature (Prewitt et al., 1976; Winter et al., 1977; 1979), whereas T-O distances in forsterite (Hazen, 1976b) and in quartz (Levien et al., 1980) are almost constant for pressures up to 50 kbar or more. Taylor and Henderson (1968) and Taylor (1972) showed that discontinuities in the thermal expansion of certain tektosilicates can be related to phase transitions, and Hazen and Finger (1979) discussed phase transitions in analcite and other tektosilicates as a function of T and P in terms of polyhedral tilting.

Structure determinations of single-phase feldspars are available at room temperature for low and high albite, analbite, sanidine, disordered RbAlSi₃O₈ (Smith, 1974; Harlow and Brown, 1980), intermediate and maximum microclines (Dal Negro et al., 1978, 1980; Blasi et al., 1981), anorthoclase (De Pieri and Quarenzi, 1974; Harlow, 1982) and synthetic sanidine Or₅₈ (Fenn and Brown, 1977), and as a function of temperature for low and high albite, analbite and monalbite (Prewitt et al., 1976; Winter et al., 1977, 1979) and for K-rich sanidine (Ohashi and Finger, 1974, 1975). No structural data are as yet available at high pressures, nor for ordered feldspars of intermediate composition. It is thus not possible at present to interpret the whole range of feldspar expansion on a highly detailed structural basis, nor to formulate a computer model, as was done for cubic sodalites by Taylor and Henderson (1978) and Dempsey and Taylor (1980).

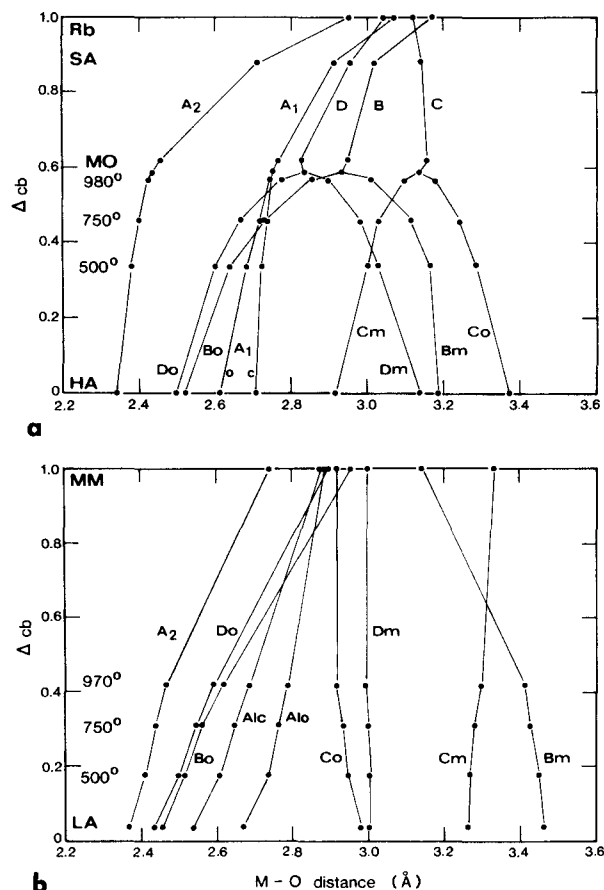


Fig. 7. Variation of the M-O distances as a function of the position of the feldspar on a b - c plot expressed as the fractional distance ($\Delta c/b$) along Trend I from analbite or low albite. (a) Data for disordered feldspars (upper figure) as in Fig. 6. (b) Data for ordered feldspars (lower figure) as in Fig. 6.

As changes in expansion behavior and phase transitions occur in tetrahedral framework structures without cavity cations (e.g., quartz, tridymite, cristobalite), it is possible that such transitions in other tectosilicates (e.g., leucite, sodalite, feldspar) are largely determined by the framework, although the cavity cations are likely to have a modifying effect. Whereas the thermal expansion of T–O bonds is effectively zero, this is not the case for Na–O, K–O and Ca–O bonds, which have relatively large mean coefficients of thermal expansion (Hazen and Prewitt, 1977b). As tectosilicates without cavity cations may have larger initial thermal expansion coefficients than most of those with cavity cations, it is possible that such cations actually hinder thermal expansion (see below and Dempsey and Taylor, 1980). Feldspar expansion is considered here as a compromise between the intrinsic freedom of the framework to twist or untwist and the effect thereon of the M-cation, resulting in the different trends observed.

The two components of Trend I expansion. Trend I is considered to be a result of two expansion components, both of which operate during Trend IA, whereas during Trend IB only one of them is operative. The *first expansion component* is a general expansion of the framework, which involves a large *a* expansion, much smaller coupled *b* and *c* expansions and a slight decrease in β . This expansion involves straightening or unwinding of the double crankshafts. The main interatomic changes in the ordered or disordered series involve the T–O–T angles (Fig. 6), and the M–O distances (Fig. 7). These changes are very similar whether the feldspar is monoclinic or triclinic, but in the latter case average T–O–T angles and M–O distances must be taken over pseudosymmetrically related o and m sites. In both series the average T–O–T angles around O_C and O_D change very little or decrease slightly on expansion, whereas those around O_B increase as do those around O_{A2} and to a smaller extent around O_{A1} . Expansion along *b* is controlled by the large angular increase in T–O–T around O_{A2} , counterbalanced by small decreases around O_C and O_D . Expansion along *c* is controlled by angular increases around O_{A1} and O_B , counterbalanced by the small decrease around O_D . Expansion along *a* is controlled mainly by the increase in the T– O_B –T angle linked to increases in M– O_{A2} and M– O_{A1} distances (Megaw, 1974; Ohashi and Finger, 1975; Henderson, 1979).

The *second expansion component* only operates in Trend IA between Or_0 and approximately Or_{35-45} corresponding to the approach to monoclinic symmetry in the disordered series (displacive transformation) or to the equivalent changes in the ordered series. The T–O–T angles and M–O distances become equivalent or approach each other (Fig. 6 and 7). α decreases (α^* increases) rapidly to 90° (or nearly so) and *b* and *c* increase more rapidly relative to *T* or to cation size than along Trend IB. This second component thus involves shearing of $3-4^\circ$ in the *b*–*c* plane and the extra coupled *b*–*c* expansion. It is this shearing which produces the very

asymmetric thermal expansion ellipsoid with α_3 negative (Willaime et al., 1974; Winter et al., 1977, 1979) in both low and high albite.

The M–O site and T–O–T angles. In low albite, sodium seems to occupy a single site with very high anisotropic thermal motion (Quarenì and Taylor, 1971; Winter et al., 1977; Harlow and Brown, 1980; Harlow, 1982), and this motion extrapolates to zero at 0 K. In high albite and analbite, sodium shows spatial disorder in response to differences in local Al,Si disorder, but its exact nature is not yet resolved (Winter et al., 1979; Harlow, 1982). In the following discussion, we assume a single site for sodium.

The rapid changes with temperature in the Na–O environment for high albite and analbite are shown by Prewitt et al. (1976, Fig. 5) and by Winter et al. (1979, Figs. 9a and b) and Na–O distances are given in Figure 7a; see also Harlow (1982). At 25°C the Na coordination polyhedron is extremely irregular, and it is difficult to decide which oxygens should be included and which excluded from the Na–O coordination polyhedron. Sodium has five close neighbors at an average Na–O distance of 2.53\AA . Differences between o and m distances are very great; they are 0.67\AA for O_B , 0.46\AA for O_C and 0.64\AA for O_D at room temperature for analbite. These distances approach each other very rapidly with increasing *T*; the shorter distances increase slightly faster than the longer ones decrease, and they become equal near 1000°C for monalbite (and at lower temperatures for anorthoclase with increasing Or-content, Harlow, 1982) or nearly so at over 1000°C for high albite (Fig. 7a and Prewitt et al., 1976; Winter et al., 1979). The closest-neighbor coordination for monalbite is 7-fold at 980°C with an average distance of 2.78\AA , the Na– O_{A2} distance being extremely short (2.436\AA). The two Na– O_C distances are 3.133\AA .

The T–O–T bond angles and M–O distances for slightly triclinic anorthoclase at room temperature (De Pieri and Quarenì, 1974) are very close to those of high albite near 850°C . In high sanidine and disordered $\text{RbAlSi}_3\text{O}_8$ (Fig. 6a and Smith, 1974) T–O–T angles are almost the same as for monalbite except for T– O_{A2} –T, which is markedly larger. M–O distances also increase from those of monalbite (except for M– O_C) and this is especially the case for M– O_{A2} , which for Rb is nearly the same as the other Rb–O distances (Fig. 7a). In high sanidine and $\text{RbAlSi}_3\text{O}_8$ the M-cation is in 9-fold coordination with average distances of 2.97\AA and 3.08\AA respectively. The thermal expansion of the K–O polyhedron was determined by Ohashi and Finger (1974, 1975). The mean M–O distance increases from 2.967\AA at 25°C to 3.034\AA at 800°C , giving a mean expansion from 25 to 800°C of 2.3% or of 2.8% from 25 to 1000°C (estimated).

Very similar changes in T–O–T angles and M–O distances occur in ordered low albite with increasing *T* (Fig. 6b, 7b and Winter et al., 1977) and K substitution. In ordered feldspars monoclinic symmetry is not reached, but the T–O–T angles for microcline also converge and

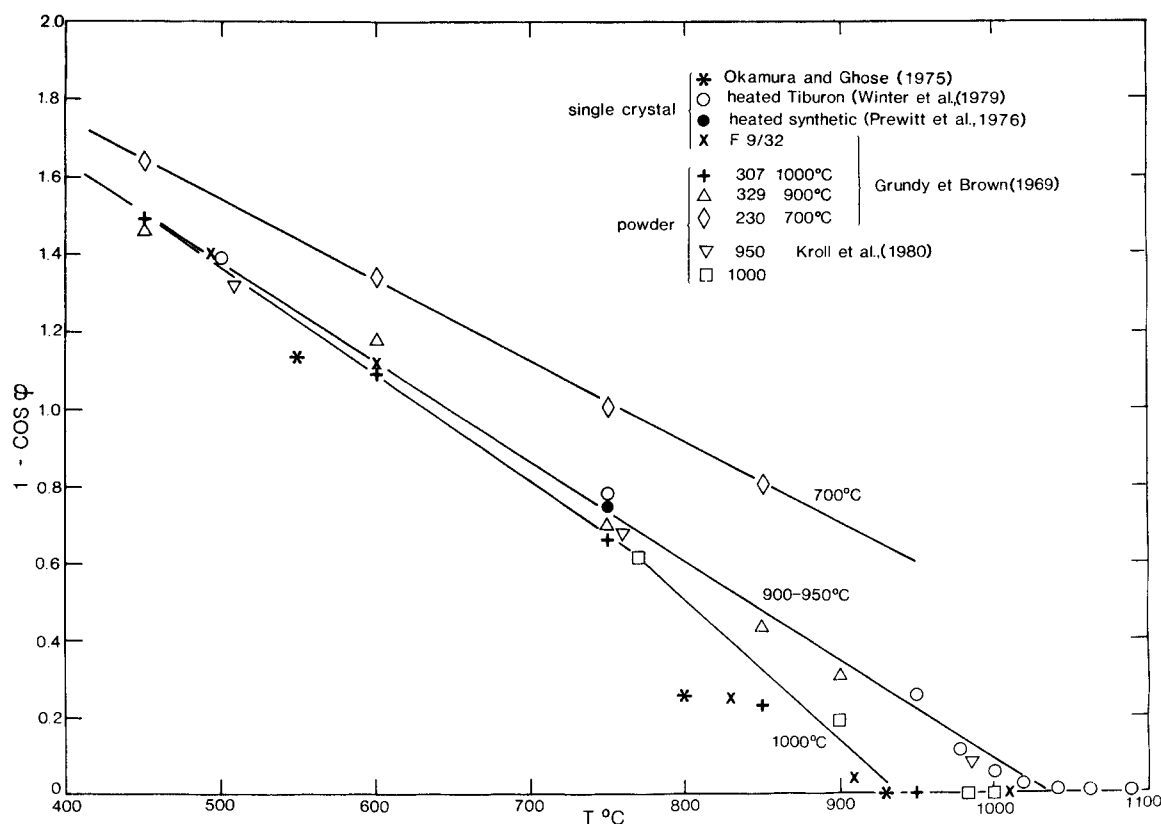


Fig. 8. Plot of $1 - \cos \phi$ against temperature for some high albites and for analbite-monalbite. The precision for the single-crystal data is not affected by the smallness of the deviation from monoclinic geometry; this is not the case for the powder data, which become imprecise as $\phi \rightarrow 0^\circ$ due to peak overlap. Single-crystal data show clearly that analbite differs from high albite above about 750°C . Temperatures on lines or in legend (Grundy and Brown, 1969; Kroll et al., 1980) are synthesis temperatures.

are very similar to those of high sanidine at room temperature (Fig. 6b and Smith, 1974). This is also the case for K-O distances with the exception of the O_C distances, which diverge (see Fig. 7b and later section on the microcline phase transition). In low albite at room temperature Na has five closest neighbors with an average Na-O distance of 2.49\AA . At 970°C the coordination is either 5-fold (average Na-O distance of 2.63\AA) or 7-fold (average Na-O distance of 2.72\AA). In microcline at room temperature the K coordination is 7-fold (average K-O distance of 2.89\AA) or 8-fold (average K-O distance of 2.92\AA). If one includes the distant O_C atom, the coordination is 9-fold with the same average K-O distance as in high sanidine. It is interesting that a change of Al,Si order from high sanidine (Smith, 1974) to maximum microcline (Dal Negro et al., 1978; 1980) has little effect on average T-O-T angles and K-O distances, except that the K- O_C distances diverge in microcline.

The nearest-neighbor distances in analbite and high albite thus change rapidly and markedly with temperature and the polyhedra become more symmetrical as they expand. At 25°C Na has five closest neighbors with an average Na-O distance of 2.53\AA , whereas at 980°C the

average distance of the *same* bonds is 2.63\AA . This corresponds to a 7.1% increase over almost 1000°C . A similar expansion occurs for low albite between room temperature and 970°C . These expansions of the closest Na-O distances in albite are three times larger than those quoted for Na-O in inorganic compounds by Hazen and Prewitt (1977b), and greater than that for Na-Cl in halite, a much softer structure, and twice the expansion of K-O in sanidine. We suggest that the Na-O expansion is abnormally large and therefore we agree with Dempsey and Taylor (1980), that the driving force for expansion lies in the untwisting of the framework, rather than in the M-O bonds (Harlow, 1982) which may in fact restrain expansion.

The thermodynamic nature of the triclinic→monoclinic transition

The transition in analbite and anorthoclase is of great importance in understanding the crystallographic properties of feldspars, but its exact thermodynamic nature is still uncertain. The transition occurs on rapid heating and does not appear to involve hysteresis on the basis of single-crystal X-ray investigations (Grundy et al., 1967;

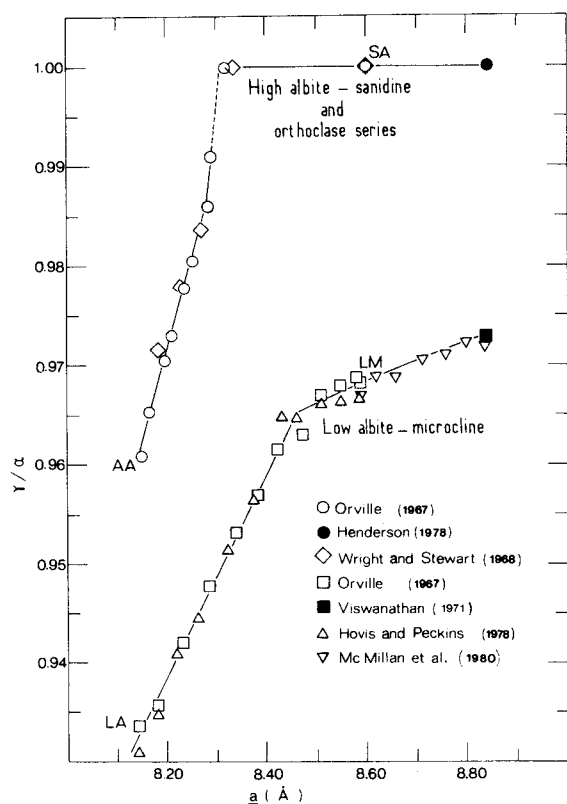


Fig. 9. Plot of the ratio of the cell angle ratio γ/α against a for the ordered and disordered series (symbols as in Fig. 3, LA etc. as in Fig. 1).

Okamura and Ghose, 1975), though MacKenzie (1952) noted that optically visible twins in anorthoclases did not reappear at the same temperature on cooling as they disappeared on heating, but such optical observations, however, are not sensitive near the transition temperature. Structurally, the transition is displacive (Prewitt et al., 1976; Winter et al., 1979; Harlow, 1982), but bulk properties must be used to determine its thermodynamic nature. There is no measurable thermal effect near the transition temperature as shown by DTA or by differential scanning calorimetry (T. R. Schneider, pers. comm., 1957; A. B. Thompson, pers. comm., 1981). In addition, there seems to be no discontinuity nor rapid change in the volume at the transition temperature, though there is probably a change in slope and hence a small discontinuity in the thermal expansion coefficient, α_p (Henderson, 1979; Kroll, pers. comm.). The transition is thus not of clear λ -type, as suggested by Helgeson et al. (1978) on the basis of what is to us an incorrect interpretation of calorimetric data on long-term heated albite from Holm and Kleppa (1968). Formally, it may closely approach a second-order transition, though there are at present too few data above the transition temperature to be sure.

In any case, it is not clear at which temperature, if any, the transition in a given feldspar begins on raising the temperature. Departures from monoclinic symmetry can be expressed by the obliquity, $\phi = b/b^*$, or the cell angles, the major departure being in α or α^* ; γ and γ^* are very nearly 90° (Fig. 2). Recently, attempts have been made to find a suitable function which varies linearly with temperature. Kroll et al. (1980) and Harlow (1982) used $\cos^2\alpha$ and Hovis (1980) used $1 - \cos\phi$. Such plots imply that the displacive transition is truly second order and has no temperature range, the departure from monoclinic symmetry decreasing smoothly as the symmetry change is approached. The lattice angles of low, intermediate and high albites change smoothly with temperature and order (Grundy and Brown, 1969, Fig. 2) and plots of $\cos^2\alpha$ or $1 - \cos\phi$ vary linearly with temperature up to about 1000°C (Fig. 8). The lattice angles of high albite and analbite are virtually indistinguishable at each temperature from room temperature up to about 750°C (Grundy and Brown, 1969; Kroll et al., 1980). This is no longer true above about 750°C for above this temperature analbite more rapidly approaches and attains monoclinic symmetry (Fig. 8; see also Kroll et al., 1980, Fig. 6); for very high albite $1 - \cos\phi$ varies linearly up to about 1000°C (Fig. 8), but monoclinic symmetry is not attained (Winter et al., 1979).

We thus suggest that the departure from pure second-order behavior is limited to the more rapid changes in the lattice angles or obliquity of analbite over about 200°C below the temperature of the symmetry change. Similar, slightly more rapid changes may also possibly occur on change of composition at room temperature in Orville's (1967) synthetic Na-K feldspars, where the more rapid change occurs over about 10% Or (Fig. 9). When these points are plotted on a temperature-composition diagram for disordered Na-K feldspars, they fall on a line parallel to and about 200°C below the line for the displacive symmetry change. The width of the zone between the two lines shows the possible departure from pure second-order behavior towards a very weak smeared λ -type transition.

To a first approximation the symmetry change occurs at very similar values of the parameters a , b , c , β and V as a function of T , P and composition (Hazen, 1976a; 1977). This fact confirms the suggestion that it is the framework which is more important than the kind of M-cation in determining the expansion and the symmetry change. This is borne out by the similar behavior of the above lattice parameters in the ordered series in which no symmetry change occurs (McMillan et al., 1980, Fig. 2)—a sharp drop in the expansion rate of b , c , and V with composition occurs near Or₄₀. The rates of change of the α and γ angles with Or-content decrease beyond Or₄₀ and are nearly constant beyond Or₇₀ (McMillan et al., 1980; Hovis, 1980) possibly suggesting a slight smearing out of the analogous changes in the ordered series (compare Fig. 9).

The microcline phase transition and change of trends

In the beginning of our work, we examined the possibility that the inferred phase transition in microcline (Openshaw et al., 1976; 1979a,b; Wyncke et al., 1981) might be the analogue in ordered feldspars of the monoclinic→triclinic inversion in highly disordered Na-rich feldspars (Thompson and Hovis, 1979). As shown above this is not the case, as changes similar to those of the triclinic→monoclinic transition occur in ordered AlSi_3 feldspar near Or_{40} at room temperature. In this section we discuss a possible interpretation of the transition. No new data are presented.

The phase transition observed in an ion-exchanged microcline (specimen 71104) was characterized by hysteresis in the specific heat, the unit cell parameters and in one far infrared absorption band (Openshaw et al., 1979a,b; Wyncke et al., 1981). One form of microcline denoted the A-form was obtained on cooling below 245 K. It slowly inverted to the normal form, denoted the B-form, at temperatures above 300 K ($27 \pm 10^\circ\text{C}$). Similar effects were observed in the far infrared spectrum of a natural microcline, and we believe that the transition may be characteristic of all highly ordered microclines. The transition was found neither in sanidine, nor in high and low albite (Openshaw et al., 1976).

The room-temperature b and c parameters of microcline sample 71104 before and after cooling in liquid nitrogen are shown in Figure 5. On reheating, the A form is temporarily preserved and has very slightly larger a and c and a considerably larger b parameter (12.976\AA). The lattice angles do not change. With time, the A-form relaxes to the more stable B-form, b contracting to about 12.965\AA . At low temperature the A-form lies on a continuation of Trend I, whereas the B-form has the same b parameter as that of the K–Rb series (Trend II).

The changes in the T–O–T angles on heating $\text{NaAlSi}_3\text{O}_8$ or on substitution of K for Na are extremely similar in ordered and disordered alkali feldspars, except that the o and m angles are not quite identical in ordered K-feldspar (Fig. 6). The M–O distances behave similarly, the main difference being the convergence of the two M– O_C distances from analbite to sanidine and their divergence from low albite to low microcline (Fig. 7). In order for the K– O_Cm distance in microcline to decrease, it would be necessary to reduce T– O_Cm –T below 131°C , the smallest angular value in the feldspars; alternatively it would be necessary to rotate the T_2O tetrahedron, thus increasing the K– $\text{O}_{\text{A}2}$ distance and hence increasing b and perhaps causing an inversion to the A-form.

We suggest that the microcline phase transition mainly involves movements of the O_C atoms and in particular O_Cm . In the A-form, K– O_Cm may be smaller than in the B-form (smaller than the value in low albite at high temperature). The transition from the B to the A-form is

rapid, whereas the reverse is slow and subject to hysteresis, and intermediate states can be obtained. Thus there is probably some steric hindrance for the reverse transition, which could involve other oxygen atoms such as $\text{O}_{\text{A}2}$, which is strongly bonded to K. This would be consistent with the far infrared data (Wyncke et al., 1981), which indicate that the transition affects one band probably associated with both T–O–T bending and K–O stretching motions (Stengel, 1977). Furthermore, because of the hysteresis the exact thermodynamic nature of the phase change is not known, though some sort of smeared (martensitic-type ?) transition seems the most probable (Openshaw et al. 1979b) involving hybrid states, which may consist of unit cell-scale mixtures of the A and B-forms. Though the transition is not a large effect, it may be a general property of feldspars which have reached the end of Trend I expansion. We tentatively predict its occurrence in other such feldspars, e.g., ordered KGaSi_3O_8 and disordered KGaSi_3O_8 , KGaGe_3O_8 and KFeGe_3O_8 and possibly disordered $\text{RbAlSi}_3\text{O}_8$ and $\text{RbAlGe}_3\text{O}_8$ on the basis of the similar behavior of the b – c parameters (see Fig. 1a and 1b).

Conclusions

1. Because of their low symmetry the expansion of the alkali feldspars is complex, but it can be broken down into different trends; these trends are characterized by the coupling or uncoupling of certain cell parameters. In many K or Rb-rich feldspars the b parameter has reached a limit of expansion. This precludes a simple representation of alkali feldspar expansion by polynomials.

2. Composition, temperature and pressure behave approximately as analogous variables (Hazen, 1977a) only in the expansion of Ab-rich feldspars along Trend I. For other feldspars their effects may be quite different.

3. The very high thermal expansion of the short Na–O bonds in low albite and analbite and the very similar values of the cell parameters at which the triclinic→monoclinic transition occurs as a function of P , T and composition in Ab-rich feldspars suggest that alkali feldspar expansion is determined largely by the framework and not by the M–O bonds.

4. The displacive monoclinic→triclinic phase transition in Na-rich feldspars may be almost purely second order, and should not be represented on phase diagrams by a binary loop.

5. The phase transition in microcline near room-temperature is probably a smeared (martensitic-type) transition and it may occur in other alkali feldspars. It probably involves movement of the O_C and $\text{O}_{\text{A}2}$ oxygens. It is suggested that such a transition may possibly occur in other K-rich feldspars with ordered GaSi_3 and disordered GaSi_3 , GaGe_3 or FeGe_3 frameworks, and in $\text{PbAl}_2\text{Si}_2\text{O}_8$, which occur at positions similar to microcline in framework expansion. Disordered $\text{RbAlSi}_3\text{O}_8$ and $\text{RbAlGe}_3\text{O}_8$ may also show such a transition.

6. The existence of the microcline phase transition and the complexity of the variation of *b* and *c* with Or content in ordered Or-rich AlSi_3 feldspars suggest that use of the *b*-*c* plot to determine Al,Si site occupancies in microclines may not be simple and unambiguous.

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