

The thermal expansion behaviour of the framework silicates

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SUMMARY. This paper gives a revision and expansion of an earlier interpretation of the thermal expansion behaviour of the framework silicates. The partially-collapsed and ideal fully-expanded structures of quartz, cristobalite, and sodalite are characterized by the geometric relationship between the angle of rotation of their tetrahedra from the ideal fully-expanded state, their cell parameters, and the length of the tetrahedron edge. Their thermal expansion behaviour is interpreted as due mainly to the effect of the rotation of the tetrahedra towards the fully-expanded state modified by anisotropic thermal motion of the framework oxygens and distortion of the tetrahedra from a regular form. With the leucite and sodalite groups the significance of the interframework cations is discussed.

TAYLOR (1968) and Taylor and Henderson (1968) developed a simple interpretation of the thermal expansion behaviour of the sodalite and leucite groups of minerals, but subsequent detailed examination of the structural changes in quartz and cristobalite on heating has indicated a need for revision and clarification.

When a crystalline substance is heated there is an increase in the amplitude of the bending and stretching modes of the different combinations of the atoms. At the same time there is a contribution to the total crystal or bond energy, which affects the interatomic distances (Drechsler and Nicholas, 1967). In addition, there is often a change in the detailed geometry, as evidenced by structural transitions, which are possibly a result of the increase in the amplitude of the bending and stretching modes. The possibility of anharmonic thermal vibrations in molecules complicates any simple model for thermal expansion behaviour because they lead to apparent shortening of the bonds (Cruikshank, 1956). The effect of heat on the total crystal or bond energy is expected to show a relationship between the degree of change in interatomic distance and the bond type; for example, one may contrast the high thermal expansions of the largely-ionic alkali halides with the low expansion of the covalent diamond crystal.

The framework silicates, as their name implies, consist of linked SiO_4 , or SiO_4 and AlO_4 , tetrahedra with all their oxygens shared. By this means a three-dimensional framework is built with spaces that can be occupied by other ions or molecules. The framework, while strong, is not rigid, for there are no forces holding it fully expanded. The general observation is, therefore, that the structures of framework silicates at room temperature are partially collapsed and distorted versions of the ideal fully-expanded structures. The structural collapse is brought about by rotation and bending of the framework tetrahedra about their shared oxygens. By rotation it is not implied that the tetrahedra rotate continuously but merely that they turn through a

small angle sufficient to bring the structure to a particular partially-collapsed state. The relationship between ideal fully-expanded states and partially-collapsed states is shown in fig. 1. Rotation of tetrahedra causing partial collapse of the structure has been reported by Pauling (1930) for sodalite, Nieuwenkamp (1935) for α -cristobalite, and Hahn and Buerger (1955) for nepheline.

It has been deduced from thermal expansion studies of the sodalite and leucite groups of minerals (Taylor, 1968; Taylor and Henderson, 1968) that the high initial rate of thermal expansion is due to the progressive rotation of the tetrahedra from their orientation in the partially-collapsed state rather than to the expansion of the (Si, Al)-O bonds of the framework. A similar interpretation was developed simultaneously by Megaw (1968) to explain the thermal expansion behaviour of the alkali niobates, which have frameworks consisting of linked NbO_6 octahedra. According to the interpretation of Taylor (1968) and Taylor and Henderson (1968) it was expected that the thermal expansion would continue at a high rate until the framework achieved the ideal fully-expanded state and then fall to a much lower rate consistent with the expansion of the (Si, Al)-O bonds. Abrupt changes in the rate of expansion have been observed for quartz (Jay, 1933; Mayer, 1960), cristobalite (Johnson and Andrews, 1956), nosean and haüyne (Taylor, 1968), and the leucite group of minerals (Taylor and Henderson, 1968). In a few cases the structures of these minerals are known at room temperature and high temperatures so that ideas relating to their thermal expansion behaviour can be tested. In this paper it is proposed to examine the thermal expansion behaviour of the framework silicates in relation to their structures and to compare the predicted behaviour with that observed.

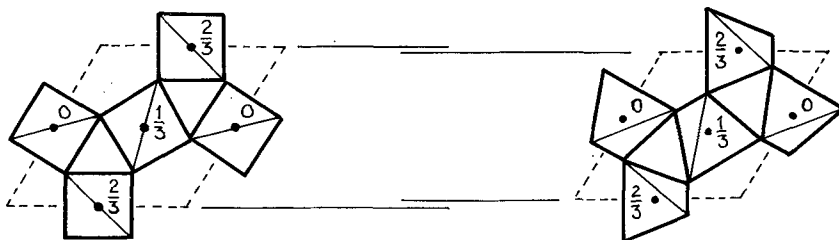
Geometric relations

For some framework silicates it has been found that the ideal fully-expanded state can be achieved by rotation of each tetrahedron about one of its $\bar{4}$ -fold axes. The relationship between the angle of rotation, ϕ , of the tetrahedron from its position in the fully-expanded structure, the cell parameters, and the length of the tetrahedron edge, X , has been determined for some framework silicates:

$$\begin{array}{l} \text{Quartz} \quad \left\{ \begin{array}{l} a = 1.225 X \cos \phi + 0.7071 X \\ c = 2.121 X \cos \phi \end{array} \right. \\ \text{Cristobalite} \quad \left\{ \begin{array}{l} a = 1.414 X \cos \phi + 1.414 X \\ c = 2.828 X \cos \phi \end{array} \right. \\ \text{Sodalite} \quad a = 2 X \cos \phi + 1.414 X \end{array}$$

For cristobalite the parameters are those for the β -cristobalite unit cell. The partially-collapsed and the ideal fully-expanded structures of quartz, cristobalite, and sodalite are shown schematically in fig. 1. The Si-O-Si bond angles for the fully-expanded structures of quartz, cristobalite, and sodalite are 156° , 180° , and 160.5° respectively.

The assumptions made in calculating the above equations can be illustrated by taking quartz as an example. The assumptions for quartz are: that the unit cell is hexagonal/trigonal, that the tetrahedra are regular, that for β -quartz the idealized structure is correct, and that the rotation of the tetrahedra about $\bar{4}$ is co-operative. The equations are then derived by simple geometric relations. Unfortunately, it is



QUARTZ

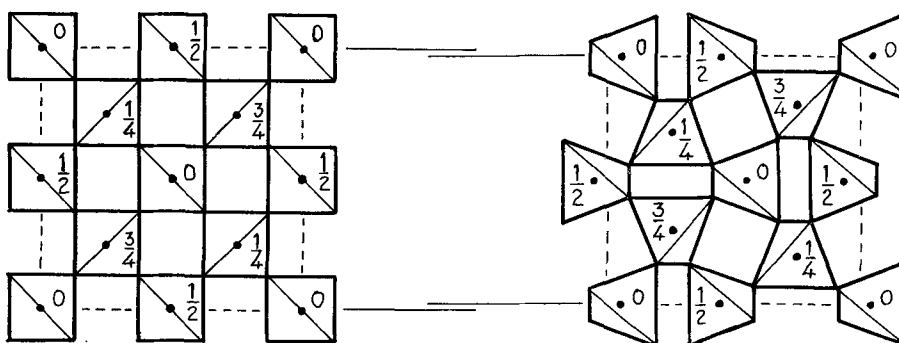
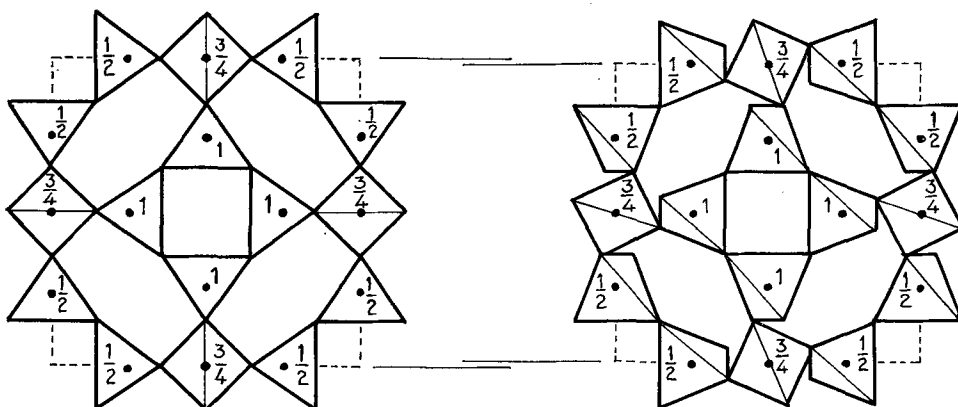
CRISTOBALITE
(β -cristobalite unit cell)SODALITE GROUP
(upper half of unit cell)

FIG. 1. Schematic diagrams for the partially-collapsed (right) and fully-expanded structures (left) of quartz, cristobalite, and sodalite, showing only the framework tetrahedra.

known that the tetrahedra are not quite regular for α - or β -quartz (Young, 1962), which in turn means that the idealized β -structure is not strictly accurate. Nevertheless, it is desirable to examine the usefulness of these simplified geometric relationships despite their imperfections.

Nepheline, kalsilite, and leucite have a more complicated geometrical relationship between the rotation and tilting of the tetrahedra and their cell parameters. Hahn and Buerger (1955) show the relationship between the partially-collapsed state of nepheline and the ideal fully-expanded structure. Megaw (1970) has deduced the geometric relations for the tilting of the tetrahedra in the feldspars.

Structural data for the framework silicates

Cell parameters, bond lengths, and angles for some framework silicates relevant to the present paper are given in table I with values of ϕ calculated from the atomic co-ordinates. When the data were not present in the original reference they were calculated from the atomic coordinates given. It is important to stress that the bond lengths and angles quoted in table I are averaged values, often of statistically significant different individual values.

The striking feature in table I is the apparent shortening of the Si–O bond in quartz with increasing temperature, and the much lower value of the mean (Si, Al)–O bond length of leucite at 635 °C compared with that of pollucite at room temperature. Cruickshank (1956) showed how anisotropic motion of atoms in molecules gives rise to an apparent shortening of the bond length, which for idealized and small amplitudes of thermal motion is given by $y = s^2/2r$, where r is the observed bond length, s^2 the mean square amplitude of oscillation of one atom relative to the other, and y is the apparent shortening.

For the purpose of the present paper this simple equation will be used to give a first approximation in allowing for the effect of thermal motion. Distinct anisotropic thermal motion of the oxygen atoms in SiO₄ and AlO₄ tetrahedra has been reported in α -quartz (Young and Post, 1962; Smith and Alexander, 1963), β -quartz (Young 1962), β -leucite (Peacor, 1968), nepheline (Hahn and Buerger, 1955), and high tridymite (Dollase, 1967). In each case the amplitude of thermal motion was greatest at right angles to the (Si, Al)–O–(Al, Si) bonding plane and least along the direction of the (Si, Al)–(Al, Si) join. This is as one would predict since it is easier to bend an (Si, Al)–O–(Al, Si) group about the oxygen than to stretch and compress an (Si, Al)–O bond.

Some caution is required, however, in interpreting all apparently short bonds as being due to thermal vibrations. Increasing the (Si, Al)–O–(Al, Si) angle increases the degree of π -bond order and so decreases the (Si, Al)–O distance (Cruickshank 1961; Brown, Gibbs, and Ribbe, 1969). This effect might be seen in cristobalite, where in the ideal fully-expanded structure the Si–O–Si angle would be 180° compared with 147° in the room-temperature structure. Liebau (1961) considers large (Si, Al)–O–(Al, Si) angles unlikely and showed that in some cases they were due to poor refinement. Brown, Gibbs, and Ribbe (1969) showed that the interframework cations may have an effect on the bond lengths of adjacent atoms in the framework.

TABLE I. Structural data for framework silicates

Mineral, reference, and temperature	Cell parameters			O—O	(Si,Al)— O—(Al, Si)	ϕ
	<i>a</i>	<i>b</i>	<i>c</i>			
α -Cristobalite				2.598 Å	146.8°	22.5°
α -Quartz	Dollase (1965)	4.978 Å	6.948 Å	1.605 Å	144.0°	15.9°
	{ Smith and Alexander (1963)	4.913	5.404	1.607	143.9°	16.2°
	{ Young and Post (1962)	4.913	5.404	1.607	148.6°	11.8°
	{ Young (1962)	4.954	5.430	1.597	2.610	
	{ 450 °C	4.965	5.438	1.594	2.600	150.0°
	{ 520 °C	4.971	5.443	1.594	2.602	150.2°
	{ 545 °C	4.975	5.445	1.594	2.603	150.4°
	{ 560 °C	4.982	5.448	1.592	2.600	151.2°
	{ 570 °C	4.998	5.458	1.590	2.597	153.0°
	{ 580 °C	4.998	5.458	1.590	2.597	152.8°
β -Quartz	Young (1962)	4.998	5.458	1.590	152.8°	0°
Tridymite	Dollase (1967)	4.998	5.457	1.590	152.9°	2.6°
		220 °C* <i>a</i> = 8.74, <i>b</i> = 5.04 <i>c</i> = 8.24	1.555	n.d.	171.7°	n.d.
Leucite	Peacor (1968)		<i>a</i>	1.618 Si, Al	145.4°	n.d.
	Newnham (1967)	13.52	13.682	1.638 Si, Al	145.1°	n.d.
	Beger (1969)	13.69	13.69	1.643 Si, Al	n.d.	n.d.
Sodalite	Löns and Schulz (1967)	8.870	8.870	1.628 Si	138.3°	23.7°
Hydrosodalite	Bukin and Makarov (1967)	8.878	8.878	1.728 Al	2.672 Å	22.4°
				1.600 Si	2.612 Å	23.8°
				1.756 Al	2.867 Å	21.9°

* indicates 3-D refinement; remaining structures refined from 2-D data.
Cell parameters for quartz above 27 °C are based on data of Jay (1933). n.d. = not determined.

In the following sections the thermal expansion behaviour of certain framework silicates will be examined and interpreted. The framework silicates of main interest are quartz, cristobalite, and the leucite and sodalite groups, for which representative volume thermal expansion curves are shown in fig. 2.

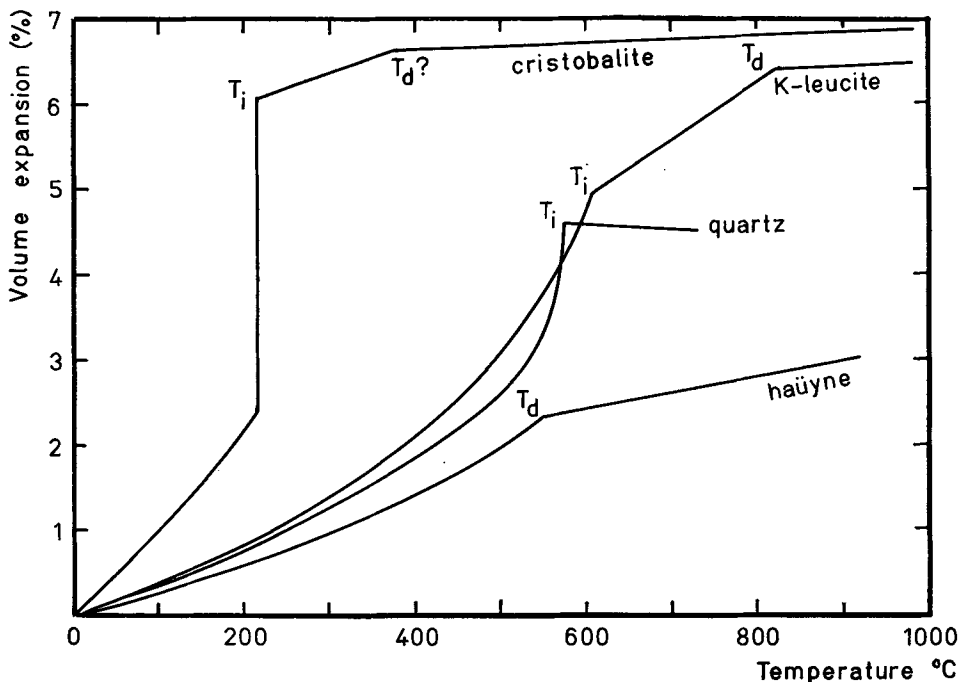


FIG. 2. Thermal expansion curves for framework silicates: cristobalite (Johnson and Andrews, 1956), K-leucite (Taylor and Henderson, 1968), quartz (Jay, 1933; Mayer, 1960), and häüyne (Taylor, 1968). T_i is the inversion temperature and T_d the discontinuity temperature after Taylor (1968).

Quartz shows an increasing rate of thermal expansion to the inversion, where there is an abrupt change from expansion to contraction on further heating. Assuming that β -quartz has the idealized fully-expanded structure shown in fig. 1, the O-O bond length, X , can be calculated using the above equations by substituting the cell parameters of β -quartz after Jay (1933) and setting ϕ to zero. The result is 2.586 Å from the equation for a and 2.573 Å, from that for c . This difference in the O-O distances with the a and c parameters means that the cell parameters of β -quartz are such that the SiO_4 tetrahedron cannot be regular. This is borne out by the observations of Young (1962). From the O-O bond lengths calculated above the mean Si-O distance is calculated to be 1.580 Å, which is in fair agreement with the value 1.590 Å observed for β -quartz by Young (1962) and given in table I. If one now applies Cruickshank's simple correction (1956) for the anisotropic thermal motion of the oxygen atom, reported by Young (1962), to the bond lengths for quartz in table I, one obtains Si-O bond lengths of 1.613 Å at 25 °C, 1.610 to 1.611 Å between 470 and 570 °C, and

1.615 to 1.616 Å at 600 and 650 °C. The most accurate determinations are those at 25 and 600 °C since the other structural determinations were based on *hko* and *hhl* intensity data only (Young, 1962). Thus, after correcting for simple anisotropic thermal motion of the oxygen atom the Si–O bond length probably shows a real increase from 1.613 to 1.616 Å between 25 and 600 °C (such a difference is equal to one standard deviation quoted by Young (1962) and so may not be significant). In reality, however, one observes an apparent contraction of the Si–O bond length of 0.017 Å, which should give rise to a contraction in the volume of approximately 3 %, and yet the cell volume increases by 4.6 % (fig. 2). The distinct expansion of quartz up to the inversion temperature is due to the rotation of the tetrahedra, albeit slightly distorted and modified by thermal motion, from the partially-collapsed state to the fully-expanded state. The situation, though, is even more complex than the latter statement implies because the rotation of the tetrahedra to the fully-expanded state does not appear to be a continuous process. If one plots the observed values of ϕ given in table I against temperature then the tetrahedron turns gradually at a slightly increasing rate until at 560 to 570 °C the value of ϕ is approximately 8°; it then suddenly falls to zero at 575 °C (fig. 3). Mayer (1960) found a discontinuity in the cell parameters immediately prior to the inversion, calculated to be 0.009 Å for Δa and 0.006 Å for Δc . Using the above equations and assuming X to be 2.600 Å then the mean rotation angle, ϕ , immediately before the inversion is calculated to be 8.6°. This is very close to that determined above from the data of Young (1962). For all Young's refinements the space group of α -quartz, $P3_221$, was used, which in part explains the non-zero values of ϕ for the 580 and 650 °C data.

Above the inversion there is a slight but measurable contraction in the cell volume. This apparently anomalous behaviour is interpreted as being due to an increase in the amplitude of the anisotropic motion of the oxygen atoms on increasing the temperature, resulting in a further apparent shortening of the Si–O distance. Since the structure is already in the fully-expanded state, in which the tetrahedra can no longer rotate, the observed result is a real contraction in the cell volume.

The phenomena of the thermal expansion of quartz can, therefore, be attributed to a combination of a major effect, the rotation of the framework tetrahedra, and a minor effect, the increasing amplitude of thermal motion of the oxygen atoms. Taking these two factors into account and using the above equations it should be possible to calculate a thermal expansion curve by successive approximations until it is similar to the observed curve. This has been done using as reference points the anisotropic motions of the oxygen atoms at 25 and 600 °C and the observed angle of rotation of the tetrahedra at room temperature based on the data of Young (1962), Young and Post (1962), and Smith and Alexander (1963). It is of interest to comment here that if a linear rate of rotation of the tetrahedra is assumed then the rate of expansion will decrease rather than increase with increasing temperature. Fig. 3 shows a calculated thermal expansion curve similar to the observed curve. The changes in the rotation angle of the tetrahedra and the thermal motion of the oxygens with temperature on which the calculated curve was based are also shown. Up to the inversion temperature the rotation of the tetrahedra is the predominant factor and thereafter the thermal

motion of the oxygens. The discrepancy between the calculated and observed percentage volume expansions at the inversion is due to the distortion of the tetrahedra,

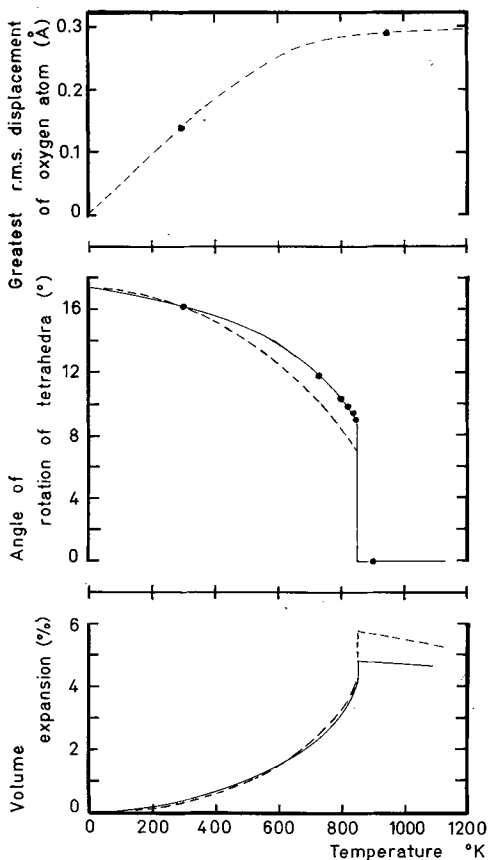


FIG. 3. Thermal vibration, tetrahedral rotation, and thermal expansion of quartz. The points indicated were based on the data of Young (1962), Young and Post (1962), and Smith and Alexander (1963). The broken curves in the upper and middle part of the figure were those finally obtained by successive approximations, which gave the calculated expansion curve shown by a broken line in the lower part of the figure. The solid expansion curve was drawn from the data of Jay (1933) and Mayer (1960).

plane using Cruickshank's equation (1956) is 0.46 Å for β -cristobalite at the inversion. This value may be compared with the greatest amplitude of vibration of 0.40 Å for the oxygen atoms in leucite at 635 °C (Peacor, 1968) and high tridymite at 220 °C (Dollase, 1967). It is also close to the 0.4 Å radius circle about the Si-Si join on which Nieuwenkamp (1937) would have the oxygen atom rotating continuously or statistically distributed. It is difficult to know when cristobalite achieves the ideal fully

which was neglected in the calculations.

The slope of the calculated curve above the inversion would be closer to that of the observed curve if slight expansion of the Si-O bond had been included. In fitting a calculated expansion curve for quartz it is found that the observed expansion curve requires the abrupt change in the angle of rotation of the tetrahedra at the inversion. Fig. 3 also extrapolates the rotation angle to 0 °K where the thermal vibration of the atoms is assumed to be zero.

Cristobalite. The O-O bond length, X , for β -cristobalite at the inversion has been calculated to be 2.522 Å from the above equations by setting ϕ to zero and substituting the cell parameters of Johnson and Andrews (1956) for β -cristobalite. For a regular tetrahedron this gives an Si-O bond length of 1.544 Å. This exceptionally short Si-O distance has led some authors to suggest that the idealized structure of β -cristobalite is not in fact possible.

There may be nothing more unusual about the short Si-O bond length in β -cristobalite than high anisotropic thermal motions of the oxygen atoms, possibly combined with further shortening caused by increased π -bond order due to the large Si-O-Si bond angle. An estimate of the oscillation of the oxygen atom and hence the thermal vibration perpendicular to the Si-O-Si bonding

expanded state since the inversion to cubic symmetry need not necessarily mean that the ideal fully-expanded state has been achieved. Single-crystal studies do appear to indicate that the ideal fully-expanded state is achieved at the inversion, but the relatively high thermal expansion rate above the inversion and up to 380 °C suggests that it has not been achieved and that the tetrahedra are still rotating towards the fully-expanded state. Once the ideal fully-expanded state is achieved one might expect the volume to contract as is observed with quartz, and for the same reasons. Instead, Johnson and Andrews (1956) observed a slight expansion at high temperatures. This discrepancy between the expected and the observed may be due to a limit on the degree of anisotropy of the thermal motions of the oxygens or even a trend to isotropic vibration allowing the expansion of the Si–O bond to become significant, or may be due to impurities in the structure.

The leucite group of minerals. The thermal expansion curve for synthetic leucite, fig. 2, shows two changes of rate. The first occurs at the tetragonal–cubic inversion. From the relatively high rate of expansion above the inversion it was concluded that leucite did not achieve the ideal fully-expanded state at the inversion (Taylor and Henderson, 1968) but at the temperature of the second change of rate, T_d . The structural study by Peacor (1968) of a leucite at 635 °C appears to support this conclusion, since the tetrahedra were found to be displaced from the more ideal positions. The apparent uncorrected mean (Si, Al)–O bond length obtained by Peacor (1968) was considerably shorter than that predicted by Smith and Bailey (1963) for the Si/Al ratio of leucite. However, the apparent shortening of the (Si, Al)–O bond length in leucite can be accounted for by the anisotropic thermal motion observed. It is probable that there is a slight increase in the (Si, Al)–O bond length in leucite on heating, which may be of the same order as that in quartz.

Taylor and Henderson (1968) studied the thermal expansion of other synthetic leucites with larger cations (Rb and Cs) in place of potassium. It could be argued that if these leucites were to achieve the ideal fully-expanded state then their cell parameters would be identical because the frameworks have identical constitutions. However, this neglects the anisotropic thermal motions of the oxygens in the tetrahedra and the influence of the interframework cations on this motion. For example, Cs-leucite, or pollucite, is cubic at room temperature due to the large size of the Cs atom, whereas leucite is not cubic until at least 600 °C. The isotropic temperature factor for the oxygen atom in pollucite at room temperature is approximately 2 Å² (Newnham, 1967; Beger, 1969), whereas that in leucite, when it is cubic at 635 °C, is 7.3 Å². Thus for a similar cubic structural state (in terms of the rotation of the tetrahedra and of the atomic coordinates) the oxygen atoms in leucite have a much greater thermal motion than those in pollucite. Therefore, leucite will have a greater shortening of its (Si, Al)–O bond length due to thermal motion than will pollucite, and so the framework of pollucite will actually be significantly larger than that of leucite when the ideal fully-expanded state is achieved. It is believed that the combined effect of cation size and thermal motion of the oxygen atoms explains why the volumes of synthetic leucites at the temperature T_d (Taylor and Henderson, 1968) are not identical but show

some relationship to the interframework cation size. This suggests that the effect of an increase in temperature in increasing the cell volume should not be likened to the increase in cell volume at a given temperature produced by the substitution of a larger interframework cation, since temperature affects not only the degree of rotation of the tetrahedra but also the amount of anisotropic thermal motion of the framework oxygens.

Taylor and Henderson (1968) assumed by analogy with the sodalite group of minerals that the ideal fully-expanded state was not achieved in the leucite minerals at temperature T_d . This hypothesis was based on the discrepancy between the calculated cell size of the fully-expanded sodalite structure and that achieved experimentally at temperature T_d . It is now clear that such a discrepancy is to be expected and that the ideal fully-expanded state was probably achieved at the temperature T_d . Where Taylor and Henderson (1968) refer to the 'maximum uncollapsed state' the term 'ideal fully-expanded state' should now be substituted.

The sodalite group of minerals are aluminosilicates amongst which sodalite and hydro-sodalite have ordering of the Si and Al atoms (Löns and Schulz, 1967; Bukin and Makarov, 1967). The most recent information on nosean suggests that Al and Si are disordered (Schulz and Saalfeld, 1965), and the structure of häüyne has not yet been refined. An ordered distribution of Si and Al in the ideal fully-expanded structure would give rise to some distortion since the sequence Si-Al-Si-Al-Si across the unit cell gives a calculated cell edge of 9.308 Å (Si-O = 1.628 Å, Al-O = 1.728 Å) whereas the sequence Al-Si-Al-Si-Al at right angles gives a cell edge of 9.403 Å. It has been calculated that varying the tetrahedral angle, O-(Si, Al)-O, by $\pm 0.5^\circ$ is all that is required to prevent this distortion due to ordering, which is less than that observed at room temperature in sodalite (Löns and Schulz, 1967). It is known that both the interframework cations and anions present in the sodalite group of minerals affect the room-temperature cell parameters, and also that the interframework cations appear to have some effect on the cell parameters at high temperatures (Taylor, 1968). The same type of argument as applied to the leucite group of minerals with regard to the combined effect of cation size and thermal motion of the oxygen atoms of the framework can also be applied to the sodalite group. Using the above equation it is possible to calculate the mean (Si, Al)-O bond lengths for the cell parameters of noseans and häüynes at the temperature T_d given by Taylor (1968). The amount of thermal motion of the oxygens necessary to bring these calculated bond lengths to the value according to Smith and Bailey (1963) for the particular Si/Al ratio, after making a nominal correction for room-temperature thermal motion, varies from 0.37 to 0.40 Å. Such a degree of thermal motion, which is similar to that observed for leucite and tridymite at high temperatures, is sufficient to allow the noseans and häüynes to achieve the ideal fully-expanded state. However, the space group evidence (Taylor, 1968) suggests that the ideal fully-expanded state is not achieved at T_d . The realization of the true situation must await a structural refinement of this group at a temperature above T_d . Until such information is available it is the writer's opinion that the ideal fully-expanded state is achieved at T_d . The low rate of expansion of the

noseans and haiynes above the temperature T_d is believed to correspond to the expansion of the framework bonds and from fig. 3 is seen to be similar to that observed for cristobalite and leucite at high temperatures.

Conclusions

This paper has attempted to provide a unified interpretation of the thermal expansion behaviour of framework silicates. The main part of the expansion is shown to be due to the rotation of the framework tetrahedra from a partially-collapsed structure modified by the increasing amplitude of the anisotropic thermal motion of the framework oxygens. However, detailed correspondence between calculated and observed expansion curves may only be achieved by taking into account factors such as the distortion of the tetrahedra from a regular form, the effect of the variation in the (Si, Al)-O-(Al, Si) bond angle, the slight expansion of the (Si, Al)-O bond, and the influence of the interframework cations. This interpretation may satisfy the question of how the thermal expansion takes place but does not attempt to show why it takes place. The present writer's opinion is that the fundamental cause of the type of thermal expansion seen in the framework silicates may be thermally-induced changes in the (Si, Al)-O-(Al, Si) angles of the framework, or simply a result of the changes in the thermal motion of the oxygen atoms with temperature. The study has indicated the need for further accurate thermal expansion studies and shown the type of information that may be derived from such studies when coupled with high-temperature structure determinations. Studies of this kind need not be restricted to the framework silicates since similar structural controls on thermal expansion are likely amongst all the silicates and other inorganic polymers such as borates and phosphates.

Acknowledgement. The author wishes to thank Dr. W. L. Brown for critically reviewing the manuscript.

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[*Manuscript received 12 January 1971*]