

THE STRUCTURES OF THE PRINCIPAL FELSPARS

By

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Abstract

Moderately accurate structure analyses have been completed for most of the "key" feldspar structures—sanidine, orthoclase, microcline; low-temperature and high-temperature albites; celsian; anorthite. Their salient features are described and discussed, and applied to the consideration of the series of "mixed" K–Na, K–Ba and Na–Ca feldspars. Some information is also available about the structures of oligoclase, andesine and bytownite, important members of the Na–Ca (plagioclase) series.

Certain important generalisations which emerge from these structural studies are summarised.

The paper also provides a basis for the detailed accounts, to be given by following contributors, of the maximum microcline structure, of the three-dimensional analysis of low albite, of the peristerites, of transitional anorthite, and of order-disorder effects in feldspars.

1. Introduction

The structural scheme common to all feldspars was determined, and many of the features which differentiate one feldspar from another were discovered, more than 25 years ago (Taylor (1933); Taylor, Darbyshire and Strunz (1934)). These structural studies, though very inaccurate by modern standards, also served to explain the nature of the perthitic structures of K, Na feldspars, to reveal the complexity of the plagioclase series of Na, Ca feldspars, and to account for the striking twinning behaviour characteristic of both orthoclases and plagioclases.

The framework of linked tetrahedral SiO_4 and AlO_4 groups established as the basis of all feldspar structures is familiar and need not be described here. Its relative flexibility was shown to allow either large cations (K, Ba) or small ones (Na, Ca) to occupy a set of interstices

in the framework; the distinction between orthoclases [1] and plagioclases arises in this way. The perthitic structure could then be regarded, in broad general terms, as essentially a continuous framework held in extended configuration in K-rich regions, somewhat collapsed in Na-rich regions. The plagioclases, previously thought to provide a perfect example of a continuous isomorphous series, were found to include very complex structural modifications, now known to depend, in part, on the distribution of Al and Si among the tetrahedral groups—an important feature of feldspar structures for which experimental evidence was first obtained with orthoclase (Chao, Hargreaves and Taylor (1940)). With the early work already mentioned should be included the discovery and partial elucidation of the twinned orientations of Na-rich lamellae in perthite (Chao and Taylor (1940a)), and the discovery and first attempted explanation of the diffraction pattern characteristic of the intermediate (low-temperature) plagioclases (Chao and Taylor (1940b)).

After a gap of nearly ten years, the publication of a new analysis of sanidine (Cole, Sørum and Kennard (1949)) may be regarded as marking the beginning of a series of structure analyses all at least moderately accurate, some very accurate, when judged by modern standards. They include also orthoclase, intermediate and maximum microclines, celsian, low-temperature and high-temperature albites, anorthite, oligoclase, andesine, bytownite, and the intermediate (low-temperature) plagioclases; references will be given in following paragraphs.

In discussing all these structures a generalised and systematic nomenclature (Megaw (1956)) will be used; and, since many of the features which differentiate one feldspar from another are not easily distinguished in diagrams, tabulated lists of interatomic distances, temperature factors, etc. will be used instead (Tables 1 to 11).

The gradual accumulation of accurate factual information about the feldspar structures, in combination with information from optical and thermal studies, is now beginning to reveal many of the principles governing the feldspar structures, and it is convenient to review some

[1] The term orthoclase may be used to denote *either* the monoclinic or nearly-monoclinic group of feldspars *or* one particular mineral of that group, characterised by its composition and optical properties. The meaning to be attached to the term in the present paper will be obvious from the context.

of the more important general concepts, which will find repeated application, before proceeding to the discussion of individual structures.

(a) The optical (and other physical) properties of a feldspar of given composition may depend very markedly on the thermal history of the material. In general, a range of "transitional" states exists between the extremes of so-called "low-temperature" and "high temperature" materials. The term "low-temperature" may mean *either* formed at low temperature *or* brought into its present state by very prolonged annealing (under geological conditions) at low temperature; the term "high-temperature" means formed at high temperature or brought into that state by treatment at high temperature for a suitable period. Some materials may be preserved in the high-temperature state on quenching to a low temperature; conversion from low-temperature state to high-temperature state is usually carried out by simple heating; but complete conversion from high-temperature to low-temperature form has not been achieved under laboratory conditions, and, similarly, synthesis never results in the low-temperature form.

(b) Since the atoms Al and Si have very similar scattering powers for X-rays, the distribution of Al and Si atoms in the tetrahedral groups must be deduced from the sizes of the tetrahedra, AlO_4 being slightly larger than SiO_4 . The difference in size is small, and it is usually necessary to undertake a (laborious) three-dimensional refinement of the structure in order to obtain a reasonably accurate and reliable measure of the Al, Si distribution.

A graph of tetrahedron size (i.e. mean distance Si (or Al) to O) against Al, Si occupancy, published by Smith (1954), has been used in all but the most recent studies of Al, Si distribution. Evidence now seems to be accumulating, however, that in the feldspars the relationship is better represented by a slightly different straight line between end-points 1.61 Å for SiO_4 and 1.75 Å for AlO_4 . This relationship has been used to derive the tetrahedron occupancies quoted throughout the present paper; they thus differ a little from the published values; but the indirect nature of the argument implicit in this procedure must be kept in mind in studying the figures given.

(c) The physical significance to be attached to a fractional tetrahedron occupancy deduced in this way is not always obvious. An occupancy $\text{Al}_{\frac{1}{3}}\text{Si}_{\frac{1}{2}}$ must in the simplest interpretation represent an

average for two tetrahedra, one pure Si and one pure Al, and other fractional Al, Si occupancies correspond to more complicated averages. For most purposes it is sufficient to think of all such cases as simple space averages over an extended region of the structure, but more sophisticated interpretations will be considered later.

(*d*) In refined structure analyses the theoretical scattering powers of the atoms are modified by the application of "temperature factors" representing the effect of thermal vibration, either isotropic or anisotropic. The effective "shape" of the atom may thus vary from nearly spherical to highly elongated, according to the nature of any thermal anisotropy which exists.

Somewhat similar apparent "shapes" for an atom may result if the apparent position of the atom in the structure, as determined, really represents slightly different atomic positions averaged over an extended region of the structure, possibly as a consequence of partial disorder.

Any such anisotropy observed in a structure determined at room temperature should disappear or be reduced at low temperature if it is really due to anisotropic thermal vibration; if it remains unchanged, a model must be sought for the space average of atomic positions which reproduces the observed anisotropy. Prominent effects of this kind, observed in several feldspars, have been discussed in terms of faulted structures or disordered domains; they will be considered later.

2. Potassium feldspar, KAlSi_3O_8

Accurate structure analyses have been completed for a sanidine, an orthoclase and two microclines. The unit cell dimensions and space group for each of the materials studied are included in Table 1. The ideal unit cell content $4(\text{KAlSi}_3\text{O}_8)$ is assumed although in fact a little Na replaces some K in each material. Details of the structures are given first, their significance is discussed afterwards.

2.1 *Sanidine*

The specimen used for the accurate structure analysis (Cole, Sørum and Kennard (1949)) was prepared (Spencer (1930, 1937)) by heating for 300 hours at 1075 °C a colourless, optically homogeneous orthoclase, Spencer *C*, with composition 92% (by weight) potassium feldspar. The heated material has the optical properties characteristic of sanidine

Table 1. Unit cells and space groups for feldspars

Ideal Composition	Material	<i>a</i>	<i>b</i>	<i>c</i>	α	β	γ	Space Group
KAlSi ₃ O ₈	Sanidine	8.564	13.030	7.175	90	115.99	90	C2/m
	Orthoclase	8.562	12.996	7.193	90	116.01 ₅	90	C2/m
	Intermediate microcline	8.578	12.960	7.211	90.30	115.97	89.13	C $\bar{1}$
	Maximum microcline	8.561	12.966	7.216	90.65	115.83	87.70	C $\bar{1}$
NaAlSi ₃ O ₈	Low albite	8.138	12.789	7.156	94.33	116.57	87.65	C $\bar{1}$
	High albite	8.149	12.880	7.106	93.37	116.30	90.28	C $\bar{1}$
BaAl ₂ Si ₂ O ₈	Celsian	8.627	13.045	14.408	90	115.22	90	I2/c
CaAl ₂ Si ₂ O ₈	Anorthite							
	(low temperature)	8.177	12.877	14.169	93.17	115.85	91.22	P $\bar{1}$
Ab ₇₄ An ₂₂	Oligoclase	8.169	12.836	7.134	93.83	116.45	88.99	C $\bar{1}$
Ab ₅₂ An ₄₈	Andesine							
	(high temperature)	8.176	13.879	7.107	93.40	116.17	90.40	C $\bar{1}$
Ab ₂₀ An ₈₀	Bytownite	8.171	12.869	14.181	93.37	115.97	90.53	I $\bar{1}$

Notes

[1] The origin of the material, and its actual composition, are given in the text in each case.

[2] Axial lengths in Å, axial angles in °. The values quoted are accurate to within ± 0.005 Å., $\pm 0.03^\circ$ for all materials.

and is perfectly homogeneous and monoclinic in symmetry, under both X-ray and optical examination.

In the unit cell 16 tetrahedral groups, containing 4Al + 12Si atoms, are located on two sets of crystallographically-distinct sites, T₁ and T₂, each 8-fold. The 32 oxygen atoms in the unit cell comprise 4OA₁ (on diad rotation axes), 4OA₂ (on reflexion planes), 8OB, 8OC, 8OD. The K atom occupies a 4-fold site on the reflexion planes.

The interatomic distances are listed in Tables 2 and 3. The two tetrahedral groups, around T₁ and T₂, are identical in size whether measured in terms of average T–O or average O–O distances; each site is therefore assumed to contain (on the average) Al₁Si₃, the Al, Si distribution being completely random. There is, however, a considerable variation in the lengths of individual tetrahedron edges (i.e. O–O distances), even within the same tetrahedron; this departure from regularity of form lies quite outside the estimated error of the measurements. In the group of 9 O atoms around K, one bond K–OA₂ is much

Table 2. Tetrahedron dimensions in potassium feldspars (in Å).

Material	Bond	Group		Bond	Group					
		T ₁	T ₂		T ₁	T ₂				
Sanidine	T-OA	1.642	1.644	OA-OB	2.618	2.689				
	T-OB	1.640	1.640	OA-OC	2.743	2.600				
	T-OC	1.647	1.642	OA-OD	2.623	2.674				
	T-OD	1.640	1.644	OB-OC	2.707	2.686				
				OB-OD	2.718	2.716				
				OC-OD	2.680	2.721				
	Mean	1.642	1.642	Mean	2.682	2.681				
Orthoclase	T-OA	1.651	1.639	OA-OB	2.639	2.674				
	T-OB	1.650	1.624	OA-OC	2.781	2.588				
	T-OC	1.654	1.634	OA-OD	2.645	2.661				
	T-OD	1.651	1.635	OB-OC	2.710	2.690				
				OB-OD	2.726	2.689				
				OC-OD	2.692	2.690				
	Mean	1.652	1.633	Mean	2.699	2.665				
Intermediate Microcline		T ₁ (0)	T ₁ (m)	T ₂ (0)	T ₂ (m)		T ₁ (0)	T ₁ (m)	T ₂ (0)	T ₂ (m)
	T-OA	1.697	1.643	1.618	1.611	OA-OB	2.690	2.650	2.624	2.603
	T-OB	1.698	1.646	1.607	1.608	OA-OC	2.834	2.744	2.577	2.552
	T-OC	1.698	1.644	1.616	1.609	OA-OD	2.704	2.640	2.630	2.613
	T-OD	1.705	1.647	1.615	1.614	OB-OC	2.830	2.714	2.650	2.649
						OB-OD	2.794	2.700	2.656	2.647
					OC-OD	2.795	2.665	2.673	2.703	
	Mean	1.700	1.645	1.614	1.611	Mean	2.775	2.686	2.635	2.628
Maximum Microcline	T-OA	1.738	1.592	1.614	1.644	OA-OB	2.761	2.592	2.639	2.607
	T-OB	1.739	1.608	1.574	1.617	OA-OC	2.910	2.690	2.556	2.563
	T-OC	1.745	1.629	1.633	1.593	OA-OD	2.770	2.598	2.613	2.639
	T-OD	1.741	1.628	1.621	1.592	OB-OC	2.901	2.632	2.648	2.631
						OB-OD	2.864	2.667	2.649	2.648
						OC-OD	2.840	2.638	2.665	2.685
	Mean	1.741	1.614	1.611	1.612	Mean	2.841	2.636	2.628	2.629

Notes

[1] Sanidine: estimated errors less than 0.008 Å for T-O, less than 0.012 Å for O-O.

[2] Orthoclase: standard deviations approx. 0.015 Å for T-O, 0.018 Å for O-O.

[3] Intermediate microcline: standard deviations less than 0.009 Å for T-O, 0.012 Å for O-O.

[4] Maximum microcline: standard deviation 0.008 Å for T-O, 0.011 Å for O-O.

Table 3. Coordination and temperature factors of K atom in potassium feldspars.

Bonds	Sanidine (Å)	Orthoclase (Å)	Intermediate Microcline (Å)	Maximum Microcline (Å)
K-OA ₁	2.919	2.898	2.882, 2.897	2.877, 2.881
K-OA ₂	2.698	2.698	2.758	2.750, 3.402
K-OB	3.026	3.029	3.105, 3.096	2.962, 3.136
K-OC	3.129	3.135	3.054, 3.209	2.908, 3.336
K-OD	2.948	3.111	2.920, 2.985	2.893, 2.994
Temperature Factors	(Å ²)	(Å ²)	(Å ²)	(Å ²)
B _x	1.3	1.0	0.7	1.0
B _y	2.3	1.8	1.2	1.6
B _z	1.9	1.5	1.0	1.6

*Notes**Contact distances*

- [1] Sanidine: estimated error less than 0.008 Å; 2 contacts to each kind of atom except OA₂.
- [2] Orthoclase: standard deviation approximately 0.015 Å; 2 contacts to each kind of atom except OA₂.
- [3] Intermediate microcline: standard deviation less than 0.009 Å; contacts as shown.
- [4] Maximum microcline: standard deviation 0.008 Å; contacts as shown.

Temperature factors

- [1] Orientation B_x along a*, B_y along b, B_z along c.
- [2] Orthoclase and maximum microcline: B-values determined in course of structure analysis.
- [3] Sanidine and intermediate microcline: estimated from difference syntheses, assuming the ratios B_x:B_y:B_z to be the same as for orthoclase.

shorter than the others, and only a little greater than the sum of the accepted radii 1.33 Å (K⁺) and 1.32 Å (O²⁻).

The electron densities at the peaks T₁ and T₂ are identical.

2.2. Orthoclase

The interatomic distances listed in Table 2, determined (Jones and Taylor (1961)) for the same orthoclase (Spencer C) as was used to prepare the sanidine described above, correspond to partial Al, Si

ordering represented by occupancies (0.3Al + 0.7Si) in T_1 and (0.2Al + 0.8Si) in T_2 , compared with (0.25Al + 0.75Si) in all sites T_1 and T_2 in sanidine. The very short K-OA₂ bond is seen again (Table 3).

It is perhaps useful to emphasise that a material with optical properties which identify it as an orthoclase is not necessarily structurally identical with Spencer C; it would not be surprising to find varying degrees of Al,Si order in different orthoclases.

2.3. *Microcline*

For convenient comparison with the monoclinic feldspars, the base-centred cell is retained for microcline (Table 1). The space group is now $C\bar{1}$, and the 8-fold sites T_1 and T_2 are replaced by pairs of 4-fold sites $T_1(0) + T_1(m)$ and $T_2(0) + T_2(m)$; similarly for the sites occupied by oxygen atoms OB, OC, OD. (For a full account of the systematic nomenclature, see Megaw (1956)). The orientation of the triclinic axes proposed by Laves (1951) is used throughout this paper.

From the series of microclines (Bailey (1954), Goldsmith and Laves (1954), MacKenzie (1954)) extending from near-monoclinic to markedly triclinic, structures are now known for an intermediate and for a maximum microcline.

(a) *Intermediate microcline*

Bailey and Taylor (1955) analysed a transparent and colourless micropertthite, Spencer U from Kodarma, containing about 85% (by weight) potassium feldspar; the weak sodium-feldspar reflexions of the double diffraction pattern did not interfere with the measurement of the main potassium-feldspar (microcline) reflexions.

The interatomic distances in Table 2 correspond to partial Al, Si ordering with Al-contents $T_1(0)$ 0.65, $T_1(m)$ 0.25, $T_2(0)$ 0.03, $T_2(m)$ 0.01, total 0.94 Al. The tetrahedral groups again show marked departures from regular form and the K-atom environment (Table 3) is again similar to that in sanidine and orthoclase.

(b) *Maximum microcline*

Tables 2 and 3 also summarise the results of the analysis of a maximum microcline by Brown and Bailey (1961), (also, Bailey (1961)) which will be described by Brown. The site-occupancies are $T_1(0)$ 0.93, $T_1(m)$ 0.03, $T_2(0)$ 0.01, $T_2(m)$ 0.01, total 0.98 Al.

2.4. Discussion of potassium feldspars

The following important points emerge from a study of the structures of sanidine, orthoclase and microcline.

(a) In all the structures the (Al,Si) O_4 tetrahedra are distorted from the regular form. The distortion of a given tetrahedron is similar for all the structures and (as will be seen below) is also found in celsian and anorthite where the Al,Si ordering is totally different. It may be deduced that the distortions are a consequence of the general balance of forces between tetrahedral bonds on the one hand and interactions between O and the cation K (or Ba, or Ca) on the other. It also follows that the nature of the Al,Si ordering may be relatively unimportant in determining the detailed geometry of the structure.

(b) In passing from sanidine to maximum microcline, the Al,Si ordering process consists of the transfer of Al out of sites $T_2(0)$ and $T_2(m)$ first, and out of $T_1(m)$ afterwards, into $T_1(0)$. (In monoclinic orthoclase, of course, all T_1 sites are equivalent and are slightly Al-rich.) The tendency for Al to be concentrated in $T_1(0)$ in preference to $T_1(m)$ suggests a simple and natural explanation for the multiple albite-twinning and pericline-twinning seen as cross-hatching in microclines. For in a given region of the crystal, the T_1 sites of sanidine divide into two sets of sites as the symmetry becomes triclinic in microcline; either of these sets of sites may become $T_1(0)$ if it acquires a high proportion of Al, and adjacent localities take up the twin relationship as one or other of the sets of sites becomes identified as $T_1(0)$.

(c) Consideration of the fractional Al,Si occupancies of the T_1 and T_2 sites in orthoclase has led to the tentative conclusion that these may indicate *either* local near-randomness of Al,Si distribution on a unit cell scale, the fractional occupancy representing the probability of finding AlO_4 or SiO_4 at a given tetrahedral site; *or* the consequence of stacking disorder of a structure based on a larger unit cell—a concept which will be discussed by Megaw. The evidence is not regarded as providing any support for the view (Laves (1952)) that orthoclase is merely sub-microscopically twinned microcline.

(d) The nature of the forces responsible for the Al,Si ordering is not fully understood, though the simplest feldspar model suggests a possible

reason for the concentration of Al in sites T_1 at the expense of sites T_2 . For the oxygen atom OA_1 has contacts with *two* K atoms and is the only oxygen atom to do so; since it also links two tetrahedra of type T_1 the requirement of local valency-balance obviously favours Al in these sites if the bonding forces are even partly ionic in character.

(e) The environment of K is very similar in all structures (Table 3), and in particular the bond $K-OA_2$ is very short. Since, in the ordered structures, OA_2 links two tetrahedral groups containing little or no Al, it can contribute little or no bonding to K in these structures; the short bond thus demonstrates the importance, in this instance, of packing considerations in relation to ionic forces.

Table 3 also shows that there is some anisotropy of "thermal vibration" for the K atom. Whether this really represents anisotropy of thermal vibration within the surrounding group of nine oxygen atoms, or whether it really represents a space average of positions for the K atom which vary slightly according to the Al, Si distribution in the immediate locality, remains for consideration (see also, section 3.3 on the sodium feldspars).

3. Sodium feldspar, $NaAlSi_3O_8$

Moderately accurate structure analyses have been carried out, by two-dimensional methods, for both low-temperature and high-temperature albites; the cell dimensions for the materials examined are given in Table 1, and the interatomic distances in Tables 4, 5. In addition, a three-dimensional analysis for low albite is nearly completed and will be described by Ribbe; while a three-dimensional study of high albite has been begun.

3.1. *Low-temperature albite*

The results obtained by Taylor, Darbyshire and Strunz (1934) have been confirmed and greatly refined in two independent studies of a typical low albite from Ramona, containing about 98% $NaAlSi_3O_8$. The first (Ferguson, Traill and Taylor (1958)) carried out at room temperature, led to the interatomic distances listed in columns (1) of Tables 4 and 5; the second (Williams (1961)) carried out at liquid air temperature, gave the figures of columns (2) in Tables 4 and 5. The following features of the structure emerge partly from these numerical

Table 4. Tetrahedron dimensions in sodium feldspars (in Å).

Material	Bond	Group							
		$T_1(0)$		$T_1(m)$		$T_2(0)$		$T_2(m)$	
		[1]	[2]	[1]	[2]	[1]	[2]	[1]	[2]
Low albite	T-OA	1.762	1.738	1.599	1.611	1.618	1.649	1.640	1.633
	T-OB	1.702	1.741	1.626	1.629	1.627	1.580	1.597	1.600
	T-OC	1.762	1.724	1.553	1.626	1.649	1.607	1.618	1.617
	T-OD	1.744	1.745	1.583	1.603	1.649	1.652	1.611	1.613
	Mean	1.742	1.737	1.590	1.617	1.636	1.622	1.616	1.616
High albite	T-OA	1.652	1.662	1.646	1.639	1.646	1.622	1.637	1.712
	T-OB	1.654	1.686	1.633	1.646	1.635	1.615	1.633	1.598
	T-OC	1.645	1.642	1.641	1.673	1.640	1.636	1.668	1.635
	T-OD	1.658	1.683	1.636	1.567	1.645	1.680	1.649	1.671
	Mean	1.652	1.668	1.639	1.631	1.642	1.638	1.647	1.654

Notes

Columns [1] refer to Ferguson, Traill and Taylor (1958), measurements at room temperature.

Columns [2] refer to Williams (1961), measurements at -180°C .

For all distances, standard deviation not greater than 0.019 Å.

data, partly from the electron density distribution in Fourier syntheses: the notation for the atoms in the (triclinic) cell is the same as for microcline.

(a) There is a high concentration of Al in site $T_1(0)$, very little Al in $T_1(m)$ and $T_2(m)$; the room-temperature analysis shows a significant amount of Al in $T_2(0)$, while the low-temperature analysis shows only half this amount. The figures for Al-content at room temperature are $T_1(0)0.95$, $T_1(m)0$, $T_2(0)0.19$, $T_2(m)0.04$, total 1.18 Al; and at -180°C the corresponding occupancies are 0.91, 0.05, 0.09, 0.04, total 1.09 Al. The three-dimensional treatment (to be described by Ribbe) for the same material, at room temperature, gives occupancies 0.96, 0, 0.04, 0.02, total 1.02 Al, and must be regarded as superseding the earlier (two-dimensional) room-temperature study. [It is useful to stress that all Al-occupancies are derived from the (empirical) curve of tetrahedron-size against Al-content, which is itself constructed on the basis of measurements for structures in which the Al-content is thought to be known. Internal self-consistency for all structures considered is thus the strongest evidence that the Al-occupancies are correctly derived.]

Table 5. Coordination of Na atoms in sodium feldspars.

Bond	Low albite [1]	High albite [1]			High albite [2]
	Na	Na	Na'	Na''	Na
OA ₁	2.615 [4]	2.65 [4]	2.46 [3]	2.85 [5]	2.61 [4]
OA ₁	2.681 [5]	2.72 [5]	2.92 [6]	2.53 [3]	2.67 [5]
OA ₂	2.363 [1]	2.39 [1]	2.44 [2]	2.37 [1]	2.31 [1]
OA ₂	3.663	3.49	3.49	3.52	3.58
OB(0)	2.460 [2]	2.62 [3]	2.80 [5]	2.45 [2]	2.52 [3]
OB(m)	3.424	3.16	2.98 [7]	3.35	3.19
OC(0)	3.247	3.31	3.57	3.05 [7]	3.31
OC(m)	2.889 [6]	2.91 [6]	2.68 [4]	3.15	2.97 [6]
OD(0)	2.465 [3]	2.48 [2]	2.35 [1]	2.64 [4]	2.49 [2]
OD(m)	2.997 [7]	3.03 [7]	3.22	2.87 [6]	3.10 [7]

Notes

- [1] All distances in Å.
- [2] Columns (1) refer to measurements at room temperature (Ferguson, Trail and Taylor (1958)), column (2) at -180°C . (Williams (1961)).
- [3] Low albite from Ramona, and high albite from Amelia, inverted, for (1); synthetic pure albite for (2).
- [4] Columns headed Na give interatomic distances from central position of Na atom; columns headed Na' and Na'' give interatomic distances from extreme positions separated by 0.6 Å in y -direction.
- [5] Numbers in parentheses, following interatomic distances, show order of increasing Na-O distance for the seven closest O atoms in each case.
- [6] Standard deviations ~ 0.023 Å for low albite, ~ 0.06 Å for high albite.

(b) The electron density syntheses for -180°C show a small anisotropy in each of the T peaks, corresponding to half-atoms separated by ~ 0.06 Å; this important feature, if confirmed for room temperature when the three-dimensional study is completed, is most important in relation to a discussion of stacking faults, which will follow in the paper by Megaw (page 104).

(c) The Na atom appears markedly elongated in both room-temperature and low-temperature analyses. The effect must arise from a space-average, and a split-atom model is therefore adopted, to represent random occupation of either of two sites. The constants for this model, and for a model which assumes that the effective elongation at room temperature represents true anisotropic thermal vibration, are set out in Table 6.

Table 6. Interpretation of anisotropy of Na atom in sodium feldspars

Low albite

- (1) Ramona, measurements at room temperature.
- (a) Assuming anisotropic thermal vibration of single atom, $B \sim 2.5 \text{ \AA}^2$ parallel to elongation (approximately along b), $B \sim 1.3 \text{ \AA}^2$ perpendicular to elongation: corresponding amplitudes 0.18 \AA , 0.13 \AA .
- (b) Assuming random occupation of either of two sites, separation is $0.2\text{--}0.3 \text{ \AA}$ parallel to elongation, $B \sim 1.3 \text{ \AA}^2$ (isotropic) for atom in either site. (The estimate $\sim 0.1 \text{ \AA}$ (Ferguson, Traill and Taylor (1958)) was almost certainly too small).
- (2) Ramona, measurements at $-180 \text{ }^\circ\text{C}$.
Assuming random occupation of two sites, separation is 0.27 \AA at $\sim 20^\circ$ to b , and $B \sim 0.55 \text{ \AA}^2$ isotropic for atom in either site.

High albite

Assuming split-atom occupation of two sites:

- (1) Amelia (inverted), measurements at room temperature. Extreme positions Na' and Na'' (see Table 5), separation $\sim 0.6 \text{ \AA}$ along b , $B > 1.5 \text{ \AA}^2$ (isotropic) in either position.
- (2) Synthetic pure albite, measurements at $-180 \text{ }^\circ\text{C}$.
Sites Na₁ and Na₂, separation 0.65 \AA at 35° to b (nearly in (100) plane);

either	Na ₁ contains 0.5 Na, B (isotropic) 0.7 \AA^2 }	} B (isotropic) 1.0 \AA^2
	Na ₂ contains 0.5 Na, B (isotropic) 1.3 \AA^2 }	
or	Na ₁ contains 0.55 Na }	} B (isotropic) 1.0 \AA^2
	Na ₂ contains 0.45 Na }	

Notes

- (a) (1) refers to measurements at room temperature (Ferguson, Traill and Taylor (1958)), (2) at $-180 \text{ }^\circ\text{C}$. (Williams (1961)).
- (b) environment of sites Na₁ and Na₂ is very similar to that for Na' and Na'' (Table 5).

It may be noted, in addition, that partial measurements repeated by Williams (1961) at room temperature confirmed the distance of separation of the two sites (0.27 \AA) and the temperature factor for isotropic vibration in either site (B $1.3\text{--}1.4 \text{ \AA}^2$).

The three-dimensional study, not yet complete, also provides evidence of the effective anisotropy of the Na atom.

More complete discussions of these observations will be given by Megaw and by Ribbe (pages 104 and 152).

3.2. *High-temperature albite*

The only information so far available is from two independent two-

dimensional studies. In the first, at room temperature, Ferguson, Traill and Taylor (1958) examined Amelia albite (about 98% $\text{NaAlSi}_3\text{O}_8$) after inversion to the high-temperature form by heating at 1065 °C for 16 days. For the second analysis, at -180 °C, by Williams (1961), a synthetic high albite (pure $\text{NaAlSi}_3\text{O}_8$) was used.

(a) The tetrahedron dimensions (Table 4) measured in these two investigations correspond to Al occupancies $T_1(0)$ 0.31 and 0.42, $T_1(m)$ 0.21 and 0.16, $T_2(0)$ 0.24 and 0.20, $T_2(m)$ 0.27 and 0.32, totals 1.03 and 1.10 Al. (Figure for inverted Amelia first, for synthetic albite second, for each tetrahedral group.) The interpretation of these tetrahedron dimensions for the two materials is far from clear; there is a fairly definite indication of excess Al in $T_1(0)$ and a slight indication of excess in $T_2(m)$, but these must be regarded as somewhat tentative until the three-dimensional refinement has been completed. The anisotropy seen in all T peaks with low albite was *not* seen in the comparable study of high albite.

(b) The Na atom is elongated to a much greater extent than in low albite and the low-temperature measurements confirmed the obvious expectation that the "anisotropy" must be interpreted as arising from a space average for a split-atom model (Tables 5, 6). The magnitude of the separation, and the magnitudes of the (isotropic) temperature factors for atoms occupying these sites, are reasonable in relation to the corresponding quantities for low albite. It is a little surprising that the electron density syntheses do not show separate peaks corresponding to Na_1 and Na_2 : the three-dimensional analysis may be expected to throw light on this point.

3.3. *Discussion of sodium feldspars*

Although the structures of low and high albites are less accurately known, at present, than those of sanidine and the microclines, it is already possible to discuss the real physical significance of the fractional Al, Si occupancies of the tetrahedral sites, and of the large anisotropy of Na and relatively small anisotropy of K, in the alkali feldspars. As an alternative to the obvious interpretation in terms of positional disorder, a hypothesis based on a faulted structure has some advantages: this will be considered by Megaw.

4. Barium feldspar, $\text{BaAl}_2\text{Si}_2\text{O}_8$

The first structural study of celsian (Taylor, Darbyshire and Strunz (1934)) proved it to be essentially similar to sanidine, with Ba atoms taking the place of K in the same sites within the same tetrahedron framework. Observation of weak additional reflexions confirmed the anticipated doubling of the c -axis (Gay (1956)), and an accurate structure analysis was described for material of composition $\text{Ba}_{0.84}\text{K}_{0.18}\text{Al}_{1.90}\text{Si}_{2.11}\text{O}_8$ from Broken Hill (Newnham and Megaw (1960)). Cell dimensions are quoted in Table 1 and structural details in Table 7.

Table 7. Interatomic distances, and anisotropy of Ba, in celsian.

	Tetrahedron dimensions (Å.)				Ba-O distances (Å.)	
Bond	$T_1(0)$	$T_1(z)$	$T_2(0)$	$T_2(z)$	Ba-OA ₁	2.850, 2.850
T-OA	1.651	1.722	1.702	1.640	Ba-OA ₂	2.667, (3.421)
T-OB	1.640	1.703	1.698	1.645	Ba-OB	2.927, 2.939
T-OC	1.633	1.711	1.714	1.617	Ba-OC	3.112, 3.135
T-OD	1.630	1.733	1.733	1.637	Ba-OD	2.909, 2.909
Mean	1.639	1.717	1.712	1.635		

Temperature factors for Ba. (Å²)

$B_x 0.5$, $B_y 1.0$, $B_z 1.2$

($Y = b$; $X = a^*$ approximately)

Note

Standard deviations 0.012Å for T-O, 0.011Å for Ba-O.

(a) The Al occupancies are $T_1(0) 0.21$, $T_1(z) 0.77$, $T_2(0) 0.73$, $T_2(z) 0.18$. total 1.89 Al in very close agreement with the chemical composition. The Al,Si distribution thus corresponds to a high degree of ordering: moreover, it is of the simplest possible type—each Al-rich tetrahedron is surrounded by four Si-rich tetrahedra, and vice-versa. This arrangement seems likely to be electrostatically the most stable for a ratio Al:Si of 1:1 and is also found in anorthite $\text{CaAl}_2\text{Si}_2\text{O}_8$. It is this alternating network of Al and Si tetrahedra which requires the doubling of the feldspar (7 Å) c -axis, observed both in celsian and in anorthite. For the 7 Å axis corresponds to an odd number (3) of alternations Al-Si-Al-Si and is thus only a pseudo-repeat periodicity.

(b) The pattern of Al,Si ordering in this alternating network is essentially different from the Al,Si pattern in the most highly ordered

alkali feldspars, in which the ratio Al:Si is 1:3. In these every Al is surrounded by 4Si, while two-thirds of the Si have 1 Al and 3Si neighbours, and the remaining one-third have 2Al and 2Si. Nevertheless, individual tetrahedra show departures from regularity of form (*not* illustrated by the dimensions quoted in Table 7) strictly comparable to those observed in potassium feldspars. The interpretation, in terms of balance of forces, has already been suggested in the discussion of potassium feldspars.

(c) The oxygen atom environment is very similar for Ba (Table 7) and K (Table 3), and the anisotropy in the effective "shape" of the two atoms is also very similar. The rather surprising observation that Ba-O bonds are shorter than the corresponding K-O bonds suggests a more strongly homopolar character for the bonding in celsian; supporting evidence is discussed by Newnham and Megaw (1960).

(d) Nothing is known of any variations between low-temperature and high-temperature forms of $\text{BaAl}_2\text{Si}_2\text{O}_8$. Information on the series KAlSi_3O_8 - $\text{BaAl}_2\text{Si}_2\text{O}_8$ is also incomplete, though unpublished work by Gay (1962) has established the existence of a series of mixed K-Ba feldspars (hyalophanes) from pure K to $\text{K}_{\frac{3}{4}}\text{Ba}_{\frac{1}{4}}$ and again from $\text{K}_{\frac{1}{4}}\text{Ba}_{\frac{3}{4}}$ to pure Ba. No K, Ba feldspars have been found in the gap from $\text{Ba}_{\frac{1}{2}}$ to $\text{Ba}_{\frac{3}{4}}$, and it is permissible to suggest that the gap is a consequence of the totally different Al, Si ordering schemes for the K and Ba feldspars which flank it.

5. Calcium feldspar, $\text{CaAl}_2\text{Si}_2\text{O}_8$

Anorthite is the most complex of all the feldspar structures. Taylor, Darbyshire and Strunz (1934) first showed that the true *c*-axis (14 Å) is twice that of the other feldspars, though there is a well-defined pseudo-periodicity of 7 Å. Cole, Sørum and Taylor (1951) established the existence of two anorthite-type patterns, one corresponding to a primitive unit cell, the other to a body-centred cell, both with the doubled (14 Å) *c*-axis, and with axes $a \sim 8$ Å, $b \sim 13$ Å the same as for other feldspars. The X-ray reflexions from this unit cell are classified in groups (a) ($h+k$) even, l even; (b) ($h+k$) odd, l odd; (c) ($h+k$) even, l odd; (d) ($h+k$) odd, l even. In both forms of anorthite reflexions (a) are strong and sharp. In the body-centred pattern (b) are sharp but considerably weaker than (a), but (c) and (d) are absent; while

for primitive anorthite (c) and (d) are seen (in addition to (a) and (b)) —both sharp, and (c) comparable with (b) in intensity. Later work, mostly due to Gay and his collaborators, showed that the primitive anorthite structure is characteristic of low-temperature materials with composition at or near An_{100} (i.e. pure $CaAl_2Si_2O_8$); that the body-centred pattern is observed in low-temperature materials with 10% to 30% Na replacing Ca, the Al, Si ratio being adjusted accordingly, of course (i.e. $An_{90}Ab_{10}$ to $An_{70}Ab_{30}$); and that high-temperature material near An_{100} gives a pattern (“transitional anorthite”) tending towards, but not attaining, the body-centred type, for reflexions (d) have disappeared, reflexions (c) are very diffuse but still visible. These structures are considered in turn.

5.1. Primitive anorthite

The cell dimensions for a typical low-temperature anorthite (nearly pure $CaAl_2Si_2O_8$) from Monte Somma, Italy, are quoted in Table 1, and Tables 8, 9, 10 summarise, very briefly, the information needed for the following discussion of the structure.

(a) The cell contains 16 (crystallographically) distinct tetrahedra,

Table 8. Mean T–O bond lengths (Å) for tetrahedral groups in anorthite and bytownite

Number	Atom	Mean T–O Bond		Number	Atom	Mean T–O Bond	
		Anorthite	Bytownite			Anorthite	Bytownite
1	$T_1(0000)$	1.613	1.620	9	$T_1(0z00)$	1.758	1.742
2	$T_1(00i0)$	1.616	1.622	10	$T_1(0zic)$	1.746	1.749
3	$T_1(mz0c)$	1.608	1.618	11	$T_1(m00c)$	1.752	1.738
4	$T_1(mzic)$	1.626	1.610	12	$T_1(m0ic)$	1.741	1.724
5	$T_2(0z00)$	1.613	1.621	13	$T_2(0000)$	1.746	1.710
6	$T_2(0zi0)$	1.610	1.611	14	$T_2(00i0)$	1.753	1.728
7	$T_2(m00c)$	1.602	1.608	15	$T_2(mz0c)$	1.744	1.728
8	$T_2(m0ic)$	1.628	1.628	16	$T_2(mzic)$	1.752	1.732
	Mean (1–8)	1.614	1.617		Mean (9–16)	1.749	1.731

Notes

[1] For anorthite, standard deviation in T–O bond is 0.0041 Å; for bytownite 0.0065 Å.

[2] The 16 tetrahedral groups are not symmetry-related.

Table 9. Variation of T-O bond length (\AA) with environment of O in anorthite

Group	Number of O atoms in group	Number of Ca neighbours	Mean bond length (\AA)	
			Si-O	Al-O
1	4	2	1.632	1.780
2	20		1.622	1.755
3	8		1.588	1.719

Note

Each of the 32 O atoms is coordinated with 1 Al, 1 Si and 2, 1 or 0 Ca atoms.

Table 10. Coordination of Ca atoms in anorthite and Na, Ca atoms in bytownite (distances in \AA)

Material	Atom Ca	OA ₁	OA ₂	OB	OC	OD	Mean
Anorthite	(000)	2.500, 2.618	2.279	2.368	3.088	2.423, 2.532	2.544
	(zi0)	2.471, 2.586	2.322	2.421	2.807	2.391, 2.771	2.538
	(z0c)	2.476, 2.720	2.350	2.464, 2.491	2.565	2.397	2.495
	(0ic)	2.459, 2.822	2.335	2.413, 2.496	2.568	2.382	2.495
Bytownite	Na,Ca						
	(000)	2.526, 2.669	2.324	2.407		2.447, 2.565	2.486
	(zi0)	2.387, 2.727	2.331	2.388	2.707	2.422	2.493
	(z0c)	2.434, 2.853	2.361	2.419, 2.484	2.500	2.455	2.501
	(0ic)	2.439, 2.845	2.312	2.413, 2.627	2.609	2.422	2.524

Notes

[1] For anorthite, standard deviation in Ca-O distance is 0.004\AA .; for bytownite, 0.008\AA .

[2] Ca in anorthite is 7-coordinated; in bytownite the groups (000) and (zi0) are 6-coordinated and may contain Na, Ca while groups (z0c) and (0ic) are 7-coordinated and contain Ca only.

8 small, 8 larger. The mean T-O distances correspond to nearly complete ordering; thus 1.614\AA and 1.749\AA corresponding to occupancies 0.03 Al and 0.99 Al. The alternation of AlO_4 and SiO_4 groups is the same as in celsian. Oxygen atoms may be linked to 2Ca atoms, to 1Ca atom, or may have no Ca linkage, with a corresponding marked variation in Si-O and Al-O bond-lengths (Table 9). Departure from the regular tetrahedral form is again found: all tetrahedra T₁

show the same distortion, whether they contain Si or Al, and similarly for T_2 ; moreover these distortions are very similar to those already noted in other structures (e.g. edge of tetrahedron OA–OC is long in T_1 but short in T_2).

(b) The Ca atom is located within an irregular cavity bounded by about 10 oxygen atoms: Table 10 lists the 7 nearest O atoms for each kind of Ca atom, and it is seen that all Ca atoms are tightly coordinated. Features of the cation environment already noted in other feldspars are seen again here, in spite of the irregularity of the group of O atoms—e.g. the bond Ca–OA₂ is exceptionally short. The temperature factor $B \sim 1.0 \text{ \AA}^2$ for each Ca atom: this, and the tight coordination group around the atom, make it clear that there is no significant randomness or disorder in the Ca positions.

(c) The small temperature factors $B \sim 0.6 \text{ \AA}^2$ for O and $B \sim 0.2 \text{ \AA}^2$ for T atoms confirm the perfection of the Al,Si ordering in this structure; in maximum microcline, which also has a highly ordered structure, the B values are only slightly larger.

5.2. “Body-centred anorthite” (*bytownite*)

The material examined, (Chandrasekhar (1957), Fleet (1962)) a bytownite Ab₂₀An₈₀, should really be described as a mixed Ca–Na feldspar: it is, however, convenient to regard it as anorthite-type by contrast with the intermediate plagioclases.

The only reflexions observed are of (a) and (b) types, and the structure derived from these is an average in which Na,Ca atoms are split among sites obviously related to the Ca atoms in (primitive) anorthite, while T and O atoms are anisotropic and can be replaced by split atoms. From the structures which might be constructed from these half-atoms (T and O), one is chosen which is most like primitive anorthite, and it is then found that the tetrahedral groups are nearly regular in form (as in primitive anorthite). (This is discussed by Megaw in another paper.) The structural details are listed in Tables 8, 10. (It is, of course, assumed that this truly primitive structure exists only when it is faulted to emulate body-centring, so that no reflexions (c) and (d) are observed.)

(a) The structure is highly ordered, with mean T–O distances

corresponding to occupancies 0.05 Al and 0.87 Al, total 0.92 Al in close agreement with the chemical composition. The Si-rich and Al-rich tetrahedra alternate throughout the framework exactly as in primitive anorthite, and the departures from regularity of form are generally similar to those in primitive anorthite.

(b) Two of the four cation sites are now 6-coordinated while the other two remain 7-coordinated as in primitive anorthite: it is suggested that the smaller Na atoms are concentrated in the former.

5.3. *Transitional anorthite*

The analysis of this structure will be described by Ribbe and Megaw in another paper. (This volume pp. 158).

6. The plagioclase series

The complex sequence of structures between $\text{NaAlSi}_3\text{O}_8$ (albite) and $\text{CaAl}_2\text{Si}_2\text{O}_8$ (anorthite) occurs as a consequence of the nearly equal sizes of the cations Na and Ca, combined with the completely different Al,Si ordering schemes required in the end members for the Al:Si ratios 1:3 and 2:2.

6.1. *High-temperature plagioclases*

Transitional anorthite and high albite have already been discussed (sections 5.3 and 3.2); the existence of a monoclinic high albite has been reported (Brown (1960)) but its structure has not been examined; the only other high plagioclase for which structural information is available is an andesine from Linosa, though it is known that other high plagioclases over the composition range from Ab_{100} to $\text{Ab}_{10}\text{An}_{90}$ (approximately) all give diffraction patterns of the triclinic high albite type.

Andesine (High-temperature form)

Table 11 summarises the results of a partial analysis (Kempster (1957)) of an andesine $\text{Ab}_{52}\text{An}_{48}$. The tetrahedron dimensions correspond to Al-contents $T_1(0)$ 0.56; $T_1(m)$ 0.28; $T_2(0)$ 0.27; $T_2(m)$ 0.34; total 1.45 Al in close agreement with the composition 1.48 Al. The cations Na and Ca occupy a group of sites in positions corresponding to those occupied by the 4Ca atoms in anorthite.

Table 11. Mean T-O bond lengths (\AA) in oligoclase and andesine (Linosa)

Material	$T_1(0)$	$T_1(m)$	$T_2(0)$	$T_2(m)$
Oligoclase	1.682	1.634	1.632	1.652
Andesine	1.688	1.648	1.646	1.657

Coordination of Na, Ca atoms in oligoclase and andesine (Linosa)
(distances in \AA)

Material	Na, Ca	OA ₁	OA ₂	OB	OC	OD
Oligoclase	site [1]	2.78, 2.46	2.40	2.52	(3.00)	2.45, 2.77
	site [2]	2.41, 2.82	2.37	2.46, 2.94	2.86	2.51
Andesine	site (i)	2.80, 2.42	2.35	2.51	(3.07)	2.44, 2.74
	site (iv)	2.71, 2.57	2.37	2.51	(3.03)	2.37, 3.02
	sites (ii) (iii)	2.34, 2.48	2.37	2.50, 2.73	2.62	2.52

Notes

[1] Standard deviation for T-O bonds is $< 0.02\text{\AA}$. for oligoclase, and probably similar for andesine at this stage of refinement.

[2] For Na, Ca-O distances no standard deviation is quoted since refinement is not complete.

The Al-concentration in $T_1(0)$, reminiscent of that in ordered alkali feldspars, is surprising in a high-temperature material; it must be accepted with reserve, since the material may not be in the true high-temperature state, and the analysis is in any case incomplete. Nevertheless, it is interesting in relation to the similar (though smaller) concentration of Al in $T_1(0)$ in high albite—which also awaits confirmation from a three-dimensional analysis.

An interpretation of the high plagioclase structures in terms of faulted domains of anorthite-type sub-cells will be discussed by Megaw.

6.2. Low-temperature plagioclases

The composition range Ab_{100} to $Ab_{75}An_{25}$ includes the peristerites, which will be treated in a separate paper (Ribbe), and oligoclase for which a two-dimensional analysis has been completed and is described below. The range from $Ab_{75-70}An_{25-30}$ to An_{100} includes the “inter-

mediate plagioclases", which will be discussed by Megaw, bytownite ("body-centred anorthite") and primitive anorthite, (see sections 5.2 and 5.1).

Oligoclase

The two-dimensional analysis (Waring (1961)), summarised in Table 11, is for a material $\text{Ab}_{74}\text{An}_{22}\text{Or}_4$. The Al-contents are $T_1(0)$ 0.52; $T_1(m)$ 0.18; $T_2(0)$ 0.16; $T_2(m)$ 0.31; total 1.17 Al, in agreement with the chemical composition. It is probably best to accept the concentration of Al in $T_1(0)$ as certain, but to regard the precise Al-contents for the other groups as subject to confirmation. To a first approximation the cations Na, Ca occupy the sites (1) and (2) of Table 11, with greater scattering power associated with site (2); but further refinement breaks up site (2) into three distinct sites, adjacent to the Ca site (000) of anorthite, while site (1) replaces the three other Ca sites in anorthite.

Thus the Al-Si distribution is similar to that in low albite, though with a less marked concentration of Al in $T_1(0)$; while the Na, Ca split-atom sites correspond to an average structure with domains of anorthite type. The importance of these features in relation to peristeritic unmixing is obvious.

7. Conclusion

This survey of the structures of the principal feldspars may conclude with a list of the features now seen to be of importance for an understanding of the general principles governing their atomic architecture.

The Al, Si distribution in the tetrahedral groups ranges from fully ordered to completely random, according to the thermal history of the material and its composition. There are two different ordered patterns, one for the Al:Si ratio 1:3, the other for the Al:Si ratio 2:2. For the former, the *c*-axis periodicity is 7 Å, but for the latter this is only a pseudo-repeat and the true *c*-axis is 14 Å.

The form of the tetrahedral AlO_4 or SiO_4 group in a feldspar is not perfectly regular. The departure from regularity does not depend upon occupancy by Al or Si, but is controlled by the balance between forces within the tetrahedral group on the one hand, and forces external to the tetrahedron on the other.

The group of O atoms surrounding the cation K, Ba, Ca or Na is

rather irregular, but its characteristics remain surprisingly constant for most of the feldspar structures. The effective "shape" of the cation itself shows departures from the spherical form which may be large or small; low-temperature measurements show that, in some cases at least, this anisotropy is not due to anisotropic thermal vibration but must correspond to a space-average of domains with nearly identical structures.

The need to provide a physically-realistic interpretation of fractional Al, Si occupancies of tetrahedral sites, together with the observations of anisotropy of the cations, mentioned above, has led to the discussion of various models. Probably the most favoured is similar to that developed for application to the intermediate plagioclases, being based on a large unit cell and a domain structure with stacking faults.

Many of the tentative ideas now emerging, about the nature of the feldspar structure, depend upon obtaining the highest possible accuracy in the structure analysis; they will be confirmed or modified as accurate information becomes available for more and more feldspar minerals.

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