

STATISTICAL ANALYSIS AND DISCUSSION OF MEAN  
Al/Si-O BOND DISTANCES AND THE ALUMINUM  
CONTENT OF TETRAHEDRA IN FELDSPARS

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ABSTRACT

The aluminum content of aluminous feldspars is linearly related to the grand mean tetrahedral distance ( $\overline{T-O}$ ) by the equation

$$\text{Al}/(\text{Al}+\text{Si}) = 6.58 [(\overline{T-O}) - 1.605]$$

Because the data are restricted to the range 0.25–0.50 Al, the errors associated with the prediction of Al<sub>i</sub> content of tetrahedron T<sub>i</sub>, on extrapolated portions of the curve, increase with increasing difference from the mean value ( $\overline{T-O} = 1.56 \text{ \AA}$ ; Al=0.33) according to the equation

$$\text{var}(\text{Al}_i) \approx 44.0 \text{ var}(\overline{T_i-O}) + 0.034 \{(\overline{T_i-O}) - 1.65\}^2.$$

If only random errors are important, there is no statistical basis for rejecting the linear model proposed by Smith and Bailey. Using the equation above, this model predicts partial disorder for low albite and maximum microcline, which by analogy with reedmergnerite should be completely ordered. However, until electrostatic and covalent effects on the T-O bond are quantitatively understood, the linear model must be used with caution.

In a detailed study of Al-O and Si-O tetrahedral distances Smith and Bailey (1963) found that the mean tetrahedral bond length in feldspars varies linearly with percentage aluminum from 1.61 Å for Si-O to 1.75 Å for Al-O. They estimated that the average Al content of a tetrahedron can be predicted to within  $\pm 5$  percent. This relationship, determined from two- and three-dimensional refinements of twelve aluminous feldspar structures and reedmergnerite, has since been used to predict Al-Si distribution in feldspars and other framework structures, thereby permitting an estimate of the degree of long-range order or structural state. To facilitate comparison of the structural states of alkali feldspars, Smith and MacKenzie (1961) defined a "distant order function"

$$S = \sum_{j=1}^{j=4} \frac{|0.25 - S_j|}{1.50}$$

where  $S_j$  is "the distant order Al/(Al+Si) in the four different types of tetrahedra." This function ranks a feldspar between 0.0 for complete long-range disorder and 1.0 for complete order, and has been used to relate the structural state of potassium feldspars to such readily-measured physical properties as optic angle, birefringence and extinction angle (Brown, 1962; Finney and Bailey, 1964) and the ratio  $c^*/b^*$  (Jones, 1966).

Wright and Stewart (1968) measured the cell parameters of a series of alkali feldspars and their ion-exchanged equivalents and found that  $b$  and  $c$  can be used to estimate both composition and structural state. In re-

lating these cell parameters to the Smith-MacKenzie distant-order function, it became apparent that an assessment of the propagation of errors was needed to establish the statistical significance of the relationship (Stewart and Ribbe, 1967). This in turn necessitated a recalculation of the Smith-Bailey curve relating Al content to mean tetrahedral distance ( $\bar{T-O}$ ) and an evaluation of the associated errors. The results of this calculation produced new values for the slope and intercept of the Al vs ( $\bar{T-O}$ ) curve (Ribbe and Gibbs, 1967) and has led to a reexamination of the linearity of the model. Furthermore, because there are a number of different distant-order parameters (Bragg and Williams, 1934; Smith and MacKenzie, 1961; Christie, in press) and because they all are second-order in nature, it is proposed that the physical properties of feldspars no longer be related to distant-order parameters but that they be related to the mean sizes of the individual tetrahedra (or the predicted Al/Si distribution) from which these parameters were calculated (*cf.* Stewart and Ribbe, 1967; Colville and Ribbe, 1968b; Smith, 1968; Ribbe, 1968).

#### CALCULATIONS

To begin our study we compiled the grand-mean tetrahedral distances and Al contents for 13 three-dimensionally refined aluminous feldspar structures, and reedmergnerite, the boron analog of albite (Table 1). Fol-

TABLE 1. FELDSPARS USED IN CALCULATING LINEAR REGRESSIONS CURVES I AND II, IN ORDER OF INCREASING Al CONTENT

Feldspar	Al content	Grand mean T-O bond length (Å)	Reference
Reedmergnerite	0.0	1.615 <sub>2</sub> <sup>a</sup>	Appleman and Clark, 1965
Igneous max. microcline	0.25	1.644 <sub>2</sub>	Brown and Bailey, 1964
Authigenic max. microcline	0.25	1.644 <sub>1</sub>	Finney and Bailey, 1964
Low albite	0.25 <sub>1</sub> <sup>b</sup>	1.645 <sub>8</sub> <sup>b</sup>	Ribbe <i>et al.</i> , in press
High albite	0.25 <sub>2</sub>	1.643 <sub>3</sub>	Ribbe <i>et al.</i> , in press
Sanidine	0.25 <sub>3</sub>	1.642 <sub>1</sub>	Ribbe, 1963a
Orthoclase	0.25 <sub>3</sub>	1.641 <sub>9</sub>	Colville and Ribbe, 1968
Intermediate microcline	0.25 <sub>4</sub>	1.642 <sub>6</sub>	Bailey and Taylor, 1955
Adularia	0.25 <sub>4</sub>	1.642 <sub>8</sub>	Colville and Ribbe, 1968
Oligoclase	0.32 <sub>2</sub>	1.653 <sub>6</sub>	Colville and Ribbe, 1968a
Bytownite	0.45	1.673 <sub>7</sub>	Fleet <i>et al.</i> , 1966
Celsian	0.47 <sub>4</sub>	1.675 <sub>6</sub>	Newnham and Megaw, 1960
Primitive anorthite	0.49 <sub>4</sub>	1.680 <sub>5</sub>	Kempster, 1966
Transitional anorthite	0.49 <sub>8</sub>	1.681 <sub>5</sub>	Ribbe, 1963b

<sup>a</sup> For the Si sites only.

<sup>b</sup> Values from a refinement of low albite by Wainwright and Starkey (pers. comm.) are 0.25 and 1.644<sub>4</sub>.

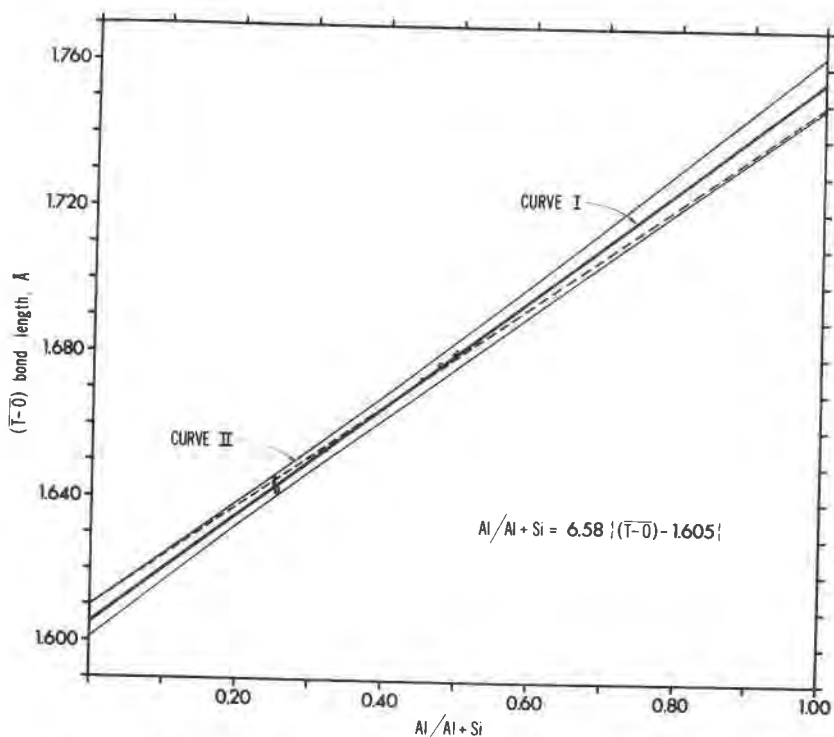


FIG. 1.  $(\overline{T-O})$  bond length as a function of  $Al/(Al+Si)$ , showing Curve I (solid heavy line) and Curve II (dashed line). Slightly curved light lines represent the 95 percent confidence band for Curve I; the equation for Curve I is at the lower right.

following the procedures outlined in Brownlee (1965, pp. 334-342) and Bennett and Franklin (1954, pp. 222-232) and assuming a linear model, we computed two regression lines, one using only aluminous feldspars, the other using these and reedmergnerite. Because of the unreliable estimates of standard errors in mean T-O distances associated with certain anorthite-rich feldspars, all data were given unit weights. The Al content was chosen as the independent variable in this calculation (*contra* Jones, 1968) because the  $(\overline{T-O})$  distance is dependent on the Al content. However, since the slopes of the calculated regression lines are about forty times greater than their respective standard errors, it makes very little difference which parameter is chosen as the independent variable.

The two regression lines are shown in Figure 1. Curve I was calculated using only data for aluminous feldspars; Curve II also includes the data for reedmergnerite and is statistically identical to that determined by Smith and Bailey (1963). The slopes, intercepts, their standard errors and the coefficients of correlation are listed in Table 2. It is important to re-

TABLE 2. INTERCEPTS, SLOPES AND COEFFICIENTS OF CORRELATION FOR CURVES I, II AND III COMPARED WITH CURVES OF JONES (1968)  
Standard errors in parentheses

	Intercept (Al=0.0)	Slope	Intercept (Al=1.0)	Coefficient of correlation
Feldspar data only				
Curve I	1.605 (1)	0.152 (4)	1.757	0.997
Jones (1968) <sup>a</sup>	1.603 <sub>3</sub>	.157 <sub>5</sub>	1.760 <sub>3</sub>	.997
Feldspar and Reedmergnerite Data				
Curve II	1.610 (2)	.139 (5)	1.749	.992
Non-feldspar data only				
Curve III, without keatite (Data from Jones, 1968)	1.606 (2)	.149 (5)	1.755	.991
With keatite, Jones (1968)	1.603 <sub>3</sub>	.155 <sub>5</sub>	1.759 <sub>1</sub>	.987

<sup>a</sup> Two values used in this paper are worth noting as different from those used by Jones. In bytownite the overall tetrahedral mean is 1.673; not 1.674<sub>2</sub>, the latter having been transmitted from an error in the original paper by Fleet *et al.* (1966). For celsian and Al-content used is 0.474 rather than Jones' 0.460. Jones (written communication, 1968) points out "... that the value of 0.460 is the amount of aluminum equivalent to a new microprobe determination of barium. This unfortunately was not explained in my paper."

alize that the observed data used to compute Curve I are confined to the narrow region bracketed by 0.25 and 0.50 Al and to understand that errors associated with the extrapolated portions of the curve will be significantly larger than those in the bracketed region. If the model is linear and if only random errors are important, then for feldspars the relationship between the variance of the mean T-O bond length of the  $i^{\text{th}}$  tetrahedron (obtained from a structure analysis) and its Al-content is given by the following expression:

$$\text{var}(\text{Al}_i) \approx 44.0 \text{ var}(\overline{\text{T}_i - \text{O}}) + 0.034\{(\overline{\text{T}_i - \text{O}}) - 1.65\}^2$$

Figure 2 is a graphical representation of this equation; notice that the estimated error in Al-content increases with increasing difference from the mean value [ $\overline{\text{T}-\text{O}} = 1.65 \text{ \AA}$ ; Al=0.33] for Curve I.

The slopes and intercepts of Curve I and II (Table 2, Fig. 1) differ at the "possibly significant" level (Cruickshank, 1949); however, they give different results when applied to feldspar structures. It is therefore useful to decide on criteria for selecting one of these curves or, if need be, rejecting *both* of them on the basis that a linear model is not valid. The criteria might be as follows:

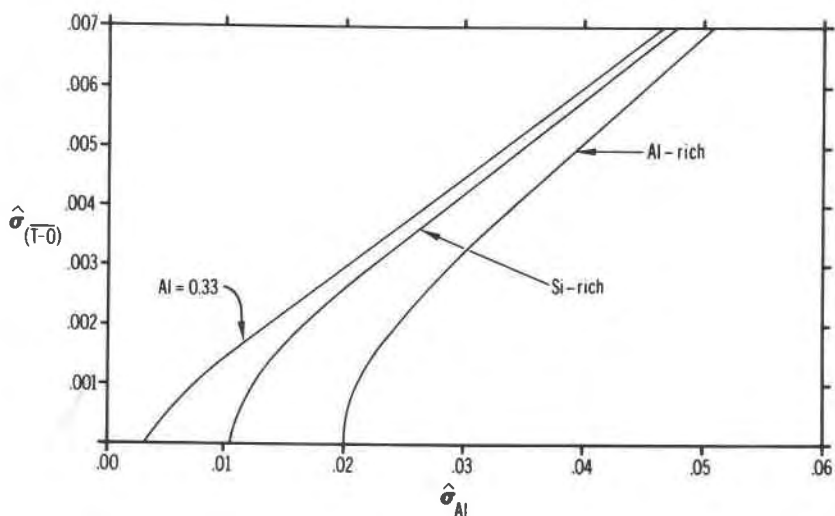


Fig. 2. Graphical representation of the equation (see text) relating the standard error in Al content of an individual tetrahedron to the standard error in  $(\overline{T-O})$  bond distance determined by crystal structure analysis. Notice that the estimated error increases with increasing difference from the mean value [ $(\overline{T-O}) = 1.65 \text{ \AA}$ ;  $Al = 0.33$ ] for Curve I.

(1) That Al determined by chemical methods agree with the total Al predicted by the curves using the mean tetrahedral bond distances calculated from the atomic coordinates.

(2) That no mean tetrahedral distance be greater than the intercept of the curve at  $Al = 1.0$  or less than the intercept at  $Al = 0.0$ . These criteria should hold if the linear model is strictly correct.

For the eight alkali feldspars listed in Table 3 Curve I clearly gives better average total Al ( $1.00_4$ ) than Curve II ( $0.97_0$ ). Furthermore, on the average, Curve I gives the theoretical Al content for four Al-rich feldspars, whereas Curve II on the average gives 2.5 percent excess total Al for the same Al-rich feldspars. Notice that in this test of Curve I, 97 percent of the individual mean tetrahedral bond distances fall on the *extrapolated* portion of the curve. By contrast only 33 percent fall outside the range of data points used in constructing Curve II, yet Curve II does not satisfy the first criterion as well as Curve I. By the second criterion, which says that no individual mean tetrahedral distances observed in feldspars should be significantly greater or less than the intercepts of the curve at  $Al = 1.0$  and  $Al = 0.0$ , Curve I also is the better of the two: 17 percent of the data points fall outside the range of Curve II, whereas only 5 percent fall outside the range of Curve I.

Thus Curve I is apparently better than Curve II for predicting the Al content of individual tetrahedra in aluminous feldspars. This is not sur-

TABLE 3. ALUMINUM CONTENT FROM CURVE I  
 (See text for discussion of standard errors)

Alkali feldspar	T <sub>1</sub> (0)	T <sub>1</sub> (m)	T <sub>2</sub> (0)	T <sub>2</sub> (m)	Total Al (Curve I)	Total Al (Curve II)
Low albite	0.92 <sub>5</sub>	0.03	0.06 <sub>5</sub>	0.04 <sub>5</sub>	1.06 <sub>5</sub> <sup>a</sup>	1.03 <sup>a</sup>
High albite	.28	.25	.22 <sub>5</sub>	.25	1.01	.97
Authigenic maximum microcline	.85	.05	.09	.02 <sub>5</sub>	1.01 <sub>5</sub>	.99
Igneous maximum microcline	.89	.06	.03 <sub>5</sub>	.04	1.02 <sub>5</sub>	.99
Intermediate microcline	.62	.26	.06	.04	.98	.94
Adularia (Spencer B)	.38 <sub>5</sub>	.38 <sub>5</sub>	.11	.11	.99	.96
Orthoclase (Spencer C)	.33 <sub>5</sub>	.33 <sub>5</sub>	.15	.15	.97	.93
Sanidine (heated Spencer C)	.26	.26	.23	.23	.98	.95
Average total Al:					1.00 <sub>4</sub>	0.97

<sup>a</sup> These values are from Ribbe *et al.* (in press). A recent refinement of low albite by Wainwright and Starkey (pers. comm.) gives 1.02 total Al using Curve I and 0.99 using Curve II. Recalculation of average total Al gives 0.99<sub>5</sub> for Curve I and 0.96<sub>5</sub> for Curve II.

prising, because reedmergnerite, NaBSi<sub>3</sub>O<sub>8</sub>, which does not contain Al in tetrahedral coordination, was used in determining Curve II. It may be that distortions of the tetrahedral framework caused by replacing aluminum ( $r \sim 0.5$  Å) by boron ( $r \sim 0.2$  Å) account for the fact that the mean Si-O bond length of reedmergnerite (1.615 Å) is longer than that predicted by either curve. In fact danburite, CaB<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>, containing twice as much tetrahedral boron, has an even longer mean Si-O bond length of 1.619 Å (Johansson, 1959).

It is interesting that the slope and intercept of Curve I are statistically identical to those calculated for non-feldspar framework structures (Curve III, Table 2), using data from 18 three-dimensionally refined structures tabulated by Jones (1968). This suggests that Curves I and III are applicable to all framework structures containing tetrahedrally coordinated Al and Si.

The correlation coefficient for Curve I indicates that within the range of observed data (Al=0.25 to 0.50), approximately 99.5 percent of the variation in  $(\overline{T-O})$  can be explained by a linear dependence on the Al content. The linearity of the extrapolated portions of Curve I is more difficult to establish, although the Si-rich end of the curve coincides with

that of Curve III whose correlation coefficient (0.991) indicates that in the range  $Al=0.00$  to  $0.50$ , 98 percent of the variation in  $(\overline{T-O})$  can be explained by a linear dependence on Al content. It may be that a higher order equation defines the  $(\overline{T-O})$ -Al relationship more perfectly; however, the tests of criteria (1) and (2) indicate that the simpler linear model is consistent with our data, and if only random errors are important, there is no apparent justification for rejecting it.

#### DISCUSSION

It should be emphasized that the relationship for determining the mean Al content of an individual tetrahedron is based on the *mean* T-O bond length for that tetrahedron, and not on any single bond length. In ordered feldspar structures (Clark and Papike, 1967) an individual T-O bond length for an Si-rich tetrahedron is  $\sim 1.59$  Å when the oxygen is shared with an Al-rich tetrahedron; when it is shared with another Si-rich tetrahedron, the T-O bond length is  $\sim 1.62$  Å. However, in agreement with Cruickshank's (1961) empirical rule that a long (or short) bond in a tetrahedron is compensated by corresponding change in the other T-O bonds to preserve the  $(\overline{T-O})$  bond characteristic of the structure type and its chemistry, the  $(\overline{T-O})$  bond for the three tetrahedra containing these bonds is  $1.612$  Å. This is  $0.007$  Å longer than the bond length predicted by Curve I for a pure Si tetrahedron. Furthermore, the  $(\overline{T-O})$  bond length for all allegedly "pure" Si tetrahedra in supposedly fully ordered feldspars (primitive and transitional anorthite, low albite, maximum microcline) is an even longer  $1.614$  Å. This indicates one of two possibilities: either these structures are not *completely* ordered or the linear model assumed for the  $(\overline{T-O})$ -Al relationship is incorrect.

Since the so-called completely ordered feldspars have had their "equilibrium" Al/Si distribution frozen in at  $400$ - $800^\circ K$ , and since the energy barrier to Al-Si exchange at lower temperatures is probably much greater than the thermal energy, perfect order in feldspars may be impossible. In spite of this, there is a tendency to make simplified statements about structural state; for example, because maximum microcline shows maximum deviation from the disordered monoclinic modifications, it has been assumed to be completely ordered. This is clearly a subjective judgment.

Appleman and Clark (1965) have deduced that low albite is completely ordered, because low albite and reedmergnerite have remarkably similar Si-O bond lengths and O-Si-O and Si-O-Si angles. Since their reedmergnerite contains no Al in the  $T_1(m)$ ,  $T_2(0)$ ,  $T_2(m)$  sites, they concluded that in both feldspars variations of  $(\overline{T-O})$  from the value predicted for pure Si occupancy by a linear regression curve is controlled by factors other than site occupancy. This is a cogent argument, but it is also reasonable to ask

whether or not larger Al atoms might occasionally be found in the T sites in low albite which are somewhat larger than predicted for pure Si occupancy.

The linear model predicts disorder for low albite and maximum microcline. However, the aluminum avoidance principle (Loewenstein, 1954; Goldsmith and Laves, 1955) and the intolerance of low albite and maximum microcline for Al in excess of 1.00 (evidenced by the peristerite solvus,  $An_2$ - $An_{15}$ , and the extremely limited solid solution of An in Or) indicate that substitutional disorder does not occur. Since substitutional disorder is not unlike site disorder it may be that these structures are in fact perfectly ordered and that the linear model does not correctly predict site occupancy for the ordered feldspars. Thus it is clear that until there is a method of quantitatively predicting electrostatic, chemical coordination and packing effects on the  $(\overline{T-O})$  bond, the linear model must be used with judicious caution; next-nearest neighbor effects may introduce unpredictable systematic variations in the individual T-O bond distances that are *not* compensated for by other bond distances to maintain a mean T-O distance characteristic of the chemistry of the tetrahedron.

This uncertainty about the linear character of the model implies that the use of any long-range order parameter to characterize the structural state of feldspars is of questionable value. In addition there are ambiguities in long-range order parameters: the Bragg and Williams (1934) parameter is sensitive only to the Al content of  $T_1(0)$  and does not differentiate between ordered and disordered Al distribution among  $T_1(m)$ ,  $T_2(0)$  and  $T_2(m)$ . The Smith and MacKenzie (1961) parameter is insensitive to Al interchange between  $T_1(0)$  and  $T_1(m)$  at  $(S) = 0.67$ . The Christie (in press) parameter  $S^*$  arbitrarily resolves these ambiguities by a complex calculation, but all of these parameters retain the disadvantage of being dependent on a knowledge of the *exact*  $(\overline{T-O})$ -Al relationship which has yet to be satisfactorily established.

Calculations of configurational entropy,  $S_{conf}$ , are extremely sensitive to small differences in Al site-occupancy for ordered feldspars. For example, in low albite  $S_{conf} = 0.0$  for complete order, 0.81 cal/deg mole for the Al/Si distribution predicted by the Smith-Bailey curve ( $\equiv$  Curve II) and 1.81 cal/deg mole for the distribution predicted from Curve I. If the low  $\rightarrow$  high albite transformation is first order (as assumed by Holm and Kleppa, 1968), then "equilibrium transformation temperatures" calculated in the manner of Holm and Kleppa are found to be 490°C, 670°C and 1035°C, respectively, for the three different Al/Si distributions. It is apparent from this example that a knowledge of the propagation of errors and the *exact*  $(\overline{T-O})$ -Al relationship are requisite to meaningful configurational entropy calculations.



In conclusion it is suggested that in systematizing structurally-related data for K-rich feldspars, the bulk chemistry and the physical properties—such as birefringence, optic angle and cell parameters—be related directly to the (T–O) dimensions of the individual tetrahedra (see Colville and Ribbe, 1968) and that the Al/Si content predicted from these dimensions be accepted with the foregoing reservations. The structural state of a non-end-member alkali feldspar should only be predicted by extrapolation along alkali-exchange paths to the known feldspar structures (*cf.* Wright and Stewart, 1968). This approach must also be used with caution because different Al/Si distributions are to be expected for a triclinic Na-rich modification and for its monoclinic K-rich alkali-exchange equivalent. Thus if a natural alkali feldspar is triclinic its structural state should be predicted by extrapolation to the nearest triclinic exchange equivalent, not to be the monoclinic equivalent.

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