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WHENCE ORTHOCLASE AND MICROCLINE? A CRYSTALLOGRAPHER'S INTERPRETATION OF POTASSIUM FELDSPAR PHASE RELATIONS*

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ABSTRACT

The widely held view that the potassium feldspar polymorphs constitute an essentially temperature-dependent series (from high-temperature silicon–aluminum (Si–Al) disordered monoclinic (high) sanidine through intermediate-temperature partly ordered monoclinic orthoclase to low-temperature fully ordered triclinic maximum microcline) is questioned: (1) Structurally the Si–Al distribution in orthoclase is not that to be expected between disordered sanidine and ordered maximum microcline; (2) genetically much orthoclase (and closely related adularia) occurs in low- rather than intermediate-temperature settings. Based on a quantitative bond-strength interpretation of the Si–Al distributions in nearly two dozen published potassium-feldspar structures, it is found that such interpretation better accounts for many of the structural and genetic characteristics of the potassium feldspars. This theory has two implications, in the potassium-rich region of the alkali feldspar phase diagram: (1) a temperature-dependent series exists from high-temperature Si–Al disordered monoclinic (high) sanidine to low-temperature partly ordered monoclinic orthoclase, which is thus interpreted as a low-temperature polymorph; (2) triclinic potassium feldspar (microcline) arises through the influence of sodium (or calcium) on the aluminosilicate framework at the (higher-temperature) stage of Si^{4+}–Al^{3+} segregation ("ordering") of the cooling alkali feldspar. Thus, there exists a series of low-temperature potassium feldspars from partly ordered monoclinic orthoclase formed in an original potassium-rich sodium-poor environment, through partly ordered triclinic intermediate microclines to largely ordered triclinic maximum microcline formed in a potassium-poor sodium-rich environment. It follows that in many instances physical–chemical processes operative after Si–Al segregation resulted in the more or less complete separation of microcline from the earlier associated sodium feldspar.

SOMMAIRE

D'après une opinion très répandue, les polymorphes des feldspaths alcalins constitueraient une série dépendant surtout de la température, série passant de la sanidine, de haute température, monoclinique, à désordre Si–Al; — par l'orthose, de température intermédiaire, monoclinique, partiellement ordonnée; — au microcline dit maximum, de basse température, triclinique, complètement ordonné. Cette opinion est ici remise en question parce que: (1) au point de vue structural, la distribution Si–Al dans l'orthose n'est pas celle que l'on s'attendrait à trouver dans une structure de transition entre la sanidine désordonnée et le microcline ordonné; (2) au point de vue génétique, beaucoup d'orthoses (et d'adulaires) se rencontrent dans des conditions de température plutôt basse qu'intermédiaire. En se fondant sur l'interprétation quantitative de la distribution Si–Al dans quelque vingt structures de feldspath potassique, au moyen de la théorie des valences de liaisons, on conclut que pareille interprétation rend le mieux compte de beaucoup d'aspects structuraux et génétiques des feldspaths potassiques. Au voisinage du pôle potassique du diagramme

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de phase, la théorie entraîne deux conséquences: (1) il existe une série, fonction de la température, entre un polymorphe monoclinique totalement désordonné (sanidine de haute température) et l'orthose monoclinique partiellement ordonnée, orthose interprétée ici comme polymorphe de basse température; (2) le microcline est stabilisé sous l'effet du sodium (ou du calcium) sur la charpente anionique de l'aluminosilicate à un stade antérieur du refroidissement, au début de la mise en ordre. Il existerait donc une série de feldspaths potassiques de basse température allant de l'orthose partiellement ordonnée, formée en milieu riche en K et pauvre en Na, au microcline maximum, fortement ordonné, formé en milieu pauvre en K et riche en Na, en passant par des microclines intermédiaires, tricliniques, partiellement ordonnés. Il s'ensuit aussi que, dans beaucoup de cas, des processus physico-chimiques mis en oeuvre après la ségrégation Si–Al auraient comme résultat de séparer plus ou moins le microcline d'un feldspath sodique qui lui était antérieurement associé.

(Traduit par la Rédaction)

INTRODUCTION

Whence orthoclase and microcline? What is the origin of these two closely related members of the feldspars both of which occur as essential and widespread constituents of granitic-type rocks, and which, with sanidine and adularia, make up the potassium feldspars, ideally, $KAlSi_3O_8$? As is well known, sanidine is the high-temperature "quenched" form of potassium feldspar characteristic of volcanic rocks like rhyolites and trachytes, and adularia is a form with wedge-shaped crystals that occurs as a late-stage mineral in vugs. The characteristic of orthoclase and microcline that is perhaps unusual and perplexing is that both commonly occur in petrogenetic settings that are seemingly the same, namely, in deep-seated, slowly cooled granitic or related rocks. Why for a given granitic rock with a particular composition is the potassium feldspar not always orthoclase *or* microcline? For some of us who have wrestled with this problem, we might well ask as the Lord did of Satan (in relation to long-suffering Job): "Whence comest thou?" And sometimes it seems that orthoclase and microcline are giving us the same answer Satan gave the Lord: "From going to and fro in the earth, and from walking up and down in it!"

Perhaps the principal reason the relationship between orthoclase and microcline is still ambiguous despite vast amounts of work on the alkali feldspars is that it has never proved possible to synthesize these two minerals in the laboratory and to establish their stability fields experimentally. Thus, it is necessary to try to deduce their interrelationship by some inductive method that utilizes a crystal–chemical, thermodynamic or other theoretical approach in conjunction with petrological evidence.

Most modern workers attempt to utilize and most modern textbooks describe a set of relationships between orthoclase and microcline based on the distribution of the structurally very similar silicon (Si) and aluminum (Al) atoms in the crystal structures of the different potassium feldspars. Any theory based on the different distributions of these two kinds of atom in the different polymorphs is known as the "order-disorder" theory, although it would be more precise to call it the "Si–Al disorder–order" theory.

The particular theory on which the presently accepted orthoclase–microcline relationships are based was first proposed by Laves (1952a, b) and Goldsmith & Laves (1954a, b). I call this the "Si–Al full-ordering" theory. Like any such theory, this one holds implications with respect to crystal chemistry on the one hand and to petrogenesis on the other, *i.e.*, to the conditions under which orthoclase and microcline could be expected. A few years after the appearance of the Laves & Goldsmith papers, Ferguson *et al.* (1958) and Ferguson (1960) proposed a significantly different set of orthoclase–microcline relationships based on a different theory but one still related to Si–Al distributions. I call this the "bond-strength" theory.

I felt at that time and I still feel today that Laves & Goldsmith's full-ordering theory has certain weaknesses, both crystal–structural and petrological, and that the bond-strength theory is more soundly based. This Presidential Address seemed an appropriate occasion to review both theories and their implications with respect to orthoclase and microcline in the light of today's knowledge of these minerals.

THE POTASSIUM FELDSPARS INTERPRETED IN TERMS OF THE SI-AL FULL-ORDERING THEORY

First, let us consider in its simplest terms the Si–Al full-ordering theory of Laves & Goldsmith, which will subsequently be referred to simply as the full-ordering theory. It is known from X-ray diffraction analyses that, in the crystal structures of the high-temperature forms of these feldspars, sanidine and high albite, the four tetrahedral atoms (the Al and 3 Si of the chemical formulae $KAlSi_3O_8$ and $NaAlSi_3O_8$) are randomly distributed over the four different, or two pairs of different, tetrahedral sites available; *i.e.*, the structures of both these minerals are Si–Al disordered. It is also known that in forms of these feldspars known to be low-temperature, namely, maximum microcline and low albite, the two kinds of atom are largely or completely segregated; most or all of the Al atoms are concentrated into one of the four sites and most or all of the Si atoms into the other three. Laves & Goldsmith in their 1952 and 1954 papers proposed that for both the potassium and sodium feldspars the Si and Al atoms that are disordered in the high-temperature forms would, under ideal conditions of slow cooling, progressively order themselves, ultimately achieving a fully ordered configuration in the presumed low-temperature forms. Thus, in the simpler sodium feldspars, the high-temperature form is (neglecting monalbite) Si–Al-disordered triclinic high albite, and the low-temperature form is, according to this theory, fully ordered triclinic low albite, the familiar albite of many igneous and metamorphic rocks.

In the potassium feldspars, Si–Al disordered monoclinic sanidine is of course the high-temperature form. Because orthoclase has a partly ordered monoclinic structure, it is seen as an intermediate-temperature form. Similarly, because they are partly ordered triclinic, the intermediate microclines are seen as intermediate-to-low temperature forms, and presumed fully ordered maximum microcline is seen as the low-temperature form. The important petrogenetic implication of this crystal–chemical interpretation is that orthoclase is an intermediate-temperature polymorph, and maximum microcline is ideally the only stable low-temperature potassium feldspar.

It is this interpretation of the potassium feldspars to which I take serious exception. My principal criticisms of it are the following: (1) Although a triclinic maximum microcline can be converted to monoclinic sanidine by prolonged heating in the laboratory, it has never been shown that any of the intermediate products has the properties of natural monoclinic orthoclase as this theory suggests should be the case. (2) The Si–Al distributions in several orthoclase or orthoclase-like potassium feldspars determined by crystal-structure analyses do not, in my view, fall logically between the Si–Al distribution in maximum microcline and that in sanidine as one might expect for a transition from one of these structures to the other. (3) Very similarly, the Si–Al distributions in ten structurally analyzed intermediate microclines also do not, in my view, fall logically between the distributions in maximum microcline and sanidine. (4) Monoclinic orthoclase, interpreted here as an intermediate-temperature mineral, has not classically been differentiated from low-temperature triclinic microcline as a constituent of slowly cooled granitic rocks, and present-day petrologists continue to describe monoclinic potassium feldspars, both orthoclases and adularias, occurring in settings that appear to be of low- rather than intermediate-temperature.

THE POTASSIUM FELDSPARS INTERPRETED IN TERMS OF THE BOND-STRENGTH THEORY

An alternative to the full-ordering concept of the potassium feldspars is, as indicated earlier, the bond-strength theory proposed by Ferguson *et al.* (1958) and Ferguson (1960). Details of this theory may be found in those papers. In the present paper I attempt to (1) summarize the general nature of the bond-strength theory and some of its implications, and (2) interpret the Si–Al distributions in about two dozen structurally analyzed potassium feldspars in terms of this theory. Figure 1 is a diagrammatic representation of four alkali feldspar structures which, with its detailed description, is intended to help convey the bond-strength interpretation of these structures.

This interpretation of the alkali feldspars invokes the widely known and applied crystal–chemical principle known as Pauling's second rule relating to inorganic structures, described in such books as Bloss (1971) and Hurlbut & Klein (1977). The essence of this principle is that, in minerals like the feldspars, the atoms are thought of as *ions*, so that a potassium feldspar can be regarded as an assemblage of the cations K^+, Al^{3+} and Si^{4+} and of the anions O^{2-}. Electrostatic charges or bonds are considered to be distributed from the different cations to their surrounding oxygen anions. Pauling's second rule says that in a stable structure each anion such as oxygen would be expected to receive from all its neighboring cations a total bond-strength equal to its negative charge or valence (in the case of oxygen, 2). In the case of the Si^{4+}–Al^{3+} disordered high-temperature form of sodium feldspar, high albite, the simple calculation of the bond strengths to each oxygen shows that they depart somewhat from the ideal of 2.

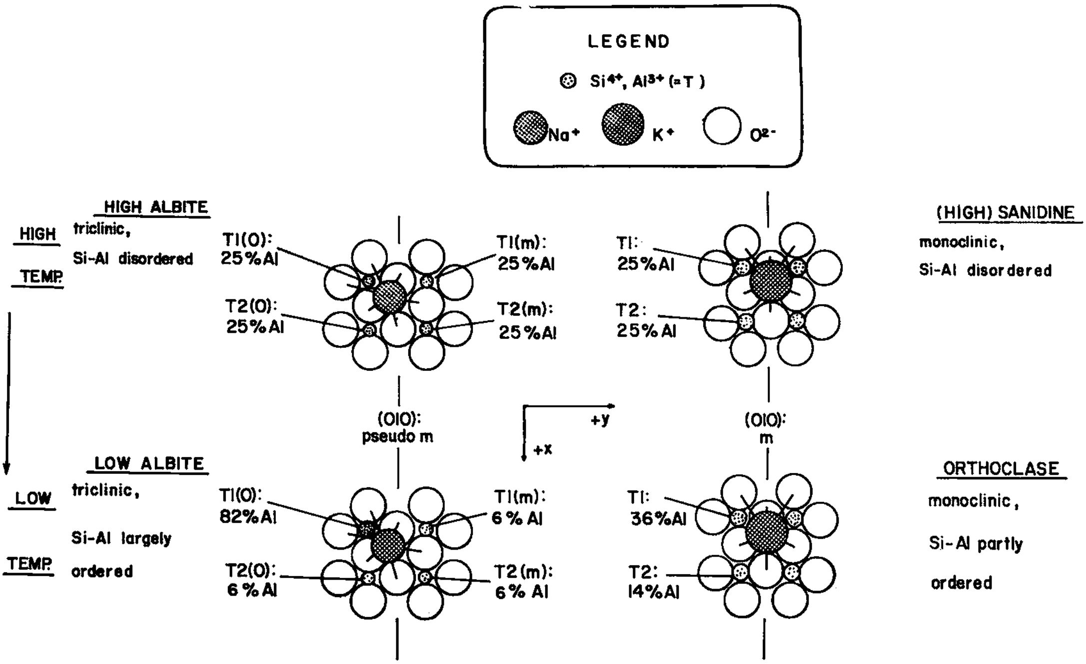

FIG. 1. Diagrammatic representation of the structures of high albite, low albite, sanidine and orthoclase projected onto (001). The lack of planar symmetry *m* parallel to (010) in the two albites resulting in their triclinic character is due to the relatively small size of Na^+ and its coordination by six O^{2-} anions which allows the aluminosilicate framework to collapse around it. In contrast, the presence of *m* parallel to (010) in these two potassium feldspars resulting in their monoclinic symmetry is due to the relatively large size of K^+, which distends the framework, and its coordination by nine or ten O^{2-} anions, and to the symmetrical Si–Al distributions. According to the bond-strength interpretation of these structures, in high albite and sanidine with disordered Si–Al, each *T* site contains statistically $Si^{4+}{}_{0.75}Al^{3+}{}_{0.25}$; consequently each has effectively the same charge and contributes the same bond-strength to its four O^{2-} anions. From the diagram it can be seen that the additional bonds to the O^{2-} anions from Na^+ (solid lines) are, because of the small size of Na^+, directed mainly to the four O^{2-} anions coordinating site *T*1(0). Because of this, as high albite cools slowly, lower charged Al^{3+} migrates from *T*1(*m*), *T*2(0), *T*2(*m*) into *T*1(0) at the expense of higher charged Si^{4+}, which migrates in the opposite direction, until the bond strengths to the O^{2-} anions become ideal. Calculations show that this occurs for a largely ordered (triclinic) low albite with ideally 82% Al in *T*1(0) and 6% Al in the other three sites. In contrast, monoclinic sanidine has a bond distribution that is symmetrical across (010) but with more bonds directed to the O^{2-} anions around *T*1 than to those around *T*2. To offset the excess bond-strengths to the *T*1 oxygens, Al^{3+} ions migrate, with slow cooling, into *T*1, in a symmetrical fashion at the expense of Si^{4+} until the bond strengths to the oxygen anions reach ideality. Calculations show that this corresponds to 36% Al in *T*1 and 14% in *T*2 in a monoclinic structure, "ideal orthoclase" (*cf.*, Ferguson 1960, Fig. 1).

Because the Si^{4+} and Al^{3+} cations are differently charged, if they change in any way from the disordered configuration in high albite, *i.e.*, if they are even partly ordered or segregated in some way, then the bond strengths to each oxygen anion must also inevitably change from their values in sanidine. It can in fact be shown that, if the Si^{4+} and Al^{3+} cations start ordering in a particular way, that is, if the Al^{3+} cations start to segregate into a particular one of the four triclinic tetrahedral sites *T*1(0), *T*1(m), *T*2(0), *T*2(m) (with an accompanying reverse segregation of Si^{4+} cations), then the bond strengths to the oxygens change concurrently closer to the ideal of 2. It can further be shown that in the sodium feldspars the bond strengths to the oxygens achieve the ideal of 2 not for a *complete* segregation of Al^{3+} ions into the one site, *i.e.*, not for a fully ordered structure, but rather for one with only about 82% of the Al^{3+} ions concentrated into that site, the rest of the ions being of course Si^{4+}. Such a structure would have an ideal bond-strength distribution and could be expected to be a stable low-temperature form. The bond-strength theory postulates this as the structure for low albite, and this structure can be described as *largely* rather than fully Si–Al ordered.

Regarding the potassium feldspars, the oxygen anions in high-temperature Si^{4+}–Al^{3+}-disordered sanidine receive, like those in high albite, bond strengths somewhat removed from the ideal of 2. Again, a certain type of segregation of the differently charged Si^{4+} and Al^{3+} cations results in the bond strengths to the oxygens approaching 2 more closely. Now, in this bond-strength interpretation of the alkali feldspars, the fact that Na^+ is relatively small (radius 1.02Å) and K^+ is relatively large (1.55Å) is of fundamental importance, because the manner in which the positive bond-strengths are distributed to their oxygens by Na^+ or K^+ in their respective minerals has a direct bearing on the Si^{4+}–Al^{3+} configuration or ordering pattern that results in ideal bond-strengths to the oxygens, and hence on the presumed stable low-temperature form. Thus, in the sodium feldspars, the fact that ideal bond-strengths to the oxygens are achieved for an 82% Al^{3+}-ordered triclinic configuration is a direct consequence of the relatively small size of Na^+.

In the potassium feldspars, an analysis of the bond strengths relative to disordered monoclinic sanidine reveals that, because of the large size of K^+ relative to Na^+, ideal bond-strengths to each oxygen anion result *not* for Si^{4+}–Al^{3+} fully ordered triclinic microcline but rather for a partly ordered monoclinic structure. Because of its ideal bond-strengths, this partly ordered monoclinic structure could be presumed to be a low-temperature form, and in our original paper we concluded that it was none other than classical monoclinic orthoclase. We called it "ideal orthoclase".

For triclinic microcline to originate in a slowly cooled igneous (granitic) rock, we proposed that a triclinic character is imposed upon it by appreciable amounts of smaller Na^+ ions present with the larger K^+ ions in the homogeneous crystal or possibly even in the melt at the high temperatures at which Si^{4+}–Al^{3+} segregation is presumed to take place. Potassium-rich environments would yield, because of the dominance of the relatively large K^+ ions, a symmetrical monoclinic aluminosilicate framework like that of "ideal orthoclase", whereas sodium-rich environments would yield, because of the dominance of the relatively small Na^+ ions, an asymmetrical triclinic aluminosilicate framework like that of low albite. In the latter case, the resulting K-feldspar, say after exsolution into a perthite at some lower-temperature stage, would be triclinic because of the previously sodium-imposed triclinic Si–Al configuration of the framework. In such a way we proposed plutonic microcline to have arisen. A diagrammatic representation of this proposed origin for microcline (Fig. 1) is similar to that given in Ferguson (1960, Fig. 1).

Much microcline originates in another way that is well established and well known, namely, by potassium replacement of sodium/calcium in a plagioclase, *e.g.*, under conditions of potassium metasomatism. Such replacement has indeed been carried out experimentally: *cf.*, Laves (1951), Wyart & Sabatier (1956a, b), and others more recently. The two feldspar theories discussed in this paper are equally compatible with the replacement origin for much microcline, although the bond-strength theory may suggest that more microcline originates in this way than is now thought to be the case.

GENERAL NATURE OF THE ALKALI FELDSPAR PHASE DIAGRAM IMPLIED BY THE BOND-STRENGTH THEORY

The foregoing crystal–chemical interpretation of the alkali feldspar minerals holds implications for their origin and for their positions within the alkali feldspar phase diagram.

The high-temperature pure potassium feldspar is, of course, monoclinic sanidine. If sanidine cools slowly in a sodium-poor environment, it will, according to this theory, ideally transform to monoclinic *orthoclase as the stable low-temperature form derived from this original sodium-poor environment.* If a potassium feldspar forms from an environment containing appreciable sodium feldspar, say for example, in a 1:1 ratio, the resulting potassium feldspar, after separation by perhaps perthitization, will have a framework halfway between that of monoclinic orthoclase and triclinic low albite, actually slightly triclinic, and such a potassium feldspar would be a triclinic intermediate microcline. And *if the potassium feldspar forms in a sodium-rich environment, then the resulting separated potassium feldspar will have a triclinic framework like that of low albite; this feldspar will be maximum microcline,* whose aluminosilicate framework will be identical to that of low albite.

The bond-strength theory implies then that, rather than maximum microcline being the only stable low-temperature potassium feldspar,

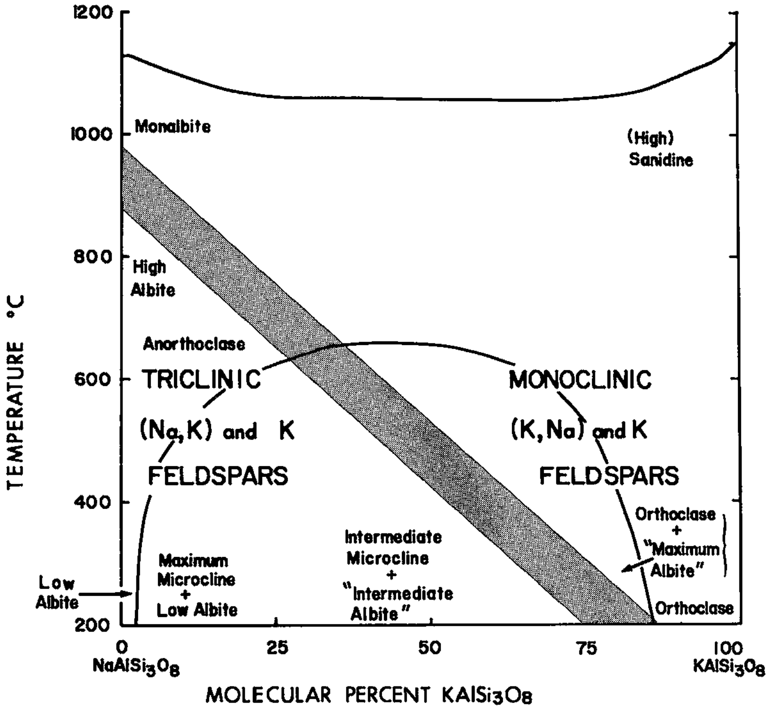

FIG. 2. Alkali feldspar phase diagram to convey the idea of the general regions of triclinic (Na,K)- and K-feldspars (anorthoclase, microclines) and of monoclinic (K,Na)- and K-feldspars (sanidine, orthoclase) implied by the bond-strength interpretation of the structures. All Na-feldspars except monalbite are triclinic. All changes from one phase to a contiguous one (sanidine to orthoclase, high albite to low albite, *etc.*) are regarded as transitional. The solidus and the solvus are from the "dry" system (1 atm) of Bowen & Tuttle (1950). The centre of the triclinic–monoclinic boundary (represented as a band because of the general nature of the diagram) extends from the high albite–monalbite transition temperature (at 1 atm) of 930°C of Okamura & Ghose (1975) to the theoretically derived low-temperature monoclinic orthoclase–triclinic microcline transition composition of the "original environment" of 80% KAlSi$_3$O$_8$ of Ferguson (1960). The terms "intermediate albite" and "maximum albite" are explained in the author's 1960 paper.

there exists a *series of low-temperature potassium feldspars;* monoclinic orthoclase formed in an original potassium-rich environment progresses with increasing sodium:potassium ratio in the original environment through slightly triclinic intermediate microclines to triclinic maximum microcline formed in an original sodium-rich environment.

These proposed phase-relationships for the potassium feldspars are shown in Figure 2, the alkali feldspar phase diagram implied by bond-strength considerations. This general phase-diagram may be contrasted with others based on the Si–Al full-ordering theory, some examples of which are those of Martin (1974, Fig. 2), Smith (1974a, Fig. 5-1) and Hurlbut & Klein (1977, Fig. 10.77). For this discussion of the general potassium-feldspar relationships, the particular solidus and solvus used in the phase diagram of Figure 2 are not important, but in any more detailed discussion these and other characteristics of the phase diagram would assume great importance.

EVIDENCE SUPPORTING THE BOND-STRENGTH INTERPRETATION OF THE POTASSIUM FELDSPARS

I submit the following as the principal crystal-structural and petrogenetic lines of evidence to support the bond-strength interpretation of the potassium feldspars.

Si–Al distributions in orthoclase-like potassium feldspars

The Si–Al distributions determined by crystal-structure analysis for five monoclinic orthoclase or orthoclase–like potassium feldspars are all reasonably close to the Si–Al distribution predicted by Ferguson *et al.* (1958) for "ideal orthoclase" using this theory. In the two different monoclinic tetrahedral sites *T*1 and *T*2, the authors predicted respective statistical numbers of Al atoms as 36% and 14%. Five subsequent X-ray diffraction analyses of orthoclase-like feldspars yielded values varying from 32 to 40% Al in *T*1 and from 11 to 18% Al in *T*2. These Al contents, along with others, are plotted in Figure 3 where solid lines join the Al contents in tetrahedra *T*1 and *T*2 in high-temperature (high) sanidine (*T*1 = *T*2 = 25% Al; Si–Al disordered) to the Al contents predicted by Ferguson *et al.* (1958) in low-temperature "ideal orthoclase" (*T*1 = 36% Al, *T*2 = 14% Al; Si–Al partly ordered). This theory assumes that sanidine–orthoclase constitutes a high-temperature – low-temperature series formed in an original K-rich, Na-poor environment. In this model, any monoclinic potassium feldspar would be expected to have Al contents in *T*1 and *T*2 of or between these two extreme amounts, and any such feldspar with Al contents between the two extremes would be interpreted as an intermediate-temperature form. The results shown in Figure 3 would thus suggest that specimens coded as CRSC, PRa and CRSB could reasonably be regarded as "low-temperature", those coded as BHPSs, Wns, JTSC and PRls as "intermediate-temperature", and those coded as RhSC, Whs and CSK as "high-temperature". I have purposely omitted a temperature scale on Figure 3, but "high-temperature" might reasonably be 800–1000°C depending upon P(H_2O), and "low-temperature" would be ordinary temperature, say, 0°C. Of the orthoclase-like feldspars whose structures have been refined, only the pegmatitic orthoclase refined by neutron diffraction (Prince *et al.* 1973) gave Al contents substantially different from the theoretical and X-ray values, namely, 50% Al in *T*1 and 0% Al in *T*2 (plots coded as PDMn in Fig. 3). The authors called this feldspar "ordered orthoclase", and they proposed an explanation for it. I cannot account for this exception, but it

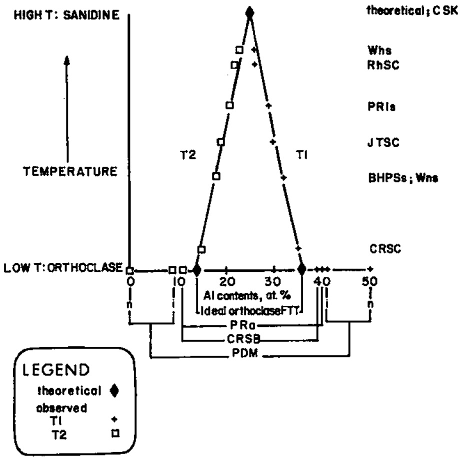

FIG. 3. Plot of Al contents of the tetrahedra *T*1, *T*2 in the presumed high-temperature (high) sanidine–low-temperature orthoclase monoclinic potassium feldspar series against temperature (on an arbitrary scale) according to the bond-strength theory of the structures. These monoclinic potassium feldspars are considered to have formed, according to the bond-strength interpretation of their structures, in an original environment that was K-rich and Na-poor. Solid lines join the Al contents in the tetrahedra derived theoretically for high-temperature (high) sanidine, *T*1 = *T*2 = 25% Al (see Cole *et al.* 1949, point CSK), and the Al contents in low-temperature "ideal orthoclase", *T*1 = 36% Al, *T*2 = 14% Al (Ferguson *et al.* 1958, point FTT). Mathematically fitted to the curves on the temperature axis are the Al contents in monoclinic potassium-rich feldspars derived from structure analyses by Brown *et al.* (1974) of a sanidine (BHPSs); Colville & Ribbe (1968) of Spencer B (CRSB) and of Spencer C (CRSC); Jones & Taylor (1961) of Spencer C (JTSC); Phillips & Ribbe (1973) of low sanidine #7002 (PRls) and of adularia #7007 (PRa); Prince *et al.* (1973) of an orthoclase by neutron scattering (PDMn) and by interatomic distances from the neutron diffraction refinement (PDMi); Ribbe (1963) of heated Spencer C (RhSC); and Weitz (1972) of a natural sanidine (Wns) and the same sanidine heated (Whs). Because, according to the bond-strength theory, no monoclinic potassium-rich feldspar can have a higher degree of Si–Al ordering than that in "ideal orthoclase", namely, 36% Al in *T*1 and 14% Al in *T*2, these values are taken to define the lower limit of the diagram. Of those specimens that have an apparently greater degree of Si–Al order than this, three of the results, PRa, CRSB and PDMi, are close to the theoretical if error and uncertainty in both the experimental and theoretical values are taken into account. Only one result (PDMn) differs appreciably from the predicted; it is discussed in the text.

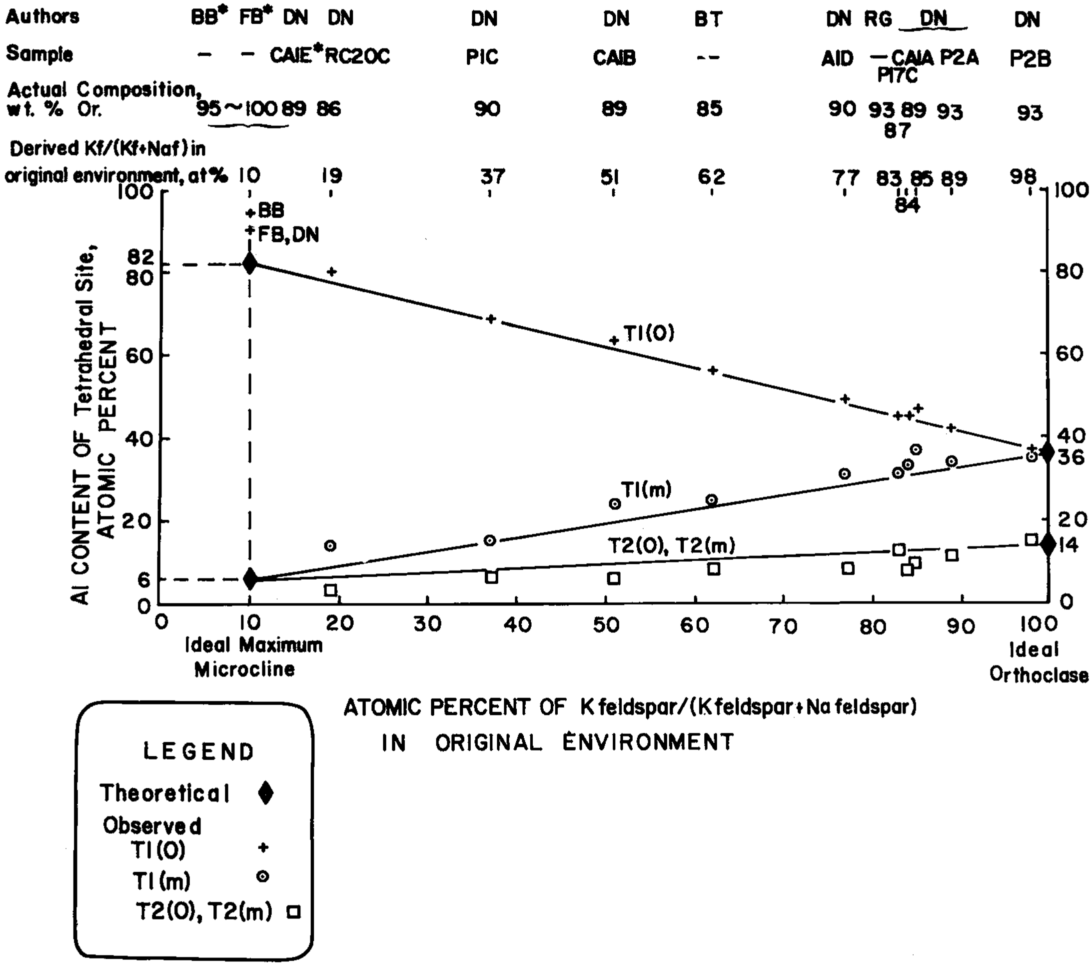

FIG. 4. Plot of Al contents of the tetrahedra *T*1(0), *T*1(*m*), *T*2(0), *T*2(*m*) in the presumed low-temperature potassium feldspar series maximum microcline–intermediate microclines–orthoclase against atomic percent K-feldspar/(K-feldspar+Na-feldspar) in the original environment (at the stage of Si–Al segregation) according to the bond-strength theory of the structures. Solid lines join the Al contents in the tetrahedra *T*1, *T*2 derived theoretically for monoclinic orthoclase (Ferguson *et al.* 1958) to those in the tetrahedra *T*1(0), *T*1(*m*), *T*2(0), *T*2(*m*) interpreted for triclinic maximum microcline (Ferguson 1960, modified by Gait *et al.* 1970). The left-hand side of the diagram has been constructed in a manner which arbitrarily assumes that maximum microcline (whose Si–Al distribution is the same as that of low albite) forms in an original environment of 10% Kf/(Kf+Naf). Mathematically fitted to the theoretical curves are the observed Al contents in intermediate microclines derived from structure analyses by Bailey & Taylor (1955), Ribbe & Gibbs (1975) and Dal Negro *et al.*. (1978), shown as BT, RG and DN, respectively. Plotted on the 10% Kf/(Kf+Naf) vertical axis are the Al contents in maximum microclines BB, FB and DN refined by Brown & Bailey (1964), Finney & Bailey (1964) and Dal Negro *et al.* (1978), respectively. The Al contents in *T*1, *T*2 in monoclinic potassium feldspars that plot on or near the horizontal axis in Figure 3 (orthoclases and adularias) should logically be included on the right-hand vertical axis in this Figure but are omitted for clarity. For each sample are given the actual Or content (in wt. %, from the original authors) and the proportion Kf/(Kf+Naf) in the original environment (in at. %), as deduced from the fitting of the Al contents to the curves. For maximum microclines BB*, FB* and DN (sample CAIE*), the al contents in *T*1(*m*), *T*2(0), *T*2(*m*) have not been plotted (for clarity); they are, respectively: 3, 1, 2; 2, 6, 0; and 5, 3, 3.

is worth noting that the Al contents in even this orthoclase deduced by the authors from the mean tetrahedral cation oxygen interatomic distances (as distinct from neutron diffraction), namely, $T1 = 41\%$ Al, $T2 = 9\%$ Al (plots coded as PDMi in Fig. 3), are reasonably close to the predicted.

Si–Al distributions in intermediate microclines

In my view, the second line of evidence supporting this theory is that the Al contents in the four tetrahedral sites in the ten intermediate microclines that have been refined fall logically between the Al contents in the end-members of the proposed low-temperature series, namely, orthoclase–maximum microcline corresponding to K-rich – increasing Na:K – Na-rich environments at the time of Si–Al segregation. Furthermore, an inherent feature of this interpretation is the derivation, from the crystal structure or geometry of a particular intermediate microcline, of the K-feldspar/(K-feldspar + Na-feldspar) ratio of the original environment.

The Al contents in the four triclinic tetrahedra $T1(0)$, $T1(m)$, $T2(0)$, $T2(m)$ of the ten intermediate microclines (and of three maximum microclines) are shown in Figure 4. These are mathematically fitted to solid lines that show the variation (assumed to be straight-line) in Al contents of the four tetrahedra between the end-members of the assumed low-temperature series orthoclase–maximum microcline *versus* the atomic percent K-feldspar/(K-feldspar+Na-feldspar) in the original environment. Sources of both the theoretical and the experimental data are given in the description of Figure 4.

Figure 4 shows that the Al contents in the four tetrahedra in the ten refined intermediate-microcline structures can be fitted in a reasonable way to the theoretical curves. Figure 4 also shows (across the top) the derived atomic percent Kf/(Kf+Naf) in the environment at the time of Si–Al segregation in relation to the actual Or content (in wt. %) quoted by the original authors. Only for those specimens which are slightly triclinic (those plotting near the right-hand side of the diagram) are the derived composition for the original environment and the actual composition roughly the same. In general, the more triclinic the microcline (the further to the left on the diagram), the greater the gap between the two compositions. This interpretation of these structures suggests that the greater the proportion of Na-feldspar:K-feldspar in the original environment, the lesser the Na-feldspar proportionally retained in the resulting K-rich feldspar. The extreme case is represented by maximum microclines which would have formed in a Na-rich environment, here arbitrarily taken to be 90 at. % Naf/(Kf+Naf), whereas many maximum microclines actually contain <10 % Na-feldspar. Because many granitic rocks containing maximum microclines do not show a great predominance of sodium-rich plagioclase over the microcline, this interpretation implies that some petrogenetic process must have operated between the Na–feldspar-rich stage of Si–Al segregation in the alkali feldspar(s) and the final stage where the K-rich and the Na-rich feldspars coexist, perhaps in nearly equal amounts.

Common occurrence of orthoclase and adularia in low-temperature environments

The third line of evidence is the general petrological one, that monoclinic potassium feldspar (orthoclase or adularia) commonly occurs, contrary to the expectations of the full-ordering theory, under conditions that can only be reasonably construed as low-temperature. The discrepancy between the occurrence of these monoclinic K-feldspars and that to be expected according to the full-ordering theory has resulted in several explanations to try to account for the existence of this seemingly metastable orthoclase or adularia. The types of occurrence that fall into this category are late-stage hydrothermal orthoclase or adularia, authigenic orthoclase or adularia and plutonic orthoclase in granites. Following are the three principal explanations that have been proposed to account for one or another of these presumed metastable occurrences of orthoclase or adularia (or both), and all assume the fundamental premise of the full-ordering theory that monoclinic orthoclase inverts with cooling to triclinic microcline at a temperature generally agreed to be about 500°C.

The first of these explanations is that the feldspar crystallized in the presumed stability field of microcline, *i.e.*, below the orthoclase–microcline inversion temperature, but at such a rapid rate that metastable orthoclase rather than stable microcline was formed. This explanation has been invoked by, among others, Laves (1952a) for adularias generally, by Baskin (1956) for authigenic orthoclases and by Marmo (1971) for some granitic orthoclases.

The second explanation, the one most frequently suggested, is that the potassium feldspar crystallized in the stability field of orthoclase above the inversion temperature, and then subsequently cooled too rapidly to allow the Si–Al atoms to segregate into the ordered configuration of microcline. Laves (1952a) proposed this or the first explanation to account for orthoclase in granitic rocks, as did Marmo (1971), both authors stressing the importance of time in the inversion process. Prince *et al.* (1973) invoked a slightly modified version of this mechanism to account for the occurrence of the late-stage hydrothermal orthoclase referred to earlier, on which they carried out their structure analysis by neutron diffraction.

The third explanation is that of Parsons (1978), who argues strongly (p. 1) that "... the presence of aqueous fluids during the cooling of the feldspar from magmatic temperatures ... is pre-eminently the factor that dictates the nature of the feldspar in all but the smallest of intrusive igneous bodies ..." and, as he explains later (p. 13), this includes the persistence of orthoclase in plutonic rocks.

The bond-strength theory suggests a different explanation for the occurrence of orthoclase in granitic rocks, one that differs in principle from the above three that are based on the premise that orthoclase is an intermediate-temperature form existing metastably in a low-temperature setting. The bond-strength theory says that, if for an orthoclase existing in a granitic rock, the environment at the time of Si–Al segregation could reasonably be construed as having been potassium-rich and sodium-poor, then this is the potassium feldspar to be expected as the low-temperature form under those circumstances.

IN CLOSING

In view of the extreme variability and complexity of the potassium and sodium feldspars so completely described by Smith (1974a, b), any general theory of these minerals such as the two crystal–chemical theories discussed in this paper will inevitably have certain real or apparent weaknesses, and I acknowledge concerns about certain aspects of the bond-strength theory. My principal concern is that it is difficult in some cases to see how a particular orthoclase could have formed in a potassium-rich environment or a particular microcline in a sodium-rich environment. In spite of this, however, I feel that the detailed crystal–structural characteristics as well as certain petrological features of these minerals provide strong evidence for the bond-strength interpretation of the alkali feldspars and hence for the origins proposed for orthoclase and microcline. In this paper I am suggesting that, in seeking an explanation for the occurrence of orthoclase or microcline in particular geological settings, consideration should be given to a factor not yet seriously taken into account, namely, the possible potassium-feldspar:sodium-feldspar ratio of the environment at the time of Si–Al segregation. It is my hope that such an approach might help answer the question, "Orthoclase and microcline, whence comest thou?"

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