THE LOW-TEMPERATURE PHASES OF THE ALKALI FELDSPARS AND THEIR ORIGIN

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ABSTRACT

New relationships among the low-temperature alkali feldspars were deduced in part and implied in whole by recent crystal structure analyses of low and high albite and consideration of the electrostatic charge distributions in all the known alkali feldspar structures by Ferguson, Traill & Taylor. In general the most stable low-temperature forms are not those most highly ordered with respect to Al-Si as suggested by Goldsmith & Laves, but those most highly balanced electrostatically: slightly ordered orthoclase and largely ordered low albite. Because of the mobility of the alkali atoms at higher temperatures, appreciable amounts of Na in a cooling K-Na feldspar tend to make the Al-Si framework triclinic so that, after exsolution, the K component is triclinic microcline. There thus exists a series of low-temperature K feldspars varying from orthoclase formed in the presence of relatively little Na, through intermediate microcline to maximum microcline formed in the presence of considerable excess of Na over K. It is shown that the (dimensional) monoclinic to triclinic change in the low-temperature K feldspars occurs at an original alkali feldspar composition of about 20% $NaAlSi_3O_8$. Intermediate microclines very similar to the low-temperature forms may also result from processes involving rapid cooling from intermediate temperatures of K feldspars in which Na again played an earlier essential role.

The theory also predicts an analogous series of low-temperature Na feldspars varying from ordinary low-temperature albite formed in the presence of relatively little K, through low-temperature "intermediate albites" to a "maximum albite" formed in the presence of considerable excess of K over Na. Evidence for the existence of such a series is discussed. Intermediate albites formed by rapid cooling from intermediate temperatures apparently also exist.

The ionic picture of the alkali feldspars thus leads to the following low-temperature phases on the phase diagram: orthoclase, orthoclase + maximum albite, intermediate microcline + intermediate albite, maximum microcline + low albite, low albite. Some of the many implications of the theory are discussed.

INTRODUCTION

In a recent paper describing the crystal structures of low-temperature and high-temperature albites, Ferguson, Traill & Taylor (1958) developed some new ideas regarding the long-disputed phase relationships of the alkali feldspars which are different in important respects from the fairly widely accepted ideas of Goldsmith & Laves (1954*a*, *b*). The new theory has broad implications for the mineralogy and petrology of the alkali feldspars as a whole, and the writer intends to consider them in some detail at a later date. In the meantime, it is desirable to bring to the attention of mineralogists and petrologists generally the main ideas of the new theory which originated from crystal-structural studies. The writer also wishes to present further evidence in support of the ideas of

Ferguson, Traill & Taylor (1958) and to consider briefly some of the many implications. In a later paper, Ferguson, Traill & Taylor (1959) clarify certain points and answer criticisms of their first paper made by MacKenzie & Smith (1959). An abstract giving the essentials of the present ideas has already been published by the writer (Ferguson, 1957).

The relationships between the different alkali feldspars rest upon the exact distributions of the Al and Si atoms which occupy structurally equivalent positions in these minerals. It is thus necessary to understand the possible distributions of these two kinds of atom in the alkali feldspars before the ideas can be developed in detail. For this reason, a general description of the alkali feldspar structures with particular reference to the possible Al-Si distributions is given first.

THE ALKALI FELDSPAR STRUCTURES

The general features of the feldspar structures were described first by Taylor (1933) and by Taylor, Darbyshire & Strunz (1934) and the description is readily available in Bragg (1937). Each (Al, Si) atom is surrounded tetrahedrally by four oxygen atoms, and each oxygen is linked to two (Al, Si) atoms to form a three-dimensional framework characterized by distinct zig-zag chains running parallel to the *a* axis of the crystal. These chains form large cavities in the structure which are occupied by the large alkali or alkaline earth atoms. In the early structure analyses by Taylor and his colleagues, no attempt was made to distinguish between the very similar Al and Si atoms. However, Barth (1934) pointed out that the well-known polymorphism of the K feldspars probably results from different distributions of the Al and Si atoms among the tetrahedral sites in the different structures. It is with reference to the exact distributions of these atoms that the ideas of Ferguson, Traill & Taylor (1958) differ from those of Goldsmith & Laves (1954*a*, *b*) and lead them to a set of phase relationships different from those of the latter authors.

The conventional unit cell of all alkali feldspars contains $4[(K,Na)AlSi_3O_8]$, and there are thus 4 Al + 12 Si atoms to be accommodated in each unit cell. In the triclinic members of the group, the microclines and the albites, the symmetry is such as to repeat any one atomic position three other times to make it a 4-fold position, and therefore four such 4-fold positions are required to accommodate the 4 Al + 12 Si atoms in any one unit cell. In the monoclinic members, orthoclase and sanidine, the higher symmetry results in each position being an 8-fold position, and thus only two of these 8-fold positions are required in the monoclinic members to accommodate the 4 Al + 12 Si atoms. The triclinic and monoclinic members are all very closely related to each other structurally despite their difference in symmetry, and two of the 4-fold positions in the triclinic members correspond to one of the 8-fold positions in the monoclinic members. Different symbols are used to designate the four triclinic or the two monoclinic sites. Table 1 gives the designations used by Taylor and his colleagues in the original papers, a recently improved designation suggested by Megaw (1956), and a still more recent designation suggested to the writer by Dr. Megaw in a personal communication. In this last designation T stands for "Tetrahedral," and because it implies nothing about the presence of Al or Si to the exclusion of the other, the writer prefers this to the previous two, and he therefore uses it throughout this paper.

TABLE 1. DIFFERENT DESIGNATIONS OF THE TETRAHEDRAL SITES IN THE ALKALI FELDSPARS

	Taylor, Darbyshire, & Strunz, (1934)	Megaw (1956) (in brief)	Megaw (personal communication, 1958) (in brief)
Monoclinic 8-fold	Si_1	Si_1	T_1
	Si_2	Si_2	T_2
Triclinic 4-fold	Si_1	$Si_1(0)$	$T_1(0)$
	Si_1'	$Si_1(m)$	$T_1(m)$
	Si_2	$Si_2(0)$	$T_2(0)$
	Si_2'	$Si_2(m)$	$T_2(m)$

Laves (1950) has summarized the various ways of distributing the Al and Si atoms throughout these different sites in the monoclinic and triclinic cases. Thus in the triclinic case, all the Al might go into site $T_1(0)$ and all the Si into the other three sites; or all the Al might go into site $T_1(m)$ and the Si into the other three sites, etc. It is important to note, however, that a given site need not contain only Al or Si—it may be statistically occupied by a certain proportion of both kinds of atom. For example, Bailey & Taylor (1955) found the following distribution in their intermediate microcline: .56 Al in $T_1(0)$, .25 Al in $T_1(\mathrm{m})$, .08 Al in $T_2(0)$ and .07 Al in $T_2(m)$.[1]

It is now well established that in the high-temperature forms of the alkali feldspars, sanidine (or "high sanidine")[2] (Cole, Sörum & Kennard, 1949) and high-temperature albite (Traill in Ferguson, Traill & Taylor,

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[1] It is convenient to refer to the proportion of only Al in a particular site and the remainder must be Si. Thus site $T_1(0)$ in Bailey & Taylor's microcline contains .56 Al and .44 Si, site $T_1(m)$ .25 Al and .75 Si, etc.

[2] The term "high sanidine" was proposed by Tuttle (1952*a*) to describe K-rich alkali feldspars in which the optic plane is parallel to the symmetry plane in order to distinguish them from "normal" sanidine with low $2V$ and the optic plane perpendicular to the symmetry plane. The former are probably more highly disordered with respect to Al-Si than the latter. After prolonged heating at high temperatures most natural K-rich alkali feldspars are probably high sanidine, but in this paper all high-temperature K-feldspars are called simply sanidine.

1958) the Al and Si atoms are randomly distributed, with each of the two 8-fold sites in the former and each of the four 4-fold sites in the latter occupied by .25 Al. The reason for this complete disordering of the Al and Si atoms is generally accepted to be the thermal vibration of the Al-Si atoms at high temperatures which keeps them moving from site to site and prevents them from segregating to particular sites which they will do at lower temperatures in response to specific bonding requirements. It is with respect to the nature of the Al-Si segregation that accompanies the slow cooling of a sanidine or a high albite that the ideas of Ferguson, Traill & Taylor (1958) diverge from those of Goldsmith & Laves (1954*a*, *b*).

THE "FULL-ORDERING" THEORY OF GOLDSMITH AND LAVES

Goldsmith & Laves (1954*a*) have drawn an analogy between the behaviour of the Al and Si atoms in the K feldspars and the metal atoms in the alloys Cu_3Au and CuZn. In the latter compounds there is complete disorder of the atoms at high temperatures, but with cooling, the two kinds of atom proceed to order themselves until, at the lowest temperatures, the two kinds of atom are fully ordered. Applying this idea to the K feldspars Goldsmith & Laves (1954*a*, *b*) have concluded that there is a progressive change in the Al and Si atoms from complete disordering in sanidine to complete ordering in the most triclinic of the microclines, maximum microcline, in which all the Al will be in one tetrahedral site and all the Si in the other three tetrahedral sites of the triclinic structure. They thus recognize only sanidine and maximum microcline as stable phases of the K feldspars, and they therefore had to discard as a stable polymorph the long-accepted low-temperature monoclinic phase of K feldspar, orthoclase. By analogy they concluded that in the Na feldspars there would be a similar graduation of the Al and Si atoms from complete disorder in high albite to complete order in low albite. The writer believes that there is no sound crystal-chemical basis for drawing an analogy between the behaviour of certain of the atoms in a silicate and all the atoms in an alloy. Ferguson, Traill & Taylor (1958) have pointed out weaknesses in the attempt of De Vore (1956) to explain why maximum microcline should be fully ordered with respect to Al and Si. Furthermore, neither Goldsmith & Laves (1954*a*, *b*) nor De Vore (1956) have attempted to predict or explain into which particular tetrahedral site the Al can be expected to migrate.

LOW-TEMPERATURE ALBITE

The first experimental evidence that strengthened the writer's doubts about the correctness of the full-ordering theory was his finding that the typical low-temperature albite whose structure he refined has not all, but only about three-quarters, of the Al concentrated into one tetrahedral site. An obvious theory to consider in seeking an explanation for the lack of full Al-Si ordering and for the particular Al distribution in low albite was the ionic theory, and with Traill and Taylor, he therefore worked out the electrostatic charges contributed to the oxygens in both low albite and high albite. When we did this, we found that the particular Al-Si distribution which would give rise to the most highly balanced charge distribution in low albite is one with 75% of the Al concentrated in site $T_1(0)$ and the remaining 25% distributed equally among the other three sites. Within the accuracy of our 2-dimensional Fourier analysis, our low albite has this distribution. We found further that the charge unbalance is much less in low albite (for a reasonable co-ordination of the Na atom) than it is in high albite, and this suggests a reason why high albite tends to invert to low albite under the proper cooling conditions. Finally when we sought an explanation for the concentration of most of the Al into the particular site $T_1(0)$, we found that the charge distribution in high albite is such as to tend to make Al^{+3} migrate into this particular site rather than into any of the other three sites in order to give the oxygens a more satisfactory charge. These observations then, suggested to us that the atoms in the Na feldspars behave as ions, and that the Al-Si distribution which low albite adopts is the one which tends to make the structure most highly balanced electrostatically. We thus tentatively equated stability at low-temperature with electrostatic neutrality of the crystal.

All the detailed figures leading to these conclusions are given in the paper by Ferguson, Traill & Taylor (1958). The only figures that enter into the later discussion of this paper are the particular Al-Si contents of the tetrahedra in the theoretical low albite with the most highly balanced charge distribution, a distribution which our analysed low albite probably has. These figures, along with others, are given in Table 2

TABLE 2. AL-SI CONTENTS (GIVEN AS AL-CONTENTS) OF ALKALI FELDSPAR STRUCTURES

Monoclinic 8-fold sites		T_1			T_2		Degree of order or disorder
Triclinic 4-fold sites	$T_1(0)$		$T_1(m)$	$T_2(0)$		$T_2(m)$	
High albite	.25		.25	.25		.25	Completely disordered
Low albite* Maximum microcline*	.75		.08	.08		.08	Largely ordered
Sanidine		.25			.25		Completely disordered
Ideal orthoclase* "Maximum albite"*		.36			.14		Slightly ordered
Intermediate microcline of Bailey & Taylor (1955)	.56		.25	.08		.07	Partially ordered

*Deduced theoretically. See detailed discussion in text.

and shown with diagrammatic representations of the structures in Figure 1. It is also helpful in following the argument below to realize that in the cooling crystal, the Al^{+3} and Si^{+4} ions migrate in such a way as to offset the charges distributed by the Na^{+} ions. Thus the Al-Si distribution of low albite is in a sense characteristic of Na^{+} in (probably) 6-co-ordination. One would therefore expect a different Al-Si distribution for a balanced K feldspar because K^{+} is larger and probably has 9-co-ordination in the feldspars.

ORTHOCLASE AND MICROCLINE

The Most Balanced K-Feldspar: Orthoclase

If electrostatic charge controls the Al-Si distribution in the stable low-temperature Na feldspar, then it seems reasonable that it should also control the Al-Si distribution in the corresponding K feldspar, and in fact the success with which the ionic theory can be applied to the K feldspars is a further test of its validity for these minerals. We then calculated the charges contributed to the oxygens in the two K feldspars whose structures have been refined, sanidine (Cole, Sörum & Kennard, 1949) and an intermediate microcline (Bailey & Taylor, 1955). Both structures have an appreciable charge unbalance, but we found somewhat to our surprise that microcline has the greater charge unbalance. It was an easy matter to work out from these structures the symmetry and the particular Al-Si distribution that would give rise to a perfectly balanced charge distribution in a pure K feldspar. We did this and found that the most balanced structure is one that is monoclinic and slightly ordered[3] with respect to Al and Si, having .36 Al in site T_1 and .14 Al in site T_2. We naturally concluded that this is the long-accepted low-temperature monoclinic form of K feldspar, orthoclase. We call this as yet theoretical K feldspar "ideal orthoclase." Its Al-Si distribution is given in Table 2 and shown diagrammatically in Figure 1, and this distribution is then the one characteristic of K^{+} in 9-co-ordination.

We were therefore led to the conclusion that a slowly cooling pure sanidine will retain its monoclinic symmetry and adopt as the stable low-temperature form the electrostatically balanced, slightly ordered orthoclase structure.

The Exsolution Origin of Microcline

Satisfactory as our deductions were to that point, we were left with microcline to be explained, and further thought led us to an origin for

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[3] The writer uses the term "slightly ordered" in order to emphasize the small difference between this Al-Si distribution and that in completely disordered sanidine. The difference between the two is such that it could be just distinguished by a careful 2-dimensional Fourier structure analysis.

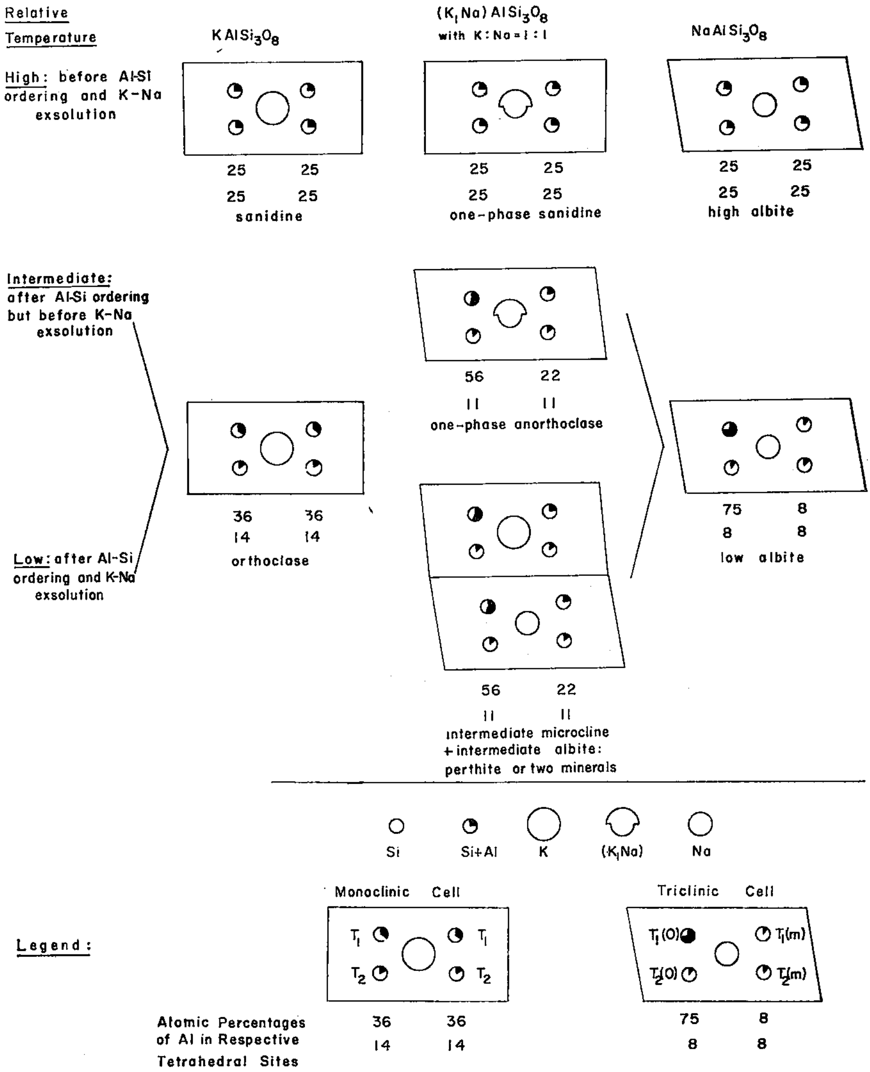

FIG. 1. Diagrammatic representation of the principal alkali feldspar structures on the assumption that the bonding in the low-temperature forms is ionic.

this mineral that seems to us to follow almost naturally from two well-known facts about the alkali feldspars: first, the ease with which the K and Na atoms can move through an alkali feldspar framework (Spencer, 1937), and second, the great difficulty in moving the Al and Si atoms of the framework, several workers having shown that it takes many days at high temperature to invert a microcline or an orthoclase to sanidine.

If then, keeping these two facts in mind, we envisage the cooling of an alkali feldspar containing appreciable amounts of both K and Na, at the relatively high temperatures at which Al-Si ordering takes place, the K and Na atoms will almost certainly be randomly distributed throughout all the large cavities in the feldspar. Since, as we have shown for the pure K and Na members, the Al-Si distribution in the framework is in effect a consequence of the kind of large atom in the structure, the nature of the framework in the alkali feldspar with appreciable amounts of both K and Na should be determined by the relative amounts of these two atoms present in the structure at the time of Al-Si ordering. If the alkali atoms are mostly K, the Al-Si ordering should approximate to that for our slightly ordered monoclinic "ideal orthoclase," whereas if Na predominates, the Al-Si ordering should be more like that of largely ordered triclinic low-albite; for more nearly equal proportions of K and Na, an ordered arrangement intermediate between the two would be expected. Presently some point in the cooling process must be reached where the Al-Si atoms can no longer migrate, and presumably later at some still lower temperature and for some reason not yet understood, perthitization takes place with the segregation of the K atoms into one part of the framework and of the Na atoms into another part. In such a structure the K-rich component of a perthitic microcline will show unbalance of charge since the K^{+1} ion is within an Al-Si distribution appropriate to the average K-Na cation of the high-temperature structure. (Similarly the Na-rich component will show unbalance of charge. This matter is discussed below.) If the cooling alkali feldspar contained sufficient Na initially, then the Al-Si distribution in the framework will be a triclinic one, and this triclinic symmetry will persist in the K-rich component of the perthite forming a microcline. In such a way we believe many triclinic microclines with an appreciable charge unbalance to have formed. (Another closely-related mode of origin for microcline is described below.) This origin for microcline is illustrated diagrammatically in Figure 1.

The Low-Temperature K Feldspar Series

This theory thus implies that there will be a series of low-temperature K feldspars varying from monoclinic orthoclase formed by the slow cooling of a sanidine in the presence of little or no Na, through a series of intermediate microclines, to maximum microcline with the greatest triclinicity formed by the slow cooling and eventual exsolution of an alkali feldspar with a great excess of Na over K, say with Na: K $\geqslant$ 9:1.[4] This theory further implies that a maximum microcline will have virtually the

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[4] This approximate ratio was arrived at by taking account of the Δ value of Goldsmith and Laves (1954*a*); the application of their Δ value to the present ideas is given below.

same Al-Si distribution as low-temperature albite because, in a cooling alkali feldspar that has a great excess of Na, the relatively small amount of K present will have a negligible effect on the Al-Si distribution in the framework. Therefore the Al-Si distribution for maximum microcline given in Table 2 is that of low albite.

The Na Content of Orthoclase and Microcline

Although the present theory implies that the monoclinic or triclinic nature of a low-temperature K feldspar is directly related to the Na content at the time of Al-Si ordering, no sympathetic relationship need exist between the Na content of a perthite *specimen* and the symmetry of its K component, for the following reason. There is very good petrological evidence that the process of exsolution may extend beyond the perthite stage to give a fairly pure K feldspar (frequently microcline) and a fairly pure Na-feldspar existing side-by-side in the rock. Faessler & Tremblay (1946) and Tuttle (1952*b*) have described K feldspars occurring in this way which they believe have resulted from complete exsolution. Their observations thus suggest that in at least some orthoclases and microclines no correlation is to be expected between the Na content of the particular mineral and its monoclinic or triclinic symmetry. Regarding the amount of Na held *in solid solution* in an orthoclase or a microcline, certain characteristics of perthites such as the rarity of antiperthites and the coarseness of many microcline-perthites compared with orthoclase-perthites[5] suggest that the exsolution forces, whatever they may be, are greater for an alkali feldspar with an initial high Na content than for one with an initial high K content. If this is so, one would expect more complete exsolution in the case of initial high Na, and this would explain why in general, microclines contain in solid solution *less* Na than orthoclases, an observation which MacKenzie & Smith (1959) suggest contradicts the present theory.

The Al-Si Distribution in Intermediate Microcline

Our theory for the origin of orthoclase and microcline implies that there should be a regular gradation in the Al-Si distribution from that in ideal orthoclase corresponding to no Na present originally, to that in maximum microcline corresponding to a great excess of Na over K originally (see Table 2 and Figure 1). Figure 2a shows curves giving the variations in Al contents in the four tetrahedral sites against the atomic proportion of Na:(K+Na)(or Na feldspar: (K+Na) feldspar) in the original mineral

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[5] With the help of John C. Davies the writer has *x*-rayed 12 coarse pegmatitic perthites from different localities, and in all of them the K feldspar component is maximum microcline.

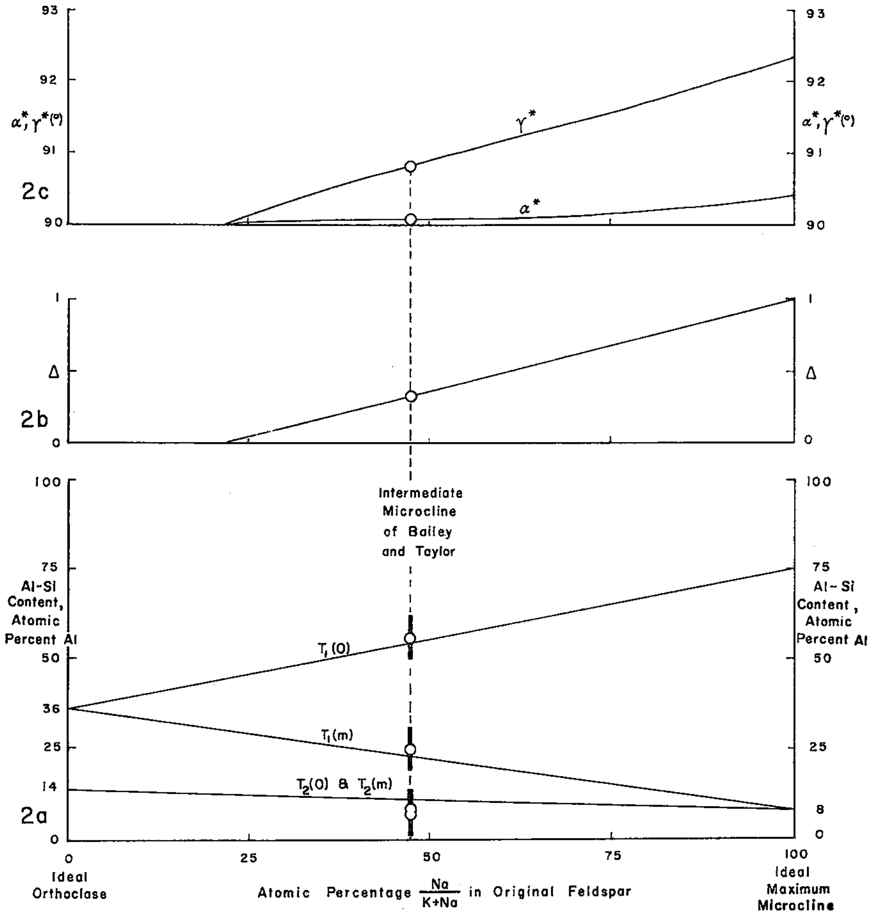

FIG. 2. Variations of the following properties of low-temperature K feldspars with the alkali composition of the original feldspar before exsolution:
2a: the Al-Si contents of the tetrahedra;
2b: $\Delta = 12.5\ (d(131) - d(1\bar{3}1))$ of Goldsmith and Laves (1954*a*) but modified in meaning as described in the text;
2c: α^* and γ^* of microcline.

before exsolution. In this figure straight-line relationships are assumed; in actual fact these may not be exactly straight-line, but there seems no reason for believing they should not be nearly so.

Now no refinements of orthoclase or maximum microcline structures have yet been published in order to prove or disprove the theoretical Al-contents in the extreme members of this series, but an intermediate microcline, that of Bailey & Taylor (1955), has been refined, and a further test of the ionic theory for the alkali feldspars is whether it offers a reasonable explanation for the Al-Si distribution observed by Bailey & Taylor in their intermediate microcline. Ferguson, Traill & Taylor (1958) showed that in a qualitative way it does. Figure 2a shows that, when account is taken of their standard deviation of error in the tetrahedral sizes and hence in the Al-contents of the four tetrahedra, the Al-Si distribution fits very nicely on the theoretical curves, the best fit occurring at an original alkali feldspar composition of 47% $NaAlSi_3O_8$.[6] The ionic theory thus indicates that this particular microcline is a low-temperature form of K feldspar which has resulted from the exsolution of an alkali feldspar which, at higher temperatures before exsolution, contained 47 atomic per cent Na feldspar and 53 atomic per cent K feldspar. The chemical analysis quoted by Bailey & Taylor (1955) from Spencer (1937) for this specimen shows that it contains only about 14 atomic per cent (Ab + An).[7] The present theory thus suggests that in this particular microperthite most of the Na must have migrated outside the K feldspar "host" during the exsolution stage.

The full ordering theory of Goldsmith & Laves (1954*a*, *b*) implies to the writer that there should be a regular gradation in Al-Si content from 25% Al in the two tetrahedral sites of completely disordered monoclinic sanidine, to 100% Al in one and 0% Al in the other three triclinic sites of maximum microcline, the gradation corresponding to a change from the highest temperature to the lowest temperature forms. It can be readily shown that the particular Al-Si distribution observed by Bailey & Taylor (1955) for their intermediate microcline will not fit onto a set of curves of this kind if the relationship is again straight-line or nearly so. The agreement of the ionic picture of the alkali feldspars with Bailey & Taylor's results and the disagreement of Goldsmith & Laves' implied ideas with these results are presented by the writer as further evidence in support of the theories proposed by Ferguson, Traill & Taylor (1958, 1959).

Goldsmith and Laves' Δ Value and the Present Theory

Figure 2a shows that the smallest amount of Na present with K in a cooling alkali feldspar tends to cause different amounts of Al to segregate into the two parts of the one monoclinic 8-fold site T_1, giving rise to the two 4-fold triclinic sites $T_1(0)$ and $T_1(m)$, and thus tending to make the framework triclinic and the resulting K phase a microcline. However, it

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[6] The best fit has been arbitrarily taken on the two upper curves because their slopes are so much greater than the slopes of the two identical lower curves. The best mathematical fit on all four curves occurs at 49% $NaAlSi_3O_8$.

[7] Throughout this paper no account is taken of the effect of any Ca present. In most perthites very little Ca is present, and its effect is probably negligible.

is to be expected that some appreciable proportion of Na to total alkali feldspar would be necessary before the framework would become sufficiently distorted to make the final K feldspar noticeably triclinic. An estimate of the proportion of Na feldspar required to do this can be arrived at in the following way. Goldsmith & Laves (1954*a*) have shown that, when a maximum microcline is heated at high temperatures for a long period, there is a progressive change from triclinic to monoclinic symmetry, and they have described a very simple way of measuring this change of symmetry from the *x*-ray powder photographs. They have set up a Δ value as a measure of the triclinicity where Δ equals 1 for maximum microcline with the greatest triclinicity and, of course, 0 for a monoclinic structure. Δ is very simply related to the spacings in Å of two of the prominent *x*-ray reflections, thus; $\Delta = 12.5\ (d(131) - d(1\bar{3}1))$. Our theory suggests that intermediate microclines which occur in slowly cooled igneous rocks such as many granites and many pegmatites are true low-temperature forms and do not represent a temperature-dependent transition between maximum microcline and sanidine as Goldsmith & Laves suggest, but rather a "compositional" transition between maximum microcline and orthoclase dependent upon the Na content at the time of Al-Si ordering. If this is so, then one can plot as a part of Figure 2b a curve for Δ against original Na: (K+Na) with a value of one for maximum microcline at the right-hand side. It is then a simple matter to calculate Δ for Bailey & Taylor's intermediate microcline using their very accurate cell dimensions. This comes to .33, and this point can be plotted at the composition previously deduced for this microcline from its Al-Si distribution, 47% Na feldspar. If this point and that for $\Delta = 1$ are joined by a straight line, it intersects the abscissa at an alkali composition of 21.6% Na feldspar. Let us call this 20%. This seems to the writer a reasonable value for the proportion of original Na feldspar required to make the resultant K feldspar noticeably triclinic, and in the writer's opinion this provides still further evidence in support of the ionic theory for the alkali feldspars. This Δ curve can be conveniently used with the *x*-ray power data of a microcline from a slowly cooled rock to determine the relative amounts of Na feldspar and K feldspar present in the original alkali feldspar at the time of Al-Si ordering and prior to exsolution. When single-crystal data are available for a microcline, the curves for α^* and γ^*, plotted as Figure 2c from Bailey & Taylor's cell dimensions, can be used.

Qualifications Regarding the Composition of 20% Na Feldspar for the Orthoclase-Microcline Transition

Several points need be made about the figure of 20% for the proportion of Na feldspar required to make the framework noticeably triclinic.

First, as with the Al-Si distribution curves the Δ curve may not be a perfectly straight line, but again there seems no reason for expecting it to be greatly different from one. Thus the figure of 20% Na feldspar is given as only an approximate and not an exact one for the orthoclase to microcline transition. Second, strictly speaking the figure of 20% Na feldspar is for only a *dimensional* and not a truly structural change from monoclinic to triclinic symmetry, because all K feldspars to the left of 20% in Figure 2a except those at the extreme will have different amounts of Al in sites $T_1(0)$ and $T_1(m)$, and will thus be *structurally* triclinic. Such a departure from true monoclinic symmetry might be discernible from intensity differences on single-crystal photographs of some orthoclases, but for most purposes it will likely prove satisfactory to regard a dimensionally monoclinic orthoclase as truly monoclinic.

Third, according to the present theory a K feldspar that has cooled slowly to a low-temperature form and that still remains monoclinic cannot have contained during cooling, or cannot contain at room temperature, more than 20% total Na feldspar. If a monoclinic K feldspar contains more than this amount of Na feldspar it must, if the ionic picture is the correct one, be a higher temperature form since Donnay & Donnay (1952) have shown that, in the synthetic high-temperature alkali feldspar series of Bowen & Tuttle (1950), the monoclinic to triclinic transition takes place at about 67% Ab. Fourth, it is well known that crystals that are only slightly triclinic have a strong tendency to twin in order to simulate monoclinic symmetry. This happens in the K feldspars and makes many slightly triclinic microclines seem monoclinic under the microscope. Thus from an *optical* viewpoint the apparent change from monoclinic to triclinic symmetry in the low-temperature K feldspars may appear to be at a somewhat greater value than the 20% Na feldspar. The writer believes that the third and fourth observations offer an explanation why the figure of 20% is so much smaller than the 52% Na feldspar that Spencer (1937) found for his orthoclase-microperthite series. The careful *x*-ray work of MacKenzie & Smith (1955) on Spencer's specimens revealed that in several of his orthoclase-microperthites of higher Na content, the K phases are triclinic. Further reference is made to some of MacKenzie & Smith's (1955) results below, and the writer plans to consider them in greater detail in the light of the present ideas at a later date.

Intermediate Microclines other than Low-Temperature

Although as pointed out above, the writer believes that many intermediate microclines represent low-temperature forms of K feldspars that are intermediate between maximum microcline and orthoclase, this picture of the feldspars does not rule out natural intermediate microclines analogous to those made by Goldsmith & Laves (1954*a*) by heating maximum microclines. They would form, according to the present theory, if an alkali feldspar containing more than 20% Na feldspar cooled slowly to some intermediate temperature, and then cooled rapidly; the product would be an intermediate microcline-microperthite. They might also result from the reheating to an intermediate temperature, followed by rapid cooling, of a maximum microcline from whose environment the original Na feldspar was in some way removed before reheating took place. The second of these processes would lead to an intermediate microcline virtually free of any associated albite; such intermediate microclines might conceivably be found in a metamorphosed sediment. These considerations lead to problems of nomenclature, or at least of implied origin, of intermediate microclines. The writer feels that if such a K feldspar is found in a plutonic rock, it will be a low-temperature form and, according to these ideas, transitional between the two extreme low-temperature forms, orthoclase and maximum microcline. It could be described as a "low-temperature intermediate microcline." If on the other hand such a K feldspar, probably as a microperthite, is found in a volcanic, possibly in a hypabyssal, or possibly even in a pegmatitic rock (as some of MacKenzie & Smith's (1955) results suggest to the writer), then it may be an intermediate temperature form in which case it could be described as an "intermediate temperature microcline." The same term would probably also describe some low-Na intermediate microclines in metamorphosed sediments. In a particular case the nature of a microcline would have to be described by either of the long terms only once, after which it could be called simply intermediate microcline. In certain cases it might require a detailed *x*-ray investigation to determine whether a given intermediate microcline is a low-temperature or an intermediate-temperature form, if indeed its true nature could be determined at all.

OTHER ALBITES

One of the interesting aspects of the ionic theory for the alkali feldspars is that it implies the existence of a series of low-temperature albites slightly different from ordinary low albite. The series will be analogous to the orthoclase-intermediate microcline-maximum microcline series in the K feldspars and will result from the same process, namely exsolution from homogenous alkali feldspars of different composition. Thus at one extreme, if an Na feldspar is cooled slowly in the presence of considerable excess of Na over K, the resulting Na feldspar will be an ordinary low-temperature albite with the Al-Si distribution given in Table 2 and Figure 1. If at the other extreme, the original alkali feldspar contains a considerable excess of K over Na, the Na feldspar formed after exsolution will have an Al-Si distribution similar to that in our ideal orthoclase, also given in Table 2 and Figure 1. If the alkali feldspar contains some intermediate proportion of K and Na before exsolution, then the framework will have an Al-Si distribution somewhere between these two extremes, and the curves in Figure 2a will again give the variations in Al-Si distribution between the two extreme low-temperature forms of Na feldspar. Now although the albite formed in the presence of excess K will have the Al-Si distribution of monoclinic orthoclase, it does not follow that this albite will be monoclinic, the reason for this being that the small Na^+ ion allows the aluminosilicate framework, whatever its Al-Si distribution, to collapse into a triclinic structure. (High-temperature albite has a monoclinic Al-Si distribution but is triclinic for the same reason.) Thus in the low-temperature Na feldspar series there will not be the symmetry change from monoclinic to triclinic that characterizes the corresponding K feldspar series and which makes it fairly easy to define a member of that series. Despite the fact that all members of this low-temperature series of Na feldspars will be triclinic, it should be possible eventually to characterize different members of the series by their cell dimensions. The albite formed under conditions of excess K will not necessarily be "more triclinic" than ordinary low albite, but as far as naming the members of this series is concerned, it is desirable to use names analogous to those for the corresponding K feldspars. Thus the obvious name for the Na feldspar formed in the presence of excess K is "maximum albite" which implies nothing about the geometry of the crystal but implies only that this particular albite was formed under conditions of "maximum K." Regarding the name of the intermediate members of this low-temperature series, the problem is almost exactly the same as in the case of intermediate microcline, for albites intermediate between the high-temperature and low-temperature forms can apparently result from the incomplete cooling of a pure Na feldspar or of one containing some K initially. The writer suggests that terms analogous to those suggested above for intermediate microclines be used, that is "low-temperature intermediate albite" for intermediate albites occurring in plutonic rocks, and "intermediate temperature albite" for intermediate albites occurring in volcanic and possibly in some hypabassal and pegmatitic rocks.[8] The first of these intermediate albites will be transitional between ordinary low albite and maximum albite which has the same Al-Si distribution as ideal

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[8] In a previously published abstract (Ferguson, 1957) the writer suggested the term "interalbite" for low-temperature intermediate albites in order to reserve the term "intermediate albite" for the intermediate temperature forms. He now prefers the terms proposed above.

orthoclase, whereas the second will be transitional between ordinary low albite and high-temperature albite. As in the case of intermediate microclines, once a particular intermediate albite has been described as one type or the other, then it need only be called "intermediate albite." Now because, as Table 2 and Figure 1 show, the Al-Si distribution in ideal orthoclase and hence in maximum albite is only slightly different from that in high-temperature albite, these two extreme forms of Na feldspar will be very similar to each other in most respects. Similarly the intermediate members of these two Na feldspar series, both extending to ordinary low-temperature albite, will be very like each other. For some intermediate albites the nature of the rock environment may indicate which of the two possible intermediate types they are, but for others the most detailed *x*-ray investigation may be required to distinguish between the two possible forms if, again, they can be distinguished at all.

It is reasonable to enquire at this point whether there is any evidence for the existence of this low-temperature Na feldspar series implied by the ionic theory of the alkali feldspars. In the writer's view there is some possible evidence for such a series. Because of the close similarity of all members of this low-temperature series to each other and to the presumed low albite-high albite series, such new albites (or high-Na plagioclases) could be distinguished only by a careful *x*-ray investigation of different specimens which have been analysed chemically and whose geologic environments are known. MacKenzie & J. V. Smith (1955), J. R. Smith & Yoder (1956), and J. V. Smith & Gay (1958) have all found from careful *x*-ray work on specimens of this kind that all plagioclases of a given composition occurring in plutonic rocks are not identical. The first of these authors carried out a very careful single-crystal *x*-ray investigation of the alkali feldspar specimens which had been so thoroughly examined in other ways by Spencer (1937). As mentioned earlier, the writer plans to consider MacKenzie & Smith's (1955) results in some detail later on, but here it is worth considering their observations and conclusions relating to Spencer's specimens D, E and F, all of which contain less than 23 weight % (Ab + An) and all of which show the K phase to be monoclinic, although specimen E apparently gives some evidence of being triclinic. If these three specimens are all lowest-temperature forms of K feldspar and if they all crystallized with a considerable excess of K over (Na + Ca), then according to the ionic theory, the Na feldspar present in these microperthites should be close to "maximum albite" and should therefore have cell dimensions which are reasonably close to those of high-temperature albite. MacKenzie & Smith (1955) found that the Na components in these microperthites do indeed have lattice angles which are similar to those of high albite. They say (page 720): "The existence of a high-temperature soda phase in these specimens, which are presumed to be from low-temperature environments, presents a problem for which no completely satisfactory explanation is at present available." The writer believes that their "high-temperature" soda phases may well be low-temperature "maximum albites." Smith & Yoder (1956) and Smith & Gay (1958) carried out careful *x*-ray work, mainly by powder photographs, on specimens of chemically analysed plagioclases from known geologic environments. The former authors conclude (page 646): "Similarly, plagioclases which have been hitherto presumed to be of 'low-temperature' origin do not fall into a unique low-temperature series." The latter authors say (page 744): "Consideration of the angular separation and subsidiary reflections of soda plagioclases from a wide range of geological environments has led to the novel hypothesis that in this composition range even plutonic specimens are in a metastable state with slightly disordered structures."[9] The writer believes that the exsolution theory for the origin of microcline, by implying the existence of a new series of low-temperature high-Na plagioclases dependent upon the proportion of K feldspar in the cooling rock, offers a possible explanation for the "metastable" plagioclases found in plutonic rocks by the above authors, although much more careful work will have to be done to test this possible explanation. The ionic picture of the alkali feldspars suggests that, when a rock contains appreciable amounts of both K feldspar and (Na,Ca) feldspar, the true nature of one cannot be determined without taking account of the possible part played by the other in its earlier history.

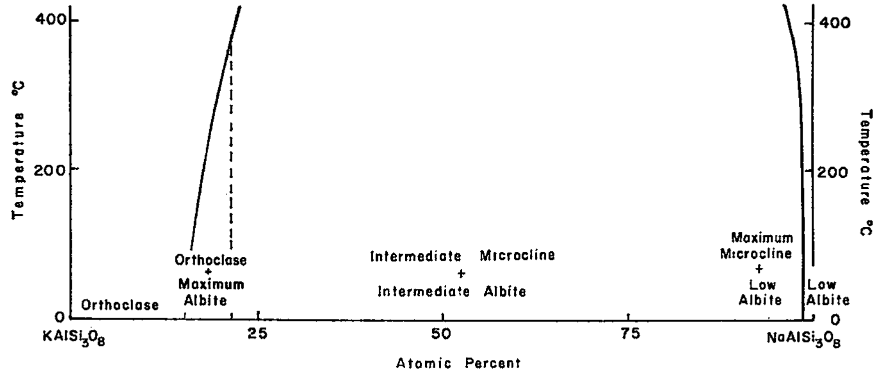

FIG. 3. The (slowly-cooled) low-temperature phases on the alkali feldspar phase diagram as they are implied by an ionic picture of the structures. The changes between the sub-solvus phases are all transitional. A fuller discussion is given in the text.

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[9] It will be apparent from the earlier part of this paper that the writer disagrees with the implication in this statement that, in general, the degree of stability is related to the degree of Al-Si order.

THE LOW-TEMPERATURE PHASES ON THE ALKALI FELDSPAR PHASE DIAGRAM.

The assumption of ionic bonding in the alkali feldspars for which there is strong structural evidence, and the resultant exsolution theory for the origin of microcline described by Ferguson, Traill & Taylor (1958, 1959) and by the writer in this paper, have led to the following stable phases in the low-temperature region of the alkali feldspar phase diagram: orthoclase, orthoclase + maximum albite, (low-temperature) intermediate microcline + (low-temperature) intermediate albite, maximum microcline + low albite, low albite. These relationships are shown on the diagram in Figure 3 in which the limit of solid solution of Na feldspar in orthoclase is taken from MacKenzie & Smith (1959), and this boundary joined to the solvus deduced by Bowen & Tuttle (1950) for synthetic (high-temperature) alkali feldspars. In the low albite region the solvus is extended from that of Bowen & Tuttle, but its exact position is not significant and it is intended to indicate only that the solubility of K feldspar in low albite is very small. The solvus may, in fact, intersect the vertical line representing pure $NaAlSi_3O_8$ at a temperature of several hundred degrees. Within the solvus all phase changes are transitional including that from orthoclase to microcline; the boundary between these two is shown by a broken line at the composition deduced earlier, 20% $NaAlSi_3O_8$.

The writer is aware that one, and perhaps two, aspects of the phase relationships shown here are unorthodox on a phase diagram. The first is that, at a given temperature, the structure of one phase within the solvus varies with the overall composition—thus at room temperature orthoclase in the high-K region and maximum-microcline in the high-Na region. The second is that if, as suggested earlier, the amount of Na retained in solid solution in a low-temperature K-feldspar is a function of original Na:K ratio, then in the high-K region in Figure 3, the solvus would have to be represented not by one line as shown, but by a broad band (probably narrowing towards higher temperature), one side of which would represent the composition of the final K phase (orthoclase) for an original low-Na content, and the other that of the final K phase (maximum microcline) for an original high-Na content. This matter the writer hopes to explore more fully later. Despite these unorthodox aspects, the phase relationships shown here have a strong crystal-structural basis which is the writer's justification for proposing them.

Whether the different pairs of phases within the solvus will exist as (micro) perthite or as two separate minerals side by side is not clear to the writer, but if initial high Na does tend to give more complete exsolution than initial low Na as suggested previously, then orthoclase + maximum albite will probably occur only as (micro) perthites whereas maximum microcline + low albite are more likely to occur as two separate minerals.

SOME FURTHER IMPLICATIONS

Although the writer plans to explore more of the broad implications of these ideas in a later paper, he wishes to consider two important implications here.

Authigenic K Feldspars

This type of K feldspar apparently forms as a fairly pure chemical compound, and if this is the case, one would expect from the present theory that all authigenic K feldspars would be either sanidine if the mineral crystallizes metastably as the high-temperature form, or orthoclase if it crystallizes as the stable low-temperature form. Observations such as those of Baskin (1956), however, indicate that although many authigenic K feldspars are monoclinic, some are triclinic microcline with apparently no albite lamellae or other indication of the exsolution process required by the present theory to produce a microcline. The existence of authigenic microclines appears to either contradict the ionic theory for the alkali feldspars or to indicate that microclines can arise in some way other than in the presence of Na as suggested by the ionic theory. However, in view of the uncertainty and disagreement regarding the mode and temperature of origin of authigenic feldspars suggested by Ingerson (1955) for example, it is possible an explanation for the apparent discrepancy with the present theory will eventually be found.

Series in the Alkali Feldspars

The second implication concerns possible series between the K feldspars and the Na feldspars. Spencer (1937), Tuttle (1952*a*) and MacKenzie & Smith (1955) have all concluded, mainly on the basis of optical observations, that there are two low-temperature alkali feldspar series, one extending from low albite to orthoclase and the other from low albite to microcline. All these authors found large gaps in these two proposed series. The present theory suggests that only one incomplete low-temperature series is to be expected in the K-Na feldspars. The writer believes, as the earlier part of this paper makes clear, that there must exist a low-temperature K feldspar series varying from orthoclase to maximum microcline, and a low-temperature Na feldspar series varying from low albite to maximum albite, but he does not believe there is any complete series involving both of these alkali feldspars. For such a series with steadily changing optical properties to exist, one would expect the following characteristics of the members either to remain unchanged or to vary regularly with composition throughout the series: (1) the crystal structures of both phases; (2) the nature and amount of twinning, if it is present, in one or both phases; and (3) the relative amounts of the two phases. It is mainly the third condition which, in the writer's opinion, prevents the series from being a complete one. If, as suggested earlier, the (unknown) exsolution forces are greater for initial high Na than for initial high K, then more and more complete exsolution would tend to take place with increasing initial Na. Thus, one might expect the low-temperature alkali feldspar series to exist in the high-K composition range, but when the proportion of Na reached some particular amount, complete exsolution would tend to take place and the resultant K feldspar, a microcline, would not reveal its original Na content. In a sense microcline would represent a member of the orthoclase—low albite series in the high-Na region, but because of the loss of some or all of its original Na, neither its final composition nor its optical properties would place it anywhere in the orthoclase-low albite series. This interpretation would account for the fact that Spencer (1937) and Tuttle (1952*a*) were able to find a fairly smooth gradation of properties in the orthoclase-microperthite series up to a composition of about 50% (Ab + An), but that their microcline-microperthite series covers a very limited range in the low-Na region, about 15 to 40% (Ab + An).

SUMMARY

The assumption of ionic bonding in the Na feldspar structures offers an explanation for the lack of complete Al-Si ordering in low albite, for the relative stabilities at ordinary temperatures of high albite and low albite, and for the tendency of Al to segregate into one particular site in low albite. If it is then assumed that the K feldspars are ionic, it can be shown that the most balanced and presumably most stable K feldspar structure is monoclinic orthoclase, but that if a K feldspar cools slowly in the presence of appreciable Na, the resultant exsolved K component will be triclinic microcline. The particular Al-Si distribution observed in an intermediate microcline by Bailey & Taylor (1955) supports this theory, and permits one to estimate that about 20% $NaAlSi_3O_8$ is required in the original alkali feldspar to make the final exsolved K feldspar triclinic. The exsolution origin for microcline implies that the Na component of an exsolved perthite will be slightly different from ordinary low albite, and it thus implies also that there will be a series of low-temperature K feldspars and one of low-temperature Na feldspars,

both depending on the Na:K ratio in the feldspar before exsolution. A number of the broad implications are examined.

Although many of the details of the picture presented here require confirmation by further careful investigations, particularly on material from rocks whose thermal and chemical history can reasonably be deduced, this theory does account for the known detailed structural features of the alkali feldspars, and it leads to a reasonable set of phase relationships among the alkali feldspars.

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