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Distribution of potassium feldspar polymorphs in intrusive sequences

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SUMMARY. K-feldspar variation is described for four salic plutonic igneous complexes that show normative mineral variation in keeping with the course of fractionation predicted from experimental studies. These are a sodic syenite (Loch Ailsh), potassic syenites (Loch Loyal), a peralkaline syenite and soda granite complex (Puklen), and a calc-alkaline pluton (Foyers). Additional data are given for examples from the literature. Microcline becomes more abundant relative to orthoclase as members of these intrusive sequences approach thermal minima on the liquidus in the system Ab–Or–An–Qz. This relation exists irrespective of the compositional range of the rock suites, the bulk composition of the alkali feldspars, grain size, or field relations. Early orthoclase-bearing rocks retain orthoclase when enclosed as xenoliths in microcline-bearing rocks, or when cut by minor bodies of microcline-bearing rocks. The K-feldspar variation may reflect increasing water content or the peralkaline or peraluminous character of the fractionating magmas. These factors affect the feldspar structure at the time of initial crystal growth and dictate whether microcline will develop on cooling.

THE observation that the potassium feldspar in plutonic acid and alkaline rocks may be either monoclinic orthoclase or triclinic microcline, usually showing the characteristic cross-hatched twinning, is familiar to petrologists. The physical factors controlling the presence of either polymorph are, however, unclear. Experimental studies of ordering in synthetic albitic alkali feldspars have been carried out (MacKenzie, 1957; Parsons, 1968; Martin, 1969) but the synthesis of highly ordered K-feldspars has not been achieved under controlled conditions. (Euler and Hellner, 1961, produced material of adularia habit and slight obliquity in a pressure vessel with a temperature gradient.) The factors controlling the presence of either polymorph can, at present, be established only from studies of the distribution of these feldspars in natural rocks, and such evidence is summarized by MacKenzie and Smith (1961). Dietrich (1962) subsequently carried out a generalized study of the distribution of K-feldspars in a wide variety of rock types. A large number of more specialized studies have been made of variation in the K-feldspars in individual igneous bodies, and many of these will be referred to later. Temperature of crystallization, cooling rate, presence of volatile fluxes, shearing stress, and variations in bulk composition are all factors that seem likely to influence the eventual identity of the K-feldspar in igneous rocks. More recently Martin (1969) has demonstrated the possible importance of peralkaline solutions in facilitating the ordering process, and the inhibiting effect of a peraluminous environment. The precise nature of the orthoclase–microcline transformation is itself not clear and the views of MacKenzie and Smith (1961) may be contrasted with those of Wright (1967), Tilling (1968), and Ragland (1970). A clear discussion of the physical reality of Al–Si order–disorder and domain textures in alkali feldspars is given by Smith (1970).

The present study concerns the distribution of K-feldspar polymorphs in a number of slightly alkaline plutonic bodies on which one of us (Parsons) has worked, and in the calc-alkaline Foyers intrusion investigated by the second author. Supported by data obtained from the literature we demonstrate that the identity of the K-feldspar in many igneous rocks is strongly dependent on the relative bulk composition of the rock in terms of normative *ab*, *or*, *an*, and *Q*, and on the rock's position in a scheme of fractionation appropriate to each intrusion. Each of the instrusive complexes described exhibits variation in bulk chemistry that gives a sequence of rock types corresponding to the course of fractionation established by experimental studies in the system Ab–Or–An–Qz; on these grounds *only* each series of rocks is referred to as a fractionated sequence. Whether fractional crystallization has occurred in all or any of these examples is not under consideration; we are concerned only with the relationship between the position of different rocks in these sequences and the nature of their K-feldspars.

Techniques. The new chemical analyses utilized in this paper were determined by X-ray fluorescence analysis using crushed and compressed lithium metaborate fusions and appropriate wet-analysed standards. Auxiliary determinations of FeO, CO_2, and H_2O were also made. Analytical detail will be given in papers in preparation, only C.I.P.W. norms being presented here. These were calculated using a computer programme written by D. C. Smith. The conventionl symbols *ab*, *or*, *an*, and *Q* are used to indicate normative minerals; Ab, Or, An, and Qz indicate end-member molecules such as $NaAlSi_3O_8$, etc.

A crude X-ray diffractometer technique was used to establish the nature of the K-phase of the feldspars in the rocks studied. Pressed mounts of powdered whole rock samples were used and the diffractometer caused to complete one oscillation over the region 29°–32° 2θ (Cu-$K\alpha$). Standard instrument settings used are given in fig. 1. The traces were then visually ranked, using a transparent overlay as indicated in fig. 1. These traces include the 131 reflection of orthoclase at about 29·8° 2θ (Cu-$K\alpha$ radiation) and the 131 reflection of microcline between that angle and about 29·45° for a maximum microcline.

Because the bulk feldspar of most plutonic syenites and anorthite-poor granites covers a restricted range of composition (Tuttle and Bowen, 1958) from about $Ab_{50}Or_{50}$ to $Ab_{75}Or_{25}$, diffractometer traces of feldspar reflections from whole rock samples look similar whether the rock is hyper- or sub-solvus (the terms of Tuttle, 1952) in type. Over this compositional range variation in the relative intensities of reflections from Na- and K-phases does not greatly alter the general appearance of traces in the region 29°–32° 2θ. Only the anorthite-rich members of the Foyers tonalite-granite sequence provide diffraction patterns markedly different from those in fig. 1 and these will be described below.

Fig. 1 shows seven variations (referred to as 'reflection types') in the appearance of the diffractometer traces in the region 29·25°–30·25° 2θ. The traces show both K-phase and Na-phase reflections, which may be phases in perthitic intergrowth or discrete albitic plagioclase and perthitic K-feldspar crystals. The Na-phases give reflections in all cases close to those of low albite. Type 1 traces show a single sharp K-feldspar 131 reflection at about 29·8° 2θ indicating a dominantly monoclinic K-feldspar and thin sections of rocks with feldspars in this category do not show cross hatched microcline twinning. For the purposes of this paper such feldspars will be called 'orthoclase' although, strictly, these criteria do not rule out feldspars that would be called sanidine on the basis of 2V. Traces of types 2 and 3 indicate coexistence of orthoclase and microcline. In the case of type 2a traces, microcline type twinning can be seen in thin section although a broadened 131 reflection is the only X-ray evidence of the presence of microcline. Types 2b and 2c clearly show diffuse microcline 131 reflections. Type 3 traces are distinguished by a distinct microcline 131 peak. Both type 2 and type 3 traces show the presence of both orthoclase and microcline and will be said to indicate 'variable' obliquity within the sample. Type 4 traces do not show reflections in the region between the 131 microcline reflection and 29·8° and for the purposes of this study are considered to contain only microcline. Similar variation in the K-feldspars of plutonic rocks has been illustrated by Christie (1962) and Tilling (1968). Dietrich (1962) measured the half-height width of the 131 reflection in traces like the present type 2a and used this as a measure of the obliquity of the microcline; we prefer to regard such a trace as indicating dominantly orthoclase with minor microcline of variable obliquity. When a distinct microcline 131 reflection can be seen an estimate of obliquity (usually measured in terms of $2\theta_{131}-2\theta_{1\bar{3}1}$) can be made in traces in which albite reflections obscure microcline $1\bar{3}1$ by considering the position of microcline 131 with respect to low albite $02\bar{2}$ (Parsons, 1965).

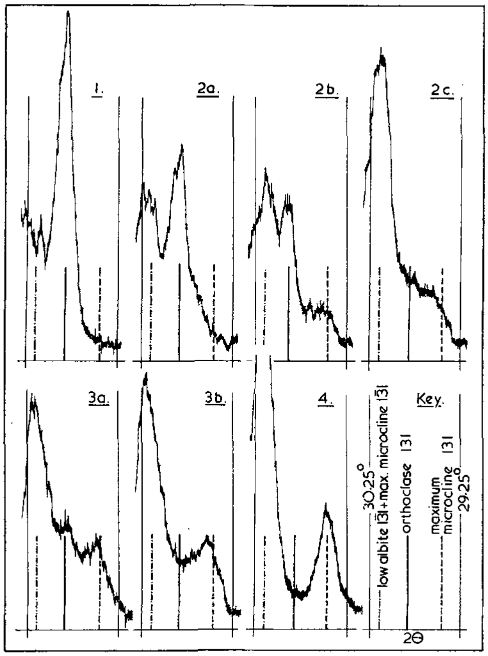


FIG. 1. Examples of X-ray diffractometer traces defining the K-feldspar 131 reflection types used in this paper. These patterns are appropriate for mixtures of the K-feldspar and low albite. If a more calcic plagioclase is present the orthoclase 131 reflection may be obscured (see fig. 5). For the purposes of this paper the reflection types are: Type 1 = Orthoclase only, always confirmed optically. Type 2 = K-feldspar of variable obliquity. Type 3 = Variable obliquity, but a distinct microcline 131 peak. Type 4 = Microcline only. Instrument settings used were: Ratemeter 2; time constant 8; Slits 1°–0·1–1°; Scanning speed ⅛°/min; Chart speed 400 mm/hr Cu-$K\alpha$ radiation.

Range of variation in the K-*feldspar*

Only 8 samples gave type 1 reflections; 135 gave types 2 and 3 and 65 gave type 4. This distribution mainly reflects the number of samples collected within the individual intrusions to be described, but it is in keeping with the findings of Dietrich (1962). When a distinct microcline 131 reflection is present (types 3 and 4) it usually indicates microcline of high obliquity and, as Barth (1959) and Dietrich (1962) show, microclines with obliquities less than 60 % of the maximum are rarities. Similarly, diffuse reflections of feldspars of variable obliquity (type 2) normally extend over a 2θ range indicating high obliquity for some parts of the structure of the K-feldspars. Despite their variability in size and composition the instrusions to be described below all show a range of the K-feldspar types illustrated (fig. 1). There is no straightforward tendency for microcline to be most abundant in the larger bodies, for example, and there will be described large orthoclase-bearing intrusions with which are associated minor intrusives that carry microcline.

Measurements of the $20\bar{1}$ spacings of the feldspars investigated were also made. These always indicated compositions close to K-free low albite and nearly pure Or, although reliance cannot be placed on the $20\bar{1}$ method as an indication of composition for feldspars in perthitic intergrowth because of strain effects (Smith, 1961) and some examples gave 'impossible' compositions. There was a very slight tendency for the $20\bar{1}$ spacings of feldspars with a dominantly monoclinic K-phase to indicate K-phase compositions more sodic than those with a wholly triclinic K-phase. A similar result was obtained by Ragland (1970); it is in keeping with the theoretical predictions of Smith and MacKenzie (1961) and the experimentally determined relationships for albitic anorthoclases (Parsons, 1968).

Variation within individual intrusive complexes

The Loch Ailsh complex, Assynt, Scotland, is a small (4 km by 2·5 km) composite intrusion dominantly of highly feldspathic syenite. The feldspar variation within the mass has been described previously (Parsons, 1965) and is summarized here, together with some new rock and separated-feldspar analyses (fig. 2).

The syenites comprise medium- to coarse-grained hypersolvus rocks, the majority with >90 % modal alkali feldspar. Three phases of intrusion can be recognized in the field, early phases S1 and S2, and a later, almost monomineralic alkali-feldspar rock, S3. Quartz is most abundant in S2 but is usually less than 3 % by volume, only rarely reaching 10 % in individual thin sections. Normative *an–ab–or* for rocks from each phase of intrusion are plotted in fig. 2, together with An–Ab–Or for feldspars separated from some of the analysed rocks. One of the S2 syenites contains 3 % normative *Q*, but the remaining rocks are *Q* free. Feldspars in the earliest rocks are more *ab* and *an* rich than the later rocks so that they form a sequence trending towards the field boundary in An–Ab–Or and lying within the plagioclase field. All the feldspars are microperthitic with a range of coarseness in the perthite from 2 μm spindles in S1 to 0·5 mm patches in some marginal varieties of S3. The variation in the K-phases of the perthites is summarized in fig. 2 using the classification of 131 reflection types shown as fig. 1. Highly sheared rocks are excluded; in some instances shearing leads to increase in microcline obliquity but in highly mylonitized rocks K-feldspar may have recrystallized as orthoclase (Parsons, 1965).

There is a striking tendency for microcline to become increasingly abundant as the rock types progressively approach the field boundary. Orthoclase does not occur

Syenite:	Reflection types: 1	2	3	4
x S1	1	3	1	
● S2			18	12
Δ S3				43

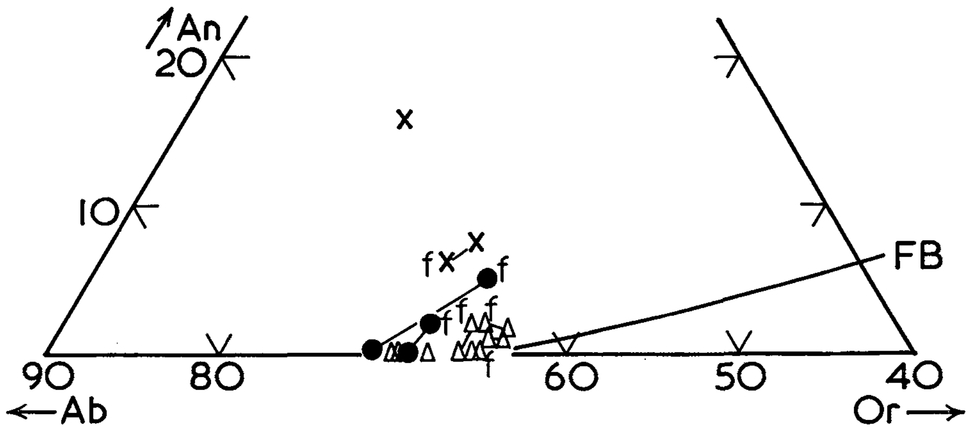


FIG. 2. Feldspar variation in the Loch Ailsh complex (new analyses, see also Parsons, 1965), plotted on part of Ab–Or–An. f = analysed separated feldspar. Other points represent normative feldspar for analysed whole rocks (strictly, *ab–or–an*). FB is the field boundary in Ab–Or–An given by Yoder, Stewart, and Smith (1957). The table shows the number of specimens of each phase of intrusion giving the listed K-feldspar reflection types (see fig. 1)

in any S3 rocks but where S3 encloses feldspar xenocrysts from S2, the S2 crystals preserve their monoclinic K-phase. Single-crystal X-ray studies showed that S1 xenoliths held in S3 contained dominantly monoclinic K-feldspar up to within less than 2 cm of a sharp contact. Even though the earlier S1–S2 feldspar must have had the same cooling history as the enclosing S3 the feldspars on unmixing and ordering did not reach the same final condition.

The Loch Loyal syenites, Tongue, Scotland. Three syenite intrusions, of Caledonian age, lie close together around Loch Loyal. No contacts are seen between them, but the larger Ben Loyal intrusion is separated by less than 1 km of unexposed terrain from the smaller Cnoc-nan-Cuilean and Ben Stumanadh bodies. Although no age relations can be established between these intrusions their petrographic and chemical similarities (fig. 3) and spatial closeness suggest that they are related to one genera. period of intrusive activity. King (1942) has described the Cnoc-nan-Cuilean mass in some detail, but only a brief account (Read, 1931) of the Ben Loyal body has been made, and there is no published detail of the Ben Stumanadh rocks. The Cnoc-nan-Cuilean body outcrops over about 4 km^2 with a vertical section of at least 400 m, and differs from the other bodies in containing extensive xenolithic bodies of basic

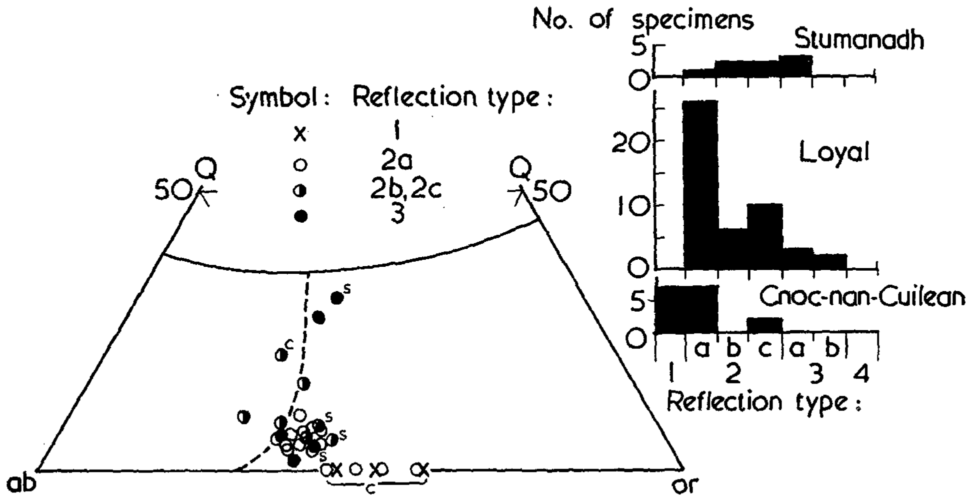


FIG. 3. Feldspar variation in the Loch Loyal intrusions. Analysed rocks from each intrusion are plotted in terms of normative *ab–or–Q*. C = Cnoc-nan-Cuilean, S = Ben Stumanadh, Unlettered = Ben Loyal. The quartz–feldspar field boundary and unique fractionation curve in Ab–Or–Qz at P_{H_2O} = 2000 kg/cm^2 (Tuttle and Bowen, 1958) are also shown. The histograms show the number of specimens in each intrusion giving the various K-feldspar reflection types (see fig. 1).

syenites shown by King (1942) to be metasomatized Moine schist, the local country rock. Ben Loyal covers about 15 km^2 and is exposed over a vertical section of over 700 m. Like Cnoc-nan-Cuilean its form in depth is obscure, but R. Robertson (personal communication) has mapped an inward dipping lamination in its marginal syenites. The syenites of Ben Stumanadh outcrop over about 8 km^2 and map as a pair of eastward thinning sheets, the larger, upper member attaining a thickness of at least 250 m.

A detailed account of the variation within the Loch Loyal bodies is in preparation (I.P.), including thirty-four new analyses, which are presented in terms of normative *ab–or–Q* in fig. 3. These medium- to course-grained leucosyenites contrast with the Loch Ailsh syenites, with which they have previously been compared (Read, 1931) by being more potassic (particularly Cnoc-nan-Cuilean) and more quartzose (except Cnoc-nan-Cuilean). It is possible that the high quartz contents of some of the rocks plotted are due to incorporation of silica from country rock, but the analysed material is free from obvious xenoliths or 'ghosts'. The one Cnoc-nan-Cuilean rock that contains *Q* is from an aplitic vein in massive basic xenoliths. The majority of the Loch Loyal rocks show no normative *an*, but a maximum of 2·6 % *an* is reached in examples from Cnoc-nan-Cuilean, which averages 1·1 % *an*. Ben Stumanadh averages 0·8 % *an*, and Ben Loyal 0·2 % *an*. All Ben Loyal and Ben Stumanadh rocks show $>83\%$ $(ab+or+Q)$. Cnoc-nan-Cuilean rocks may be more basic, with, however, $>72\%$ $(ab+or+Q)$. The variation in the Loch Loyal rocks is reasonably summarized, therefore, in fig. 3.

The feldspar in the syenites may be almost entirely a microperthitic alkali feldspar together with minor amounts of finer grained interstitial albite in discrete grains, or, in many Ben Stumanadh and Ben Loyal rocks, coarse plagioclase crystals may be present together with the perthitic alkali feldspar. Fig. 3 does not indicate bulk alkali feldspar compositions and these may vary between specimens that have similar normative *ab–or–an*. These syenites may be hypersolvus or subsolvus types and this difference does not appear to be related to their bulk composition. The K-phases of the alkali feldspars show rather limited variation and are never entirely microcline. The majority of rocks give K-feldspar reflections of types 1 and 2a. Although many of these type 2a reflections are fairly sharp 131 reflections, diffuse microcline-type cross-hatched twinning can always be seen in thin section. The few specimens classed as type 1 do not show recognizable twinning, although even in these specimens the K-feldspar occasionally shows patchy extinction.

Fig. 3 shows the distribution of feldspars of the various reflection types between the three intrusions, and also a relationship between the reflection type and normative *ab–or–Q*. Although the distribution between the three intrusions overlaps, there is a general tendency for microcline to be more conspicuous in Ben Stumanadh than in Cnoc-nan-Cuilean while the larger Ben Loyal body lies dominantly between the two. There is a real tendency for specimens giving reflection types 2b, 2c, and 3 to plot near to the thermal valley (Tuttle and Bowen, 1958) in Ab–Or–Qz. The Cnoc-nan-Cuilean rocks, which lie on the *ab–or* sideline, are those that contain little or no microcline. The relative amount of microcline in the Loch Loyal rocks is not a function of the size of the intrusion, and does not vary systematically with the locality of the specimens within each intrusion or their grain size. No simple relationship could be found between modal plagioclase and identity of the K-phase in the coexisting alkali feldspar, and this implies that the orthoclase or microcline content is not related to the bulk composition of the alkali feldspar. The only systematic element that can be discerned in the variation described is the tendency for microcline to develop in rocks that approach most closely to the thermal valley in Ab–Or–Qz. In the Loch Loyal syenites the direction of the sequence of fractionation implied by the plotted normative compositions is quite different from Loch Ailsh, and the feldspar variation less well defined, but the same general relationship is observed between relative normative composition and the nature of the K-feldspar.

The Puklen complex, Nunarssuit, SW. Greenland, is a minor member of the Pre-Cambrian Gardar suite of alkaline intrusions in SW. Greenland. It has been briefly described by Pulvertaft (1961) and is currently the subject of a more detailed study (Parsons, in press). It covers an area of about 8 km² and comprises an oval body of coarse augite–fayalite syenite and quartz syenite mixed in an intricate fashion and sometimes showing evidence of gravitational banding, into which are intruded two bodies of coarse soda granite, the largest circular body being about 1·5 km in diameter. This is cut by, or in places grades into, fine grained, almost flinty granophyre. Contacts between syenites and soda granite are usually gradational but sharply defined granite veins cut augite syenite up to 1 km outside the main soda granite mass. The Puklen analyses show normative *ac* (up to 8 %) and there are petrographic indications of peralkalinity in the form of strongly coloured amphiboles and accessories such as aenigmatite and astrophyllite. Such signs of peralkalinity are most apparent in the later soda-granites. The most mafic augite syenites show >70 % (*ab*+*or*+*Q*), while the most leucocratic granophyres give >93 % (*ab*+*or*+*Q*). Only the granophyres show slight (<2 %) normative *an*. Eleven new analyses are plotted in terms of *ab–or–Q* on fig. 4. They form a clear series parallel to the unique fractionation curve (Tuttle and Bowen, 1958) in the system Ab–Or–Qz and there seems every reason to believe that the Puklen sequence represents the result of fractional crystallization of a quartz syenitic magma lying close to the thermal valley in Ab–Or–Qz.

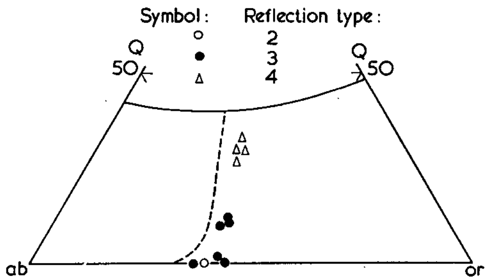


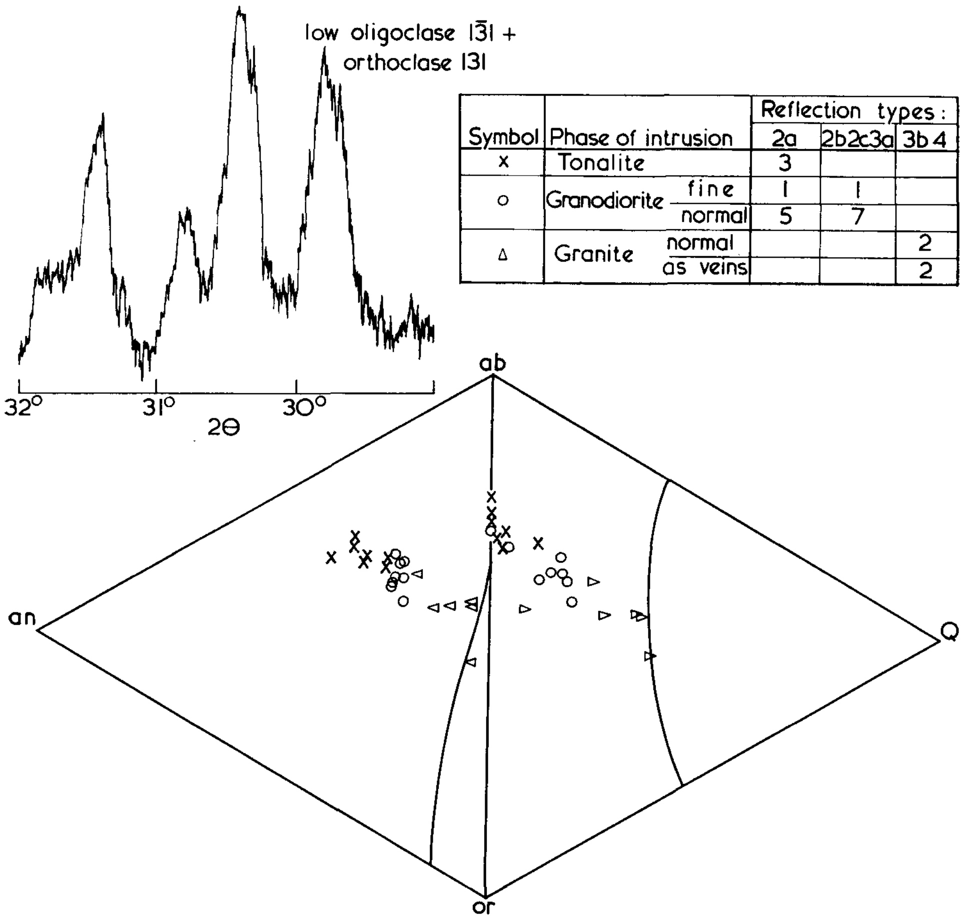
FIG. 4. Feldspar variation in the Puklen intrusion (Parsons, in press). Analysed rocks are plotted in terms of normative *ab–or–Q*. The quartz–feldspar field boundary and unique fractionation curve in Ab–Or–Qz at P_{H_2O} = 500 kg/cm² (Tuttle and Bowen, 1958) are also shown.

The feldspar is almost all microperthitic alkali feldspar, although some specimens show discrete grains of albite or albitic rims at the margins of perthitic alkali feldspars. Fig. 4 shows the variation in the 131 reflections of the K-phases. Type 4 (wholly microcline) traces are strikingly restricted to the soda-granite–granophyre group and all other rocks have K-feldspars of variable obliquity. This sharp distinction extends to a narrow (30 cm) microgranite vein cutting augite syenite 3/4 km outside the main body of soda granite. The K-feldspar of the vein is type 4, the coarse syenite that it cuts is type 3a.

In the Puklen intrusion microcline is restricted to the later, most fractionated phase of the intrusion, even though this part of the intrusion is in places very fine-grained and exhibits intricate granophyric intergrowths. Microcline is the only K-feldspar in later granite veins cutting orthoclase-bearing syenite.

The Foyers complex, Inverness-shire, Scotland, is a concentrically zoned calc-alkaline tonalite-granite pluton outcropping over an area of about 65 km². It lies near the east bank of Loch Ness 30 km south of Inverness and is faulted on its NW. margin. The complex has recently been studied by Marston (1967, 1970). We are indebted to Dr. Marston for allowing us to use his data prior to their publication.

Three phases of intrusion are present: a tonalite (about 50 % plagioclase, 25 % hornblende plus biotite, 20 % alkali feldspar), a granodiorite (30 to 40 % alkali



Symbol	Phase of intrusion		Reflection types: 2a	2b2c3a	3b4
x	Tonalite		3		
o	Granodiorite	fine	1	1	
		normal	5	7	
Δ	Granite	normal			2
		as veins			2

FIG. 5. Feldspar variation in the Foyers complex. Analyses of Marston (1967, 1970) are plotted in terms of normative *ab–or–an* and *ab–or–Q*. An example of the diffractometer pattern of a granodiorite between 29° 2θ, Cu-$K\alpha$, and 32° is shown. Because the plagioclase is calcic, the 131 orthoclase peak is obscured. Absence of a distinct microcline 131 reflection can be confirmed, however. This trace is classified as 2a, because faint grid twinning is seen in section. The table shows the number of specimens falling into the various reflection types (fig. 1).

feldspar), and a leucogranite (50 % alkali feldspar). A locally highly potassic migmatite occurs south of the main intrusion.

The normative data plotted show the normal course of fractionation of a calc-alkaline magma and the field relations are in keeping with an order of intrusion parallel to the course of fractionation suggested by fig. 5. We have not X-rayed feldspars from the analysed specimens but the well-defined grouping of the rock types (particularly in terms of *ab–or–an*) shown in fig. 5 suggests that it is reasonable to assume that our specimens from the various phases of intrusion fall in the same compositional areas as the analysed specimens.

Diffractometer traces of feldspar reflections between 2θ 29°–32° (Cu-$K\alpha$) for rocks with a calcic plagioclase are different from those for the anorthite-poor rocks shown in fig. 1. Fig. 5 shows an example of a trace from a Foyers tonalite in which the orthoclase 131 reflection is obscured by the calcic plagioclase 131 reflection, but no reflections exist in the region of 29·5° 2θ, indicating that microcline of high obliquity is not present. All such rocks show feldspars exhibiting faint microcline twinning in thin section, and they are thus classed as type 2a in the table given with fig. 5. This shows that the granites contain microcline with little or no orthoclase, while all the other phases of the intrusion contain orthoclase with subsidiary microcline.

The development of microcline relative to orthoclase is again demonstrated to parallel the course of fractional crystallization. At Foyers, as at Loch Ailsh, shearing stress has modified K-feldspars of all phases of the intrusion and sheared rocks gave type 4 traces, irrespective of rock type.

Examples from the literature

Oslo alkaline series. Dietrich (1962, fig. 2) shows average obliquity values for Oslo rocks in the sequence kjelsåsite–larvikite–nordmarkite–ekerite (i.e. from calcic syenite towards alkali granite) and noted that average obliquity increased sytematically with increasing fractionation towards ekerite. Dietrich's lower obliquity values correspond essentially to type 2 traces of the present scheme, so that K-feldspars of the Oslo series behave like those in the previous sections.

Enchanted Rock batholith. Very recently Ragland (1970) has described in some detail the K-feldspar variation in this calc-alkaline granodiorite-leucogranite composite intrusion and its associated aplites and pegmatites. A clear correlation is shown between the K-feldspar present and the lithology, the degree of order in the K-feldspars increasing with increasing fractionation, as measured by the Thornton–Tuttle differentiation index (Thornton and Tuttle, 1960). By considering the composition of biotites in the different portions of the batholith he suggests that the marginal parts of the intrusion, which essentially contain microcline only, crystallized under conditions of higher water-fugacity and lower temperature than the interior, orthoclase- and microcline-bearing rocks. Water is believed to have catalysed the transformation of orthoclase to microcline during the cooling of the rocks. Ragland points out that the feldspars completed crystallization at high temperatures (he suggests 693 to 740 °C) well above the upper limit of stability of microcline around 400 °C (see discussion below). The sharp characterization of particular lithologic units by particular types of K-feldspar also implies that the volatiles remained localized within the rock types during cooling. We have here pointed out the striking precision of this limitation of certain K-feldspar types to particular members of intrusive sequences and its implications are discussed below.

Herefoss granite. Nilssen and Smithson (1965) showed that the K-feldspar of this circular foliated diapiric calc-alkaline body shows increasing obliquity with decreasing CaO, Fe_2O_3, TiO_2, and MgO. The rocks range from quartz diorite to granite, and dominantly comprise quartz monzonites. Most specimens examined contained both orthoclase and microcline of variable obliquity.

Boulder batholith. Dietrich (1962) also shows increase in obliquity with fractionation in a series comprising three granodiorites, ademellite, and two-mica granite. As in the Oslo province K-feldspar obliquity increases with the quartz content of the rocks, and comments that 'Δ-values [obliquity] commonly increase with "normal" magmatic evolution, lowering of temperature of consolidation, increase of volatile content etc.'. Tilling (1968), describing in detail the Rader Creek granodiorite within the Boulder batholith, showed that rocks with more sodic alkali feldspars (bulk Or 66 to 78) contain dominant microcline whilst more potassic feldspars (Or 84 to 85) showed orthoclase only. Increasing Ab in alkali feldspar is the trend expected with fractionation in a granodiorite, and again the evolution of microcline is in parallel with this trend.

Tatoosh pluton. Wright (1964) described feldspar variation in this pluton, which comprises granodiorite and quartz monzonite. All examples of granodiorite K-feldspars gave sharp 131 peaks for monoclinic feldspar whilst some quartz monzonite traces gave 'broadened' 131 reflections. 2V also increases from granodiorite to quartz monzonite.

Wiborg rapakivi granites. Vorma (1971) has described feldspar variation in the large Wiborg rapakivi complex, which is at least 160 km $\times$ 100 km in size, and which may be subdivided into three or four intrusive phases. The different members follow a calc-alkaline trend from within the plagioclase volume in Ab–Or–An–Qz, with a final phase of injection of aplites and quartz porphyries lying close to the field boundary in Ab–Or–Qz. Both amongst and within these intrusive units there is a close relationship between falling calcium content, increasing silica, and the appearance of microcline in the perthitic alkali feldspars, at the expense of orthoclase. Vorma ascribes this relationship to the parallel processes of fractionation and concentration of volatiles. It is striking that within this large intrusive complex the same range of feldspar variation exists as is found in the comparatively tiny masses described in the present paper, and that increase in feldspar order (Vorma shows comparable variation in the Na-phase) should similarly parallel changes in rock composition.

Kûngnât syenite. Upton (1960) shows that the rocks of the layered Gardar alkaline pluton are compositionally similar to those of the small Puklen body described above. The volume of soda granite relative to syenite is much smaller, however, and the late acid rocks occur as sheets or small lens shaped pods in various members of the syenite suite. Upton notes that cross-hatched microcline appears only in the later soda granite members of the complex. Many of these rocks have a pegmatitic aspect; the Puklen acid members are normal even-grained or fine-grained rocks. Despite these differences microcline seems to evolve in the later acid members of both these peralkaline intrusions.

Minor intrusives associated with granites. Marmo (1959) noted that orthoclase-bearing granites are frequently cut by aplitic veins that contain microcline of high obliquity. Marmo concluded that 'once orthoclase has been formed it seems to be as stable as microcline at low temperatures'. MacKenzie and Smith (1961, and see also discussion by Laves of their paper) explain this as an effect of composition, believing that solid solution of Na will promote Al–Si disorder in K-feldspars (an hypothesis supported by the experimental data of Parsons, 1968, which shows that the converse applies for Na-rich feldspars). The sodium-bearing orthoclase of the granites will have a lower equilibrium degree of order than the nearly pure microcline of the aplites. The data in the present paper show that persistence of orthoclase in early rocks intimately associated with later microcline-bearing rocks is a common phenomenon, and it cannot be explained as a direct effect of composition because in many cases the K-phases are *unmixed phases* in alkali feldspars of similar bulk composition. The composition of the K-phases should be the same at any particular temperature irrespective of the bulk composition of the feldspar.

Discussion of observed distribution

The twelve intrusions or groups of intrusions described above have one feature in common. Microcline is the dominant K-feldspar (in many cases the only K-feldspar) in those members of the intrusive sequences that have bulk compositions approaching most closely the composition with the lowest melting temperature for the particular range of compositions covered by the intrusion or group of intrusions. Less fractionated members contain orthoclase or orthoclase and microcline. Fig. 6 summarizes the variation in bulk chemistry (in terms of *ab–or–an–Q*) exhibited by the four complexes described in detail. The arrowheads show the direction both of increasing fractionation and also of the relative development of microcline at the expense of orthoclase. At Loch Ailsh, Puklen, and Foyers they also show the order of emplacement of the distinct members of the complexes (no field evidence exists for the Loch Loyal suite). Although this distribution may be modified by shearing stress (Loch Ailsh and Foyers) it seems to be a very generally observed relationship and search of the literature has not revealed any examples of intrusive sequences showing the opposite relationship, although in some cases a systematic distribution of orthoclase and microcline appears to be lacking.[1]

Fig. 6 illustrates that the same relative development of microcline occurs in intrusive sequences following different paths in Ab–Or–An–Qz. This diagram does not, in all cases, indicate the bulk composition of the alkali feldspars because all the Foyers and many of the Loch Loyal rocks are subsolvus in type, so that fig. 6 shows *bulk feldspar* composition. The Puklen and Loch Ailsh rocks are hypersolvus so that fig. 6 suggests their bulk *alkali feldspar* composition. The Ben Loyal rocks are all very *an*-poor, so that their two-feldspar nature implies crystallization at higher P_{H_2O} and lower temperatures than the hypersolvus quartz syenites and soda granites of Puklen. Bulk *alkali feldspar* composition is not directly related to microcline content, except

[1] The Loch Borralan complex near to Loch Ailsh is of this type. Normative *ab–or–an–Q* obtained from eight new analyses of Cnoc-na-Sroine leuco-syenites plot on both the plagioclase and alkali feldspar sides of the two-feldspar surface (MNAB, Fig. 6). Other members of the complex extend into the *ne–ks* volume (Tilley, 1958). No simple fractionation scheme in terms of *ab–or–an–Q* can therefore be suggested for this intrusion; neither is there a systematic distribution of K-feldspar types.

in so far as it locates the sample in a fractionation scheme. This lack of relationship between microcline-abundance and Na:K ratio in these rocks does not support the suggestion of Ferguson (1960) that microcline would evolve more readily in feldspars initially triclinic because of a high Na content.

The grain size of the rock is not related to the presence or absence of microcline; the microcline-bearing Puklen granophyres are very fine-grained rocks enclosed by coarsely crystalline orthoclase-rich augite syenite. The relative sizes of the various units of the intrusions are not related to microcline content. At Puklen the microcline-bearing soda granite body 1·5 km in diameter is enclosed by a 0·25-km thick sheath of orthoclase bearing syenite; at Loch Ailsh the orthoclase-rich rocks are enclosed by the larger, wholly microcline-bearing S3 intrusion. The aplites cited by Marmo (1959) are microcline-bearing veins cutting orthoclase granites of batholithic proportions.

Superficially it might seem that, as falling temperature of crystallization parallels fractionation, so we would expect the polymorph stable at lower temperatures (microcline) to form (for discussion of the evidence for the relative stabilities of orthoclase and microcline see Smith and MacKenzie (1961). The present authors accept the view that the more highly ordered polymorph, microcline, is the stable polymorph at the lowest temperatures). This is oversimple. Although the stability range of triclinic K-feldspars has not been determined experimentally, all available evidence (summarized by Barth, 1969, p. 114) points to a maximum temperature of about 400 °C for microcline to be a stable phase, and maximum microcline may be the stable form at much lower temperatures. Certainly the hypersolvus, and, in all probability, the subsolvus rocks completed crystallization at higher temperatures (Parsons, 1965, suggests 750 °C as the minimum temperature for crystallization of the now microcline-bearing syenite, S3, at Loch Ailsh). Furthermore, the cross-hatched twinning commonly shown by microcline was demonstrated by Laves (1950) to imply that the microcline had formed from crystals initially of monoclinic symmetry, and synthesis studies of alkali feldspars (MacKenzie, 1957; Parsons, 1968; Martin, 1969) show that these rapidly grown crystals, at least, always form with a low degree of order at all temperatures. Even if slowly grown natural crystals do not behave in this way it is unlikely that all

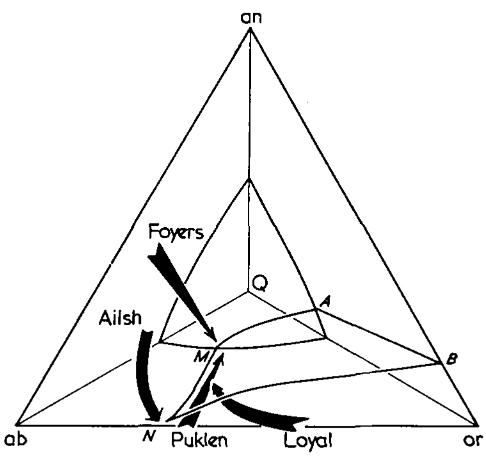


FIG. 6. Quaternary diagram based on Carmichael (1963) showing the primary phase volumes in Ab–Or–An–Qz and the general variation in normative mineralogy of the four complexes in terms of this diagram. ABMN is the two feldspar surface. Loyal and Puklen lie near the base of the tetrahedron, Ailsh on its *ab–or–an* face, Foyers within the plagioclase volume. The arrowheads indicate in each case the theoretical course of fractionation, the observed order of emplacement (except at Loyal where evidence is lacking) and the progressive increase in abundance of microcline at the expense of orthoclase.

the intrusive sequences described here terminated crystallization at precisely the same temperature (see previous paragraph), and therefore the systematic variation in the degree of order attained cannot merely be a reflection of falling temperature. It is not possible that the plutons described that have only microcline (Loch Ailsh, Puklen) or dominant microcline (Foyers) in their final, distinct intrusive phase, yet have orthoclase dominant in earlier phases, all completed crystallization below some common critical temperature above and below which the K-feldspar structures differed in some way that could control their ability to order during subsequent cooling.

Cooling rate will, of course, be one control of the identity of the K-feldspar in the rocks and slow cooling will favour the evolution of the stable form at low temperatures. However, in the intrusions described here (and in many descriptions in petrographic literature) there is no systematic relationship between microcline development and features related to cooling rate, such as grain size or the size of the intrusion. Details of the field data presented here show that feldspars that have shared the same cooling history do not always yield the same final product once cooling is completed. This observation is discussed below.

The catalytic effect of volatiles has been invoked previously by various authors (for example Emeleus and Smith, 1959; Caillère and Kraut, 1960) as an important control of the ordering and unmixing processes in alkali feldspars. Parsons (1965) suggested that volatiles were responsible for feldspar differences at Loch Ailsh, in particular for the very coarse perthites concentrated in certain marginal regions of the intrusion. The observed increase in relative development of microcline with increasing fractionation can be explained as an effect of volatile constituents (and perhaps, more particularly, peralkaline components) and is in keeping with the well-known idea (Tuttle and Bowen, 1958; Bailey and Schairer, 1966; Thompson and MacKenzie, 1967) that these components will increase in relative amount during the normal course of magmatic evolution. Volatiles may be expected to affect the feldspar structure both during the initial growth of the feldspar from the melt, and during the later stages of annealing as the pluton cools. Consideration of the field relations in a number of the intrusions described suggests that the former stage is a most important control of whether microcline will form or orthoclase persist on cooling.

A striking feature of the distribution of K-feldspar polymorphs found for two of the intrusions described (Loch Ailsh, Puklen) is the precision with which the wholly microcline-bearing examples are located in the latest, most fractionated member of the complex, irrespective of its juxtaposition with earlier members, which carry orthoclase as the K-feldspar. Marmo (1959) was also impressed by his similar observations on aplites, and, as reasoned in a previous section, the explanation of MacKenzie and Smith (1961), based on compositional differences, does not seem satisfactory in many instances. At Loch Ailsh orthoclase persists in xenoliths, or even in xenocrysts, enclosed in syenite with highly oblique microcline as the only K-feldspar. Similarly, at Puklen, the later microcline granites are intruded into orthoclase-bearing syenites and even fine-grained granite veins cutting the augite syenites contain microcline while the syenites, with which they are in contact, carry orthoclase. In both these instances it seems that adjacent feldspars (all of which have sodic bulk compositions in the range $Ab_{70}Or_{30}$–$Ab_{50}Or_{50}$, see preceding detailed sections) must have shared the same final cooling history into the temperature range of stability of microcline, and also must have entered this temperature range in the presence of the same intergranular volatiles. Similar considerations apply to the aplites of Marmo (1959) and to the Cnoc-nan-Cuilean body where the only abundantly microcline-bearing rocks are aplites cutting orthoclase-rich syenite. Ragland (1970), while proposing that volatiles present during annealing control the relative abundance of orthoclase and microcline, also points to the difficulties implied by the localization of microcline to particular rock units.

It seems to the present writers that some special mechanism must exist to explain the metastable preservation of orthoclase under these conditions and we suggest that the presence of volatile (or peralkaline or peraluminous) components in the melt *at the time of initial crystal growth* can control details of the structure of the feldspar that determine the ease with which microcline can evolve on cooling. Shearing stress can cause feldspars that would otherwise persist as orthoclase to order to microcline (well shown at Foyers, and see also Bordet and Chauris, 1965), and perhaps prolonged annealing (as in regional metamorphism for example) may have the same effect. Certainly the presence or absence of volatiles during this part of the history of a K-feldspar will be one control of the degree of order achieved. For feldspars having otherwise similar cooling histories, however, the eventual development of microcline is predetermined by the bulk composition of the melt as the crystals grow. This implies that the eventual product of slow cooling is decided at comparatively high temperatures (at least 700 °C in the case of the hypersolvus syenites and granites), well above the range of temperature in which microcline is a stable phase, and also in feldspars considerably more sodic than the almost pure K-feldspar produced on unmixing.

Experimental evidence

The acceptability of the preceding ideas hinges on whether Al–Si order can occur in mixed Na–K-feldspars at magmatic temperatures, and upon whether any mechanism exists whereby bulk composition may influence the degree of order achieved, or the size and shape of ordered regions in the structure. Recent experimental work offers support on both counts. Parsons (1968) showed that ordering could take place in anorthoclases with up to 10 % Or in the region above the solvus, and unpublished work in progress has shown the existence of such ordered forms up to at least $Ab_{70}Or_{30}$. It seems reasonably established that in the temperature ranges in which these plutons would have crystallized some degree of order would exist in the feldspars at magmatic temperatures.

Martin (1969) has presented data on the effect of water content and, more comprehensively, on the effect of sodium disilicate and excess alumina on ordering in albite. His data suggest that albite will order more rapidly in the presence of a rather small percentage of water at 350 °C and 1 kb pressure (about 3 % by weight) than with higher water contents, but he does not present data at higher temperatures or evaluate the equilibrium obliquity of the albite. The synthetic anorthoclases made by one of us (Parsons, 1968) were crystallized in the presence of between 10 and 25 wt % water and no systematic relationship between water content and obliquity can be discerned in these products. MacKenzie (1957) noted that the heating of albites in the absence of water at 700 °C did not produce a change in obliquity. Water is certainly a prerequisite for ordering to occur but the effect of different water contents on the kinetics of ordering needs further study.

Martin (1969) showed that the presence of sodium disilicate has a pronounced effect on the obliquity of albite, and forms close to low albite could be synthesized in two weeks at 300 °C and 10 kb P_{H_2O}. His data do not allow the evaluation of the effect of less than 5 % of disilicate, but his fig. 4 suggests that even small amounts may considerably increase the rate of ordering. A peraluminous environment has the converse effect. Martin suggests that the obliquity of albite depends 'on the a_{Na^+}/a_{H^+} of the fluid in the environment of recrystallization'. Our petrological evidence of the precise restriction of microcline-only rocks to clearly defined phases of intrusion requires a mechanism that operates at the time of crystal growth, rather than during the subsolidus changes, which take place at low temperatures. It seems possible that the bulk composition of the melt during crystallization may influence the feldspar structure in such a way as to control its later ability to order to microcline, either in controlling whether or not an equilibrium degree of order was achieved at the temperature of crystallization, or by controlling the size and shape of the domains of order that must exist at higher temperatures and which, on cooling, subsequently enlarge to appear as the familiar microcline grid twins.

Summary and conclusions

The basic observation of this paper is that in many suites of salic igneous rocks that exhibit variation in bulk composition consistent with current ideas of fractionation in the system Ab–Or–An–Qz, there is a strong tendency for microcline to develop at the expense of orthoclase with progressive fractionation.

A puzzling detail of this link between the identity of the K-feldspar and the relative bulk composition of the rock in which it formed is that the feldspar differences are often sharply defined between rock types and the differences persist even when earlier, less fractionated rocks are intimately associated, in the field, with later, more highly fractionated rocks that have only microcline as the K-feldspar. It is argued that in this setting the feldspar crystals will have locally shared the same cooling history and will have reached the temperature range of stability of microcline in the presence of the same vapour phase.

We are forced to postulate that the metastable preservation of orthoclase in the early feldspars is due to structural features impressed upon them at the time of initial crystal growth at magmatic temperatures. The ability of the crystals to achieve the degree of order of maximum microcline on later cooling is a reflection of conditions in the magma from which they crystallized. We suggest that it is the amount of water, or the peralkaline or peraluminous character of the magma that dictates the eventual product of slow cooling, and that the increase in relative abundance of microcline with increasing fractionation is a reflection of the well-known tendency for water content and peralkalinity to increase with progressive magmatic evolution. Only exceptional treatment, such as application of shearing stress or exceptionally prolonged annealing (as in regional metamorphism or along certain intrusive contacts), can obliterate these early-imposed structural differences.

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REFERENCES

BAILEY (D. K.) and SCHAIRER (J. F.), 1966. *Journ. Petrology*, **7**, 114 [M.A. **18**–21].
BARTH (T. F. W.), 1959 [M.A. **14**–504].
—— 1969. *Feldspars*. New York (Wiley-Interscience).
BORDET (P.) and CHAURIS (L.), 1965. *Bull. Soc. franç. Min. Crist.* **88**, 527 [M.A. **17**–508].
CAILLÈRE (S.) and KRAUT (F.), 1960. Ibid. **83**, 21 [M.A. **15**–49].
CARMICHAEL (I. S. E.), 1963. *Quart. Journ. Geol. Soc.* **119**, 95 [M.A. **16**–579].
CHRISTIE (O. H. J.), 1962. *Norsk Geol. Tidsskr.* **42**, 2, Feldspar vol. 383 [M.A. **16**-464].
DIETRICH (R. V.), 1962. Ibid. 394 [M.A. **16**–462].
EMELEUS (C. H.) and SMITH (J. V.), 1959. *Amer. Min.* **44**, 1187 [M.A. **15**–140].
EULER (R.) and HELLNER (E.), 1961. *Zeits. Krist.* **115**, 433 [M.A. **15**–440].
FERGUSON (R. B.), 1960. *Canad. Min.* **6**, 415 [M.A. **15**–140].
KING (B. C.), 1942. *Quart. Journ. Geol. Soc.* **98**, 147 [M.A. **9**–165].
LAVES (F.), 1950. *Journ. Geol.* **58**, 548 [M.A. **11**–327].
MACKENZIE (W. S.), 1957. *Amer. Journ. Sci.* **255**, 481 [M.A. **14**–38].
—— and SMITH (J. V.), 1961. *Estudios Geológicos, Cursillos y Conferencias*, **8**, 53 [M.A. **16**–75].
MARMO (V.), 1959. *Bull. Comm. Géol. Finlande*, **29**, 133 [M.A. **14**–416].
MARSTON (R. J.), 1967. *Nature*, **214**, 159.
—— 1970. *Quart. Journ. Geol. Soc.* **126**, 331.
MARTIN (R. F.), 1969. *Contr. Min. Petr.* **23**, 323.
NILSSEN (B.) and SMITHSON (S. B.), 1965. *Norsk. Geol. Tidsskr.* **45**, 367 [M.A. **17**–610].
PARSONS (I.), 1965. *Journ. Petrology*, **6**, 365 [M.A. **19**–147].
—— 1968. *Min. Mag.* **36**, 1061 [M.A. 69–311].
—— in press. *Medd. Grønland.*
PULVERTAFT (T. C. R.), 1961. *Medd. Grønland*, **123**, No. 6, 36 [M.A. **16**–82].
RAGLAND (P. C.), 1970. *Lithos*, **3**, 167.
READ (H. H.), 1931. *Mem. Geol. Surv. Scotland* (explanation of sheets 108 and 109).
SMITH (J. V.), 1961. *Amer. Min.* **46**, 1489 [M.A. **16**–76].
—— 1970. *Lithos*, **3**, 145.
—— and MACKENZIE (W. S.), 1961. *Estudios Geológicos, Cursillos y Conferencias*, **8**, 39.
THOMPSON (R. N.) and MACKENZIE (W. S.), 1967. *Amer. Journ. Sci.* **265**, 714.
THORNTON (C. P.) and TUTTLE (O. F.), 1960. Ibid. **258**, 664 [M.A. **15**–220].
TILLEY (C. E.), 1958. *Trans. Edinb. Geol. Soc.* **17**, 156 [M.A. **14**–353].
TILLING (R. I.), 1968. *Journ. Petrology*, **9**, 331 [M.A. 69–2309].
TUTTLE (O. F.), 1952. *Journ. Geol.* **60**, 107 [M.A. **12**–256].
—— and BOWEN (N. L.), 1958. *Geol. Soc. Amer. Mem.* **74** [M.A. **14**–89].
UPTON (B. G. J.), 1960. *Medd. Grønland*, **123**, No. 4, 1 [M.A. **15**–301].
VORMA (A.), 1971. *Bull. Comm. Géol. Finlande*, 246.
WRIGHT (T. L.), 1964. *Amer. Min.*, **49**, 714 [M.A. **17**–88].
—— 1967. Ibid. **52**, 117 [M.A. **19**–308].
YODER (H. S.), STEWART (D. B.), and SMITH (J. R.), 1957. *Ann. Rep. Geophys. Lab. Carnegie Inst. Washington*, **56**, 206.

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