

Homogeneity in alkali feldspars

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Summary. Curves relating $20\bar{1}$ spacing, Or content, and structural state of alkali feldspars are reviewed. Discrepancies between the compositions of heated natural perthitic alkali feldspars determined by this method and by chemical analysis are discussed and shown to be largely consistent with incomplete homogenization.

THE apparently simple relationship between $d_{20\bar{1}}$ and composition in synthetic homogeneous alkali feldspars was first demonstrated by Bowen and Tuttle (1950). Since then a number of new determinative curves have appeared, and the technique has been utilized by numerous workers for the estimation of bulk composition of naturally perthitic alkali feldspars that had been 'homogenized' by heating near the solidus. Some authors have noted discrepancies between compositions determined in this way and those determined by chemical methods; this paper reviews and suggests a reason for these discrepancies.

Curves relating $20\bar{1}$ spacing to composition for synthetic alkali feldspars have been produced by a number of authors, and are compared on fig. 1. Bowen and Tuttle (1950) originally showed a straight line relationship between $d_{20\bar{1}}$ and composition in weight %. Their more recent curve (Tuttle and Bowen, 1958) exhibits a slight departure from a straight line in the form of a flat S-shaped curve. Orville (1963) shows two straight lines for mol. % Or intersecting at Or₇₅. This curve is shown recalculated to weight % in fig. 1, which produces a slight curvature in the two lines. Carmichael and MacKenzie (1964) show a straight line relation for weight % Or for sodium-rich compositions using material supplied by Orville, and Luth and Tuttle (1966, details communicated personally) have produced another S-shaped curve relating 2θ and composition.

The biggest differences between these curves are of the order of 4 weight % Or or 0.05 2θ (Cu- $K\alpha$ radiation). Also shown on fig. 1 are 2θ values for a series of alkali feldspars synthesized by the writer from gels at 850° C and 5000 lb/in² for periods of between 2 and 21 days. All

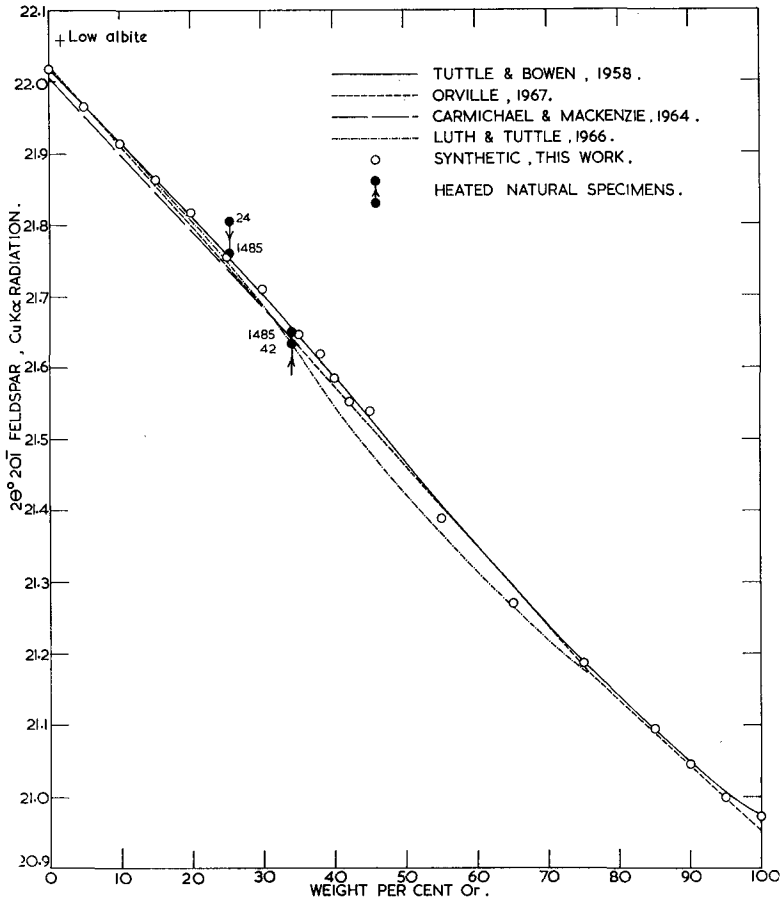


FIG. 1. Published curves relating $2\theta_{201}$ and composition in alkali feldspars, together with 2θ values of a further suite of synthetic alkali feldspars and 2θ values showing change in position of the single 201 reflection of two natural perthites heated for times given (in hours) at 1040°C .

but two lie within 1% Or or $0.01^\circ 2\theta$ of Tuttle and Bowen's (1958) curve. For this reason the latter curve will be adopted subsequently in this paper.

The value of d_{201} is largely sensitive to changes in a since

$$1/d_{201} = 4a^*2 + c^*2 - 4c^*a^* \cos \beta$$

Orville (1967) shows that whilst c and β are only slightly composition dependent in the alkali feldspars, the a cell parameter is strongly

affected by composition, showing a very slight S-shaped departure from a straight line relationship with composition. Particularly in view of the interpretation that will be placed on fig. 2 of the present paper it is interesting to speculate that such a small departure from a straight line relationship could be produced by lack of complete homogeneity even in these apparently homogeneous high-temperature feldspars. The X-ray method is certainly not sensitive enough to detect the requisite small quantity of unmixed material itself.

The $20\bar{1}$ spacing is slightly affected by variations in the structural state of the alkali feldspar, the $20\bar{1}$ spacing of low albite being slightly different from that of high albite (Smith, 1956). MacKenzie (1957) demonstrated that the separation of the 131 and $1\bar{3}1$ reflections of albite was characteristic of the temperature of synthesis of the crystals. A slow change in 131 – $1\bar{3}1$ from values characteristic of high-temperature forms to those characteristic of the temperature of synthesis takes place as crystallization proceeds. Because of the sluggish nature of these reactions, and because of the inability to synthesize microcline directly, the $20\bar{1}$ spacings of synthetic alkali feldspars used to construct the determinative curves have been assumed to be representative of an approximation to a high-temperature structural state. Orville (1967) has recently published lattice parameters for alkali feldspars crystallized at 800°C , 1000 bars H_2O pressure for 5 to 7 days. Experiments at present in progress by the writer show that small differences in the diffraction patterns analogous to those obtained for albite may be found in anorthoclases crystallized at different temperatures, and Orville's values will not apply strictly for feldspars in a fully high structural state, at least for albite-rich compositions. Variations in the value of $20\bar{1}$ for anorthoclases crystallized at different temperatures found by the writer are small, however, so that the assumption that the $20\bar{1}$ curves are appropriate for a high structural state appears to be reasonable.

Orville (1967) has shown a plot of $20\bar{1}$ spacing against composition for a range of synthetic metastable feldspars having a high degree of ordering of Al and Si but a supposedly homogeneous distribution of K and Na. These were made by an ion-exchange procedure involving natural low albite and microcline end members. That the $20\bar{1}$ spacings of apparently homogeneous heat-treated naturally perthitic feldspars may approximate to some such curve joining the $20\bar{1}$ spacings of low albite and microcline has been suggested by the writer (1965) to account for an apparent lack of fit between certain homogenized perthites and

the 'high temperature' $20\bar{1}$ curve. Orville (1967) presents convincing data that the dry homogenization of certain Or-rich perthites gives $20\bar{1}$ values fitting the 'low temperature' curve whilst hydrothermal homogenization of the same material gives values on the 'high temperature' curve. However this effect does not account wholly for the very common discrepancies between $20\bar{1}$ and composition of heated perthitic feldspars, the cause of which will be discussed in the following section.

Applicability to heated natural specimens. Many naturally perthitic alkali feldspars may be rendered apparently homogeneous to X-rays by suitable heat-treatment. An estimate of bulk composition can then be obtained using the $20\bar{1}$ method. This technique has been applied by a number of workers some of whom have pointed out discrepancies between compositions determined in this way and those obtained by chemical analysis. In fig. 2 a total of 120 such analyses are plotted against 2θ of the $20\bar{1}$ reflection. The data of most authors required some recalculation to bring it into the form used in this diagram. A few naturally apparently homogeneous specimens are also included. Sources used were: Smith (1956); Tuttle and Bowen (1958); Orville (1958); Hakli (1960); Carmichael and MacKenzie (1964); Christie and Nilsen (1964); Wright (1964); Parsons (1965); Wilkinson (1965); Heier (1966); Kempe (1966).

Although much of the departure from the curves for synthetic high- and low-temperature feldspars shown by this diagram must be ascribed to the expected shortcomings of chemical methods, imperfect separation of material for analysis, and inaccuracies in the X-ray measurements, there is a notable systematic element that cannot be explained in this way. Relative to both curves there is a pronounced excess of analyses with $2\theta_{20\bar{1}}$ appropriate to more potassic feldspars at the orthoclase-rich end of the series, and a rather less striking excess of analyses with 2θ values appropriate to more sodic feldspars at the albite-rich end. This effect is most pronounced relative to Orville's (1967) low-temperature curve, to which many of these alkali feldspars, which come from plutonic rocks, might be expected to approximate on homogenization.

Reasons that have been suggested for differences in composition determined by chemical and X-ray methods are the influence of ions other than K and Na, especially Ca (Wright, 1964; Kempe, 1966), and the effect of the structural state of the feldspar (Parsons, 1965; Orville, 1967). Neither of these hypotheses satisfactorily accounts for the observed distribution of fig. 2. As Hakli (1960) and Carmichael and MacKenzie (1964) point out, Ca should not affect the $20\bar{1}$ spacing of

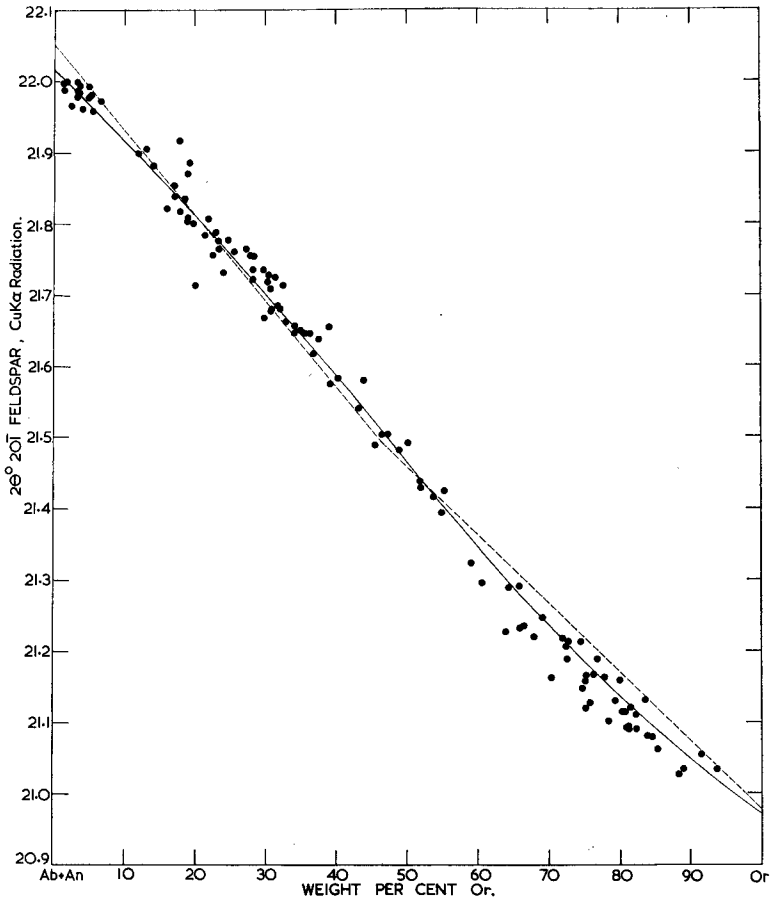


FIG. 2. 2θ $20\bar{1}$ plotted against composition for 120 analysed feldspars from the literature (see text). The solid curve is that for synthetic high-temperature feldspars (Tuttle and Bowen, 1958), the broken curve that for 'low-temperature' feldspars (Orville, 1967), made by ion-exchange in natural low albite and microcline mixtures.

alkali feldspars, as $20\bar{1}$ does not change position significantly throughout the plagioclase series. However, it may act effectively to inhibit the homogenization of unmixed ternary feldspars, as movements of strongly bonded Al will be implied if Ca as well as Na and K are to be distributed at random throughout the structure. Orville's (1967) demonstration that differences in structural state can account for the discrepancies may well be a contributory factor but does not account fully for the

values observed. The effect of a random distribution of K and Na in an Al-Si framework with a high degree of ordering would be to give $20\bar{1}$ spacings approximating to Orville's low-temperature curve, although in fact many of the published values appear to fit the high-temperature curve better.

The two hypotheses that appear to explain the distribution seen in fig. 2 are partial melting and incomplete homogenization. Since the minimum melting composition in Ca-free alkali feldspars is between Ab_{70} and Ab_{60} (Tuttle and Bowen, 1958) and moves towards Or-rich compositions in ternary feldspars, the apparent relative enrichment in albite or orthoclase for crystals on the albitic or Or-rich sides of $Ab_{60}Or_{40}$ on the high-temperature curve could be accounted for by partial melting during the homogenization treatment. However, heat treatments used, in some cases under hydrothermal conditions, have in all cases been below the solidus and glass has not been reported. It seems unlikely that partial melting will have occurred.

Fig. 2 could however be explained if the heat treatments used had been inadequate to fully homogenize the perthitic feldspars. It is well known that perthites respond in different ways and at different rates to heat treatment. Factors governing the ability to homogenize may be complex and related to Ca content, size and shape of perthitic intergrowths, ordering of Al and Si, twinning, and perhaps other features. The criterion of homogeneity usually used, the presence of a single sharp $20\bar{1}$ reflection, is possibly not sensitive enough to detect the small departures from homogeneity needed to account for fig. 2. Durations of heat treatment used by the authors compiled in fig. 2 vary from 2 hours (Wright, 1964) to 1485 hours (Parsons, 1965). The course of change in $20\bar{1}$ in two examples of perthites from the Loch Ailsh intrusion (Parsons, 1965) is shown in fig. 1. Change in $20\bar{1}$ spacing continues long after only one $20\bar{1}$ reflection is detectable by X-rays. After 120 hours the reflection is sharp and symmetrical, but nevertheless still increases slightly in intensity when heating is continued up to 1485 hours. From fig. 1 it will be noted that the direction from which these two samples approach their final $20\bar{1}$ position is different. In the more sodic example the reflection representing the albite phase of the perthite remains the strongest reflection and is joined by the $20\bar{1}$ reflection of the potassium phase of the perthite. In the more potassic example the originally stronger sodium phase reflection of the perthite becomes relatively weaker as homogenization proceeds, eventually joining the now stronger potassium phase reflection as a limb on its higher 2θ side. This difference

in behaviour might be due to the more albitic of these perthites having a monoclinic potassium phase whilst that in the other example is triclinic, close to maximum microcline, or to the difference in bulk compositions. A similar effect has been noted by Czamanske (1965) for a suite of perthites that did not homogenize fully on heating, but nonetheless underwent the same change in relative intensity of the $20\bar{1}$ reflections.

Conclusions. The $20\bar{1}$ spacing may well offer the best method of estimating the bulk Or content of alkali feldspars, as Carmichael and MacKenzie (1964) claim. However, discrepancies between chemical analysis and the $20\bar{1}$ method cannot be ascribed directly to the effect of calcium or to variations in the structural state of the feldspars. It seems that the technique of homogenization adopted by numerous authors fails to fully homogenize the feldspars and that the practical criteria for homogeneity may be inadequate to detect small amounts of unmixed material.

There is great variation in the way perthites from different environments will respond to heating and clearly this diversity is sufficient to merit a degree of experimentation to find the most satisfactory conditions for homogenization for any particular suite of specimens. Indeed some perthites do not homogenize at all (Czamanske, 1965; Tuttle and Bowen, 1958), whilst others, like those described in this work, respond differently to heat treatments. Systematic study of the way a large number of perthites respond to heating has not been carried out, so that simple application of heating procedures to perthites in an endeavour to determine bulk composition may give unreliable results. Without carrying out some checks on composition by wet chemical methods, compositions based on the X-ray method alone may be low by as much as 25 % of the amount of Ab+An present in more potassic feldspars, whilst much better agreement may be expected for albite-rich alkali feldspars. The X-ray method is then most likely to give results high in albite.

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