

An X-ray examination of some potash-soda-felspars.

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[Read January 26, 1939; communicated by Prof. W. L. Bragg, F.R.S.]

I. INTRODUCTION.

THE systematic classification of the potash-soda-felspars is of the greatest interest and importance to the mineralogist, but problems demanding physical investigation are raised in work carried out recently by Dr. Edmondson Spencer,¹ who has described the results of exhaustive chemical, optical, and thermal investigations of a representative collection of these minerals. Dr. Spencer has very kindly allowed us to examine by X-ray methods a large number of his specimens, so that for the first time it has been possible to make a fairly complete survey of potash-soda-felspars of known chemical composition and with accurately measured optical properties.

This paper presents the results of our general survey of potash-soda-felspars with compositions ranging from nearly pure potash-felspar to approximately 50 % soda-felspar. The natural minerals, and the same minerals after specified heat-treatment, have been examined. We do not discuss in detail the interpretation of our results in terms of the atomic structure of felspars;² the subject is extremely complicated, and it is necessary to await the results of other investigations now in progress in this laboratory before attempting a complete structural interpretation.

Spencer's measurements, and especially those concerned with the effects due to various heat-treatments, show conclusively that the classification of the natural minerals can only be achieved when the physico-chemical problem of their constitution has been solved, a mere descriptive classification being impossible by reason of the large number of variables involved. Those of his conclusions which are important for

¹ E. Spencer, *Min. Mag.*, 1930, vol. 22, p. 291; 1937, vol. 24, p. 453; 1938, vol. 25, p. 87.

² W. H. Taylor, *Zeits. Krist.*, 1933, vol. 85, p. 425 [M.A. 5-473]; W. H. Taylor, J. A. Darbyshire, and H. Strunz, *ibid.*, 1934, vol. 87, p. 464 [M.A. 6-177].

the present work may be summarized very briefly as follows: (i) The naturally occurring perthitic potash-soda-feldspars may be divided into two main groups, the orthoclase-micropertthites and the microcline-micropertthites; within each group the optical properties and specific gravity vary in a regular way with the chemical composition, but the variation laws are different for the two groups. (ii) On heating micropertthites for short periods at temperatures not exceeding 850° C. (approx.), the perthitic structure disappears, the schiller vanishes, the specific gravity and refractive indices are reduced, but there is no change in the optic axial angle. On cooling, the perthitic structure reappears and the other changes are reversed. The extent and rapidity of the changes depend upon the composition and the micropertthite type (orthoclase or microcline), but they are essentially reversible and represent solution (on heating) or separation (on cooling) of the soda-rich micropertthitic lamellae. (iii) On heating for long periods at temperatures approaching the melting-point an irreversible change takes place, and the resulting homogeneous or 'sanidinized' feldspars have the low optic axial angle which is associated with natural sanidine. On cooling slowly through the normal perthite exsolution range (800–400° C.), these 'sanidinized' feldspars show no trace of exsolution.

It has already been suggested (Taylor, Darbyshire, and Strunz, 1934) that the lamellae in perthitic structures represent segregations of cations in a tetrahedron framework which is essentially continuous throughout the crystal but is modified locally to accommodate the large potassium ions and small sodium ions respectively. It seems clear that in the processes described in detail by Spencer and summarized above, we are concerned with the problem of diffusion of ions in a crystalline structure and with the slight modifications of structure which are a consequence of the diffusion.

The results of earlier X-ray examinations of perthitic feldspars have been reviewed by Spencer (1937, p. 484) and need not be discussed here. Ito and Inuzuka have recently shown¹ that X-ray powder photographs of micropertthites possess features which are not explicable on the assumption that the perthitic structure is due to sub-microscopic twinning of structurally identical individuals, but are explained if it is supposed that two structures are present, one similar to pure potash-orthoclase, the other similar to albite. This conclusion is in agreement with the results of earlier work, and is confirmed by our results.

¹ T. Ito and H. Inuzuka, *Zeits. Krist.*, 1936, vol. 95, p. 404 [M.A. 7-32].

II. UNTREATED ORTHOCLASE-MICROPERTHITES.

We have determined the axial lengths of orthoclase-micropertthites with compositions which range from 96 % potash-felspar (specimen A) to 44 % potash-felspar (specimen R). The axes have been determined from measurements on X-ray photographs obtained by rotating small fragments of crystal about each of the axes a , b , and c in turn, using molybdenum K_{α} radiation of wave-length $\lambda = 0.710 \text{ \AA}$. The axial lengths for both untreated and heat-treated materials are collected in table I and represented graphically in fig. 1. The accuracy of the measurements is discussed later (section IV). The specimens are lettered in accordance with the lists in Spencer's second paper (1937, p. 455 and pl. XVIII). The conclusions which may be drawn from the experimental data are set forth in the following paragraphs.

(i) *The nature of the lamellar structure.*

Specimens A and C are free from schiller and show no trace of perthitic structure under optimum conditions of illumination (Spencer, 1930). In conformity with this optical behaviour, rotation photographs from these specimens are typical of a single crystal of monoclinic symmetry. By contrast, specimens M and R show a clear blue schiller and perthitic structure, and the rotation photographs contain, in addition to sets of reflections similar to those in photographs from specimens A and C, extra sets of reflections. The extra reflections must be due to the increased soda-felspar content of the crystal, since they become gradually more intense in passing along the series of specimens, from E (17 % soda-felspar), where they first appear, to F (21.5 %), I (30.5 %), J (36 %), M (42.5 %), R (56 %).

The arrangement of the extra reflections due to soda-felspar is relatively simple. On rotating an orthoclase crystal about the a -axis, the principal reflections (due to pure potash-felspar) are arranged in straight horizontal layer-lines (cylindrical film) with separations corresponding to $a_1 = 8.6 \text{ \AA}$. approximately. The additional reflections (due to soda-felspar) are also arranged on layer-lines, parallel to the principal layer-lines, but corresponding to an axial length $a_2 = 8.1 \text{ \AA}$. approximately. The soda-felspar is therefore present in the form of crystals with axis a_2 parallel to the principal axis a_1 of the potash-felspar component.

On rotating about the orthoclase c -axis, no additional layer-lines are seen, all reflections being included in a single set of layer-lines; this

indicates that c_1 (for potash-felspar) and c_2 (for soda-felspar) are parallel and equal in magnitude, the axial length being 7.2 Å. approximately.

On rotating about the orthoclase b -axis, the principal reflections are arranged in layer-lines corresponding to $b_1 = 13.0$ Å. approximately, but additional reflections are observed which increase in relative intensity as the percentage of soda-felspar increases. In these photographs the additional reflections are not arranged in straight layer-lines, but are grouped as would be expected for a crystal with axis b_2 equal in magnitude to b_1 but inclined at a small angle ($\sim 3^\circ$) to b_1 .

The micropertthitic crystal thus contains two components: one has axes $a_1 = 8.6$ Å., $b_1 = 13.0$ Å., $c_1 = 7.2$ Å., and angle $\beta = 64^\circ$, conforms to monoclinic symmetry, and corresponds to pure (or nearly pure) potash-felspar; the other has axes $a_2 = 8.1$ Å. (parallel to a_1), $b_2 = 13.0$ Å. (inclined at a small angle to b_1), $c_2 = 7.2$ Å. (parallel to c_1), and angle $\beta = 64^\circ$, cannot conform dimensionally to monoclinic symmetry, and corresponds to pure (or nearly pure) soda-felspar.

(ii) *The structure of the soda-felspar lamellae.*

Since the unit-cell dimensions of the soda-felspar lamellae are identical, within the accuracy of our measurements, with the dimensions of the unit cell of albite (Taylor, Darbyshire, and Strunz, 1934), it would be natural to assume that the structure is also identical with that of albite. This is not the case.

In a rotation photograph of Amelia albite (Spencer's specimen S), with rotation about the a -axis, the distribution and relative intensities of reflections for layer-lines corresponding to $h = 0, 1, 2$, are similar to those in the additional layer-lines in a photograph of an orthoclase-micropertthite; in layer-lines corresponding to $h = 4, 5$, there are quite obvious differences. The two structures must therefore be similar but not identical. Further examination of the soda-felspar lamellae is to be undertaken, with the aid of Weissenberg photographs. It already seems probable, however, that the major difference between the pure potash-felspar structure and that of the soda-felspar lamellae must be represented by differences in the atomic co-ordinates parallel to the a -axis. For in a photograph of specimen M or specimen R, with rotation about the a -axis, the reflections on the additional layer-lines for $h = 0, 1$, or 2 , correspond in position and relative intensity with those on the principal layer-lines, while reflections on the additional layer-lines for $h = 4, 5$, do not match those on the corresponding principal layer-lines.

The conclusion that the soda-felspar lamellae are not structurally

identical with albite is in agreement with Spencer's statement (1937, pp. 485, 486) that these microperthite lamellae appear to have optical properties different from those of ordinary albite, and that they never show multiple albite twinning.

It is highly probable that the various known forms of soda-felspar are similar but not identical in structure: with the help of further experimental data it may be possible to determine the precise structural features which differentiate ordinary (triclinic) albite, the (triclinic) soda-felspar lamellae of the orthoclase microperthites here described, the monoclinic soda-felspar known as barbierite, and the soda-felspar lamellae of the 'antiperthite' described by Barth.¹ In a private communication, Dr. Edmondson Spencer has suggested that the soda-felspar lamellae of our orthoclase-microperthites may in fact prove to be identical with the soda-felspar lamellae of Barth's 'antiperthite'.

(iii) *Optical and X-ray evidence of lamellar structure.*

It is of interest to examine whether the first appearance of optical inhomogeneity² in the series of orthoclase-microperthites coincides with the first appearance of additional reflections in the X-ray photographs.

Specimens A and C are optically homogeneous even under optimum conditions of illumination, show little or no sign of the changes on heat-treatment which characterize the lamellar microperthites, and give simple X-ray photographs corresponding to a single crystal having monoclinic symmetry.

Specimen D shows a faint blue-white schiller, and lack of homogeneity when examined microscopically, and in its behaviour under heat-treatment falls into the microperthitic group. No additional reflections can be detected on X-ray photographs with rotation about either a -axis or b -axis.

Specimen E is also lamellar, and shows a faint white schiller, but in b -axis photographs no trace of additional reflections can be detected, while in a -axis photographs the additional layer-lines (if present) are exceedingly faint—so faint that their real existence is doubtful.

Specimen F is lamellar with a blue schiller, and in a -axis photographs there are definite, though weak, traces of the additional layer-lines due to soda-felspar lamellae; in b -axis photographs, however, there is still no trace of additional reflections—i.e. all reflections, from both potash-felspar lamellae and soda-felspar lamellae, correspond to unit cells with identical and parallel b -axes.

¹ T. Barth, *Zeits. Krist.*, 1929, vol. 69, p. 476 [M.A. 4-127].

² Optical data are taken from Spencer, 1937.

Specimen I is lamellar with clear blue schiller, and additional reflections are now clearly visible on both a -axis and b -axis photographs.

In the remaining specimens J, K, M, R the additional reflections become increasingly intense as the percentage of soda-felspar increases, but no new phenomenon is encountered.

It appears, therefore, that although specimens D, E, F are optically inhomogeneous, and specimens E (?) and F show additional layer-lines on a -axis photographs, the soda-felspar lamellae only assume their final stable form when the amount of soda-felspar is greater than 21.5 % by weight (specimen F); in specimen I (30.5 % soda-felspar) additional reflections appear in both a -axis and b -axis photographs. Additional evidence for this view is found in the comparison of values of the principal axis a_1 with the axis a of the heat-treated materials (table I and fig. 1): for specimens A, C, D, F the two axes are almost identical (no data for treated E); for specimens I, J, K, M, R the 'treated' a -axis is quite different from the 'untreated' a_1 -axis. (See also section IV.)

It is probable that the soda-felspar lamellae exist, in D as well as in E and F, with a structure having a secondary axis $a_2 = 8.1 \text{ \AA}$., but the axis b_2 identical in magnitude and direction with the principal axis b_1 of the potash-felspar. If this is the case, a -axis photographs of specimen D fail to reveal additional layer-lines merely because their intensity is too small.

It is obviously impossible to decide experimentally whether the same lamellation occurs in A and C as in D, but Spencer (1937) gives reasons for supposing that these specimens (containing 4 % and 9 % of soda-felspar respectively) hold the soda-felspar in solid solution, while D (16 % soda-felspar) is unable to do so.

(iv) *The sanidine structure.*

Specimen G (Eifel sanidine) is glass-clear and free from schiller (although it contains more soda-felspar than specimens E and F) and has an abnormally low optic axial angle. Rotation photographs show no additional reflections due to soda-felspar. Its properties are thus similar to those of specimens which have been homogenized by prolonged heating at a temperature approaching the melting-point.

III. HOMOGENIZED ORTHOCLASE-MICROPERTHITES.

Table I and fig. 1 include data obtained from specimens which have been heated for 300 hours at 1075°C . and so have been homogenized or

'sanidinized'. All the specimens show a reduction in optic axial angle, the reduction being greater the smaller the percentage of soda-felspar (Spencer, 1937).

Rotation photographs about the a -axis now show a single set of layer-lines, corresponding to an axial length which varies from 8.6 Å. for specimen A (4 % soda-felspar) to 8.35 Å. for specimen R (56 % soda-felspar); the axial length diminishes continuously as the percentage of soda-felspar increases.

Similarly, rotation photographs of specimens I, J, K, and M about the b -axis show a single set of layer-lines, the additional reflections due to the soda-felspar lamellae in the untreated material having disappeared; with specimen R, the additional reflections are still visible, but they are relatively much weaker than in the corresponding photograph of the untreated material, and appear to indicate a smaller angle between principal axis b_1 and secondary axis b_2 . (No additional layer-lines can be seen on a -axis photographs of this specimen.)

It seems clear that we are here concerned with two processes. In the first place the lamellae disappear, rendering the felspar optically homogeneous; this change presumably represents the consequence of diffusion of the sodium and potassium ions through the tetrahedron framework, and the gradually diminishing a -axis (in the treated materials) is a measure of the increasing importance of the small sodium ions in determining the dimensions of the structure. In the second place, some additional modification of the structure occurs which prevents the exsolution of soda-felspar and formation of lamellae, and is responsible for the striking change in optic axial angle. This change, it will be noted, cannot be due to the interdiffusion of sodium and potassium ions, since it is observed in specimens A and C (4 % and 9 % soda-felspar, respectively), which contain relatively few sodium ions.

Further work is needed to elucidate the nature of this second change, but we may point out that no marked alterations in axial length occur when specimens A and C are sanidinized, nor do the relative intensities of reflection suffer any marked alteration.

IV. AXIAL LENGTHS OF UNTREATED AND HOMOGENIZED FELSPARS.

In the preceding sections we have described qualitatively the changes observed when orthoclases suffer the specified heat-treatment. A quantitative discussion may be attempted, but it should be emphasized that the experimental method does not permit the attainment of great

accuracy in determining axial lengths, nor does it supply any direct measurement of interaxial angles.

The individual measurements of axial lengths for untreated and homogenized materials are collected in table I and represented graphically in fig. 1. The following conclusions may be drawn:

(i) *The c-axis.*—All experimental values for both untreated and treated specimens lie close to the simple mean $c = 7.17 \text{ \AA}$. Specimens K and R, untreated and treated, may perhaps be exceptions: their points lie below the mean at distances slightly greater than would be attributed to experimental error.

(ii) *The a-axis.*—The length of the unique axis a of the homogenized materials diminishes steadily as the percentage of soda-felspar increases. The straight line YY' represents the variation of axial length with composition, only one point (M treated) lying at a distance from the line greater than that corresponding to probable experimental error.

For specimens A, C, D, E, F, G the lengths of the principal axis a_1 for the untreated materials also lie very closely on the same line, but for specimens I, J, M, R (untreated) the values of a_1 lie quite away from the line YY' and are all extremely close to a mean value $a_1 = 8.635 \text{ \AA}$. (For specimen K the experimental value is much lower than for J which has nearly the same composition: we assume that this discrepancy arises from some unexplained experimental error, and neglect K in finding mean values.)

The gradual decrease in length of the a -axis of the treated materials, with increase of soda-felspar, might be anticipated; the abrupt change in behaviour of the principal axis a_1 of the untreated materials, on passing from F to I, was not anticipated. The secondary layer-lines in a -axis photographs are visible for specimen E (probably) and F (certainly), as well as for I, J, K, M, R, but additional reflections in b -axis photographs (indicating a secondary axis b_2 not coinciding with b_1) appear for the first time in specimen I; these facts (already discussed in section II), considered in conjunction with the observed abrupt change in the magnitude of a_1 , appear to indicate that the properties of the potash-felspar constituent of the perthitic structure are independent of the soda-felspar lamellae only when the amount of soda-felspar exceeds 21.5 % (specimen F).

(iii) *The b-axis.*—The significance of these measurements is less certain than for the axes a and c , and the 'spread' of the experimental points is wider. Of various possible interpretations the following appears to us to be the most probable (p. 348).

TABLE I. Axial Lengths and Densities of Untreated and Homogenized Felpars.

Specimen.	Sym- bol.	Composition by wt. %.		Axial lengths (determined experimentally).						Axial lengths from graphs in fig. 1.		Densities.		Homogenized.		
		Or.	An.	a_1	b_1	c_1	a	b	c	a_1	b_1	$D_{\text{calc.}}$	$D_{\text{obs.}}$	$D_{\text{calc.}}$	$D_{\text{obs.}}$	
Madagascar yellow orthoclase ...	A	96	4	8.60	13.06	7.19	8.57	13.02	7.19	A.	A.	2.538	2.563	0.025	2.543	—
Burma colourless	C	91	6.5	8.60	13.02	7.22	8.60	13.03	7.16	8.56	13.03	2.546	2.563	0.017	2.545	2.555
Burma blue, no. I	D	84	14	8.52	12.96	7.17	8.52	13.06	7.16	8.53	13.00	2.559	2.567	0.008	2.543	2.555
Kalahandi ortho- clase ...	E	83	15	8.52	13.01	—	—	—	—	8.525	12.995	2.560	2.570	0.010	2.543	—
Burma blue, no. II	F	78.5	19	8.50	13.07	7.18	8.49	13.04	7.15	8.505	12.98	2.569	2.571	0.002	2.542	—
Eifel sanidine ...	G	76	21	8.53	13.08	7.17	8.53	13.08	7.17	8.51	13.03	2.559	2.576	0.017	2.559	2.572
Ceylon blue, no. I	I	69.5	29	8.64	12.92	7.19	8.46	13.01	7.17	8.635	12.94	2.541	2.578	0.037	2.538	—
Ceylon blue, no. II	J	64	34	8.61	12.92	7.19	8.42	13.07	7.21	8.635	12.92	2.546	2.580	0.034	2.537	2.555
Burma blue, no. III	K	64.5	34	8.53	12.98	7.13	8.41	12.98	7.11	8.635	12.92	2.545	2.578	0.033	2.537	—
Ceylon blue, no. III	M	57.5	40	8.65	12.84	7.18	8.48	13.01	7.19	8.635	12.895	2.554	2.585	0.031	2.537	2.555
Fredriksvårn anorthoclase ...	R	44	51.5	8.64	12.87	7.12	8.36	12.82	7.12	8.635	12.84	2.566	2.595	0.029	2.537	2.569

The description of the specimens, the lettering, and percentage compositions, are taken from Spencer, 1937. The axial lengths and densities are discussed in detail in section V of the text.

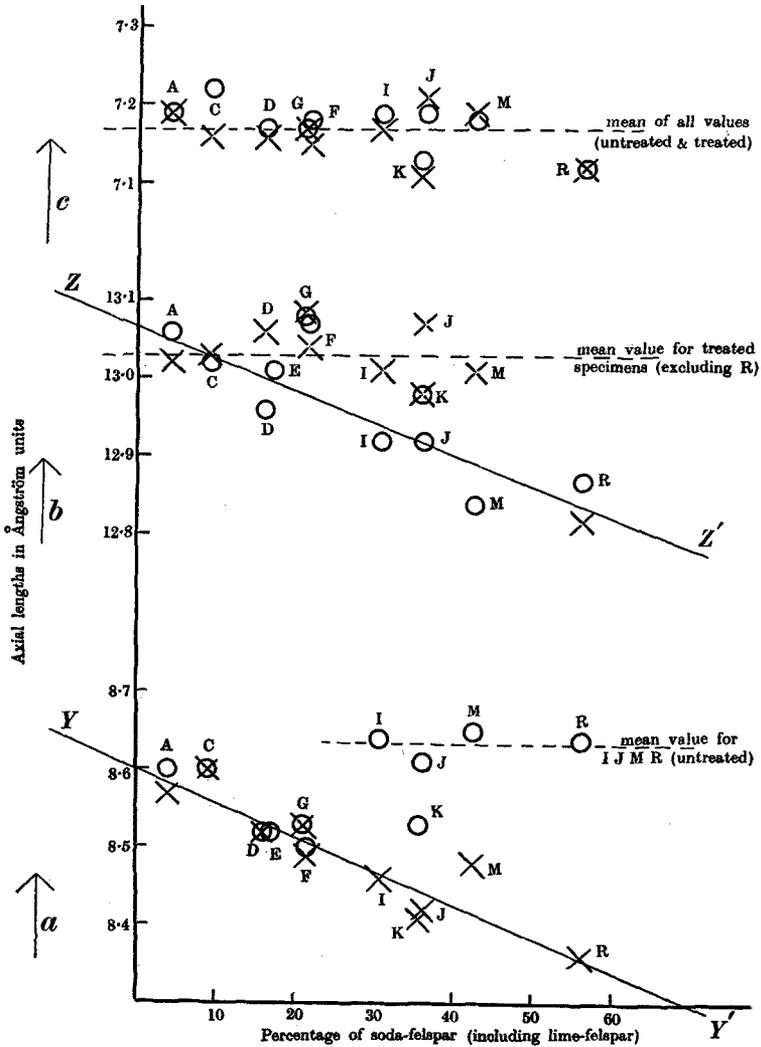


FIG. 1. Axial lengths of untreated and homogenized feldspars.

The axial length in Å. is plotted vertically, the percentage of soda-felspar (including lime-felspar) horizontally. Circles and crosses represent untreated and homogenized (treated) specimens respectively. The lettering of the specimens corresponds to that of table I.

For the homogenized materials A to M, individual values are grouped about a mean value $b = 13.03 \text{ \AA.}$; for R the observed value $b = 12.82 \text{ \AA.}$ is markedly lower. This difference probably corresponds to the fact (discussed in section III) that in b -axis photographs of R (treated), additional reflections can be distinguished which are similar to those in photographs of untreated material, whereas in specimens up to M homogenization is apparently complete.

For the untreated materials, excepting F and G, the experimental values are grouped about the straight line ZZ' . Specimen G (sanidine) is naturally homogeneous, and would therefore be expected to conform to the law for *treated* materials, as in fact is the case. Specimen F remains as the only specimen showing unexplained departure from the variation law represented by the line ZZ' .

To test the correctness of the above interpretations of the experimental data we have calculated the densities of untreated and homogenized materials and compared these values with those determined directly. The results are collected in table I.

The procedure adopted in finding the density by calculation ($D_{\text{calc.}}$) is as follows. For specimens A and C (untreated), which are homogeneous, a unit cell with axes a_1, b_1, c_1 , and angles $\alpha = \gamma = 90^\circ, \beta = 64^\circ$, is assumed to contain amounts of potash-felspar and soda-felspar, in the proportions given by Spencer's analyses, such that there are altogether 32 oxygen atoms per unit cell. For specimens D, E, F, I, J, K, M, R, (untreated), the potash-felspar and 8% of the soda-felspar¹ are assumed to have a structure with a monoclinic unit cell a_1, b_1, c_1, β , and the remaining soda-felspar a structure with monoclinic unit cell a_2, b_1, c_1, β ; the density of the micropertthitic crystal is finally deduced by considering the relative amounts of the two structures present. Lime-felspar and barium-felspar are assumed to have the same structures as soda-felspar and potash-felspar respectively. The values $a_2 = 8.12 \text{ \AA.}, c_1 = 7.17 \text{ \AA.}, \beta = 64^\circ$, are assumed throughout, and values of a_1 and b_1 appropriate to the composition of each specimen are deduced from the lines YY' and ZZ' of fig. 1.

For all treated specimens and for specimen G (untreated) a single monoclinic unit cell is assumed with axes $b = 13.03 \text{ \AA.}, c = 7.17 \text{ \AA.}, \beta = 64^\circ$, and axis a deduced from the line YY' .

For the untreated specimens the observed densities ($D_{\text{obs.}}$) are taken from Spencer's data; we have determined the densities of six

¹ Potash-felspar may retain 8% of soda-felspar in solid solution (Spencer, 1930, pp. 313, 330-331).

treated specimens by flotation in a mixture of chloroform and bromoform.

It is clear from table I that all calculated densities are lower than the corresponding observed densities; this is presumably due to a slight inaccuracy in the calibration of our apparatus. (It should be noted that each axial length a , b , c must be determined to 1 part in 750 if the density is to be accurate to 1 part in 250, i.e. if the difference ($D_{\text{obs.}} - D_{\text{calc.}}$) is not to exceed ± 0.010).

For the untreated specimens A to G the difference ($D_{\text{obs.}} - D_{\text{calc.}}$) ranges from 0.025 to 0.002, with a mean value 0.013; for untreated specimens I to R the difference ranges from 0.037 to 0.029, with a mean value 0.033. The abrupt change in passing from F to I is again evident (G is omitted from consideration of the series of micropertthitic structures); the discrepancy between calculated and observed densities is increased in specimens I, J, K, M, R because in making the calculation a *monoclinic* cell a_2 , b_1 , c_1 , β is assumed, whereas the soda-felspar cell is really triclinic (see section II), and so for given axial lengths and given angle β is of smaller volume than the assumed monoclinic cell.

For the treated specimens, with the exception of G, all calculated densities are nearly identical, the actual figures showing a slight decrease from A to R. By direct observation, the density is 2.569 for R, 2.572 for G, and 2.555 for C, D, J, M, which may be taken as representative of the range from A to R, excluding G. The high value calculated for G is due to the barium content of the specimen, and the observed value is correspondingly high. The high value observed for R is probably due to the abnormally short b -axis, 12.82 Å. against the mean value of 13.03 Å. assumed in calculating the density.

The general agreement between observed and calculated densities, for both untreated and treated specimens, encourages us to believe that our attempt to give a quantitative interpretation to the measurements of axial lengths is based on something more than pure speculation, and that the regularities which are apparent in fig. 1 are not due to coincidence arising out of experimental inaccuracies.

V. CONCLUSION AND SUMMARY.

We have examined by X-ray methods a series of micropertthitic potash-soda-felspars of varying composition, in their normal state and after homogenization by prolonged heating at high temperatures. In the untreated micropertthite the crystal contains two closely related structures, one (containing the potash-felspar) being monoclinic, the

other (containing most of the soda-felspar) being triclinic but different from ordinary albite. In the homogenized specimens only one single structure is present. The cell-dimensions of both untreated and treated specimens depend in a rather complicated way upon the amount of soda-felspar present.

We wish to express our thanks to Dr. E. Spencer for the supply of materials, and for helpful discussion during the course of the work. One of us (S. H. C.) is indebted to the Government of Honan for the grant of a scholarship during the tenure of which this work was carried out.
