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X-RAY STUDY OF THE STRUCTURE OF MOONSTONES

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1. INTRODUCTION

MOONSTONES which represent the potash-soda series of mixed feldspars lying in the compositional range Or_{85} Ab_{15} to Or_{40} Ab_{60}, exhibit a characteristic optical effect, commonly known as schiller. To all external appearances, they are monocrystals with symmetry elements characteristic of the monoclinic system. The optical phenomenon displayed by them, however, betray their complex nature, for the phenomenon arises due to optical heterogeneity of the medium. X-ray methods may therefore be expected to throw light with regard to the precise nature of the material responsible for the heterogeneity. Indeed, earlier X-ray investigations of Kozu and Endo (1921), Chao, Smare and Taylor (1939), Chao and Taylor (1940) and Ito (1950) have shown that moonstones possess a complex structure; the soda-feldspar having segregated as a distinct crystalline phase with triclinic symmetry, from the monoclinic potash-feldspar. The fact that the existence of the former feldspar as a separate phase is directly connected with the schiller phenomenon is shown by heat-treatment studies on moonstones. In recent years, X-ray studies by Mackenzie and Mackenzie and Smith (1955, 1956, 1958) on alkali feldspars have shown that there exist high and low temperature perthites. Further, the unmixed soda-phase has a considerable amount of potash feldspar in solid solution as is found from α^* and γ^* values of the former phase. All previous X-ray studies have established the fact that while the potash feldspar phase exist with monoclinic symmetry, the soda-feldspar crystallites in the moonstones have triclinic symmetry and are twinned either according to pericline or albite law. However, the explanations advanced with regard to the factor influencing the type of twinning assumed by the soda-feldspar crystallites are contradictory and have not provided the correct answer to this interesting question. Further, there are some points with regard to the structure of Korean Moonstones needing clarification, in view of the different interpretations given by Chao and Taylor on the one hand and by Ito on the other.

The optical effect exhibited by moonstones was investigated in great detail in this laboratory several years ago and an account of the same appeared in these *Proceedings* (1950, 1953). The need for an independent X-ray investigation was keenly felt at that time to elucidate on the complex nature of moonstones whose optical behaviour was thoroughly understood. For various reasons such a study could not be carried out then. Recently another type of moonstone from the Coimbatore District in South India came to our notice which exhibits, apart from a golden yellow schiller, other highly characteristic optical effects, namely, polarised transmission and forward diffusion effects. Subsequent to our previous studies, a fair number of Korean moonstones exhibiting schiller were added to our collection. These circumstances and for the reasons already set forth, a thorough investigation by X-ray methods was undertaken recently on all the above-mentioned materials by single crystal methods. Of these, the Coimbatore moonstone is studied for the first time. As a precise investigation was called for, the Weissenberg method was extensively employed along with rotation methods. Both natural and heat-treated materials were studied and accurate measurements of the lattice constants were obtained. The results have greatly aided in putting forth an explanation for the differences in the twinning behaviour, based on the differences between the lattice constants of the constituent feldspar in moonstones. Further, the studies by the Weissenberg method have led to a determination of the precise nature of the crystallographic relationships of the two feldspars and the structural characters of the soda-feldspar phase. In this communication the results of the experimental study and the conclusions drawn from them would be set forth in detail.

2. ROTATION DIAGRAMS OF MOONSTONES

In the oscillation and rotation photographs of all moonstones, two kinds of reflections are distinguishable; a set of strong, sharp reflections and a set of weak and somewhat diffuse ones. They evidently have different origin, the stronger ones arising due to the potash-feldspar phase and the latter due to soda-feldspar. Certain common features are noticeable in the X-ray rotation photographs of the three moonstones (Coimbatore, Ceylon and Korean varieties). The principal reflections in the *b*-axis rotation photographs are symmetrically disposed with respect to the zero layer, revealing thereby the presence of a symmetry plane perpendicular to that axis. The *a*-axis photographs of all moonstones examined consist of a double set of layer lines, one set comprising of strong reflections and another of weaker reflections. The latter are spaced wider than the former and the corresponding axes lie very nearly parallel. In every case, the morphological axes of the moonstone are found to coincide with the crystallographic axes of the potash feldspar phase.

The *b*- and *c*-axis rotation photographs divide these three moonstones into two types. In one type, while the X-ray photograph for the *c*-axis rotation exhibits features characteristic of a simple layer line diagram, the *b*-axis rotation photograph is complex. In the latter, strong reflections fall accurately upon layer lines and weak subsidiary reflections accompany these. In general, each principal reflection is accompanied by a pair of satellites, one lying above and the other below the layer line, on curves of constant ζ. The distribution of the subsidiary reflections is characteristic of an oscillation photograph with misset axes and their symmetrical appearance about the layer line indicates the presence of two such axes, equally inclined to the principal axis. In the second type, the position is reversed, namely, that while the *b*-axis rotation picture has the characteristics of a simple layer line diagram, the rotation diagram for the *c*-axis is complex. The distribution of the subsidiary reflections in the photograph is indicative of the existence of two subsidiary *c*-axes, symmetrically inclined to the principal axis.

To the first type belong the Ceylon and Coimbatore moonstones and to the second type the Korean variety. Figures 1 and 2 reproduce *b*-axis oscillation photographs of the Coimbatore and Korean moonstones respectively. In recording these, the crystal was oscillated through a range of 15° with (001) plane set parallel to the X-ray beam at the middle of the oscillation range (*see* Mackenzie and Smith, 1955). In the oscillation photograph of the Korean moonstone, the principal reflection can be seen accompanied by weaker subsidiary reflections. The features described for the Coimbatore and Ceylon moonstones are clearly to be seen in Fig. 1.

The foregoing facts become intelligible, if it is assumed that in all moonstones, the potash feldspar exists as a truly monoclinic component and that the soda-feldspar crystallites possess triclinic symmetry and are arranged in the mutual orientation characteristic of albite twin law in the case of Coimbatore and Ceylon moonstones, and in the orientation characteristic of the pericline twin law in the case of Korean moonstone.

3. WEISSENBERG X-RAY DIAGRAMS

Zero layer Weissenberg photographs about *a*, *b* and *c* axes were recorded for all the varieties of moonstones, employing CuK$_\alpha$ radiation and a Unicam Weissenberg X-ray Goniometer.

a-axis.—In the *a*-axis photograph of the Coimbatore moonstone, two subsidiary reflections appear close to the principal ones from (*okl*) and (*ool*) planes, while in the case of Korean moonstones, the *oko* and *okl* spots are accompanied by two such reflections. Single spots appear for the *oko* reflections in the former moonstone and for the *ool* reflections in the latter. The *a*-axis zero layer photograph of the two moonstones are reproduced in Figs. 3 and 4. In the photograph for the Korean moonstone the *oko* reflections apear as long streaks but these actually consist of three spots merging into each other. The triple character of the reflections can be seen in *okl* reflections.

b-axis.—In photographs for the Coimbatore moonstones, the principal reflections from the (*ool*) and (*hol*) planes have two close satellites and the corresponding reflections in the case of Korean moonstone consist of single and double spots. In both cases the *hoo* reflections are accompanied by single subsidiary reflections which lie more or less in the same line as the principal reflection.

c-axis.—In the *c*-axis zero layer photograph of the Coimbatore moonstone all the reflections except those from (*oko*) planes are doubled. But in the photographs for the Korean moonstone it is seen that the principal reflections from the (*hko*) and (*oko*) planes have two subsidiary reflections, the *hoo* reflections appearing doubled.

The X-ray photographs of Ceylon moonstones of both the blue and white schiller variety exhibit characters similar to that described for the Coimbatore variety. The differences in the features observable on the Weissenberg photographs arise due to the different twinning laws of the soda-feldspar phase. The two individuals of the albite twin have the (010) plane common and are related to each other by a rotation through 180° about the normal to this plane. While this operation leaves the b^* axes coincident, the c^* axes of the reciprocal lattice do not coincide and are symmetrically inclined with respect to the c^* axis of the monoclinic phase. Therefore planes involving l indices of the soda-feldspar have two points in the reciprocal lattice, giving rise to two subsidiary reflections. In the case of pericline twinning, the individuals are related by a rotation through 180° about the *b*-axis, which operation, while bringing the a^* and c^* axes into coincidence leave the b^* axes non-coincident. The latter are symmetrically inclined with respect to the b^* axis of the monoclinic phase. Now, the planes involving k indices are doubled and give rise to two subsidiary reflections. The (*hoo*) spacings differ for the two feldspars and therefore reflections from these are doubled.

A noteworthy feature observable on Weissenberg photographs is that only single spots appear from the triclinic soda-feldspar phase accompanying the principal reflections from (*hoo*) planes. This result leads to the important conclusion that the interaxial angle γ is very nearly equal to 90° for the soda-feldspar phase in these moonstones.

The zero and *n* layer Weissenberg photographs of the Korean moonstone rotated about the *b*-axis reveal that a monoclinic potash phase and a pericline twinned soda-phase coexist in it. No evidence for the presence either of a triclinic potash phase or monoclinic soda-phase could be found in the photographs. This is in agreement with the results obtained by Chao and Taylor (*loc. cit.*) for Korean moonstone.

4. X-RAY PATTERNS OF HEAT-TREATED MOONSTONES

When moonstones are heated to about 850° for a few hours, the schiller vanishes. This fact was reported earlier by E. Spencer (*loc. cit*). Some of the heat-treated specimens were examined by Taylor and collaborators (*loc. cit.*).

In the present series of studies, the moonstones were heat-treated, and their X-ray patterns were recorded both by rotation and Weissenberg methods. The heat-treatment was carried out at 900° C. for about three hours and the specimens were then allowed to cool to room temperature. In X-ray photographs of heat-treated moonstones, all subsidiary reflections characteristic of the untreated material vanish. The detailed characteristics of the rotation diagrams which consist of single set of layer lines, and the Weissenberg photographs reveal that the heat treated material possess monoclinic symmetry. The lattice constants of the heat-treated moonstones were obtained from a measurement of the high-order reflections in the respective Weissenberg photographs and are presented in Table I along with the data for the potash-feldspar phase of the untreated material.

TABLE I

Lattice Constants of Natural and Heat-treated Moonstones in A.U.

Lattis Con-stant	Coimbatore moonstone		Ceylon moonstone		Korean moonstone	
	Natural	Heat-treated	Natural	Heat-treated	Natural	Heat-treated
a^*	7·70	7·58	7·72	7·619	7·63	7·50
b^*	12·94	12·94	12·84	12·84	12·94	12·94
c^*	6·40	6·42	6·42	6·45	6·39	6·40
a	8·57	8·46	8·59	8·47	8·49	8·34
b	12·94	12·94	12·84	12·84	12·94	12·96
c	7·20	7·14	7·17	7·17	7·12	7·13

It will be seen from a scrutiny of the data presented in the table that a significant contraction along the *a*-axial dimension has taken place on heat-treatment. This change is found to be proportional to the amount of soda-feldspar that has gone into solution. In estimating the contraction, the value for the natural material should be taken as 8·61 Å which is the value for pure orthoclase. If a certain percentage of soda-feldspar has already gone into solution in the potash phase of the untreated material, the actual value for the latter would be brought down. This is what has happened in the case of Korean moonstone. The difference in the *a*-axial dimension between heat-treated Korean moonstone and orthoclase is nearly double that of such differences for the other two moonstones, as is to be expected, in view of the higher percentage of soda-feldspar present in it, nearly 60% as against 30 to 33% in the others.

5. DISCUSSION OF THE RESULTS

The results obtained raise the interesting question, namely, what is the factor that is responsible for the soda-feldspar crystallites in the moonstones assuming a orientation characteristic of albite law in one case and pericline law in the other. Chao and Taylor (*loc. cit*) classified Spencer's specimens into two groups; those having albite twinned soda-phase whose lattice angles were close to low-albite, and those having pericline twinned soda-phase whose lattice angles did not agree with low-albite. They were not aware of the existence of high albite at that time which was proved beyond doubt by Tuttle and Bowen (1950), and gave a tentative explanation for this difference based on bulk chemical composition. Ito and Sadanaga (1952) put forward a similar suggestion. Laves (1952) thought that the type of twinning was controlled, depending upon whether the soda-phase has low or high albite structure. This suggestion was however contradicted by Smith and Mackenzie (1954).

Table II presents the *a*-axial dimensions of the potash phase and the lattice angles α^* and γ^* of the soda-feldspar phase for the three moonstones studied. The latter were obtained from measurements on Weissenberg photographs. The lattice angles for low albite are also given in the last column.

Mackenzie and Smith have shown that from the α^* and γ^* values of the soda-phase, the chemical composition of that phase could be determined. As remarked earlier, the present investigation shows that the composition of the monoclinic potash phase is revealed by the *a*-axial dimension of that phase. The *a*-axial dimension of the Korean moonstone whose bulk chemical composition is $Or_{40}\,Ab_{60}$ indicates that the potash phase in it holds an appreciable proportion of soda-feldspar, amounting to nearly 25 to 30% in solid

TABLE II

Phase	Lattice constant	Korean moonstone	Ceylon moonstone	Coimbatore moonstone	Albite
Potash ..	a	8·49Å	8·59Å	8·57Å	..
Soda ..	a^*	88° 30′	86° 30′	87° 24′	86° 24′
Soda ..	γ^*	90°	90° 20′	90° 20′	90° 29′

solution. The a^* and γ^* values of the soda-phase in the same moonstone correspond to a chemical composition $Or_{30}Ab_{70}$.

It will be seen from Table II that in the case of Korean moonstones, the differences in respect of both the a-axial dimensions and the lattice angles a^* and γ^* between the two constituent feldspars are lessened, this essentially arising out of solid solution of an appreciable proportion of one phase in the other. Such circumstances favour pericline twinning of the soda-phase. On the contrary, these differences are greater in the case of Coimbatore and Ceylon moonstones and the two feldspar phases are very close to orthoclase and low-albite. When the soda-feldspar phase thus demands full-triclinic symmetry of low-albite and the a-axial dimension of the potash phase correspond to that of orthoclase, twinning takes after the albite law. The principal factor that seems to influence the nature of twinning is therefore the narrowing down of the differences in respect of the a-axial dimension of the two feldspars.

In conclusion the author wishes to express his sincere thanks to Sir C. V. Raman, F.R.S., N.L., for his kind interest in this work.

SUMMARY

X-ray studies on three different types of moonstones, namely, Ceylon, Coimbatore and Korean varieties have been made by single crystal rotation and Weissenberg methods. The results are consistent with the explanation that the triclinic soda-feldspar crystallites which has segregated out of monoclinic potash feldspar are twinned according to albite law, in the former two, while the twinning is according to pericline law in the latter moonstone. The view is put forward that the narrowing down of the differences, firstly of the a-axial dimension between the soda-feldspar and potash feldspar phases, and secondly between the lattice angles of the two feldspars result in pericline

twinning of the soda-phase, while albite twinning results when the soda-phase demands full triclinic symmetry and the potash phase has the *a*-axial dimension corresponding to that of orthoclase. The detailed features of the Weissenberg X-ray diagrams of Korean moonstones reveal that there exist in it a monoclinic potash phase and a pericline twinned triclinic soda-phase, in agreement with the results obtained by Chao and Taylor. X-ray results also indicate that the lattice angle γ of the triclinic soda-phase in these moonstones is very nearly equal to 90°.

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EXPLANATION OF PLATES

PLATE VI

FIG. 1. *b*-axis oscillation photograph of Coimbatore moonstone, oscillated through a range of 15° with the (001) plane in the middle of the oscillation range. Fainter reflections can be seen accompanying many of the spots. The (242) reflection is enclosed in square brackets and the subsidiary reflections can be seen, one on either side of the layer line.

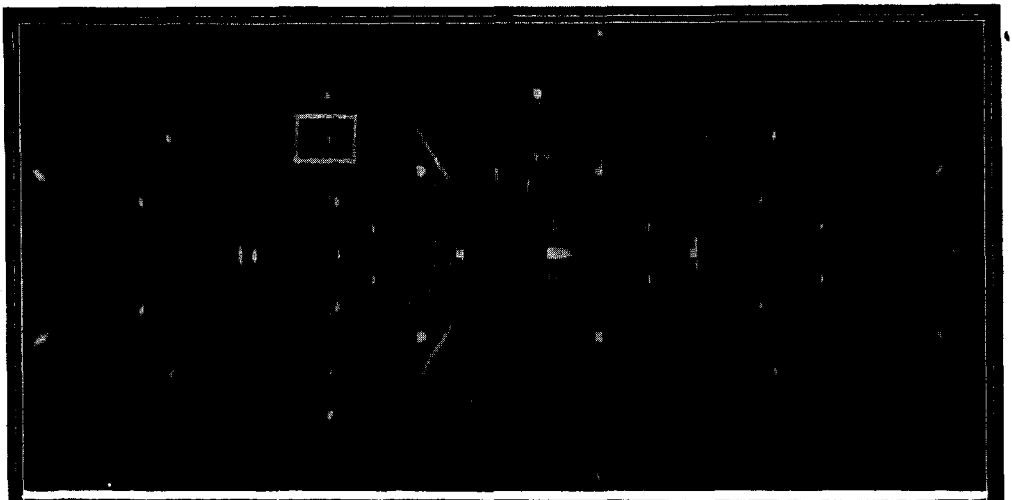

FIG. 1

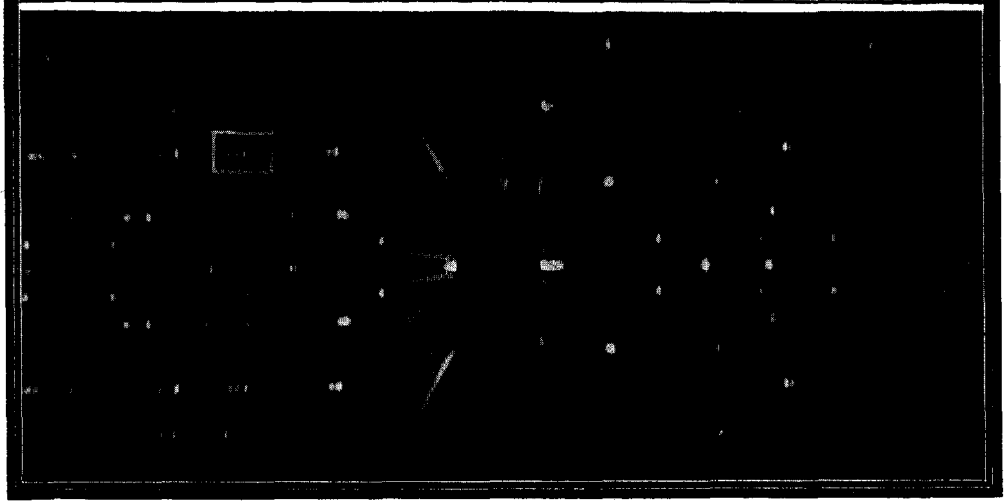

FIG. 2

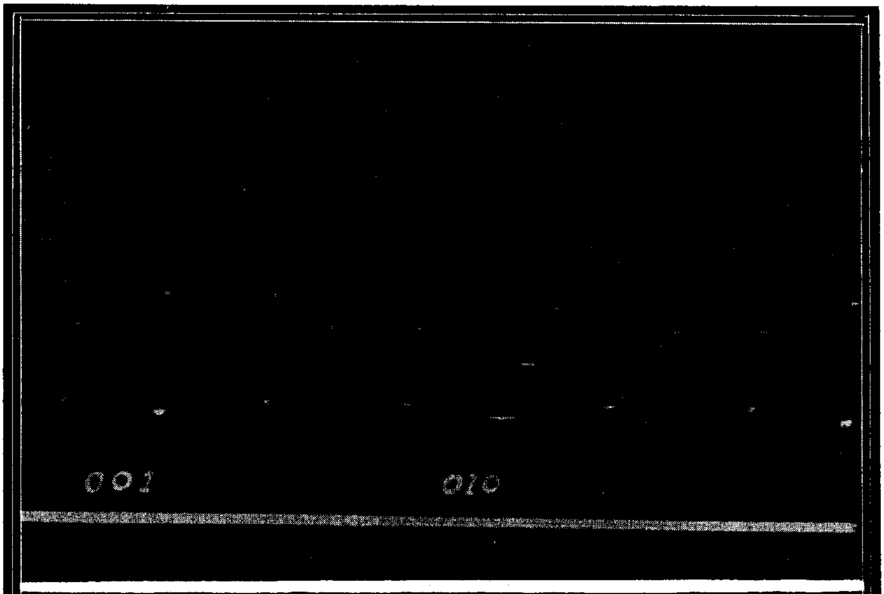

FIG. 3

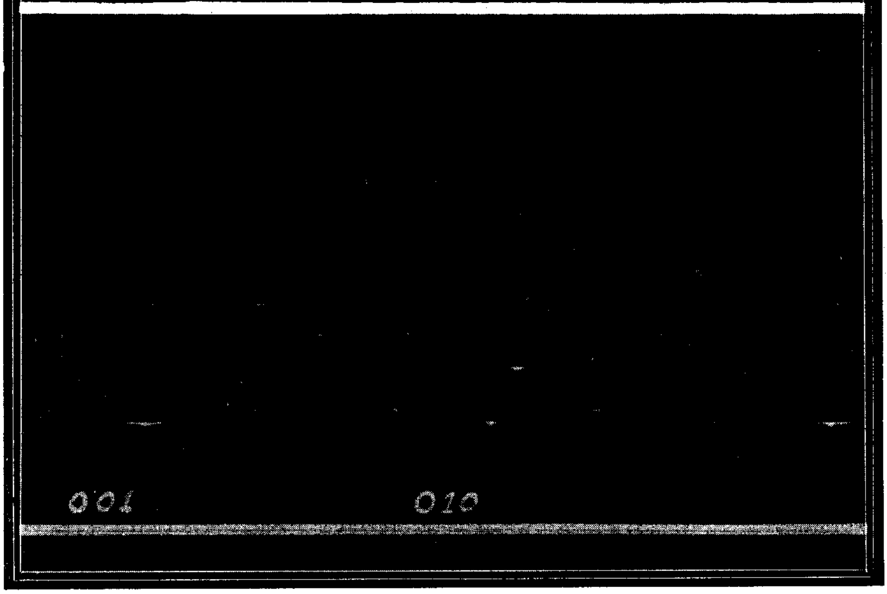

FIG. 4

FIG. 2. Same oscillation photograph as Fig. 1 with Korean moonstone. Principal reflections are accompanied by subsidiary reflection, but the latter fall in the layer line. The (442) reflection is enclosed in square brackets. The two subsidiary reflections can be seen on the left.

PLATE VII

FIG. 3. *a*-axis zero layer Weissenberg photograph of the Korean moonstones. The 001 and 010 reflections are marked. The triple character of the 010 reflections can be seen clearly on high angle spots.

FIG. 4. Same as Fig. 3, but with Coimbatore moonstone. The tripling of the 001 reflections can be clearly seen in the photograph.