STUDIES IN THE MICA GROUP; RELATIONSHIP BETWEEN POLYMORPHISM AND COMPOSITION IN THE MUSCOVITE-LEPIDOLITE SERIES*†

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Abstract

A determination of the crystal structures of about 50 muscovites and lepidolites of known composition has demonstrated the existence of a complete series between muscovite and lepidolite. The polymorphic variation in this series can be correlated with chemical composition, particularly with Li_2O content. A new variant of the muscovite structure has been discovered for which the term *lithian muscovite* is proposed. This modification crystallizes with a structure very similar to that of normal muscovite and apparently links heptaphyllite muscovite with octophyllite lepidolite. Normal musocvite may have as much as 3.3% Li_2O. Micas with 3.4–4.0% Li_2O are generally characterized by poor crystal development and anomalous optical properties owing to their transitional structures. Lepidolites with 4.0–5.1% Li_2O generally crystallize as the 6-layer monoclinic polymorph whereas those with more than 5.1% Li_2O have usually crystallized as the 1-layer monoclinic polymorph. Hexagonal lepidolite is commonly associated with the 1-layer monoclinic polymorph, and both have essentially identical compositions. Twinning may be responsible for the hexagonal form. Polymorphic modifications of lepidolite may occur across sheets and also along the *c*-axis directions in single books. Owing to considerable overlapping of indices and 2V, the various polymorphs may not be distinguished on the basis of optical constants alone.

Introduction

On the basis of chemical evidence it has been suggested by several investigators that a series exists between muscovite and lepidolite. This series is identified particularly by an increase in the Li_2O content, although other chemical changes, such as an increase in SiO_2 and F, and a decrease in Al_2O_3 are also characteristic. However, no serious effort has previously been made to correlate successive chemical changes with the four polymorphs of these micas described by Hendricks and Jefferson (1939). This paper proposes to correlate the successive change in Li_2O content with polymorphism in the muscovite-lepidolite series.

In order to attempt this correlation about 50 specimens of analyzed muscovites and lepidolites with more than 1.5% Li_2O were obtained from about a dozen investigators, museums and laboratories throughout the world. Most of the micas used in this study have been analyzed within about the last 25 years and the results of most have been recorded in the literature. The structures of these, in addition to the structure of about

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450 specimens of unanalyzed muscovites and lepidolites, were investigated primarily by the Weissenberg method. In those cases where crystal development or optical extinctions were poor, the powder *x*-ray method was used. Copper radiation was used throughout.

The first *x*-ray studies on the micas were made by Mauguin (1927, 1928*A*) who measured the units of structure of several varieties of known composition and density. He observed that the *c*-axis of biotite was apparently only half as long as that of muscovite; thus he was the first to establish polymorphism in the micas by means of *x*-ray analysis. Jackson and West (1930, 1933) made the first detailed study of the structure of mica (muscovite) and, in addition, confirmed in greater detail the structure of mica and other layered silicates proposed by Pauling (1930) on the basis of his coordination theory. The most detailed study of the mica group as a whole was conducted by Hendricks and Jefferson (1939) who found numerous polymorphs. In all, seven different polymorphic modifications, embracing 3 crystal systems were found among 100 specimens. In discussing their observations on the lepidolites analyzed by Stevens (1938), Hendricks and Jefferson (1939) stated that there was no evident correlation of composition with the 3 different polymorphs found in these micas. Muscovite they noted was unique among the micas in that it crystallized with but one structure, the 2-layer monoclinic muscovite type. However, Axelrod and Grimaldi (1949) have described a hydrothermal muscovite that has a small, variable 2V and has crystallized with 3 layers in a monoclinic (?) unit cell. This determination is being restudied, but in any case, such muscovite is exceedingly rare and for the purposes of this discussion only the 2-layer muscovite structure (normal muscovite) need be considered.

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STRUCTURE OF MUSCOVITE

The distinguishing feature of the muscovite structure (space group $C2/c$) is that it is distorted from the ideal mica structure. This is revealed by the presence of certain reflections, $(06l)$ with l odd, which should normally be absent in the ideal arrangement on the basis of the structure factor calculations of Jackson and West (1930, 1933). This distortion results from an incomplete filling of the octahedral positions and is considered by Hendricks and Jefferson (1939) to be the factor permitting only the two-layered structure for muscovite (Fig. 1). Herein also lies the reason why Winchell's grouping of the micas into heptaphyllite and octophyllite divisions is correct, for these terms simply mean that the unit cell contains seven and eight atoms respectively excluding (O, H, and F). Muscovite, $KAl_2(AlSi_3)O_{10}(OH,F)_2$, is the type heptaphyllite and phlogopite, $KMg_3(AlSi_3)O_{10}(OH,F)_2$, the type octophyllite. This relation may also be expressed by saying that the heptaphyllites have only 2/3 of the octahedral positions filled, whereas the octaphyllites have all such positions occupied. The presence of $(06l)$ reflections with l odd must imply departure from the ideal muscovite structure given by Jackson and West (1930, 1933). Hendricks and Jefferson (1939) report these reflections absent in the two-layered biotite-like micas. Thus muscovite (2-layered) has a structure different from that of the 2-layered octophyllite micas.

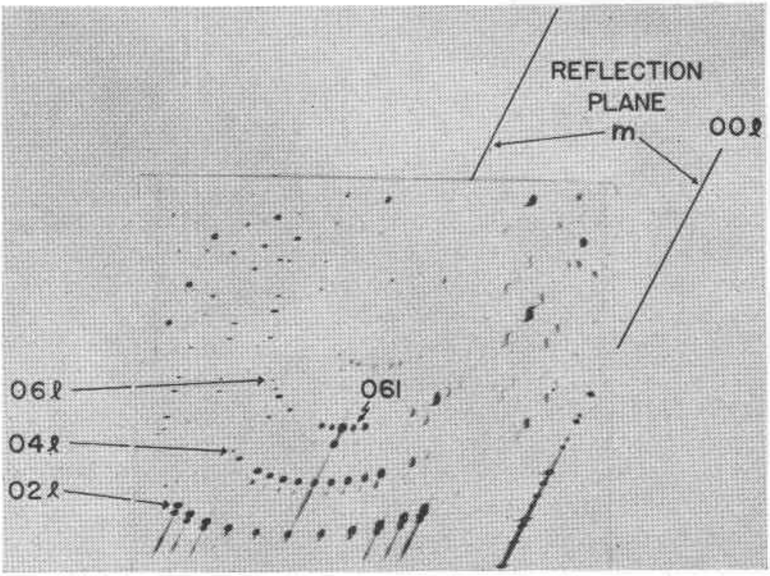

FIG. 1. 0-level a-axis Weissenberg photograph of normal muscovite.

LITHIAN MUSCOVITE

In the course of x-ray studies of so-called lepidolites a new variation of the muscovite polymorph has been discovered. A 0-level a-axis Weissenberg photograph of this form is shown in Fig. 2. It is apparently confined to "lepidolites" with a low Li_2O content and to muscovites with a relatively high Li_2O content. For this variation, the term lithian muscovite is proposed, to distinguish it from normal muscovite. Lithian muscovite is not to be confused with lithium muscovite, the hypothetical end-member used by Stevens (1938) and Berggren (1941). This variety is not common, having been found in only 10 micas of the approximately 500 studied. The following characteristics of lithian muscovite illustrate its close structural similarity to normal muscovite. Both have:

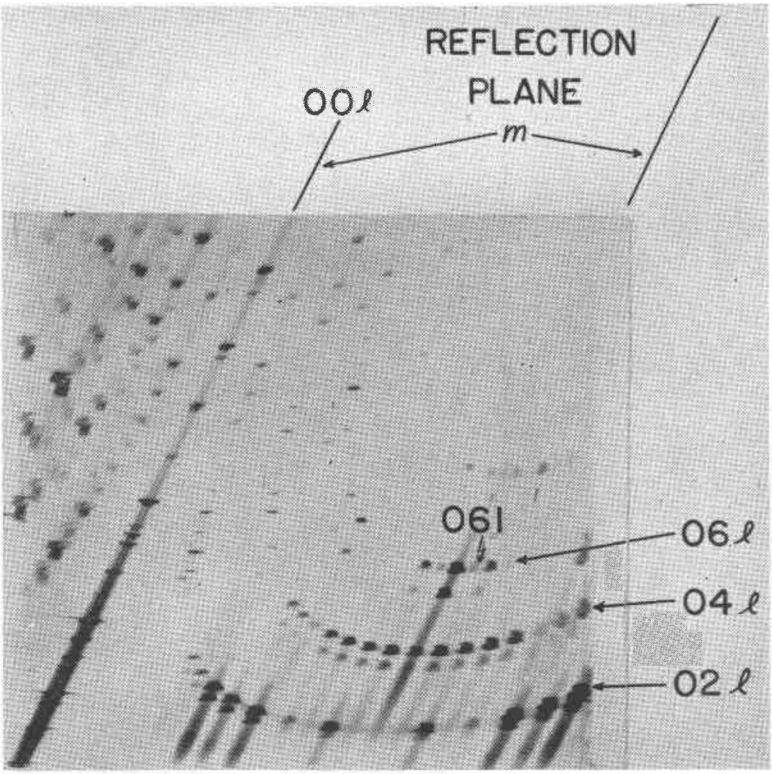

FIG. 2. 0-level a-axis Weissenberg photograph of lithian muscovite.

1. Space group $C2/c$.
2. Cell dimensions (approximate; measured on Weissenberg photographs) $a_0=5.2$ Å; $b_0=9.0$ Å; $c_0=20.0$ Å; $\beta=95°30'$.

3. (06 *l*) reflections with *l* odd present.
4. Optic plane perpendicular to (010).

The following points are different:

1. Indices are in the normal lepidolite range
   $\alpha=1.532$; $\beta=1.552$; $\gamma=1.556$.
2. Several differences occur in intensity of reflections.

The more important intensity differences for (0*kl*) reflections in normal muscovite and lithian muscovite are given in Table 1. The observed intensities of normal muscovite are those of Hendricks and Jefferson (1939). The lithian muscovite structure has been found in pegmatitic micas from: Tördal, Norway; Newry, South Portland and Topsham, Maine; Eight Mile Park, Colorado; Gunnison County, Colorado; Pala, California; Usakos, South West Africa; Kimito, Finland; and Londonderry, Western Australia.

TABLE 1. APPROXIMATE OBSERVED INTENSITIES OF SOME (0*kl*) REFLECTIONS OF NORMAL MUSCOVITE AND LITHIAN MUSCOVITE

Plane	Normal Muscovite	Lithian Muscovite
020	w	a
022	mw	vw
026	a	vw
045	a	vw
061	w	vw
065	vw	vvw
066	w	vvw
067	vw	vvw
069	w	vvw

The data in Table 1 demonstrate that on the basis of the presence of (06*l*) reflections with *l* odd, lithian muscovite must be considered as having crystallized with the muscovite type structure, but with lesser distortion, than in normal muscovite, for most of the (06*l*) reflections with *l* odd recorded are extremely weak. This indicates that lithian muscovite approaches more closely the octophyllite micas in structure and composition for, as Hendricks and Jefferson (1939, page 738) note: these reflections:

> ". . . are absent for the two layer biotite-like micas and none is observed for any of the micas that give (*h*0*l*) intensities of the single layer structure (except muscovite)."

A "lepidolite" No. 1 analyzed by Stevens (1938), which contains only 2.70% Li_2O, has crystallized with the 2-layer muscovite structure, according to *x*-ray analysis by Hendricks and Jefferson (1939). Inasmuch as it contains such a relatively high Li_2O content, Winchell (1942, p. 116) referred to this specimen as a lithium-bearing muscovite. It seemed possible, therefore, that the lithian muscovite structure shown in Fig. 2 was identical with the "lepidolite" analyzed by Stevens (1938) and that Hendricks and Jefferson (1939) neglected to report the intensity differences. Weissenberg photographs of a sample of Stevens No. 1, confirm that this "lepidolite" has the normal muscovite structure as stated by Hendricks and Jefferson (1939). Therefore, Fig. 2, of lithian-muscovite illustrates a new variation of muscovite which must contain at least more than 2.70% Li_2O.

Rowledge (1945) has briefly described a dozen micas and made partial analyses, including Li_2O. *X*-ray studies of portions of these analyzed muscovites and "lepidolites" show that all have crystallized as the normal 2-layer muscovite polymorph. Five of the "lepidolites" have Li_2O contents between 2.17% and 2.60%. Similarly three partially analyzed "lepidolites" from Western Australia described by Murray and Chapman (1931) were *x*-rayed and found to have crystallized with the normal muscovite structure. These micas contain 3.18%, 3.24% and 3.32% Li_2O. This information, coupled with the results obtained from Stevens No. 1 (2.70% Li_2O), and the micas described by Rowledge (1945), substantiates the conclusion that much more lithium may enter the muscovite structure without sensible distortion than has generally been realized.

In a specimen of mica from Newry, Maine, labeled "lepidolite," the lithian muscovite structure grades into that of normal muscovite. The dozen Weissenberg photographs of flakes from this specimen show the critical reflections to vary in intensity between those of normal and those of lithian muscovite. Spectrographic analysis of this material indicated 4.1% Li_2O. A spectrographic analysis of a lithian muscovite from Tördal, Norway shows 4.3% Li_2O. (These quantitative spectrographic analyses are of a preliminary nature. Future wet chemical analyses may indicate they are in need of revision.) Therefore lithian muscovite probably must contain at least 3.3% Li_2O and can contain possibly as much as 4.3% Li_2O.

In attempting to explain a form of muscovite with such a high lithium content, one must inquire if (1) it is possible for such a large amount of lithium to replace aluminum isomorphously in the muscovite structure, and if (2) the increased number of lithium atoms are sufficient to cause a reflection, such as (020), which is present in the muscovite structure, to disappear in the lithian muscovite structure. Since the atomic radii of these elements are similar, Al = 1.43Å, Li = 1.51Å, the substitution is possible and the valence difference may be countered by other substitutions, such as Fe^{+3} for Fe^{+2}. However, it is not necessary to have isomorphous replacement of aluminum by lithium. Since the muscovite (heptaphyllite) structure has only $\frac{2}{3}$ of its octahedral positions filled, with increasing Li content, the vacant positions gradually can be occupied by additional lithium atoms. This implies the existence of a structural series between heptaphyllite muscovite with a distorted structure, and octophyllite lepidolite with an undistorted structure. The presence of extremely weak (06*l*) reflections with *l* odd in lithian muscovite supports this concept. If these reflections are entirely absent, the mica has the undistorted 2-layer octophyllite type structure.

It is concluded, therefore, that Li ions, in terms of as much as 3.3% Li_2O, can enter the muscovite structure without causing any determinable structural variation. More lithium, probably occupying normally vacant octahedral positions, shifts the structure toward that of the octophyllite micas. In lithian muscovite these changes are already probably of sufficient magnitude to cause a reflection such as (020) to disappear.

Polymorphic Variation in Lepidolite

Most of the specimens analyzed by Stevens (1938), which were structurally investigated by Hendricks and Jefferson (1939, p. 761), were restudied. Table 2 illustrates the high degree of correlation between Hendricks and Jefferson (1939) and the writer's data. The two sets of results are in general agreement. However, some very significant variations may be observed. In Stevens (1938) No. 3 two very small pieces of lepidolite were found from which Weissenberg patterns could be obtained. Both of these gave identical 6-layer monoclinic diffraction patterns. Inasmuch as an optical orientation method is mainly used, the mass of the sample could not be studied by the Weissenberg method because most of the crystals had wavy extinction and gave distorted interference figures. For Stevens No. 6, Hendricks and Jefferson (1939) report the 6-layer monoclinic structure. Two Weissenberg photographs of this sample indicated respectively the presence both of the 6-layer lepidolite polymorph and the 2-layer lithian muscovite type. Therefore, the chemical analysis presented by Stevens (1938) is a composite analysis of the two forms.

A systematic study was made of one book of lepidolite for the purpose of determining the extent of polymorphic variation within a single crystal. Specimen No. 514 from the Opportunity pegmatite, in Gunnison County, Colorado was chosen. The specimen is approximately one and one-half inches across and three-quarters inch thick. It has a uniform typical pink color and is embedded in a mass of cleavelandite with grains of microlite. The sheets are a composite of many crystals. Between many of these crystal units are fine grained aggregates of lepidolite with irregular form and anomalous extinction. The optic planes of the various crystal units in a single sheet are always at 30°, or some multiple thereof, to each other. This appears to be the case regardless of the shape of the crystals in question, and whether or not they are separated by the fine grained aggregate. 2V on all sections of the sheet, regardless of polymorph, remains almost invariant. In all, 27 Weissenberg photographs of small crystals from two sheets, one near the top and the other in the center of the book were made. Of the 14 photographs taken from the sheet near the top, 12 show the 6-layer monoclinic structure and two the 1-layer monoclinic structure. However, on seven of the photographs of the 6-layer monoclinic form, the pattern of lithian muscovite was superimposed, in some cases only faintly present. Of the 13 photographs obtained from crystals in the central sheet, 12 had the 6-layer monoclinic structure and one had the 1-layer monoclinic pattern; only one lithian muscovite pattern was recorded and this again was superimposed faintly on a 6-layer monoclinic pattern. This shows the wide polymorphic variation possible in lepidolite both within sheets and also parallel with the *c*-axis direction. Yet one polymorph, in this case the 6-layer type, is dominant throughout the book. An inspection of Table 2 indicates that except for

Table 2. Structure of Micas Analyzed by Stevens (1938)

Stevens (1938) No.	Li_2O	Hendricks and Jefferson (1939)	Levinson
No. 1	2.70	Muscovite	Normal muscovite
No. 2	3.51	Too fine grained for study	Too fine grained
No. 3	3.70		6-layer monoclinic
No. 4	3.81		Too fine grained
No. 5	3.96		Too fine grained
No. 6	5.04	6-layer monoclinic	6-layer monoclinic and lithian muscovite
No. 7	5.05	6-layer monoclinic	not available
No. 8	5.11	single layer	single layer·
No. 9	5.33	single layer·	single layer
No. 10	5.39	single layer	not available
No. 11	5.51	none available	not available
No. 12	5.64	6-layer monoclinic	not available
No. 13	5.78	single layer·	single layer·
No. 14	5.89	3-layer hexagonal	3-layer hexagonal
No. 15	6.18	single layer·	single layer
No. 16	6.84	single layer	single layer
No. 17	7.26	single layer	single layer

numbers 12 and 14, those lepidolites with more than 5.1% Li_2O have crystallized with the one layer structure. It is necessary therefore to explain the apparently erratic results obtained in the case for No. 12, which has the 6-layer structure and No. 14 with the 3-layer hexagonal structure.

In the case of No. 12, unfortunately no material was available for restudy. It seems possible however, that further investigation could reveal 1-layer forms present in addition to the 6-layer type found by Hendricks. A re-determination of the Li_2O content also would be desirable.

UNIAXIAL LEPIDOLITE

Uniaxial lepidolite (No. 14) which has crystallized with the 3-layer hexagonal structure has a composition identical with that of lepidolites that have crystallized as the 1-layer polymorph. Such uniaxial lepidolites or nearly uniaxial lepidolites are very rare. A review of the literature revealed that Baumhauer (1903, 1912) first described such material from Mursinsk. He proposed the term microlepidolite for lepidolites of this character in contrast to macrolepidolites with large 2V. No chemical analyses were reported. Uniaxial lepidolite (5.83–6.15% Li_2O) from Western Australia has been described by Winchell (1925), Simpson (1927), Murray and Chapman (1931), Stevens (1938) and Hendricks and Jefferson (1939). This uniaxial lepidolite is associated with a normal biaxial mica of almost identical composition. Winchell (1925, p. 424), in agreement with Simpson (1927), suggests that the uniaxial character "may be due to fine twinning on (001)." Hendricks and Jefferson (1939) reported the structure of uniaxial material from Londonderry as the 3-layer hexagonal type, whereas the biaxial material was the 1-layer polymorph.

Jakob (1927) analyzed uniaxial lepidolite (4.93% Li_2O) from Usakos, and Pehrman (1945) analyzed and described similar material from Kimito (4.99% Li_2O). Specimens of both of these analyzed micas were found to have small areas of a biaxial phase. *X*-ray studies show that the uniaxial portions have crystallized as the 3-layer hexagonal polymorph but that the biaxial parts have the lithian muscovite structure. Since it has been demonstrated that the lithian muscovite structure is confined to a lower Li_2O range, the Li_2O contents of the Usakos and Kimito micas represent composite results which are lower than those obtainable from strictly uniaxial specimens.

Lundblad (1942, p. 58) notes that an analyzed specimen of lepidolite from Varuträsk has both uniaxial and biaxial portions. The analysis of this material seems unreliable; 65.62% SiO_2 in a lepidolite is too high. Miss Berggren (the analyst), according to Lundblad (1942), seems to suspect quartz contamination. Under these conditions it seems best to eliminate this mica from consideration.

Recently Macgregor (1945) described a lepidolite from Southern Rhodesia with a small 2V. A specimen similar to the described material surrounds a core of muscovite and in turn is enclosed by lepidolite with a large 2V. The muscovite structure is normal, and the lepidolite with the large 2V has crystallized as the 1-layer polymorph, whereas the uniaxial portion has crystallized as the 3-layer hexagonal form. The association of the 1-layer and 3-layer hexagonal polymorphs is identical with that of the Western Australia material. Chemical analyses of these lepidolites are not available.

If one takes into account the presence of lithian muscovite in the Usakos and Kimito lepidolites, it seems reasonable to infer that the Li_2O content of lepidolite of entirely uniaxial character may approach or even exceed the lowest Li_2O content of any of the samples with the 1-layer structure.

Structurally the 3-layer hexagonal polymorph can be geometrically interpreted as three 1-layer forms "twinned" on (001) with succeeding layers rotated 120° in the same direction (either clockwise or counterclockwise). Hendricks and Jefferson (1939, p. 746) with relation to the 3-layer hexagonal structure state that, "a particular layer is repeated about a three-fold screw axis normal to the cleavage." The powder patterns (Fig. 3) of the 3-layer hexagonal polymorph and the 1-layer monoclinic polymorph are almost identical. The cause of this rotation remains to be explained.

Reusch (1869) first obtained an almost perfectly uniaxial figure in muscovite by stacking rotated sheets in this fashion. Although his sheets were of considerable thickness, the over-all optical effect appears to be the same.

Ford (1932) explains the uniaxial effect on the basis of rotation of the plane of polarization. He notes (p. 328):

> "A particularly interesting case, . . . is that of the special properties of superposed cleavage-sections of mica. If three or more of these, say of rectangular form, be superposed and so placed that the lines of the axial planes make equal angles of 60° (45°, etc.) with each other the effect is that polarized light which passed through the center suffers circular polarization, with a rotation to the right or left according to the way in which the sections are built up. The interference-figure resembles that of a section of quartz cut normal to the axis. . . . Further, it is easy from this to understand how it is possible to have in sections of certain crystals (e.g., of clinochlore) portions which are biaxial and others that are uniaxial, the latter being due to an intimate twinning after this method of biaxial portions."

POWDER *X*-RAY STUDIES

Hendricks and Jefferson (1939) note that Stevens (1938) numbers 2, 3, 4 and 5, which appear to be in the interval between the 2-layer muscovite type and the 6-layer lepidolite type, are too fine-grained for study and speculate (p. 763):

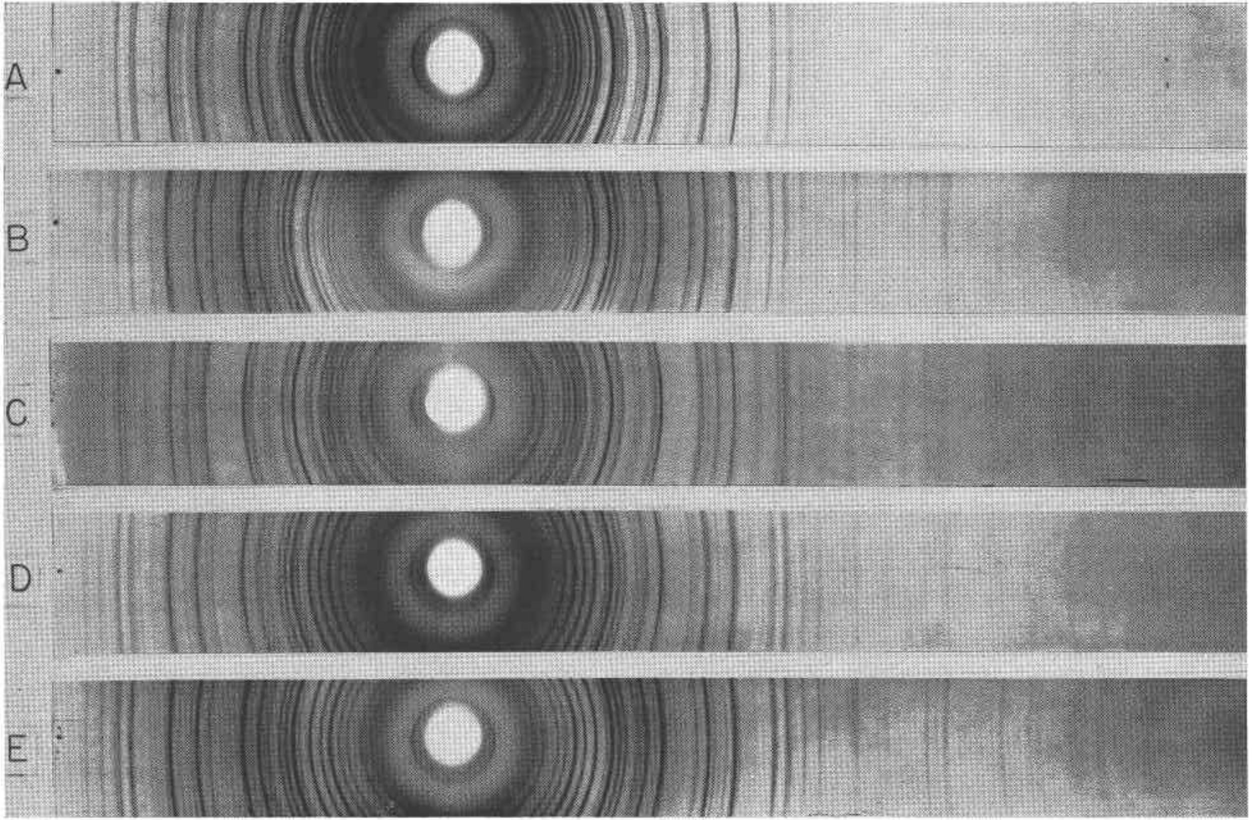

Fig. 3. *X*-ray powder photographs of polymorphs in muscovite-lepidolite series. *A*, normal muscovite, Varuträsk, Sweden; *B*, lithian muscovite, South Portland, Maine; *C*, 6-layer lepidolite, Gunnison County, Colorado; *D*, 1-layer lepidolite, Skuleboda, Sweden; *E*, 3-layer hexagonal lepidolite, Londonderry, Western Australia.

> "It is tantalizing to think that samples 2 to 5 owe their poor crystal development to their close approach to the limit of the lepidolite solid solution in muscovite."

It seems very likely that these fine-grained micas owe their macrostructural defects to small-scale variations in their crystal structure which itself probably is assignable to their chemically transitional position in the muscovite-lepidolite series. In order to test this possibility, powder *x*-ray studies on these micas were attempted.

It was first necessary to obtain powder *x*-ray data for each of the known polymorphs. The only powder data on muscovites or lepidolites found in the literature were for normal muscovite (Nagelschmidt, 1937) and for the 3-layer muscovite polymorph (Axelrod and Grimaldi, 1949). Grim and Bradley (1951) list partial data for normal muscovite and a 1-layer lepidolite and give photographs of the two patterns. A complete set of powder pictures was compiled by *x*-raying powdered single crystals whose structure had first been determined by the Weissenberg method. This set of standard photos, which includes the polymorphs of muscovite and lepidolite under study, is shown in Fig. 3; the *d*-spacings of forms not previously published are given in Table 3.

The structures of the fine-grained lepidolites analyzed by Stevens (1938) have been determined:

Stevens No.	Per Cent Li_2O	Structure by Hendricks and Jefferson	Levinson	
			Weissenberg	Powder
2	3.51	too fine-grained	too fine-grained	6-layer lepidolite+ 2-layer muscovite
3	3.70		6-layer	6-layer lepidolite+ 2-layer muscovite
4	3.81		too fine-grained	6-layer lepidolite+ 2-layer muscovite
5	3.96		too fine-grained	6-layer lepidolite+ (2-layer muscovite?)

That these fine-grained lepidolites are combinations of forms and not single structures confirms the idea that the poor crystal development is related to their composition. Micas with these combined structures may be termed transitional. It is noteworthy that crystals of Stevens No. 3 large enough for Weissenberg photographs are 6-layer forms, but the poorly developed crystals from the same specimen have a combination 6-layer lepidolite and 2-layer muscovite structure. It is likely that the

TABLE 3. SPACINGS OF POLYMORPHIC FORMS
Cu $K\alpha_1$, $\lambda = 1.53736$

2-layer lithian muscovite (monoclinic) South Portland, Maine		1-layer lepidolite (monoclinic) Topsham, Maine		3-layer lepidolite (hexagonal) Londonderry, Western Australia		6-layer lepidolite (monoclinic) Gunnison County, Colorado	
I	*d*-spacing	*I*	*d*-spacing	*I*	*d*-spacing	*I*	*d*-spacing
m	9.95	s	9.91	m	9.91	ms	9.89
m	5.01	s	4.98	m	4.97	m	4.99
m	4.50	w	4.94	m	4.66	m	4.49
vvw	3.95	vw	4.34	m	3.84	w	3.84
vvw	3.87	vw	4.12	m	3.58	m	3.61
w	3.71	vw	3.86	vs·	3.31	m	3.47
w	3.46	s	3.61	s	3.09	m	3.31
ms	3.20	vs·	3.33	s	2.86	m	3.19
m	3.21	s	3.07	w	2.651	m	3.07
m	2.98	s	2.87	s	2.571	m	2.88
s	2.84	m	2.675	mw	2.455	m	2.775
w	2.755	s	2.573	mw	2.375	vs·	2.572
ms	2.571	m	2.468	vw	2.243	m	2.416
m	2.474	m	2.387	vw	2.186	vvw	2.248
m	2.387	vw	2.253	mw	2.126	vvw	2.190
vw	2.247	m	2.132	vw	2.052	vw	2.039
vvw	2.196	ms	1.988	m	1.984	s	1.985
mw	2.132	vvw	1.956	vvw	1.955	vvw	1.684
m	2.081	w	1.748	w	1.718	vvw	1.633
vw	1.951	vw	1.715	mw	1.643	vvw	1.572
w	1.742	m	1.646	w	1.611	m	1.506
vw	1.720	vvw	1.581	w	1.576	vvw	1.393
m	1.644	vvw	1.544	w	1.547	vvw	1.355
vvw	1.596	m	1.511	mw	1.511	vvw	1.319
vvw	1.557	vw	1.493	vvw	1.481	w	1.300
vvw	1.510	vvw	1.420	vvw	1.457	vvw	1.239
w	1.500	vvw	1.375	vvw	1.435		
w	1.487	vvw	1.352	vvw	1.411		
vvw	1.453	vw	1.337	w	1.341		
vw	1.427	vw	1.299	vw	1.295		
w	1.340	vvw	1.242	vvw	1.285		
w	1.296	vvw	1.199				
vvw	1.269	vvw	1.136				
vw	1.243						
vvw	1.220						
vvw	1.199						

well developed crystals have a higher lithium content than the intergrown portions. In all cases the 6-layer lepidolite polymorph predominates over the muscovite type in the transitional types.

Owing to the close similarity between the powder patterns of normal muscovite and lithian muscovite the exact type of muscovite in the combined powder patterns of the transitional micas is difficult to determine with certainty. However, careful study of the intensities indicates the probable presence of the lithian muscovite type.

CORRELATION

On the basis of the structural data obtained it seemed advantageous to plot Li_2O content against the polymorphic types of the muscovite-lepidolite series: normal muscovite, lithian muscovite, transitional micas, 6-layer lepidolite, 1-layer lepidolite and 3-layer hexagonal lepidolite. The result of plotting the Li_2O contents of 35 analyzed micas against their structures is presented in Fig. 4. Many more micas with less than 2% Li_2O were studied, but there is no need to record these, as they all have the normal muscovite structure.

The graph shows that micas with less than ca. 3.3% Li_2O have the normal muscovite structure; those with 3.4%–4.0% L_2iO have transitional structures; those with 4.0%–5.1% generally have crystallized with the 6-layer lepidolite structure; whereas those biaxial micas with more than 5.1% Li_2O generally have the 1-layer lepidolite structure. The uniaxial lepidolites as discussed previously have relatively large Li_2O contents, probably in the same range with those of the 1-layer form, and this structure may be explained by "twinning." Too few data are now available to place the lithian muscovite type of structure accurately in the series. However, the available evidence shows that it belongs somewhere between normal muscovite and 6-layer lepidolite. The above conclusions are graphically represented in Fig. 5.

Analyzed micas described by Berggren (1940, 1941) and Lundblad (1942) do not fit well into the conclusions represented by Fig. 5. The results obtained from *x*-ray studies of these micas are presented in Table 4. Inconsistencies appear with regard to Berggren's (1940) micas in *A*, *B*, *C*, and Lundblad's (1942) No. 10 and No. 13. Micas *C* and No. 13, which are reported to have 3.9 and 5.7% Li_2O, respectively, have crystallized with the normal muscovite structure. As much as 5.7% Li_2O, or even 3.9% Li_2O, in normal muscovite appears improbable on the basis of present knowledge. It is possible that the lithium content of the mica samples varies so much that the portions supplied represent extreme structural variations of the analyzed materials. Several of the Swedish micas are optically non-homogeneous, which may indicate the presence of several crystal phases and one may even be contaminated with quartz (Lundblad, 1942). Under such circumstances an attempt to correlate polymorphism with chemistry would be fruitless. Baumhauer (1912) described several lepidolites with a wide range of 2E and several distinctive types of etch figure in single specimens. Both of these features probably indicate structural variations. Except for the Swedish micas the correlation is good; indeed better than might be expected, considering the presence of structural variation in single books and the probable wide range in quality of the Li_2O determinations.

Legend for Figure 4

1. Wodgina, Western Australia; No. 36, Rowledge (1945).
2. Tabba Tabba, Western Australia; No. 52, Rowledge (1945).
3. Varuträsk, Sweden; Analysis D, Berggren (1940).
4. Wodgina, Western Australia; No. 6, Rowledge (1945).
5. Tabba Tabba, Western Australia; No. 6, Rowledge (1945).
6. Karibib, South Africa; 2.5% Li_2O, courtesy of Foote Mineral Company.
7. Wodgina, Western Australia; No. 2, Rowledge (1945).
8. Manitoba, Canada; No. 1 Stevens (1938).
9. Karibib, South Africa; 3.0% Li_2O, courtesy of Foote Mineral Company.
10. Wodgina, Western Australia; Murray and Chapman (1931).
11. Tabba, Western Australia; Murray and Chapman (1931).
12. Ravensthorpe, Western Australia; Murray and Chapman (1931).
13. Bear Claim, Manitoba; Spec. XXXIII, Ellsworth (1932).
14. Pala, Calif.; No. 2, Stevens (1938).
15. Pala, Calif.; No. 3, Stevens (1938).
16. San Diego Co., Calif.; No. 4, Stevens (1938).
17. Pala, Calif.; No. 5, Stevens (1938).
18. Ubini, Western Australia; Murray and Chapman (1931).
19. Poona, Western Australia; Murray and Chapman (1931).
20. Pala, Calif.; No. 6, Stevens (1938).
21. Ohio City, Colorado; No. 7, Stevens (1938).
22. Pala, Calif.; No. 12, Stevens (1938).
23. San Diego Co., Calif; No. 8 Stevens (1938).
24. Mesa Grande, Calif.; No. 9, Stevens (1938).
25. Antsongombato, Madagascar; Mauguin (1928*B*).
26. Wakefield, Canada; No. 10, Stevens (1938).
27. Mesa Grande, Calif.; No. 13, Stevens (1938).
28. Maharitra, Madagascar; Mauguin (1928*B*).
29. San Diego Co., Calif.; No. 15, Stevens (1938).
30. Antsongombato, Madagascar; No. 16, Stevens (1938).
31. Greenland; No. 17, Stevens (1938).
32. Usakos, Southwest Africa; Jakob (1927).
33. Kimito, Finland; Pehrman (1945).
34. Calgoorie, Western Australia; No. 14, Stevens (1938).
35. Londonderry, Western Australia; Murray and Chapman (1931).

The structures of all the above micas, with the exception of numbers 21, 22 and 26, have been determined by the author. In the case of the three exceptions the structures reported by Hendricks and Jefferson (1939) have been used.

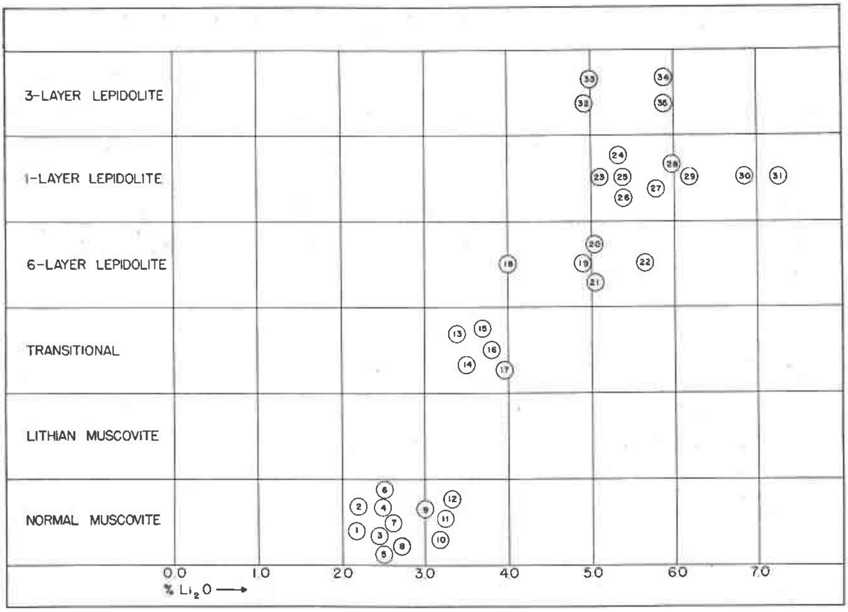

FIG. 4. Plot of the Li_2O contents of 35 micas in muscovite-lepidolite series against the various polymorphs.

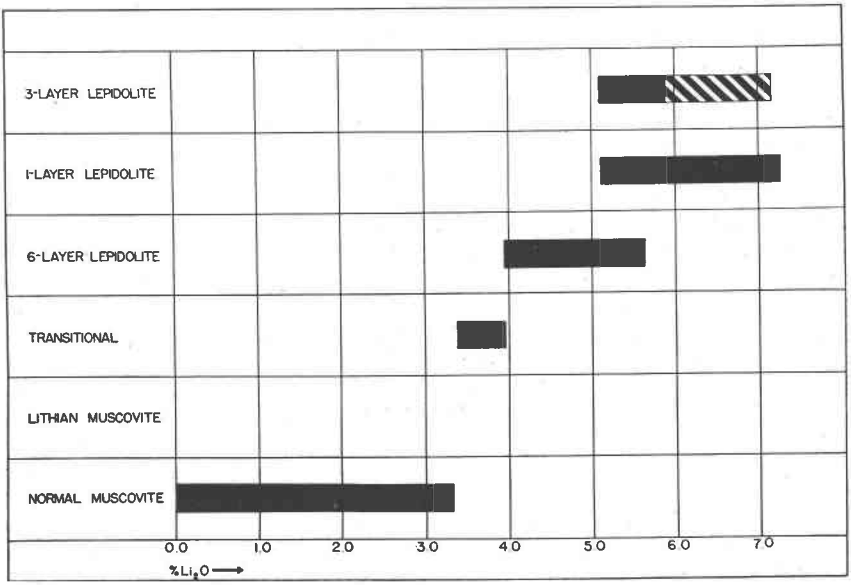

FIG. 5. Idealized representation of relationship between Li_2O content and polymorphism in the muscovite-lepidolite series.

TABLE 4. STRUCTURE OF MICAS DESCRIBED BY BERGGREN (1940, 1941) AND LUNDBLAD (1942)

Analysis	% Li_2O	Structure	Locality
A	5.95	6-layer lepidolite	Varutraäk
B	4.35	1-layer lepidolite	Varuträsk
C	3.9	normal muscovite	Varuträsk
D	2.45	normal muscovite	Varuträsk
E	1.80	normal muscovite	Varuträsk
G	0.73	normal muscovite	Varuträsk
H	0.69	normal muscovite	Varuträsk
I	0.22	normal muscovite	Varuträsk
J	0.76	normal muscovite	Varuträsk
K	1.10	normal muscovite	Varuträsk
L	1.1	normal muscovite	Varuträsk
No. 10	4.55	1-layer lepidolite	Varuträsk
No. 13	5.7	normal muscovite	Utö
No. 14	5.5	6-layer and 1-layer lepidolite	Rozena

OPTICAL DATA

Optical constants were measured on individual flakes whose structures first had been determined by the Weissenberg method. The results are presented in Table 5. The indices of refraction were measured on the Abbé refractometer or were obtained by the immersion method and are reproducible to ±0.001. The Mallard method was used in the determination of 2E and 2V was calculated. The only other determinations of the optical properties of lepidolite polymorphs have been by Hendricks and Jefferson (1939). Their results, though scanty, are in close agreement with those of this paper.

The lepidolite polymorphs are not distinguishable on the basis of optical properties alone; it follows therefore their general chemistry cannot be deduced from the optical constants. A trend is that the indices, and particularly 2V, of the 1-layer polymorph tend to be slightly higher than those of the 6-layer polymorph, but the overlap is broad. This can be accounted for by the optical inactivity of the Li-ion. Although variations in Li content influence the layer stacking pattern, changes in optical constants are more sensitive to variations in Fe^3, Ti, Fe^2 and Mn, both in muscovite and the lepidolites. The indices of lepidolite vary within books (note values obtained from spec. 514 and 452 in Table 5). However the indices of lithian muscovite are in the range of normal lepidolites whose values are considerably below those of normal muscovite.

TABLE 5. INDICES OF REFRACTION AND 2V OF LEPIDOLITES AND LITHIAN MUSCOVITE*

Specimen Number	Locality	α	β	γ	2V
	Six-layer lepidolite				
502	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.555	1.560	30°
514 (2)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.553	1.557	30°
514 (3)	Opportunity Pegmatite, Ohio City, Colorado	1.533	1.555	1.560	33°
514 (4)	Opportunity Pegmatite, Ohio City, Colorado	1.533	1.557	1.560	33°
514 (5)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.555	1.558	—
514 (6)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.553	1.560	33°
505 (*b*)	Brown Derby Pegmatite, Ohio City, Colorado	1.530	1.556	1.560	34°
967 (*a*)	Stewart Mine, Pala, California	1.530	1.552	1.559	33°
967 (*b*)	Stewart Mine, Pala, California	1.529	1.554	1.558	34°
970 (*a*)	Stewart Mine, Pala, California	1.534	1.555	1.559	33°
452 (*a*)	Varuträsk, Sweden	1.532	1.556	1.561	33°
452 (*b*)	Varuträsk, Sweden	1.535	1.556	1.561	35°
452 (*c*)	Varuträsk, Sweden	1.532	1.552	1.557	37°
	Three-layer hexagonal lepidolite				
539 (*f*)	Pope mining claim, Southern Rhodesia	—	1.568	1.568	0°
539 (*g*)	Pope mining claim, Southern Rhodesia	—	1.566	1.566	0°
	One-layer lepidolite				
514 (*k*)	Opportunity Pegmatite, Ohio City, Colorado	—	1.556	1.558	—
464 (*b*)	Newry, Maine	—	1.556	1.558	50°
471 (*b*)	South Portland, Maine	1.533	1.553	1.560	41°
476 (*b*)	Skuleboda, Sweden	—	1.562	1.564	—
476 (*c*)	Skuleboda, Sweden	—	1.564	1.566	37°
535 (*c*)	Dogon Daji, Nigeria	—	1.557	1.562	47°
539 (*a*)	Pope Mining Claim, Southern Rhodesia	—	1.562	1.565	38°
539 (*b*)	Pope Mining Claim, Southern Rhodesia	—	1.562	1.566	37°
	Two-layer lithian muscovite				
556 (*b*)	Tördal, Norway	—	1.553	1.556	34°
556 (*c*)	Tördal, Norway	1.532	1.554	1.558	34°
465 (*a*)	Newry, Maine	1.533	1.555	1.559	34°
465 (*f*)	Newry, Maine	1.534	1.555	1.561	37°
471 (*a*)	South Portland, Maine	1.529	1.551	1.556	32°
679 (*a*)	Usakos, S. W. Africa	1.534	1.559	1.566	—

* Measurements by Mr. Charles H. Hewitt.

Specimens numbered such as 415 (2) through 514 (6), or 452 (*a*) through 452 (*c*) represent measurements obtained from different flakes in the same book.

SUMMARY

The discovery of the lithian muscovite type of structure, the interpretation of the structural origin of the 3-layer hexagonal lepidolite polymorph and good correlation between Li_2O content and the various polymorphs shows that chemical and structural transitions occur between Li-free muscovites and Li-rich lepidolites. It seems probable, however, that the relationship between a single ion variation and polymorphism (Fig. 5) in a structure as complex as the micas is somewhat oversimplified. Overlapping undoubtedly takes place and it may be possible to get even more accurate results by calculating the number of atoms with octahedral coordination against the various polymorphs; at present too few complete analyses and specimens are available to attempt this approach.

The study shows also that structural variations in single lepidolite crystals are common and can be coupled with compositional variations. Optical data are insufficient to distinguish the various polymorphs. The correlation may prove valuable in estimating the approximate Li_2O content of the mica without a chemical analysis.

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