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QUARTERLY PROGRESS REPORT NO. 2

NATURAL MICA STUDIES

By

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Project M978

SIGNAL CORPS, DEPARTMENT OF THE ARMY
CONTRACT DA-36-039 sc-15357, SC PROJECT NO. 152B-0, DA PROJECT NO. 3-99-15-022

March, 1952

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INTRODUCTION

Research on the natural micas has been expanded along the lines outlined in Quarterly Report No. 1. Many additional specimens have been studied by means of the Weissenberg and powder methods. Complete powder data are now available for all the various polymorphous types in the muscovite-lepidolite series. Their use in connection with studies of fine-grained micas and also of micas that appear to fall in the transition areas of the various polymorphs is discussed in detail in this report. Additional Weissenberg studies indicate that the "lithium muscovite" variety of the 2-layer muscovite polymorph reported in the first quarterly report has not been described previously. Furthermore investigations on lepidolites with a small 2V have disclosed a new polymorph.

High-temperature work has recently been started with the purpose of investigating the effect of heat on crystallization. Apparatus capable of maintaining a constant temperature (within 2°-3°) up to about 1200°C, is being used. This work is as yet in the preliminary stages.

The entire mica collection has been renumbered to simplify cataloguing and usage. Henceforth the new numbers will be used. Where it is necessary to refer to micas mentioned in Quarterly Report No. 1 the old numbers will also be indicated. Selected specimens continue to be prepared for chemical analysis.

WEISSENBERG X-RAY STUDIES

In Quarterly Report No. 1 (page 9), reference was made to Stevens' analyzed lepidolite No. 1, which was assumed to have the "lithium muscovite" structure on the basis of a high (2.70%) Li_2O content. (The structure is illustrated on page 20 of that report.) Dr. S. B. Hendricks, who studied this material by means of x-rays (1939), has graciously supplied some of the original analyzed sample for restudy at this laboratory. Further x-ray work on the material has shown that it crystallized as the normal muscovite polymorph. Thus it was incorrect to assume that Hendricks and Jefferson (1939) had found anything but the normal muscovite polymorph, especially since no reference was made to any irregularities in the Weissenberg photographs. Therefore the "lithium muscovite" reported in the first quarterly report is a hitherto unreported variation of the normal muscovite polymorph. "Lithium muscovite" is the variety of the muscovite structure referred to as occurring in Specimen No. 514 (lepidolite M29), which was studied in detail and discussed on pages 6 and 7 of the first report, where it is called "2-layer muscovite type". The following characteristics of "lithium muscovite" illustrate the close structural similarity between it and normal muscovite. Both have:

1. Space group C2/c
2. Cell dimension (approximately)

$$a = 5.2\ \text{Å}$$
$$b = 9.0\ \text{Å}$$
$$c = 20.0\ \text{Å}$$
$$\beta = 95°30'$$

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

They differ in the following points:

1. Indices for "lithium muscovite" are in the normal lepidolite range:

$$\alpha = 1.532$$
$$\beta = 1.552$$
$$\gamma = 1.556$$

2. Several differences occur in intensities of reflections. The more important intensity differences for $(ok\ell)$ reflections in normal muscovite and "lithium muscovite" are given below. The observed intensities of normal

muscovite are those of Hendricks and Jefferson (1939), whereas the calculated intensities for muscovite are those of Jackson and West (1930).

Plane	Normal Muscovite Observed	Normal Muscovite Calculated	"Lithium Muscovite"
020	w	4	a
022	mw	8	vw
026	a	7	vw
045	a	4	vw
061	w	0	vw
065	vw	0	vvw
066	w	4	vvw
067	vw	0	vvw
069	w		vvw

This table demonstrates that on the basis of the presence of 06 ℓ reflections with ℓ odd, "lithium muscovite" must be considered as having crystallized in a muscovite type of structure. The distortion, however, is probably not as great as in normal muscovite, for most of the 06ℓ with ℓ odd reflections recorded are extremely weak. This would seem to indicate that "lithium muscovite" is close to the octophyllite micas in structure and composition, for as Hendricks and Jefferson note (1939, page 738) these reflections "are absent for the two-layer biotite-like micas and none is observed for any of the micas that give (hoℓ) intensities of the single-layer structure (except muscovite)".

This variety is apparently found in lepidolites with a low Li_2O content or muscovite with a high Li_2O content. On the basis of this, the varietal term "lithium muscovite" is tentatively proposed. Spectrographic analyses are now being conducted on two micas which have crystallized with this structure. Because Stevens' No. 1, with 2.70% Li_2O, has crystallized in the normal muscovite polymorph, it is assumed that "lithium muscovite" will have a still higher Li_2O content. It is hoped that the extent to which lithium may enter the muscovite structure will be determined through spectrographic analysis.

In this connection, data obtained from x-ray studies of analyzed micas received from Dr. H. P. Rowledge, Government Mineralogist of Western Australia, and Dr. Frans Wickman, Director, Royal Mineralogical Museum of Stockholm, Sweden, are pertinent. Rowledge (1945) has described briefly a dozen micas with their Li_2O contents. In about half the cases he also gave the percentages of Na_2O and K_2O, along with optical data. X-ray studies of portions of these analyzed muscovites and "lepidolites" show that these micas have all crystallized as the normal 2-layer muscovite polymorph. Five of the "lepidolites" have lithium contents that range from 2.17% to 2.60% Li_2O. This information, coupled with the results obtained from Stevens No. 1 (2.70% Li_2O), substantiates the suggestion that much more lithium may enter the muscovite structure than has been generally realized.

Dr. Frans Wickman has supplied lithium micas, mainly from Varuträsk, analyzed by Berggren (1940, 1941). The optical properties of these micas have been described by Lundblad (1942). The following table presents the results obtained from x-ray studies of these micas.

Project No. M978	Swedish Analysis	% Li_2O	Structure	Locality
519	A	5.95	6-layer lepidolite	Varuträsk
520	B	4.35	1-layer lepidolite	Varuträsk
521	C	3.9	normal muscovite	Varuträsk
522	D	2.45	normal muscovite	Varuträsk
523	E	1.80	normal muscovite	Varuträsk
524	G	0.73	normal muscovite	Varuträsk
525	H	0.69	normal muscovite	Varuträsk
526	I	0.22	normal muscovite	Varuträsk
528	J	0.76	normal muscovite	Varuträsk
529	K	1.10	normal muscovite	Varuträsk
530	L	1.1	normal muscovite	Varuträsk
531	No. 10	4.55	1-layer lepidolite	Varuträsk
532	No. 13	5.7	normal muscovite	Utö
533	No. 14	5.5	6+1-layer lepidolite	Rozena

From the standpoint of correlating polymorphs with Li_2O content, the results obtained from these Swedish micas are not in good agreement with the data obtained from investigations of the lepidolites analyzed by Stevens (see Quarterly Report No. 1, page 8). The inconsistencies are in the following:

No. 519
No. 520
No. 521
No. 531
No. 532

Two of the above immediately arouse suspicion. Nos. 521 and 532, which are reported to have 3.9% Li_2O and 5.7% Li_2O, respectively, have crystallized in the normal muscovite structure. As much as 5.7% Li_2O, or even 3.9% Li_2O, in the muscovite structure is to be questioned on the basis of present knowledge. It seems possible that the lithium contents of these samples vary so much that the portions supplied represent extreme variations of the analyzed materials. Thus, under such circumstances, any attempt to correlate polymorph with chemistry would be fruitless. At present the compositions of micas 521 and 532 are being redetermined, and further discussion of the Swedish micas must await the new results.

The 2V of lepidolites has been reported as varying from 0° to about 60°. The vast majority are in the range from 35° to 60°. Several occurrences of lepidolite with a very small 2V have been reported. Winchell (1925) describes a sensibly uniaxial lepidolite from Londonderry, Western Australia, and suggests (page 424) that the uniaxial character "may be due to fine twinning on(001)". Axelrod and Grimaldi (1949) report that the 3-layered muscovite polymorph has a variable 2V (3° to 15°) and state (page 559), "Variation in the observed optic axial angle is attributed to the coalescence of the optical effects of superposed twin elements."

Hendricks and Jefferson (1939) studied a uniaxial lepidolite from the Western Australia locality and report its structure as the 3-layer rhombohedral polymorph. It is intimately associated with a lepidolite with a large 2V which has crystallized in the 1-layer form. The two types occur in the same sheet and chemical analysis of both sections (Winchell 1925, page 424) are almost identical. Recently Macgregor (1945) has described a lepidolite from Southern Rhodesia with a small 2V. A specimen (No. 539) similar to the described material has been obtained from Dr. Macgregor. It surrounds a core of muscovite and in turn is enclosed by lepidolite with a large 2V. The muscovite structure is normal, and the mica with a large 2V has crystallized in the 1-layer polymorph, whereas the uniaxial portion has crystallized in the 3-layer rhombohedral form. The association of the 1-layer and 3-layer rhombohedral polymorphs is identical with that of the Western Australia material.

Specimen No. 679 from Usakos, South Africa, obtained from Harvard University (Harvard No. 13879), consists of uniaxial and biaxial segments. The mica has the appearance of a slightly brownish muscovite. Weissenberg photographs obtained from several crystals taken from the uniaxial section reveal the existence of another 3-layer rhombohedral mica. The biaxial portion, however, has crystallized with the "lithium muscovite" structure. Before assigning a name (muscovite or lepidolite) to the uniaxial section, it will be necessary to obtain a chemical analysis, or at least a determination of the lithium content.

A specimen of lepidolite from Skuleboda, Sweden, (No. 476) was also found to contain a uniaxial section. Again the biaxial part has crystallized in the 1-layer polymorph (space group Cm). X-ray studies of the uniaxial portion, however, reveal the discovery of a new 3-layer polymorph, which likewise has crystallized in the monoclinic space group Cm. O-level Weissenberg photographs have been taken about all a- and pseudo a- axes as well as b- and pseudo b- axes. A plane of symmetry in the position of b^* as well as c^* was recorded on only one of the photographs; therefore this pattern is considered as having been obtained from rotation about the true a- axis. Coupled with information from other Weissenberg photographs, this establishes the crystal system as monoclinic. The Weissenberg pattern obtained by rotation about the o-level a- axis is very similar to o-level a- axis photographs obtained from the 3-layer rhombohedral polymorph. The o-level b- axis (Fig. 1) and 1-level a- axis (Fig. 2) photographs show some differences. Differences along the 13ℓ reciprocal lattice line between the 3-layer rhombohedral and 3-layer monoclinic may be seen by comparing Fig. 2 with a first-level Weissenberg photograph illustrated by Axelrod and Grimaldi (1949, page 569). Indexing showed all reflections with h + k odd and (00ℓ) with $\ell \neq 3n$ were absent. The possible monoclinic space groups with the information thus far are Cm, C2, and C2/m. In comparing a- and b- axes photographs of the new form and the 1-layer monoclinic lepidolite (space group Cm), a very interesting relationship is observed. When the photographs are superimposed, every third reflection of the new form corresponds exactly with a reflection of the 1-layer polymorph. In between, two additional reflections will almost always be found on the photographs of the new polymorph. This relationship also applies to rotation about the pseudo a- and b- axes. Therefore it is logical to assume that the 3-layer monoclinic lepidolite contains the same symmetry elements as the 1-layer polymorph and has also crystallized in the monoclinic space group Cm. The unit cell dimensions are (approximately):

a = 5.2 A	c = 30.0 A
b = 9.0 A	β = 91° (±30')

Powder data for the 3-layer monoclinic lepidolite are given in Table 1 and the pattern is illustrated in Fig. 4.

POWDER X-RAY STUDIES

Before any attempt could be made to study polymorphism in the fine-grained micas, it was necessary to obtain powder x-ray data for each of the known polymorphs. The only complete powder data on micas found in the literature are 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 single-layer lepidolite and give photographs for the two forms. A set of powder pictures was compiled by photographing powdered crystals whose structure had been determined by the Weissenberg method. The set of standard photographs, which includes all known polymorphs of muscovite and lepidolite, is shown in Figs. 3 and 4; and the d-spacings of five forms are given in Table 1 in the appendix.

Powder x-ray photographs were taken of approximately 40 specimens, most of which are too fine-grained to study by the Weissenberg method. The photographs were then compared with the standard photographs to determine the type of structure. A complete list of the specimens and their structures may be found in Table 2 of the appendix. All but two of the micas are known forms. One of the exceptions is a barium muscovite (described by Bauer and Berman, 1933), the structure of which has not yet been determined; the other is the 3-layered monoclinic lepidolite described in this report.

The structure of the fine-grained lepidolites analyzed by Stevens in 1938 (page 8, First Quarterly Report) has been determined. The complete results are listed below.

Stevens No.	% Li_2O	Structure by Hendricks	Project M978 Weissenberg	Project M978 Powder
2	3.51	too fine-grained	too fine-grained	6-layer plus 2-layer
3	3.70		6 layer	6-layer plus 2-layer
4	3.81		too fine-grained	6-layer plus 2-layer
5	3.96		toc fine-grained	6-layer (plus 2-layer (?))

The fact that these fine-grained lepidolites are combination forms (Fig. 5) and not single structures confirms the idea that the poor crystal development is in some way related to the chemistry. It is interesting to note 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 and 2-layer "lithium muscovite" form. It is probable that the larger crystals have a higher lithium content than the intergrown portions. Specimens 514, 488, 489, 500, 456, and 533 are either 6-layer plus 2-layer "lithium muscovite" forms or 6-layer plus 1-layer forms with either the 6-layer or 1-layer form dominant. In all cases these specimens are fine-grained and single crystals show poor extinction. With the exception of such fine-grained muscovites as oncosine, the only uncombined pattern obtained was that of the 6-layer polymorph. The 1- or 2-layer "lithium muscovite" form was never found alone in the fine-grained micas studied. Closer correlation between chemistry and structure will be attempted as soon as the results from quantitative spectrographic analyses are available.

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APPENDIX

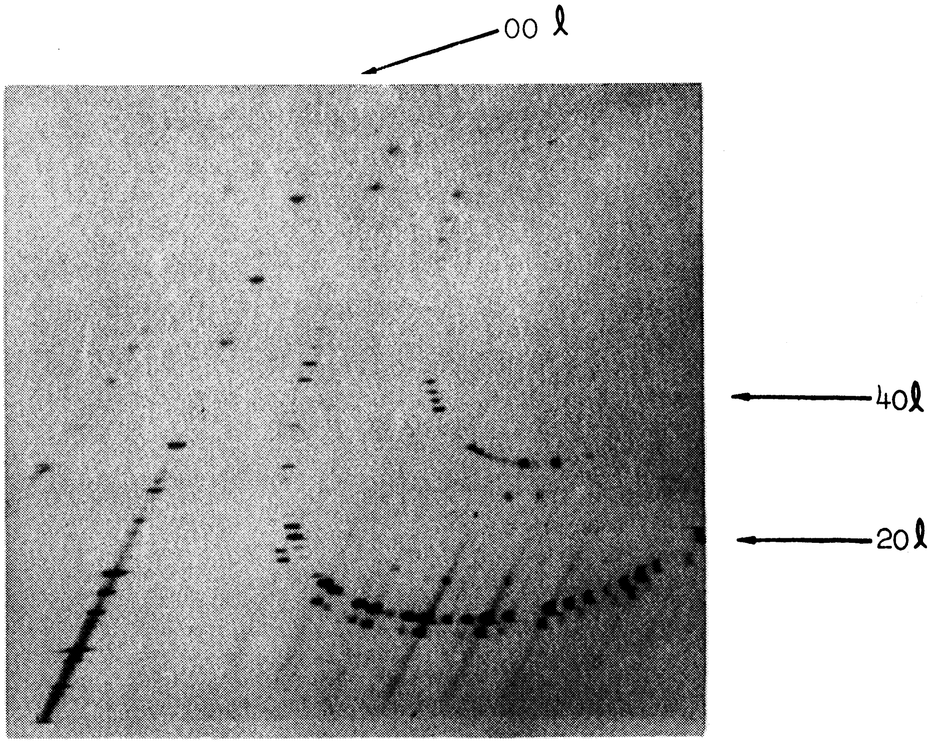

Fig. 1. 3-layer monoclinic lepidolite
0-level b- axis Specimen No. 476.

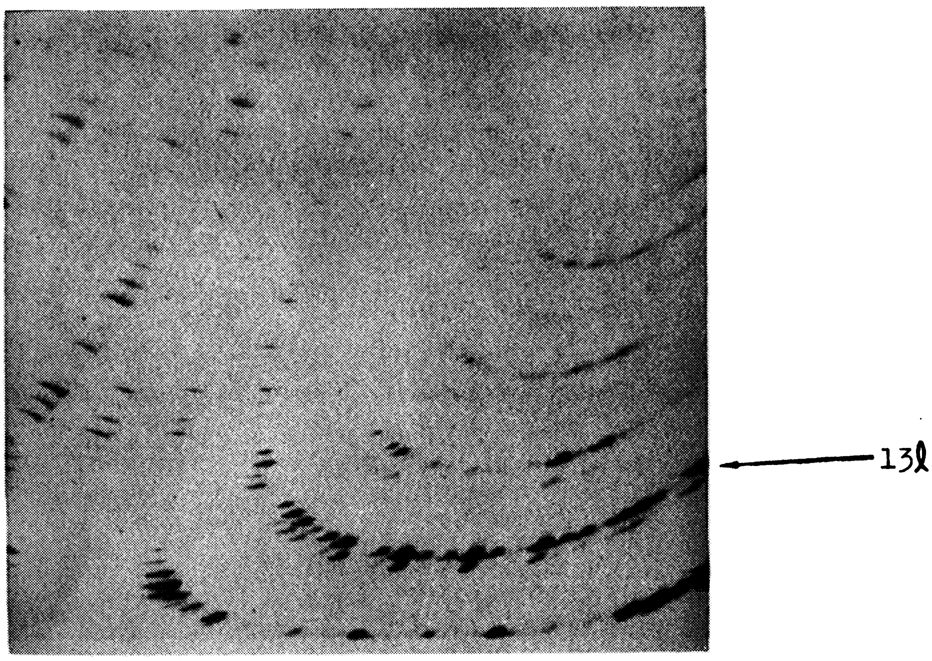

Fig. 2. 3-layer monoclinic lepidolite
1-level a- axis Specimen No. 476.

A.

B.

C.

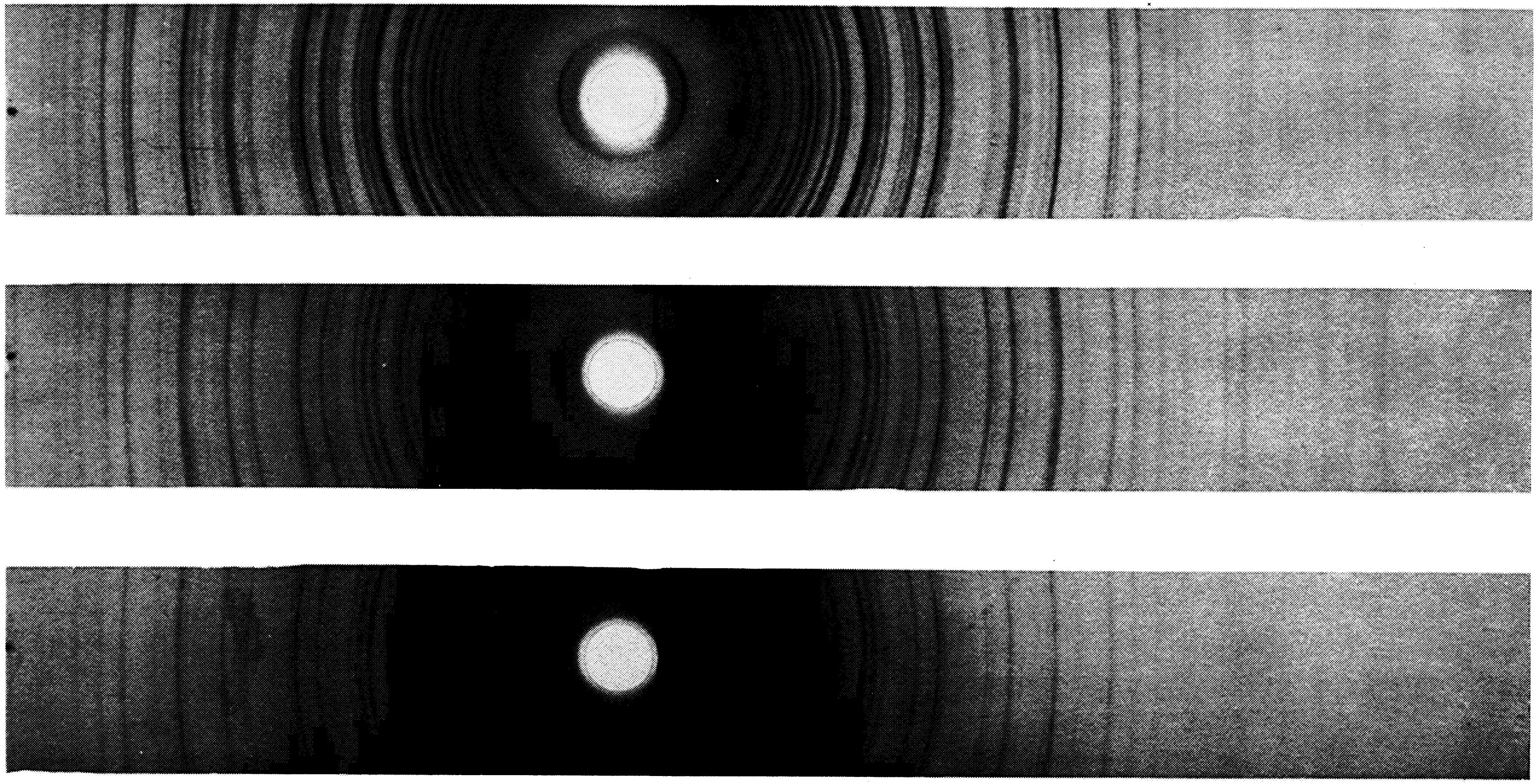

Fig. 3

A. Normal muscovite Varutrask, Sweden Specimen No. 525.

B. "Lithium muscovite" South Portland, Me. Specimen No. 471a (M-98a).

C. 3-layer muscovite Sultan Basin, Wash. Specimen No. 615 (M239).

A.
B.
C.
D.

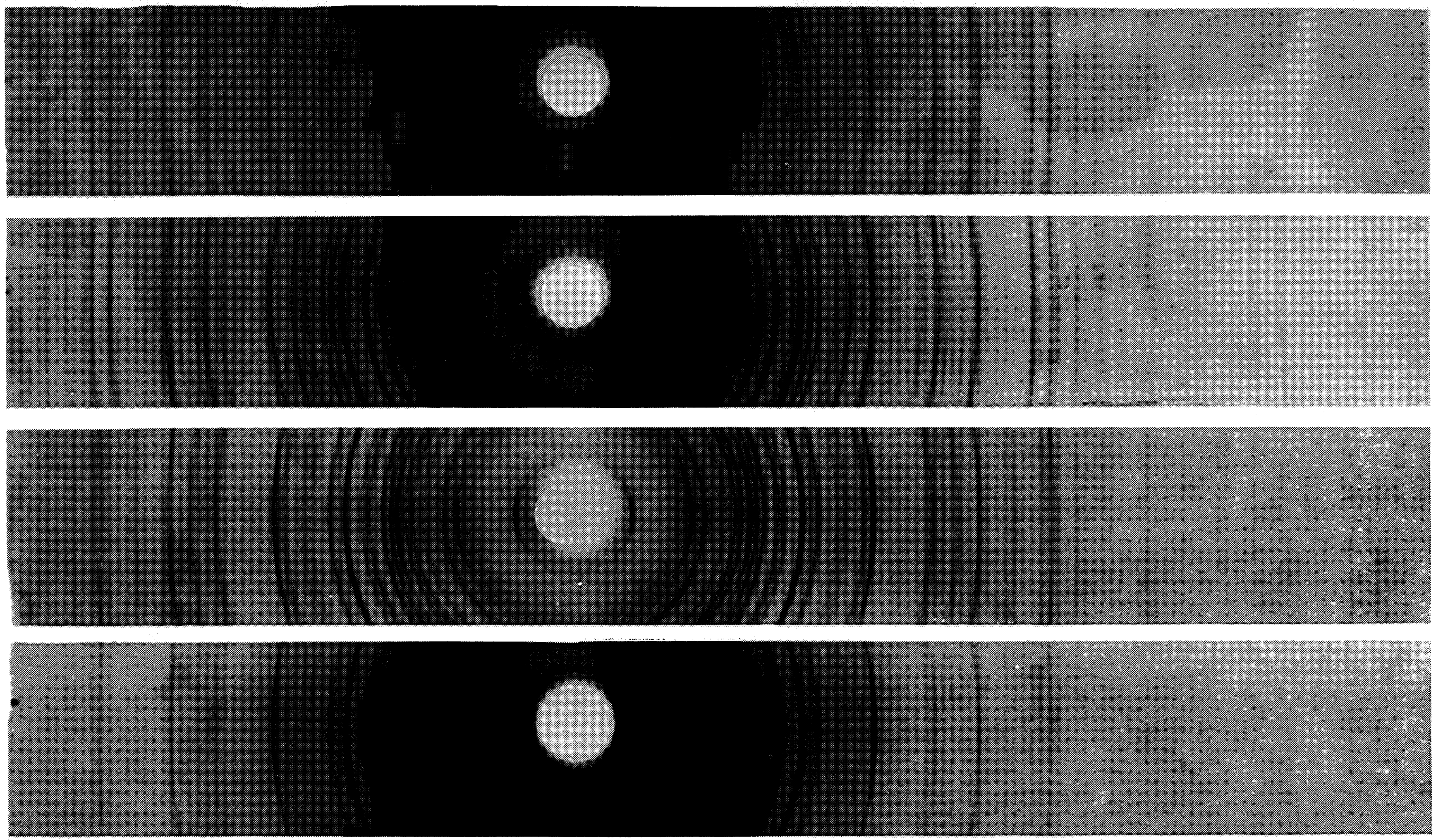

Fig. 4

A. 1-layer lepidolite Topsham, Me. Specimen No. 467b (M-94b).

B. 3-layer rhombohedral lepidolite Western Australia Specimen No. 978 (M-74).

C. 6-layer lepidolite Opportunity pegmatite, Gunnison Co., Colo. Specimen No. 514 a and j (M-29 a and j).

D. 3-layer monoclinic lepidolite Skuleboda, Sweden Specimen No. 476a (M-103a).

A.

B.

C.

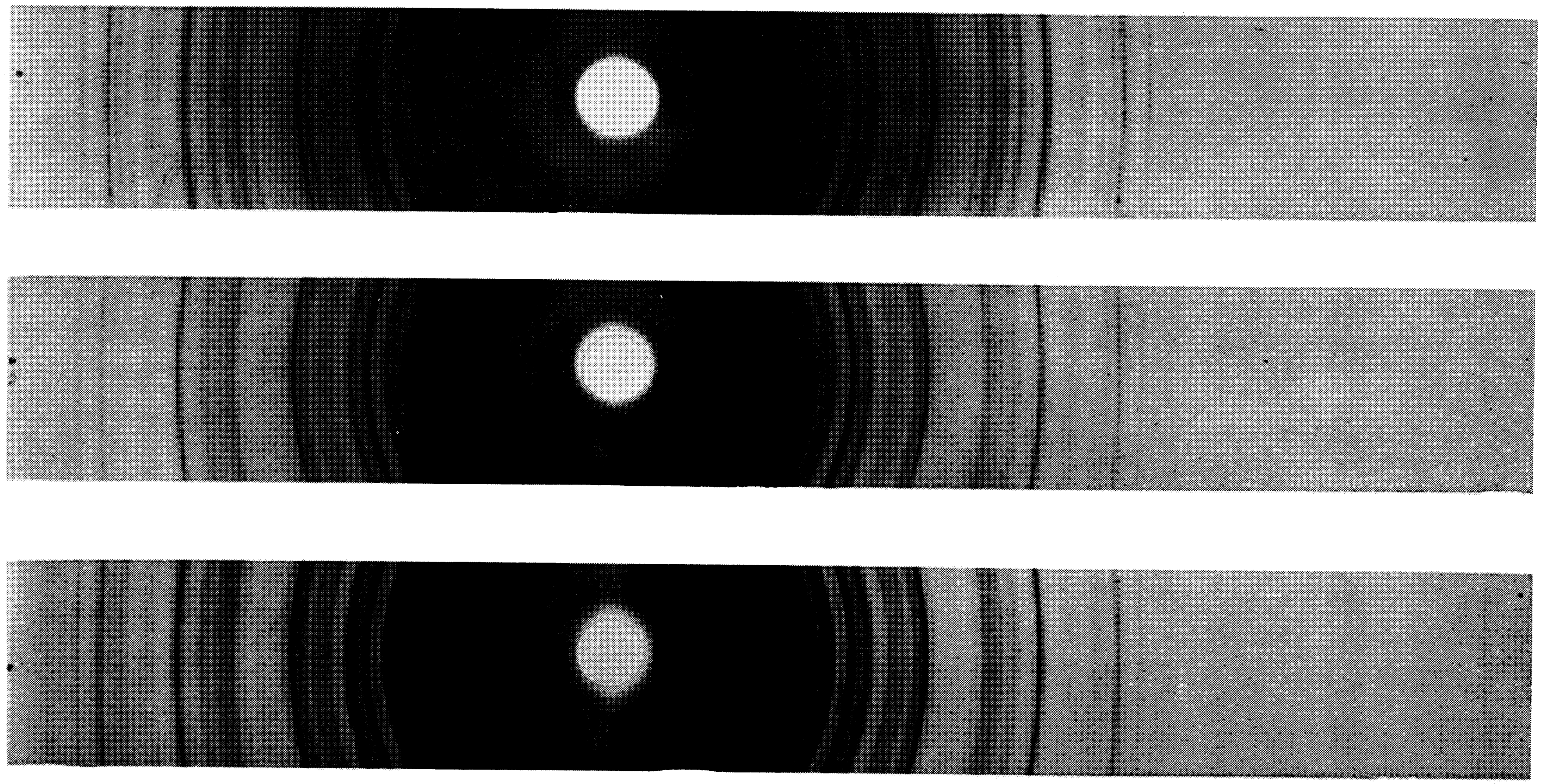

Fig. 5

A. Standard 6-layer lepidolite plus 2-layer "lithium muscovite" Specimen No. 514c (M-29c).

B. Stevens No. 2 - 6-layer lepidolite plus 2-layer "lithium muscovite" Specimen No. 976 (M-72).

C. Stevens No. 4 - 6-layer lepidolite plus 2-layer "lithium muscovite" Specimen No. 977 (M-73).

TABLE 1

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

2-layer lithium muscovite No. 471 (M-98a)		1-layer lepidolite No. 467 (M-94b)		3-layer rhombohedral lepidolite No. 978 (M-74)	
I	d spacing	I	d spacing	I	d spacing
m	9.952	s	9.908	m	9.908
m	5.007	s	4.980	m	4.968
m	4.449	w	4.494	m	4.657
vvw	3.945	vw	4.341	m	3.839
vvw	3.866	vw	4.121	m	3.580
w	3.707	vw	3.861	vs	3.309
w	3.462	s	3.608	s	3.091
ms	3.320	vs	3.333	s	2.861
m	3.208	s	3.074	w	2.651
m	2.976	s	2.867	s	2.571
s	2.841	m	2.675	mw	2.455
w	2.755	s	2.573	mw	2.375
ms	2.571	m	2.468	vw	2.243
m	2.474	m	2.387	vw	2.186
m	2.387	vw	2.253	mw	2.126
vw	2.247	m	2.132	vw	2.052
vvw	2.196	ms	1.988	m	1.984
mw	2.132	vvw	1.956	vvw	1.955
m	2.081	w	1.748	w	1.718
vw	1.951	vw	1.715	mw	1.643
w	1.742	m	1.646	w	1.611
vw	1.720	vvw	1.581	w	1.576
m	1.644	vvw	1.544	w	1.547
vvw	1.596	m	1.511	mw	1.511
vvw	1.557	vw	1.493	vvw	1.481
vvw	1.510	vvw	1.420	vvw	1.457
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				

TABLE 1 (cont)

6-layer monoclinic lepidolite
No. 514 (M-29a and j)

I	d spacing
ms	9.386
m	4.985
m	4.493
w	3.839
m	3.608
m	3.470
m	3.308
m	3.189
m	3.070
m	2.876
m	2.775
vs	2.572
m	2.416
vvw	2.248
vvw	2.190
vw	2.039
s	1.985
vvw	1.684
vvw	1.633
vvw	1.572
m	1.506
vvw	1.393
vvw	1.355
vvw	1.319
w	1.300
vvw	1.239

3-layer monoclinic lepidolite (new form)
No. 476 (M-103a)

I	d spacing
s	9.909
m	4.984
m	4.539
w	3.867
m	3.561
s	3.323
w	3.177
vw	3.109
vw	2.951
vw	2.873
vw	2.831
ms	2.604
vvw	2.542
w	2.442
vvw	2.335
vw	2.257
vw	2.171
vw	2.091
vvw	2.024
m	1.986
vw	1.711
w	1.667
vw	1.635
vw	1.596
vvw	1.551
w	1.512
vvw	1.416
vvw	1.333
vw	1.304
vvw	1.283
vvw	1.241

TABLE 2

MICAS X-RAYED BY POWDER METHODS

Number	Structure
450 (M-77)	6-layer lepidolite
456 (M-83)	6-layer lepidolite plus 2-layer "lithium muscovite"
459 (M-86)	6-layer lepidolite
467 (M-94b)	1-layer lepidolite
470 (M-97b)	1-layer lepidolite
471 (M-98a)	2-layer "lithium muscovite"
472 (M-99)	6-layer lepidolite
473 (M-100)	6-layer lepidolite
475 (M-102b)	1-layer lepidolite
476 (M-103a)	3-layer monoclinic lepidolite (new form)
481 (M-247)	2-layer normal muscovite
488 (M-254)	1-layer lepidolite plus 6-layer lepidolite
489 (M-255)	6-layer lepidolite plus 1-layer lepidolite
500 (M-262)	6-layer lepidolite plus 1-layer lepidolite
514 (M-29-1)	6-layer lepidolite plus 1-layer lepidolite
(M-29-2)	6-layer lepidolite
(M-29-3)	6-layer lepidolite plus 1-layer lepidolite
(M-29-c)	6-layer lepidolite plus 2-layer lepidolite
(M-29-a)	6-layer lepidolite
(M-29-j)	6-layer lepidolite
519 (M-272)	6-layer lepidolite
520 (M-273)	1-layer lepidolite
521 (M-274)	2-layer normal muscovite
525 (M-278)	2-layer normal muscovite
528 (M-281)	2-layer normal muscovite
529 (M-282)	2-layer normal muscovite
532 (M-285)	2-layer normal muscovite

TABLE 2 (cont)

Number	Structure
533 (M-286)	6-layer lepidolite plus 1-layer lepidolite
534 (M-287)	2-layer normal muscovite
540 (M-292)	structure undetermined (barium muscovite)
541 (M-294)	2-layer normal muscovite
542 (M-295)	2-layer normal muscovite
543 (M-296)	2-layer normal muscovite
544 (M-297)	2-layer normal muscovite
545 (M-298)	2-layer normal muscovite
546 (M-299)	2-layer normal muscovite
548 (M-301)	2-layer normal muscovite
550 (M-303)	2-layer normal muscovite
552 (M-305)	2-layer normal muscovite
553 (M-306)	2-layer normal muscovite
813 (M-109)	2-layer normal muscovite
967 (M-55)	6-layer lepidolite plus 2-layer lepidolite
975 (M-71)	6-layer lepidolite
976 (M-72)	6-layer lepidolite plus 2-layer lepidolite
977 (M-73)	6-layer lepidolite plus 1-layer lepidolite
978 (M-74)	3-layer rhombohedral lepidolite