FURTHER STUDIES OF THE LEPIDOLITE SYSTEM

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About ten years ago the writer made his third attempt[1] to correlate the chemical composition and optic properties of the lithia micas. At that time he wrote: "An attempt to solve this problem (the formulas of these micas) has met with little success," and "some examples do not agree well with the diagram. Apparently these micas are dimorphous, and that condition would doubtless entail variations in optical properties, the extent and character of which are at present unknown." Since then he has continued work on the problem and has obtained the kind cooperation of several colleagues who have generously supplied samples from various localities, in some cases, of analyzed material. It is a pleasure to acknowledge the assistance of Dr. Harry Berman of Harvard University, Dr. W. F. Foshag of the United States National Museum, Miss Jewell Glass, Dr. W. T. Schaller, and Dr. R. E. Stevens of the U. S. Geological Survey, Edward S. Simpson, Government Mineralogist of Western Australia, Dr. C. H. Stockwell of the Geological Survey of Canada, and H. Gordon Taylor of Boston, Mass.

In order to eliminate the possibility of confusion on account of including more than one crystal phase in the study, all available samples were studied by x-ray methods to determine the crystal symmetry; some samples were analyzed, and many were measured optically. This work was made possible by generous grants from the Wisconsin Alumni Research Foundation, continued for three years. In this work the author has had the efficient assistance of E. J. Lyons, R. G. Comer, J. J. Marais, John J. Ronan, B. E. Steierman and Sturges W. Bailey.

The following new analyses of lithia micas have been made for this study. Analyses 10*a*, 17*a*, 24*a* and 26*a* are included from the literature for comparison.

Two recent studies of lithia micas are of much importance. R. E. Stevens[2] published seventeeen new analyses of lithia micas and discussed their interpretation. He found that the old analyses of polylithionite were inaccurate and a new analysis leads to $K_2Li_4Al_2Si_8O_{20}F_4$ as the correct formula of this end-member of the lepidolite system. This confirms the conclusion of Mauguin that lithia micas have 24 negative $(O+F,OH)$ ions in the crystals and that they are like biotite (and unlike muscovite) in containing sixteen positive ions. The lithia micas are accordingly octophyllites, but, nevertheless, they seem to intercrystallize with biotite

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[1] Winchell, A. N., *Am. Mineral.*, **17**, 551 (1932).

[2] Stevens, R. E., *Am. Mineral.*, **23**, 607 (1938).

TABLE 1

Sample No.	10	10a	17	17a	18	24	24a	26	26a
SiO$_2$	49.19	49.58	49.14	51.11	50.16	46.74	45.23	51.88	50.31
TiO$_2$	0.08	0.06	0.00		0.00	0.00	n.d.	0.21	0.22
Al$_2$O$_3$	24.81	23.87	27.66	25.26	23.54	21.78	21.78	20.65	19.95
Fe$_2$O$_3$	0.24	0.21	0.43	0.20	0.46	1.19	0.47	0.79	0.99
FeO				0.07	1.27	10.22	11.25	1.99	2.55
MnO	2.51	2.78	1.22	0.17	0.79	0.37	1.73	2.01	2.63
MgO	0.05	0.00	0.05	0.01	0.06	0.00	0.23	0.00	0.02
CaO	tr.	0.00	tr.	0.12	0.04	0.00	n.d.	0.00	tr.
Li$_2$O	5.10	5.05	3.95	4.98	5.34	3.72	3.23	5.26	5.39
Na$_2$O	0.52	0.57	0.40	1.43	1.10	0.54	0.78	0.51	0.59
K$_2$O	10.25	10.14	10.13	10.51	10.97	10.37	9.98	10.55	10.14
Rb$_2$O	1.78	1.62	1.17	n.d.	n.d.	n.d.	n.d.	n.d.	0.97
Cs$_2$O	0.19	0.09	0.62	n.d.	n.d.	n.d.	n.d.	n.d.	0.06
H$_2$O$-$	1.21	n.d.	2.64	n.d.	0.78	n.d.	n.d.	n.d.	n.d.
H$_2$O$+$		1.22		0.94	1.39	0.89	0.97	1.89	0.88
F	6.89	7.49	5.21	6.57	6.75	7.54	7.98	7.65	7.65
Cl	n.d.	n.d.	n.d.	n.d.	0.05	n.d.	n.d.	n.d.	n.d.
	102.82	103.19	102.62	103.11	102.70	103.36	103.63	103.39	102.51
O$=$F	2.90	3.15	2.19	2.76	2.85	3.17	3.35	3.22	3.22
	99.92	100.04	100.43	100.35	99.85	100.19	100.28	100.17	99.29

10. Lepidolite, Ohio City, Colorado. R. B. Ellestad, analyst. U. S. Nat. Mus. No 97893.

10a. Lepidolite, Ohio City, Colorado, R. E. Stevens, analyst, *Am. Mineral.*, **23,** 607 (1938). U. S. Nat. Mus. No. 97893.

17. Lepidolite, Mt. Apatite, Auburn, Maine. R. B. Ellestad, analyst, U. S. Nat. Mus. No. 80230.

17a. Lepidolite, Auburn, Maine. R. B. Riggs, analyst. *U. S. Geol. Surv., Bull.* **42,** 17 (1887).

18. Lepidolite, Alabaschta, Urals. F. A. Gonyer, analyst.

24. Zinnwaldite, Zinnwald, Erzgebirge. F. A. Gonyer, analyst.

24a. Zinnwaldite, Zinnwald, Erzgebirge, W. Kunitz: *N. Jahrb. Mineral., Bl. Bd.,* **50,** 365 (1924).

26. Zinnwaldite, Wakefield, Quebec. F. A. Gonyer, analyst.

26a. Zinnwaldite, Wakefield, Quebec. R. E. Stevens: *Am. Mineral.*, **23,** 607 (1938).

little, if at all. Being octophyllites, they do not intercrystallize with the heptaphyllite micas (the muscovite system), just as is to be expected, although they are frequently closely associated with muscovite—perhaps even interleaved with it in some cases.

A second paper of great importance in regard to lithia micas was published very recently by Hendricks and Jefferson.[3] They show that micas

[3] Polymorphism of the micas: *Am. Mineral.*, **24,** 729 (1939).

are not only dimorphous, but polymorphous, there being one hexagonal, four monoclinic and two triclinic phases. They find evidence that a small amount of lithia (at most probably about 3%), can enter the muscovite structure, but this produces a lithium-bearing muscovite and not a member of the lepidolite system. Only three structures have been found in that system, namely the three layer hexagonal (uniaxial), the single layer monoclinic hemihedral and the six layer monoclinic hemihedral. But the optical properties of some lithia micas make it probable that they can also take on either the six or the twenty-four layer triclinic structure.

In attempting to solve the problem of the composition of the end members of the lepidolite system it is important, first, to select the best analyses of the lithia micas, and, second, to eliminate those which are lithium-bearing muscovites, or biotites, rather than true members of the lepidolite system. There seems to be no infallible method to select the best analyses, but it is probably prudent to omit all those more than twenty years old and also certain others which seem incomplete or unsatisfactory. Stevens' excellent analyses include one (No. 1) which Hendricks and Jefferson have proved to be a lithium-bearing muscovite and four (Nos. 2–5) which certainly are not lepidolites. Omitting these, the analyses of most importance for the purpose may be listed as follows:

1. Lepidolite, Epprechtstein, Fichtelgebirge. W. Kunitz: *N. Jahrb. Mineral., Bl. Bd.* **50,** 365 (1942). Kunitz No. 3.
2. Lepidolite, Penig-Chursdorf. W. Kunitz: *loc. cit.*
3. Lepidolite. Penig-Auersdorf. W. Kunitz: *loc. cit.*
4. Lepidolite, Indiakawa, Mursinsk. W. Kunitz: *loc. cit.*
5. Lepidolite, Rozna, Moravia. J. Sekanina and J. Vyslonzil: *Mineral. Abst.,* **4,** 379 (1930).
6. Lepidolite, Stewart mine, Pala, California. R. E. Stevens: *Am. Mineral.,* **23,** 607 (1938). Stevens No. 6.
7. Lepidolite, Ohio City, Colorado. R. E. Stevens: *loc. cit.* No. 7.
8. Lepidolite, Himalaya mine, Mesa Grande, California. R. E. Stevens: *loc. cit.* No. 8.
9. Lepidolite, San Diego mine, Mesa Grande, California. R. E. Stevens: *loc. cit.* No. 9.
10. Lepidolite, Ohio City, Colorado. R. B. Ellestad., analyst. See Table 1.
11. Lepidolite, Stewart mine. Pala, California. R. E. Stevens: *loc. cit.* No. 11.
12. Lepidolite, Stewart mine, Pala, California. R. E. Stevens: *loc. cit.* No. 12.
13. Lepidolite, Himalaya mine, Mesa Grande, California. R. E. Stevens: *loc. cit.* No. 13.
14. Lepidolite, Calgoorlie, W. Australia, R. E. Stevens: *loc. cit.* No. 14.
15. Lepidolite, Little Three mine, Ramona, California. R. E. Stevens: *loc. cit.* No. 15.
16. Lepidolite, Antsongombato, Madagascar. R. E. Stevens: *loc. cit.* No. 16.
17. Lepidolite, Mt. Apatite, Maine. R. B. Ellestad, analyst. See Table 1.
18. Lepidolite, Alabaschta, Urals. F. A. Gonyer, analyst. See Table 1.
19. Polylithionite, Kangarluarsuk, Greenland. R. E. Stevens: *loc. cit.* No. 17.
20. Protolithionite, Zinnwald, Erzgebirge. W. Kunitz: *loc. cit.* No. 8 (p. 413) and No. 11 (pp. 394, 395).
21. Zinnwaldite, Altenberg, Erzgebirge. W. Kunitz: *loc. cit.* No. 7 (p. 413) and No. 10 (pp. 394, 395).

22. Zinnwaldite, Zinnwald, Erzgebirge. W. Kunitz: *loc. cit.* No. 6 (p. 413) and No. 9 (pp. 394., 395).

23. Zinnwaldite, Schlaggenwald, Bohemia. W. Kunitz: *loc. cit.* No. 5 (p. 413) and No. 8 (pp. 394, 395).

24. Zinnwaldite, Zinnwald, Erzgebirge. F. A. Gonyer, analyst. See Table 1.

25. Zinnwaldite, Wakefield, Quebec. R. E. Stevens; *loc. cit.* No. 10.

26. Zinnwaldite, Wakefield, Quebec. F. A. Gonyer, analyst. See Table 1.

27. Zinnwaldite, Morefield mine, Amelia, Virginia. J. J. Glass: *Am. Mineral.*, **20,** 741 (1935).

28. Taeniolite, Magnet Cove, Arkansas. H. D. Miser and R. E. Stevens: *Am. Mineral.*, **23,** 104 (1938).

With the formula of one end-member (polylithionite) of the lepidolite system now well established as $K_2Li_4Al_2Si_8O_{20}F_4$, the formulas of other end-members can be written with somewhat less uncertainty. The lithia micas are now known to be octophyllites, that is, they have eight positive ions for twelve negative ions; therefore formulas like $K_4Li_6Al_6Si_{15}O_{40}F_8$ are unsatisfactory. $K_4Li_7Al_7Si_{14}O_{40}F_8$ is correct, but it is not an end-member, since it gives neither maximum nor minimum tenor of any element. So far as known $K_2Li_3Al_5Si_6O_{20}F_4$ represents the minimum tenor of Li for lepidolites containing no divalent atoms, and $K_2LiFe_4Al_3Si_6O_{20}F_4$ (protolithionite) seems to represent the maximum tenor of divalent atoms together with minimum tenor of lithium. The second end-member ($K_2Li_3Al_5Si_6O_{20}F_4$) has no name and no varietal name in the literature seems to be appropriate. In these circumstances the writer would suggest that it be called paucilithionite.

Including the new analyses of Stevens, more than two dozen high grade analyses of lithia micas are now available (see Table 1). If they are calculated into the end-members named above (see Table 2) a surprising result is obtained: Li_2O is deficient in all cases (except one) and the average deficiency is almost one per cent by weight (0.91). This result demands an explanation. It seems that it must be due to one (or more) of the following causes:

1. The formulas of the end-members are incorrect. At first thought this seems to be the most probable explanation. However, Stevens' analysis of polylithionite is the only one that shows no deficiency in lithia and therefore that formula seems to be incontestable. With the known structure of micas in mind and the requirement of eight positive ions for each twelve negative ions a reduction in lithium below the tenor shown in $K_2Li_3Al_5Si_6O_{20}F_4$ (assuming no other elements present) leads to the formula $K_2Li_2Al_8Si_4O_{20}F_4$.

This formula requires decidedly more alumina and less silica than are found in any mica, and, moreover, if it is used as an end member with $K_2Li_4Al_2Si_8O_{20}F_4$ computation of analyses so as to use all the silica and

TABLE 2. DISCREPANCIES IN SELECTED ANALYSES OF LITHIA MICAS

Assuming as end-members: $K_2Li_4Al_2Si_8O_{20}F_4$, $K_2Li_3Al_5Si_6O_{20}F_4$ and $K_2LiFe_4Al_3Si_6O_{20}F_4$

Anal- ysis No.	Li₂O		K₂O+Na₂O+ Rb₂O+Cs₂O		(F, OH)		Remarks
	Rel. Mol.	Weight %	Rel. Mol.	Weight %	Rel. Mol.	Weight %	
1	− 29	− .87	+10	+ .94	+ 84	+ 1.60	Explanation of Table 2.
2	− 51	− 1.53	+10	+ .94	− 24	− .45	The relative numbers of
3	− 58	− 1.74	+16	+1.50	− 26	− .49	molecules (Rel. Mol.)
4	− 33	− .99	+13	+1.22	+ 8	+ .15	are obtained by divid-
5	− 31	− .93	+11	+1.03	− 4	− .08	ing each weight per-
6	− 52	− 1.56	− 4	− .38	+ 14	+ .27	centage of each analy-
7	− 26	− .78	+ 2	+ .19	+ 40	+ .76	sis by its molecular
8	− 56	− 1.68	−11	−1.03	− 14	− .27	weight, multiplied by
9	− 36	− 1.08	+ 7	+ .66	+ 42	+ .80	one thousand to avoid
10	− 25	− .75	+ 3	+ .28	+ 2	+ .04	decimals, and then
11	− 12	− .36	0	0	+ 12	+ .23	making as many as
12	− 29	− .87	0	0	+ 44	+ .84	possible of each end-
13	− 19	− .57	0	0	− 2	− .04	member using all the
14	− 13	− .39	0	0	− 10	− .19	silica and alumina.
15	− 11	− .33	+ 3	+ .28	+ 40	+ .76	With F, H₂O+ is in-
16	− 1	− .03	+ 2	+ .19	+ 30	+ .57	cluded (as 2 OH) in all
17	− 66	− 1.98	− 5	− .47	+ 60	+ 1.14	cases and H₂O− is in-
18	− 22	− .66	+ 2	+ .19	+ 14	+ .27	cluded (as 2 OH) in the
19	+ 4	+ .12	− 1	− .09	− 34	− .65	calculations of Nos. 8,
20	− 25	− .75	+ 6	+ .56	+ 48	+ .91	10, 13, 14, 17, and 27.
21	− 34	− 1.02	+ 8	+ .75	+ 38	+ .72	The weight percentage
22	− 34	− 1.02	0	0	+ 32	+ .61	of K₂O+Na₂O+Rb₂O
23	− 28	− .84	+ 5	+ .47	+ 88	+ 1.67	+Cs₂O is obtained by
24	− 30	− .90	0	0	+ 20	+ .38	assuming that the defi-
25	− 10	− .30	+ 1	+ .09	+ 18	+ .34	ciency or excess is
26	− 24	− .72	− 4	− .38	+118	+ 2.24	solely K₂O. Similarly
27	− 69	− 2.07	+ 2	+ .19	− 22	− .42	(F, OH) is assumed to be F for this calcula-
	−820	−24.60	+76	+7.13	+616	+11.71	tion.
Aver- age	− 30.4	− 0.91	+ 2.8	+ .264	+ 22.7	+ .43	

alumina shows that the deficiency in lithia remains unchanged. It seems reasonable to conclude that this method of reducing the lithium required by the formulas is not correct.

2. In spite of the great inequality in the atomic domains is it possible that potassium may proxy for lithium to a limited extent? This explanation is clearly unsatisfactory, since the average number of atoms of potassium apparently available is less than one-tenth of the number needed

to supply the average deficiency of lithium, and the excess of potassium does not increase with the deficiency of lithium; on the contrary two of the analyses with the largest deficiencies of lithium (No. 6 and 8) are also deficient in potassium.

3. It is doubtless quite unnecessary to point out that fluorine cannot proxy for lithium since these ions have opposite electric charges. However, even if this were possible, it would not solve the problem since the average excess of fluorine (22.7 ions) is only about one-third the average deficiency of lithium (60.8 ions).

4. Can the deficiency in lithia be due to errors in the chemical analyses? The quantitative determination of Li_2O is not easy and some old analyses are clearly inaccurate, but the careful analyses of recent years are unquestionably good and it is doubtful whether even one-tenth of the average deficiency actually found can be reasonably ascribed to errors.

5. Can the deficiency in lithia be due to impure samples? It is only necessary to read the description of the precautions used by Stevens to ensure the purity of his samples to reach the conclusion that any impurities (in the ordinary sense) in his samples were far less than necessary to explain the deficiency in lithia. It is true that the average deficiency in lithia (0.66 weight %) in his analyses is less than the average found in others, but two of his analyses show deficiencies exceeded only rarely in other analyses, so that his precautions failed to eliminate the main cause of the difficulty.

6. Can the deficiency in lithia be due to another end-member of an isomorphous system, not replacing, but supplementing, those already used? It seems to be difficult, if not impossible, to prove or to disprove this hypothesis, but the writer has failed to discover another end-member which has eight positive ions for twelve negative ions and will eliminate the lithia deficiency without introducing other important discrepancies. Apparently no such a thing exists.

7. Can the deficiency in lithia be explained by assuming the presence of some heptaphyllite mica such as muscovite, $H_2KAl_3Si_3O_{12}$, containing only seven positive ions for twelve negative ions? The results of this assumption are shown in Table 3.

It appears that this assumption suffices to eliminate the deficiency in lithia in all cases (in one case it is cut down from -69 only to -3) without introducing any other inconsistencies except for the fact that the average excess of K_2O molecules is increased from 2.8 to 4.1, and the average excess of F ions is increased from 22.7 to 27.3. However, the average excess of K_2O in the twelve analyses of Stevens is only 0.6, an amount probably within the limits of accuracy that can be expected. And, since Stevens has proved that Rb_2O and Cs_2O are normal constituents of lithia micas, it is evident that analyses in which these elements are included

TABLE 3. DISCREPANCIES IN SELECTED ANALYSES OF LITHIA MICAS ASSUMING SOME MUSCOVITE, $H_2KAl_3Si_3O_{12}$, PRESENT WITH $K_2Li_4Al_2Si_8O_{20}F_4$, $K_2Li_3Al_5Si_6O_{20}F_4$ AND $K_2LiFe_4Al_5Si_6O_{20}F_4$

Analysis No.	$K_2O+Na_2O+Rb_2O$ $+Cs_2O$		F, OH		Mol. % $H_2KAl_3Si_3O_{12}$	Remarks
	Rel. Mol.	Weight %	Rel. Mol.	Weight %	(Muscovite)	
1	+ 12	+ 1.13	+ 90	+ 1.71	16.0	After the calculations
2	+ 12	+ 1.13	− 14	− .27	27.5	for Table 2 it is neces-
3	+ 20	+ 1.88	− 10	− .19	31.7	sary merely to com-
4	+ 15	+ 1.41	+ 14	+ .27	17.8	pute from the original
5	+ 7	+ .66	− 22	− .42	15.4	analysis two-thirds as
6	− 2	− .19	+ 16	+ .30	27.2	many muscovite mole-
7	+ 2	+ .19	+ 38	+ .72	14.6	cules as the number of
8	− 8	− .75	− 1	− .02	29.9	lithia deficiency mole-
9	+ 1	+ .09	+ 20	+ .38	18.2	cules in order to elimi-
10	+ 3	+ .28	+ 6	+ .11	13.0	nate this deficiency—
11	+ 3	+ .28	+ 13	+ .25	6.6	though the percentages
12	+ 1	+ .09	+ 48	+ .91	16.0	of the lithia end-mem-
13	− 3	− .28	+ 10	+ .19	9.3	bers do not remain un-
14	+ 1	+ .09	− 6	− .11	6.6	changed.
15	+ 4	+ .38	+ 41	+ .78	6.5	(In 27 the deficiency
16	+ 2	+ .19	+ 31	+ .59	0.8	can be reduced from 69
17	− 3	− .28	+ 68	+ 1.29	34.4	to 3, but not to 0.)
18	+ 13	+ 1.22	+ 62	+ 1.18	11.4	
19	− 1	− .09	− 34	− .65	0.0	
20	+ 7	+ .66	+ 52	+ .99	15.0	
21	+ 10	+ .94	+ 46	+ .87	19.3	
22	+ 2	+ .19	+ 42	+ .80	18.9	
23	+ 7	+ .66	+ 72	+ 1.37	15.4	
24	+ 1	+ .09	+ 25	+ .48	17.0	
25	+ 2	+ .19	+ 22	+ .42	5.8	
26	− 3	− .28	+121	+ 2.30	13.0	
27	+ 7	+ .66	− 12	− .23	37.7	
	+112	+10.53	+738	+14.02	445.0	
Average	+ 4.1	+ 0.39	+ 27.3	+ 0.32	16.5	

with K_2O must show an apparent excess of that oxide. In fact, if the percentages of Rb_2O and Cs_2O determined by Stevens are assumed to be K_2O his analyses will then show an apparent excess of K_2O very similar to (though even greater than) the average excess found in the other analyses. Since water is included with fluorine, and water is notoriously difficult to determine accurately, the increase in the average excess of this constituent from 22.7 to 27.3 ions does not seem important.

Accordingly, it seems that all the best analyses of lithia micas can be explained satisfactorily in terms of these four molecules. However, the writer accepts only three of these as end-members of the lepidolite system and considers that the muscovite molecule enters into crystal solution in this system only very little, if at all. The reasons for this view need careful and full statement.

1. Theoretically, it seems improbable that a crystal structure containing eight positive ions for twelve negative ions can change *gradually* into one containing seven positive ions for twelve negative ions. One can get a mental picture of the problem by trying to imagine a *gradual* change from the structure shown in Hendricks and Jefferson's[4] Fig. 1*a* to that shown in their Fig. 1*b*. Or, assuming that the structure remains that of Fig. 1*a*, how can the eight ions of that figure change *gradually* into the seven ions of Fig. 1*b*? It seems probable the layer of ions having octo-hedral coordination must be uniform—either the one pattern or the other.

2. Actually it is well known to every student of rocks that muscovite and biotite often exist side by side. If heptaphyllites and octophyllites can intercrystallize to form one substance their existence side by side should be rare and due to formation at different times. Actually it is very common and there is every reason to think that the two minerals formed at the same time, at least in many cases.

3. Analyses of micas show only rare and doubtful cases that are supposed by some writers to represent types intermediate between muscovite and biotite. A sample of "alurgite" from California has been described[5] as such a type, but the evidence does not seem conclusive to the writer nor to Schaller.[6]

4. Lepidolite and muscovite are also often closely associated. Such a condition is described by many writers including Scherizer,[7] Baumhauer,[8] Bowman,[9] Stockwell,[10] and Kunitz.[11] In some cases they are described as formed at the same time; in other cases they have zonal arrangement showing formation at different times. If they can intercrystallize to form one substance they should never form simultaneously side by side. An especially good illustration of this condition is furnished by Stevens who writes: "The lepidolite samples were furnished by Dr. Schaller, of the

[4] *Am. Mineral.*, **24,** 131 (1939).

[5] Webb, R. W., *Am. Mineral.*, **24,** 123 (1939)

[6] *Am. Mineral.*, **24,** 127 (1939).

[7] *Zeits. Kryst.*, **12,** 5 (1886); **13,** 22, 464 (1887).

[8] *Zeits. Kryst.*, **51,** 344 (1912).

[9] *Mineral. Mag.*, **13,** 97 (1902).

[10] *Trans. Roy. Soc. Canada*, **27,** 27 (1933).

[11] *Chemie der Erde*, **4,** 231 (1930).

U. S. Geological Survey, who examined them optically to be sure that no impurities were present which could not be easily removed. Five samples were discarded because the small flakes of lepidolite were found to contain numerous minute inclusions of muscovite . . . This occurrence of minute inclusions of muscovite in lepidolite is apparently not unusual."

It is the writer's theory that such intergrowths of muscovite and lepidolite are very common and of all sizes from those which are apparent in museum-size crystals, through those observed by Schaller microscopically to units which are too small to be seen microscopically, but nevertheless are quite different in character from crystal solution, since they are *two* substances, whereas crystal solution of two (or more) end-members leads to *one* substance. The writer[12] demonstrated some years ago that quartz may exist as an aggregate of units which are too small to be seen with a microscope and so small that many exist in the thickness of a thin section and, being in random position, compensate each other in birefringence effects so that the aggregate is isotropic. Moreover, by chance the quartz units at a given point in the thin section may be dominantly in or near one orientation and then that point will show very weak birefringence. Similarly, the units of muscovite and lepidolite may be in entirely irregular positions, but are probably in most cases more or less parallel, especially as to the vertical axes, so that the basal cleavages are parallel. Thus a cleavage plate, apparently from one single crystal, may actually contain both minerals in submicroscopic intergrowth, but not at all in crystal solution.

Unfortunately this conception must be presented as a theory whose validity the writer is unable to demonstrate at the present time. However, various facts tend to support the theory.

1. It is known that muscovite and lepidolite form aggregates (with parallel cleavages) when both micas are in large units (*e.g.*, one or more centimeters across). It is likewise known that they form aggregates when one or both are in small units—units which are easily visible microscopically, but probably *about* one-hundredth of a millimeter in diameter since they are described as "minute." That is, the known range in size is from about 10 mm. to about .01 mm. If the range of size actually extends downward a tenth again as much as is known, it would be quite sufficient for the theory.

2. The common existence of these minerals side by side suggests that they cannot intercrystallize to form one substance.

3. A similar condition of a mineral in submicroscopic units has been proved to exist in quartz.

[12] Winchell, A. N., *Am. Mineral.*, **9**, 235 (1924).

4. If muscovite and lepidolite actually intercrystallize to form one substance that substance should have optical properties which are the same in all parts of a crystal, or any single mass of crystals formed at the same time (assuming no zonal growth to be present). On the other hand, if muscovite and lepidolite form interleaved aggregates it is to be expected that they would be present in variable proportions so that optical properties would vary in different parts of a crystal or mass of crystals. The results of careful measurements of refractive indices of lithia mica samples from various localities are assembled in Table 4.

TABLE 4. MEASURED PROPERTIES OF LITHIA MICAS

No.	Calc. % Musc.	G	N_g	N_m	N_p	N_g-N_p	2V	Measured by
1	16.0	2.820	1.5563	1.5525	1.5295	0.0268	46°6'	W. Kunitz: *Neues Jahrb.*
2	27.5							*Min. Bl. Bd.50,* 365 (1924(
3	31.7	2.837	1.5617	1.5574	1.5329	0.0288	43°46'	W. Kunitz: *loc. cit.*
4	17.8	2.881	1.5661	1.5626	1.5371	0.0291	40°28'	W. Kunitz: *loc. cit.*
5	15.4	2.832		1.558			39°	J. Sekanina: *Min. Abst.,* 4, 379
6	27.2		1.560[1]				25–30°	M. E. Jefferson: *Am. Mineral.,* 24, 729
6	27.2		1.5581	1.5553	1.5307 calc.	0.0274	37.2°	J. J. Marais[2]
7	14.6		1.562				25–40°	M. E. Jefferson: *loc. cit.*
7	14.6		1.5668	1.5635	1.5398	0.0270	36.8°	J. J. Marais
7	14.6		1.5630	1.5616	1.5480 calc.	0.0150	35°	J. J. Ronan[2]
7	14.6		1.5632	1.5600	1.5292 calc.	0.0340	35°	J. J. Ronan
7	14.6		1.5636	1.5612	1.5262 calc.	0.0275	35°	J. J. Ronan
8	21.8		1.56				30–40°	M. E. Jefferson: *loc. cit.*
8	21.8		1.5633	1.5575	1.5356	0.0277	50.5°	J. J. Marais
8	21.8		1.5582	1.5561			34°	J. J. Marais
			−1.5599	−1.5568				
8	21.8		1.5607	1.5578	1.5452 calc.	0.0175	47°	J. J. Ronan
8	21.8		1.5611	1.5568	1.5381 calc.	0.0230	50°	J. J. Ronan
8	21.8		1.5634	1.5582	1.5409 calc.	0.0225	50°	J. J. Ronan
8	21.8		1.5634	1.5593	1.5414 calc.	0.0220	49°	J. J. Ronan
9	18.2		1.555				55–58°	M. E. Jefferson: *loc. cit.*
9	18.2		1.5577	1.5530	1.5331	0.0246	52.5°	J. J. Marais
9	18.2		1.5590	1.5552	1.5336	0.0254	50.2°	J. J. Marais
9	18.2		1.5591	1.5540	1.5318	0.0273	50.6°	J. J. Marais
10	13.0		1.5647	1.5627	1.5335	0.0292	35°	R. G. Comer[2]
10	13.0		1.5668	1.5635	1.5398	0.0270	36.8°	J. J. Marais
10	13.0		1.5620	1.5586	1.5315 calc.	0.0305	38°	J. J. Ronan
10	13.0		1.5620	1.5598	1.5398 calc.	0.0222	36°	J. J. Ronan
11	6.6							R. E. Stevens: *Am. Mineral.,* 23, 607
12	16.0		1.554				35°	M. E. Jefferson: *loc. cit.*
12	16.0		1.5567	1.5545	1.5344	0.0233	35°	J. J. Marais
13	9.3		1.558				25–30°	M. E. Jefferson: *loc. cit.*
13	9.3		1.5590	1.5542	1.5330	0.0260	56°	J. J. Marais
13	9.3		1.5591	1.5568	1.5372	0.0219	30.6°	J. J. Marais
13	9.3		1.5594	1.5547	1.5356	0.0238	46.2°	J. J. Marais
13	9.3		1.5603	1.5554	1.5333	0.0270	50°	J. J. Marais
14	6.6		1.558	1.558	1.535	0.033	0°	M. E. Jefferson: *loc. cit.*
14	6.6		1.5546	1.5546	1.5324	0.0222	0°	J. J. Marais
			−1.5556	−1.5556	−1.5334	−0.0234		
15	6.5		1.555	1.5515	1.533	0.022	45°	M. E. Jefferson: *loc. cit.*
15	6.5		1.5546	1.5519	1.5315	0.0232	45°	R. G. Comer

TABLE 4. MEASURED PROPERTIES OF LITHIA MICAS—*Continued*

No.	Calc. % Musc.	G	N_g	N_m	N_p	$N_g - N_p$	2V	Measured by
15	6.5		1.5546	1.5518	1.5318	0.0228	46.5°	J. J. Marais
15	6.5		1.5556	1.5522	1.5325	0.0231	44.2°	J. J. Marais
15	6.5		1.5542	1.5512	1.5452 calc.	0.0190	46°	J. J. Ronan
16	0.8		1.5562	1.5522	1.5318	0.0244	45.5°	Wunder and Sabot[a]
16	0.8		1.558					M. E. Jefferson: *loc. cit.*
16	0.8		1.5540	1.5508	1.5345	0.0195	48.2°	J. J. Marais
16	0.8		1.5555	1.5528	1.5346	0.0209	49.3°	J. J. Marais
16	0.8		1.5571	1.5537	1.5343	0.0228	48.2°	J. J. Marais
16	0.8		1.5574	1.5537	1.5334	0.0240	48.3°	J. J. Marais
17	34.4		1.5863	1.5804	1.5516	0.0347	45°	R. G. Comer
17	34.4		1.5584	1.5565	1.5330	0.0254	32°	J. J. Marais
17	34.4		1.5604	1.5588	1.5430 calc.	0.0174	36°	J. J. Marais
17	34.4		1.5624	1.5594	1.5332	0.0292	43°	J. J. Marais
18	11.4		1.5574	1.5540	1.5325	0.0249	45.6°	J. J. Marais
18	11.4		1.5577	1.5562	1.5390	0.0187	32°	J. J. Marais
18	11.4		1.5581	1.5547	1.5314	0.0267	45.6°	J. J. Marais
18	11.4		1.5607	1.5564	1.5310	0.0297	46.5°	J. J. Marais
19	0.0	2.81[4]	1.569				40°	M. E. Jefferson: *loc. cit.*
19	0.0		1.5640	1.5607	1.5404	0.0235	43.6°	J. J. Marais
19	0.0		1.5648	1.5634	1.5465	0.0183	38.2°	J. J. Marais
19	0.0		1.5660	1.5636	1.5441	0.0219	38.8°	J. J. Marais
19	0.0		1.5692	1.5673	1.5541	0.0151	38°	J. J. Marais
19	0.0		1.5620	1.5596	1.5380 calc.	0.0240	36°	J. J. Ronan
19	0.0		1.5634	1.5595	1.5253 calc.	0.0381	37°	J. J. Ronan
19	0.0		1.5650	1.5616	1.5350 calc.	0.0300	39°	J. J. Ronan
19	0.0		1.5670	1.5638	1.5430 calc.	0.0240	42°	J. J. Ronan
19	0.0		1.5673	1.5640	1.5375 calc.	0.0298	39°	J. J. Ronan
19	0.0		1.5674	1.5642	1.5383 calc.	0.0291	38°	J. J. Ronan
19	0.0		1.5678	1.5631	1.5362 calc.	0.0316	44°	J. J. Ronan
19	0.0		1.5708	1.5672	1.5482 calc.	0.0226	46°	J. J. Ronan
19	0.0		1.5736	1.5686	1.5631 calc.	0.0375	42°	J. J. Ronan
20	15.0	3.148	1.6057	1.6057	1.5724 calc.	0.0333	0°	W. Kunitz: *loc. cit.*
21	19.3	3.018	1.5876	1.5850	1.5572	0.0304	20.7°	W. Kunitz: *loc. cit.*
22	18.9	2.987	1.5812	1.5777	1.5511	0.0301	29.6°	W. Kunitz: *loc. cit.*
23	15.4	2.97	1.5792	1.5754	1.5485	0.0307	31.3°	W. Kunitz: *loc. cit.*
24	17.0		1.5750	1.5729	1.5435	0.0315	32°	R. G. Comer
24	17.0		1.5757	1.5737	1.5450	0.0307	30°	R. G. Comer
25	5.8		1.562				30°	M. E. Jefferson: *loc. cit.*
			1.5628	1.5596	1.5357	0.0271	39°	R. G. Comer
			1.5674	1.5637	1.5440	0.0234	38°	R. G. Comer
26	13.0		1.5701	1.5671	1.5415	0.0286	34°	J. J. Marais
27	37.7		1.580	1.580	1.550	0.030	0–33°	J. J. Glass
			−1.590	−1.590	−1.558	−0.032		
27	37.7		1.5770	1.5754	1.5463	0.0307	28°	R. G. Comer
27	37.7		1.5781	1.5762	1.5424	0.0357	30°	R. G. Comer
27	37.7		1.5783	1.5760	1.5400	0.0383	30.4°	J. J. Marais
27	37.7		1.5784	1.5767	1.5488	0.0296	30.1°	J. J. Marais
27	37.7		1.5786	1.5762	1.5450	0.0336	30.4°	J. J. Marais
28	0.0		1.553	1.553	1.522	0.031	0°±	Miser and Stevens: *Am. Mineral.*, 23, 104 (1938)

[1] Corrected from 1.550 in personal communication from M. E. Jefferson, June 3, 1940.

[2] Measures by Marais, Ronan and Comer were made by universal stage immersion methods at the University of Wisconsin.

[a] *Mem. Soc. Phys. Hist. Nat. Genève*, 36, 367 (1910).

[4] Lorenzen: *Zeits. Kryst.*, 7, 610 and 9, 251.

In Table 4 the most accurate measures are those for N_g (and N_m in some cases). As made at the University of Wisconsin by Comer and by Marais using the double variation method with the five axis universal stage, these measures are believed to be accurate within ± 0.0005. Now the measures on a single sample (and even on a single tiny flake in some cases) made by the same person differ as much as several times this amount, reaching ten times as much in sample 19. Miss Glass reports a variation of .01 in sample 27, but the accuracy of her measures is not reported. If the measures of the same sample (that is, from the same source and in some cases from the same large U. S. National Museum sample) made by different persons be compared, the differences are still greater, being .005 in sample 19, .006 in sample 7, .0081 in samples 25, 26, .013 in sample 27, and .0279 in sample 17. It is therefore well established that a single sample varies considerably in optical properties. And it is also well established that such a variation is found in so many of the samples that it may be said to be the usual condition.

When it is remembered that the lowest value for N_g measured on any lithia mica is 1.554 while N_g in muscovite is 1.588, it is readily seen that 5 to 35 per cent of muscovite interleaved with lithia mica will not raise the index a great deal. In fact, 5 per cent would raise the index from 1.554 to 1.5557, 15 per cent would raise it to 1.5597, and even 35 per cent would raise it only to 1.5659. Irregularities in the distribution of the muscovite may be expected to be ordinarily no more than a minor fraction of the per cent present. Accordingly, the differences in index due to this cause should be commonly a minor fraction of the increases just noted, that is, less than half of: .0017 for 5 per cent, .0051 for 15 per cent, .0085 for 25 per cent, and .0119 for 35 per cent. The differences actually observed are of this order of magnitude. The fact that sample 19 with no calculated muscovite shows similar differences may be regarded as an exception, but this may be explained on the ground that the analysis does not represent the same material that was studied optically, just as the two analyses (25 and 26) of lithia mica from the same source (Wakefield, Quebec) are not exactly alike, but one requires the assumption of twice as much muscovite as the other.

As previously noted, zonal and other coarse intergrowths of muscovite and lepidolite have been described many times, and minute intergrowths were found in five samples by Schaller, but there is evidence which suggests that such intergrowths are much commoner than indicated by these reports. It is usually assumed that the two minerals can be distinguished by their colors, but this is often not true as shown by the fact that samples carefully selected for chemical analysis and study of the lithia micas have been shown in several cases to be muscovites, in some instances almost

completely devoid of lithia. The writer has had this experience and it is indicated in the writings of Stockwell and others. Pink and purple muscovites have been described by Schaller[13] and green muscovite by Stockwell. It seems probable that the same influences which cause the color of lithia mica may cause similar color in muscovite; if that be true, then muscovite intergrown with lepidolite is much more likely to be colored like it than is muscovite not closely associated with lithia mica. Again, Comer's measures of the optical properties of "lepidolite" from Mt. Apatite, Maine, indicate that he measured those of a flake of musco- vite, though the measures of Marais and also the analysis prove that the sample is largely lepidolite. If the theory of the writer as to the correct interpretation of the analyses of lithia micas be accepted, then the analyses themselves prove either an isomorphous system or an inter- growth of two minerals in nearly all cases.
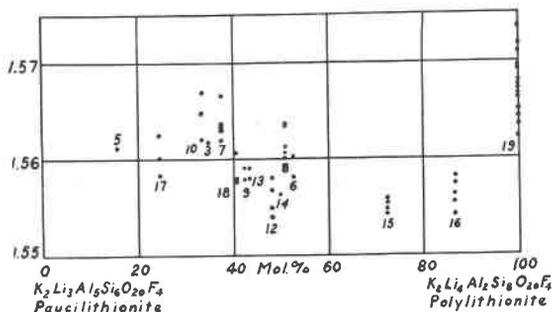
FIG. 1

It is desirable next to study the relations between variations in com- position and in optical properties. These can be seen much more clearly by dealing with the binary series, and we may begin with the paucilithio- nite-polylithionite series ($K_2Li_3Al_5Si_6O_{20}F_4$-$K_2Li_4Al_2Si_8O_{20}F_4$). The first nineteen analyses belong to this series, but samples 2 and 11 were not measured optically and sample 4 contains about 16 per cent of proto- lithionite and is therefore omitted. The others[14] are shown in Fig. 1 as calculated into these two molecules, disregarding the deficiencies in Li_2O, and also the protolithionite, which is less than 10 per cent in all cases and averages only 4.8. Samples 1, 3, and 5 appear at only one point each on the diagram, probably not because they do not vary, but because only one measure was made (or, at least, only one reported). The value of

[13] *Am. Mineral.*, **11**, 5 (1926).

[14] The value, 1.5863 for N_g measured by Comer on sample 17 is not included in the figure since it is considered to be evidence of nearly pure muscovite.

N_g for sample 5 was calculated from $N_m = 1.558$ and $2E = 63°$ (as reported), assuming $N_g - N_p = 0.025$, which is an approximation for lepidolite. It is obvious that this diagram leads to no satisfactory correlation. The only safe conclusions are that in ordinary lepidolite $N_g = 1.56 \pm 0.01$ and that the index varies very little from paucilithionite to polylithionite.
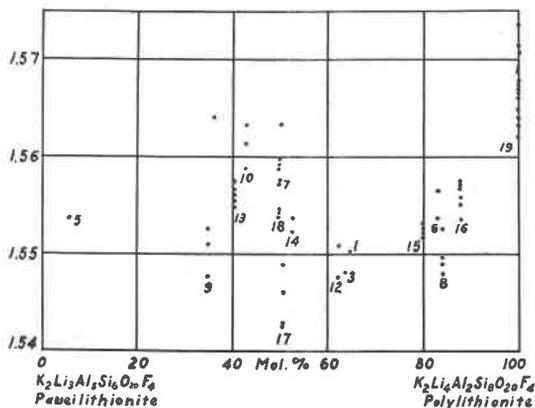

Fig. 2

However, study of the chemical analyses has led to the theory that lithia micas commonly contain variable amounts of muscovite in an intimate intergrowth with lepidolite proper. If sufficient muscovite be subtracted from each analysis to eliminate the deficiency of lithia and the remainder be calculated into the three lithia mica end-members, the percentages of these (for paucilithionite and polylithionite) are not the same as at first computed because these percentages depend upon the ratios of Al_2O_3 to SiO_2 and these ratios are changed by subtracting muscovite. It is obvious that there is no way to know whether the flake used to determine the optic properties contained the same proportion of muscovite as the material analyzed. If it be assumed that such was the case, it is possible to calculate the index of refraction (N_g) necessary in the lithia mica to give the measured index in the aggregate. The results of plotting the indices (N_g) thus derived against the computed percentage of polylithionite (disregarding protolithionite, as was done before) are shown in Fig. 2. It is evident that the correlation is even worse than in Fig. 1. This result suggests that (as might have been expected) the flake used to determine the index did not contain the same proportion of muscovite as the mass that was analyzed. In some large units it is known that lithia mica is surrounded by muscovite, or vice versa, when viewed in a cleavage piece. If this condition is common, any small flake used to measure

the index is probably much more nearly muscovite-free than is an aggregate used for chemical analysis. Accordingly, it is desirable to plot the percentage of polylithionite as corrected for muscovite against the indices (N_g) as measured. The results are shown in Fig. 3.
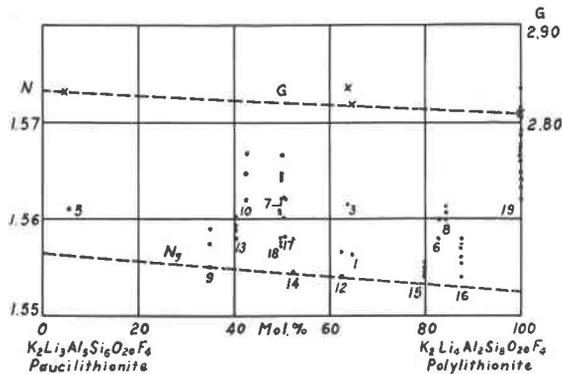

FIG. 3

It is clear that any mixture of the paucilithionite-polylithionite series ($N_g = 1.56+$) with muscovite ($N_g = 1.588$) will have an index higher than the muscovite-free mineral. Therefore, in any such mixture (presumably irregular) the fragment with the lowest index has the least admixed muscovite. It may be reasonable to assume that the lowest values in the diagram were obtained from fragments containing no muscovite. If so, N_g in all kinds of lepidolite containing no protolithionite is nearly equal to 1.555. It is perhaps significant that the five samples (9, 12, 14, 15, 16) which are almost on the minimum line have an average muscovite tenor of 9.6 while the remaining eleven samples have nearly twice as much (18.3), even though they include one (19) the analysis of which indicates that it contains no muscovite at all. Furthermore, samples 15 and 16 are reasonably slightly above the line because they contain a little protolithionite.

Many measures were made on samples available to determine the amount of variation in optical properties, since that may serve as a measure of their variability in composition. The results as shown in Table 4 prove that these lithia micas vary in properties very much more than is known in most minerals, and, in all cases measured, the variation is toward muscovite. In one case (No. 17) one measure gave results which indicate pure muscovite. Sample 19—the polylithionite from Greenland— varies in index (N_g) more than any other sample (disregarding the approximate figures of Glass on sample No. 27) except for one measure on
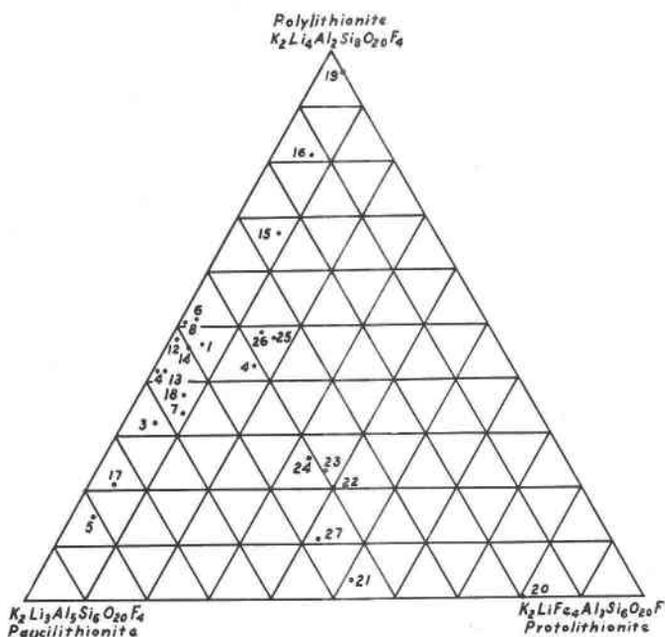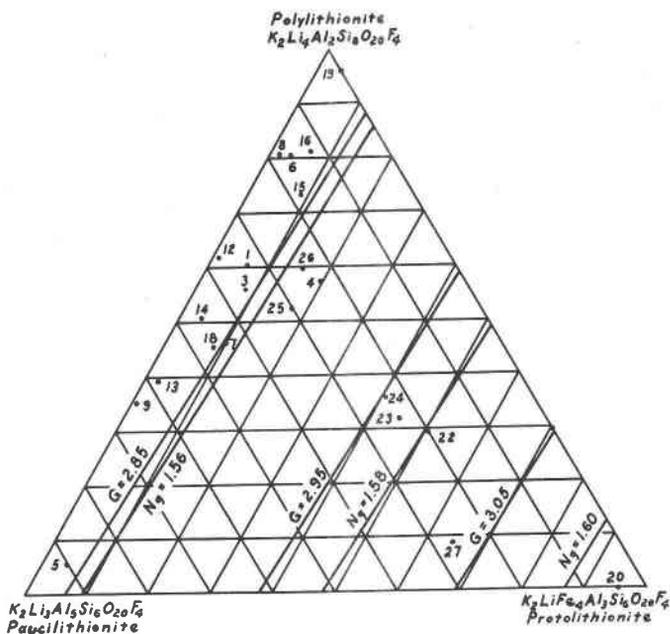
Fig. 4

Fig. 5

sample 17, just cited. Therefore the sample analyzed from Greenland seems to have been unlike the sample from the same place which is available for measures of the optical properties.

Before studying another binary series it is desirable to get a picture of the ternary system: paucilithionite-polylithionite-protolithionite. If the analyses are calculated into these three formulas, disregarding deficiencies in lithia, and plotted on a triangle, the results are shown in Fig. 4. But it
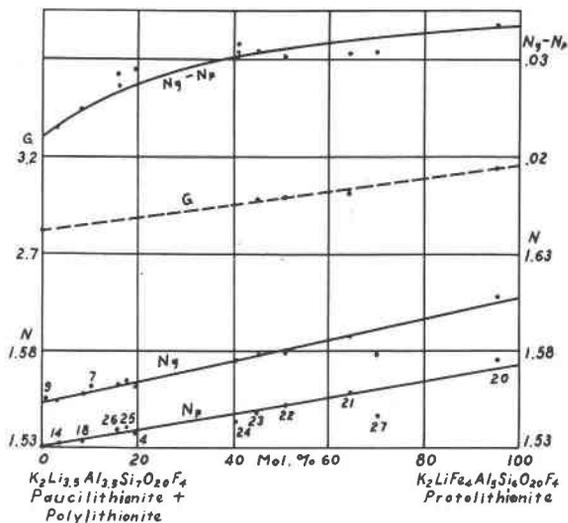

FIG. 6

seems probable that muscovite is present in most of the samples analyzed; if the analyses are recalculated, after eliminating the indicated muscovite, the diagram resulting is shown in Fig. 5. Since the variations in properties in the paucilithionite-polylithionite series are slight, they may be disregarded, and all samples with an appreciable tenor of protolithionite may be considered to belong to a series from 50 paucilithionite–50 polylithionite to pure protolithionite as shown in Fig. 6. From these two binary series the properties of the ternary system may be approximated by extrapolation, as shown by the lines in Fig. 5. Unfortunately, these lines indicate the properties in muscovite-free lithia micas, and lithia micas usually contain admixed muscovite, according to the theory proposed in this paper. The writer is unable to present a diagram which includes this molecule; it would seem to require the use of a tetrahedron.