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NEW ANALYSES OF LEPIDOLITES AND THEIR INTERPRETATION*

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ABSTRACT

Seventeen new analyses of lepidolites are presented. These analyses seem to be in agreement with the structural requirements of micas, as revealed by the *x*-ray studies of other investigators. The analyses indicate that the rare alkalis, rubidium and cesium, are normal constituents of lepidolites.

The interpretation of a new analysis of polyolithionite, an end member of the lepidolite series, shows it to approximate closely the formula $\text{K} \cdot \text{Li}_2\text{Al} \cdot \text{Si}_4\text{O}_{10}\text{F}_2$. Columbium and titanium seem to be normal constituents of lepidolites near polyolithionite in composition.

The composition of the seventeen lepidolites closely approximates that of isomorphous mixtures of polyolithionite [$\text{K} \cdot \text{Li}_2\text{Al} \cdot \text{Si}_4\text{O}_{10}\text{F}_2$] with biotite [$\text{K} \cdot \text{R}'_2 \cdot \text{AlSi}_3\text{O}_{10}(\text{OH}, \text{F})_2$],

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lithium muscovite $[K_4 \cdot Li_6Al_6 \cdot Al_4Si_{12}O_{40}(OH, F)_8]$, and muscovite $[K \cdot Al_2 \cdot AlSi_3O_{10}(OH, F)_2]$. The selection of biotite as an end member is arbitrary and other end members may be taken to account for the bivalent ions present.

Evidence is presented indicating discontinuous isomorphism. Theoretical consideration of this evidence suggests the following formulas for lepidolites: ideal formula A, $K \cdot Li_2Al \cdot Si_4O_{10}F_2$, the formula of polyolithionite; ideal formula B, $K_4 \cdot Li_6Al_6 \cdot AlSi_{18}O_{40}(F, OH)_8$; ideal formula C, $K_4 \cdot Li_7Al_6 \cdot Al_2Si_{14}O_{40}(F, OH)_8$; ideal formula D, $K_4 \cdot Li_6Al_6 \cdot Al_3Si_{13}O_{40}(OH, F)_8$; and ideal formula E, $K_4 \cdot Li_6Al_6 \cdot Al_4Si_{14}O_{40}(OH, F)_8$. The analyses may be divided into the types, A, B, C, D, and E with limits of composition expressed by the corresponding ideal formulas. One of the samples analyzed is close to polyolithionite (type A) in composition; eight samples are of type C, and five of the samples are of type D. Three samples do not correspond to any of the above types because of their bivalent ion content.

THE MICA STRUCTURE

The modern concept of the mica structure began with Mauguin's¹ measurements of the dimensions of mica unit cells with x -rays. Knowing the composition and density of a mica and the weights of the atoms, he was able to calculate the number of atoms of each element in the unit cell. Mauguin found that the number of atoms of oxygen plus fluorine in the mica unit cell is forty-eight, within the errors of analysis, and upon this value of the anion content of the unit cell are based modern mica formulas.

The concept of the mica structure is due to Pauling,² who showed that micas contain SiO_4 groups in sheets of indefinite extent. In these groups silicon has a coordination number of four, i.e., each silicon atom is surrounded by four oxygen atoms at the corners of a tetrahedron and the group is therefore referred to as the tetrahedral group. The silicon-oxygen sheet has the composition Si_4O_{10} , silicon atoms being linked through oxygen to form a hexagonal network. In the common micas a fourth of the silicon is replaced by aluminum so that the silicon-oxygen sheet has the composition $AlSi_3O_{10}$.

Two tetrahedral silicon sheets are held together firmly by cations in six-fold coordination, each cation being surrounded by six anions at the corners of an octahedron. Such a group is called an octahedral group. The octahedral groups may be built around aluminum, magnesium, titanium, manganese, iron, and lithium. Thus two silicon layers are joined through octahedral groups to form a double silicon layer. Many of the oxygen atoms are included in two groups.

These double silicon layers in turn are joined to other similar layers by large cations in twelve-fold coordination, consisting of potassium,

¹ Mauguin, C., Orientation of liquid crystals by mica sheets: *Compt. Rend.*, vol. 156, p. 1246, 1913. Also *Compt. Rend.*, vol. 186, pp. 879, 1131, 1928.

² Pauling, Linus, Structure of the micas and related minerals: *Nat. Acad. Sci. Proc.*, vol. 16, p. 123, 1930.

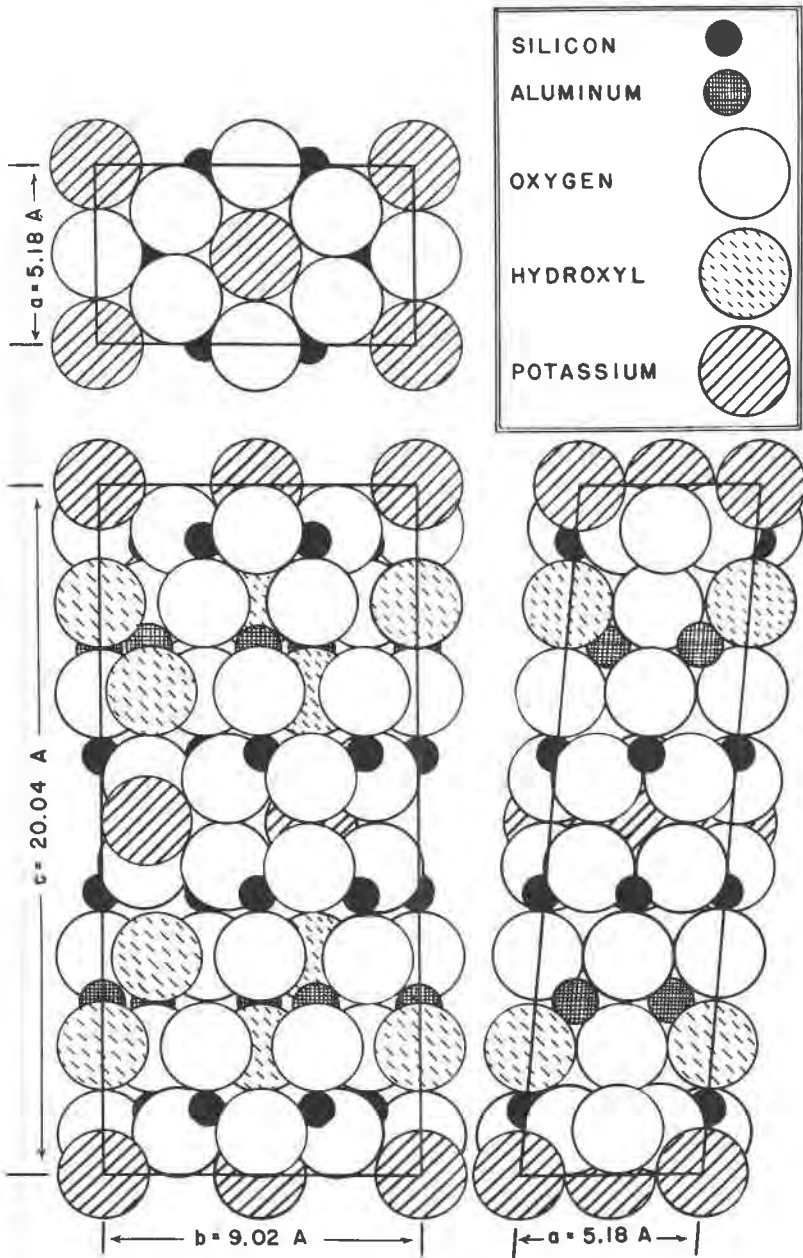


FIG. 1. Unit cell of muscovite.

replaceable to a limited extent by sodium, rubidium, and cesium. In the brittle micas calcium takes the place of potassium.

A mica thus has a layered arrangement, the sequence of layers being: tetrahedral, octahedral, tetrahedral, large cation, tetrahedral, octahedral, tetrahedral, large cation, etc.

Figure 1 shows the unit cell of muscovite. The layered arrangement can be seen; cleavage occurs along the planes of potassium atoms. In figure 2, one of the octahedral layers is shown in detail, the other octahedral layer being a mirror image of the one pictured. The mica unit cell thus can be seen to have twelve octahedral positions, only eight of which are occupied in muscovite.

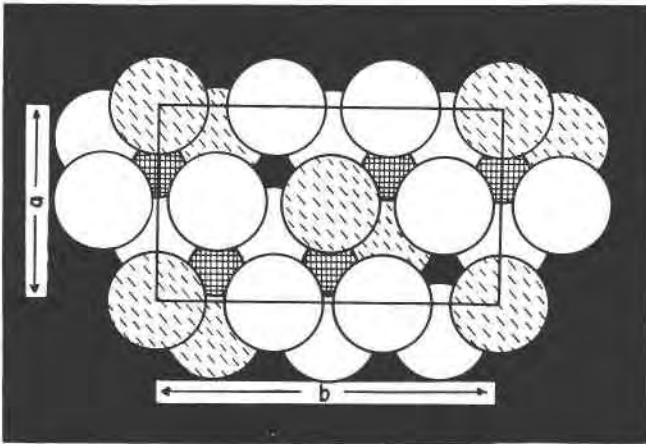


FIG. 2. Octahedral group in muscovite.

UNIT CELL CONTENT AND FORMULAS OF MICAS

X-ray data show that a mica unit cell has four silicon layers (or two double layers) and contains: 4 positions for large cations occupied by atoms of potassium, sodium, rubidium, and cesium; 8-12 octahedral positions for atoms holding the silicon layers together, occupied by aluminum, magnesium, manganese, iron, titanium, and lithium; 16 tetrahedral positions for silicon (and aluminum); 40 oxygen atoms; and 8 hydroxyls or fluorines.

The unit cell content of muscovite, with the groups in the above order, is $K_4 \cdot Al_8 \cdot Al_4Si_{12}O_{40}(OH)_8$, which simplifies to the established formula $K \cdot Al_2 \cdot AlSi_3O_{10}(OH)_2$. Similarly the unit cell content of phlogopite is $K_4 \cdot Mg_{12} \cdot Al_4Si_{12}O_{40}(F, OH)_8$, and the formula simplifies to $K \cdot Mg_3 \cdot AlSi_3O_{10}(F, OH)_2$.

Although the composition of common micas is given approximately by formulas representing one-fourth of the unit cell, the composition of lepidolite, apparently, cannot be so simply represented. Bragg³ says "The constitution of lepidolite diverges even more widely from that represented by a simple formula. It appears highly probable that Li is in six-coordination in the second of the groups. The formula $K \cdot Li_2 Al(Si_4O_{10})(OH, F)_2$ [this is the formula of polyolithionite. R.E.S.] is approached, but it implies too high a lithium content. We must assume a substitution of part of the Si by Al, balanced by a reduction of the lithium in six-coordination."

In the present paper are presented seventeen new analyses of lepidolites, from different localities, made with the more recent analytical methods.

The interpretation of these analyses is based on the mica structure outlined above. Two interpretations will be presented: in the first, the composition is regarded as representing isomorphous mixtures of end members with simple mica formulas; in the second, some evidence will be presented which seems to show that the series is discontinuous, resulting in several types of lepidolite with limits of composition given in ideal formulas representing the entire unit cell.

SAMPLES AND THEIR PREPARATION FOR ANALYSIS

The lepidolite samples were furnished by Dr. Schaller, of the U. S. Geological Survey, who examined them optically to be sure that no impurities were present which could not be readily removed. Five samples were discarded because the small flakes of lepidolite were found to contain numerous minute inclusions of muscovite. Samples discarded for this reason were: two from San Diego County, California; one from Auburn, Maine; one from Tin Mountain, South Dakota; and one from the Harding mine, New Mexico. This occurrence of minute inclusions of muscovite in lepidolite is apparently not unusual.

The quartz, feldspar, and other commonly associated minerals were separated from the lepidolite in the following way: small pieces of the lepidolite and associated minerals were crushed on a steel plate by means of a steel roller, crushing to a fine powder all the minerals except the more resistant lepidolite. The sample was then put in a sieve and the powder passing through discarded. This treatment was repeated until the lepidolite was freed of the associated minerals. The mica was washed several times with distilled water, decanting and discarding the fine

³ Bragg, W. L., *Atomic Structure of Minerals*, p. 125, Cornell University Press, *Ithaca*, N. Y., 1937.

material that did not immediately settle to the bottom. The lepidolite was then examined optically by Dr. Schaller and when any impurities were found the treatment was repeated. The samples were then ground to pass an 80 mesh sieve.

The lepidolites analyzed, in order of increasing content of lithia, were as follows:

1. Large spherulitic plates, gray purple, Manitoba, Canada, U.S.N. Mus. 97635.
2. Coarse-medium grained, purple, Katerina mine, Pala, San Diego County, Calif.
3. Fine, compact, purple, Stewart mine, "40 acres," Pala, Calif.
4. Medium grained, purple, Panama-Pacific-Exposition mine, Chihuahua Valley, east of Oak Grove, San Diego County, Calif.
5. Medium grained, pink, west lens, Stewart mine, Pala, Calif.
6. Coarse, pale blue, silky or talc-like, Stewart mine, Pala, Calif.
7. Large plates, pale purple, Ohio City, Colo., U.S.N. Mus. 97893.
8. Coarse, purple, iron stained, Himalaya mine, Mesa Grande, San Diego County, Calif.
9. Coarse, purple, San Diego mine, Mesa Grande, Calif.
10. Large plates, gray purple, Wakefield, Canada, U.S.N. Mus. 96012.
11. Fine, almost white, Stewart mine, Pala, Calif.
12. Coarse, pale blue, Stewart mine, Pala, Calif.
13. Very coarse, purple, Himalaya mine, Mesa Grande, Calif.
14. Large transparent sheets, pale lavender, optically uniaxial, Calgoorie, W. Australia, U.S.N. Mus. R-4365.
15. Terminated crystals, pink, Little Three mine, Ramona, San Diego County, Calif., U.S.N. Mus. 93924.
16. Large plates, pale blue, Antsongombato, Madagascar, collected by Lacroix.
17. Polyolithionite, large plates, greenish, Greenland, U.S.N. Mus. 94314.

In addition, the alkalies alone were determined on a sample of purple muscovite from the Harding mine, New Mexico, described by Schaller and Henderson.⁴ The percentages of alkalies found were: Li_2O 0.38; Na_2O 0.91; K_2O 10.07; Rb_2O 0.82; Cs_2O 0.09.

METHODS OF ANALYSIS

A number of methods not commonly used in rock analysis had to be employed in analyzing the lepidolites. Rubidium was found in all of the samples and cesium in all but one; their determination was made by methods described by Wells and Stevens.⁵ Lithium was determined by the Palkin⁶ method.

Silica and fluorine were separated by the Berzelius method as modified

⁴ Schaller, W. T., and Henderson, E. P., Purple muscovite from New Mexico: *Am. Mineral.*, vol. 11, pp. 5-16, 1926.

⁵ Wells, R. C., and Stevens, R. E., Determination of the common and rare alkalies in mineral analysis: *Ind. Eng. Chem., Anal. Ed.*, vol. 6, pp. 439-442, 1934.

⁶ Palkin, S., Separation of lithium from the other alkali metals: *Am. Chem. Soc. Jour.*, vol. 38, p. 2326, 1916.

by Hoffman and Lundell,⁷ except that neutralization was made with hydrochloric acid instead of nitric. The silica was then obtained by acid dehydration in the usual way. Fluorine was determined nephelometrically.⁸

The determination of silica in polyolithionite (sample 17) gave with the silica a residue, non-volatile with HF, of nearly 12 milligrams from a half gram sample, as if decomposition of the mineral had not been complete. However, a second determination yielded an equally high residue and the figures for fluorine and silica checked well with the previous results (the figures for fluorine were 7.76% and 7.69%, for silica 59.52% and 59.59%). The residues were then examined spectrographically by George Steiger, of the Geological Survey, and found to consist essentially of columbium oxide. The columbium oxide was determined on a separate sample by fusion with sodium carbonate, evaporating to dryness with hydrochloric acid, filtering, and igniting the separated silica and columbium oxide. The silica was then removed with hydrofluoric acid. The columbium oxide was further purified by fusion with sodium carbonate, leaching the cake with water, adding to the extract hydrochloric acid to 5% strength, and digesting on the steambath to coagulate the columbium oxide and to dissolve the aluminum hydroxide. The columbium oxide was then filtered, ignited, and weighed. It was examined spectrographically by Mr. Steiger and found to be free of tantalum, which commonly accompanies columbium in other minerals.

In most of the lepidolites iron was present in such small quantity as to make its determination by reduction and titration with permanganate solution impracticable, and on all samples except 1, 10 and 17 the iron was determined by the thiocyanate colorimetric method. In the samples containing an appreciable quantity of iron it was found for the most part to be in the ferrous condition, so where total iron only was determined it is reported as ferrous oxide.

Manganese was determined colorimetrically after oxidation to permanganate. The manganese was found not to oxidize ferrous sulphate solutions; therefore most of it is present in the bivalent state and is reported as MnO.

For the determination of total water a half gram of the mineral was mixed with 1.5 grams of sodium tungstate and the water determined by the Penfield glass tube method. The mineral and flux were heated strongly with two blast lamps directed at opposite sides of the tube. An

⁷ Hoffman, J. I., and Lundell, G. E. F., Determination of fluorine and of silica in glasses and enamels containing fluorine: *Bur. Standards Jour. Research*, vol. 3, p. 581, 1929.

⁸ Stevens, R. E., Nephelometric determination of fluorine: *Ind. Eng. Chem., Anal. Ed.*, vol. 8, p. 248, 1936.

asbestos shield was used to prevent heating of the condenser end of the tube, which was covered with wet paper. By turning the tube over and over collapse of the glass tube was prevented until the charge had been heated sufficiently long for the volatilization of all water.

ANALYTICAL RESULTS

In Table 1 the results of the analyses are listed from left to right in order of increasing content of lithia. Table 3 gives corresponding relative equivalents and Table 4 atomic ratios. A gradation from a composition approaching that of muscovite (No. 1) to that of polyolithionite (No. 17) is indicated.

An increase in lithia content generally accompanies an increase in silica and a decrease in alumina, but there are some exceptions (note samples 6, 8, 13, 14), and other factors must be considered in explaining the composition of all lepidolites. In other words, lepidolites are not simply a solid solution of muscovite, or a hypothetical fluorine muscovite, with polyolithionite. With increase in lithia the fluorine usually increases, and with increase in fluorine the combined water decreases. The uncombined water (H_2O-) is taken as that given off below $110^\circ C.$, but the removal of all uncombined water at that temperature is doubtful. If the total loss in weight up to $160^\circ C.$ is taken as uncombined water, the combined water and fluorine approach closer the requirements of a mica.

Titanium and columbium were found to be constituents of those lepidolites which approach polyolithionite in composition (No. 17 from Greenland and No. 16 from Madagascar). As the micas had been found to be pure by optical examination the columbium is apparently a constituent of the mica.

Rubidium oxide was present in all samples, reaching a maximum of 2 per cent in sample 13 from Mesa Grande, California. Cesium oxide in smaller quantity, usually less than 0.5 per cent, was found in all the lepidolites except the polyolithionite from Greenland. These figures indicate that these rare alkalies are normal constituents of lepidolites, and the figures for K_2O given in most previous analyses may be too high.

In none of the lepidolites was more than a trace of calcium found. Calcium was precipitated as the oxalate and the precipitate would include any strontium in the mica.

Hahn, Strassmann, and Walling⁹ report the finding of strontium 87, radioactive disintegration product of rubidium, in a lepidolite from Manitoba, Canada. They found 2-3 per cent of rubidium and .0148 per

⁹ Hahn, Otto, Strassmann, Fritz, and Walling, Ernst, Herstellung wägbarer Mengen Strontiumisotops 87 als Umwandlungsprodukt des Rubidiums aus einem kanadischen Glimmer: *Naturwissenschaften*, vol. 25, p. 189, 1937.

TABLE 1

Sample No.	1	2	3	4	5	6	7	8	9
SiO ₂	47.00	49.50	48.58	50.20	49.29	53.45	49.58	54.69	51.70
Al ₂ O ₃	30.60	28.06	28.93	28.18	28.40	22.15	23.87	22.83	23.97
Cb ₂ O ₅	—	—	—	—	—	—	—	—	—
TiO ₂	Trace	Trace	Trace	Trace	Trace	Trace	.06	Trace	Trace
Fe ₂ O ₃	.26	—	—	—	—	—	—	—	—
FeO	.41	.11 ^a	.04 ^a	.04 ^a	.05 ^a	.16 ^a	.21 ^a	.11 ^a	.04 ^a
MgO	.13	None	None	None	.12	.14	None	None	None
MnO	2.04	.54	.92	.28	.65	.52	2.78	.14	.17
Li ₂ O	2.70	3.51	3.70 ^b	3.81	3.96	5.04	5.05	5.11	5.33
CaO	Trace	None	Trace	Trace	Trace	None	None	None	Trace
Na ₂ O	.77	1.27	.87	.64	.77	.74	.57	.57	.89
K ₂ O	9.52	10.32	10.02	9.91	9.93	9.58	10.14	9.53	10.79
Rb ₂ O	1.93	1.11	.91	1.55	1.56	1.62	1.62	1.64	.42
Cs ₂ O	.18	.13	.16	.11	.12	.48	.09	.17	.41
H ₂ O—	.25	.27	.54	.18	.14	.46	.51	.31	.26
H ₂ O+	2.18	2.15	2.56	2.18	1.76	1.28	1.22	1.02	1.24
F	4.09	5.98	4.93	4.97	6.52	7.22	7.49	6.86	7.76
O = F	102.06 1.72	102.95 2.52	102.16 2.08	102.05 2.09	103.27 2.75	102.78 3.04	103.19 3.15	102.98 2.89	102.98 3.27
H ₂ O 160°C.	100.34 .35	100.43 .38	100.08 .73	99.96 .27	100.52 .22	99.74 .65	100.04 .81	100.09 .50	99.71 .37

Sample No.	10*	11	12	13	14*	15	16	17
SiO ₂	50.31	51.10	52.58	51.25	51.07	54.40	57.03	59.56
Al ₂ O ₃	19.95	23.98	22.82	23.71	22.05	17.95	15.55	12.04
Cb ₂ O ₅	—	—	—	—	—	—	.14	1.52
TiO ₂	.22	Trace	Trace	.01	.06	.02	.03	.48
Fe ₂ O ₃	.49	—	—	—	—	—	—	.13
FeO	2.55	Trace	.01 ^a	.07 ^a	.23 ^a	.21 ^a	.12 ^a	.42
MgO	.02	None	None	.08	.09	.30	.22	.34
MnO	2.63	.13	.28	.61	.76	2.06	.72	.03
Li ₂ O	5.39	5.51	5.64	5.78	5.89	6.18	6.84	7.26
CaO	Trace	Trace	Trace	Trace	Trace	Trace	Trace	Trace
Na ₂ O	.59	.63	.59	.65	.82	.72	.44	.53
K ₂ O	10.14	10.25	10.11	9.90	9.70	10.28	10.65	11.05
Rb ₂ O	.97	1.38	1.04	2.00	1.38	1.22	1.35	1.14
Cs ₂ O	.06	.48	.67	.08	.09	.24	.40	None
H ₂ O—	.66	.56	.30	.34	.96	.81	.74	.73
H ₂ O+	.88	1.50	1.35	.90	—	.58	.49	.47
F	7.65	7.36	7.56	8.08	7.13	9.19	9.00	7.73
O = F	102.51 3.22	102.88 3.10	102.95 3.18	103.46 3.40	100.23 3.00	104.16 3.87	103.72 3.79	103.43 3.26
H ₂ O 106°C.	99.29 .98	99.78 .88	99.77 .46	100.06 .45	97.23 1.74	100.29 1.18	99.93 .93	100.17 .76

^a Total iron reported as FeO.

^b Results of other analysts suggest that this figure may be too high.

* Contaminated by organic matter during grinding.

cent of strontium. The age of this mineral is supposed to be about 2,000 million years. Most of the lepidolites in the present paper are probably a good deal younger and would contain less of strontium 87. Such small quantities of strontium could not be accurately determined without the use of very large samples.

EQUIVALENTS AND ATOMS IN THE MICA UNIT CELL

The structure of mica calls for the following requirements:

1. *Number of atoms in the unit cell:* (a) 40 oxygen atoms; (b) 8 positions for fluorine and hydroxyl; (c) 4 positions for potassium atoms (including sodium, rubidium, and cesium); (d) 8–12 octahedral positions for atoms of aluminum, iron, magnesium, manganese, titanium, and lithium; (e) 16 tetrahedral positions for silicon and aluminum.

The total number of atoms in the unit cell, therefore, varies from 76 to 80, counting the hydroxyl radical (OH) as one atom for simplicity. As the number of atoms in a unit cell is variable the total atoms cannot be used as a basis for calculating formulas. The number of tetrahedral cations (Si and Al) in the unit cell is fixed at 16.¹⁰

2. *Number of equivalents in the unit cell:* The unit cell must be neutral. This means that anion equivalents (the atomic weight divided by the valence of the atom) must be balanced by an equal number of cation equivalents. Anion equivalents in a unit cell of a mica are 88 (40 bivalent oxygen atoms and 8 univalent fluorines or hydroxyls). In the unit cell of muscovite $K_4 \cdot Al_8 \cdot Al_4Si_{12}O_{40}(OH, F)_8$, the 88 anion equivalents are balanced by 4 equivalents of potassium, 36 equivalents of aluminum, and 48 equivalents of silicon, making a total of 88 cation equivalents. Presumably the number of cation equivalents in a mica unit cell is an invariable quantity, always 88.

As the large cation in mica is filled by 4 equivalents of potassium there remain 84 equivalents to be distributed between the octahedral and tetrahedral groups. In the common micas which contain tetrahedral $Al_4Si_{12}O_{40}$, the tetrahedral group comprises 60 equivalents, leaving 24 equivalents for the octahedral group. In taeniolite,¹¹ however, and in polyolithionite, whose formula is derived in the section to follow, all 16 tetrahedral positions are filled by silicon, so that the tetrahedral group contains 64 equivalents which leaves only 20 equivalents for the octahedral group.

¹⁰ Bragg uses 4 tetrahedral atoms in $\frac{1}{4}$ unit cell. See Bragg, W. L., *Atomic Structure of Minerals*, p. 215, Cornell University Press, *Ithaca, N. Y.*, 1937.

¹¹ Miser, H. D., and Stevens, R. E., Taeniolite from Magnet Cove, Arkansas: *Am. Mineral.*, vol. 23, p. 104, 1938.

Relative Equivalents in Lepidolites

The assignment of 84 equivalents to be distributed between the tetrahedral and octahedral groups is used in this paper as the basis for calculating the formulas and for comparing the compositions of the lepidolites.

TABLE 2. RELATIVE EQUIVALENTS (octahedral+tetrahedral=84)

		Eq. Wt.	1	2	3	4	5	6	7	8
octahedral and tetrahedral	SiO ₂	15.08	50.57	53.20	52.10	53.27	52.53	57.18	54.00	57.30
	Al ₂ O ₃	17.03	29.12	26.70	27.46	26.50	26.79	20.97	23.01	21.19
	Cb ₂ O ₅	26.58	—	—	—	—	—	—	—	—
	TiO ₂	19.98	—	—	—	—	—	—	.05	—
	Fe ₂ O ₃	26.61	.16	—	—	—	—	—	—	—
	FeO	35.92	.18	.05	.02	.02	.03	.06	.10	.05
	MgO	20.16	.10	—	—	—	.10	.11	—	—
	MnO	35.45	.94	.24	.42	.13	.29	.24	1.28	.06
	Li ₂ O	14.95	2.93	3.81	4.00	4.08	4.26	5.44	5.56	5.40
		84.00	84.00	84.00	84.00	84.00	84.00	84.00	84.00	84.00
large cation	Na ₂ O	31.00	.41	.66	.45	.34	.40	.39	.30	.28
	K ₂ O	47.10	3.27	3.55	3.45	3.36	3.39	3.29	3.53	3.19
	Rb ₂ O	93.48	.34	.19	.16	.27	.27	.27	.28	.29
	Cs ₂ O	140.86	.02	.02	.02	.02	.02	.05	.02	.02
			4.04	4.42	4.08	3.99	4.08	4.00	4.13	3.78
F+OH	H ₂ O+	9.00	3.92	3.87	4.61	3.87	3.15	2.29	2.24	1.79
	F	19.00	3.48	5.11	4.19	4.19	5.51	6.13	6.48	5.70
			7.40	8.98	8.80	8.06	8.66	8.42	8.72	7.49

		9	10	11	12	13	14	15	16	17
octahedral and tetrahedral	SiO ₂	55.41	55.58	55.08	56.18	54.88	55.68	58.78	61.18	63.12
	Al ₂ O ₃	22.72	19.50	22.88	21.61	22.50	21.28	17.18	14.76	11.27
	Cb ₂ O ₅	—	—	—	—	—	—	—	.08	.91
	TiO ₂	—	.18	—	—	.02	.05	.02	.03	.38
	Fe ₂ O ₃	—	.30	—	—	—	—	—	—	.08
	FeO	.02	1.18	—	—	.03	.10	.10	.05	.19
	MgO	—	.02	—	—	.06	.07	.24	.18	.27
	MnO	.08	1.23	.06	.13	.27	.34	.95	.32	.02
	Li ₂ O	5.77	6.01	5.98	6.08	6.24	6.48	6.73	7.40	7.76
	84.00	84.00	84.00	84.00	84.00	84.00	84.00	84.00	84.00	84.00
large cation	Na ₂ O	.47	.32	.33	.31	.34	.44	.37	.23	.27
	K ₂ O	3.70	3.58	3.54	3.47	3.39	3.39	3.55	3.66	3.76
	Rb ₂ O	.08	.17	.24	.18	.34	.25	.21	.23	.19
	Cs ₂ O	.05	.01	.05	.08	.02	.02	.03	.05	—
		4.30	4.08	4.16	4.04	4.09	4.10	4.16	4.17	4.22
F+OH	H ₂ O+	2.23	1.63	2.71	2.42	1.61	—	1.04	.87	.83
	F	6.60	6.71	6.29	6.42	6.86	—	7.87	7.67	6.51
		8.83	8.34	9.00	8.84	8.47	—	8.91	8.54	7.34

TABLE 3. ATOMIC RATIOS IN LEPIDOLITES

		1	2	3	4	5	6	7	8	9
tetrahedral	Si	12.64	13.30	13.03	13.32	13.13	14.30	13.51	14.33	13.85
	Al	3.36	2.70	2.97	2.68	2.87	1.70	2.49	1.67	2.15
octahedral	Al	16.00	16.00	16.00	16.00	16.00	16.00	16.00	16.00	16.00
	Cb	6.35	6.20	6.18	6.15	6.06	5.29	5.19	5.39	5.42
	Ti	—	—	—	—	—	—	—	—	—
	Fe ⁺⁺⁺	—	—	—	—	—	—	.01	—	—
	Fe ⁺⁺	.05	—	—	—	—	—	—	—	—
	Mg	.09	.03	.01	.01	.02	.03	.05	.03	.01
	Mn	.05	—	—	—	.05	.06	—	—	—
	Li	.47	.12	.21	.07	.15	.12	.64	.03	.04
	Li	2.93	3.81	4.00	4.08	4.26	5.44	5.56	5.40	5.77
large cation		9.94	10.16	10.40	10.31	10.54	10.94	11.45	10.85	11.24
	Na	.41	.66	.45	.34	.40	.39	.30	.28	.47
	K	3.27	3.55	3.45	3.36	3.39	3.29	3.53	3.19	3.70
	Rb	.34	.19	.16	.27	.27	.27	.28	.29	.08
	Cs	.02	.02	.02	.02	.02	.05	.02	.02	.05
		4.04	4.42	4.08	3.99	4.08	4.00	4.13	3.78	4.30
	H	3.92	3.87	4.61	3.87	3.15	2.29	2.24	1.79	2.23
	F	3.48	5.11	4.19	4.19	5.51	6.13	6.48	5.70	6.60
	O	44.24	43.59	44.25	43.84	42.86	42.08	41.95	41.94	41.97
	O+F	47.72	48.70	48.44	48.03	48.37	48.21	48.43	47.64	48.57

		10	11	12	13	14	15	16	17
tetrahedral	Si	13.90	13.77	14.05	13.72	13.92	14.70	15.30	15.78
	Al	2.10	2.23	1.95	2.28	2.08	1.30	.70	.22
octahedral	Al	16.00	16.00	16.00	16.00	16.00	16.00	16.00	16.00
	Cb	4.40	5.40	5.25	5.22	5.01	4.43	4.22	3.54
	Ti	—	—	—	—	—	—	.02	.18
	Fe ⁺⁺⁺	.04	—	—	—	.01	—	.01	.10
	Fe ⁺⁺	.10	—	—	—	—	—	—	.03
	Mg	.59	—	—	.02	.05	.05	.02	.10
	Mn	.01	—	—	.03	.04	.12	.09	.14
	Li	.62	.03	.07	.14	.17	.48	.16	.01
	Li	6.01	5.98	6.08	6.24	6.48	6.73	7.40	7.76
large cation		11.77	11.41	11.40	11.65	11.76	11.81	11.92	11.86
	Na	.32	.33	.31	.34	.44	.37	.23	.27
	K	3.58	3.54	3.47	3.39	3.39	3.55	3.66	3.76
	Rb	.17	.24	.18	.34	.25	.21	.23	.19
	Cs	.01	.05	.08	.02	.02	.03	.05	—
		4.08	4.16	4.04	4.09	4.10	4.16	4.17	4.22
	H	1.63	2.71	2.42	1.61	—	1.04	.87	.83
	F	6.71	6.29	6.42	6.86	—	7.87	7.67	6.51
	O	41.50	42.29	42.02	41.42	—	40.67	40.69	41.27
	O+F	48.21	48.58	48.44	48.28	—	48.54	48.36	47.78

In Table 2 the relative equivalents are given, on the basis of 84 tetrahedral and octahedral equivalents, and in Table 3 the corresponding *atomic ratios* are listed. Correlation of the analyses on this basis allows a precise evaluation of octahedral groups, on which the interpretation largely depends. Satisfactory agreement with the mica requirements may be seen in Tables 2 and 3.

FORMULA OF POLYLITHIONITE

Polyolithionite was first described by Lorenzen,¹² whose analysis gives the alkali content as follows: Li_2O 9.04, Na_2O 7.63, and K_2O 5.37 per cent. So far as is known sample 17 in this paper is the same as Lorenzen's sample. Flink¹³ described a mica from Narsarsuk, Greenland, similar to polyolithionite, and his figures for alkalies are Li_2O 8.24, Na_2O 1.61, K_2O 11.05 per cent. Neither of these analyses list rubidium, cesium, or columbium.

Detailed study of the mica unit cell shows that there are only twelve octahedral positions (Fig. 2), and apparently lithium occupies octahedral positions in the mica. Sufficient octahedral positions are not available to account for the high lithium content reported for polyolithionite by Lorenzen and by Flink. Lorenzen's figures for Na_2O and K_2O also seem questionable. These two analyses do not give the same formula for polyolithionite as that derived from sample 17 in the present paper.

The scheme described in the previous section was used in calculating the formula of polyolithionite from the analysis of sample 17. This calculation is shown in Table 4. The inclusion of columbium in the octahedral group is logical though not as yet confirmed by *x*-ray data.

The unit cell content is closely represented by $\text{K}_4 \cdot \text{Li}_3\text{Al} \cdot \text{Si}_{16}\text{O}_{40}\text{F}_8$, and the formula simplifies to $\text{K} \cdot \text{Li}_2\text{Al} \cdot \text{Si}_4\text{O}_{10}\text{F}_2$. The analysis shows minor substitution of aluminum for silicon and of columbium, titanium, iron, magnesium and manganese for octahedral aluminum and lithium. A slight deficiency of fluorine and hydroxyl for the required F_8 may indicate a substitution of oxygen in this anion group.

A brief study of mica unit cells will suffice to show that polyolithionite has the maximum ratio of lithium in a mica, as the octahedral group is limited to 12 atoms and valency must be balanced, unless it is possible to substitute lithium in another group. For a pure completely fluorinated polyolithionite the percentage of Li_2O is 7.65 and with substitution of hydroxyl for fluorine a slightly higher figure (7.73%) may be obtained.

¹² Lorenzen's analyses are given under zinnwaldite on p. 627 of Dana's System of Mineralogy.

¹³ Flink, Gustav, On the minerals from Narsarsuk on the Firth of Tunugdliarfik in southern Greenland: *Meddelelser om Grönland*, vol. 24, pp. 110-115, 1901.

TABLE 4. CALCULATION OF FORMULA OF POLYLITHIONITE

	Per cent	Eq. Wt.	Equivalent Ratios		Atomic Ratios	
SiO ₂	59.56	15.08	3.949	63.12	15.78	Si ₁₆
Al ₂ O ₃	12.04	17.03	.706	11.27	3.76	Al ₄
Cb ₂ O ₆	1.52	26.58	.057	.91		
TiO ₂	.48	19.98	.024	.38		
Fe ₂ O ₃	.13	26.61	.005	.08		
FeO	.42	35.92	.012	.19		
MgO	.34	20.16	.017	.27		
MnO	.03	35.45	.001	.02		
Li ₂ O	7.26	14.95	.486	7.76	7.76	Li ₈
			5.257	84.00		
Na ₂ O	.53	31.00	.017	.27		
K ₂ O	11.03	47.10	.235	3.76		
Rb ₂ O	1.14	93.48	.012	.19		
			.264	4.22	4.22	K ₄
H ₂ O+	.47	9.00	.052	.83		
F	7.73	19.00	.407	6.51		
			.459	7.34	7.34	F ₂

Unit cell content: $K_4 \cdot Li_8 Al_4 \cdot Si_{16} O_{40} F_8$.

Formula: $K \cdot Li_2 Al \cdot Si_4 O_{10} F_2$.

The high percentages of Li₂O reported by Lorenzen and by Flink do not seem to be theoretically possible.

INTERPRETATION OF THE LEPIDOLITES AS ISOMORPHOUS MIXTURES

The percentages found in the analyses in Table 1 are very close to those that would be obtained in an isomorphous mixture of four end member micas, namely: polyolithionite, $K \cdot Li_2 Al \cdot Si_4 O_{10} F_2$; biotite, $K \cdot (Mn, Fe, Mg)_3 \cdot AlSi_3 O_{10} (F, OH)_2$; lithium muscovite, $K_4 \cdot Li_6 Al_6 \cdot Al_4 Si_{12} O_{40} (F, OH)_8$; and muscovite, $K \cdot Al_2 \cdot AlSi_3 O_{10} (OH, F)_2$.

The formulation of polyolithionite in the previous section supplies one of the missing end members in the series. The formula for lithium muscovite, as far as the writer knows, is hypothetical and represents the maximum lithium that may substitute for aluminum in the muscovite formula. It must be included to account for the lithium content found in the lepidolites. The biotite formula is represented by the bivalent cations in the analysis and is usually that of a manganese biotite with substitution of iron and magnesium for manganese in some of the samples. The

biotite may be divided, if desired, into a manganese biotite, an iron biotite, and a magnesium biotite (phlogopite), but for simplicity they have been taken as one component in the calculations.

Method of Calculating Ratios of End Member Micas

The calculation of the unit cell formula ratios is simple and involves few assumptions so that they should be accurate. These formula ratios may be calculated either from the relative equivalents (Table 2), the atomic ratios (Table 3), or, with more difficulty, the writer thinks, from the percentages in Table 1. The equations used to calculate the formula ratios are as follows:

$$\text{Polyolithionite} = \frac{\text{SiO}_2 - 48}{16}$$

$$\text{Biotite} = \frac{R''}{24}$$

$$\text{Lithium muscovite} = \frac{\text{Li} - 8P}{6}, \text{ where SiO}_2, R'', \text{ and Li are given in rela-}$$

tive equivalents for 84 octahedral and tetrahedral equivalents, and P is the unit cell formula ratio of polyolithionite. The muscovite was found by difference.

These ratios are of the unit cell formulas so that all are comparable with the larger formula for lithium muscovite.

To calculate the weight percentages of the end members from the unit cell formula ratios a number of assumptions are involved. The unit cell formula ratios were divided by the respective unit cell weights in obtaining the weight percentages of end members. With which end member the hydroxyl, sodium, rubidium, and cesium should be counted could not be told, so the end members taken were the potassium and fluorine unit cells, thus distributing the hydroxyl, sodium, rubidium, and cesium equally. The biotite unit cell weight was calculated for each sample from the ratios of the bivalent ions present (manganese, iron, and magnesium).

The figures obtained in the above calculations were then adjusted to add to 100 per cent. No correction was made for uncombined water.

Table 5 gives the unit cell formula ratios and Table 6 the weight percentages of the end member micas found by calculation. In Table 7 the silica, alumina, and lithia are calculated from the weight percentages of the end members. A close agreement between the calculated values and those determined shows that the compositions may be given satisfactorily in terms of the four end members chosen. The determined values for sample No. 17 in Table 7 are lower than those calculated because of the heavy elements columbium and titanium present in the mineral.

TABLE 5. UNIT CELL FORMULA RATIOS OF END MEMBER MICAS IN LEPIDOLITES

Sample No.	Polyolithionite	Biotite	Lithium Muscovite	Muscovite
1	.1606	.0508	.2742	.5144
2	.3250	.0121	.2017	.4612
3	.2562	.0183	.3251	.4004
4	.3294	.0063	.2408	.4235
5	.2831	.0175	.3325	.3669
6	.5739	.0171	.1415	.2675
7	.3750	.0575	.4267	.1408
8	.5815	.0046	.1247	.2892
9	.4630	.0042	.3443	.1885
10	.4737	.1012	.3698	.0551
11	.4425	.0025	.4067	.1483
12	.5115	.0054	.3313	.1518
13	.4300	.0151	.4667	.0883
14	.4800	.0213	.4400	.0587
15	.6737	.0538	.2234	.0491
16	.8237	.0229	.1351	.0183
17	.9450	.0200	.0334	.0016

TABLE 6. WEIGHT PERCENTAGES OF END MEMBER MICAS IN LEPIDOLITES
(uncorrected for H₂O—)

Sample No.	Polyolithionite	Biotite	Lithium Muscovite	Muscovite
1	15.5	6.4	27.1	51.1
2	31.8	1.6	20.2	46.5
3	25.0	2.3	32.4	40.2
4	32.3	.8	24.1	42.8
5	27.7	2.2	33.2	36.9
6	56.6	2.1	14.3	27.1
7	36.3	7.3	42.3	14.1
8	58.2	.6	12.4	28.9
9	45.7	.5	34.7	19.1
10	45.5	12.8	36.3	5.4
11	43.6	.3	41.0	15.1
12	50.4	.7	33.4	15.4
13	42.3	1.9	46.9	8.9
14	47.2	2.7	44.2	5.9
15	66.0	6.7	22.4	5.0
16	81.6	2.8	13.7	1.9
17	94.2	2.4	3.4	.2

TABLE 7. COMPARISON OF CALCULATED AND DETERMINED VALUES FOR
 SiO_2 , Al_2O_3 , AND Li_2O
 (corrected for H_2O -, OH, Na, Rb, and Cs)

Sample No.	SiO_2		Al_2O_3		Li_2O	
	Calc.	Found	Calc.	Found	Calc.	Found
1	47.0	47.00	30.8	30.60 + .26 Fe_2O_3	2.7	2.70
2	49.7	49.50	28.0	28.06	3.5	3.51
3	48.4	48.58	28.8	28.93	3.7	3.70
4	50.2	50.20	28.2	28.18	3.8	3.81
5	48.9	49.29	28.2	28.40	4.0	3.96
6	53.6	53.45	22.2	22.15	5.1	5.04
7	49.6	49.58	24.0	23.87 + .06 TiO_2	5.1	5.05
8	54.5	54.69	22.6	22.83	5.1	5.11
9	52.2	51.70	24.2	23.97	5.4	5.33
10	50.2	50.31	20.4	19.95 + .49 Fe_2O_3 + .22 TiO_2	5.4	5.39
11	51.3	51.10	24.1	23.98	5.5	5.51
12	52.8	52.58	22.8	22.82	5.7	5.64
13	51.2	51.25	23.8	23.71	5.8	5.78
14	51.8	51.07	22.4	22.05	6.0	5.89
15	54.4	54.40	18.0	17.95	6.2	6.18
16	57.2	57.03	15.75	15.55	6.9	6.84
17	60.5	59.56	13.7	12.04 + 1.52 Cb_2O_3 + .48 TiO_2	7.4	7.26

The selection of end members is an arbitrary one. Other combinations of four end members which express the compositions equally well are: polyolithionite, zinnwaldite (assuming its formula to be $\text{K} \cdot \text{LiR}^n\text{Al} \cdot \text{AlSi}_3\text{O}_{10}\text{F}_2$),¹⁴ lithium muscovite, and muscovite; polyolithionite, taeniolite ($\text{K} \cdot \text{LiR}^{n_2} \cdot \text{Si}_4\text{O}_{10}\text{F}_2$), lithium muscovite, and muscovite. In addition five or six of the above end members may be taken, depending on what end members are used to express the bivalent ion content. Selection of polyolithionite, biotite, lithium muscovite, and muscovite as end members serves to express the composition in the simplest way.

EVIDENCE OF DISCONTINUOUS ISOMORPHISM AND ITS INTERPRETATION

In crystalline substances there seems to be a tendency toward simple atomic ratios. This tendency is illustrated by the fact that in many solu-

¹⁴ Bragg gives $\text{K} \cdot \text{LiFeAl} \cdot (\text{Al} \cdot \text{Si}_3\text{O}_{10})(\text{OH}, \text{F})_2$. See Bragg, W. L., Atomic Structure of Minerals, p. 215, 1937.

tions of salts the individual salts crystallize separately, as solids having simple compositions.

If the lepidolite series is one of complete isomorphism of end members there would be no tendency to form simple ratios of atoms. In the process of their crystallization a surface of one composition would be covered by a layer of a different composition. For like to crystallize on like seems more plausible and this tendency would result in incomplete isomorphism. It seems well to examine the lepidolite analyses further to see if isomorphism is or is not continuous.

Simple Mica Unit Cells

A large number of mica unit cells having simple ratios of whole atoms are possible. Starting with muscovite with 12 atoms of silicon and 4 atoms of tetrahedral aluminum in a unit cell and replacing aluminum by silicon, atom for atom, the tetrahedral groups would be $Al_4Si_{12}O_{40}$, $Al_3Si_{13}O_{40}$, $Al_2Si_{14}O_{40}$, $AlSi_{15}O_{40}$, and $Si_{16}O_{40}$. As the tetrahedral and octahedral equivalents total 84, the equivalents in the octahedral group depend on which of the tetrahedral groups is present. With tetrahedral $Al_4Si_{12}O_{40}$ (60 equivalents) the octahedral equivalents would be 24; tetrahedral $Al_3Si_{13}O_{40}$ (61 equivalents) leaves 23 equivalents for the octahedral group; tetrahedral $Al_2Si_{14}O_{40}$ (62 equivalents) leaves 22 equivalents for the octahedral group; tetrahedral $AlSi_{15}O_{40}$ (63 equivalents) leaves 21 equivalents for the octahedral group; and tetrahedral $Si_{16}O_{40}$ leaves 20 equivalents for the octahedral group.

Then, starting with the octahedral groups filled with aluminum and replacing the aluminum atoms with other octahedral components, all combinations of whole atoms in unit cells may be devised.

The simple unit cell combinations are shown in figure 3 by the spaces containing the numbers. The numbers represent the octahedral aluminum atoms in the unit cell, corresponding octahedral equivalents other than aluminum are at the left, and the corresponding tetrahedral group is given below. Several unit cells may be represented by each square filled by a number, depending on what elements other than aluminum are in the octahedral group. Octahedral elements other than aluminum are in equivalents as these elements differ in valence.

The unit cells of muscovite (M), zinnwaldite (Z), biotite (B), taeniolite (T), and polyolithionite (P) are shown in figure 3. These so-called end member unit cells are non-polar, one side being a mirror image of the side opposite. Most of the intermediate unit cells are polar and for that reason would probably not have as great a tendency to repeat in a crystal as would the non-polar end member unit cells.

As a means of referring to the different unit cells in figure 3 the tetra-

hedral and octahedral constituents will be given in the following order: 1. octahedral *equivalents* other than aluminum; 2. octahedral aluminum *atoms*; 3 silicon *atoms*. The unit cell of muscovite, $K_4 \cdot Al_8 \cdot Al_4Si_{12}O_{40}(OH)_8$ in the lower right corner in figure 3 is given by the symbols 0-8-12.

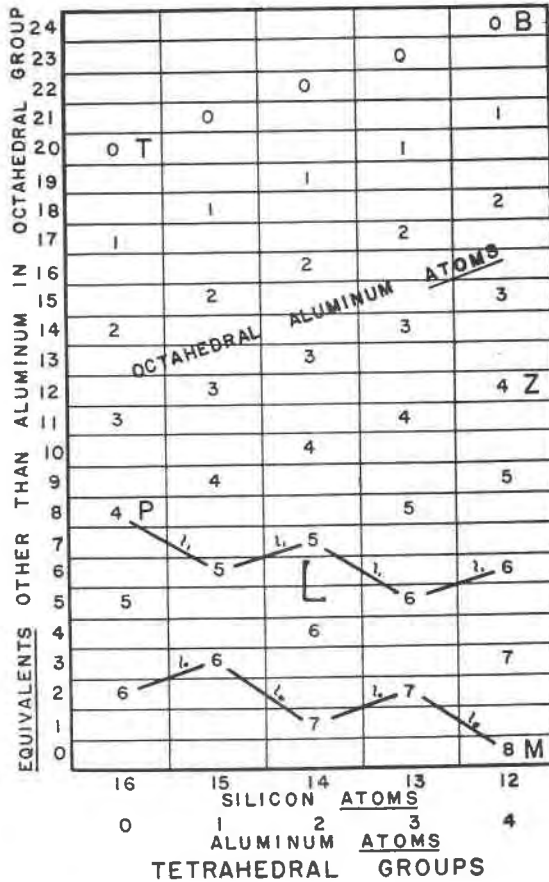


FIG. 3. Octahedral and tetrahedral groups in mica unit cells, ranging from tetrahedral $(AlSi_3O_{10})_4$ to $(Si_4O_{10})_4$. Numbers within the diagram represent octahedral aluminum atoms, the remaining octahedral equivalents being at the left, and corresponding tetrahedral groups at the bottom.

l_0 : Formulas below this line contain less than the minimum octahedral positions (8).

l_1 : Unit cells containing only lithium and aluminum in octahedral positions are below this line.

M = muscovite. Z = zinnwaldite. B = biotite.

T = taeniolite. P = polyolithionite.

L = probable lepidolite area.

Limits of Substitution in Lepidolites

Certain limits of substitution in figure 3 have theoretical significance. No unit cells are shown below the line marked l_0 because below that line the total number of octahedral positions occupied is less than 8, the minimum as far as is known. If the octahedral group consists only of lithium and aluminum the unit cell composition cannot be above l_1 , for above that line the octahedral atoms (Li+Al) would exceed 12, and there

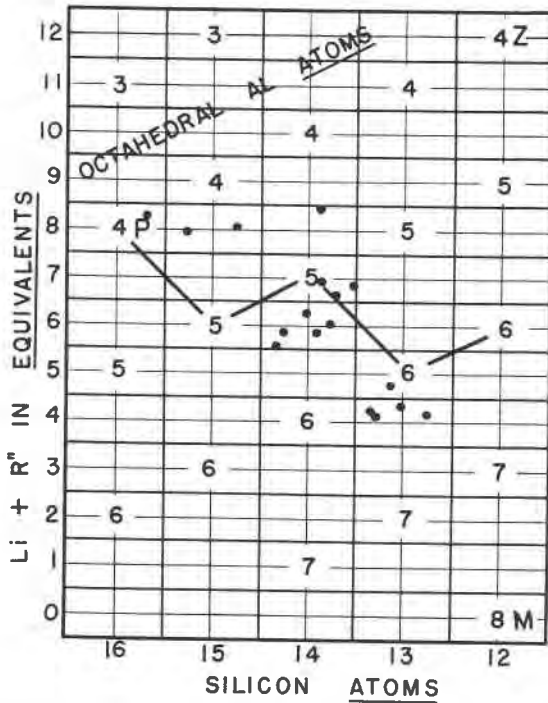


FIG. 4. Tetrahedral Si plotted against octahedral Li and R'' in lepidolites. Unit cells are designated by the numbers within the diagram as in figure 3.

are only 12 octahedral positions available in a mica unit cell. Therefore lepidolites containing no octahedral ions other than lithium and aluminum, as is essentially true of most of the lepidolites analyzed, would necessarily fall between the limits l_0 and l_1 , in figure 3. With a bivalent ion present in the octahedral group the unit cell composition may be above the limit l_1 , for one atom of a bivalent ion (such as Mn, Fe^{++} , and Mg) may take the place of two atoms of univalent lithium, ions in octahedral positions thus being reduced in numbers.

As the octahedral group in most of the lepidolites consists essentially of only lithium and aluminum, their compositions, for the most part, would lie under the limit l_1 , and somewhere between muscovite and polyolithionite, probably near the area marked with the large L in figure 3.

Figure 4 is a diagram consisting of the lower part of figure 3 and on which the silicon atoms per unit cell, calculated from the analyses, are plotted against the lithium and bivalent ion content in equivalents. The small quantities of other octahedral ions with valences greater than two are considered as substituting for aluminum.

As predicted in the discussion of figure 3 the compositions fall below the limit l_1 , except where bivalent ions cause them to go above that limit. A grouping of compositions may be seen in figure 4 immediately below the unit cells 7-5-14 and 5-6-13, which, together with polyolithionite, 8-4-16, the unit cell 6-5-15, and the unit cell 6-6-12, outline the limit l_1 . Theoretical considerations, therefore, indicate that the above unit cells represent ideal formulas, three of which are approximated by the analyses.

Ideal Formulas for Lepidolites

The ideal formulas, which are suggested by this discussion, are as follows:

Ideal formula A is polyolithionite, $K \cdot Li_2Al \cdot Si_4O_{10}F_2$, unit cell 8-4-16 in figures 3 and 4.

Ideal formula B, $K_4 \cdot Li_6Al_5 \cdot AlSi_{15}O_{40}(F, OH)_8$, unit cell 6-5-15 in figures 3 and 4.

Ideal formula C,¹⁵ $K_4 \cdot Li_7Al_5 \cdot Al_2Si_{14}O_{40}(F, OH)_8$, unit cell 7-5-14 in figures 3 and 4.

Ideal formula D, $K_4 \cdot Li_5Al_6 \cdot Al_3Si_{13}O_{40}(F, OH)_8$, unit cell 5-6-13 in figures 3 and 4.

Ideal formula E,¹⁵ $K_4 \cdot Li_6Al_6 \cdot Al_4Si_{12}O_{40}(F, OH)_8$, unit cell 6-6-12 in figures 3 and 4. This is the formula for lithium muscovite, proposed as an end member in the previous section.

Three points in figure 4, representing samples 10, 15, and 16, are well above the limit l_1 due to their content of bivalent ions. Samples well above the limit l_1 probably represent more nearly a condition of complete isomorphism as above that limit neighboring unit cells may be more nearly alike.

That the compositions approximate only roughly the ideal formulas is apparent, but nevertheless the lepidolites may be divided into groups

¹⁵ Niggli has proposed these two as ideal formulas for lepidolites, with Na replacing Li. See Niggli, Paul, Über die chemische Zusammensetzung der Alkaliglimmer: *Zeits. Kryst.*, vol. 96, p. 96, 1937.

whose limits of composition are given by the ideal formulas. These limits of composition may also be expressed in terms of end member micas. It seems apparent that in such a solid solution the end members would not be present as individual end member unit cells, but that the ions of which the end members are composed would be distributed uniformly throughout the structure. Such a uniform distribution of ions would involve the limits of composition expressed in the ideal formulas.

Types of Lepidolites

The lepidolites, therefore, may be divided into types as follows:

Type A. Limit of composition ideal formula A, polyolithionite, $K \cdot Li_2Al \cdot Si_4O_{10}F_2$. Presumably this formula has the maximum content of lithium in a mica. Sample 17 is essentially polyolithionite.

Type B. Limit of composition ideal formula B, $K_4 \cdot Li_6Al_5 \cdot AlSi_{16}O_{40} \cdot (F, OH)_8$, one formula of muscovite to three of polyolithionite. Further substitution of Li_3 for Al in the formula would require 13 octahedral positions and only 12 are contained in a mica unit cell. Substitution of LiMn for Al is structurally possible (octahedral positions filled would then be 12) and this substitution seems to have occurred in samples 15 and 16.

Type C. Limit of composition ideal formula C, $K_4 \cdot Li_7Al_5 \cdot Al_2Si_{14}O_{40} \cdot (F, OH)_8$. This type has approximately one unit cell formula of polyolithionite to not more than one unit cell formula of lithium muscovite. Samples 6, 7, 8, 9, 11, 12, 13, and 14 have this composition, as shown in Table 5 and in figure 4.

Type D. Limit of composition ideal formula D, $K_4 \cdot Li_5Al_6 \cdot Al_3Si_{13}O_{40} \cdot (OH, F)_8$. This type has approximately one unit cell formula of polyolithionite to not more than two unit cell formulas of lithium muscovite and not less than one unit cell formula of muscovite. Samples 1, 2, 3, 4, and 5 have this composition, as shown in Table 5, and figure 4.

Type E. Limit of lithium content ideal formula E, $K_4 \cdot Li_6Al_6 \cdot Al_4Si_{12}O_{40}(OH, F)_8$, the formula of lithium muscovite. None of the samples is of this composition.

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