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## FURTHER STUDIES IN THE MICA GROUP

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The heptaphyllite micas include two distinct systems which may be designated from their commonest types, the muscovite system and the lepidolite system. In a former study<sup>1</sup> of the mica group the best solution then discovered for the variations in composition in these systems involved the assumption of molecules with unequal numbers of oxygen atoms. It was stated at that time that: "The variations in tenor of oxygen are considered improbable and unsatisfactory, but, nevertheless, no better solution has been found." Further study of the problem seems to show that it is unnecessary to assume molecules with unequal numbers of oxygen atoms. The following paper is, then, an attempt to modify the writer's explanation of the constitution of the heptaphyllite micas so as to bring it more fully into harmony with the theory of atomic isomorphism. The two systems included in heptaphyllite will be discussed separately, beginning with the muscovite system.

### 1. THE MUSCOVITE SYSTEM

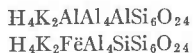
In the earlier discussion of the muscovite system it was suggested that the system has three end-member molecules, namely:

Muscovite	$H_4K_2Al_2Al_4SiSi_5O_{24}$
Protolithionite	$H_4K_2Fe_2Al_4FeSi_6O_{22}$
"Phengite"	$H_4K_2Si_2Al_4SiSi_4O_{25}$

It is only necessary to add two "phengite" molecules to one protolithionite molecule to obtain a molecule which is more satisfactory, since it contains the correct number of oxygen atoms. The resulting molecule is  $H_4K_2FeAl_4Si_7O_{24}$ , whose relationship to the muscovite molecule may be made more obvious by writing the molecules as follows:

<sup>1</sup> A. N. Winchell: *Am. Jour. Sci.*, **CCIX**, 1925, p. 415.

Muscovite  
Phengite



This last molecule is probably a better expression of the composition of the mineral called phengite by Tschermak than the formula previously assigned to it; it should be understood that the iron in the formula is ferrous and that magnesium may proxy for it in any amount—in fact, magnesium is more abundant than ferrous iron in nearly all modern analyses of phengitic micas.

The proposition that the muscovite-phengite series varies in composition from  $\text{H}_4\text{K}_2\text{Al}_6\text{Si}_6\text{O}_{24}$  to  $\text{H}_4\text{K}_2(\text{Mg, Fe})\text{Al}_4\text{Si}_7\text{O}_{24}$  implies that the points representing analyses in the triangle ABC of Figure 4 of the earlier article on the micas should all be grouped along or near a line running from B to a point at 33 1/3% from A toward C. In general, the analyses, as plotted, show 5–20% too much of the protolithionite molecule (C) to fall along this line; this is due, at least in large part, to the fact that in the computations for Figure 4 it was assumed<sup>2</sup> that titanium and ferric iron and manganese as well as magnesium all proxy for ferrous iron, although it was stated that “there is no evidence now available to determine the role of the minor constituents in this system,” and the assumption was made only because such a condition seems to exist in biotite. Further study of muscovite indicates that this assumption was probably incorrect. In computations for the present work it has been assumed that calcium and sodium proxy for potassium, ferrous iron for magnesium, ferric iron for aluminum, and titanium for silicon.

Many more analyses have been studied for the present occasion than were employed for the first discussion of heptaphyllite; indeed 110 analyses have been computed for the muscovite system alone, including all the good (and some poor) analyses listed by Doelter, and all the new analyses of Kunitz,<sup>3</sup> Jakob<sup>4</sup> and others. It has been found that 53 of these analyses can be computed into the molecules  $\text{H}_4\text{K}_2\text{Al}_6\text{Si}_6\text{O}_{24}$  and  $\text{H}_4\text{K}_2(\text{Mg, Fe})\text{Al}_4\text{Si}_7\text{O}_{24}$  with discrepancies<sup>5</sup> of less than 3% in alumina, less than 1% in potassa, 0% in magnesia and less than 5% total (irrespective of sign);

<sup>2</sup> *Am. Jour. Sci.*, CCIX, 1925, p. 421.

<sup>3</sup> *N. Jahrb. Min. Beil. Bd.*, L, 1924, p. 365.

<sup>4</sup> *Zeit. Kryst.*, LXII, 1925, p. 443.

<sup>5</sup> Discrepancies in fluorine and water are disregarded; Ca is calculated as Na, MnO as FeO and  $\text{Mn}_2\text{O}_3$  as  $\text{Al}_2\text{O}_3$ .

these 53 analyses are plotted in Fig. 1; 24 other analyses<sup>6</sup> correspond with these formulas except for abnormal tenor of alkalis. Most of the other analyses are old and very probably inaccurate. It is worth noting that 16 out of the 20 recent analyses of Kunitz and Jakob correspond well with the formulas here proposed and the 4 others are abnormal only in their tenor of alkali.

Penfield's<sup>7</sup> "alurgite" (No. 128 of Fig. 1) is apparently even more siliceous than typical phengite. That is, phengite is produced when one MgSi group proxies for one AlAl group of the muscovite molecule. In alurgite, and also in analysis 104, this replacement is carried to completion and about twenty percent of a second AlAl group is replaced by MgSi. Therefore the points which represent the composition of these micas are a short distance outside of the triangle of Fig. 1, as suggested by the arrows.

Of course, every change of composition produces some effect upon the optic properties; since there are many minor variations in composition in the muscovite system, such as, a variable tenor of Na which proxies for K, and of Ti which proxies for Si, and of Mn and still other elements, it is impossible to prepare a diagram which will express accurately the relations between variations in composition and variations in optic properties, because no diagram can show the effects of so many variables. However, study of the data shows that, as usual, iron is especially important in its effects on the optic properties and the effects of ferrous and ferric iron are quite different. The chief effect of ferric iron is to raise the index of refraction and apparently also the birefringence, while the chief effect of ferrous iron is, like that of magnesium, to decrease the optic angle. Apparently ferrous iron has a greater effect than magnesium on the optic angle. The diagram (Fig. 1) is based on available data and represents phengite with a little ferrous iron. It may be estimated that iron-free phengite would have the optic plane normal to (010), as in muscovite, with  $2V = 25^\circ \pm$ ,  $N_m = 1.59 \pm$ , and  $N_g - N_p = 0.04 \pm$ , while pure iron (magnesium-free) phengite would have the optic plane parallel with (010),  $2V = 10^\circ$

<sup>6</sup> These are Doelter's Nos. 9, 20, 22, 33, 36, 38, 46, 59, 61, 62, 71, 76, 78, 79, 102, 110, 113, 119, and my No. 126 (P. P. Pilipenko: *Min. Abst.*, II, 1923, p. 112), 127 (E. V. Shannon: *U.S. Nat. Mus. Bull.* 131, 1926, p. 372), 135 (W. Kunitz: *loc. cit.*, No. 5), and 152, 154, 156 (J. Jakob: *Zeit. Kryst.*, LXII, 1925, p. 443, Nos. 2, 4, and 6).

<sup>7</sup> *Am. Jour. Sci.*, CXLVI, 1893, p. 288; Larsen (*U. S. Geol. Surv. Bull.* 679) gives for alurgite  $(- )2V = \text{near } 0^\circ$ ,  $\rho > \nu$  weak,  $N_m = 1.594$ ,  $N_g - N_p = \text{like muscovite}$ .

or more, and  $N_m =$  about 1.61. It should be understood that Fig. 1 is not supposed to be highly accurate so far as the relations between composition and optic properties are concerned, but it represents an average picture of existing data. It does not express accurately the optic data of Jakob, but these seem to be decidedly less accurate than the optic data of Kunitz.

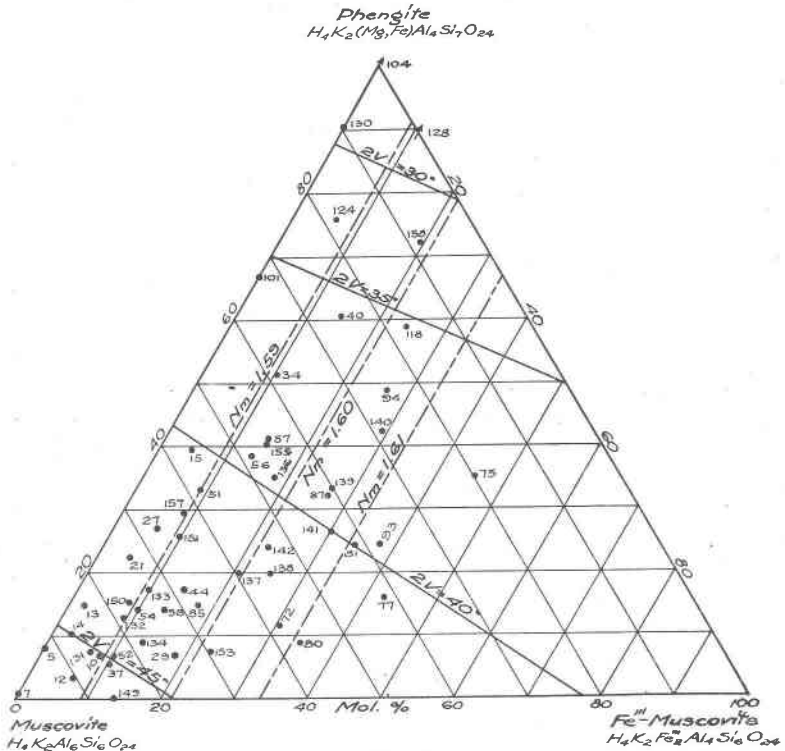


FIG. 1.

## REFERENCES FOR FIG. 1.

For the chemical analyses, all numbers below 124 refer to the same numbers as listed by Doelter (HANDB. MINERALCH., II, 2, 1917, pp. 418-431). 124, H. Sigg: *Zeit. Kryst.*, LVII, 1923, p. 418. 127, E. V. Shannon: *U. S. Nat. Mus. Bull.* 131, 1926, p. 372. 128, S. L. Penfield: *Am. Jour. Sci.*, CXLVI, 1893, p. 288. 130, W. Kunitz: *N. Jahrb. Min. Beil. Bd.*, L, 1924, p. 365. 131-142, W. Kunitz: *N. Jahrb. Min. Beil. Bd.*, L, 1924, p. 412, Nos. 1-12 in order. 149-158, J. Jakob: *Zeit. Kryst.*, LXII, 1925, p. 443, Nos. 9-18 in order. For the optic data: No. 10, R. Scharizer: *Zeit. Kryst.*, XIII, 1888, p. 463, gives  $2E = 73^\circ 52'$  Na for muscovite from Auburn, Me. 13, A. Hutchinson and W. C. Smith: *Mineral Mag.*, XVI, 1912, p. 264, give

$2E=68^{\circ} 50'$ ,  $N_g=1.594$ ,  $N_m=1.589$ . 14, R. Scharizer: *Zeit. Kryst.*, **XIII**, 1888, p. 463 gives  $2E=73^{\circ} 52'$ . 21, G. Tschermak: *Zeit. Kryst.*, **III**, 1879, p. 127, gives  $2E=68^{\circ} 24'$  Na. 52, R. Scharizer: *Zeit. Kryst.*, **XIII**, 1888, p. 460, gives  $2E=70^{\circ} 58'$  Na. 117, G. Tschermak: *Zeit. Kryst.*, **XIII**, 1879, p. 147 gives  $2E=60^{\circ} 12'$  Na. 118, G. Tschermak: *loc. cit.*, gives  $2E=55^{\circ} 0'$  red. 124, H. Sigg: *Zeit. Kryst.*, **LVII**, 1923, p. 418 gives  $2V=36^{\circ} 47'$ ,  $N_g=1.5976$ ,  $N_m=1.5936$ ,  $N_p=1.5574$ . 128, E. S. Larsen: *U. S. Geol. Surv. Bull.* **679**, 1921, p. 39, gives for alurgite from Piedmont  $2V$ =nearly  $0^{\circ}$ ,  $\rho > \nu$  weak,  $N_m=1.594$ ,  $N_g-N_p$ =like muscovite. For optic data on 130-141 see W. Kunitz: *N. Jahrb. Min. Beil. Bd.*, **L**, 1924, p. 365. For optic data on 149-158 see J. Jakob: *Zeit. Kryst.*, **LXII**, 1925, p. 443.

Since the publication of the first article<sup>8</sup> on the constitution and optic properties of the micas, Hallimond<sup>9</sup> has published an interesting discussion of the micas in which he emphasizes two points, namely, the idea that RO can replace  $R_2O_3$  and the constancy of the  $K_2O:SiO_2$  ratio. The writer believes that the RO: $R_2O_3$  ratio is variable and that  $K_2O$  is (nearly) constant, but he can not agree with Hallimond's interpretation of either of these facts. The writer believes that the RO: $R_2O_3$  ratio varies because  $Al_2O_3$  replaces  $MgSiO_3$  and *vice versa*. He believes that potash is constant (apparently with some unexplained exceptions), but does not believe that silica is constant; therefore the  $K_2O:SiO_2$  ratio is not constant.

Hallimond<sup>10</sup> rejects the writer's view that the  $K_2O:SiO_2$  ratio varies from 1:6 in muscovite and phlogopite to 1:5 in protolithionite and siderophyllite and to 1:8 in "phengite."<sup>11</sup> The evidence offered by him to prove that the  $K_2O:SiO_2$  ratio is always 1:6 does not seem conclusive; it consists of three diagrams showing the relative abundance of all sorts of ratios of  $K_2O$  to  $SiO_2$  in "muscovite," "lithia micas" and "biotite." Hallimond's own diagram shows that the  $(K_2O:SiO_2=)1:7$  ratio is nearly as common in "muscovite" as the 1:6 ratio. However, the frequency of occurrence of various ratios is not the question at issue. It seems strange that it should be necessary to point out that it is not safe to conclude that the more uncommon ratios are all due to errors and do not actually occur. If such an assumption were correct it would be possible to prove by the same sort of method that marialite is not a mineral, but only a figment of the imagination, because it is quite rare, and the common ratio between

<sup>8</sup> *Amer. Jour. Sci.*, **CCIX**, 1925, p. 309 and 415.

<sup>9</sup> A. F. Hallimond: *Mineral Mag.*, **XX**, 1925, p. 305, and **XXI**, 1926, p. 25.

<sup>10</sup> *Mineral. Mag.*, **XXI**, 1926, p. 26.

<sup>11</sup> This ratio is 1:7 in phengite as defined previously in this article.

$\text{Na}_2\text{O}$  and  $\text{SiO}_2$  in scapolite is far less than that found in marialite. Many other minerals could be proved to be non-existent by the same method.

It is not difficult to test the question at issue. So far as known to the writer no one has suggested that in muscovite proper the  $\text{K}_2\text{O}:\text{SiO}_2$  is not 1:6. Consequently data in regard to muscovite proper have no bearing on the question. It has long been recognized that there is a gradation from muscovite proper to a very similar substance, called phengite by Tschermak, which is distinguished from muscovite by its small optic angle and by its high tenor of silica. The writer has suggested that in phengite the  $\text{K}_2\text{O}:\text{SiO}_2$  is not 1:6. Doelter gives twenty-one analyses which he classifies as phengite. Testing these analyses by Hallimond's method of computing the number of  $\text{K}_2\text{O}+\text{Na}_2\text{O}$  molecules in each when the  $\text{SiO}_2+\text{TiO}_2$  molecules are calculated to be 600, the following list may be obtained, taking Doelter's analyses 102-122 in regular order: assuming  $\text{SiO}_2+\text{TiO}_2=600$ ,  $\text{K}_2\text{O}+\text{Na}_2\text{O}+\text{Ca}_2\text{O}_2^{12}=74, 86, 73, 56, 75, 45, 55, 80, 58, 79, 85, 74, 40, 37, 92, 85, 83, 68, 74, 66, 63$ . It is unfortunately true that all of these analyses are old and most of them probably inaccurate, but it is interesting that no one of them shows as high a tenor of alkalis as required by Hallimond's theory, the average being only 69, instead of 100. The only modern analysis of a uniaxial "muscovite," that is, a phengite, known to the writer is that published in 1924 by Kunitz;<sup>13</sup> this analysis contains 83 molecules of  $\text{K}_2\text{O}+\text{Na}_2\text{O}$  (and no lime) to 600 molecules of  $\text{SiO}_2+\text{TiO}_2$ . On the basis of the theory suggested by the writer this mineral should have 87 molecules<sup>14</sup> of alkali to 600 of  $\text{SiO}_2+\text{TiO}_2$ . Jakob<sup>15</sup> published in 1925 an analysis of a potash mica with a small optic angle ( $2V=29^\circ 20'$ ); this analysis contains 83 molecules of alkali (and no lime) to 600 molecules of  $\text{SiO}_2+\text{TiO}_2$ ; the theory of the writer would require 86 molecules of alkali. Penfield's "alurgite" is apparently a phengite; it contains 84 molecules of alkali (and no lime) to 600 of  $\text{SiO}_2+\text{TiO}_2$ ; the writer's theory would require 83 molecules of alkali.

<sup>12</sup> The writer believes that Ca proxies for K in mica as in feldspar; if Ca were omitted, according to the practice of Hallimond, the results would be slightly more unfavorable to his theory.

<sup>13</sup> *N. Jahrb. Min. Beil. Bd.*, I, 1924, p. 383.

<sup>14</sup> Calculated on the assumption that the iron oxide (5.92%) reported by Kunitz is all FeO.

<sup>15</sup> *Zeit. Krist.*, LXII, 1925, p. 443, No. 18.

In summary, every analysis of phengite on record shows that the mineral does not contain 100 molecules of alkali (plus lime) to 600 molecules of  $\text{SiO}_2 + \text{TiO}_2$ ; the old analyses give an average of only 69 molecules of alkali; the actual number present is probably greater as indicated by recent accurate analyses on carefully purified material. The data from the latest analyses accord well with the theory that the phengite molecule contains alkali in the ratio  $\text{K}_2\text{O}:\text{SiO}_2 = 1:7$ .

#### THE LEPIDOLITE SYSTEM

Another attempt to solve the problems of the constitution of the lepidolite system has given results which are not entirely satisfactory. However, a study of the 53 available analyses (with special attention to the best ones) seems to establish a difference between the muscovite system and the lepidolite system in the number of oxygen (plus fluorine) atoms in the unit cell or crystal. All formulas of the muscovite system contain twenty-four atoms of oxygen to two of potassium. The number of oxygen atoms in the formulas of the lepidolite system is clearly less than twenty-four; it is probably twenty-two, although twenty-one is not excluded by the data. Muscovite and lepidolite do not seem to be miscible to any important extent, and this condition is perhaps due to the different number of oxygen atoms.

If the number of oxygen atoms be disregarded (as was done in the previous study) it is reasonable to regard the muscovite molecule as one end member of the lepidolite system. Since the muscovite molecule is not satisfactory in the number of its oxygen atoms a different molecule should take its place as an end member of the lepidolite system. Hallimond<sup>16</sup> gives  $\text{H}_4\text{K}_2\text{Li}_2\text{Al}_4\text{Si}_6\text{O}_{22}$  as the formula of lepidolite; this is exactly the composition expressed by a point in figures 4 and 5 of the writer's<sup>17</sup> earlier discussion of the micas at one third the distance from B toward E; it may be accepted as probably correct, at least for one kind of lepidolite. This formula has the same number of oxygen atoms and the same number of other atoms as the writer's formula for protolithionite, namely,  $\text{H}_4\text{K}_2\text{Fe}_3\text{Al}_4\text{Si}_5\text{O}_{22}$ , which seems to be correct for another end member of the lepidolite system.

<sup>16</sup> *Mineral. Mag.*, XX, 1925, p. 305.

<sup>17</sup> *Am. Jour. Sci.*, CCIX, 1925, p. 415.

The correct formulas of the other end members of the lepidolite system are quite doubtful. Only two analyses of polyolithionite are on record and they are old and of very questionable accuracy. Hallimond has suggested the formula  $K_2O \cdot 2Li_2O \cdot Al_2O_3 \cdot 6SiO_2 \cdot 2H_2O$ , which is exactly the composition expressed by a point in figures 4 and 5 of the writer's earlier article on the micas at two thirds the distance from B toward E. This formula is deficient in oxygen atoms; data at present available are quite insufficient to determine whether the formula should be written with 4  $H_2O$  (including fluorine) or in some other way (such as with more silica) to provide the lacking oxygen atoms. For the present occasion the writer has used the formula  $H_8K_2Li_4Al_2Si_6O_{22}$ .

The fourth end member of the lepidolite system seems to be approximately represented in the rare mineral called cryophyllite, of which only a few old analyses are available. The formula of this mineral is given by Hallimond as follows:  $H_4K_2Li_2FeAl_2Si_6O_{20}$ ; the data in this case indicate rather clearly that this should be written  $H_4K_2Li_2FeAl_2Si_7O_{22}$ ; the evidence on this point will be given later.

In summary, the end members of the lepidolite system seem to be the following molecules:

Lepidolite	$H_4K_2Li_2Al_4Si_6O_{22}$
Polyolithionite	$H_8K_2Li_4Al_2Si_6O_{22}$
Protolithionite	$H_4K_2Fe_3Al_4Si_6O_{22}$
Cryophyllite	$H_4K_2Li_2FeAl_2Si_7O_{22}$

The way in which atoms proxy for one another can be made more evident by writing the formulas as follows:

Lepidolite	$H_4K_2Li_2AlAl_2AlSiSi_6O_{22}$
Polyolithionite	$H_8K_2Li_2LiAl_2LiSiSi_6O_{22}$
Protolithionite	$H_4K_2Fe_2FeAl_2AlAlSi_6O_{22}$
Cryophyllite	$H_4K_2Li_2FeAl_2SiSiSi_6O_{22}$

Ordinary lepidolite contains 50 to 100 per cent of the first molecule, the remainder being chiefly polyolithionite with or without minor amounts of protolithionite or cryophyllite.



Ordinary lepidolite is miscible in all proportions with protolithionite and these intermediate types are known as zinnwaldite (the name lithionite has been used occasionally for some of these). The end member (protolithionite) is rather rich in iron (or magnesium) so that it is usually dark colored or black and has been called biotite probably in some cases. Polyolithionite is of doubtful stability, but mica of approximately this composition has been described from Greenland. Cryophyllite is also somewhat doubtful, but has been described from Cape Ann, Massachusetts; it seems to be fairly common as a minor constituent in the lepidolite system, but almost unknown as a dominant constituent.

The chief constituents of the lepidolite system are shown in figure 2. Analyses represented by points contain no cryophyllite; the size of small triangles (or lines along the left hand edge) is proportional to the tenor of cryophyllite in the sample. Six analyses are too rich in cryophyllite to be useful in the figure; ten analyses are rather clearly inferior; thirty-three of the remaining thirty-seven are shown in Fig. 2. Many of the analyses and some of the optic data are not very accurate and analyzed samples with recorded optic data are not numerous; therefore the lines representing the relations between optic data and composition are not supposed to be very accurate; however, making a suitable allowance for the influence of cryophyllite in some cases, they give the index within about  $\pm 0.01$ , the optic angle within about  $\pm 5^\circ$ , and the birefringence within about  $\pm .005$  in all cases carefully determined, except for Nos. 3, 10, and 22.<sup>18</sup> The diagram would require that polyolithionite have its optic plane normal to the position found in lepidolite and zinnwaldite, that is, parallel to (010) and this is the position reported by Lorenzen;<sup>19</sup> his statement of the dispersion, namely  $\rho < \nu$ , is the reverse of that found in lepidolite and zinnwaldite, as it should be if the optic angle varies through zero as the composition varies from lepidolite to polyolithionite.

<sup>18</sup> As noted in the first discussion of the micas (*op. cit.* p. 418) two of these are probably incorrect; the third was reported in 1874.

<sup>19</sup> *Zeit. Kryst.*, IX, 1884, p. 252.

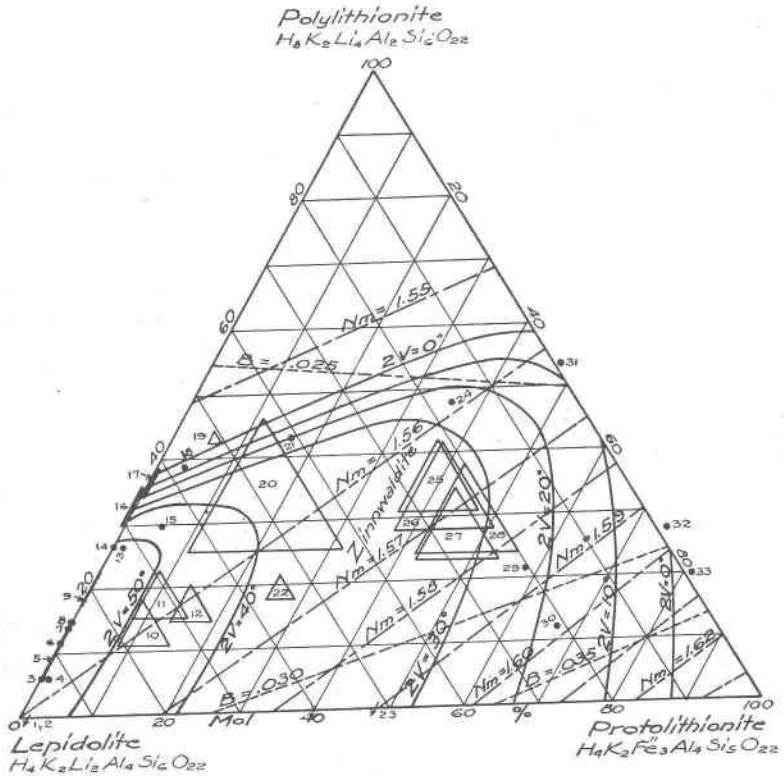


FIG. 2.

## REFERENCES FOR FIG. 2.

- 1, 2, 3, 4, R. B. Riggs: *U. S. G. S. Bull.* 42, 1887, p. 18 (white and brown), p. 14 and p. 17, No. II.
- 5, 6, W. T. Schaller: *U. S. G. S. Bull.* 419, 1910, No. K and L.
- 7, F. Berwerth: *Min. Mit. Beil. J. R. A.*, 1877, p. 343.
- 8, W. T. Schaller: *U. S. G. S. Bull.* 419, 1910, p. 287, No. 1.
- 9, R. B. Riggs: *op. cit.* p. 17, No. I.
- 10, W. Kunitz: *N. Jahrb. Min. Beil. Bd.*, L, 1924, p. 394, 395, No. 4.
- 11, W. Kunitz: *op. cit.*, p. 413, No. 2.
- 12, V. Dürfeld: *Zeit. Kryst.*, XLVI, 1909, p. 573.
- 13, C. Doelter: *Min. Chem.*, II, 2, 1917, p. 450, No. 11.
- 14, W. T. Schaller: *op. cit.*, p. 287, No. H.
- 15, R. Scharizer: *Zeit. Kryst.*, XII, 1887, p. 1 and XIII, 1888, p. 467.
- 16, W. Kunitz: *op. cit.*, p. 394, 395, No. 3.
- 17, R. B. Riggs: *op. cit.*, p. 12. No. I.
- 18, A. N. Winchell: *Am. Jour. Sci.*, CCIX, 1925, p. 424, No. 51.
- 19, A. N. Winchell: *op. cit.*, No. 21a.
- 20, W. Kunitz: *op. cit.*, p. 394, 395, No. 5.
- 21, R. A. Johnston: *Am. Jour. Sci.*, XI, 1901, p. 149.
- 22, W. T. Schaller: *Am. Jour. Sci.*, XXII, 1907, p. 158.
- 23, E. S. Simpson: *Geol. Surv. W. Australia, Bull.* 48, 1915, p. 95, and A. N. Winchell; *Am. Jour. Sci.*, CCIX, 1925, p. 426, No. 31.
- 24, C. F. Rammelsberg:

*Min. Chem. Sup.*, 1895, p. 418. 25, Duparc, Wunder et Sabot: *Mem. Soc. Phys. Hist. Nat. Geneve*, XXXVI, 1910, p. 369. 26, W. Kunitz: *op.cit.*, p. 394, 395, No. 8. 27, F. Berwerth: *op. cit.*, p. 343. 28, W. Kunitz: *op. cit.*, p. 394, 395, No. 9. 29, W. Kunitz: *op. cit.*, p. 394, 395, No. 10. 30, S. Haughton: *Min. Cornwall*, 1876, p. 72, quoted by E. S. Simpson: *Geol. Surv. W. Australia Bull.* 48, 1912, p. 95. 31, Stein: *Jour. prak. Chem.*, XXVIII, 1883, p. 295. 32, H. Schulze: *Zeit. prak. Geol.*, 1896, p. 377. 33, W. Kunitz: *op. cit.*, p. 394, 395, No. 9.

Hallimond<sup>20</sup> has recently discussed the composition of the micas and reached the conclusion that the K<sub>2</sub>O:SiO<sub>2</sub> ratio has the constant value 1:6 in all of them. In support of this conclusion he presents diagrams showing that this ratio is commoner than any other in "muscovite," "lithia micas" and "biotite." According to this theory such molecules as suggested above for protolithionite and cryophyllite are impossible. It is therefore necessary to examine the available data bearing on this question as applied to protolithionite and cryophyllite.

Hallimond gives a list of eleven analyses of "lepidolite-protolithionite (zinnwaldite)." One of these (Dana No. 8) is evidently included by mistake since the analysis cited does not correspond even approximately with the ratios given by Hallimond; four others are too old to be worthy of serious consideration. The six remaining analyses give the following results (assuming SiO<sub>2</sub>+TiO<sub>2</sub>=600):

ANALYST	DATE	K <sub>2</sub> O+Na <sub>2</sub> O+Ca <sub>2</sub> O <sub>2</sub>		
		Hallimond's theory	Found	Winchell's theory
Schaller	1907	100.	98.5	104.5
Dürrfeld	1909	100.	107.4	105.9
Kunitz 8	1924	100.	99.7	105.6
Kunitz 9	1924	100.	93.9	106.4
Kunitz 10	1924	100.	106.0	111.5
Kunitz 11	1924	100.	108.5	116.0

Three of these analyses are favorable to Hallimond's theory as compared with the writer's theory; two of them are as good for one theory as the other and one of them is favorable to the writer's theory. The differences in all cases are rather small; it is easy to demonstrate that modern analyses report more alkali than old analyses; is it possible that even in modern analyses the tenor of

<sup>20</sup> *Mineral. Mag.*, XX, 1925, p. 305 and XXI, 1926, p. 25.

alkali reported is actually a little too low? It seems possible, also, that Hallimond is correct in his idea that  $\text{Li}_2\text{O}$  can proxy, *in small amount*, for  $\text{K}_2\text{O}$ ; if so, this might account for the discrepancies found. Finally, it seems to the writer that other ratios are as important in micas as the  $\text{K}_2\text{O}:\text{SiO}_2$  ratio, and he has been unable to find any simple formula for protolithionite containing 6  $\text{SiO}_2$  which satisfies these other ratios as well as does the formula with 5  $\text{SiO}_2$ —this is especially true for the analysis richest in the protolithionite molecule, namely Kunitz No. 11.

In regard to the formula of cryophyllite, Hallimond gives a list of six analyses of "lepidolite-cryophyllite"; the first of the list contains only 8% of the cryophyllite molecule and therefore should not be considered in attempting to determine the formula of that mineral. The data for the remaining five analyses are as follows (assuming  $\text{SiO}_2 + \text{TiO}_2 = 600$ ):

ANALYST	DATE	REF.	$\text{K}_2\text{O} + \text{Na}_2\text{O} + \text{Ca}_2\text{O}_2$		
			HALLIMOND'S THEORY	FOUND	WINCHELL'S THEORY
Cooke <sup>21</sup>	1867	Dana 3	100	94.3	93.8
Riggs	1887	Dana 4	100	89.6	96.4
Riggs	1887	Dana 5	100	89.4	96.7
Riggs	1887	Dana 6	100	84.2	96.9
Kunitz	1924	Kunitz 5	100	97.0	99.2

In this case the data clearly favor the theory of the writer as compared with that of Hallimond. It is worthy of note that  $\text{K}_2\text{O}$  is insufficient to equal the requirements of the writer's theory in every case studied, namely, phengite, protolithionite and cryophyllite. This is perhaps due to one or more of the reasons given in connection with the case of protolithionite.

Summarizing the conclusions reached in this article it may be said that although the formulas of the end-members of the lepidolite system are still in doubt, the formulas proposed in this article correspond reasonably well with the best analyses. These formulas are characterized by having a constant number of  $\text{K} (+ \text{Na} + \text{Ca})$  atoms, a constant number of  $\text{O} (+ \text{F})$  atoms, and a constant number of other atoms. (Such a constancy seems to be required in molecules which are miscible in the crystal state by our present knowledge of crystal space lattices.) The writer is

<sup>21</sup>  $\text{SiO}_2$  increased to 53.46 (as done by Riggs and Doleter) by adding  $\text{SiO}_2$  from  $\text{SiF}_4$  reported.

unable to accept the conclusion of Hallimond that the  $K_2O:SiO_2$  ratio is 1:6 in all mica molecules and gives the evidence to show that it is 1:7 in phengite and cryophyllite. He believes that the  $K_2O:SiO_2$  ratio is 1:5 in protolithionite, partly because he has been unable to find any simple formula for that molecule containing 6  $SiO_2$  which satisfies the other ratios as well as does the formula with 5  $SiO_2$ .

This article has benefited by the constructive criticism of Professor R. C. Emmons.

### ROEMERITE FROM CALIFORNIA

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Roemerite occurs as one of the interesting sulphates developed by the alteration of pyrrhotite at Island Mountain, Trinity County, California. Several very fine specimens of this rare sulphate were collected by Mr. Vonsen of Petaluma, California, and since no American locality has hitherto been given for its occurrence, it was thought worthy of note. It was mentioned as one of the interesting secondary minerals from the pyrrhotite with claudetite, hieberite, goslarite, fibroferrite, and morenosite by Dr. Eakle<sup>1</sup> in his bulletin, but no description of the crystals was given.

The first report of roemerite appears in 1858,<sup>2</sup> when it was found in the Rammelsberg mine in the Hartz mountains, near Goslar. Grailich, who was the first to describe the mineral, measured the interfacial angles with a contact goniometer, and concluded that the mineral was monoclinic in crystallization. Chemical analyses were made of the same material by Tschermak. J. Blaas<sup>3</sup> reported roemerite from Persia in 1885. His material came in smaller but better crystals, so that it was possible to use a reflection goniometer. The results of Blaas' work showed that the mineral was not monoclinic, but triclinic. In 1888<sup>4</sup> roemerite was discovered in Chile, where it occurs with several other natural sulphates. The

<sup>1</sup> Minerals of California, A. S. Eakle; *Bull.* 91, *Calif. State Min. Bureau*, p. 280.

<sup>2</sup> Der Römerit, ein neues Mineral aus dem Rammelsberge, Joseph Grailich; *Sitzungsberichte d. k. Akademie der Wissenschaften, Wien*, 28 (1858), pp. 272-288.

<sup>3</sup> Beiträge zur Kenntniss natürlicher wasserhaltiger Doppelsulfate, J. Blaas; *Zeitschr. für Kryst.*, 10, (1885), pp. 409-412.

<sup>4</sup> Beiträge zur Kenntniss der Sulfate von Tierra ammarilla bei Copiapo in Chile, G. Linck; *Zeitschr. für Kryst.* 15 (1889), pp. 22-26.