ART. XL.—*Researches on the Lithia Micas*;* by F. W. CLARKE.

I. THE LEPIDOLITES OF MAINE.

IN the western part of Maine, along a line running southeasterly from the Rangeley Lakes to a point on the seaboard between Brunswick and Portland, there are a series of veins of albitic granite which are noted for their contents in lithia mica and colored tourmalines. These localities, in the towns of Rumford, Paris, Norway, Hebron and Auburn, are all within a narrow belt of about forty miles in length; and with them, as a probable part of the same system, may be classed the spodumene locality in the town of Peru. The northernmost of the known localities is that on Black Mountain in Rumford; but a few fragments of inferior green tourmaline have been found about five miles farther north, in Roxbury—a fact which indicates a prolongation of the belt in that direction. Similarly, a southern extension of the belt is suggested in the territory covered by the towns of Pownal, Yarmouth and Freeport, a region from which a few casual specimens of lepidolite have been reported. The total width of the belt, so far as has been observed, appears to be not much over fifteen miles, although sharp definition of the area is not yet possible.

In general character the several localities are much alike, although in points of minor detail they vary considerably. With the tourmaline and lepidolite, quartz, muscovite, cleavelandite, cassiterite and amblygonite are always found, together with a wide range of other species which cannot be considered here. Some of the differences are probably due to the fact that certain localities have been more thoroughly opened up than others, and these differences would probably be eliminated by more complete exploration. In other points, however, the variations are notable and characteristic. For convenience we may briefly consider the several localities separately, beginning with the northernmost and proceeding southward in regular order.

Rumford.†—This locality, discovered in 1878 by Mr. E. M. Bailey, of Andover, Maine, is situated on the northern slope of Black Mountain, at an estimated elevation of about one thousand feet above the valley. The excavation, so far, has been merely superficial. A part of the lepidolite, which is very abundant, is fairly pure, lilac-purple in color and finely granular; but the larger portion of the mineral is coarser in structure and thickly sprinkled with small, opaque, red tourmalines.

*Abstract of a paper to appear in full in a forthcoming Bulletin of the U. S. Geological Survey.

† See G. F. Kunz, Proc. Amer. Assoc., 1885.

The color of the latter is very rich and the appearance of the material carrying the associated minerals is strikingly characteristic. It resembles nothing from the other localities of the region. Green tourmaline occurs here but sparingly, while the spodumene is enormously developed, showing faces over a metre in length. At the other lepidolite localities spodumene is one of the scarcer minerals.

Paris.—The famous locality known as Mt. Mica has been so thoroughly studied by mineralogists that no detailed account of it is needed here. The lepidolite occurs both in the ordinary, purple, granular form and in broad foliations resembling muscovite. Analyses of it have been published by Berwerth and by Rammelsberg,* which agree fairly well with the results obtained in this laboratory.

Hebron.—The deposit at this point, about seven miles southeast of Mt. Mica, has long been well known to collectors. It has only been superficially opened, however, and deserves more systematic exploration. The lepidolite, which is coarsely granular and purple, is especially interesting on account of its yield in cæsia and rubidia, which, after their identification by Johnson and Allen here, were also found in the lepidolite of Rozena. As yet no complete analysis of the Hebron mineral seems to have been published. For the specimens examined in this laboratory I am indebted to the kindness of Professor G. J. Brush.

Associated with the Hebron lepidolite there have been found many specimens of red and green tourmaline, which, while preserving their crystalline form, have undergone an alteration into a softer mineral of an opaque, talcose appearance. Some of the material so derived from tourmaline has been supposed to be lepidolite; and as it was possible that a study of it might have interest, an analysis was made by Mr. R. B. Riggs. The specimen chosen was originally rubellite, and its specific gravity, as determined by Mr. T. M. Chatard, was 2.87. Analysis as follows:

SiO ₂	43.90
Al ₂ O ₃	38.71
Fe ₂ O ₃	.58
FeO	.25
MnO	.04
CaO	.41
MgO	.05
Na ₂ O	1.05
K ₂ O	10.92
H ₂ O	4.25
F	none
B ₂ O ₃	trace
	<hr/> 100.16

* See third supplement to Dana's Mineralogy, pp. 78, 79.

These results show clearly that the alteration product is not lepidolite but damourite, a fact which could hardly be altogether unexpected. The material analyzed was received from Mr. N. H. Perry of South Paris, Maine.

Norway.—The specimens from this locality were partly collected during a field expedition, and partly received from N. H. Perry. The lepidolite was of two varieties; one, pale brown and very finely granular, the other much coarser in structure and white. Most of the associated tourmaline has a peculiar, dark oily-green tint; and with many of the specimens a rose red earthy clay was associated. A partial analysis of the latter was published in Bulletin No. 9 of the U. S. Geological Survey, but a re-analysis seemed to be desirable and was therefore made by Mr. Riggs. The results are subjoined and show that the mineral approaches nearly to cimolite. The difference from the earlier analysis indicates a non-homogeneity of composition. As to the genesis of the clay, nothing can be said with certainty.

SiO ₂	66.86
Al ₂ O ₃	22.23
Fe ₂ O ₃	.47
FeO	.18
MnO	.07
CaO	.42
MgO	.33
Li ₂ O	.29
Na ₂ O	.46
K ₂ O	.18
H ₂ O	8.26
F	.06
	<hr/> 99.81

Formula, $\text{Al}_2\text{H}_4\text{Si}_6\text{O}_{18}$.

Auburn.—In the western part of this town, near the Minot line, there are two localities for lepidolite, less than half a mile apart. The one longest known is small and unimportant, but the other on the farm of G. C. Hatch, has been quite thoroughly developed.* It has yielded many gem tourmalines, mostly of the paler tints, some remarkable apatites, and perhaps the best crystallizations of lepidolite so far known. The latter mineral occurs in the ordinary purplish, coarsely granular form and also in extraordinary perfection as a border upon muscovite; the broad plates of the latter being practically encircled by aggregations of small crystals of the lithia mica. Some specimens of this type have also been found at Paris.

As it was hoped that this mode of occurrence of lepidolite might throw some light upon its genesis, three analyses of Auburn material were made by Mr. Riggs: first, of the com-

* See G. F. Kunz, this Journal, III, xxvii, 303.

mon granular variety, secondly of the border upon muscovite, and finally of the latter mineral from the center of the second specimen. The muscovite analysis came out as follows:

SiO ₂	44.48	Na ₂ O	2.41
Al ₂ O ₃	35.70	K ₂ O	9.77
Fe ₂ O ₃	1.09	H ₂ O	5.50
FeO	1.07	F	.72
MnO	trace		
CaO	.10		100.84
MgO	trace	Less oxygen -	.30
Li ₂ O	trace		100.54

About these results there is nothing unusual.

For convenience the analyses of lepidolite, all by Mr. Riggs, may be given in one table. The water in every case was determined directly by means of the Gooch tubulated crucible, and most of the lithia separations were effected by a new process, lately devised in this laboratory by Dr. F. A. Gooch, and shortly to be published in full. In the Auburn and Norway lepidolites, however, the old phosphate method was used. The fluorine estimations were all by the Berzelian process. In the figures given for (KRbCs)₂O, we have actual determinations based upon the amount of chlorine in the mixed chlorides, and not a mere computation as K₂O from the weight of the chloroplatinates. All important estimations were checked in duplicate, and the table of results represents mean values. Seven analyses were made, as follows:

Analyses of Lepidolite.

	A. Purple. Rumford.	B. Foliated. Paris.	C. Granular. Hebron.	D. Border. Auburn.	E. Granular. Auburn.	F. White. Norway.	G. Brown. Norway.
SiO ₂	51.52	50.92	48.80	49.62	51.11	49.52	50.17
Al ₂ O ₃	25.96	24.99	28.30	27.30	25.26	28.80	25.40
Fe ₂ O ₃	.31	.30	.29	.31	.20	.40	.87
FeO	undet	.23	.09	.07	.07	.24	.45
MnO	.20	trace	.08	.55	.17	.07	.23
CaO	.16	trace	.10	----	.12	.13	undet
MgO	.02	trace	.07	----	.01	.02	undet
Li ₂ O	4.90	4.20	4.49	4.34	4.98	3.87	4.03
Na ₂ O	1.06	2.11	.74	2.17	1.43	.13	13.40
(KRbCs) ₂ O	11.01	11.38	12.21	11.19	12.25	12.63	
H ₂ O	.95	1.96	1.73	1.52	.94	1.72	2.02
F	5.80	6.29	4.96	5.45	6.57	5.18	5.05
	101.89	102.38	101.86	102.52	103.11	102.71	101.62
Less oxygen	2.44	2.64	2.02	2.29	2.76	2.18	2.13
	99.45	99.74	99.84	100.23	100.35	100.53	99.49

As regards cæsium and rubidium the Maine lepidolites offer some points of difference. In the Rumford material an ordinary analysis shows neither; but a special examination upon

the alkalis from 150 grams of mineral gave a spectroscopic trace of rubidium. The Paris lepidolite showed traces of both metals, while the material from Hebron, Norway and Auburn was much richer. On these lepidolites approximate estimations were made, as follows:

	Hebron. Granular.	Norway. White.	Auburn. Granular.	Auburn. Border.
K ₂ O	11.44	8.82	10.51	8.03
Rb ₂ O }	.77	3.73	1.29	2.44
Cs ₂ O }		.08	.45	.72
	12.21	12.63	12.25	11.19

In these determinations the cæsium was separated as stannic chloride, and the other two metals were computed indirectly from the amount of chlorine in the mixed chlorides. The results have only a comparative value, and no sharp accuracy can be imputed to them. The methods of separation for the three metals are still far too defective.

The foregoing analyses of the Maine lepidolites, covering several distinct types of the mineral from five different localities, indicate a great constancy of composition. The essential identity of the mineral with that from Rozena and Cornwall is also shown by the several published analyses of the latter, while the lepidolite from Juschakova is slightly different, in that it contains some manganese replacing aluminum, higher fluorine and a little chlorine. In most of the analyses the water and fluorine appear to vary reciprocally, suggesting the ordinary replacement of the latter element by hydroxyl. With this slight assumption the formula for lepidolite may be written thus:

a formula which has long had general acceptance, but which now rests upon the surer basis of a wider range of analytical data. It corresponds to the following theoretical composition:

	Calculated.	Found.	(Riggs).
SiO ₂	49.18	48.80	to 51.52
Al ₂ O ₃	27.87	24.99	28.80
Li ₂ O	4.09	3.87	4.98
K ₂ O	12.81	(NaKRbCs) ₂ O	12.07
F	9.84	(FH ₂ O)	6.69
			8.25

Most of the variations are no greater than we should expect to find in material so difficult to secure in absolute purity as lepidolite. The granular structure of that species is peculiarly favorable to the presence of inclusions, as for example of albite, to which probable impurity some of the soda shown by the analyses may be due. The greatest difference is in the case of fluorine; although some of the foreign analyses, notably Rammelsberg's analysis of the Juschakova mineral, contain very nearly the full theoretical amount.

II. THE IRON-LITHIA MICAS OF CAPE ANN.

In the granite quarries of Rockport, Massachusetts, near the extremity of Cape Ann, there are occasional feldspathic veins which contain the rarer minerals danalite, cyrtolite, fergusonite, and amazon-stone, together with certain remarkable micas. One of the latter, cryophyllite, was described by Cooke in 1867;* who also analyzed an associated "lepidomelane," to which Dana afterward gave the name of annite. The vein from which Cooke obtained his material was long ago blasted away or covered up; but other veins of like nature are still accessible, and from one of them the micas examined in this laboratory were obtained. They were collected by the original discoverer of the locality, Mr. W. J. Knowlton of Boston, and were, to all appearance, identical with the micas described by Cooke. Among the specimens the two types were clearly recognizable; the one a dark-greenish-black lithia mica, cryophyllite; and the other a black, brilliant lepidomelane. In some specimens the cryophyllite formed a border upon the broad plates of the other mica, precisely as in the association of lepidolite and muscovite at Auburn. The resemblance in this particular was curiously striking; only the Rockport minerals are less conspicuous than those from Maine, since they lack the contrast of color which gives the latter their beauty.

The cryophyllite varies considerably in outward character, and three well marked types of it were analyzed by Mr. Riggs. They may be described as follows: A. Broadly-foliated, brilliant, blackish-green. B. Paler, dull-green, less lustrous, seemingly altered. C. An aggregation of minute six-sided prisms, dark green, almost granular in appearance, resembling some chlorite. Mean results are given.

	A.	B	C.	Cooke's Analysis.
SiO ₂	51.96	51.46	52.17	51.49
Al ₂ O ₃	16.89	16.22	16.39	16.77
Fe ₂ O ₃	2.63	2.21	4.11	1.97
FeO	6.32	7.63	5.99	7.98
MnO	.24	.06	.32	Mn ₂ O ₃ .34
CaO	.12	trace	trace	----
MgO	.03	.17	trace	.76
Li ₂ O	4.87	4.81	4.99	4.06
Na ₂ O	.87	.89	.63	trace
K ₂ O	10.70	10.65	10.48	13.15
H ₂ O	1.31	1.12	1.46	----
F	6.78	7.44	7.02	SiF ₄ 3.42
	102.72	102.66	103.56	99.94
Less oxygen,	2.86	3.11	2.95	
	99.86	99.55	100.61	

* This Journal, II, xliii, 217.

It will at once be seen that the three samples examined by Mr. Riggs were sensibly identical, and not far in composition from the material analyzed by Cooke. In the latter the fluorine amounted to 2.49 per cent, which is only about one-third of the figure found in this laboratory, while the silica, plus that represented in the SiF_4 , is sensibly higher. The integrity of the species, however, seems to be clear; and it cannot be identified with zinnwaldite. Empirically, as deduced from the new analyses, the formula of cryophyllite may be written

With the "lepidomelane" or annite, quite unexpected results were obtained. The material analyzed by Mr. Riggs was black, brilliant, broadly foliated, and apparently very pure. His figures are given in contrast with those published by Cooke.

	Riggs.	Cooke.
SiO_2	31.96	39.55
TiO_2	3.42	----
Al_2O_3	11.93	16.73
Fe_2O_3	8.06	12.07
FeO	30.35	17.48
MnO	.21	Mn_2O_3 .60
CaO	.23	----
MgO	.05	.62
Li_2O	trace	.59
Na_2O	1.54	trace
K_2O	8.46	10.66
H_2O	4.25	1.50
F	trace	SiF_4 .62
	<hr/> 100.46	<hr/> 100.42

It is at once evident that two entirely distinct micas are here represented, and the question is raised whether the Rockport granites may not contain a series of complex isomorphous mixtures. Cooke, indeed, pointed out the isomorphism of cryophyllite with his samples of annite, and showed that the lithia and fluorine in the latter were probably due to admixtures of the former. We now see that at least three micas are involved in the problem to be solved, and the difficulty of establishing definite formulæ is enormously increased. For the present, approximate formulæ only can be assigned to these micas, involving several assumptions, and representing probabilities rather than complete interpretations of the facts. If we unite the groups SiO_2 and TiO_2 in the new annite, and regard the ferric iron as belonging partly with the alumina, and partly as having been derived by oxidation from the ferrous state, we have the following general formulæ for the two analyses.

For the former, the equivalent of R'_8 is approximately $\text{K}_2\text{H}_2\text{Fe}''$, and for the latter it is $\text{K}_2\text{H}_4\text{Fe}_4$. These values correspond to the following percentage compositions.

	Cooke.	Riggs.
SiO_2	39.5	36.6
Al_2O_3	26.8	12.4
FeO	18.9	35.1
K_2O	12.4	11.5
H_2O	2.4	4.4
	<hr/> 100.00	<hr/> 100.00

Attention has already been called to the fact that some specimens of cryophyllite form borders upon plates of annite, precisely as the lepidolite of Auburn is arranged about nuclei of muscovite. It accordingly becomes quite probable that similar relations connect the two pairs of minerals; and upon these relations the formulæ so far deduced shed some light. In each case we have a mineral with metasilicate ratios implanted upon an orthosilicate; and a derivability of the one from the other is forcibly suggested. Structural analogies also appear; for in each couplet we have evidence of a common type of nucleus, which may be represented thus:

The development of complete structural formulæ from these nuclei is rendered difficult by our ignorance of the part which fluorine plays in such compounds. In the ordinary lepidolites it is easy to conceive of it as united with the aluminum in the univalent group AlF_2 ; but in the iron micæ and phlogopite a similar representation is unsatisfactory. A different solution of the problem is therefore to be sought, and it may be found by an application of the well-recognized principle that fluorine and hydroxyl can replace each other isomorphously.

If now we start from orthosilicic acid, $\text{Si}(\text{OH})_4$, and regard the hydroxyl groups as successively replaceable by atoms of fluorine, we can conceive of a series of acids ranging from $\text{Si}(\text{OH})_4$ to SiF_4 ; and with the aid of such a supposition many of the fluoriferous silicates may be rationally explained. For example, the acid $\text{SiF}(\text{OH})_3$ may be considered, and its nucleus, SiFO_3 , a trivalent residue, can be applied to the discussion of the lithia micæ. Upon this basis the formulæ for lepidolite and cryophyllite become curiously significant, especially when written in direct comparison with the structures assignable to muscovite and annite respectively. In every case the assump-

tion is made that double salts are to be represented as substitution derivatives of normal salts of similar type, and the results appear as follows:

To zinnwaldite, as represented by the published analyses, a probable structure is not easily assignable; and the variations in its composition suggest that the specimens examined have been mixtures of isomorphous compounds. It needs to be thoroughly reinvestigated, not only for itself, but also in its relations to whatever other micas may be associated with it. The matter of association can never, in the study of the micas, be safely neglected.

Although the formulæ herein assigned to annite and cryophyllite are to be regarded merely as trial hypotheses, there can be little doubt that the ratios between the sesquioxides and the silica are correctly given. These ratios are the ratios indicated by the published analyses of phlogopite; with which, therefore, rather than with ordinary lepidomelane, the Rockport micas are chemically to be classed. Zinnwaldite is already so classed by Tschermak and others, and cryophyllite falls easily into the same category. The subjoined formula fairly represents a typical phlogopite.