

GEOLOGINEN TUTKIMUSLAITOS

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N:o 149

**CONTRIBUTIONS TO THE KNOWLEDGE OF  
THE CHEMISTRY OF THE RADIOACTIVE  
MINERALS OF FINLAND**

**BY**

**LAURI LOKKA**

**WITH 7 FIGURES AND 35 TABLES IN TEXT**

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**HELSINKI**

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## PREFACE

This work deals with the chemistry of radioactive minerals found in Finland before the year 1939, excluding the radioactive accessory constituents occurring to a negligible degree in rocks. To my knowledge, no radioactive minerals have been found in Finland after 1939.

The minerals in question are: gadolinite, allanite, monazite, and the tantaloniobates nuolaitite, loranskite and wiikite. Some have been found in several varieties and as alteration products representing various stages of alteration. Of the occurrences three are in southwestern Finland: Lövböle in Kemiö, Åva in Brändö and Varala in Kangasala. Two are in the vicinity of the earlier (1940) boundary between Finland and Russia: Impilahti on the northern shore of Lake Laatokka, best known and most remarkable because of the occurrences of wiikites and certain other radioactive minerals found there, and Silmäpuolivaara in Salla, a new wiikite occurrence in East-Lapland. As the sixth, though not quite in the same sense as those previously mentioned, Ivalojoiki in North-Lapland may be mentioned (sand containing monazite). Several rock-minerals from these localities have also been analysed and also these analyses will be presented here.

The methods used in the analyses of these radioactive minerals are described in the respective places. The methods used in the new analyses are chiefly the same as I have used before, with the exception of some items, such as the determination of niobium and tantalum. In the new analyses of wiikite I have determined niobium photometrically (tantalum indirectly).

Since the mineral-analyses should be very carefully performed and accurate, more accurate than good rock-analyses, it is also logical and important that as accurate values as possible be used in the calculations and not approximate and obsolete molecular weights, which are sometimes still used. These can lead to considerable errors in the calculations.

The molecular proportions are calculated here to the fourth decimal place, using the latest and most accurate atomic weights. The tables

contain the molecular proportions multiplied by 10000. In some cases, when less accuracy suffices, the molecular proportions are multiplied by 1000 and the last decimal place neglected.

I am indebted to my wife, Mrs. Esteri Lokka, for having translated this work and to Mrs. Lily Björling for having kindly revised the English.

Geological Survey of Finland.

Lauri Lokka

Helsinki, January 1950.

## RADIOACTIVE MINERALS FROM SOUTHWESTERN FINLAND

In southwestern Finland, especially in its southern part, there are both on the mainland and in the archipelago several occurrences of minerals, which in the course of time have been examined and treated in many publications. In some of them also rarer minerals occur. The following deals with radioactive minerals. Two of these minerals have been found in this region, viz. gadolinite and allanite, the occurrences of which are as follows:

Gadolinite. Lövböle in Kemiö.

Allanite. Åva in Brändö.

Allanite. Varala in Kangasala.

### GADOLINITE FROM LÖVBÖLE, KEMIÖ

Lövböle is situated in the western part of the large isle of Kemiö (Kimito), abt. 35 km. to the S. S. E. of the town of Turku. Gadolinite was found in the occurrences of pegmatite from Lövböle in 1922 and it is the first discovery of gadolinite in Finland (Kranck 1924, p. 3). To my knowledge gadolinite has not later been found in Finland.

According to the investigations of Kranck, the following may be mentioned concerning the gadolinite of Lövböle. The colour is pitchy, the fracture glassy. Generally it occurs in the form of lumps, crystal faces are very seldom seen and they are badly formed. There are two varieties, a dark green and red-brown one, which may occur in the same crystal. In the main they are both isotropic (op. c., pp. 10—11).

Kranck has carried out an analysis (op. c., p. 14); of the first mentioned variety this is recorded in Table 1. He says of the substance used that it was only partly possible to separate the pyrite that occurred as impurity in it. The content of sulphur is also fairly high in the analysis. From his analysis Kranck has reduced such an amount of pyrite as corresponds to the amount of sulphur. The analysis is also given in this form in Table 1.

Of a similar variety of gadolinite, as analysed by Kranck, he kindly placed a sample at my disposal. My purpose was to try to ascertain whether it was possible to determine the age of this mineral, as, due to the low content of thorium, the content of lead could be supposed to be fairly low. Therefore determinations had to be made not only of the content of lead, which Kranck had not done in his analysis, but also of



Table 1. Gadolinite from Lövböle, Kemiö

	FeS <sub>2</sub> subtr., red. to 100		Mol. prop.	At. prop.	
	%	%			
SiO <sub>2</sub>	23.53	24.15	4021	Si	4021 = 2 × 2011
BeO	8.81	9.03	3609	Be	3609 = 2 × 1805
Y <sub>2</sub> O <sub>3</sub>	46.71	47.95	1918	Al	240
Ce <sub>2</sub> O <sub>3</sub>	2.82	2.96	90	Fe··	88
ThO <sub>2</sub>	0.60	0.61	23	Fe··	1709
Al <sub>2</sub> O <sub>3</sub>	1.20	1.22	120	Mg	5
Fe <sub>2</sub> O <sub>3</sub>	0.69	0.71	44	Ca	164
FeO	13.50	12.28	1709	Y	3836
MnO	tr.	—	—	Ce	180
MgO	0.02	0.02	5	Th	23
CaO	0.90	0.92	164		
Na <sub>2</sub> O	0.15	0.15			
S	0.88				
H <sub>2</sub> O	0.31				
	100.12	100.00			
Sp. gr.	4.208				
H.	c. 6.5				

Y<sub>2</sub>Fe Be<sub>2</sub>Si<sub>2</sub>O<sub>10</sub>

Anal. E. H. Kranck

that of thorium, as they ought, of course, to be determined for this purpose from the same powder. Since the chemical composition of the mineral in different samples may vary considerably, even when they are taken from the same occurrence, I carried out a total analysis in order to establish it in this case.

In preparing the substance for analysis, one could see that the pyrite occurred as impurity also in this sample, but evidently in lesser amount. The sample was crushed into fairly small grains and from these the purest were carefully selected, while endeavouring to exclude the pyrite. As Th and Pb in this case had each to be determined from different portions and many determinations of both had to be carried out and, moreover, each of them from several grammes of the powder, many tens of grammes of the powder were prepared from which all determinations were performed.

In order to gain greater accuracy in the determination of age, we carried out four determinations of Th and four of Pb. For each of the former determinations we used three and for each of the latter ten grammes of the powder. These determinations were performed chiefly according to Fenner (1928). The results are:

0.723, 0.728, 0.692 and 0.719, average 0.716 % ThO<sub>2</sub> = 0.629 % Th.  
0.0397, 0.0383, 0.0374 and 0.0421, average 0.0394 % PbO = 0.037 % Pb.

The determinations of thorium generally give fully consistent values also on those cases when fairly small amounts are in question. Thus also in this case the maximum difference between the values obtained is only abt. 5 %, which result may be considered quite normal. In the determinations of lead the maximum difference is over 10 %, but this cannot be

regarded as exceptionally great, where such a very low content of lead is involved as in this case.

The calculation of age is described in connection with Table 25, where the result is given.

The analysis in Table 2 is carried out according to standard methods. For the oxides  $\text{ThO}_2$  and  $\text{PbO}$  the above mentioned values are given in the table, but with two decimals.

Table 2. Gadolinite from Lövböle, Kemiö

	%	Mol. prop.	At. prop.	
$\text{SiO}_2$	24.29	4044	Si	4044 = $2 \times 2022$
$\text{TiO}_2$	none	—	Be	3725 = $2 \times 1863$
$\text{BeO}$	9.32	3725	Al	176
$\text{Y}_2\text{O}_3$	47.34	1894	Fe···	80
$\text{Ce}_2\text{O}_3$	2.57	71	Fe··	1797
$\text{ThO}_2$	0.72	27	Mg	7
$\text{Al}_2\text{O}_3$	0.90	88	Ca	109
$\text{Fe}_2\text{O}_3$	0.63	40	Y	3788
$\text{FeO}$	12.91	1797	Ce	142
$\text{MnO}$	tr	—	Th	27
$\text{MgO}$	0.03	7		
$\text{CaO}$	0.61	109		
$\text{Na}_2\text{O}$	0.08			
$\text{PbO}$	0.04			
S	0.09			
F	0.08			
$\text{H}_2\text{O}$	0.28			
	99.89			
—0 = $\text{F}_2$	0.03			
	99.86			
Sp. gr.	4.195			

Anal. Lauri Lokka

$\left. \begin{array}{l} \text{Fe} \cdots \cdots 80 \\ \text{Fe} \cdots \cdots 1797 \end{array} \right\} 2060 = \text{Y}$

$\left. \begin{array}{l} \text{Mg} \cdots \cdots 7 \\ \text{Ca} \cdots \cdots 109 \\ \text{Y} \cdots \cdots 3788 \\ \text{Ce} \cdots \cdots 142 \\ \text{Th} \cdots \cdots 27 \end{array} \right\} 4066 = \text{X} = 2 \times 2033$

$\text{Y}_2\text{FeBe}_2\text{Si}_2\text{O}_{10}$

The analyses in Tables 1 and 2 are very much alike, if the pyrite is reduced from the former. As far as may be concluded from these two analyses, the chemical composition of the gadolinite of Lövböle does not vary much, except in the content of sulphur (pyrite).

Table 3 shows the analysis of the first gadolinite found in Canada (Ellsworth 1932, p. 97). It is taken here for the sake of comparison and as an example of a gadolinite analysis uncommonly poor in iron.

When comparing the chief constituents of the analyses of the gadolinite of Lövböle with the corresponding values in older and newer analyses of gadolinite, we get the following results.

$\text{SiO}_2$ , generally 23—24 %, sometimes somewhat more, as in the analysis in Table 3, has a normal value in the gadolinite of Lövböle.

$\text{BeO}$  also occurs in quite a normal amount, generally 9—10 %. Some values as low as 5—7 % may partly be due to errors in analysis.

With regard to the amount of rare earths — generally 50—52 %, a

Table 3. Gadolinite from Loughborough, Ontario

		Mol.		At.		Anal. H. V. Ellsworth
	%	prop.		prop.		
SiO <sub>2</sub>	25.97	4324	Si	4324	= 2 × 2162	
BeO	10.29	4113	Be	4113	= 2 × 2057	
Y <sub>2</sub> O <sub>3</sub>	46.47	1860	Al	62	} 1467 = Y	
Ce <sub>2</sub> O <sub>3</sub>	2.85	86	Fe··	294		
ThO <sub>2</sub>	0.14	5	Fe··	810		
Al <sub>2</sub> O <sub>3</sub>	0.32	31	Mg	136		
Fe <sub>2</sub> O <sub>3</sub>	2.34	147	Mn	165		
FeO	5.82	810	Ca	421	} 4318 = X = 2 × 2159	
MnO	1.17	165	Y	3720		
MgO	0.55	136	Ce	172		
CaO	2.36	421	Th	5		
Na <sub>2</sub> O } K <sub>2</sub> O } H <sub>2</sub> O }	0.09 1.23					
	<u>99.60</u>					
Sp. gr.	4.101					

limit with few and only slight exceptions — the two above mentioned analyses are quite normal.

If the iron, which in the analyses of gadolinite is for the most part FeO, is calculated totally as FeO, a very normal value of 12—14 % is obtained. The iron-content of the gadolinite of Lövböle agrees well with these limits. Higher and lower values are rare, some of them are very exceptional, such as the content of iron in analysis 3.

In the calculation of the analyses no attention is paid to the alkalis, S, F and H<sub>2</sub>O, their amounts are of no importance. Th, being near the rare earths, is indicated in group X.

In the calculations of the gadolinite of Lövbele the corresponding values of different groups are fairly consistent. The chemical composition of each mineral agrees well with the formula



Since yttrium earths almost exclusively form group X and Fe likewise almost exclusively group Y, the formula may be written



The analysis in Table 3 — although, without doubt, correctly carried out — is a good instance of the fact that the chemical composition of the mineral cannot be given with that formula which in most cases is the most suitable. The calculation of the analysis shows that the group Y is abt. 30 % lower as to its value than the corresponding fairly consistent values of the other groups. The unusually great difference is chiefly due to the uncommonly low iron-content, as already stated.

This gadolinite is not, however, unique in this respect, as there are even such where the calculations of the analyses give still worse results; this being chiefly due to the exceptionally low content of iron or beryllium.

## ALLANITE FROM ÅVA, BRÄNDÖ

In southwestern Finland in the archipelago between the mainland and Åland lies the parish of Brändö, to which belong a great many larger (2—3 km. long) and smaller isles. The isle of Åva, one of the largest, where the allanite in question occurs, is situated about 65 km. to the west of the town of Turku.

Red porphyritic granite is the prevailing rock in the occurrence of the allanite in Åva, but there is also found a rock similar to the typical rapakivi. The allanite occurs in pegmatite as larger and smaller lustrous, jet black lumps. The interface between the mineral and the surrounding rock is generally very distinct. The influence of this weakly radioactive mineral upon the surrounding rock cannot be seen.

The sharp and distinct interface of the mineral and the unaltered appearance of the surrounding rock support the opinion that this mineral is fresh and its black colour also indicates this. These qualities do not, however, belong only to the unaltered allanite. An altered allanite, changed in colour from black to brown, may also be limited sharply to the surrounding rock, where the feldspar may have its own colour near the interface. However, the feldspar has frequently altered for a somewhat reddish colour in the contact of the surrounding rock, whether the limit be sharp or indistinct.

No signs of alteration could be seen in the samples of this allanite. It must also be mentioned that even allanite in crystals has been found in Åva. The mineral is isotropic, hardness *abt.* 5 and specific gravity 3.487, which value is very common amongst allanites. No impurities could be seen in the substance, which was prepared from one piece of the mineral, a fairly large quantity, when considering the amounts of powder required for the age determination. Colour of powder is dark grey.

Also here four determination of both thorium and lead were performed for age determination. In the case of the former 3 g of the powder of analysis was used for each determination and in the case of the latter 10 g for each. The determinations have been performed as before (gadolinite) chiefly according to Fenner (1928) with the following results: 0.519, 0.540, 0.548 and 0.529 %, average 0.534 %  $\text{ThO}_2 = 0.467$  % Th. 0.0290, 0.0278, 0.0315 and 0.0279 %, average 0.0291 %  $\text{PbO} = 0.027$  % Pb.

These different determinations of thorium and lead differ from each other to about the same extent as do the afore mentioned corresponding determinations of gadolinite.

The total analysis in Table 4 is carried out as usual. The values mentioned before are given to the oxides  $\text{ThO}_2$  and  $\text{PbO}$ , but with two decimals. The analysis shows that this allanite is very common as regards its chemical composition. Some facts may, however, be pointed out.  $\text{TiO}_2$ , which in a way belongs to the constituents of allanite, though also

Table 4. Allanite from Åva, Brändö

	%	Mol. prop.		At. prop.	
SiO <sub>2</sub>	31.26	5205	Si	5205 = 3×1735	
TiO <sub>2</sub>	0.71	89	Al	2688	}
Y <sub>2</sub> O <sub>3</sub>	0.68	27	Ti	89	
Ce <sub>2</sub> O <sub>3</sub>	21.03	637	Fe <sup>···</sup>	440	} 5159 = Y = 3×1720
ThO <sub>2</sub>	0.53	20	Fe <sup>··</sup>	1604	
Al <sub>2</sub> O <sub>3</sub>	13.70	1344	Mg	263	}
Fe <sub>2</sub> O <sub>3</sub>	3.52	220	Mn	75	
FeO	11.52	1604	Ca	2152	} 3500 = X = 2×1750
MnO	0.53	75	Y	54	
MgO	1.06	263	Ce	1274	
CaO	12.07	2152	Th	20	
Na <sub>2</sub> O	0.29				
K <sub>2</sub> O	0.06				
PbO	0.03				
S	none				
F	0.21				
CO <sub>2</sub>	none				
H <sub>2</sub> O+	2.53				
H <sub>2</sub> O—	0.04				
	99.77				
—O = F <sub>2</sub>	0.09				
	99.68				
					X <sub>2</sub> Y <sub>3</sub> Si <sub>3</sub> (O, OH, F) <sub>13</sub>
Sp. gr.	3.487				
H.	c. 5.0				

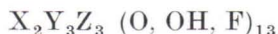
titanium-free allanites are found, is here represented in moderate amount. The same can be said of fluorine, which for its part compensates the water.

The opinion that the chemical composition of this mineral has not changed remarkably, is supported by the following.

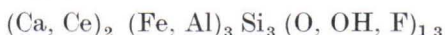
The iron is chiefly FeO, as should be the case with fresh allanite. The water-content of fresh allanite changes somewhat 1.5—3 %. The water-content of this allanite is also between these limits, viz. 2.57 %. The giving up of only 0.04 % of water at 110°C shows that the water is primary. As a surer proof may, however, be considered that it has given off only 0.19 % of water at so high a temperature as 260°. The difference 0.19—0.04 = 0.15 % corresponds only to 5.9 % of total water which has been given off at temperatures over 110°C. This is an unusually low value and a sure proof that the mineral does not contain secondary water. At the determinations of dehydration performed similarly, fresh minerals, allanites as well as others, may give up at 260° a double amount of water (compared with the above mentioned) and even somewhat more.

The calculation of the analysis in Table 4 shows that in the different groups the values, which theoretically are equal, are so consistent that the greatest difference between them is only 1.7 %, which result is unusually good.

The chemical composition of the allanite of Åva may thus very well be given by the general formula of the allanite



or by a special formula



where the chief constituents of the groups are indicated.

In this case Si alone without Al forms group X, which is not common in calculations of the allanites and other silicates.

Earlier this mineral has erroneously been regarded as *gadolinite*, and no wonder, as it resembles *gadolinite* in appearance, though it recalls also some other radioactive minerals. But it is strange that it has been called *gadolinite* also in the case of an analysis from which may be concluded that not a *gadolinite* but an *allanite*<sup>1</sup> is in question.

This mineral may still be found under name of *gadolinite* in the collections of institutions and of private persons in Finland and probably also in other countries. To my knowledge *gadolinite* has not been found in Åva in the parish of Brändö.

Rankama (1944, p. 32) has spectrographically established that the »*gadolinite*», Åva, Brändö, does not contain tantalum. The erroneous name of the mineral is perhaps due to the faulty name in the collection he has used.

#### ALLANITE FROM VARALA, KANGASALA

The feldspar quarry of Varala in the parish of Kangasala in southwestern Finland is abt. 30 km. to the E. S. E. of the town of Tampere. Fresh allanite has been found there and many different products of alteration of same. The allanite and other minerals of Varala I have dealt with earlier (1935, 1943); the following shorter treatment of allanite is chiefly founded on this earlier work. We may, however, first make mention of the other minerals of Varala.

#### Rock forming minerals.

Besides allanite the following minerals have been found in Varala: Quartz, potash and soda feldspar, biotite, chlorite, seladonite, beidellite, spessartite and pyrite. Referring to my earlier treatment of these mi-

<sup>1</sup> In the »*Teknillinen Aikakauslehti*» 1927, Nos. 5—6, p. 320 (Helsinki), the *gadolinite*, Åva, Brändö, is mentioned and an analysis of it is given. For the correction of the name it may be said that one may clearly see from the analysis — in spite of its defectiveness and faultiness — that this mineral is not a *gadolinite* but an *allanite*; as in the analysis no mention is made of beryllium, and such constituents as rare earths (21.61 %),  $Al_2O_3$  (13.48 %) and CaO (10.86 %) are not as to their amount even nearly suited to the values of *gadolinite*, but are nearest to those of *allanite*. Among the constituents are also mentioned  $(Ta, Nb)_2O_5 = 1.55$  %. Specific gravity 3.2683.

nerals (1935, pp. 6—8, 24—38 and 1943, pp. 10—11, 17—19) they will here be mentioned shortly and the analyses given (Table 5) without calculations; in the original publication they are given with calculations. The brackets refer to the original publication.

Table 5. Rockforming Minerals from Varala, Kangasala

	Anal. Lauri Lokka									
	I	II	III	IV	V	VI	VII	VIII	IX	X
SiO <sub>2</sub> . . . . .	65.76	66.30	35.88	33.98	35.15	34.38	36.58	39.44	33.22	38.01
TiO <sub>2</sub> . . . . .	none	none	none	2.23	2.58	2.43	0.72	none	none	none
Al <sub>2</sub> O <sub>3</sub> . . . . .	18.66	20.69	19.52	18.32	16.45	17.04	17.39	17.90	21.78	21.10
Fe <sub>2</sub> O <sub>3</sub> . . . . .	0.20	0.21	6.48	5.95	7.40	11.13	19.08	1.40	15.69	23.72
FeO . . . . .	none	none	25.92	22.63	20.02	16.55	9.94	20.25	10.32	none
MnO . . . . .	tr.	tr.	0.35	0.46	0.38	0.37	0.44	0.35	0.31	0.23
MgO . . . . .	0.04	0.05	0.85	2.83	3.87	3.60	1.36	11.54	2.87	1.23
CaO . . . . .	0.15	1.43	0.07	0.07	0.10	0.15	0.38	0.05	0.83	0.75
Na <sub>2</sub> O . . . . .	3.69	9.55	0.54	0.92	0.67	0.78	0.60	0.26	0.29	0.19
K <sub>2</sub> O . . . . .	11.12	1.68	3.79	7.45	7.71	7.86	5.69	0.09	1.34	1.35
F . . . . .			0.07	0.49	0.31	0.38	0.13	0.06		
H <sub>2</sub> O+ . . . . .	0.12	0.13	6.10	3.92	4.70	4.72	5.85	8.57	8.85	7.70
H <sub>2</sub> O— . . . . .	0.18	0.16	0.65	0.85	1.11	1.11	2.19	0.46	4.72	5.52
	99.92	100.20	100.22	100.10	100.45	100.50	100.35	100.37	100.22	99.80
—O = F <sub>2</sub>			0.03	0.21	0.13	0.16	0.05	0.03		
			100.19	99.89	100.32	100.34	100.30	100.34		
Sp. gr. . . . .			3.033	3.021	2.935	2.916	2.852		2.742	2.564

- I. Potash feldspar, fresh (1935, p. 7).
- II. Soda feldspar, » »
- III. Biotite, fresh, jet black, lustrous, fineflaky (1935, p. 29).
- IV. Biotite, fresh, black, lustrous, flakes somewhat larger (1935, p. 29).
- V. Biotite, fresh, large flakes, somewhat bent, the black colour of the flakes has a weak greyish tinge (1935, p. 29).
- VI. Biotite, fresh, much like the former (1935, p. 29).
- VII. Biotite, somewhat altered, the flakes much smaller than those of the two lastmentioned (1935, p. 29).
- VIII. Chlorite, iron-rich, fresh, green (1943, p. 17).
- IX. Seladonite-like, green, secondary mineral. An alteration and decomposition product of the biotite III-like mineral (1935, p. 35).
- X. Beidellite, iron-rich, almost cinnabar red, omorphous, secondary mineral. A more complete alteration and decomposition product of the former (1935, p. 35).

Referring to the more detailed presentation of these minerals in the original publication some points may nevertheless be noticed also in this connection.

It is remarkable that the other minerals in Table 5 are titanium-free, except the large flaky biotites (IV—VII), of which the first three (IV—VI)

are titanium-bearing to that amount which is usual in the biotites of granitic rocks.

In analysis III the alkali-content is exceptionally low, corresponding only to about half of the theoretic amount, and the content of water is unusually high. It could be supposed that the mineral is secondary and therefore its water-content has increased abundantly while at the same time half of the alkalies has been washed away. Such is not the case, however, because only 6.4 % of the water-content ( $H_2O+ = 6.10\%$ ) is lost in so high a temperature as  $260^\circ$ , which is an uncommonly low value in the case of biotites and of minerals in general, and also a reliable proof that this biotite does not contain secondary water. »Dieser Fall zeigt, wie ungewöhnlich niedrig der Alkaligehalt und wie hoch der Wassergehalt ( $H_2O+$ ) auch bei einem frischen Biotit sein kann, wahrscheinlich infolge der primären Unvollkommenheit der Kristalle» (Lokka 1943, p. 10).

#### On allanite in general.

This mineral occurred in Varala both as prismatic crystals and as individuals of indefinite habit. Particular attention was paid to its occurrence in spots of a strange shape on the walls of the quarry. From the centres of the spots, which were of different sizes, irregular branches different as to length and shape (Fig. 1) ran in several direction. The



Fig. 1. Allanite from Varala. Black=fresh, black allanite; dark grey=altered, brown allanite; light grey=feldspar. Nat. size.



greatest dimension of such spots could be about 30 cm. They were either quite uni-coloured, black or reddish brown, or multicoloured, when the centre usually was black and different colours varied irregularly in the branches. The limit between black and brown could be clear or the colour changed to another little by little.

The boundary between the black, fresh allanite and the surrounding rock was usually clear. Sometimes it was more indistinct and generally the face of the feldspar was then, owing to the allanite, changed into more reddish ( $\text{Fe}_2\text{O}_3$ ). Also the reddish brown allanite and the changed allanite of other colours too could at these spots be sharply bounded against the feldspar, the colour of the face of which was unchanged. However, the face of the feldspar was often reddish and then the colour could reach deeper than in the case of fresh allanites.

#### Fresh allanite.

Jet black, columnar, dark grey powder. The analysis published earlier by the writer (1935, p. 12) is given in Table 6. It shows that the mineral is fresh. The ratio  $\text{Fe}_2\text{O}_3 : \text{FeO}$  is small, as should be the case with fresh allanite.  $\text{H}_2\text{O}+ = 1.74\%$  belongs to the lowest water-content of fresh allanites and is due to secondary water not being found in this mineral. At  $260^\circ$  it has given off  $11.5\%$  of water, which is a double value compared

Table 6. Fresh Allanite from Varala, Kangasala

				Anal. Lauri Lokka	
	%	Mol. prop.		At. prop.	
$\text{SiO}_2$	30.86	5138	Si	5138	} 5192 = Z = 3 \times 1731
$\text{TiO}_2$	none	—		54	
$\text{Y}_2\text{O}_3$	2.17	87	Al	3 038 <	} 5191 = Y = 3 \times 1730
$\text{Ce}_2\text{O}_3$	20.88	633		2984	
$\text{ThO}_2$	2.04	77	Fe···	300	} 3317 = X = 2 \times 1659
$\text{Al}_2\text{O}_3$	15.48	1519	Fe··	1823	
$\text{Fe}_2\text{O}_3$	2.39	150	Mg	84	
FeO	13.10	1823	Mn	464	
MnO	3.29	464	Ca	1336	
MgO	0.34	84	Y	174	
CaO	7.49	1336	Ce	1266	
BaO	none		Th	77	
$\text{Na}_2\text{O}$	0.16				
$\text{K}_2\text{O}$	0.09				
$\text{ZrO}_2$	none				
PbO	0.114				
S	none				
F	0.06				
$\text{CO}_2$	none				
$\text{H}_2\text{O}+$	1.74				
$\text{H}_2\text{O}-$	0.11				
	<u>100.314</u>				
$-\text{O} = \text{F}_2$	<u>0.03</u>				
	100.284				
Sp. gr.	3.521				
H.	c. 5.0				

$\text{X}_2\text{Y}_3\text{Z}_3 (\text{O}, \text{OH}, \text{F})_{13}$

with the corresponding low value (5.9 %) of the allanite of Åva, but quite common as the value of fresh allanite.

The content of thorium and manganese may specially be mentioned.

There are both thorium-free and thorium-bearing allanites, which often contain only some tenths %  $\text{ThO}_2$ , such as for instance the allanite of Åva (Table 4). The  $\text{ThO}_2$ -content seldom rises to 2 % and to my knowledge only in some few cases beyond it (see Lokka 1935, p. 22). Of these highest values the next highest is 3.48 %  $\text{ThO}_2$  and the highest of all that of a Madagascar allanite analysed by F. Pisani, the  $\text{ThO}_2$ -content of which, 5.60 %, has been known a long time (Lacroix 1912, p. 232). Thus the allanite of Varala (2.04 %  $\text{ThO}_2$ ) belongs to the most thorium-rich allanites, as I have mentioned earlier (1935, p. 23).

The content of manganese is also very subject to variation by allanites. Earlier I have mentioned, in accordance with my then knowledge, that the highest manganese-content of allanites was about 2.50 %  $\text{MnO}$ ; thus that of the allanite of Varala, 3.29 %  $\text{MnO}$ , was to be regarded as the highest manganese-content of allanites (1935, p. 22). Afterwards, however, I had the opportunity to establish that some years before an analysis of West Australian allanite had been performed by D. G. Murray, where the content of manganese was remarkably higher, 4.48 %  $\text{MnO}$  (Simpson 1931, p. 137). This analysis is given in Table 7. A higher value is not mentioned in the literature, and even if such may be mentioned, the fresh allanite of Varala nevertheless belongs to the most manganese-rich allanites.

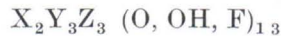
The high manganese-content of the above mentioned Australian allanite and that of Varala are of great importance, which is clearly shown by the calculations in Tables 6 and 7.

Table 7. Allanite from Woodstock, N. W. Division, W. Australia

		Mol.		At.	
	%	prop.		prop.	
$\text{SiO}_2$	30.60	5095	Si	5095	$= 3 \times 1698$
$\text{TiO}_2$	0.36	45	Al	3174	} 5025 = Y = $3 \times 1675$
$(\text{Y, Er})_2\text{O}_3$	2.22	89	Ti	45	
$\text{Ce}_2\text{O}_3$	19.36	586	Fe $\cdots$	440	
$\text{La}_2\text{O}_3$	none	—	Fe $\cdots$	1197	
$\text{DiO}_2$	none	—	Mg	114	} 3350 = X = $2 \times 1675$
$\text{ThO}_2$	1.02	39		55	
$\text{Al}_2\text{O}_3$	16.18	1587	Mn	632	<
$\text{Fe}_2\text{O}_3$	3.52	220		577	} 3350 = X = $2 \times 1675$
$\text{FeO}$	8.60	1197	Ca	1384	
$\text{MnO}$	4.48	632	Y	178	
$\text{MgO}$	0.46	114	Ce	1172	
$\text{CaO}$	7.76	1384	Th	39	
$(\text{Na, K})_2\text{O}$	none				
$\text{H}_2\text{O}^+$	5.07				
$\text{H}_2\text{O}^-$	0.22				
	<u>99.85</u>				
Sp. gr.	3.52		$\text{X}_2\text{Y}_3\text{Si}_3(\text{O, OH})_{13}$		

Amounts of rare earths are very common in these allanites, but the CaO-content looks rather low. As may be concluded from the amount of rare earths, they should in both analyses be approximately about 11 %, accordingly over 3 % higher than shown by the analyses. These deficiencies are, substituted by the manganese, which as to ionic radius in the calculation of the analysis may be recorded as belonging either to the group X or Y. In one analysis (Table 6) the deficient calcium is substituted by manganese to its total amount and in the other (Table 7) to the largest part of it, so that only an insignificant part of the manganese falls into group Y. It is uncommon that in the analyses of allanite manganese is found as substitute for such large amounts of calcium as in these two analyses.

The analysis of the allanite of Varala may best be represented by the general formula of the allanite



or when giving the chief constituents, by the special formula



and so also the analysis of the very similar allanite of Woodstock, but or course without fluorine.

#### Altered allanites.

They are of various kinds, both as to colour and degree of alteration. In Table 8 we see the analyses of five such alteration products of allanite published before (Lokka 1935, p. 17).

Table 8. Altered Allanites from Varala, Kangasala

	Anal. Lauri Lokka				
	I	II	III	IV	V
SiO <sub>2</sub>	23.14	25.23	22.43	28.49	24.99
TiO <sub>2</sub>	none	none	none	none	none
Y <sub>2</sub> O <sub>3</sub>	3.24	2.75	2.33	2.50	2.69
Ce <sub>2</sub> O <sub>3</sub>	13.40	16.24	19.74	21.70	20.59
ThO <sub>2</sub>	1.98	2.01	2.45	2.39	2.56
Al <sub>2</sub> O <sub>3</sub>	18.18	18.75	17.50	20.58	17.11
Fe <sub>2</sub> O <sub>3</sub>	15.60	3.61	9.98	2.99	7.20
FeO	none	2.51	3.59	none	none
MnO	0.26	0.19	0.22	0.23	0.12
MgO	0.95	3.19	1.94	0.89	2.28
CaO	2.54	2.09	0.83	0.65	0.71
Na <sub>2</sub> O	0.70	0.62	0.51	0.95	0.85
K <sub>2</sub> O	1.08	0.95	0.97	4.46	0.62
CO <sub>2</sub>	5.13	6.16	5.89	6.69	5.84
H <sub>2</sub> O+	10.23	9.96	9.06	5.99	9.70
H <sub>2</sub> O—	3.63	5.61	2.39	1.28	4.98
	100.06	99.87	99.83	99.79	100.24
Sp. gr.	2.975	2.678	3.099		3.011
H.	3—4	3—4	4—5	4—5	c. 3

- I. Altered allanite, reddish brown.
- II. » » inclusion in the foregoing, bluish grey.
- III. » » prismatic, 5 cm long broken crystal, almost black. The unbroken end light brown. The analysis is made from the black broken end.
- IV. The light brown end of the preceding crystal.
- V. Weathered, 15 cm. long, 3—4 cm. thick, rounded allanite crystal, broken at each end.

The alteration of these allanites is clearly shown by the analyses. The water-content is very high and they give up considerable water already at 110° (H<sub>2</sub>O—). At 260° H<sub>2</sub>O+ has given off already 26—38 %, corresponding to 39—53 % of the total water. These are certain indications of the alteration, which is confirmed also by the following facts. CO<sub>2</sub> does not belong to fresh allanites, but we find it in these allanites and even very abundantly. The alkali-content of fresh allanite is insignificant, but in these allanites we find alkalies in considerable amount. These changes are caused by the circulating water, containing CO<sub>2</sub> and alkalies, and it has also brought about that many constituents have either increased or decreased in amount.

These five alteration products of allanites are titanium-free, as well as all the other minerals of Varala (Tables 5 and 6), except four biotites, the titanium-content of which has been mentioned above. It seems as though the titanium in Varala has concentrated only in biotites, and not even in all of these.

In allanites ThO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> are the most resistant constituents of the alteration products, each being as to amount fairly the same in all five analyses. No remarkable changes have occurred in ThO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>. In one case (Table 8, Anal. IV) Al<sub>2</sub>O<sub>3</sub> has probably increased from its original amount through the alkali-aluminium-silicate immigration, when alkalies have come very abundantly into the mineral — at one end of it. Rare earths are probably found in all the five alteration products in original amount.

As is well known, the iron of fresh allanite is only to a small part Fe<sub>2</sub>O<sub>3</sub>. Since the iron is totally oxidized in three of the five altered allanites and in two of them for the most part, also this indicates a far-going alteration. As both FeO and Fe<sub>2</sub>O<sub>3</sub> dissolve somewhat in water containing CO<sub>2</sub>, a part of the iron has probably leached away as easily soluble bicarbonate. In connection with a more abundant alkali-immigration (Anal. IV) it is likely that most of the iron has leached away.

It would be strange, if these altered allanites also originally had been so poor in manganese, as the analyses show, because in the same occurrence the manganese-content of fresh allanite is unusually high, 3.29 % MnO. It may be regarded as most probable that from the manganese, which

is more easily soluble than iron, the main part of the original amount has leached away and also in this case in the form of bicarbonate.

The very low CaO-amounts, which most likely correspond to only a part of the original amount, are also results of the alteration and the leaching. Evidently MgO has also originally been higher as to its amount in these allanites than in fresh, and by the alteration of the mineral also part of it may have leached away.

## RADIOACTIVE MINERALS FROM IMPILAHTI

In the preceding occurrences of radioactive minerals in S. W. Finland these minerals are one-sidedly represented, one mineral in each of three localities. Instead the radioactive minerals are much more many-sidedly represented in Impilahti—belonged earlier to Finland, now to Russia—on the north coast of Lake Laatokka in some pegmatite occurrences, which for a long time have been known for their rarer minerals.

The occurrences in Impilahti, where such minerals are found, are Hunttila, Lokansaari, Nuolainniemi and Mursula. (Fig. 2 and 3). Both thorium and uranium minerals have been found there. The thorium minerals are: allanite, monazite, nuolaite and loranskite. In all four localities the uranium mineral wiikite is found as the chief radioactive mineral containing also some thorium.

According to Trüstedt (1907) the pegmatite occurrences in Impilahti are of different ages. Ramsay (Ramsay and Zilliacus 1897, p. 59) divides those four occurrences according to their mineralogical composition in to two chief types.

To the type of Hunttila belong Hunttila, Lokansaari and Nuolainniemi. Their minerals are quartz, red microcline, red oligoclase, biotite, magnetite and some minerals containing rare earths, among them monazite and wiikite.

Ramsay does not particularly mention the allanite, but also this mineral containing rare earths belongs to the minerals of the type of Hunttila, as established later. Besides, the later discovered titanite and spessartite must also be mentioned.

To the type of Mursula belongs Mursula, the minerals of which are quartz, light red, light grey or white microcline, light obligoclase, muscovite, black tourmaline and garnet.

Some minerals, which have become known later, must be added to the minerals also of this type. First wiikite has to be mentioned; it has probably been found in Mursula very early (Lokka 1928, p. 5, footnote 3). Later also monazite and ilmenite have been found there.

More or less complete analyses have been carried out, we may say, of almost all the radioactive minerals of Impilahti, their varieties and

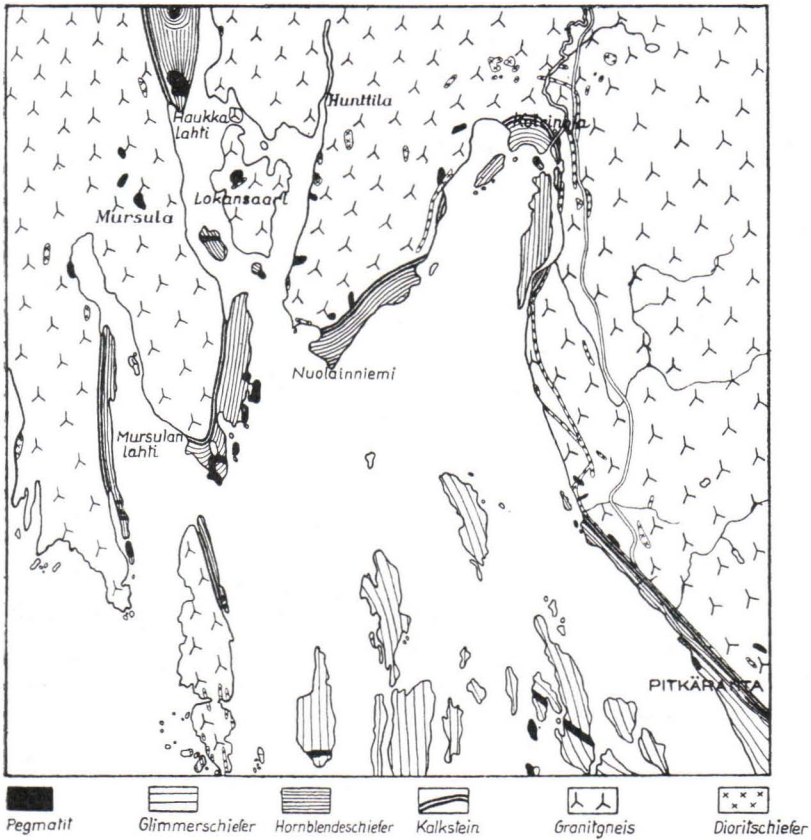


Fig. 2. The pegmatites of Impilahti. 1:80 000. According to Trüstedt.



Fig. 3. Mursulanniemi and Nuolainniemi, Impilahti.  
Photo by Erkki Kanervo.

alteration products. In the following these minerals are listed according to their localities, and those non-radioactive minerals of which total analyses are made are also mentioned. Only Impilahti, without particulars, has unfortunately been stated as the locality for some minerals, according to which they also are given in the list.

Hunntila. Wiikite, Spessartite.

Lokansaari. Wiikite, Monazite.

Nuolanniemi. Wiikite, Nuolaite, Allanite, Titanite.

Mursula. Wiikite, Monazite, Ilmenite.

Impilahti. Wiikite, Loranskite, Allanite.

The non-radioactive (rockforming) minerals of Impilahti of which total analyses have been made will first be treated shortly, then the thorium minerals and lastly the uranium mineral wiikite with its different types.

#### ROCKFORMING MINERALS

The analyses of the three rockforming minerals, ilmenite, titanite and spessartite, found in the last mentioned occurrences, are given in Table 9. For comparison also the analysis of the ilmenite of Leppäkylä has been given in this table. Leppäkylä is also in the parish of Impilahti, but somewhat outside the afore mentioned occurrences.

Table 9. Rockforming Minerals from Impilahti

	I	II	III	IV
SiO <sub>2</sub>	0.96	1.36	27.80	36.61
TiO <sub>2</sub>	50.00	50.00	35.71	0.49
Al <sub>2</sub> O <sub>3</sub>	0.22	0.31	1.96 <sup>1)</sup>	21.28
V <sub>2</sub> O <sub>5</sub>	0.044	0.044		
Fe <sub>2</sub> O <sub>3</sub>	4.10	5.11	1.91	0.44
FeO	43.55	42.56	0.69	17.73
MnO	0.98	0.52		21.93
MgO	0.09	0.23		0.07
CaO	none	none	24.18	1.56
Alk.				tr.
H <sub>2</sub> O+				0.11
H <sub>2</sub> O—				0.04
Loss on ign.			3.40	
	<u>99.944</u>	<u>100.134</u>	<u>100.32</u> <sup>2)</sup>	<u>100.26</u>

<sup>1</sup> Incl. the lanthanide oxides from Pr<sub>2</sub>O<sub>3</sub> to Lu<sub>2</sub>O<sub>3</sub>.

<sup>2</sup> Incl. spectrogr. determ.: Ta<sub>2</sub>O<sub>5</sub> = 1.75 (Th. G. Sahama); Nb<sub>2</sub>O<sub>5</sub> = 1.60, V<sub>2</sub>O<sub>5</sub> = 0.05, ZrO<sub>2</sub> = 0.06, Sc<sub>2</sub>O<sub>3</sub> = 0.03, Y<sub>2</sub>O<sub>3</sub> = 0.96, La<sub>2</sub>O<sub>3</sub> = 0.01, Ce<sub>2</sub>O<sub>3</sub> = 0.19, SrO = 0.01 and BaO = 0.01 (Oiva Joensuu). Sum = 4.67.

Sp. gr. I = 4.723, III = 3.591

I. Ilmenite from Mursula, Impilahti. Anal. Pentti Ojanperä.

II. » » Leppäkylä, » » » »

III. Zoned titanite from Nuolanniemi, Impilahti. Anal. Oleg v. Knorring (Sahama 1946, p. 117).

IV. Spessartite from Hunntila, Impilahti. Anal. Lauri Lokka (Lokka 1943, p. 38).

### Ilmenite, Mursula.

The chemical composition of this mineral, found in 1935, approaches the theoretical composition of ilmenite, no uncommon case in that respect. The analysis of the ilmenite of Leppäkylä is very similar,  $V_2O_3$ -amounts are quite the same. MnO-amounts are certainly not equal but they belong, nevertheless to the same class and are both considerable as manganese-content of ilmenite. The ilmenite of Mursula belongs to the most manganese-rich minerals of Impilahti.

### Titanite, Nuolainniemi.

Borgström points out (1910, p. 1531) that the specific gravity 3.591, which he has determined, is, as that of titanite, unusually high; he thinks probable that the mineral contains rare earths, all the more because it occurs associated with wiikite. Besides he mentions (p. 1531) that the mineral contains, according to the determination by G. Eberhard, only traces of scandium, but yttrium rather abundently.

This mineral has been examined later also by Sahama (1946), who had at his disposal a fairly large crystal individuum. He says of this i. a.: »The titante sample, representing a single crystal individuum, shows a marked zoned structure where a dark brown kernel is surrounded by alternating dark and light zones in regular arrangement.» Of the microscopic examination Sahama says: »— — — the brown kernel of the crystal contained numerous almost opaque inclusions. It is very possible that these inclusions are wiikite, this mineral not, however, occurring as separate grains in the sample» (op. c., p. 116).

The chemical and spectrographic analysis in Table 9, made of the unaltered brown kernel, establishes what, according to Borgström, has been mentioned above as to rare earths and scandium. »— — it seems most probable that the greatest part, if not all, of alumina actually is rare earths» (op. c., p. 117).

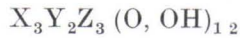
### Spessartite, Hunttila.

This spessartite belongs, as the analysis in Table 9 shows, to the more manganese-rich members of the almandite-spessartite series ( $MnO = 5-40\%$ ) being as to its chemical composition fairly normal.<sup>1</sup> Its  $TiO_2$ -

<sup>1</sup> In Bull. Comm. géol. de Finlande No. 97, 1932, p. 10 it is mentioned about this garnet that it is »un nouveau type de grenat» and that »d'après l'analyse il serait principalement manganoferrifère». This conclusion is quite correct, founded on the analysis which I have obtained from the analyst. He placed also at my disposal, according to his information at that time, some of the same material (fine powder) from which he had made his analysis. Unfortunately, this analysis is rather erroneous; an analysis of the same material very carefully performed by the writer (Anal. IV, Table 9) shows that the mineral is a normal spessartite. To my knowledge garnet has not been found, being »principalement manganoferrifère».



content may, however, be particularly pointed out. So far as the  $\text{TiO}_2$ -content of spessartites is given in the analyses — not even in all newer analyses is it mentioned — it is very low, at most 0.2 %, very seldom more. Accordingly, the  $\text{TiO}_2$ -content 0.49 % of the spessartite of Hunttila is to be regarded as being comparatively high. The calculation of the analysis (Lokka 1943, p. 38) shows, that the chemical composition may best be presented with the formula



or by indicating the chief constituents with the special formula



#### THORIUM MINERALS

The analysed thorium minerals found in the pegmatites of Impilahti are: allanite, monazite, nuolaite and loranskite.

#### ALLANITE

Three analyses of this mineral have been published. They are given in Table 10.

The occurrence at Impilahti has not been particularized more closely for allanites I and II in Meyer's publication (1911). According to him

Table 10. Altered Allanites from Impilahti

	I	II	III
$\text{SiO}_2$	30.52	23.53	23.02
$\text{TiO}_2$			1.25
$\text{Y}_2\text{O}_3$	} 16.45	16.80	0.78
$\text{Ce}_2\text{O}_3$			19.40
$\text{ThO}_2$	1.32	1.10	2.04
$\text{Sc}_2\text{O}_3$	0.80	1.00	
$\text{Al}_2\text{O}_3$	12.88	15.21	16.10
$\text{Fe}_2\text{O}_3$	10.01	12.05	8.01
$\text{FeO}$	6.02	none	none
$\text{MnO}$	tr.	tr.	0.67
$\text{MgO}$	3.80	4.39	2.14
$\text{CaO}$	10.25	4.72	4.98
$\text{Na}_2\text{O}$			1.40
$\text{K}_2\text{O}$	0.22	0.51	0.92
$\text{P}_2\text{O}_5$			0.06
$\text{CO}_2$	0.90	6.90	6.53
$\text{H}_2\text{O}+$	} 7.34	14.01	9.64
$\text{H}_2\text{O}-$			3.16
	<u>100.51</u>	<u>100.22</u>	<u>100.10</u>
Sp. gr.	3.20	2.84	2.913

I and II. Allanites from Impilahti. Anal. R. J. Meyer (Meyer 1911, p. 380).

III. Allanite from Nuolainniemi, Impilahti. Anal. Lauri Lokka (Lokka 1928, p. 49).

allanite I is black, pitchy, hard and fairly fresh; allanite II is brown, rather fragile and in appearance very altered.

Allanite III is from Nuolainniemi and it resembles erroneously a wiikite which has altered considerably. Also the sample I investigated went in the collection I used by the name of wiikite among samples of wiikite (1928, p. 14). The mineral is yellowish, massive, an alteration product.

Table 10 shows that these three allanites have altered considerably. Allanite I has altered least, but in spite of its black colour, also it is as to its chemical composition different in some respects from what it originally undoubtedly has been. Thus the water-content is about 2—3 times as great as it usually is in fresh allanites and  $\text{CO}_2$ , an unknown constituent in fresh allanites, is considerable as to amount. The ratio between the oxides  $\text{Fe}_2\text{O}_3$  and  $\text{FeO}$  also indicates that this allanite has altered, as in fresh allanite  $\text{Fe}_2\text{O}_3$  forms only a small part of the total amount of iron. Water, containing  $\text{CO}_2$ , has probably caused the above mentioned changes in the chemical composition of the mineral, and may be it has caused small changes also in other constituents. As a consequence of the changes also the specific gravity has somewhat decreased from its original value.

Allanites II and III are typical far advanced alteration products. Their analyses are quite comparable to those given in Table 8. The water-content has increased almost double compared with that of allanite I, and the  $\text{CO}_2$ -content is very high. The consequence has been the total oxidation of iron and the partial leaching of some constituents, among them iron. Other constituents have possibly also increased in amount. The specific gravities have decreased very remarkably chiefly because of the abundant content of water and  $\text{CO}_2$ .

In allanite III  $\text{TiO}_2$  is considerable as to amount. It has not been determined in analyses I and II, but instead  $\text{Sc}_2\text{O}_3$  is determined in them. Further particulars are given in connection with the chemistry of the wiikites of Impilahti.

#### MONAZITE

Monazite has been found at Impilahti in Lokansaari already 50 years ago and in Mursula over 10 years ago.

#### Monazite, Lokansaari.

Both fresh and weathered monazite of Lokansaari has been examined. According to Ramsay and Zilliacus (1897, p. 62), the specific gravity of fresh yellow-brown monazite is 5.163 and the powder light yellow. The analysis made by Zilliacus (op. c., p. 64) is shown in Table 11 (I).

The common constituents  $\text{TiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MnO}$  and  $\text{MgO}$  are not determined in the analysis, but since they are found probably only in small amounts in this mineral — so far as they are found at all — the analysis gives, however, a fairly good conception that the chemical composition of the mineral is common.

Table 11. Monazite from Impilahti

	I	II	III
$\text{SiO}_2$ .....	1.22	3.62	
$\text{P}_2\text{O}_5$ .....	26.81	21.50	
$\text{SnO}_2$ .....	0.84		
$\text{ThO}_2$ .....	5.65	9.50	8.80
Yttrium earths .....	2.86	3.22	
$\text{Ce}_2\text{O}_3$ .....	31.63	53.31	60.64
Didymium (lanthanum) earths	29.68		
$\text{Fe}_2\text{O}_3$ .....	0.68		
$\text{CaO}$ .....	0.39		
$\text{PbO}$ .....			0.50
Loss on ign. ....	0.40	2.76	
	100.16		
Sp. gr. ....	5.163		5.177

- I. Fresh monazite from Lokansaari, Impilahti. Anal. Allan Zilliacus (Ramsay and Zilliacus 1897, p. 64).  
 II. Decomposed monazite from Lokansaari, Impilahti. Anal. Allan Zilliacus (*ibid.*, p. 65).  
 III. Fresh monazite from Mursula, Impilahti. Anal. Lauri Lokka.

To prove in what direction the chemical composition of this mineral has changed by weathering, Zilliacus has carried out some determinations of the red-brown, apparently fairly decomposed monazite of Lokansaari. This partial analysis (*op. c.*, p. 65) is also found in Table 11 (II).

Zilliacus concludes from the result of the analysis — supposing probable that the chemical composition of the decomposed monazite has originally been fairly the same as that of the fresh mineral (Anal. I, Table 11) — that  $\text{P}_2\text{O}_5$  has decreased owing to the decomposition and the mineral has taken up a considerable amount of water,  $\text{CaO}$  and also iron, which have not been determined.

Zilliacus also points out that  $\text{SiO}_2$  and  $\text{ThO}_2$  have been enriched by the weathering. Thus he establishes, what in relation to these constituents in such cases can generally be established, namely, that they — as well as  $\text{Al}_2\text{O}_3$  — are most able to resist leaching and are thus the most permanent constituents. They can, of course, indicate enrichment or decrease depending on, whether other constituents have been reduced by the weathering of the mineral, leached by the water, or increased through the substances brought by the water. More exact conclusions cannot, of course, be drawn in this case, as the analysis of the fresh mineral is, as we know, partly incomplete and only a few determinations have been made of the decomposed mineral. In any case, the conclusions of Zilliacus are interesting and also possible as regards the constituents he mentions.

## Monazite, Mursula.

The examined sample of this mineral, found in 1935, was fresh and free from other substances, colour reddish brown, specific gravity 5.177, powder reddish grey. Since the actual purpose was the determination of age, I did not perform a total analysis, but only some determinations, specially with a view to age determination. The results are given in Table 11 (III).

For age determination I regarded it as sufficient in this case to use for determination of thorium 1 g and for that of lead 3 g of the substance. These determinations are carried out in the same way as the corresponding ones in the analysis of gadolinite. The results are:

8.78, 8.82, 8.84 and 8.75 %, average 8.80 %  $\text{ThO}_2 = 7.73$  % Th.

0.488, 0.537, 0.505 and 0.483 %, average 0.503 %  $\text{PbO} = 0.467$  % Pb.

Age calculation is mentioned in connection with Table 25, where also the age is given.

In order to be able to compare the chemical composition of the fresh monazites of Mursula (III) and Lokansaari (I), the sum of rare earths 60.64 % in the monazite of Mursula has also been determined. It is somewhat less than the corresponding value of the monazite of Lokansaari 64.17 %, but the sums of rare earths and  $\text{ThO}_2$  69.82 % (Lokansaari) and 69.44 (Mursula) are fairly equal. Thus it may be concluded that the  $\text{P}_2\text{O}_5$ -contents of these monazites are fairly equal too. As for the other constituents, to the share of which only a few per cent remain in both analyses, — as in the case of monazites in general — the possible changes of their amounts in these two monazites are of no great importance. Still it may be noticed that the specific gravities are fairly equal.

## NUOLAITE

The writer has used the name of nuolaite (1928, p. 21) for a mineral found in Nuolainniemi at Impilahti. In chemical composition it is rather similar to wiikites — therefore I have treated it in connection with these — with the exception of uranium being deficient. To my knowledge a mineral similar to nuolaite has nowhere been found if we do not take into consideration the following one, loranskite, the reports of the thorium-content of which are inconsistent. Referring to the earlier account, the following will briefly be mentioned re nuolaite.

The two nuolaites examined are named in the following nuolaite I and II. Both are black and veined. Fracture of nuolaite I is almost massive with metallic lustre. Face of the vein is here and there asphalt-like. Powder of the unaltered mineral is dark grey. Fracture of nuolaite II is somewhat more lustrous. On the surface of the vein a resin-like,

yellowish tinged brown substance is seen, which in some places penetrates vertically into the black main part.

Microscopic examination of nuolaite I (Fig. 4) shows that the fresh black main part is not homogeneous, but is for the most part a black, nontransparent, opaque substance. The other part is almost colourless, translucent and isotropic, forming irregular veins, in which lightly brown-



Fig. 4. Nuolaite from Nuolainniemi. Black=opaque; white=colourless. 100  $\times$ .

sh tinged small spots and points with double refraction are found here and there. Nuolaite II is under the microscope similar to nuolaite I.

The analyses made of the unaltered, fresh part of both nuolaites are given in Table 12. An analysis of the enrichment product is also there, containing the opaque substance and besides an considerable amount of the colourless substance, which could not entirely be separated from the opaque one. These three analyses were performed according to the same method as the analyses I—VI in Table 21.

The analysis of the enrichment product (III) shows that the oxides have more or less increased or decreased. There is not, however, a very great difference between the analyses II and III. On the ground of analysis III nothing for certain be concluded as to the chemical composition of the black opaque part or the colourless translucent part. It may be thought possible that there have been two somewhat different mineral substances, of which one has penetrated into the other, as if they in the contact had melted together. Because of this they cannot be separated

completely pure, and also because their specific gravities are probably fairly the same. It might also be possible that changes in the originally homogeneous mineral might have occurred in some places.

Table 12. Nuolaite from Nuolainniemi, Impilahti

	I	II	III
Nb <sub>2</sub> O <sub>5</sub>	39.34	38.03	} 63.88
Ta <sub>2</sub> O <sub>5</sub>	24.56	22.41	
TiO <sub>2</sub>	6.47	7.45	7.40
SiO <sub>2</sub>	2.45	2.92	3.29
SnO <sub>2</sub>	tr.	tr.	tr.
ZrO <sub>2</sub>	0.32	0.48	0.40
ThO <sub>2</sub>	3.30	4.40	2.03
Uranium	none	none	
Y <sub>2</sub> O <sub>3</sub>	6.73	7.02	6.62
Ce <sub>2</sub> O <sub>3</sub>	3.57	2.02	1.93
Al <sub>2</sub> O <sub>3</sub>	0.35	0.24	0.33
Fe <sub>2</sub> O <sub>3</sub>	3.23	4.25	8.23
FeO	3.67	2.55	none
MnO	0.34	0.37	0.29
MgO	0.63	1.07	0.97
CaO	1.58	2.06	2.21
PbO	0.21	0.27	0.22
S	none	none	
H <sub>2</sub> O+	2.67	3.61	} 2.66
H <sub>2</sub> O—	0.61	1.22	
	<u>100.03</u>	<u>100.37</u>	<u>100.46</u>
Sp. gr.	5.149	4.860	
H.	6.0	6.0	

I and II. Nuolites. Anal. Lauri Lokka (Lokka 1928, p. 47).

III. Opaque enrichment product of nuolaite II. Anal. Lauri Lokka (ibid., p. 48).

#### LORANSKITE

The first reports of loranskite date from the last years of the past century and the first decade of the present. They are fairly incomplete and contradictory, especially with regard to the chemical composition, as the analyses in Table 13 shows. In so far as I know, no complete examinations have later been published.

More for historical reasons I have earlier (1928) presented the more complete of these two analyses (II, Table 13) among those of wiikite, pointing out only that it differs greatly from the analyses of wiikites. Due to reasons which will be mentioned further on it is appropriate to treat here the loranskite as well as the nuolaite, belonging to the thorium minerals of Impilahti — as being nearest the nuolaites. On the basis of available original publications, general information relating to loranskite, its physical and chemical properties and as correct an analysis of it as possible will be presented in the following.

A short description of 143 minerals, found in Russia, some of them even analysed, is given in a French survey by Melnikow-Pokrowsky (1900)-

Among the minerals are also several from Finland. At the end of the publication (p. 55) *Loranskite* is mentioned, which C. Pfloug<sup>1</sup> had found in a quartz vein at Impilahti and sent to Melnikow in 1896. It is said that the mineral is samarskite-like in appearance. Specific gravity 4.6. As thin flakes it is transparent, optically isotropic and of a yellowish green colour. The powder is greenish grey. Concerning the name of the mineral it is said (p. 56): »M. Melnikow lui donna le nom de loranskite en l'honneur de A. M. Loransky».

The analysis of this mineral, performed by P. D. Nikolaiew in 1898 (op. c., p. 56), is the first (I) in Table 13. The following remarks are made re the chemical composition of the mineral. Being close to pyrochlore and koppite this mineral differs from them on account of a greater amount of metals in the cerium group and the lower CaO-content. With regard to the high content of sulphur it differs from the columbites so far known.

Table 13. *Loranskite* from Impilahti

	I	II	Anal. P. D. Nikolaiew
Nb <sub>2</sub> O <sub>5</sub>	60		
Ta <sub>2</sub> O <sub>5</sub>		47.00	
TiO <sub>2</sub>		tr.	
ThO <sub>2</sub> <sup>1</sup>		20.03	
Y <sub>2</sub> O <sub>3</sub>		10.00	
Ce <sub>2</sub> O <sub>3</sub>	20 à 25	3.00	
FeO		c. 4.00	
MnO		tr.	
CaO	5 à 10	3.30	
S	2	something	
Volat. const.	8	8.15	
Sp. gr.	4.6	4.162	

<sup>1</sup> In original ZrO<sub>2</sub> > 20.00 %.

At the end of the publication reference is made to a publication by M. P. Melnikow — in this instance the name is written Melnikoff — »*Loranskite, nouvelle espèce minéral*», St. Pétersbourg, 1896, which has not been at my disposal.

A rather different analysis of *loranskite* is in the communication by Nikolaiew of a new mineral (1898). In the record No. 1, January 7:th 1897, § 6, p. 11, is mentioned that a letter was presented to the Society (Imp. Min. Soc., Peterborough) written by P. D. Nikolaiew and containing the results of a preliminary analysis of a new mineral, named »*loranskite*» by P. D. Nikolaiew and M. P. Melnikow. This analysis, which is given on page 12 of the record, is in Table 13 (II). The analyst is not mentioned, but very likely it was P. D. Nikolaiew. The specific gravity given is 41.62, probably it should be 4.162, which differs considerably from that of mineral I.

<sup>1</sup> In the publication the name is also written K. Flug.

With regard to the chemical composition of the mineral page 12 of the record states that it is certainly not very complicated, but that on the other hand the presence of zirconium makes the performance of the quantitative analysis particularly difficult, since it is especially difficult to separate from tantalum.

Further on the same page is mentioned, that for determination of the composition of loranskite two analyses will still be performed of the remaining powder of the mineral. I have no knowledge, if these analyses were carried out. At the end reference is made (p. 13) to M. P. Melnikow's publication: «Лоранскитъ. Новый минеральный видъ» (Loranskite, New species of mineral). The year of publication is not given, but concluding from the time the communication was presented, was probably 1896, the same year as for the above mentioned French publication of the same name by Melnikow. Neither has this Russian publication been at my disposal. Probably it is the question of the same publication, published possibly in two languages.

On the basis of the above reported contradictory information, especially as regards the analyses, the following may nevertheless be concluded.

The information relating to loranskite recorded by Melnikow and Pokrowsky deserves notice, but the chemical analysis (Table 13, I) is unfortunately very incomplete and approximate. The analysis given in the communication by Nikolaiew (Table 13, II) is certainly more complete, but nevertheless defective.

In spite of analysis I, according to the information having been performed in 1898 and a communication concerning analysis II having been made already 7:th of January 1897, it may be considered that the former was carried out earlier, since it is much more incomplete than the latter. On account of the incompleteness of analysis I, scarcely anything can be concluded as to the chemical composition of the mineral on the basis of this analysis. It differs so much from analysis II, that it is quite uncertain, whether these analyses have been made of the same or of even two minerals of approximately similar chemical composition. Accordingly analysis I in Table 13 does not give rise to further discussion.

Analysis II — preliminary, according to the above mentioned communication by Nikolaiew — is certainly more complete than analysis I but still very approximate. It still requires considerable completion and elucidation, before detailed conclusions as to the chemical composition of the mineral could be drawn on this basis. Based on two publications mentioned further on it may be said that this is possible having regard to a significant circumstance in this analysis, which nevertheless remains fairly approximate.

In analyses I and II neither U nor Th are mentioned, but some years after these analyses were published, a Russian study by Afanasiew of the influence on photographic plates by minerals containing uranium and



thorium was brought out (1900), and according to this loranskite is radioactive to a considerable degree.

In this study 50 minerals are presented, the uranium- and thorium-content of most are mentioned and the localities more or less accurately given. The minerals are listed according to their activity, beginning with the most active (60—70 % U). The writer points out that more thorium- and uranium-rich minerals generally are also more active, with only some exceptions to this rule.

Loranskite, Finland is mineral No. 42 in the list (p. 105). The locality is not more closely given. The remark is made that this mineral does not contain U or Th, but it is not stated on what this information is founded.

Such nearest minerals to loranskite, the uranium- and thorium-contents of which are mentioned, are: No. 39 walpurgite, Schneeberg, Saxony, 20.54 %  $\text{UO}_3$  and No. 44 monazite, Ilmen Mts, 5.55—17.82 %  $\text{ThO}_2$ . Thus it may be concluded that the limiting values of the activity of loranskite differ rather considerably from each other, the upper limit corresponding to quite a remarkable activity. Accordingly, on the basis of the determination by Afanasiew it can only be said with certainty, that the sample examined contains U or Th or both of them, but as to the amount of these active elements it can only be said, that it may be quite remarkable or even rather small.

Afanasiew thinks possible that the activity of loranskite may be caused by the following (p. 106). As one possibility he thinks: since loranskite is often found together with U-containing wiikite the sample examined by him could contain wiikite minutely divided or in alternate layers, though it was not possible to establish either alternative definitely. The locality of this loranskite is accordingly Impilahti, because it is said to be found together with wiikite. As another possibility Afanasiew thinks we might have to do with a new element in loranskite.

The former supposition seems fairly natural, but most probably the very remarkable activity of loranskite (influence on photographic plate) is not caused by wiikite found as impurity in it, but is due to a cause which will be mentioned more in detail in the following.

If afanasiew's remark that loranskite does not contain U or Th, is based on the analysis, the analysis is in this respect faulty, as this radioactive mineral contains, of course, at least one of these elements. The following information re loranskite may be taken into consideration as an elucidating circumstance.

Several years later than the study of Afanasiew, Szilard's Tables of the most important uranium and thorium ores were published (1909). 111 of these are mentioned in alphabetic order according to name. No mention is made of the source of the information concerning the uranium- and thorium-content of these minerals. On page 237, No. 41, is loranskite,

Finland. This mineral is mentioned as uranium-free, containing 17.6 % Th. It is very probable that the loranskite of Impilahti is in question, the analysis of which is given in Table 13 (II), and that the thorium-content belongs to this analysis, as may be gathered from the following.

In the original publication — the above-mentioned letter by Nikolaiew — in the said analysis  $ZrO_2 > 20.00\%$  is given. Besides there is a remark — as mentioned before — that the presence of zirconium made the performance of the quantitative analysis particularly difficult. These two circumstances agree fairly well. On the other hand again, considering that  $17.6\% Th = 20.03\% ThO_2$ , it seems likely that the original indication  $ZrO_2 > 20.00\%$  should be  $ThO_2 > 20.00\%$ , which error could have been corrected later in some connection or other and according to which Szilard afterwards would have recorded in his tables 17.6 % as the thorium-content of the loranskite of Impilahti. Further, it may be mentioned, according to Borgström (1910, p. 1534), that also in the Russian translation of Szilard's Tables (Odessa 1910) mention is made of a loranskite containing 17.6 % Th.

In analysis II (Table 13) the corrected value is given according to the value of Szilard.

Thus we conclude that loranskite is a radioactive mineral, the activity of which is caused by thorium. The thorium-content of 17.6 % agrees well with Afanasiew's determination.

This thorium mineral belongs, as far as may be concluded from its analysis, nearest to the more thorium-rich nuolaites.

## URANIUM MINERAL WIIKITE

### ON WIIKITE IN GENERAL AND ON ITS PHYSICAL PROPERTIES

Wiikite is the radioactive chief mineral of the earlier mentioned pegmatites of Impilahti, being the only uranium mineral found in these occurrences. It is also the only uranium mineral, found within the old boundaries of Finland, of which mention is made in the literature.

Wiikite was found in 1889 by the Mining Engineer Gr. Lisitzin (1892, p. 152) in the feldspar quarry of Lokansaari in three different varieties: a black crystalline, a dark brown partly crystalline and a brown amorphous. This mineral was then considered by him to be an euxenite.

In 1895 W. Ramsay named the dark «euxenite» of Lokansaari «the euxenite-like mineral», and to a similar but yellow mineral, which he found in 1894 at Nuolainniemi, he gave the name of wiikite after F. J. Wiik.

Such minerals have been found later in all four occurrences at Impilahti and they have in common been called wiikites.

Wiikites are of several types. Earlier (1928) I have presented in detail their mineralogical and other physical properties; brief mention may be made of them here and some additional particulars given.

**Black wiikite, with crystal faces.** The faces are rough in general, the prismatic crystals often rounded. The crystal form is rhombic  $a : b : c = 0.5317 : 1 : 0.5046$  (Borgström, op. c., p. 1543) and  $a : b : c = 0.536 : 1 : 0.528$  (Lokka, op. c. p. 16). It may further be mentioned that crystal fragments have been found in one black, amorphous, analysed wiikite (Table 20, Anal. II). According to the analyst, they probably belong to the rhombic system.

As the crystalline form of wiikite is in question, there is reason to consider what the x-ray analysis later has brought to light in this respect. When examining betafites of Kragerö Björlykke (1931), using this method, has examined also two of the wiikites of Impilahti, one of which was black and the other brown to yellow (braun bis gelb). Both (ignited) proved to be very similar in structure to betafite, which he had established as being regular (op. c., p. 82). Björlykke supposed the brown-yellow wiikite to be a nuolaite, unknown on what grounds, as the analysed nuolaites — uranium free and thorium-containing titano-tantaloniobates (Table 12) — are black, and concluding from this the mineral in question is wiikite.

Pointing out: »Als Wiikit hat L. Lokka (l. c.) auch ein schwarzes Mineral mit rhombischer Kristallform beschrieben» (op. c., p. 82), and meaning by this, of course, what can have been presented re the crystalline form of wiikite on the ground of measuring (see above), Björlykke draws the following conclusion: »Es ist daher wahrscheinlich, dass L. Lokka unter dem Gruppennamen Wiikit zwei in mineralogischer Beziehung sehr verschiedene Minerale zusammengefasst hat. Nämlich ein samarskitähnliches Mineral mit grossen Gehalt an Yttererden, das rhombisch kristallisiert, und ein reguläres Mineral mit wenig Yttererden, dass dem Betafit nahe steht und natürlich der Pyrochlorgruppe gehört.»

Since it has been found that rhombic crystals of black wiikites and most black wiikites contain about 13—30 % of yttrium earths (Tables 20 and 21), it can be concluded that such black wiikite which contains yttrium earths in greater abundance will crystallize rhombic. Since there, however, are also such black wiikites that contain only 6.5—7.5 % yttrium earths (Tables 19 and 20), and in one such amorphous wiikite (6.54 % Y. E.) rhombic crystal fragments of wiikite apparently are found, as mentioned above, it could be concluded that even such black wiikite which contains little yttrium earths may crystallize in rhombic form.

Björlykke established that the black wiikite was regular as to its original structure, in which case, according to the quotation, it would contain little yttrium earths. This supposition is not of general applica-

tion, anyhow, it finds no support in the chemical analyses, as most black wiikites contain yttrium earths in abundance as mentioned just above.

According to the analyses wiikites of other colours contain 1—11 % yttrium earths. To my knowledge no other examinations of their crystal form have been made, than the said determination of the brown-yellow wiikite (regular). On the basis of the analyses we may estimate that it contains at the most something over 10 % yttrium earths.

Machatschki (1932) has also performed x-ray analyses of betafite and doubts whether betafite belongs to the pyrochlore group. Accordingly, there ought not to exist any regular wiikite.

Björlykke's examination of the crystal form of wiikite is certainly of great importance, but there is perhaps still much to examine before this question is satisfactorily solved.

Black, crystalline wiikite. The fracture is rough. In microscopic examination (Fig. 5) is seen that there are two parts in the field of vision: the bigger brown-yellow, translucent, isotropic and the smaller more transparent, yellowish (white in the figure). Limits are not clear. Black, opaque, narrow stripes and spots with sharp limits appear irregularly divided in the field of vision. The analysis of this wiikite is the first in Table 21.

Because of a special mode of occurrence a black wiikite has to be mentioned. It appears, as radioactive minerals often do, as some cm. thick columns — the faces of which are clear — joined finger-like to each other. The fracture has almost metallic lustre, differing thus somewhat from the foregoing. This mineral is also in other respects very similar to the

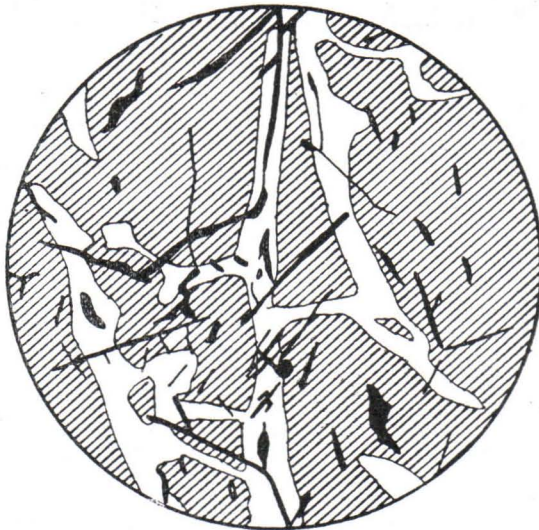


Fig. 5. Wiikite from Hunttila. Black=opaque; shaded=brown-yellow; white=yellowish. 100  $\times$ .

former, as the microscopic examination shows, the alteration, however, is more advanced (Anal. II, Table 21).

Amorphous, black wiikite. Massive, with glassy lustre. In the contact the surrounding rock (microcline) has altered, influenced by wiikite. Fracture as in the case of asphalt, powder grey, slightly brownish tinged. More altered than the foregoing (Anal. III, Table 21).

As quite a special case a black, amorphous, lead ore containing wiikite has to be mentioned. In microscopic examination (Fig. 6) the



Fig. 6. Wiikite from Lokansaari. White=brownish yellow; black=opaque; a—b and c = lead ore. 100  $\times$ .

largest part of the field of vision is brown-yellowish, isotropic (white in the figure). The other part (black in the figure) is black, opaque, appearing as thin stripes and as shapeless larger areas. Lead ore covers almost totally in the shape of a broad stripe the space between a-b and to the right the area c. This explains why the lead-content seems to be unnatural high in the analysis of this wiikite (Anal. IV, Table 21). The lead found there is without doubt for the most part common lead.

Amorphous, reddish brown wiikite. Massive, does not to any extent worth mentioning differ in appearance from the black amorphous type. Fracture as in the case of asphalt, colour of the powder brown-grey. Under the microscope, in the weakly translucent, brown, isotropic field of vision, similar narrow black stripes are seen as mentioned above (Anal. V, Table 21).

Lighter wiikites are amorphous too, colour from grey-yellow to straw-yellow, lustre often wax-like, massive, not so heavy as

the former. Powder is yellow in different shades. Under the microscope the different kinds appear somewhat unlike. In the field of vision we may see a darker and a lighter part, both of them isotropic, or even the darker part not transparent. The whole field of vision may also be grey-brown and weakly translucent. Some analyses of such wiikites are given in Tables 20 and 21.

These types of different colours may occur associated in the same way, as mentioned before concerning the allanite of Varala, viz. without regularity. Of the alteration further on.

#### ON ANALYTICAL METHOD IN GENERAL

Earlier I have in detail presented (1928, pp. 33—42) the method of analysis which I then used when analysing wiikites. With reference to this, in the following only the modifications of this method will be mentioned, which were made when performing later analyses of wiikites (Anal. VII and VIII, Table 21; Table 32). — Mention may, however, first be made of the following.

Björlykke says (1931, p. 75) that, when analysing betafites of Kragerö, he used chiefly the same method I had earlier used in analysing wiikites. As he thus in a ways has adjusted this method, there is reason to mention the results he has achieved.

He used for the analysis the usual amount 1 g. of the substance, which he fused with potassium pyrosulphate. Of the performance of the analysis he says: »Ausser der Bestimmung der einzelnen Metallsäuren verlief die Analyse sehr glatt, und die Parallelen gaben gut übereinstimmende Resultate. Ausserdem wurden alle Ausfällungen röntgenspektrographisch mit einem Vakuumspektrograph untersucht und erwiesen sich sämtlich ziemlich rein».

Björlykke establishes that the separation of earth acids from each other causes many difficulties, the separation of titanium from niobium and tantalum was specially laborious. The establishing of this is without doubt right and corresponds to what is earlier known in this respect of such minerals as wiikites and betafites. Of the salicylic acid method, which Björlykke used and which also I have used in separating titanium from niobium and tantalum, he says that it »ist nicht sehr gut geeignet, mit dem grossen Mengen von Titan, die sich in meinen Analysen vorfanden. Die Summe der Oxyde von Niob und Tantal hätten bei meinen Proben 7—8 Mal mit  $K_2CO_3$  aufgeschlossen und mit Salizylsäure gekocht werden müssen, um das Titan befriedigend abzuschneiden. Bei allen diesen Manipulationen ist es nicht zu vermeiden, dass ein Verlust eintritt, wenn auch sehr sorgfältig gearbeitet wird. Die röntgenspektrographische Untersuchung des bei dieser Methode gelösten Titan zeigte doch, dass

das Titan sehr rein sei, indem die Linien von Niob und Tantal nicht sichtbar waren» (op. c., p. 75).

Very many difficulties were undoubtedly met in the performance of this task and it required a great deal of time, but the result was good with regard to the purity of titanium. The use of salicylic acid in separation of titanium was much more difficult in the analyses of betafite than in those of wiikite — though even in them it takes much time and is a laborious task. The three betafites analysed by Björlykke contain viz. (op. c., pp. 77—79) 32—35 %  $\text{TiO}_2$  and 18—24 % earth acids, whereas most wiikites and nuolaites analysed by me (Table 21) contain only 3.5—7.5 %  $\text{TiO}_2$ , the two most titanium-rich only 11.5 and 14.6 %; wiikites contain 35—46 % and nuolaites 60—64 % earth acids.

To shorten the laborious salicylic acid method Björlykke tried to combine it with a method according to which the greatest part of titanium should remain insoluble, when  $\text{K}_2\text{CO}_3$ -melt is treated with hot water. This, however, did not give a satisfactory result, the titanium contained very remarkable amounts of niobium and tantalum.

Finally Björlykke mentions concerning the separating of titanium (op. c., p. 76): »Ich habe daher die Salizylsäuremethode allein für alle Proben angewandt, und bei jeder Probe wurde 7—8 Mal mit Salizylsäure behandelt».

As mentioned before very pure titanium was separated by this treatment. Of the sum of the oxides  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$  Björlykke says it gave a very weak titanium colour reaction, the error being at the most about some tenths of one per cent.

Björlykke has thus shown that the salicylic acid method is fit for use in separating titanium from niobium and tantalum, even when larger amounts of titanium are in question, if only the treatment is performed a sufficient number of times.

To separate niobium and tantalum from each other I used Marignac's method, Björlykke again that of Merrill, establishing that the separation was not quite quantitative.

As mentioned above I have later made modifications in the method I earlier used in analysing wiikites. Some instances follow here.

The determination of thorium. For the separation of thorium from rare earths I used the thiosulphate method, which gives a satisfactorily accurate result when carefully done. Even in such cases, when the  $\text{ThO}_2$ -content of the minerals is 5—7 % and the amount of rare earths about 20 %, I have got fairly conformable values by the thiosulphate and the hydrogen peroxide methods. In two analyses of wiikite (Anal. VII and VIII, Table 21) I have used the last mentioned method.

To separate niobium and tantalum from each other I used, as above has been said, Marignac's method. The experi-

ments I carried out with niobium and tantalum preparates showed this method, which demands great carefulness and accuracy, to be satisfactorily accurate (op. c., pp. 36—37). But it being very laborious, as known, I experimented also with other methods of separation using both inorganic and organic reagents. The last mentioned are nowadays already of considerable importance both in qualitative and quantitative analyses, also in the separation of niobium and tantalum from each other. The use of tannin for this purpose has many good points, but in the analyses of wilkite it was a laborious task requiring much time. Finally I arrived at the following photometric method.

#### PHOTOMETRIC DETERMINATION OF $Nb_2O_5$ IN MIXED OXIDES $Nb_2O_5 + Ta_2O_5$

It is known that, when using some organic reagents, very remarkable colour reactions appear in niobium solutions owing to the influence of zinc, but the corresponding tantalum solution remains colourless. This led me to make experiments, to ascertain whether niobium could be determined in mixed oxides  $Nb_2O_5 + Ta_2O_5$  in a less complicated way than by separation and gravimetric methods, either colorimetrically or photometrically.

It is not necessary to give a detailed account of the fairly numerous experiments I carried out, the following short report will suffice. Above all it was, of course, important to produce such a colour reaction of niobium by means of zinc that the colour would be permanent or at least would remain unchanged for some length of time. In the beginning I did not, however, succeed in this. In some experiments the colour of the liquid, lighter at first, changed little by little, finally becoming dark and opaque in even a thin liquid layer. In other experiments, on the contrary, the colour grew more and more faint, at last vanishing entirely. In some cases again the colour varied more quickly or more slowly, changing finally to opaque or totally disappearing.

It seemed already as it were vain to go on with the experiments. I did not succeed in preparing such a solution containing niobium which would have been permanent in colour and accordingly a suitable standard solution for the performance of colorimetric determinations. However, I at last succeeded in some experiments. After about a quarter of an hour from the beginning of the reaction the colour of the niobium solution remained unchanged for a longer time, for one or even two hours. Thus the performance of photometric determinations seemed possible.

The principal traits of these determinations may quite shortly be mentioned. Earth acids, mixed oxides  $Nb_2O_5 + Ta_2O_5$ , are in the following designated by EA.



A. **Known solutions.** They were prepared according to three different methods (I, II, III), where in dissolving the oxides  $\text{Nb}_2\text{O}_5$  and  $\text{Ta}_2\text{O}_5$  different solvents were used in different methods. Otherwise the procedure was the same in all methods.

According to each method original solutions A and B were first prepared of the oxides  $\text{Nb}_2\text{O}_5$  and  $\text{Ta}_2\text{O}_5$ , each 200 ml., containing A 250 mg.  $\text{Nb}_2\text{O}_5$  and B 250 mg.  $\text{Ta}_2\text{O}_5$ .

Comparison solutions were prepared from the original solutions (methods I and III each 12 and method II 11 solutions). The volumes of them all were equal and so also the EA-contents, but the  $\text{Nb}_2\text{O}_5$ -contents varied (Table 14).

Zinc was added to the colourless comparison solutions to produce colour reaction. The extinctions were determined and the extinction coefficients in Tables 15—17 were calculated.

B. **Test solution.** From the weighed mixture EA obtained in the analysis an original solution is prepared, according to one of the methods I—III, and of this a test solution in the same way as the comparison solutions mentioned above. The extinction coefficient is determined and the corresponding  $\text{Nb}_2\text{O}_5$ -content in mixed oxides  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$  is sought from Table 18. The  $\text{Nb}_2\text{O}_5$ - and  $\text{Ta}_2\text{O}_5$ -contents of the mineral are calculated.

The detailed performance of the above is as follows.

A. **Known solutions.**

The three methods in question are mentioned according to the solvents of the oxides  $\text{Nb}_2\text{O}_5$  and  $\text{Ta}_2\text{O}_5$ .

Method I. Ammonium Oxalate  $\text{H}_2\text{SO}_4$  Solution.

» II. Tartaric Acid Solution.

» III. Tartaric Acid  $\text{H}_2\text{SO}_4$  Solution.

Method I. Ammonium Oxalate  $\text{H}_2\text{SO}_4$  Solution.

Original solutions A and B. The necessary pure oxides were obtained from the preparations  $\text{Nb}_2\text{O}_5$  and  $\text{Ta}_2\text{O}_5$ , their pyrosulphate melts (K-pyros.) were as usual dissolved, precipitated, filtered and ignited. The original solutions A and B were prepared from these oxides in the same way. The pyrosulphate melts, for which 250 mg. oxide and 3 g. potassium pyrosulphate were used, were dissolved in a 250 ml. beaker in a solution containing 4g. ammonium oxalate. The solution, about 150 ml., was left standing in its glass over night, in order to be sure of the liquid remaining clear, as both the solutions also do, if the fusion and solution are carefully carried out.

Next day the clear solutions were transferred into 200 ml. graduated flasks, which were filled with water up to the mark. We obtained original solutions A = 250 mg.  $\text{Nb}_2\text{O}_5$  per 200 ml. and B = 250 mg.  $\text{Ta}_2\text{O}_5$  per

200 ml, which remained clear for a long time.  $\text{H}_2\text{SO}_4$  was not added until the comparison solution was being prepared, as had it been added at once, even though in such small amount as needed in this case, the solution B would have remained clear only a short time and next day it would have been turbid and unfit for photometric determinations.

Comparison solutions. From the original solutions A and B 12 comparison solutions were prepared, each 25 ml., containing 25 mg. EA. These solutions were prepared at short intervals a few in succession, according to how they — considering the time — could be used in photometric determinations. The procedure was as follows. Such amounts as Table 14 shows, 20 ml. in all, of the original solutions A and B were taken into a 50 ml. Erlenmeyer flask. To the solution was added 5 ml. sulphuric acid, containing 20 % per volume strong  $\text{H}_2\text{SO}_4$ . Each solution was thus 25 ml., containing 300 mg. potassium pyrosulphate, 400 mg. ammonium oxalate and 4 % per volume strong  $\text{H}_2\text{SO}_4$ . There was in all of them 25 mg. EA.

Table 14

Comp. Sol. No.	Orig. Sol. A		Orig. Sol. B		$\text{Nb}_2\text{O}_5$ % of EA
	ml.	$\text{Nb}_2\text{O}_5$ mg.	ml.	$\text{Ta}_2\text{O}_5$ mg.	
1			20	25.0	0
2	1	1.25	19	23.75	5
3	2	2.5	18	22.5	10
4	4	5.0	16	20.0	20
5	6	7.5	14	17.5	30
6	8	10.0	12	15.0	40
7	10	12.5	10	12.5	50
8	12	15.0	8	10.0	60
9	14	17.5	6	7.5	70
10	16	20.0	4	5.0	80
11	18	22.5	2	2.5	90
12	20	25.0			100

To produce the colour reaction 1 g. pure zinc dust was added to each solution immediately after it had been prepared. To promote the reaction the flask was shaken for 10 seconds at the same speed (20 swings). The flask was left standing with the cork on, but a little ajar. The zinc settled by and by. The colour of the clear liquid, brownish at the outset, changed to more or less yellow during 15—30 minutes, depending on the niobium-content of the liquid. Afterwards the liquid remained clear and obviously the same as to quality and intensity of colour. In one hour after the zinc had been added the comparison solution, prepared in this way, was ready for photometric determination of extinction.

With a suitable pipette clear liquid was now taken from the flask, endeavours being made to exclude the zinc, the glass cell was filled, it had to be chosen so that the transparency of the liquid in it was suitable for the determination of extinction.

The determinations were performed by using Pulfrich Photometer (Zeiss, Jena). As light source the mercury lamp »Hagephot» was used. The most suitable filter was »Hg 436».

Several parallel determinations of extinction were carried out for each comparison solution. The procedure was as follows. Beginning with the comparison solution No. 1 (Table 14) extinctions were determined for as many solutions as possible on the same day. Then new solutions A and B were made and performance of the determinations was continued until the end of the series. After this it was begun again from the beginning of the series proceeding in the same way, until 3—4, in uncertain cases 5—6, parallel determinations had been performed per each comparison solution of the series. Finally the extinction coefficients were calculated for the averages of determinations.

Table 15 shows the results of the determinations. The amounts of the oxides  $Nb_2O_5$  and  $Ta_2O_5$  are shown in columns 1 and 2, in every comparison solution 25 mg. in all, the volume of each is 25 ml. In column 3  $Nb_2O_5$  is given in percentage of EA. In column 4 the thickness of the layer of the liquid is  $d$  cm. in the glass cell. The values of extinction and their averages (below the line) are in column 5. The extinction coefficients  $k = \frac{E}{d}$  in column 6 correspond to these averages. For comparison the »recalculated» amounts of  $Nb_2O_5$  in per cent from EA (in comparison solutions), corresponding to the coefficients  $k$ , are given in column 7 according to Table 18 (k). The corresponding values in columns 7 and 3 differ from one another only to the extent of an error in observation. Of Table 18 more further on.

#### Method II. Tartaric Acid Solution.

The original solutions A and B were prepared in the same way as before, except that the pyrosulphate melts of the oxides  $Nb_2O_5$  and  $Ta_2O_5$  in this case were dissolved in tartaric acid. Each solution was 30 ml. containing 3 g. tartaric acid. Also in this case the diluted solutions, each about 150 ml., were left standing over night. The clear solutions were transferred to 200 ml. graduated flasks, which were filled with water up to the mark. Also these original solutions, 250 mg.  $Nb_2O_5$  per 200 ml. and 250 mg.  $Ta_2O_5$  per 200 ml., remained clear for a long time.

11 comparison solutions were prepared from the original solutions A and B similarly as before (Table 14), except that 5 ml. water was added instead of sulphuric acid. Thus the volume of each solution

Table 15. Extinction Coefficients. I. Ammonium Oxalate  $H_2SO_4$  Solution

Comp. sol.	25 mg. EA/25 ml.			4 d cm.	5 Ext. E	6 Ext. coeff. $k = \frac{E}{d}$	7 $\frac{Nb_2O_5}{\%}$ of EA (Tab. 18)	Comp. sol.	25 mg. EA/25 ml.			4 d m.	5 Ext. E	6 Ext. coeff. $k = \frac{E}{d}$	7 $\frac{Nb_2O_5}{\%}$ of EA (Tab. 18)		
	1 $Nb_2O_5$ mg.	2 $Ta_2O_5$ mg.	3 $\frac{Nb_2O_5}{\%}$ of EA						1 $Nb_2O_5$ mg.	2 $Ta_2O_5$ mg.	3 $\frac{Nb_2O_5}{\%}$ of EA						
1	0.00	25.0	0.0	5	.341			7	12.5	12.5	50	2	.810				
					.319								.796				
					.332								.798				
2	1.25	23.75	5	5	.372			8	15.0	10.0	60	2	.970				
					.375								.990				
					.389								1.020				
3	2.5	22.5	10	3	.379	.076	4.67	9	17.5	7.5	70	1	.584				
					.295	.095	10.22						.629				
					.278								.579				
4	5.0	20.0	20	3	.438			10	20.0	5.0	80	1	.598	.598	68.93		
					.447								.149	20.15	.724		
					.460										.772		
5	7.5	17.5	30	3	.660			11	22.5	2.5	90	0.5	.434				
					.624								.214			29.47	.431
					.638												.435
6	10.0	15.0	40	2	.612			12	25.0	0.0	100	0.5	.433	.866	90.21		
					.652								.309	40.74	.526		
					.589										.485		
					.620								.522				
					.618								.478				
													.503	1.006	99.73		

was 25 ml., everyone containing 300 mg. potassium pyrosulphate, 300 mg. tartaric acid and 25 mg. EA.

The comparison solutions were prepared for the photometric determinations (Zn) and the determinations were performed in the same way as before. The results of the determinations are given in Table 16, which is arranged in the same manner as Table 15.

#### Method III. Tartaric acid $H_2SO_4$ Solution.

Tartaric acid original solutions A and B were used here and prepared quite as before (II).

12 comparison solutions were prepared from the original solutions according to Table 14, but finally 5 ml. sulphuric acid containing 5 % per volume strong  $H_2SO_4$  was added to each of them.

Table 16. Extinction Coefficients. II. Tartaric Acid Solution

Comp. sol.	25 mg. EA/25 ml.			4 d cm.	5 Ext. E	6 Ext. coeff. $k = \frac{E}{d}$	7 $\text{Nb}_2\text{O}_5$ % of EA (Tab. 18)	Comp. sol.	25 mg. EA/25 ml.			4 d cm.	5 Ext. E	6 Ext. coeff. $k = \frac{E}{d}$	7 $\text{Nb}_2\text{O}_5$ % of EA (Tab. 18)
	1 $\text{Nb}_2\text{O}_5$ mg.	2 $\text{Ta}_2\text{O}_5$ mg.	3 $\text{Nb}_2\text{O}_5$ % of EA						1 $\text{Nb}_2\text{O}_5$ mg.	2 $\text{Ta}_2\text{O}_5$ mg.	3 $\text{Nb}_2\text{O}_5$ % of EA				
1	0.0	25.0	0.0	5	.069			7	15.0	10.0	60	2	.425		
					.082								.449		
					.067								.437		
					.073								.442		
2	2.5	22.5	10	5	.095	.015	0.00	8	17.5	7.5	70	2	.438	.219	60.59
					.108								.595		
					.099								.507		
					.112								.557		
3	5.0	20.0	20	5	.104	.021	11.00	9	20.0	5.0	80	2	.611	.276	68.75
					.170								.486		
					.146								.551		
					.145								.551		
4	7.5	17.5	30	3	.154	.031	18.33	10	22.5	2.5	90	1	.716	.357	80.14
					.173								.475		
					.148								.430		
					.182								.420		
5	10.0	15.0	40	3	.172	.056	30.67	11	25.0	0.0	100	1	.481	.446	91.71
					.169								.425		
					.291								.446		
					.245								.446		
6	12.5	12.5	50	2	.301	0.091	39.40	7	25.0	0.0	100	1	.529	.507	99.62
					.259								.480		
					.274								.465		
					.274								.465		
					.310								.540		
					.288								.469		
					.304								.507		
					.313								.507		

Thus the volume of the solutions was 25 ml. and each contained 300 mg. potassium pyrosulphate, 300 mg. tartaric acid, 1 % per volume strong  $\text{H}_2\text{SO}_4$  and 25 mg. EA.

The final preparation of comparison solutions for photometric determinations (Zn) and all the other procedures were the same as before (I, II). The results of the determinations are shown in Table 17, which is arranged in the same manner as Tables 15 and 16.

The dependence of the extinction coefficient on the  $\text{Nb}_2\text{O}_5$ -content in the comparison solutions is graphically presented in figure 7. The solutions are prepared according to the above-mentioned methods I—III.

The curves rise at first more steeply, curves I and II very steeply,

Table 17. Extinction Coefficients. III. Tartaric Acid  $H_2SO_4$  Solution

Comp. sol.	25 mg. EA/25 ml.			4 d cm.	5 Ext. E	6 Ext. coeff. $k = \frac{E}{d}$	7 $Nb_2O_5$ % of EA (Tab. 18)	Comp. sol.	25 mg. EA/25 ml.			4 d cm.	5 Ext. E	6 Ext. coeff. $k = \frac{E}{d}$	7 $Nb_2O_5$ % of EA (Tab. 18)
	1 $Nb_2O_5$ mg.	2 $Ta_2O_5$ mg.	3 $Nb_2O_5$ % of EA						1 $Nb_2O_5$ mg.	2 $Ta_2O_5$ mg.	3 $Nb_2O_5$ % of EA				
1	0.00	25.0	0.0	5	.061			7	12.5	12.5	50	0.5	.389		
					.064								.378		
					.060								.378		
					.062								.380		
					.062	.012	0.00					.381	.762	50.09	
2	1.25	23.75	5	5	.240			8	15.0	10.0	60	0.5	.474		
					.220								.497		
					.225								.487		
					.228								.480		
						.046	4.94					.485	.970	59.47	
3	2.5	22.5	10	3	.279			9	17.5	7.5	70	0.5	.624		
					.270								.604		
					.300								.600		
					.282								.634		
					.283	.094	10.00					.616	1.232	70.53	
4	5.0	20.0	20	1	.205			10	20.0	5.0	80	0.25	.365		
					.212								.388		
					.212								.374		
					.200								.362		
					.207	.207	19.77					.372	1.488	80.73	
5	7.5	17.5	30	1	.360			11	22.5	2.5	90	0.25	.446		
					.358								.421		
					.360								.420		
					.380								.426		
					.365	.365	29.72					.428	1.712	89.32	
6	10.0	15.0	40	0.5	.288			12	25.0	0.0	100	0.25	.480		
					.271								.490		
					.283								.420		
					.289								.495		
					.283	.566	40.55					.496	1.984	99.41	

curve II almost vertically, while the rise of curve III is gently sloping. As the  $Nb_2O_5$ -content amounts to about 40 %, the curvature of curve II diminishes, being almost imperceptible, and that of curve III already earlier. Curve I is most curved towards its end.

The starting point of curves II and III is practically in the zero, while that of curve I is not. This is due to the fact that the solution prepared according to method I, the  $Nb_2O_5$ -content of which is nought, has already a considerable extinction. According to the corresponding extinction coefficient 0.066, the starting point of the curve on the abscissa is rather far from the zero. This, however, has not a disturbing effect.

### B. Test solution.

If the solution is prepared according to one of the above mentioned methods from the mixed oxides EA, which is known as to weight and the extinction coefficient of which is determined, the  $Nb_2O_5$ -content of the said mixture can be determined approximately from figure 7 and more exactly from Table 18. The  $Ta_2O_5$ -content is, of course, received indirectly from the difference.

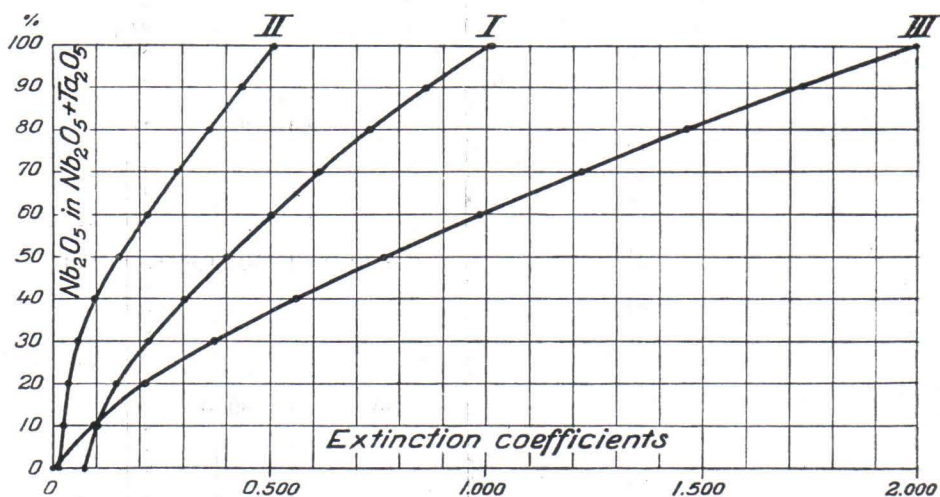


Fig. 7.

In order to achieve as reliable a result as possible, the test solution should be free from extraneous substances, above all from such causing a colour reaction in the treatment of the solution. A slight titanium-content occasions no noticeable disturbance, if the niobium-content is remarkable. But if it is very low, the colour reaction of even such an amount of titanium which as impurity is insignificant can perceptibly intensify the colour of the solution, and thus perhaps exert a deal of influence upon the value of the extinction coefficient.

Table 18 is compiled on the basis of Tables 15—17. Sufficiently accurate results are obtained when using it, as has been pointed out before. This may be seen from column 7 in Tables 15—17.

The differences on the right hand side of the values  $k$  are given to make the comparison easier. The greatest of them are of use in interpolation.

The greater the differences are, the less effect have, of course, the errors in observation, made in the photometric determinations. Thus the  $Nb_2O_5$ -contents can most accurately be determined by method III, which I have used in the analyses of two wiikites of Impilahti (Anal. VII and VIII, Table 21) and the wiikites of Salla (Table 32).

Table 18.  $\text{Nb}_2\text{O}_5$  in % of  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$ 

1 mg. EA/1 ml. k = extinction coefficient

I. Am. Oxalate  $\text{H}_2\text{SO}_4$  Solution (Table 15)

II. Tartaric Acid Solution (Table 16)

III. Tartaric Acid  $\text{H}_2\text{SO}_4$  Solution (Table 17)

$\text{Nb}_2\text{O}_5$ % of EA	k			$\text{Nb}_2\text{O}_5$ % of EA	k		
	I	II	III		I	II	III
0	0.066	0.015	0.012	30	0.218	0.054	0.370
2	.069 <sup>3</sup>	.016 <sup>1</sup>	.024 <sup>12</sup>	32	.234 <sup>16</sup>	.060 <sup>6</sup>	.406 <sup>36</sup>
4	.074 <sup>5</sup>	.017 <sup>1</sup>	.038 <sup>14</sup>	34	.250 <sup>16</sup>	.067 <sup>7</sup>	.442 <sup>36</sup>
6	.080 <sup>6</sup>	.018 <sup>1</sup>	.055 <sup>17</sup>	36	.267 <sup>17</sup>	.075 <sup>8</sup>	.479 <sup>37</sup>
8	.087 <sup>7</sup>	.019 <sup>1</sup>	.074 <sup>19</sup>	38	.284 <sup>17</sup>	.084 <sup>9</sup>	.517 <sup>38</sup>
10	.094 <sup>7</sup>	.020 <sup>1</sup>	.094 <sup>20</sup>	40	.302 <sup>18</sup>	.094 <sup>10</sup>	.555 <sup>38</sup>
12	.103 <sup>9</sup>	.022 <sup>2</sup>	.115 <sup>21</sup>	45	.349 <sup>47</sup>	.120 <sup>26</sup>	.655 <sup>100</sup>
14	.113 <sup>10</sup>	.024 <sup>2</sup>	.137 <sup>22</sup>	50	.398 <sup>49</sup>	.150 <sup>30</sup>	.760 <sup>105</sup>
16	.124 <sup>11</sup>	.026 <sup>2</sup>	.160 <sup>23</sup>	55	.448 <sup>50</sup>	.182 <sup>32</sup>	.869 <sup>109</sup>
18	.136 <sup>12</sup>	.029 <sup>3</sup>	.184 <sup>24</sup>	60	.500 <sup>52</sup>	.215 <sup>33</sup>	.982 <sup>113</sup>
20	.148 <sup>12</sup>	.032 <sup>3</sup>	.210 <sup>26</sup>	65	.554 <sup>54</sup>	.249 <sup>34</sup>	1.099 <sup>117</sup>
22	.161 <sup>13</sup>	.035 <sup>3</sup>	.238 <sup>28</sup>	70	.610 <sup>56</sup>	.285 <sup>36</sup>	.220 <sup>121</sup>
24	.174 <sup>13</sup>	.039 <sup>4</sup>	.268 <sup>30</sup>	75	.669 <sup>59</sup>	.321 <sup>36</sup>	.343 <sup>123</sup>
26	.188 <sup>14</sup>	.043 <sup>4</sup>	.300 <sup>32</sup>	80	.731 <sup>62</sup>	.358 <sup>37</sup>	.469 <sup>126</sup>
28	.203 <sup>15</sup>	.048 <sup>5</sup>	.334 <sup>34</sup>	85	.795 <sup>64</sup>	.395 <sup>37</sup>	.598 <sup>129</sup>
30	.218 <sup>15</sup>	.054 <sup>6</sup>	.370 <sup>36</sup>	90	.863 <sup>68</sup>	.433 <sup>38</sup>	.730 <sup>132</sup>
				95	.935 <sup>72</sup>	.471 <sup>38</sup>	.864 <sup>134</sup>
				100	1.010 <sup>75</sup>	.510 <sup>39</sup>	2.000 <sup>136</sup>

The procedure was as follows. 250 mg. of mixed oxides EA, which was free from extraneous substances, ignited and weighed, was taken and thence an original solution 200 ml. was prepared, according to method III. Next day three similar test solutions (1 mg.EA/1 ml.) were prepared. The extinctions were determined; in these three cases the thicknesses of layers of the liquids were different in the glass cells. At least three parallel determinations were made in each case. Three extinction coefficients were calculated. Using the same original solution, after some days three extinction coefficients were again determined in the above mentioned way. The  $\text{Nb}_2\text{O}_5$ -content in mixed oxides EA was sought from Table 18 (III), corresponding to the average of the six extinction coefficients. Finally the  $\text{Nb}_2\text{O}_5$ - and  $\text{Ta}_2\text{O}_5$ -contents of the mineral were calculated.

In the analysis VII, Table 21, this procedure, briefly given with figure values, was as follows:

$\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$ in the analysis (1 g. subst.)	392.9 mg.
Test solution	1 mg. EA/1 ml.
Extinction coefficient k	0.844
$\text{Nb}_2\text{O}_5$ -content in the mixture $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$	53.85 %
» » » » mineral	21.16 »
$\text{Ta}_2\text{O}_5$ » » » »	18.13 »



If there is less than 250 mg. EA at disposal, even then, of course, the said methods can be used, if only attention be paid to the concentration 1 mg. EA/1 ml.

The three methods presented before are not the only possible ones of their kind, though only they have been discussed here. Other similar methods may, of course, be presented for the same purpose, such, where the solvents, the concentrations and the procedures also otherwise differ to some extent from the three mentioned above.

#### ON THE CHEMISTRY OF WIIKITE

Even though the chemistry of the wiikite of Impilahti has been treated earlier, for good reason it may still be discussed. I have mentioned earlier (1928) the older investigations and given the main points of them. Attention has been paid to this mineral also in foreign literature especially to its chemistry.

In Tables 19—21 the older and newer analyses of the wiikites of Impilahti have been given.

The two oldest analyses in Table 19 are to some extent incomplete and inaccurate, also the analysts point this out. Thus, according to Borgström (1910, p. 1541), in the attestation of an analysis of wiikite, made by Holmqvist at Stockholm 1895, it is mentioned that the analysis has not given accurate results on account of the difficulties encountered. To

Table 19. Wiikites from Impilahti

	I	II
Nb <sub>2</sub> O <sub>5</sub> .....	23.67	(Ta <sub>2</sub> O <sub>5</sub> cont. some Nb <sub>2</sub> O <sub>5</sub> )
Ta <sub>2</sub> O <sub>5</sub> .....	(probably none)	15.91
TiO <sub>2</sub> .....	29.58	23.36
SiO <sub>2</sub> .....	8.75	16.98
ZrO <sub>2</sub> .....	(probably none)	(TiO <sub>2</sub> cont. some ZrO <sub>2</sub> )
ThO <sub>2</sub> .....		5.51
Sc <sub>2</sub> O <sub>3</sub> .....		1.17
UO <sub>2</sub> .....	1.86	3.56
UO .....	7.37	
Y <sub>2</sub> O <sub>3</sub> .....	} 4.06	7.64
Ce <sub>2</sub> O <sub>3</sub> .....		2.55
Al <sub>2</sub> O <sub>3</sub> .....	0.74	
Fe <sub>2</sub> O <sub>3</sub> .....	7.51	
FeO .....		15.52
Mn <sub>3</sub> O <sub>4</sub> .....	1.28	
CaO .....	4.86	
Ca, Mn, Sn, S, etc. unestim.		1.97
H <sub>2</sub> S precipitate .....	1.06	
Loss on ign. ....	<u>11.06</u>	<u>5.83</u>
	101.80	100.00
Sp. gr. ....		4.85

- I. Wiikite from Nuolainniemi (?), Impilahti. Anal. P. J. Holmqvist (Borgström 1910, p. 1541s.  
 II. Wiikite, black, amorphous, from Lokansaari, Impilahti. Anal. William Crookes (Crooke) 1908, p. 517).

my knowledge this is the first analysis of wiikite. Crookes mentions (1908, p. 517) re the wiikite he had analysed it is black, amorphous and that heating drives off 5.83 % of its weight, of which 5.82 % is water and acid vapour, the difference, 0.01 %, consisting chiefly of He, with a little H, CO<sub>2</sub> and mere trace Neon». Further on he says: »Containing so many bodies, the exact separation of which one from the other is not known, it is at present impossible to give an accurate and complete analysis of wiikite». Because of this Crookes regards his analysis as approximate.

The analyses in Table 20 are published later. The mineralogical properties of the three first wiikites (Anal. I—III) I have earlier referred to (1928, p. 11), according to the original publication (Wuorinen 1916). In the original publication (Ant-Wuorinen 1936), not mention is made of the properties of the three latter, not even the colour of the mineral. With regard to these six analyses the following may be mentioned.

In the analyses of wiikites, both in older and newer, the yttrium group is to its amount chiefly predominant compared with the cerium group.

Table 20. Wiikites from Impilahti

	Anal. Jalo Ant-Wuorinen					
	I	II	III	IV	V	VI
Nb <sub>2</sub> O <sub>5</sub>	21.17	20.53	55.62	43.53	35.83	31.59
Ta <sub>2</sub> O <sub>5</sub>	4.73	0.87	tr. in Nb <sub>2</sub> O <sub>5</sub>	2.32	3.34	1.29
TiO <sub>2</sub>	25.84	7.06	6.91	2.45	2.80	11.02
SnO <sub>2</sub>	0.13	0.09	tr.		tr.	tr.
SiO <sub>2</sub>	5.00	15.84	2.73	3.46	1.96	2.99
ZrO <sub>2</sub>	0.08	tr.	tr.	tr.		0.80
ThO <sub>2</sub>	0.50	2.08	3.58		1.60	0.33
Sc <sub>2</sub> O <sub>3</sub>			0.54			
UO <sub>2</sub>	4.69	1.36	0.25	14.12	6.63	7.12
UO <sub>3</sub>	9.97	2.88	0.52			
Y <sub>2</sub> O <sub>3</sub>	0.78	6.54	7.45	13.27	30.42	18.06
Ce <sub>2</sub> O <sub>3</sub>	2.29	8.62	3.25	1.92	6.01	3.65
Al <sub>2</sub> O <sub>3</sub>	0.58	2.40	0.29	0.26		0.15
FeO	4.38	15.46	9.53	2.58	0.63	8.95
Mn <sub>3</sub> O <sub>4</sub>	0.65	1.84	1.07			
MnO				0.41	0.38	0.21
MgO	0.40	0.76	0.87	0.56	0.04	0.65
CaO	6.76	6.84	1.36	3.44	1.98	4.58
PbO	0.75	0.74	0.22	2.18	1.45	0.65
CuO	0.08				0.15	
Bi <sub>2</sub> O <sub>3</sub>	0.06					
WO <sub>3</sub>		tr.	tr.			
Ge	?	?	?			
Se	tr.					
S	0.27	0.11		1.08	0.31	
H <sub>2</sub> O	10.09	6.90	4.73	9.01	4.65	4.58?
In original	99.20	100.92	98.92	100.59	98.18	96.62
			98.93	100.87	98.21	100.00
Sp. gr.	3.805	3.750	4.875	3.950	4.997	

I. Wiikite, straw-yellow, amorph., from Nuolainniemi, Impilahti (Wuorinen 1916, p. 16).

II. » black, amorphous, » » » » » » » » » »

III. »

IV, V and VI. Wiikites from Lokansaari, Impilahti (Ant-Wuorinen 1936, p. 215).

Analyses I and II in Table 20 are exceptions, there the case is the reverse. Pointing to the difficulties which are met with in the analyses (Anal. I—III, Table 20) when separating these groups from each other, the analyst says of the results that they are only »eine grobe Annäherung der Wirklichkeit» (Wuorinen 1916, p. 15). We can, thus, regard as a rule that in the case of wiikites the yttrium group is as to its amount predominant compared with the cerium group.

Of the iron, which in analyses I—III is given only as FeO, it is said (op. c., p. 17): »Die weiteren Analysen und besonders das Berechnen der Formel werden zeigen, inwiefern die Bestimmung des Oxydationsgrades dennoch notwendig sein wird». This has probably not been necessary, as the iron also in the later analyses (Anal. IV—VI, Table 20) is given only as FeO.

In analyses I—III the uranium is given as the oxides  $UO_2$  and  $UO_3$ , the ratio between the molecular proportions of which in all three analyses is 0.5 (0.499, 0.501 and 0.507), accordingly very much the same as the ratio  $UO_2 : UO_3 = 0.472$  in the oxide  $U_3O_8$ . This is very strange, especially as this is the case in all three analyses.

In analyses IV—VI the uranium is given only as  $UO_2$ .

Since nothing else is mentioned in the original publication of wiikites IV—VI, but only their analyses and the specific gravity for two of them are given, it is very difficult to come to conclusions as to the mineralogical properties of these minerals. It may be regarded as possible that wiikite IV, to conclude from the high water-content and the fairly low specific gravity, is remarkably altered and that it is amorphous and dark brown. Wiikite V is black, judging from the fact that it contains a moderate amount of uranium and unusually much rare earths, that its water-content is comparatively low and specific gravity high. Whether this wiikite is amorphous or crystallized, cannot be said. Any conclusion as to the properties of wiikite VI is most uncertain, since its specific gravity is not given, its water-content in the analysis (4.58 %) is fitted with a note of interrogation and the sum is 100 % though it, according to the analysis, should be 96.62 %. Whether the water content 4.58 % is too low and the difference 3.32 % contains water, cannot, of course, be said for certain, but, according to the analysis, it seems probable that a considerable part of the difference is water. On this ground it may be regarded as possible that also this wiikite has considerably altered, is amorphous and very dark.

The water-content in analyses IV—VI is given in the original publication only as  $H_2O+$ , concluding from which it is the question of total water ( $H_2O$ ), as stated in Table 20.

Analyses I—VI in Table 21, published earlier, refer to wiikites, the mineralogical properties of which I have described before in detail (1928, pp. 15—20). Above a short mention has been made of these properties

Table 21. Wiikites from Impilahti

	Anal. Lauri Lokka							
	I	II	III	IV	V	VI	VII	VIII
Nb <sub>2</sub> O <sub>5</sub>	22.75	25.64	29.70	28.98	28.34	24.36	21.16	26.41
Ta <sub>2</sub> O <sub>5</sub>	13.09	11.80	16.53	14.69	14.25	11.08	18.13	19.95
TiO <sub>2</sub>	14.35	5.25	3.64	3.45	11.52	6.06	6.76	3.42
SnO <sub>2</sub>	none	none	0.03	tr.	0.04	0.04	tr.	tr.
SiO <sub>2</sub>	2.30	2.85	4.79	4.84	4.94	12.59	12.91	7.52
ZrO <sub>2</sub>	0.20	0.17	0.33	0.32	0.20	tr.	tr.	tr.
ThO <sub>2</sub>	0.70	0.10	0.38	0.29	1.10	0.44	0.86	0.72
U O <sub>2</sub>	5.28	4.98	8.47	7.87	9.93	4.33	0.50	4.55
U O <sub>3</sub>	none	none	none	none	none	11.24	12.28	4.09
Y <sub>2</sub> O <sub>3</sub>	22.30	29.27	13.40	12.88	7.60	1.70	1.65	10.79
Ce <sub>2</sub> O <sub>3</sub>	6.24	3.93	1.88	1.50	0.52	1.12	0.77	0.73
Al <sub>2</sub> O <sub>3</sub>	0.12	0.18	0.45	0.66	0.52	0.75	0.17	0.21
Fe <sub>2</sub> O <sub>3</sub>	3.71	5.26	5.97	5.80	3.25	5.54	5.75	4.19
FeO	0.20	0.15	0.46	0.61	0.82	0.87	0.11	0.78
MnO	0.13	0.03	0.05	0.10	0.17	0.27	0.25	0.28
MgO	0.20	0.27	0.38	0.29	0.89	1.00	0.20	0.41
CaO	3.00	2.43	4.25	3.92	5.02	6.48	6.42	4.40
PbO	0.44	0.90	1.49	3.08	1.88	none	none	0.29
S	0.08	0.20	0.32	1.24	0.38	none	none	0.11
C O <sub>2</sub>	none	none	none	none	none	1.35	1.48	none
H <sub>2</sub> O+	4.19	5.91	6.12	7.23	5.97	6.21	6.00	8.00
H <sub>2</sub> O—	0.34	1.03	1.61	2.56	2.86	4.88	4.80	3.07
	99.62	100.35	100.25	100.31	100.20	100.31	100.20	99.92
Sp. gr.	4.708	4.591	4.349	4.223	4.200	3.635		4.109
H.	5.5	5.5	5.5	5.5	5.0	5.0		5.0

I.	Wiikite, black, with faces,	from	Hunttila,	Impilahti (Lokka 1928, p. 43.
II.	»	»	»	Lokansaari, » » » »
III.	»	»	amorphous	» Hunttila, » » » » 44.
IV.	»	»	»	» Lokansaari, » » » » 45.
V.	»	dark brown	»	» Nuolainniemi, » » » »
VI.	»	grey-yellow	»	» » » » 46.
VII.	»	straw-yellow	»	» » » »
VIII.	»	dark brown	»	» Mursula, »

when discussing the mineralogical properties of wiikites, so there is no reason to touch upon them any further. It may, however, be pointed out also in this connection that in the microscopic examination of one sample of wiikite (Fig. 6) abundance of lead ore was established. The lead-content of said sample (Anal. IV, Table 21) is very high in consequence. Only a small part of the lead may be of radioactive origin, the greatest part being common lead.

Analyses VII and VIII in Table 21 are new. The former is made of a wiikite which is lighter yellow than wiikite VI, but very similar as to its physical properties. I have earlier used the same powder of which analysis VII is made in the determination of radioactivity (1928, p. 20 Lg) where this mineral proved to be similar to wiikite VI. I determined then also CO<sub>2</sub>- and the water-content. Each determination gave quite the same result as in analyses VI. Under these circumstances I did not see the necessity of carrying out an analysis of wiikite VII, I supposed its chemical composition to be similar to wiikite VI, which analysis VII

also shows to be right. Analyses VIII is made of the dark brown, amorphous wiikite of Mursula, this being the only analysis of the wiikites of Mursula, which are found also black and yellow.

The wiikites are silico-titano-tantaloniobates. They are ortho-, meta- or paracompounds, as may be concluded from the analysis of the less altered wiikites.

The amounts of the basic chief constituents, uranium and rare earths, are very variable. Wiikites contain also  $\text{ThO}_2$ ,  $\text{ZrO}_2$  and  $\text{SnO}_2$ , of the two last mentioned often very little. It has also been established that wiikite contains scandium. The other usual constituents of the minerals appear generally in small amounts, with the remarkable exceptions of the iron-content in two analyses (Anal. II, Table 19 and Anal. II, Table 20).

The relative amount of yttrium earths in rare earths, calculated from the analyses, is shown in Table 22, A. The analyses are given there in order according to the colour of the mineral and the amount of rare earths. The two oldest incomplete analyses (Table 19) have been left out.

Table 22. Wiikites from Impilahti

A. Chemical Analyses							B. Spectrographic Analyses				
No.	Tab.	Anal.	Colour of Mineral	RE %	YE		No.	Colour of Mineral	RE %	YE	
					%	% of RE				%	% of RE
1	20	V	Black	36.43	30.42	84	1	Black	18.17	16.13	89
2	21	II	»	33.20	29.27	88	2	»	8.41	7.67	91
3	»	I	»	28.54	22.36	78	3	»	7.59	6.69	88
4	20	VI	»	21.71	18.06	83	4	»	6.80	6.34	93
5	21	III	»	15.28	13.40	88	5	Dark brown	12.04	10.38	86
6	20	II	»	15.16	6.54	43	6	Yellow	2.55	1.68	66
7	21	IV	»	14.38	12.88	90					
8	20	III	»	10.70	7.45	70					
9	»	IV	Dark brown	15.19	13.27	87					
10	21	VIII	»	11.52	10.79	94					
11	»	V	»	8.12	7.60	94					
12	20	I	Yellow	3.07	0.78	25					
13	21	VI	»	2.82	1.70	60					
14	»	VII	»	2.42	1.65	68					

Black and dark brown wiikites form the great majority, 11 analyses, the total amount being 14. Since 9 of these 11 contain 78—94 % of yttrium earths of the total amount of the earths, one 70 % and only one less than 50 % (43 %, No. 6), these wiikites may with good reason be considered as yttrium minerals.

Yellow wiikites contain, according to three analyses, remarkably less rare earths — only 2—3 % — than the dark kinds. Two among them are also yttrium earth minerals (Nos 13, 14), but compared with dark wiikites they contain proportionately less yttrium earths, only 60—68 %

of the rare earths. A very remarkable exception is No. 12, where this percentage is only 25, and according to which this wiikite ought to be a cerium mineral.

Sahama and Vähätalo (1939) have spectrographically examined the content of rare earths in wiikites of Impilahti. They have analysed six different samples of wiikites, which have not been chemically analysed. The content of rare earths and yttrium earths in these six minerals (op. c., p. 102, Table II) is given in Table 22 part B, which has been arranged in the same way as part A.

Table 22 shows that the spectrographic analysis has given a result very similar to that of the chemical analysis, with regard to rare earths of both darker and also yellow wiikites. Sahama and Vähätalo also point out, on the basis of their analyses, that the preponderance of yttrium earths is smaller in yellow than in other wiikites (op. c., p. 105).

Accordingly, we get the following result. In twenty different wiikites the content of rare earths has been determined, some chemically, some spectrographically. It varies very much, being in the darker about 7—37 % and in the yellow about 2—3 %. Over 50 % of the rare earths are yttrium earths even over 80 % in most of the darker wiikites, in the yellow 60—68 %. There are two exceptions (Table 22, A) to these values, No. 6 black wiikite 43 % and No. 12 yellow only 25 %. Accordingly, it can be said that wiikites are yttrium minerals.

Mention may also be made of the scandium-content, not only wiikites but also in monazites and allanites. I have had access to the following literature.

According to Crookes (1908) the scandium-content of the analysis of wiikite, 1.17 %  $\text{Sc}_2\text{O}_3$  (Table 19), is the highest scandium-content stated in the analyses of this mineral. This wiikite is balek.

Eberhard (1910), when examining the distribution of scandium throughout the globe, has determined the scandium-content of a great many different minerals, among these twelve minerals from Impilahti, of which eleven wiikites and one monazite. The colour of the wiikites was changeable, black, brown, dark grey, grey and yellow. Almost all types of wiikites of different colours were thus presented, on the basis of which it may be supposed that also the chemical composition of these wiikites is very dissimilar.

Eberhard established that all the others except the yellow wiikite contained scandium. He mentions the monazite and the four black wiikites as the most rich in scandium. »Es fanden sich — — — mit grösserem Skandiumgehalt — — — Monazit (No. 643) und einige Wiikite (648, 650, 653, 655) — — —» (op. c., p. 404). One black sample of wiikite proved to be scandium-poor.

Borgström (1910) makes mention of the scandium-content of seven wiikites, different in colour, from black to straw-yellow. Most of them

contain only traces of scandium. The scandium-content of one black wiikite is some tenths of one per cent, another black one is richer in scandium, but the content of it is probably below 1 %.

Two allanites analysed by Meyer (1911) are in a very noticeable degree scandium-containing, 0.80 % and 1.00 %  $\text{Sc}_2\text{O}_3$  (Table 10).

Of the wiikites analysed by Wuorinen (1916) one contains 0.54 %  $\text{Sc}_2\text{O}_3$  (Table 20). Also this wiikite is black.

Tschirwinsky (1922) mentions the scandium-content of both wiikite and allanite.

Of the former he says first (p. 257): »Bekanntlich hat Crookes in Wiikite Scandium entdeckt (1.2 %)». Then he remarks: »Dagegen soll nach Winter kein Scandium vorhanden sein. — — — Dem schwarzen Wiikit fehlt Scandium».

The scandium-content of allanite is according to Tschirwinsky only 0.13 %  $\text{Sc}_2\text{O}_3$ , thus only a fraction of the lowest of the above mentioned values 0.54 %  $\text{Sc}_2\text{O}_3$ . He gives a comparison between the corresponding data of the determinations by Orlov and the analysis by Meyer (Anal. I, Table 19):

Orlov:	$\text{Sc}_2\text{O}_3$	0.13,	$\text{ThO}_2$	0.75,	Rare Earths	14.58 %
Meyer:	»	0.80,	»	1.32,	»	16.45 %

Sahama and Vähätalo (1939, p. 108) mention the scandium-content of said wiikites (Table 22, B): »The optical spectra photographed with Zeiss Three Prism Spectrograph showed, however, well determined scandium lines in all of the samples».

The preceding makes it clear that the reports of the scandium-content in the wiikite and the allanite from Impilahti are very contradictory. According to some investigations the scandium-content at least of black wiikite and allanite is considerable, each about 0.5—1.0 %  $\text{Sc}_2\text{O}_3$  according to which these minerals could be regarded as scandium minerals, but account of the low scandium-content they are not, of course, to be compared to thortveitite. According to other investigations it has only been established that especially black wiikite contains scandium, but no percentages have been given. It is also mentioned that black wiikite does not contain scandium and that the scandium-content of the allanite is rather low. Thus it may, however, be regarded as probable that at least some wiikites and the allanite contain scandium, but in a small degree.

Since the wiikite as to its chemical composition is more or less altered in general, we may relate the reason for this. For comparison mention may also be made of the allanite and the monzite.

The cause of the alteration of wiikite and allanite is the radioactivity of these minerals, which property has been referred to before, when discussing their mode of occurrence and their influence on the nearest environment.

Since I earlier and more in detail (1928, pp. 22—29) have presented the determination of the radioactivity of wiikite and allanite and its results, we may here mention only the following. Of these three minerals wiikite is the most radioactive, because of the considerable uranium-content. Second comes monazite, the thorium-content of which is higher than the uranium-content of some wiikites, but nevertheless its radioactivity is, of course, remarkably lower. The third is allanite, the thorium-content of which is only a fraction of that of the monazite, and the radioactivity accordingly very weak.

Because of the radioactivity the primarily crystalline substance of the wiikite has been transformed into amorphous, glassy material, the mineral has changed to a metamict state. But this has not been caused solely by radioactivity, which in some wiikites is considerable. The cause of this internal change is that wiikites are compounds of weak bases (yttrium earths) and weak acids (earth acids). Such a material can take up much water and also other substances. At the same time also leaching takes place. Wiikite is such a typical radioactive mineral the chemical composition of which is more or less altered.

Allanites are weakly radioactive because of the low thorium-content — there are also thorium-free allanites, according to analyses. Being compounds of strong bases (cerium earths) and weak acid ( $\text{SiO}_2$ ), they can be either crystalline or metamict. In the latter case their chemical composition has more or less altered. The three allanites, described earlier (Table 10), are a good example of this.

The monazite, containing considerably more thorium than allanites in general, is accordingly more radioactive too, but it is always crystalline, being a compound of strong bases (cerium earths) and a strong acid (phosphoric acid). A total analysis — partly incomplete — of such fresh monazite is analysis I in Table 11. Like the non-radioactive minerals, the monazite may, of course, also be weathered and decomposed (Anal. II, Table 11).

#### ON THE GROUPING AND THE FORMULA OF WIIKITE

A certain regularity prevails between the characteristic physical and chemical properties of wiikites, as easily seen from those wiikites of which total analyses have been performed. But this may be established also with regard to those wiikites of which only some separate determinations have been made, when they concern only characteristic properties, such as in Borgström's investigation (1910). Different types of wiikites have been described there, the colour of which varies from black to straw-yellow, in some crystal faces have been established, some are amorphous. The specific gravities and water-contents of them all have been determined.



It has been established that the different types contains very different amounts of rare earths. On the basis of qualitative reactions also niobium, titanium and uranium are mentioned. These determinations have been performed with regard to samples, the scandium-content of which has been stated before, according to Borgström.

Earlier I have grouped the wiikites into four groups in the form of a table (op. c., p. 53, Table 14), on the basis of data mentioned previously. With the exception of some instances, all other wiikites, 10 analysed and those examined by Borgström, could be placed in these groups in such a way that, when passing from one group to another, the most characteristic properties increase or decrease.

From this grouping one wiikite has been left out, its chemical composition being (Anal. III, Table 20) to that extent unique that it cannot be placed in any of these groups. Its uranium-content is quite small compared with the corresponding lowest values of other analysed wiikites. The thorium-content again is considerably higher than in the case of wiikites in general, corresponding to that of the analysed nuolaites (Table 12). This wiikite is also otherwise near the nuolaites, according to its chemical composition. It may be said that it is an intermediate form between wiikites containing uranium and the uraniumfree kindred minerals, nuolaites. — Neither was one of the wiikites, examined by Borgström, quite suited to any group.

In Table 23 this grouping is given in its original form, with the exception of some values of limit having been somewhat changed (the original values in brackets). This has been done in order that the wiikites analysed later can be better placed in the groups. This could have been possible even without removing the limits, as these values of limit must not, of course, be considered absolute, but only as approximate values.

Table 23. Range of Properties of wiikites from Impilahti

		$\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$ $+ \text{TiO}_2 + \text{SiO}_2$	Rare earths	$\text{U}_3\text{O}_8$	$\text{H}_2\text{O}$	Sp. gr.	Hardness
I	Black, dark, with faces	45—55	25—37 (35)	tr. —7 (5)	4—7	4.5—4.8	ç. 5.5
II	Black, dark. Asphalt-like. Amorphous	45—55	10—22 (18)	4—14 (10)	6—10	3.8—4.4	ç. 5.5
III	Dark brown to brown. Asph.-like. Amorphous	50—57 (55)	5—12 (10)	(10) 9—15	9—11	3.7—4.2	ç. 5
IV	Grey-yellow to Straw-yellow. Wax-like. Amorphous	55—65	<5	(14) 13—16	10—12	3.6—3.9	ç. 4.5—5.0

If in the future new analyses give cause there will still be a possibility of removing the limits without changing the original character of the table.

The analysed wiikites (Table 19—21) can well be placed in the groups I—IV of Table 23 as follows:

	Gr. I	Gr. II	Gr. III	Gr. IV
Table 19. Analyses		II		I
» 20. »	V	II, IV, VI		I
» 21. »	I, II	III, IV	V, VIII	VI, VII

To each group have thus been added new analyses, 5 in number. Of the analysed wiikites, 16 in number, a great part (6) belongs to group II and the smallest (2) to group III. One not belonging to any group has been left out from the grouping, as mentioned above.

Holmes (1931, p. 263) presents the original table with the addition of the uranium-free nuolaites as a different group, before group I of the wiikites.

The chemical composition of wiikites cannot be given by a common and ordinary formula, on the basis of analyses. There is no real ground for it, because wiikites are generally more or less altered as regards, their original composition.

Ant-Wuorinen (1936) has, however, tried to work out such a formula for wiikites. In the beginning of his communication (p. 213) he says i. a.: »Ordnungshalber werden hier die Analysenresultate von sämtlichen dem Verfasser bekannten Wiikitanalysen beigefügt».

These 17 analyses of nuolaite, loranskite and wiikite have above been presented in these tables: two analyses of nuolaite, I and II in Table 12; one analysis of loranskite, II in Table 13; two analyses of wiikite in Table 19, six analyses of wiikite in Table 20, and six analyses of wiikite, I—VI in Table 21. Accordingly, besides the analyses of the uranium-free minerals nuolaite and loranskite, 14 analyses of wiikite.

It is very strange that, in working out the formula attention has been paid only to six analyses, namely the analyses I—VI in Table 21. With the exception of two analyses (Table 19), which are rather incomplete, the writer has in his communication totally disregarded even his own analyses, not mentioning a word of them. It is incomprehensible why these are presented only »ordnungshalber». On this account and solely for the reason mentioned earlier (alteration), there is no reason for treating the formula of wiikite, presented on the basis mentioned.

The findings and conclusions concerning the relations between some constituents, which are made in the above mentioned communication — certainly based only upon six analyses — in order to work out a formula, lead, however, to the following critical examination. Without selection all complete newer analyses of wiikite have been considered (Table 20, 21), 14 in all, leaving out only the two oldest incomplete analyses (Table 19).

On the ground of the analyses in Tables 20 and 21, Table 24 with four parts A—D has been made up for this examination. The molecular proportions of the oxides, group of oxides and sums with three decimals — which precision is here sufficient — have been given in the table multiplied by 1 000. In parts A and B under the sums of the molecular proportions their ratios are marked, keeping on each line as unit (100) the smaller molecular proportion. In parts C and D similar notes are under the quotients.

Table 24, A 1.  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$  in the original analyses,

The variety of the molecular proportions of the oxides  $\text{SiO}_2$ ,  $\text{TiO}_2$ ,  $\text{Nb}_2\text{O}_5$  and  $\text{Ta}_2\text{O}_5$  in the original analyses (I—VI, Table 21) is diagrammatically given in the communication mentioned above, and there it is stated (p. 222): »Die Niob- und Tantelmenge bleibt die ganze Zeit einigermassen konstant, während dagegen die Kieselsäure und besonders das Titanoxyd sich sichtlich verändern». This is quite right with regard to the oxides  $\text{SiO}_2$  and  $\text{TiO}_2$ , which is easily seen already from the analyses (Table 21), but neither is the variety of the molecular proportions in the case of the oxides  $\text{Nb}_2\text{O}_5$  and  $\text{Ta}_2\text{O}_5$  so very small in these six analyses, as Table 24 shows. In the analyses of Table 20 these varieties are still more remarkable. Accordingly, it can be established, without even the new analyses VII and VIII in Table 21, that the sums of the molecular proportions of the oxides  $\text{Nb}_2\text{O}_5$  and  $\text{Ta}_2\text{O}_5$  are very different, seeing that differences of over 100 % are found in the ratios.

In this connection it may be pointed out that the oxides  $\text{Nb}_2\text{O}_5$ ,  $\text{Ta}_2\text{O}_5$ ,  $\text{TiO}_2$  and  $\text{SiO}_2$ , which in the analyses of wiikites vary rather greatly as to their amount, substitute one another in the group  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5 + \text{TiO}_2 + \text{SiO}_2$ , which in the different types of wiikites varies somewhat in regard to its amount (Table 23).

A different opinion is found in the said communication. Supposing the minerals were to be free from  $\text{SiO}_2$  and  $\text{TiO}_2$ , leaving these oxides out from the analyses, the variety of molecular proportions of the mixed oxides  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$  in the analyses, which have been taken into consideration in the communication (Anal. I—VI, Table 21) are diagrammatically stated with »Summenkurve». On the basis of »Summenkurve» it is said: »Dies bringt den Gedanken nahe, dass immerhin ein beachtenswerter Teil des im Wiikit enthaltenen Titans und der Kieselsäure aus fremden Mineralien stammen könnte». Unfortunately the: »Summenkurve» is faultily drawn, giving rise to an erroneous interpretation.

Even the faulty »Summenkurve» is curved to such an extent that hardly even on the basis of it can one reason as is done in the quotation and still less on the basis of a rightly drawn »Summenkurve». It is as curved — though not, of course, of the same form — as the »Summenkurve» drawn on the basis of the original analyses, which is non presented in the communication. The same result is achieved in the following.

Table 24, A 2.  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$  in the analyses, from which  $\text{SiO}_2$  and  $\text{TiO}_2$  are excluded.

It is easily seen from the table that the sums of the molecular proportions vary as much as in the foregoing case (A 1), though not, of course, in the same order. Those six analyses are not even an exception, which has been taken into consideration in the communication. The result is the same as before, the hypothesis mentioned in the quotation cannot be based on the analyses of the minerals.

An interesting rule stated in the said communication (pp. 225—226) gives further cause to examine the ratios between the molecular proportions of some constituents of wiikites. In the rule attention has been paid only to six analyses (Anal. I—VI, Table 21), excluding  $\text{SiO}_2$  and

Table 24

	Table 20						Table 21							
	I	II	III	IV	V	VI	I	II	III	IV	V	VI	VII	VIII
A. $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$														
1. Original Analyses														
$\text{Nb}_2\text{O}_5$	79	77	209	164	135	119	85	96	112	109	107	92	80	99
$\text{Ta}_2\text{O}_5$	11	2	0	5	7	3	30	27	37	33	32	25	41	45
Sum	90	79	209	169	142	122	115	123	149	142	139	117	121	144
Ratio	114	100	265	214	180	154	146	156	189	180	176	148	153	188
2. Exclusive of $\text{SiO}_2$ and $\text{TiO}_2$														
$\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$	132	102	234	178	152	142	139	133	162	156	166	143	150	164
Ratio	129	100	229	175	149	139	136	130	159	153	163	140	147	161
B. $\text{RO} + \text{R}_2\text{O}_3$														
1. Original Analyses														
RO	204	388	194	127	57	228	65	57	99	101	134	157	125	94
$\text{R}_2\text{O}_3$	16	75	42	59	136	82	129	159	99	97	56	52	46	72
Sum	220	463	236	186	193	310	194	216	198	198	190	209	171	166
Ratio	133	279	142	112	116	187	117	130	119	119	115	126	105	100
2. Exclusive of $\text{SiO}_2$ and $\text{TiO}_2$														
RO	299	497	218	133	61	265	78	62	108	111	160	192	155	107
$\text{R}_2\text{O}_3$	23	96	46	63	145	96	155	172	108	106	67	64	57	82
Sum	322	593	264	196	206	361	233	234	216	217	227	256	212	189
Ratio	170	314	140	104	109	191	124	124	114	115	120	135	112	100
C. $\text{UO}_2 + \text{UO}_3 / \text{RO}$														
$\text{UO}_2 + \text{UO}_3$	52	15	3	52	25	27	20	18	31	29	37	55	45	31
RO	204	388	194	127	57	228	65	57	99	101	134	157	125	94
Quotient	0.25	0.04	0.02	0.41	0.44	0.12	0.31	0.32	0.31	0.29	0.28	0.35	0.36	0.33
Ratio	13	2	1	21	22	6	16	16	16	15	14	18	18	17
D. $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5 / \text{RO} + \text{R}_2\text{O}_3$														
Quotient	0.41	0.17	0.89	0.91	0.74	0.39	0.59	0.57	0.75	0.72	0.73	0.56	0.71	0.87
Ratio	24	10	52	54	44	23	35	34	44	42	43	33	42	50

TiO<sub>2</sub>. In the following examination all 14 analyses have been considered, both in their original form and exclusive of SiO<sub>2</sub> and TiO<sub>2</sub>, following otherwise the same order as in the said rule. The three parts of the rule will be mentioned in connection with the corresponding parts B, C and D in Table 24.

Table 24, B 1. RO and R<sub>2</sub>O<sub>3</sub> in the original analyses.

The molecular proportions of both oxide groups vary unusually in the different analyses, RO = 57—388 and R<sub>2</sub>O<sub>3</sub> = 16—159. The variation of them is less in the case of the sums RO+R<sub>2</sub>O<sub>3</sub>, but still very remarkable RO+R<sub>2</sub>O<sub>3</sub> = 166—463. Especially in the analyses in Table 20 do very great differences appear.

Table 24, B 2. RO+R<sub>2</sub>O<sub>3</sub> in the analyses, from which SiO<sub>2</sub> and TiO<sub>2</sub> are excluded.

Excluding the oxides SiO<sub>2</sub> and TiO<sub>2</sub> from the analyses no better result than before is obtained, as the molecular proportions of the sums RO+R<sub>2</sub>O<sub>3</sub> vary equally in each case, though not in the same order. It happens further that in those six analyses (Anl. I—VI, Table 21) which have been chosen as a basis of the above mentioned rule, the greatest difference between the molecular proportions (RO+R<sub>2</sub>O<sub>3</sub>) is in this case (256—215 = 30) greater than in the original analyses (216—198 = 18). This by no means agrees with the first part of the rule: »Die Summe der Molekülproportionen der Oxyde der zwei- und dreiwertigen Metalle ist eine Konstante».

In the two last parts, C and D, in Table 24, where the quotients of the molecular proportions are in question, it is, of course, all the same, whether the molecular proportions have been calculated on the basis of the original analyses or on such from which SiO<sub>2</sub> and TiO<sub>2</sub> are excluded. In the following the calculations have been made on the basis of the original analyses.

Table 24, C. UO<sub>2</sub>+UO<sub>3</sub>/RO.

The second part of the said rule is: »Die Summe der Molekülproportionen der Uranoxyde ist der dritte Teil der Summe der Molekülproportionen der Oxyde zweiwertiger Metalle». Such is not the case even in the chosen analyses. Regarding all 14 analyses this quotient varies very much, 0.02—0.44. Accordingly, the quotient in question is not  $\frac{1}{3}$  in the analyses of wiikites, but a rather variable figure.

Table 24, D. Nb<sub>2</sub>O<sub>5</sub>+Ta<sub>2</sub>O<sub>5</sub>/RO+R<sub>2</sub>O<sub>3</sub>.

The third part of the rule runs as following: »Die Summe der Molekülproportionen der Nioboxyde und der Tantaloxxyde ist nahezu  $\frac{3}{4}$  der Summe der Molekülproportionen der zwei- und dreiwertigen Metalle». From the value  $\frac{3}{4}$  three quotients differ considerably even in those six analyses, which have often been mentioned earlier. In all 14 analyses this quotient varies 0.17—0.91 on the basis of which it can be said that

even in this case there is no regularity, not even approximately, but that the quotient varies very much.

The critical examination presented above, based on the values in Table 24, may be summarized as follows.

$\text{Nb}_2\text{O}_5$ ,  $\text{Ta}_2\text{O}_5$ ,  $\text{TiO}_2$  and  $\text{SiO}_2$  vary quite a lot as to their amounts in the different analyses. The excluding of the oxides  $\text{SiO}_2$  and  $\text{TiO}_2$  from the analyses (Table 24, A 2) does not produce such an effect that the molecular proportions of the sums  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5$  — in the original analyses (Table 24, A 1) they are fairly variable — may come into better agreement. This would support the supposition mentioned before that  $\text{SiO}_2$  and  $\text{TiO}_2$  do not, at least to any considerable extent, belong to the ordinary constituents of wiikites.

The molecular proportions of the sums  $\text{RO} + \text{R}_2\text{O}_3$  vary a good deal in the original analyses (Table 24, B 1) and they do not agree better by excluding the oxides  $\text{SiO}_2$  and  $\text{TiO}_2$  (Table 24, B 2), even if they, according to the rule above, become equal.

The quotients of the molecular proportions  $\text{UO}_2 + \text{UO}_3 / \text{RO}$  vary 0.02—0.44. According to the said rule they are  $\frac{1}{3}$ .

The quotients of the molecular proportions  $\text{Nb}_2\text{O}_5 + \text{Ta}_2\text{O}_5 / \text{RO} + \text{R}_2\text{O}_3$  vary 0.17—0.91. According to the said rule they are »nahezu  $\frac{3}{4}$ ».

These results are an addition to what has been said above of the variety of the chemical composition of wiikites. They also indicate that the rule presented in the said communication and quoted before in parts B, C and D, is based on erroneous numerical values.

Even though an ordinary formula presenting the chemical composition of wiikites based on analyses is out of question, still considerable regularity with regard to the physical and chemical properties may be established as Table 23 shows.

## ON THE GEOLOGICAL AGE OF THE RADIOACTIVE MINERALS FROM SOUTHWESTERN FINLAND AND IMPILAHTI

Of the 35 analyses (Tables 1, 2, 4, 6, 10—13 and 14—21) of the radioactive minerals from southwest Finland and Impilahti the greatest part has to be disregarded when estimating the geological age of these minerals on the basis of the analyses, because the minerals have altered in such high degree.

As far as wiikites are concerned, I have earlier calculated and presented for comparison (1928, p. 63 Table 18) the ages of all of those of which complete analyses had been performed, with the exception of one, which, concluding from its chemical composition, had been most altered. Four of the calculated eight ages of wiikites were improbable and one of them has to be specially mentioned also here.

This wiikite, relatively rich in lead (Anal. IV, Table 21), contains a remarkable amount of common lead, as the microscopic examination showed and as mentioned already earlier. For this reason its calculated age is exceptionally high, over 2500 m.y. As this value has given rise to an erroneous interpretation, the following may in this connection be mentioned with regard to the age of this wiikite: »Auf Grund dessen, was die optische Untersuchung — — aufweist (Erzbildung), kann sein Blei nicht radioaktiven Ursprungs sein. Deshalb entspricht sein berechnetes Alter auch nicht seinem wahrscheinlichen Alter» (Lokka 1928, p. 64).

In these circumstances 10 age determinations remain which may be regarded as probable. They are given in Table 25. Three of them are new (Nos. 1, 2 and 4). The seven other determinations have first been stated in my publications: No. 3, 1935, pp. 9—10, 12 and Nos. 5—10, 1928, pp. 62—65.

In the last mentioned (p. 63, Table 18) in the case of one wiikite (No. 7 in Table 25) the uranium-content is given as 3.73 %, it should, however, be 3.60 %. The lead ratio 0.156 changes correspondingly to the value 0.162. The difference has in fact very slight significance. Also some other corrections have been made, but they are quite insignificant.

When calculating the age, the logarithmic formula

$$\text{Age} = 15150 \log \left( 1 + \frac{1.155 \text{ Pb}}{\text{U} + 0.36 \text{ Th}} \right) \text{ m. y.}$$

has been used.

Besides what has been said above of the correction of some analytical values, it must also be mentioned that when calculating the age of the minerals the earlier values of some constants in question have somewhat changed, causing slight alterations of ages calculated earlier.

For the uranium minerals that age has been denoted in the last column of the table, where the correction for actino-uranium has been considered (Holmes 1937, p. 153).

Table 25. The Age of Radioactive Minerals from S. W. Finland and Impilahti

	U %	Th %	Pb %	Pb U+0.36 Th	Age in m. y.	
					Calc. fr. log. form.	Corr. for AcU
1. Gadolinite, Lövböle, Kemjö Tab. 2 .....		0.629	0.037	0.163	1138	
2. Allanite, Äva, Brändö » 4 .....		0.469	0.027	0.160	1116	
3. » Varala, Kangasala » 6 .....		1.79	0.106	0.164	1114	
4. Monazite, Mursula, Impilahti » 11, An. III		7.73	0.467	0.168	1165	
5. Nuolaite, Nuolainniemi » 12 » I		2.90	0.19	0.182	1255	
6. » » » » » II		1.37	0.25	0.179	1239	
7. Wiikite, » » » 20, » II	3.60	1.83	0.69	0.162	1129	1081
8. » Lokansaari » » 21, » II	4.39	0.09	0.84	0.190	1305	1195
9. » Hunttila » » » III	7.47	0.33	1.38	0.182	1254	1152
10. » Nuolainniemi » » » V	8.75	0.97	1.75	0.192	1320	1207

As Table 25 shows, the ages agree rather well as regards the minerals from southwestern Finland, the gadolinite and the two allanites. These ages may be regarded as most probable, because the determination of them is based on the analyses of fresh minerals.

Of the minerals of Impilahti the monazite is without doubt fresh, therefore, its age determination has to be regarded as the most reliable. Also the other ages of the minerals from Impilahti, given in the table, are fairly equal. It must, however, be considered that these minerals have altered so much that their age determination, based on analytical results, may be regarded as only probable, but not quite exact.

The ages of the minerals from southwestern Finland and Impilahti do not differ much from one another, as is seen in Table 25. Yet it seems that the age of the minerals from Impilahti should be considered as being somewhat higher.

For comparison there is reason to pay attention in this connection also to the age of radioactive minerals discovered in Russian North Carelia, in the region of the Polar Circle, near the White Sea (Sinjaja Pala and some other localities), which question I have treated already earlier (1928, pp. 67—68). Some years previously Nenadkewitsch (1926, p. 772) had published his analyses of the uranitite from Sinjaja Pala, on the basis of which Fersman (1926, p. 776) had calculated the age of this mineral at 1950 m. y. Of this age, seemingly too high, I have earlier (1928, p. 68) said: »Es scheint, als ob der Alterswert — — — eine fühlbare Korrektur forderte, ehe wir dem wirklichen Alter nahe kommen. Denn vom geologischen Standpunkte betrachtet ist es sehr wenig wahrscheinlich, dass das Alter des Uraninits von Sinjaja Pala eine so grosse Abweichung von dem der Wiikite und Nuolaite von Impilahti aufweisen könne, wie es den Anschein hat. Also können auch keine näheren Schlüsse gezogen werden, in welchem Altersverhältnisse die Pegmatitgänge, deren Minerale die Uraninite sind, zu den Pegmatitgängen von Impilahti stehen, vorin die Wiikite und Nuolaite vorkommen».

Neither have the corrections of this apparently too high age of the uranitite from Sinjaja Pala containing common lead, performed later, led to a reliable result. Long after Fersman had first published (1926) the age of this mineral, Labuntsov reports (1935, p. 646) that it is 2125 m. y., and that the age of a monazite discovered at Tschernaja Salma, also in Russian North Carelia, is fairly similar, 2 114 m. y. Pointing to the equal ages of these minerals, rather different to their chemical composition, Labuntsov considers that on the basis of same the age of the pegmatites from Russian North Carelia may be estimated as being over 2 100 m. y.

Holmes says (1937, p. 192) of the age of these minerals i. a.: »Actually the age of the monazite is about 1520 m. y., and this estimate is likely



to be more reliable than any based on the galena-bearing uraninite of Sinyaya Pala».

This agrees well with what has been said in the quotation re the age (1 950 m. y.) of the uraninite from Sinjaja Pala and connected matters. Because of this very considerable correction the age of the minerals of pegmatites from Russian Carelia is geologically »at a natural distance» from the ages of the minerals of pegmatites from Impilahti, and the same may be said with regard also to the age of the pegmatites.

Of the age of the rocks from Sinjaja Pala Vernadsky says (1935, p. 26): »Il s'agit là de roches dont l'âge est très voisin de deux milliards d'années (1,85 à 2,0.10<sup>9</sup> années); la localité n'est pas éloignée de la Finlande — —». Of the age determinations made in Finland he says: »Jusqu'à une date récente, aucune détermination radiophysique n'avait été faite en Finlande. Mais les recherches de A. Holmes sur l'âge des granites d'Impilax, dans la région S.-E., non loin de la frontière russe, ont montré que ces roches sont, à très peu près, aussi anciennes que celles de Sinjaja Pala: les détermination de Holmes ont donné 1,7.10<sup>9</sup> années».

There is reason to mention that Impilahti is certainly not far away (about 50 km westwards) from the former boundary between Finland and Russia, but Sinjaja Pala, situated in Russian North Carelia, is about 500—600 km. to the north of Impilahti.

So far as I know Holmes has nowhere presented such information as that the granites of Impilahti have an age of 1700 m. y. Since Vernadsky does not mention whence this information comes, it cannot either with certainly be said by what this deplorable error is caused. However, the following cases may among others be regarded as possible.

When mentioning the lead ratios of wiikites and nuolaites from Impilahti, based on my analyses (Lokka 1928, p. 63, Table 18), Holmes states (1931, p. 434) i. a. the lead ratio of nuolaite 0.170. Has Vernadsky perhaps denoted — of course by an oversight — the above mentioned age (1700 m. y.), instead of the age corresponding to this value (0.170) this is not quite impossible.

The error may also have its rise in the following. In the »Bulletin de la Commission géologique de Finlande», No. 102, it says on page 14: »Mr. Arthur Holmes gives a lecture On Radioactivity and Geological Age». In the beginning of the short summary of the lecture is said: »According to the determinations quoted, we should get, in years for the Rapakivi granite 1000 millions, for granite from Moss 1250, for granite from Arendal 1600, from Impilahti 1800». With the exception of the age of the rapakivi, these three ages and all those more than 10 ages mentioned in the summary are, unfortunately, faulty, the same error is in all of them and is easily explained in the following way. In all probability the lead ratios have been mentioned in the lecture: Moss 0.125, Arendal 0.160, Impilahti 0.180 a. s. o., and they have by an oversight

been denoted as ages in the publication: Moss 1 250, Arendal 1600, Impilahti 1800 m. y., a. s. o. These ages are in fact considerably lower; thus the age, based on the lead ratio 0.180, Impilahti, is about 1240 m. y. If an uranium mineral is in question, the age is, because of the age corrected for actino-uranium, about 100 m. y. less. Perhaps the age of the granites of Impilahti, 1700 m. y., mentioned by Vernadsky, has its rise in some way or other from the above said erroneous value 1800 m. y. At the end of the summary of the lecture (p. 15) there is a comment: »(N. B. These figures from the record have not been checked by the author)».

## WIIKITE FROM SALLA, E. LAPPLAND

### A NEW OCCURRENCE OF WIIKITE

In the beginning of August 1938 Mr. Arvi Törmänen, of the church-village of Salla Parish sent to the Geological Survey specimens of minerals and rocks from the former Finnish eastern Lapland, asking for information about the specimens. Some of them were in colour and otherwise in appearance more unusual, resembling such minerals as gadolinite, allanite and wiikite, which last mentioned occurs very multi-coloured at Impilahti. These specimens I established by qualitative examinations to be uranium-bearing tantaloniobates.

In the beginning of October the same year I went to Salla and by the kind guidance of Mr. Törmänen had the opportunity to visit the occurrence of those rare minerals, at Silmäpuolivaara. It is situated in the eastern part of the former Salla Parish about 50 km. to the east of the then existing church, about 45 km. northwards from the Polar Circle and about 7 km. to the west of the earlier boundary between Finland and Russia (1940).

According to Mr. Törmänen's information muscovite, about 200 kg., had been quarried at Silmäpuolivaara for the first time already in 1932. In May 1938, when about 6 000 kg. was quarried, the occurrence of the unknown minerals was first observed. Mr. Törmänen paid special attention to them and sent specimens of same to the Geological Survey in August, as mentioned above.

It was established that these minerals are wiikites, rather similar to those from Impilahti, which further on becomes evident. Wiikite was found later in several different places at Silmäpuolivaara, when muscovite was being quarried.

This discovery of an »unknown mineral» by Arvi Törmänen in 1938 is the second discovery of uranium minerals made within the old boundaries of Finland (before 1940). The first was made by Gr. Lisitzin in 1889 at Lokansaari, Impilahti, as has been mentioned before, when dealing with the wiikites of Impilahti.

ROCKFORMING MINERALS PRESENT IN THE PEGMATITE OF SILMÄ-  
PUOLIVAARA

The rockforming minerals of Silmäpuolivaara are quartz, potash feldspar and muscovite, in some places also soda feldspar, hornblende and tourmaline occur.

## POTASH FELDAPAR

The white powder of this white mineral has an extremely feeble reddish shade. With regard to its chemical composition the mineral is an unusually pure alkali-aluminium-silicate, as Table 26 shows. The iron-content is unusually low. MgO and CaO are insignificant in amount. It makes

Table 26. Potash feldspar from Silmäpuolivaara, Salla.

	%	Mol. prop.	Anal. Lauri Lokka
SiO <sub>2</sub>	65.26	10866	5.92
TiO <sub>2</sub>	none		
Al <sub>2</sub> O <sub>3</sub>	18.73	1837	} 1.00
Fe <sub>2</sub> O <sub>3</sub>	0.12	8	
FeO	none		
MnO	tr.		
MgO	0.01	2	
CaO	0.02	4	
Na <sub>2</sub> O	3.38	545	} 1.00
K <sub>2</sub> O	12.15	1290	
H <sub>2</sub> O+	0.09		
H <sub>2</sub> O—	0.06		
	<u>99.82</u>		

almost no difference, whether in this case calcium silicate is taken into consideration or not in calculating the usual ratios. We obtain

$$(\text{Na, K})_2 \text{O} : (\text{Al, Fe})_2 \text{O}_3 : \text{SiO}_2 = 1 : 1.00 : 5.92$$

which corresponds well with the ratio 1 : 1 : 6.

## MUSCOVITE

This mineral is found both in a large foliaceous and a very small flaky form. The former occurs as broader veins, the latter as 2—3 mm. broad parallel veins, at a distance of about 5 mm. from each other in potash feldspar. Beside it is found as fine flaky aggregate.

## Large foliaceous muscovite.

Between the lamellas of the colourless mineral vary tiny magnetite and hematite spots may be found in dense group. The analysis in Table 27 is made from spotless, pur and fresh mineral. The powder is white. The

Table 27. Large foliaceous Muscovite from Silmäpuolivaara, Salla

			Anal. Lauri Lokka		
	%	Mol. prop.		At. prop.	
SiO <sub>2</sub>	44.89	7474	Si	7474	} 9892 = Z = 4 × 2473
TiO <sub>2</sub>	0.35	44		2418	
Al <sub>2</sub> O <sub>3</sub>	32.85	3223	Al	6446 <	} 4946 = Y = 2 × 2473
Fe <sub>2</sub> O <sub>3</sub>	3.76	236		4028	
FeO	2.27	316	Ti	44	} 2564 = W
MnO	0.10	14	Fe <sup>···</sup>	472	
MgO	0.29	72	Fe <sup>··</sup>	316	
CaO	0.01	2	Mg	72	
Na <sub>2</sub> O	1.65	266	Mn	14	
K <sub>2</sub> O	9.56	1015	Ca	2	
F	0.96		Na	532	
H <sub>2</sub> O+	3.32		K	2030	
H <sub>2</sub> O-	0.56				
	<u>100.57</u>				
-O = F <sub>2</sub>	0.40				
	<u>100.17</u>				

WY<sub>2</sub>Z<sub>4</sub> (O, OH, F)<sub>12</sub>

Table 28. Fine flaky Muscovite from Silmäpuolivaara, Salla

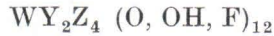
			Anal. Lauri Lokka		
	%	Mol. prop.		At. prop.	
SiO <sub>2</sub>	45.14	7516	Si	7516	} 9671 = Z = 4 × 2418
TiO <sub>2</sub>	2.08	260		2155	
Al <sub>2</sub> O <sub>3</sub>	25.88	2539	Al	5078 <	} 4835 = Y = 2 × 2418
Fe <sub>2</sub> O <sub>3</sub>	4.96	311		2923	
FeO	4.89	681	Ti	260	} 2618 = W
MnO	0.17	24	Fe <sup>···</sup>	622	
MgO	1.31	325	Fe <sup>··</sup>	681	
CaO	0.01	2	Mg	325	
Na <sub>2</sub> O	2.30	371	Mn	24	
K <sub>2</sub> O	8.83	937	Ca	2	
F	0.95		Na	742	
H <sub>2</sub> O+	3.08		K	1874	
H <sub>2</sub> O-	0.59				
	<u>100.19</u>				
-O = F <sub>2</sub>	0.40				
	<u>99.77</u>				

WY<sub>2</sub>Z<sub>4</sub> (O, OH, F)<sub>12</sub>

analysis shows that the chemical composition of this muscovite is fairly common, there is nothing special to be said of it. The content of iron and MgO of the muscovite may certainly be somewhat lower than here, but even a little higher values are not exceptional.

Only 0.56 % water is given off at 110° and at 225° only 0.79 %, the difference 0.23 % corresponding to not more than 6.93 % of the total amount of water (3.32 %), which is given off at over 110°. This 6.93 % is in such case an unusually low value, and as such a very reliable indication that the mineral does not contain secondary water, from which we conclude it is fresh.

Table 27 shows that the corresponding values in the groups Z, Y and W agree very well. The chemical composition may thus be given with the formula



#### Fine flaky muscovite.

The mineral occurs as aggregates and then looks fairly dark, almost black. Very thin flakes, smaller than one millimetre across, have a greenish shade. The mineral seems to be quite fresh. Powder is light grey. As to the chemical composition this muscovite differs in some respects rather noticeably from the large foliaceous muscovite, as Table 28 shows. As to amount  $SiO_2$  is as usual in the case of muscovites.  $TiO_2$  is fairly remarkable. Also the iron-content is considerable, almost double compared with that of the large foliaceous muscovite.  $Al_2O_3$  is remarkably lower in amount. The alkali-content is the same that of the large foliaceous form.

This muscovite gives up 0.59 % water at  $110^\circ$  and 0.80 % at  $225^\circ$ , thus the difference 0.21 % is only 6.82 % of the total amount of water (3.08 %), which is given off at over  $110^\circ$ . It may be said that quite the same result is obtained as in the corresponding case above, i. e. the mineral does not contain secondary water, whence we conclude that it is fresh.

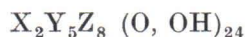
The calculation of the analysis shows that in this case the deficient part of silicium is substituted by an almost equally large part ( $\frac{3}{7}$ ) of aluminium, as also in the earlier case ( $\frac{3}{8}$ ). The alkali group W is 8 % greater than the corresponding values of the other groups, but the difference is not, however, exceptionally great, the chemical composition may thus be presented with the same formula as above.

#### HORNBLENDE

Occurs as aggregates. Dark green crystals are about 0.5 cm. long. Powder is grey. Table 29 shows that this hornblende belongs to those rich in aluminium and iron, in which  $Al_2O_3$  and  $Fe_2O_3 + FeO$  (chiefly  $FeO$ ) both are 10—15 %. Content of  $SiO_2$  is here somewhat higher than usual in hornblendes.

It may be gathered from the calculations of the analyses of these minerals that the greatest part of aluminium commonly substitutes silicium, but in this case only 44 %, which of course is caused by the higher  $SiO_2$ -content.

The chemical composition may well be given by the formula of monoclinic amphibole



## TOURMALINE

The colour of the mineral is black and it occurs both as aggregates and as group formations characteristic of tourmaline, when the thickness of the needlelike crystals is at most some millimetres. Powder very dark.

The analysis in Table 30 shows that  $\text{SiO}_2$  is a few per cent greater than it generally is in the case of tourmalines, and  $\text{Al}_2\text{O}_3$  is about as much lower than usual. The iron-content corresponds to that of the darker tourmalines. The content of alkalis is considerable, but not exceptionally high.

It is fairly common that in the calculations of the analyses of tourmaline the corresponding values of the groups do not all quite agree almost always some group or a couple of them is somewhat too high or too low compared with others. In this case group Y is too small because of a

Table 29. Hornblende from Silmäpuolivaara, Salla

Anal. Lauri Lokka

	%	Mol. prop.		At. prop.	
$\text{SiO}_2$	49.12	8178	Si	8178	} 9339 = Z = 8 \times 1167
$\text{TiO}_2$	0.80	100		1161	
$\text{Al}_2\text{O}_3$	13.44	1318	Al	2636 <	} 1475
$\text{Fe}_2\text{O}_3$	2.39	150		100	
$\text{FeO}$	9.56	1331	Ti	100	} 5837 = Y = 5 \times 1167
$\text{MnO}$	0.33	47	$\text{Fe}^{\dots}$	300	
$\text{MgO}$	10.42	2584	$\text{Fe}^{\dots}$	1331	} 2584
$\text{CaO}$	11.47	2045	Mg	2584	
$\text{Na}_2\text{O}$	1.08	174	Mn	47	} 2045
$\text{K}_2\text{O}$	0.41	44	Ca	2045	
$\text{H}_2\text{O} +$	0.59		Na	348	} 2481 = X = 2 \times 1242
$\text{H}_2\text{O} -$	0.13		K	88	
	99.74				

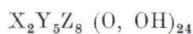
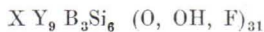


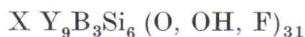
Table 30. Tourmaline from Silmäpuolivaara, Salla

Anal. Lauri Lokka

	%	Mol. prop.		At. prop.	
$\text{SiO}_2$	40.02	6663	Si	6663 = 6 \times 1111	} 2860 = 3 \times 953
$\text{B}_2\text{O}_3$	9.96	1430	B	2860	
$\text{TiO}_2$	0.53	66	Ti	66	} 5616
$\text{Al}_2\text{O}_3$	28.62	2808	Al	5616	
$\text{Fe}_2\text{O}_3$	11.20	701	$\text{Fe}^{\dots}$	1402	} 7807 = Y = 9 \times 867
$\text{FeO}$	3.95	550	$\text{Fe}^{\dots}$	550	
$\text{MnO}$	0.36	51	Mg	122	} 141
$\text{MgO}$	0.49	122	Mn	51	
$\text{CaO}$	0.79	141	Ca	141	} 714
$\text{Na}_2\text{O}$	2.21	3.57	Na	714	
$\text{K}_2\text{O}$	0.80	85	K	170	} 1025 = X
F	0.44				
$\text{H}_2\text{O} +$	1.05				} 1025 = X
$\text{H}_2\text{O}$	0.08				
	100.50				
$-\text{O} = \text{F}_2$	0.19				
	100.31				



relatively low aluminium-content, but not, however, to such extent that the chemical composition of the mineral may not be given by the formula of tourmaline



#### WIIKITE

Among the specimens of the unusual »unknown mineral» from Silmäpuolivaara four kinds could be distinguished, which as to their physical properties differ somewhat from one another. The four specimens, each representing a different kind, which were chosen for examination are in the following denoted by the letters A, B, C and D. Of the three first complete analyses have been made (Table 32), but not of the fourth, as by its physical properties and also judging from some chemical determinations it is very similar to C. A complete analysis would very likely not bring forward any further information as to the chemical composition of these minerals.

A. Black, massive. Fracture conchoidal with metallic lustre; black in thin splinters. Under the microscope thin section dark grey and translucent, here and there very dark and opaque. Powder grey. Specific gravity 3.695.

B. Reddish brown, massive. Fracture uneven, somewhat rough, lustre resinous. Thin splinters paler than larger fragments. Under the microscope yellowish brown, translucent, isotropic. Powder very light brown. Specific gravity 3.617.

In one sample A and B are in connection with each other in such a way that B appears like a brown »groundmass», in which the black mineral A forms spots of indefinite shape about 1 cm. in diameter. The boundary between the parts of different colour is in some places rather clear, in some very indistinct. It seems almost as if the black mineral A, which in some way or other has entered the brown mineral mass B, had from its surface melted together with B. Mention has before been made of matters characteristic of the radioactive minerals when discussing the allanite and the wiikite from Impilahti.

C. Very dark brown, massive. Fracture rough, vitreous. Small splinters yellow. Under the microscope evenly yellowish, translucent, isotropic. Powder light yellow having a shade of grey. Specific gravity 3.588.

D. Very similar to the former, colour somewhat darker. Powder pale yellowish, as in the former, but showing somewhat more of a grey shade. Specific gravity 3.582, very much the same as in the former case.

In Table 31 the dehydration at different temperatures is given. The results are interesting, they agree almost like parallel determinations of the same sample, even though these minerals do not seem similar. They

gave off considerable water already at lower temperatures, at 110° (H<sub>2</sub>O-) over 30 % of the total water, at 255° about 70 % of the total water and at 110°—255° over 50 % of H<sub>2</sub>O+. These high values are very characteristic of such minerals.

Generally the minerals are then already in a state of decomposition, if the same statement as above can be made of their water-content. The case is different when such radioactive metamict minerals as wiikites are in question. As already stated they can absorb fairly great amounts of water and then their chemical composition may change very much, but the minerals are not decomposed or weathered.

Table 31. Wiikites from Silmäpuolivaara, Salla  
Loss of Water

	40°	65°	95°	110° H <sub>2</sub> O—	175°	255°	Tota H <sub>2</sub> O	<sup>a</sup> 110°—255°	<sup>b</sup> over 110° H <sub>2</sub> O+	<sup>a</sup> in % of b
A	0.24	1.07	3.55	5.24	9.53	10.47	14.45	5.23	9.21	56.79
B	0.27	1.16	4.06	5.64	9.88	11.12	15.28	5.48	9.64	56.85
C	0.34	1.23	4.38	6.32	10.34	11.35	15.85	5.03	9.53	52.78
D	0.30	1.35	4.08	5.80	10.03	11.17	15.54	5.37	9.74	55.13

As has been said above, complete analyses have been made of the samples A, B and C. These analyses are given in Table 32. They are carried out according to the same method as those in Table 21 (VII and

Table 32. Wiikites from Silmäpuolivaara, Salla

Anal. Lauri Lokka

	A	B	C
Nb <sub>2</sub> O <sub>5</sub>	30.99	29.85	32.43
Ta <sub>2</sub> O <sub>5</sub>	12.07	14.82	10.50
TiO <sub>2</sub>	3.00	2.39	3.79
SiO <sub>2</sub>	3.30	5.12	3.38
SnO <sub>2</sub>	0.02	0.03	tr.
ZrO <sub>2</sub>	0.07	0.03	tr.
ThO <sub>2</sub>	0.25	0.52	0.11
U O <sub>2</sub>	3.08	0.60	0.75
U O <sub>3</sub>	6.32	7.05	10.12
Y <sub>2</sub> O <sub>3</sub>	13.35	12.64	13.87
Ce <sub>2</sub> O <sub>3</sub>	2.36	2.53	1.72
Al <sub>2</sub> O <sub>3</sub>	0.33	0.21	0.26
Fe <sub>2</sub> O <sub>3</sub>	5.42	4.31	1.76
FeO	0.29	0.22	0.18
MnO	0.21	0.21	0.28
MgO	0.05	0.03	0.19
CaO	2.16	1.99	1.79
PbO	2.23	1.86	2.75
C O <sub>2</sub>	none	none	none
H <sub>2</sub> O+	9.21	9.64	9.53
H <sub>2</sub> O—	5.24	5.64	6.32
	99.95	99.69	99.73
Sp. gr.	3.695	3.617	3.588



VIII), as already stated. All determinations are averages of two parallel determinations. With a view to the age determinations of the minerals, the determinations of lead and uranium are performed with special care, because, even though altered minerals are here in question, these determinations may, nevertheless, be of some importance.

A, B, C and D are as to their physical properties similar to the wiikites of Impilahti. A, B and C are also with regard to their chemical composition quite comparable to some wiikites of Impilahti (Table 21), re which the following may be mentioned here.

The sums  $Nb_2O_5 + Ta_2O_5$ , ratios  $Nb_2O_5 : Ta_2O_5$ , groups  $Nb_2O_5 + Ta_2O_5 + TiO_2 + SiO_2$ , groups of rare earths, and contents of uranium are in these three analyses such as regards their amounts that these analyses have parallels among those of the wiikites from Impilahti. We have good reason to specially remark that with regard to rare earths yttrium earths are predominant compared with cerium earths — yttrium earths 89, 85 and 83 % of rare earths — as in the analyses of the wiikites from Impilahti (Table 22), of which minerals this property is a characteristic. These three minerals are comparable to the wiikites of Impilahti also with regard to other constituents, except the water-content, which is remarkably higher.

These minerals A, B and C and also D — the lastnamed is as to its physical properties very similar to C and evidently also with regard to its chemical properties — may fairly well be placed in groups II and III (Table 23) of the wiikites of Impilahti on the basis of physical properties and the said chemical constituents, with the exception of water-content and specific gravity. The mineral A would belong to group II and the minerals B, C and D to group III. But on the basis of low specific gravity and in connection with it an especially high water-content, which in these minerals is remarkably higher than in any wiikite from Impilahti, all four would nearest belong to group IV.

Since these four minerals in other respects, except as to water-content and specific gravity, are similar to the wiikites of Impilahti, there is no reason to give them a new name, and they may also be called *wiikites*. As there is a slight difference, however, these wiikites from Salla, as distinguished from those from Impilahti, may be named *wiikite of Salla*.

Also the wiikites of Salla, being metamict minerals, have probably changed their chemical composition very considerably and all almost to the same extent. This may be concluded also from the equal values and heights of water-content and the determinations of dehydration which give fairly equal results.

The calculation of the age of these wiikites on the basis of lead ratio does not, of course, result in the right age of the mineral, but only in an apparent age. The ages of these wiikites, A, B and C, calculated,

according to the above mentioned logarithmic formula, are given in Table 33, in the last column of which the ages are corrected for actino-uranium (Holmes 1937, p. 153).

Doubtless these values are too high. This is due to the differential leaching of uranium. Introduction of lead is possible too, though ore

Table 33. Wiikites from Silmäpuolivaara, Salla

	U %	Th %	Pb %	Pb	Age in m. y.	
				U+0.36 Th	Calc. fr. log. form.	Correct. for AcU
A	7.98	0.22	2.07	0.257	1709	1505
B	6.40	0.46	1.17	0.264	1748	1533
C	9.08	0.10	2.55	0.280	1842	1600

was not detected in microscopic examination. Concluding from the lead ratios which are almost the same these changes have come to pass in all three wiikites practically in the same proportion.

Bearing in mind what has been said above, it may be considered very probable that the real age of the wiikites of Salla is lower than about 1500 m. y. But how much lower it is, cannot of course be said on the basis of determinations and calculations. It is probable, however, that it may be reduced at least 300 or perhaps 500 m. y., when considering that the leaching of uranium very likely has been rather considerable. Thus an age would be attained, which approximately would correspond to that of the minerals from Impilahti (Table 25).

### MONAZITE FROM IVALOJOKI RIVER, N. LAPLAND

The mineral has been separated from the sand of Ivalojoki river, which runs from the southwest into the lake Inarijärvi, situated in northern Lapland (Finland). The specific gravity 5.019 is somewhat lower than that of the monazites of Impilahti (Lokansaari 5.163 and Mursula 5.177, Table 11). Powder is light grey.

Since the real aim of the treatment of this monazite, likewise of the monazite of Mursula, was to determine the age of the mineral. I determined besides thorium and lead only the sum of rare earths, which all to some extent characterize the chemical composition of the mineral.

The determination of the sum of rare earths resulted in 54.32 %, an unusually low value for such monazites the ThO<sub>2</sub>-content of which is about 8—9 %, as it is here. Somewhat more than general falls to the share of other constituents, but how it has been divided between them, I did not consider necessary to determine in this case.

Four thorium and four lead determinations have been performed. For the former determinations 1 g. powder was used in each case for the latter 3 g. each. The determinations have been carried out chiefly according to Fenner (1928), as in the corresponding earlier cases. The results are:

9.08, 9.00, 9.09 and 8.90 %, average 9.02 %  $\text{ThO}_2 = 7.93$  % Th.

0.542, 0.520, 0.518 and 0.531 %, average 0.528 % PbO = 0.491 % Pb.

Lead ratio  $\text{Pb}/0.36 \text{ Th} = 0.172$ .

The age according to logarithmic formula is 1192 m. y.

On the basis of lead ratio 0.172 the age of this mineral is at the lower limit of the Middle Pre-Cambrian, above or below it, depending on how this limit is defined. Of this lower limit Holmes says (1931, p. 441): »The lower limit of the Middle Pre-Cambrian is less well defined geologically, but pending further discoveries, it may be provisionally defined in terms of lead ratios at between 0.17 and 0.18». The age of the monazite of Ivalojoiki would thus be at the lower limit of the Middle Pre-Cambrian.

In this connection mention may also be made of the geological age of that area, Norwegosamian, of which the Ivalojoiki and Inarijärvi area, i. e. Finnish Northern Lapland, is a part, and to which also South Waranger (Norway) and Kola Peninsula belong.

Of the geological age of this area Holmes says (1937, p. 189): »This cycle is earlier than the Karelian, but is, so far, only suspected to be earlier than the Svecofennian».

Backlund mentions the same matter (op. c., p. 64) pointing out first that some smaller scattered areas are remarkably younger than the chief part of it. Aiming at this chief part he says: »Eine absolute Altersbestimmung liegt für dieses Gebiet nicht vor — — —. Für den älteren Gebirgsgrund, der sicherlich Teile eines sehr alten Orogens representiert, sind zwei Altersstellungen möglich; Die Norwegosamiden können im Alter mit den Svecofenniden gleichgestellt werden — — — von welchen sie ein nördliches Bruchstück darstellen können. Oder sie nehmen eine selbständige Altersstellung zwischen den Svecofenniden und Märealbiden ein, entsprechend einen Alter von 1 200—1 300 Millionen Jahren».

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