Minerals of the Varuträsk Pegmatite.

I. The Lithium — Manganese Phosphates.

By

PERCY QUENSEL.

(MS. received ${}^{10}/_{2}$ 1937.)

The Varuträsk pegmatite, lying between Boliden and the Baltic coast, about 20 km SE of the great Boliden mines, was first discovered in 1933, when dr O. BAECKSTRÖM, chief geologist of the Boliden Mining Co., drew my attention to a finegrained lilac lepidolite-fels which had been observed during some temporary blasting for quartz. Since then a good deal of work has been done, the Boliden Company reckoning with an eventual economic output of the lithium minerals. I have thereby had the opportunity to follow the workings during summer visits in 1934, 1935 and 1936. The abundance and beauty of many of the pegmatite minerals speedily drew attention to the locality, and I hope in time to give a full description of the occurrence. The two last years have, however, yielded so rich an assortment of species and raised problems of such singular interest, that the final publication has been retarded, awaiting further determinations on the abundant material and final observations relating to the different stages of mineralisation. I therefore propose here to give a preliminary account of one group of minerals only, namely the triphylite series and its decomposition products, which in Varuträsk are represented in very fine development. Especially the transition from fresh blue triphylite over an intermediate stage to the brilliantly purple heterosite is to be followed in exceptional detail, even well represented in single hand specimens.[1]

Before entering on this subject I will, however, in form of a brief list, give an indication of the characteristic minerals of the locality, which up to date have been found and identified, affording an intimation of the general character of the pegmatitic mineralisation.

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[1] Further papers dealing with the minerals of the pegmatite, are planned to follow in this journal.

Native elements:	allemontite	locally frequent
Oxides:	cassiterite	abundant
Phospates:	blue Mn-apatite	frequent
	amblygonite (montebrasite)	2 modifications, one abundant, one very scarce
	triphylite	rather scarce
	ferri-sicklerite	frequent
	heterosite	frequent
	varulite (n. sp.)	scarce
	alluoudite	rather scarce
Niobate-tantalates:	columbite	frequent
	Mn-tantalite	scarce
	a mineral near stibio-tantalite	scarce
Silicates:	cleavelandite	abundant
	lepidolite	abundant
	petalite	abundant
	spodumen	locally abundant
	beryll	frequent
	coloured turmalines	abundant (black,green, red, blue)
	zeolites	frequent

The pegmatite belongs to the so-called Skellefte granite, a fine-grained outlyer of the great Revsund granite area of northern Sweden, which is now considered to belong to the closing epoch of the ancient svionian formation of the Swedish archaean.

The minerals treated in this paper are triphylite, ferri-sicklerite, heterosite, varulite and alluoudite.

The normal triphylite series.

The primary mineral of this series is in fresh state a deep sky-blue triphylite (fig. 1 A). In general one must however say that perfectly fresh material is scarce. At commencing alteration the triphylite looses its blue colour and becomes wine yellow or clove-brown (fig. 1 B), types on the whole more common than the blue variety which as yet has only been found quite locally, but there, at least in one place, relatively abundant.

The fresh blue triphylite occurs in coarsegrained cleavable masses. The luster is resinous, fracture uneven to subconchoidal. The usual cleavages parallel (001) and (010) are well developed. The mineral

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is in thin section colourless. The axial plane is parallel (001) with γ normal to (010). The character is optically +, dispersion very strong, $\varrho > \gamma$, occasioning anomalous indigo-blue interference colours. Determination of the axial angle gave: $2V_{Na} = 26°$.

MARGIT ROSENBERG.

Fig. 1. Specimen of blue triphylite (A) with alteration ring of ferri-sicklerite (C). In ring B, the blue triphylite has turned yellow, due to beginning alteration. Anal. of A and B page 81, of C p. 84.

So far the optical properties seemed to coincide with previous determinations. An instance indicating that the triphylite of Varuträsk possessed divergent qualities, was the observation of twinning in one thin section of blue triphylite (fig. 1 A). As far as I have been able to find, twins have not before been observed in triphylite, though recorded in the nearly related beryllonite. The twinning is repeated in a few broad polysynthetic lamellae. Dr VON ECKERMANN has been kind enough to corroborate the twinning law by measurements on the universal stage. The result is in several respects so unforseen and the conclusions which may be drawn, imply so farreaching consequences, that I cite VON ECKERMANNS observations in extenso:

> »At the request of Prof. QUENSEL, I have undertaken the investigation of an apparently twinned crystal, observed in a thin-section of triphylite-rock from Varuträsk. In section the mineral is colourless, the refringence and birefrigence agreeing with those of the triphylite. As no mentioning of twinned triphylite is found in the literature, the question arose whether another mineral or a haphazard growth of separate triphylite crystals were present.
>
> The investigation was carried out on the universal stage in sodium light. The crystal-section consisted of two somewhat narrower marginal and one broad central lamellae, the joints being straight and strictly parallel; not coinciding with any partings of the mineral. The optical orientation of the marginal lamellae was found to be identical.
>
> The two systems of lamellae constitute undoubtedly a true polysynthetic twinning, in the present case cut almost at right angles to the twinning-plane and slightly oblique to the acute bissectrices of both twins. The optic plane makes an angle of 4° with the trace of the twinning-plane, and the latter is not quite perpendicular to the $\alpha\beta$-plane; 87.5° being the average of several determinations. The axial angles of both twins were measured at $2V_{Na} = 26° \pm 0.5°$; the refringence of triphylite, as determined by QUENSEL, being used when calculating the corrections of the readings.
>
> Evidently, the mineral is not orthorhombic. In order to compare it with the un-twinned, presupposedly orthorhombic, triphylite of the same sample, another section was prepared where the mineral was cut almost parallel to the $\alpha\beta$-plane, exhibiting nicely developed cleavages parallel to 001. The extinction-angle $\alpha : (001)$ was found to be 4° and the axial angle was determined at $2V_{Na} = 26° \pm 0.5°$.
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> These values correspond exactly with those of the twinned crystal, assuming the twinning-plane to be (001). All other optic characteristics being equal, there seems to be small doubt of the two minerals being identical.
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> The results of my investigation, consequently, seriously question the orthorhombic character of the triphylite of Varuträsk indicating, as they do, a lower pseudo-orthorhombic symmetry. They also call for a revisal of earlier measurements of triphylite crystals.»

I must for the present restrict myself to draw attention to these facts. To definitely proof the symmetry of triphylite, especially as

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P_2O_5 . . . .

Sp. gr. . . . . .

1. Fresh blue
2. Wine-yellow
3. Triphylite,
4. Triphylite.
5. Triphylite.

the divergence from orthorhombic symmetry in any case is insignificant, it will be necessary to corroborate the new observations with Laue photographs. I hope opportunity may be offered to return to this question in a future publication. Till then we must, however, reckon with the probability that there exists a clino-triphylite.

Principally in order to determine the proportions between Fe and Mn and thereby localize the triphylite in the isomorphous series triphylite — lithiophilite a partial analysis was already at an early stage made by dr R. BLIX of the State Natural History Museum, giving 19.53 % MnO and 25.79 % FeO, all Fe reckoned as FeO. The mineral species in question can therefore be classed as triphylite. For even if one can presume a continuous series between $LiFePO_4$ and $LiMnPO_4$, existing analyses place known species rather decidedly in two groups, the triphylite group with $MnO < 20$ % and 25—40 % FeO and the lithiophilite group with $FeO < 20$ % and 25—40 % MnO. The Varuträsk species may therefore be said to represent a triphylite relatively rich in Mn.

As triphylite is a new mineral for Sweden, it was considered desirable to obtain more complete analytical data. Through the courtesy of the Boliden Mining Co. three analyses have been made by miss THELMA BERGGREN of the Companys Research Laboratories in Stockholm. The two first refer to the two modifications represented by ring A and B in fig. 1. The third represents a somewhat oxidized yellowish-brown triphylite, found at an earlier stage of the investigation.

	1.	2.	3.	4.	5.
Insoluble	1.24	5.80	0.20	—	—
$H_2O - 105°$	0.29	0.13	0.05	—	—
$H_2O + 105°$	0.31	0.36	0.64	1.47	0.42
Fe_2O_3	0.00	0.33	6.30	—	—
Al_2O_3	0.00	0.00	0.00	—	—
FeO	24.33	22.85	18.21	26.23	26.40
MnO	18.95	18.15	20.16	18.21	17.84
CaO	1.82	1.84	1.36	0.94	0.24
MgO	trace	0.11	0.00	0.59	0.47
Li_2O	8.52	7.92	6.06	9.36	9.36
Na_2O	0.16	0.58	2.28	0.12	0.35
K_2O	0.00	0.00	0.00	0.32	—
P_2O_5	44.10	42.23	44.47	44.03	44.76
	99.72	100.30	99.73	100.70	99.84
Sp. gr.	3.423	3.384	3.521	3.52	3.534

1. Fresh blue triphylite, Varuträsk (A in fig. 1).
2. Wine-yellow triphylite » (B in fig. 1).
3. Triphylite, somewhat oxidized »
4. Triphylite, Grafton, New Hampshire, U.S.A.
5. Triphylite, Norwich, Mass. ›

Analyses 1 and 2 come very near the old analyses of **Penfield** of triphylite from Grafton and Norwich, which are given above for comparison (N:o 4 and 5).

Analysis 1 indicates a very pure triphylite. Recalculated to 100 % the molecular ratios are 946: 317 or nearly exactly 3: 1. Analysis 2 represents an intermediate zone between the blue triphylite and an alteration product, ferri-sicklerite, described below. The triphylite of this zone has changed colour to wine-yellow, but shows hardly any other change than an insignificant oxidation of 0.33 % Fe_2O_3. Analysis 3 shows on the other hand more advanced oxidation, a fourth part of the iron being present as Fe_2O_3. This analysis is also of interest because of the relatively high content of Na_2O. Of former analyses of triphylite none exceeds 0.50 % Na_2O. It lies near at hand to reckon with an intermixture of a third component in the triphylite — lithiophilite series, namely the natrophilite molecule. Already **Brush** and **Dana** have shown that natrophilite is intimately associated with lithiophilite in Brancheville. On the other hand one might reckon with the possibility of a substitution of Na_2O for Li_2O during the oxidation, analogous to the formation of β-spodumen, described by **Brush** and **Dana** from Brancheville. I will here restrict myself to drawing attention to the fact, as opportunity will be given further on to return to the question, where a third explanation is discussed (p. 95).

We may now proceed to the more advanced stages of alteration of the triphylite from Varuträsk. A mineral belonging to this category at an early date drew our attention by the brilliant purple colour of its streak. It was easy to define this mineral as belonging to the heterosite — purpurite series. When I, therefore, in 1936 through the mining geologist of the Boliden Co. dr O. **Ödman** received a fine specimen of blue triphylite, surrounded by a darkbrown ring, (A and C in fig. 1), I naturally presumed that I had in my hand the transition from triphylite to heterosite, which mineral is always black to darkbrown in field specimens due to a thin coating of a hydrogenous phosphate of Fe and Mn. It soon, however, became evident that the alteration products of triphylite were of a more complex nature and that at least one definite intermediate stage of oxidation was represented at Varuträsk.

Already in the summer of 1935 dr O. **Baeckström** had in an outlying pegmatite dike near the small farm of Lövlunden, about three km NE of Varuträsk, found specimens of a mineral, which could not be identified with any other mineral of the district. A preliminary chemical investigation showed it to be a Fe-Mn phosphate with all Mn as MnO and the whole Fe content as Fe_2O_3. Later on, as my attention was drawn to the case, it was found that this intermediate product of oxidation was by no means uncommon. The dark ring round the fresh triphylite (C in fig. 1) was found to possess the same chemical qualities and where triphylite is still found unaltered, this species seems to represent a common and more initial stage of alteration, alone or intermixed with the more fully oxidized heterosite. The stages of oxidation from triphylite over the new intermediate product to heterosite are chemically characterized by practically the whole content of Mn and Fe occurring as follows:

triphylite	MnO	FeO
interm. stage	MnO	Fe_2O_3
heterosite	Mn_2O_3	Fe_2O_3

The new mineral, representing the intermediate stage, is dark brown in colour, the streak and powder brown to brownish red. The brilliant purple streak of the heterosite-purpurite series is quite lacking. Also if dipped in acid, to remove the usual thin black coating of later alteration, a brown colour appears, not the purple of the higher oxidized species, which naturally is dependant on the higher state of oxidation of Mn, there at hand.

Under the microscope the mineral is of a deep reddish brown colour and almost unpleochroitic. Subtranslucent to opaque, the deep colouring makes optical determinations difficult and unreliable. The cleavage of the original triphylite is readily distinguishable. The medium indices of refraction is well over 1.78.

Two complete analyses have been made by miss BERGGREN. N:o 1 is the outer ring surrounding the fresh triphylite, (fig. 1 C), no. 2 is from the farm Lövlunden where the mineral was first found. No. 3 is sickerite, a nearly related phosphate from Pala, Cal., first described by SCHALLER. As seen, the two analyses from Varuträsk give very close results, indicating that we have to deal with a definite mineral and not only with a link in a progressing series of alteration products.

Both analyses of the Varuträsk material correspond fairly well to the formula:

$$12\ RO\ 5\ Fe_2O_3\ 9\ P_2O_5$$

The small amount of H_2O has, however, then been left out, as the bases only suffice to bind P_2O_5. The ratios bases to P_2O_5, all Fe counted as FeO, is nearly exactly 3 : 1 (952 : 315).

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	1.	1 a. mol. prop.	2.	3.
Unsoluble	1.66	—	2.42	4.18
H_2O −105°	0.51	—	0.79	—
H_2O +105°	0.51	—	0.44	1.70
Fe_2O_3	27.20	0.170	29.08	11.20
Mn_2O_3	0.00	—	—	2.10
Al_2O_3	0.00	—	0.00	—
FeO	0.59	0.008	0.22	—
MnO	19.13	0.270	15.20	33.60
CaO	1.36	0.024	1.88	0.20
MgO	0.11	0.003	1.70	—
Li_2O	3.72	0 124	3.26	3.80
Na_2O	0.81	0.013	0.88	—
K_2O	0.00	—	0.00	—
P_2O_5	44.80	0.315	44.64	43.10
	100.40		100.51	99.95
Sp gr.	3.391		3.271	

The amount of Fe_2O_3 is naturally dependant on the accessible amount of Fe in the original triphylite and the proportions RO : R_2O_3 can therefor not be expected to present any strict molecular proportions.

In discussing the analyses, the following points may be worth notice, comparison being made between the fresh triphylite and the outer ring of alteration in the same hand specimen.

It is obvious that Mn, Fe and P_2O_5 have practically remained constant, the changes are: MnO from 18.95 to 19.13, FeO from 24.33 to 25.08, (counting all Fe as FeO) and P_2O_5 from 44.10 to 44.80. The only evident change which has taken place, refers to the almost complete oxidation of FeO and to the leaching of Li_2O which has been reduced from 8.52 % to 3.72 %, or to less than half the original content. The amount of leaching of the alkalies has to all evidence been regulated in proportion to the oxidation of FeO. The molecular ratios between bases and P_2O_5 has remained unchanged, 946 : 317 in triphylite, 952 : 315 in the alteration product.

As far as I have been able to find, no mineral in all respects corresponding to the composition given above, has been described, though several species, referred to as alteration products of triphylite or lithiophilite with Mn as MnO and Fe as Fe_2O_3 are known. I refer to the old species alluoudite of DAMOUR (1848) and to salmonsite and sicklerite of SCHALLER (1912). Of these, however, none strictly coincide with the chemical relations cited above. Rather near comes alluoudite, but the content of Na_2O substituting Li_2O places this mineral in a separate class. A mineral closely resembling alluoudite is represented at Varuträsk and I will return to the question of its position and relations

op.	2.	3.
	2.42	4.18
	0.79	—
	0.44	1.70
	29.08	11.20
	—	2.10
	0.00	—
	0.22	—
	15.20	33.60
	1.88	0.20
	1.70	—
	3.26	3.80
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further on. Salmonsite is strongly hydrated with 15.30 % H_2O, for which SCHALLER gives the formula 9MnO Fe_2O_3 $4P_2O_5$ 14 H_2O. Evidently it is not our mineral.

The sicklerite from Pala, on the other hand, in many respects comes near the Varuträsk alteration product. The formula given by SCHALLER from analysis 3 in the table above is : 6MnO Fe_2O_3 $4P_2O_5$ $3(Li, H)_2O$.

It is difficult to say, how near these two minerals are related. The Pala sicklerite, by SCHALLER considered as an alteration product of lithiophilite, is essentially poorer in iron. The relative proportions between Fe and Mn are, however, in this case of little consequence, these relations being dependant on the allotment in the primary triphylite or lithiophilite. SCHALLERS sicklerite shows, according to the only analysis published, a slightly diverging composition, the proportions RO : P_2O_5 being 9 : 4 instead of 9 : 3 in our mineral. The very brief description of sicklerite does not allow a positive conclusion as to the reliability of the analytical data. As, however, known optical properties agree and the paragenesis of both minerals is to all evidence much the same, I propose to name the mineral from Varuträsk ferri-sicklerite, indicating a relationship to the Pala sicklerite but accentuating the higher content of iron.

The two analyses of ferri-sicklerite, from two different localities, are so alike that one cannot doubt a definite species. The oxidation of triphylite and lithiophilite to all evidence does not proceed continuously, ferri-sicklerite marking a stage where all Fe is oxidized but the oxidation of Mn has not yet set in. This would denote a halting-point in the progressing alteration, the next stage would be represented by the minerals of the heterosite — purpurite series, when also all Mn has been oxidized.

We may therefore define ferri-sicklerite as representing a definite stage of oxidation in a series triphylite — ferri-sicklerite — heterosite. SCHALLER's sicklerite would probably represent the same place in the series lithiophilite — sicklerite — purpurite and might therefore be signified as Mn-sicklerite. The following table, with the percentages of the principle constituents, taken from representative analyses, indicates such a succession:

I. Triphylite series.

triphylite	Li_2O	8.52	MnO	18.95	FeO	24.33	P_2O_5	44.10
ferri-sicklerite	»	3.72	»	19.13	Fe_2O_3	27.20	»	44.80
heterosite	»	0.46	Mn_2O_3	20.42	»	27.44	»	41.93

II. Lithiophilite series.

lithiophilite	Li_2O	8.50	MnO	35.98	FeO	8.60	P_2O_5	44.40
sicklerite	»	3.80	»	33.60	Fe_3O_3	11.26	»	43.10
purpurite	»	—	Mn_2O_3	29.25	»	15.19	»	47.30

It is of interest to note, how nearly equivalent, the reduction of Li_2O is in both the two intermediate stages, from an original content of about 8.5 % in triphylite and lithiophilite to 3.72 in ferri-sicklerite and 3.80 in the Pala sicklerite. Mn and Fe also remain in their original proportions and percentage. The only change is the intermittent oxidation of Fe in the first instance and of Mn in the second and the leaching of the alkalies.

An extra indication that ferri-sicklerite represents a definite mineral species, is given by an X-ray powder photograph which prof. WESTGREN has kindly taken (fig. 2), shortly discussed further on in reference also to heterosite (p. 89).

We may now proceed to the next stage of oxidation, represented by the mineral heterosite.

Concerning the name of this phosphate, different opinions have been asserted. LACROIX[1] has referred SCHALLER's new mineral purpurite[2] to the older heterosite of ALLUOUD (1826), and is of the meaning that purpurite »n'est qu'une heterosite plus manganifère que celle du Limousin». In this case the name heterosite has naturally priority. It is, however, evident that there exists a series heterosite — purpurite with the same difference in proportions between Mn and Fe as in the series triphylite — lithiophilite. It seems therefore appropriate to retain both names, as SCHALLER also proposes in a later publication,[3] representing the same chemical distinctions as are maintained between triphylite and lithiophilite. This is all the more wellgrounded, inasmuch as the material from Varuträsk clearly indicates that no change of any importance concerning the Mn — Fe proportions has taken place during the process of alteration. In agreement with SCHALLER the mineral here in question is named heterosite, indicating the alteration and full oxidation product of triphylite.

The heterosite occurs at Varuträsk in larger or smaller nests and nodules, widely dispersed through different parts of the pegmatite. One of the largest nodules measured over 4 dm in length and 2 dm in bredth. In consequence of the strong tendency to alteration which all species of the triphylite — lithiophilite series show, the original triphylite is seldom preserved in larger quantities in the heterosite nodules. Under the microscope, however, rests of triphylite are often to be observed. The transition is in most cases singulary sharp, again indicating the absence of gradual oxidation. Ferri-sicklerite is of course often noted as an intermediate product, but between the three stages a marked hiatus is pronounced.

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[1] Min. de France, tome 4, p. 469.

[2] Am. Journal of Science, XX, 1905, p. 146.

[3] Bull. U. S. Geol. Survey, 490, 1911, p. 72.

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Attention was early drawn to the heterosite at Varuträsk because of the brilliant purple streak, produced by scratching the black surface. The mineral in field specimens always shows a dull black colour, but this was soon found to depend on a thin coating of another phosphate, which, penetrating every cleavage and crack, completely obscures the true colour of the mineral. The black fragments, when treated for a moment with cold, diluted HCl, show up in radiant purple colouring.[1]

Under the microscope the heterosite shows an intense pleochroism in yellow-gray, red and deep purple. The cleavages of the original triphylite are generally well retained. The mineral is often intimately interwoven with narrow streaks of a yellow phosphate following the cleavages. This is possibly identical with the mineral alluoudite, which will be described further on.

To control the relative proportions of Mn and Fe and the state of oxidation, a partial analysis was made by dr Blix giving the following result:

Mn_2O_3 21.14 Fe_2O_3 28.30

All Mn and Fe were present as trivalent oxides. The proportions of Mn_2O_3 and Fe_2O_3, reduced to MnO and FeO, nearly exactly correspond to the amount and proportions in the analysed triphylite:

	triphylite	heterosite
FeO	24.33	25.46
MnO	18.95	19.00

Already this early analysis indicated that no exchange or removal of these elements had accompanied the alteration.

As published analyses of the heterosite — purpurite group of minerals are scarce and the U. S. A material, treated by Schaller, has been executed on small quantities and partly impure material, whereas the material from Varuträsk was both abundant and exceptionally pure, it was desirable to get a new complete analysis made. Through the ready compliance of the Boliden Co., Miss Berggren has executed two complete analyses on singularly pure material, the one of the Varuträsk, heterosite, the other of so-called purpurite from Erongo in SW Africa on material, which prof. P. Ramdohr of the Berlin University kindly put at my disposal. The analyses are given below; for comparison the only two other analyses of later years, from Hill City, S. Dakota and Kings Mt, N. Carolina, both published by Schaller, are appended. Finally an analysis by Pisani, citied by Lacroix, on material from the original locality of Chanteloube, France, is given, concluding the available analytical data of any importance.

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[1] Cfr. Brush and Dana. Am. Journ. of Science 1879, XVII, p. 367.

	1.	2.	3.	4.	5.
Unsoluble	0.30	—	0.19	0.52	—
$H_2O - 105°$	1.67	1.08	—	1.95	—
$H_2O + 105°$	1.65	2.33	4.82	3.31	7.62
Fe_2O_3	27.44	33.42	38.36	15.89	24.60
Mn_2O_3	20.42	15.14	12.18	29.25	29.35
MnO	3.60	1.53	—	—	—
CaO	1.70	1.16	1.37	1.48	—
MgO	0.17	0.00	—	—	—
Li_2O	0.46	0.83	—	trace	—
Na_2O	1.12	0.72	—	0.84	—
K_2O	0.00	0.00	—	—	—
P_2O_5	41.93	43.79	43.45	47.30	41.60
	100.46	100.00	100.27	100.54	103.17

1. Heterosite, Varuträsk. anal. Thelma Berggren 1936 Sp. gr. 3.409.
2. Heterosite, Erongo, SW Africa. anal. Th. Berggren 1936 Sp. gr. 3.398
3. Heterosite, Hill City, S. Dakota. anal. W. Schaller 1911.
4. Purpurite, Kings Mt. N. Carolina. › › ›
5. Original ›Heterosite›, Chanteloube, France. anal. Pisani 1910.

Both the new analyses show almost identical ratios for $R_2O_3 : P_2O_5$
The heterosite from Varuträsk gives $R_2O_3 : P_2O_5$ 301 : 295
from Erongo 305 : 308
Clearly, the small amount of RO bases are bound in other ways than as phosphate.

To all evidence the oxides in the heterosite — purpurite series replace each other in as varied proportions as in the triphylite — lithiophilite series. From 6 analyses, (the 5 tabled above and one incomplete from Brancheville, Conn.) I have calculated the following allotment:

	Fe_2O_3 P_2O_5	Mn_2O_3 P_2O_5
1. Hill City, S. Dakota	76	24
2. Erongo, SW Africa	68	32
3. Varuträsk	57	43
4. Brancheville, Connecticut	53	47
5. Chanteloube	45	55
6. Kings Mt., N. Carolina	35	65

Following SCHALLERS proposal regarding nomenclature, the 4 first are to be classed as heterosite, the two last as purpurite.

The new analyses from Varuträsk and SW Africa do not confirm SCHALLERS formula respecting the content of H_2O. Miss BERGGREN, however, has drawn my attention to the fact that the mineral is highly hygroscopic after pulverisation so that it is difficult to form conclusive evidence on older analyses, made on insufficient or defective material.

4.	5.
0.52	—
1.95	—
3.31	7.62
15.89	24.60
29.25	29.35
—	—
1.48	—
—	—
trace	—
0.84	—
—	—
47.30	41.60
100.54	103.17

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Both the new analyses show a content of H_2O nearer 1/2 than 1 in proportion to P_2O_5. In fact if the ratios for the combined alkalies are deducted for R(OH), both analyses give almost exactly the ratios 2 : 2 : 1 for $R_2O_3 : P_2O_5 : H_2O$. But then the small content of MnO and CaO has been left out. SCHALLER also emphasises that the water which is given off at 105°, »goes very readily and at one time», and draws the conclusion, that it most probably is present as water of crystallization. In my calculation I have combined H_2O above and below 105°, as also SCHALLER has done. Though I can draw no conclusive evidence respecting the state in which H_2O partakes in the constitution, the new analyses rather point to the formula $R_2O_3 P_2O_5 \, {}^1/_2HO$ than to SCHALLERS R_2O_3 P_2O_5 H_2O.

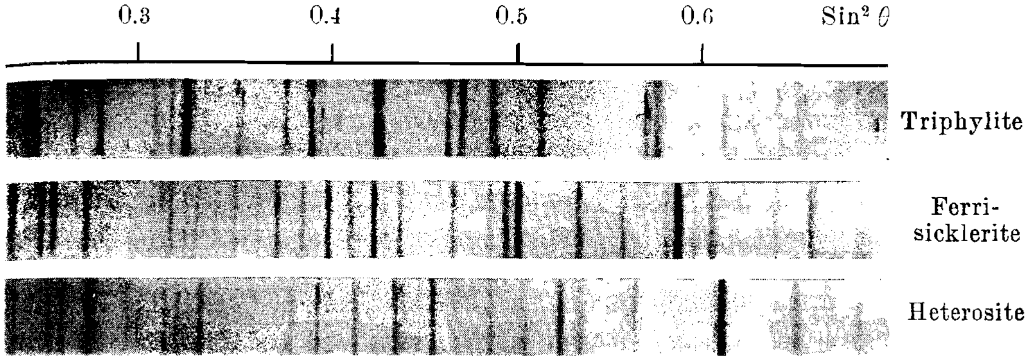

Fig. 2. X-Ray Photographs of Triphylite, Ferri-sicklerite and Heterosite obtained with Cr-*K* Radiation.

It may be of interest to note an old analysis of heterosite from Norwich, Mass. by MALLET,[1] which in respect to content of water stands near the new analyses. It will be recollected that a triphylite very near the triphylite from Varuträsk in chemical composition, also came from Norwich. The conditions at Norwich seem to have been very analogous with Varuträsk respecting formation of minerals of the triphylite series and their decomposition products.

Besides the localities, citied above, minerals of the heterosite-purpurite series have been described from Tammela in Finland[2] and Hühnerkobl, Bavaria,[3] in both cases associated with minerals of the triphylite — lithiophilite series.

Professor WESTGREN has at my request been kind enough to take some X-ray powder photographs of the series triphylite — ferri-sicklerite—heterosite. The photographs, here reproduced (fig. 2), show

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[1] Am. Journ. of Science 1854, XVIII p. 33.
[2] MÄKINEN, Bull. Com. Geol. de Finland, 35, p. 95.
[3] LAUBMANN & STEINMETZ, Zeitschr. f. Kryst. 55, p. 562, 574.

marked dissimilarities between the three minerals. Awaiting opportunity for further structural analyses, I here restrict myself to cite the following lines communicated by WESTGREN: »From X-ray powder photographs of mineral specimens, specified as triphylite, ferri-sicklerite and heterosite, may be concluded that each of them mainly represents a special phase. The strongest interferences of ferri-sicklerite show up as faint lines in the X-ray patterns of heterosite indicating that the latter specimen contains somewhat of the same phase as ferri-sicklerite.»

I will now return to the question of the black coating, which, as already mentioned, always is found on field specimens of heterosite or purpurite, masking the true colour of the minerals. This coating has probably been held for MnO_2 according to descriptions from several localities. LACROIX[1] for example speaks of psilomelan as a secondary product of alteration of triphylite occurring parallel with the transformation to heterosite or hureaulite, and MÄKINEN[2] has presumed the same interpretation for evidently quite the same phenomena regarding the heterosite from Tammela. He says that »sogar die feinsten Sprünge sind von einer schwarzen, halbmetallisch glänzenden Substanz erfüllt, die sehr deutlich auf Mn reagiert und wahrscheinlich aus Pyrolusit besteht.»

A qualitative proof showed, however, that at Varuträsk the black covering was a phosphate, containing Fe_2O_3, Mn_2O_3 and H_2O. I then noted references by BRUSH and DANA[3] and by GRATON and SCHALLER,[4] evidently referring to the same phenomena. Whereas BRUSH and DANA refer to a decomposition product of lithiophilite, GRATON and SCHALLER refer to a direct alteration product of purpurite from N. Carolina. They say »the purple mineral is always covered or surrounded by a greater or less thickness of a black or brownish-black material of pitchy luster and uneven or sub-conchoidal fracture. This material was found to contain iron, manganese, phosphoric acid and water.» The authors expressed hope that sufficient material would soon be forthcoming for an analysis, but, as far as I can find, nothing has been published since 1905, except in U. S. Geol. Survey, Bull. 490 p. 73, where SCHALLER mentions a coating of black, secondary material on lithiophilite from N. Carolina, also occurring as a narrow zone between lithiophilite and purpurite.

Miss BERGGREN has been kind enough to make some quantitative tests of the black coating on the heterosite of Varuträsk. The coating

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[1] Min. de France IV p. 363, 470.

[2] Bull. Com. Géol. de Finland, 35, p. 95.

[3] Am. Journ. of Science, 1879, XVII, p. 367.

[4] Am. Journ. of Science, 1905, XX, p. 148.

was removed by scraping the outermost layer of the black heterosite with a knife. In this way 0.0466 gr was obtained from 21 gr heterosite. The result was as follows:

	%
H_2O	14.1
Fe_2O_3	23.2
Mn_2O_3	29.1
CaO	trace
P_2O_5	31.2

The figures are only approximate. As already Brush and Dana have anticipated for Brancheville material, no definite composition of these alteration products is to be expected. The observations from Varuträsk at any rate signify that the black coating on the heterosite, penetrating the mineral by every cleavage and crevace, is a strongly hydrated Fe—Mn phosphate.

The sodium-accentuated sub-series.

We now pass over to what I think may be termed a parallel sub-series of the triphylite — lithiophilite group.

Already at an early stage of the investigation a $MnO—Fe_2O_3$ phosphate was found, which did not correspond with any species of the former series. The mineral occurs rather scarcely in well defined nodules of 2—3 cm in diameter. At Lövlunden a nodule 10 × 4 cm was observed. On weathered surface it is always of a dull canary-yellow colour, most often appearing as by weathering hollowed, oval-shaped yellow nodules. On cleavages it is of a dull black colour. Dipped in acid it readily turns yellow, contrasting to the brilliant purple of heterosite, when treated in the same way. Also streak and powder are of a dirty yellowish tinge.

Under the microscope the mineral shows itself to be remarkably pure. Translucent in yellow to yellowish-green colours and in contrast to heterosite, wholly unpleochroitic, it is crystalline in radiating fibrous or globular aggregates. The refractive indices are α 1.760, γ 1.775.

It was from the beginning apparent that this mineral represented another species of Fe — Mn phosphates than those dealt with above. Prof. Aminoff of the State Natural History Museum therefore kindly allowed an analysis to be made by the chemist of the Mineralogical Dept., dr R. Blix, which gave the result stated under 1 in the following table. Analysis 2 refers to the mineral alluoudite, of DAMOUR (1848) which is nearly related both in chemical composition and field relations and to which I am inclined to refer the Varuträsk mineral.

	1.	1 a.	2.
Unsoluble	1.24	—	0.60
H_2O − 105°	0.24	—	—
H_2O + 105°	0.66	0.037	2.65
Fe_2O_3	19.83	0.121	25.62
Mn_2O_3	—	—	1.06
MnO	28.17	0.379	23.08
MgO	1.17	0.030	0.00
Na_2O	7.20	0.116	0.47
P_2O_5	46.70	0.287	41.25
F_2	0.80	0.021	—
	100.03		99.73
—O for F	0.19		
	99.84		
Sp. gr.	3.576		Sp. gr. 3.468

1. Alluoudite, Varuträsk.
1 a. Mol. prop. of d:o .
2. Alluoudite, Chanteloube, France.

The analysis 1 works out satisfactory to the formula:

$$5\ R_3(PO_4)_2\ Na(OH,\ F).$$

All Fe and Mn have then been reckoned as bivalent and enough Na_2O has been deducted for $H_2O + 105$ and F. R represents MnO, Fe_2O_3 and remaining Na_2O. Bases to P_2O_5 are again nearly exactly 3 : 1.

It is not strictly possible to reckon the old analysis from Chanteloube analogously as the relatively high content of water has only been determined in the lump. As P_2O_5 only suffices to bind the Fe — Mn oxides, enough (OH) must be deducted for Na_2O. An approximate formula would then be

$$3\ R_3(PO_4)_2 \cdot Na(OH)$$

again counting Fe as FeO. Though the two analyses show differences, these are hardly greater than can be accounted for by impurity of material in the older work. On the other hand, so much in general character and field occurrence is in common, that I believe the two minerals to refer to the same species.

The original alluoudite from Chanteloube was supposed to be derived from triphylite through alteration. Already LACROIX[1] has, however,

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[1] Min. de France IV p. 366.

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doubted this interpretation and is more inclined to presume that natrophilite has been the original source of alluoudite, which would more easily explain the high content of Na_2O. On the other hand it is of course possible to assume an independent invasion of Na_2O, which for example naturally must have accompanied the extensive albitisation during that epoch of mineralisation. It may be remembered, how spodumen can be altered to a mixture of eukryptite and albite, inferring a substitution according to BRUSH and DANA[1] from 7.62 Li_2O and 0.39 Na_2O in fresh spodumen to 3.61 Li_2O and 8.29 NaO in β-spodumen. In our case such a process is, however, hardly acceptable, as the epoch of albitisation clearly preceeds the formation of the Li — Mn phosphates. A third explanation relating to a new mineral species will be referred to below.

To begin with, I was at a loss where to seek the primary mineral, from which the alluoudite must be derived through partial oxidation, and was most inclined to accept LACROIX's idea of natrophilite as the primary source. This idea was strengthened by the statement that natrophilite often occurs in closest association with the minerals of the triphylite — lithiophilite series. Already BRUSH and DANA[2] have emphasised this, relating to material from Brancheville, and I noticed that they mention an auburn yellow secondary phosphate, found along cleavages of the natrophilite, which might well be alluoudite.

At this stage my attention was however drawn to a mineral, described by HEADDEN[3] from Nickel Plate mine, Pennington Co., S. Dakota with an analysis which might well represent the primary source for a mineral such as alluoudite. The analysis, given as no. 2 in the following table, shows all bases as bivalent and 7.46 Na_2O against 0.28 Li_2O.

A search was now made on all material at hand from Varuträsk to try to find some rests of the primary mineral. The result was however negative. It is miss BERGGREN I have to thank for discovering the mineral I was in search of. She visited Varuträsk in september 1936 and I had asked her to look out for a probably green phosphate, nearly associated with alluoudite, by now a familar mineral to us both. MISS BERGGREN while making a collection of the minerals at the locality, succedeed, after a good deal of searching in finding half a dozen small specimens of a dark green phosphate, closely associated with alluoudite. It was soon evident that a new and distinct mineral species had been found.

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[1] Zeitschr. f. Kryst. V p. 195.
[2] Zeitschr. f. Kryst. XVIII p. 11, 13.
[3] Am. Journ. of Science 1891, XLI, p. 416.

The mineral is of a dull olive-green colour. It occurs in fine granular masses, in size and form about coinciding with the alluoudite. In one specimen one seemed able to follow a gradual transition to alluoudite, and as this observation seemed to verify my assumption relating to the connection between the two minerals, material for analysis was taken from this specimen.

Under the microscope the mineral is colourless. Two well-marked cleavages at right angles probably are the same as in the triphylite series, (001) and (010). The axial plane is, however, vertical to the best cleavage. If this also here is (001), the plane would be parallel (010). Refractive indices are α 1.720, γ 1.732. The axial angle is destinctly larger than in triphylite, $2V_{Na}$ is 70°. Dispersion distinct $\varrho > v$, but not nearly so strong as in triphylite.

Most grains are intimately interwoven with a yellowish green phosphate, without doubt alluoudite, representing beginning alteration to that mineral.

Intricately mixed with turmaline and lepidolite, it was not easy to procure pure material for analysis, but after much work miss Berggren succeeded. The analysis gave:

	1.	1 a.	2.
Unsoluble	1.80	—	2.47
$H_2O - 105°$	0.14	—	—
$H_2O + 105°$	0.75	0.042	0.73
Fe_2O_3	8.35	0.052	—
Al_2O_3	0.36	0.005	—
FeO	7.52	0.105	25.05
MnO	25.30	0.357	15.54
CaO	4.86	0.087	5.53
MgO	0.00	—	1.50
Li_2O	0.88	0.030	0.28
Na_2O	7.12	0.115	7.46
K_2O	0.12	0.013	2.00
P_2O_5	42.80	0.301	38.64
F_2	0.06	—	0.69
	100.06		99.89
Sp. gr.	3.581.		3.612

1. Varulite, Varuträsk.
1 a. Mol. prop. of d:o .
2. Unnamed phosphate (Headdenite), S. Dakota.

As the ratios $RO : R_2O_3$ again naturally are accidental, no definite formula can be expected, except reckoning all bases as bivalent. One then gets approximately the formula:

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3.612

idental, no definite
bases as bivalent.

$Na_2O\ 5RO\ 2P_2O_5$ or $Na\ 2^1/_2\ (Mn\ Fe\ Ca)\ (P\ O_4)_2$

The small amount of water and F are left out of reckoning. The proportions MnO : FeO are those of lithiophilite.

For this mineral I propose the name Varulite, the name taken from the first half of the locality of Varuträsk.

HEADDENS phosphate gives in the same reckoning, deducting enough alkalies for F only, approximatly the same formula but with other proportions MnO : FeO, indicating relation to triphylite.

The material of varulite, as yet found, is not fully unaltered. The analysis signifies, as already the thin sections under the microscope indicated, a beginning alteration to alluoudite. About just as much Fe is oxidized as in the analysis 3 of the triphylites, given above. If we therefore as yet have not found absolutely fresh material, it may be remembered that the somewhat oxidized triphylite (analysis n:o 3 p. 81) for a long time represented the freshest material I had at my disposal of that mineral. It was first at a late stage of the investigation that perfectly fresh triphylite was found.

It may, I think, be assumed that the new sodic phosphate varulite represents the original source for the alluoudite. The question now arises where to place this mineral in the larger group of triphylite — lithiophilite — natrophilite phosphates. In discussing this question I will presume that we may deal with the fully unoxidized phosphate, containing all bases as bivalent.

It seams difficult to assume that natrophilite has been the source. A reduction of Na_2O from the theoretical 17.9 % in natrophilite to 7.12 % without any oxidation or hydration would be difficult to explain. The proportions at hand are nearer the formula $^1/_2\ Na_2O\ 2^1/_2\ (Mn\ Fe)O\ P_2O_5$ than $Li_2O\ 2(Mn\ Fe)O\ P_2O_5$ in lithiophilite which would imply that half the content of alkalies in, for example, the lithiophilite molecule have been substituted for an equivalent amount of Mn—Fe. In reference to the proportions Mn — Fe HEADDEN's phosphate would represent the same condition relating to triphylite. If in future this supposition should be verified, we would have to reckon with two parallel groups: the Li-series, triphylite—lithiophilite and a Na-accentuated series, as yet represented by HEADDEN's phosphate, which provisionally may be named headdenite and varulite, the former corresponding to triphylite, the latter to lithiophilite in Fe : Mn relations. This second series would be characterized by proportions $R_2O : RO$ 1 : 5 instead of 1 : 2, in the former series. The small Na content in triphylite, as documented in analysis 3 and also referred to on page 82, may well infer a primary intermixture of varulite.

This would also explain the occurrence of alluoudite in heterosite, alluded to on page 87.

Regarding the relations of the whole group and the different stages of alteration, one might outline the following succession under progressing oxidation:

Li-series.		Na-series.	
1.	2.	3.	4.
triphylite	lithiophilite	headdenite	varulite
↓	↓	↓	↓
Fe-sicklerite	Mn-sicklerite	Fe-alluoudite	Mn-alluoudite
↓	↓	↓	↓
heterosite	purpurite	Na-heterosite	Na-purpurite

Spaced-out types are verified by analyses. Natrophilite has not been included, as analytical data of this mineral as yet only refer to one single analysis of Brancheville material. Nothing gainsays, however, that the natrophilite series may be devided in Na(Mn, Fe)PO_4 and Na (Fe Mn)PO_4 subdivisions according to the relative proportions of Mn and Fe, each followed by its series of oxidation products in accordance with the scheme above. No such species are, however, as yet known, and if the intermediate stages are missing, it would be difficult to identify the terminating alteration product from purpurite in general, as the alkalies would be missing and their character and proportions to the other bases undefined. This refers also to Na-heterosite and Na-purpurite.

Denoting the valency of Fe and Mn the relations of the minerals may be illustrated as follows:

1.	2.	3.	4.
$Li, (\overset{2}{Fe}, \overset{2}{Mn})PO_4$	$Li, (\overset{2}{Mn}, \overset{2}{Fe})PO_4$	$Na, 2\frac{1}{2} (\overset{2}{Fe}, \overset{2}{Mn})2PO_4$	$Na\ 2\frac{1}{2} (\overset{2}{Mn}, \overset{2}{Fe})2PO_4$
↓	↓	↓	↓
$(Li, \overset{3}{Fe}, \overset{2}{Mn})PO_4$	$(Li, \overset{2}{Mn}, \overset{3}{Fe})PO_4$	$(Na, \overset{3}{Fe}, \overset{2}{Mn})PO_4$	$(Na, \overset{2}{Mn}, \overset{3}{Fe})PO_4$
↓	↓	↓	↓
$(\overset{3}{Fe}, \overset{3}{Mn})PO_4$	$(\overset{3}{Mn}, \overset{3}{Fe})Po_4$	$(\overset{3}{Fe}, \overset{3}{Mn})PO_4$	$(\overset{3}{Mn}, \overset{3}{Fe})PO_4$

As mentioned, a subdivision of the natrophilite series might give analogous groups 5 and 6.

Min. Dept., Univ. of Stockholm, February 1937.

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