

I. THUCHOLITE, A REMARKABLE PRIMARY CARBON MINERAL FROM THE VICINITY OF PARRY SOUND, ONTARIO

II. CYRTOLITE INTERGROWTH ASSOCIATED WITH THE PARRY SOUND THUCHOLITE

H. V. ELLSWORTH*, *Canada Geological Survey.*

I. THUCHOLITE

A pegmatite dike which traverses the line between lots 9 and 10, conc. IX, Conger township, Parry Sound District, carries a number of rare mineral species, including uraninite,¹ calcio-samaraskite², allanite, and the two minerals described in these papers.

As seen by the writer, the dike was exposed for 70 feet along the north side of a small hill on the shore of Blackstone Lake. It had been uncovered for the most part by stripping, but as the contacts with the country rock were not exposed except for some small patches of hornblendic gneiss at the eastern end of the dike, it was not possible to ascertain the dip and strike, nor the true width. However, the exposed width on the hillside was 60 feet or more and the probable strike about N 60° E. It may be a comparatively narrow dike dipping to the north along the slope of the hill.

About one half of the exposed dike surface was massive white quartz with occasional inclusions of crystal microcline up to a foot or more in diameter, and large crystals of muscovite some of which reached a diameter of 18 inches, but were rarely if ever over an inch or two thick, i.e., along the *c* axis. These large muscovite crystals usually occurred at the contact of the massive white quartz and feldspar, sometimes parallel to the contact, sometimes normal to it, in which latter case one half the diameter of the mica book would be imbedded in quartz, the other half in feldspar. Sheets of bent and crumpled very black biotite, as much as two feet in diameter, were seen at the contact of the massive quartz with the granular quartz-feldspar-mica mixture. A few small flakes of molybdenite were found but this mineral is rare.

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¹ *Geol. Survey Canada*, Summary Rept., 1921, Part D, p. 60D et seq.

² This Journal, Volume 13, p. 66, 1928.

The other half of the exposed dike surface consisted mainly of a granular mixture of smoky quartz, pink to red feldspar in which more or less polysynthetic twinning could usually be seen, and small thin crystals of muscovite and biotite, chiefly the former. Some patches of crystals of microcline up to 2 or 3 feet in diameter and other patches in which muscovite, and to a less extent biotite, occur in irregular clusters and fan shaped aggregates of thin crystals, are also included in this area. Uraninite occurs as scattered crystals in this part of the dike, particularly in association with stringers and aggregates of thin muscovite crystals, sometimes actually embedded in the mica, sometimes in quartz or feldspar usually near the mica. The uraninite crystals vary from $\frac{1}{8}$ inch to a rare maximum of one inch in diameter, the average being $\frac{1}{4}$ inch or less. The crystals are cubic in habit showing usually only a small development of the octahedron.

Calciosamarite was found in much less quantity than uraninite though under the same conditions, except that it seems to be more closely associated with the biotite. Associated with both uraninite and calciosamarite are occasional occurrences of a black mineral, the thucholite of this paper, which might easily be mistaken for uraninite when embedded in the rock except that it never shows indications of crystal form. The thucholite occurs in exactly the same way as the uraninite, embedded in feldspar, quartz or mica, or in a mixture of all three and is certainly just as much a primary mineral as are any of those just mentioned (Fig. 1). It takes the form of irregular rounded nodules or more or less roughly globular masses from 1 millimeter to one inch in diameter (Fig. 2). In the form of small round grains, 1 to 3 millimeters in diameter, it is rather abundant in some of the finer grained parts of the granular mixture of smoky quartz, feldspar and small mica where uraninite does not occur. Occasionally a uraninite crystal is completely embedded in thucholite and the calciosamarite also is sometimes in contact with it. A few of the nodules collected were clean, lustrous and homogeneous throughout from center to outside, some have an irregular cavity in the center containing crumbly, decomposed, highly uraniferous material which appears to have been originally uraninite, and one was collected which still contained a core of fairly well preserved uraninite. The majority, however, while consisting internally of pure lustrous thucholite were outwardly more or less intergrown with the

cyrtolite intergrowth described in II. The outer surface of thucholite nodules usually also contains minute embedded scales of white and black mica. The latter is very black and opaque except in the thinnest possible cleavages. It is biaxial with $2E=50^\circ$, approximately. Such chemical tests as it was possible to make on the small quantities available did not indicate that it was different from ordinary biotite. One might suspect it to contain uranium and vanadium but the possibility of contamination from its associates would in any case render results on such small quantities dubious. Associated with the cyrtolite and black mica on some of the thucholite nodules are small patches of a powdery white mineral (Fig. 2-B) apparently a decomposition product, which so far as it has been examined appears to be a new mineral, a hydrous phosphate or silico-phosphate of calcium and yttrium earths. As this mineral can only be isolated in the most minute quantities much more work will have to be done before it can be fully described.

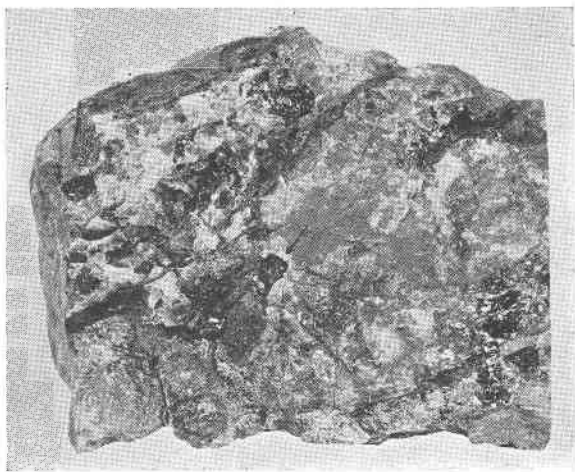


Fig. 1. Photograph about natural size showing thucholite nodule, indicated by arrow, in a mixture of feldspar, white and smoky quartz.

It appears likely that the thucholite varies considerably in composition. Nodules which are intimately in contact with uraninite naturally would be likely to carry more uranium than usual, and the ash of such specimens in fact is sometimes almost

black and highly uraniferous, while the ash at other times may be cream colored, greenish (from presence of V) or yellowish. Thucholite was first identified by the writer in 1921 but it was not until some years later that enough of the material was obtained to justify undertaking quantitative studies. Finally about 10 grams was assembled and from this lot small fragments of the most lustrous material free from any visible impurities were carefully selected for the tests under a binocular microscope equipped with a Silverman Illuminator.

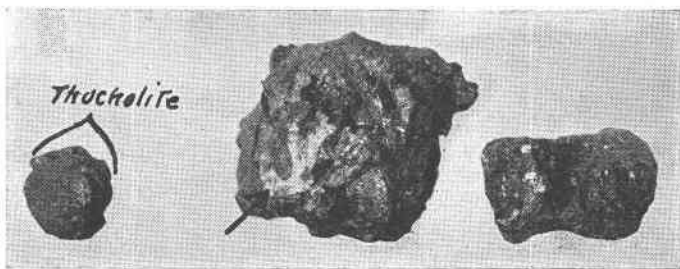


Fig. 2. Specimens of Parry Sound thucholite about twice natural size.

- A Uraninite nodule originally completely enclosed by thucholite coating, part of which is still visible at the top.
- B Thucholite mass showing the white mineral mentioned in Part II, indicated by arrow.
- C Typical thucholite nodule showing rounded outlines.

PHYSICAL PROPERTIES, PYROGNOSTICS, ETC.

Thucholite is jet black with brilliant lustre and perfect conchoidal fracture. Cleavage none. Very brittle. Hardness 3.5 to nearly 4. It is absolutely opaque even in the smallest grains with arc illumination. Sp. Gr. = 1.777 at 17.74°. In the closed tube yields much water but no hydrocarbons.³ Fragments if quickly heated may explode into powder with considerable violence, the particles being projected in all directions. It evolves gas copiously if immersed in water and heated. Digestion with 10 per cent HCl removes the greater part of the rare element constituents, and the solution if filtered and evaporated shows

³ The mineral of course shows considerable radioactivity easily detectable in the electroscope and it produces the same radial fracturing in feldspar or quartz enclosing it as do other radioactive minerals.

the yellowish green color due to uranium (yellow) and vanadium (green). Concentrated nitric acid at the boiling temperature slowly attacks the thucholite, the acid becoming brown colored. The carbon can be completely destroyed by heating with concentrated H_2SO_4 and KNO_3 . Concentrated cold H_2SO_4 seems to have little or no effect on the carbon, with long heating, however, the acid becomes brown colored. Strong hydrochloric acid (33 per cent HCl) can be boiled with the mineral for hours without becoming colored in the slightest degree by carbon compounds. Of course any of the mineral acids dissolve out the rare element compounds. The mineral burns slowly in air when maintained at a red heat yielding a light, pulverulent ashy residue. The ash retains to a great extent the shape of the original fragments, but with some reduction in size, though it crumbles to fine powder at the slightest touch. Under the binocular microscope it very closely resembles fragments of dried bentonite. The prevailing color of the ash fragments is a creamy-yellow but almost an equal number are greenish, and occasional minute black specks may be seen where no doubt there was a concentration of uranium. Some fragments may be in part greenish, in part yellowish. As obtained by ignition it shows no sign of fusion at a high red or nearly white heat in a platinum crucible heated over a large Meker burner.

Attempts to make sections thin enough to transmit light were unsuccessful, but a very good polished surface was obtained by first embedding a nodule in dentist's phosphate cement, grinding and polishing as usual. When examined by reflected light, this surface appeared to be entirely homogeneous, even under the highest powers, except for a few minute hair-like lines near the outer edge, which stood in slightly higher relief. These lines looked much like extremely minute cracks which had been filled with some harder material of the same nature. The proportion of this harder material would be much less than one per cent of the surface examined.

COMPOSITION

Three different samples of approximately 0.1 gram gave by combustion in oxygen; carbon = 45.18, 42.24, 50.48 per cent. No precautions were taken to insure complete combustion of CO and probably these results are low.

Three different samples (not corresponding to above) yielded results as follows:

	I	II	III
H ₂ O at 110°	7.64	8.63	8.40
H ₂ O total, direct.	13.20	13.02	12.96
Ash	24.48	24.20	28.06

The purpose of the foregoing experiments was to find out whether the substance has a constant composition. Unfortunately it was not practicable to obtain each sample from one nugget only, so that possibly all the samples represented more or less of an average of the lot from which they were selected.

The ash of various determinations was combined and analyzed with the following results:

ANALYSIS OF 0.40 GRAM FRESHLY IGNITED ASH OF THUCHOLITE,
PARRY SOUND, ONT.

Water soluble	(Not definitely identified)	1.60
PbO		0.20
U ₃ O ₈		5.80
ThO ₂		48.48
(Ce, La, Di) ₂ O ₃		5.45
(Yt, Er) ₂ O ₃		10.95
Fe ₂ O ₃		1.50
V ₂ O ₅		2.25
MnO		0.02
TiO ₂	doubtful	trace
Al ₂ O ₃ , etc.		1.45
¹ CaO		0.50 (3.00)
MgO		traces
ZrO ₂		0.80
² K ₂ O		0.15
³ Na ₂ O		0.22
P ₂ O ₅		3.21
SiO ₂		14.70
NiO } CoO }	Not detected on small separate amount of ash	
		97.28

¹ Probably should be about 3.00 per cent.

² Alkalies pptd. by Pt.

³ Alkalies not pptd. by Pt.

If a larger quantity of ash had been available it would doubtless have been possible to attain a more satisfactory summation. As

it was, all the constituents had to be sought for and determined in the one small lot, and as nearly half of the ash was thorium oxide all the other constituents together amounted to only about 0.2 gram. The low total is believed to be due to slight losses incident to the necessarily complicated analytical procedure rather than to failure to determine some constituent or constituents actually present. A review of the analytical notes indicates that possibly much of the deficiency was due to slight losses of lime occasioned by repeated operations on other substances from which the lime was supposed to be completely separated but actually was not. Thus it seems likely that most of the missing 3 per cent should be credited to CaO, especially as this amount was subsequently found on another sample in 1927. Owing to the presence of phosphorus, vanadium and zirconium, numerous operations and exceptional precautions were necessary to ensure pure precipitates, especially in the case of uranium and thorium, hence these are probably slightly low. With the exception of the ZrO_2 , which, being small in amount, was not especially purified, the analytical results as given therefore represent minimum percentages of the constituents present, and as thus defined are believed to be trustworthy. Vanadium was separated with certainty from uranium by cupferron. The thorium weighed was free from zirconium.

RADIOACTIVITY OF THE ASH

Previous to the analysis, the radioactivity of the ash was compared with that of standard analyzed carnotite in a U. S. Bureau of Mines type α -ray electroscope. Equal weights and surface areas of ash and 1.55 per cent U_3O_8 carnotite were compared, with the result that the ash was found to be 13.6 times as active as the carnotite, indicating an activity equivalent to 21.08 per cent U_3O_8 , which agrees quite well with the analytical results (24.9 per cent U_3O_8 equivalent), considering that radon and thoron were doubtless driven off by the ignition and that the test was made before they had again reached equilibrium. In view of the small amount of lead found it would have been interesting to have determined radium and mesothorium accurately by the emanation method to see whether these were present in amounts corresponding to the normal equilibrium ratio, or in other words, to see whether lead only is being removed faster than

it is formed, or whether not only lead but also radium and mesothorium are being leached away.

An attempt to determine the gas content by weight was carried out as follows. The sample in fragments was introduced into a small silica tube closed at one end, which was weighed before and after. Crushed and sifted, freshly ignited, transparent, fused silica was then inserted on top of the mineral to prevent loss by decrepitation and a plug of dry glass wool was inserted to hold down the fused silica powder. The whole was then weighed and attached to a mercury vapor vacuum pump. After exhausting until long after all air had been removed the mineral was gradually heated to a white heat with a gas blast lamp, the heating being continued until tests with the McLeod gauge attached to the pump indicated that only unweighable quantities of gas were coming off. The tube was allowed to cool, was detached from the pump and weighed. The results are given below:

	Per Cent
Loss by vacuum ignition	26.08
H ₂ O—direct, same sample	(12.96)
Gas by difference	(13.12)
Ash—same sample	28.06
Fixed Carbon, by difference	45.86
	100.00

Occurrences of diamonds in pegmatites have been reported⁴ and it was thought to be worth while to look for them in the thucholite ash, but none were found.⁵ During the washing of the silica of the analysis some very minute almost perfectly spherical balls attracted attention by rolling around in the platinum dish. These had withstood the action of concentrated H₂SO₄ over night at the fuming temperature. Three were secured for examination. Two measured a little less than 0.1 mm. in diameter while the largest was nearly 0.4 mm. across. Immersed in oils under the microscope the large ball was absolutely isotropic with index = 1.518 and contained numerous minute bubbles or inclusions. The color of the balls varied from colorless to greenish.

⁴ According to Mellor, *A Comprehensive Treatise on Inorganic and Theoretical Chemistry*, Vol. V, p. 737—"M. Chaper found diamonds in the pegmatites of Bellary, Madras."

⁵ After treatment with HF and H₂SO₄ any small residue of lead sulfate, etc., is completely soluble in HCl leaving not the slightest trace of insoluble material.

It was suspected that these balls might have been produced by fusion of ash around concentrations of material rich in alkalies or phosphates, but if that were so their resistance to fuming sulfuric acid is rather surprising. About 0.2 gram of the thucholite in small fragments was destroyed by heating with concentrated H_2SO_4 and KNO_3 and no balls or insoluble residue were seen in this solution. Since, however, only a half dozen balls were seen in 0.4 gram of ash they might exist in the original material and still be missed by the wet test on the small quantity.

The foregoing work was performed some years ago by the writer. Last year Dr. A. L. Day, Director of the Geophysical Laboratory, Washington, very kindly enlisted the services of Dr. E. S. Shepherd, the noted authority on volcanic gases and gas analysis, for an investigation of the gas content of thucholite. By permission of Dr. Day the report of Dr. Shepherd covering his analysis of the gases of thucholite, is reproduced below.

THE VOLATILES IN THUCHOLITE

E. S. SHEPHERD

"Dr. H. E. Mervin with the aid of the binocular microscope kindly selected from the better sample submitted some three decigrams of the most homogeneous seen.

METHODS.—A slight variation of the regular procedure was thought advisable. The mineral was placed in a small platinum capsule equipped with a fairly tight lid, which was wired on. This capsule was then placed in a Lawrence Smith Alkali tube containing pure CaO . The tube was capped and the cap secured. The combined tubes were then placed in the silica tube in the furnace and the exhaustion carried out as described in my paper on methods.⁶ Of course the platinum, silica and CaO had all been previously exhausted at 1100° before the mineral was introduced.

The lime used was intended to stop sulfur, halogens, and phosphorus, but at the temperature of evacuation would not hold CO_2 nor H_2O . At the close of the run there was no evidence of carbon deposited in the lime as one might have expected had

⁶ The Analysis of Gases Obtained from Volcanoes and from Rocks, E. S. Shepherd, *Journal of Geology*, Vol. XXXIII. Supplement to Number 3, 1925 (H. V. E.).

there been heavy hydrocarbons or tar present, nor did any such deposit appear when the lime was later dissolved for analysis. This fact combined with the results from the combustion indicates the absence of heavy hydrocarbons.

TEMPERATURE OF EVACUATION 1000–1100°C., TIME ABOUT 20 HOURS.

Material taken	0.3310	Ash = 3.48 per cent.
After	0.1909	Carbon (by difference)
		= 54.20 per cent.
Loss	0.1401 or 42.3 per cent.	
Weight of volatiles recovered	0.1395 or 42.1 per cent.*	

* Note. The agreement implies an unusual series of compensating errors since the methods claim no such precision.

Total volume of "fixed gases" at 0°, 760 mm. = 173.4 cc.

Total volume volatiles at 1200°, 760 mm. = 1016.5 cc.

Volume per cent at 1200°, 760 mm.

CO ₂	6.444
CO.....	35.827
CH ₄	1.500
H ₂	44.498
N ₂	3.562
A.....	0.010
S ₂	0.063
Cl ₂	0.033
F ₂	0.167
H ₂ O.....	7.857
	cc/gm = 3071

All rare gases are lumped as argon because there was too little to justify an attempt to separate them. The value $A/A+N \times 100 = 1.5$ is but little above the argon ratio for atmospheric nitrogen.

Phosphorus was detectable in the lime but not enough to bother with.

H₂O was 3.59 per cent by weight, thus lower than obtained by Ellsworth, but the total volatiles runs much higher. From the nature of the material no very definite ratio was to be expected. The ash varies greatly in color. Some pieces are white and some green, indicating irregular distribution of metallic oxides."

Geophysical Laboratory,
May 2nd, 1927.

E. S. SHEPHERD.

The great discrepancies in regard to ash, gas and H₂O content between Dr. Shepherd's results and the writer's can only be explained by supposing that specimens of thucholite from different parts of the dike vary greatly in quantitative composition. The specimens examined by the writer had been derived mainly from one particular small area of the dike and evidently all had much the same composition. Since the time of the writer's work, however, various additional specimens were assembled in one tray along with what was left of the original lot. The lot sent Dr. Shepherd later on was thus taken at random from a mixed assemblage representing more widely separated parts of the dike. The differences in our results apparently mean simply that thucholite is not nearly so constant in quantitative composition over the whole dike as was indicated by the writer's results for specimens from a small restricted area. In another paper⁷ it will be seen that not only the amount of ash but also its percentage composition is quite variable, though the same constituents are present in both cases.

The work of Dr. Shepherd adds much to the interest of this remarkable mineral. It was surprising enough to find a carbon containing 25 to 28 per cent of rare element compounds and yet yielding no visible evidence of their presence, but when in addition to this we find an enormous gas content in which hydrocarbons are practically absent, one cannot but feel that we have in thucholite a priceless scientific key mineral, which if carefully studied might yield important new information in the fields of both chemistry and geology. A full discussion of the possible significance of thucholite would occupy too much space here but a few important points might be briefly noted. The occurrence of carbon as a magmatic mineral is established, and it looks as if we have absorbed in thucholite a sample, preserved through some 1000 million years, of the final gases of a Precambrian granite magma, with the exception of helium which is not appreciably retained by carbon. It becomes evident also that uraninite is formed under strongly reducing conditions in the presence of carbon, carbon monoxide, and hydrogen and therefore we can be practically sure of its essentially UO₂ initial composition. The foundation for the autoxidation process in uraninite, as suggested by

⁷ Thucholite and Uraninite from the Wallingford mine, near Buckingham, Quebec. This Journal, following article.

the writer thus becomes secure. Further, one might speculate as to whether the noteworthy differences between uraninite, which occurs in crystals, and the always amorphous pitchblende, may not be due to differences in the reducing effect of the magmatic vapors or solutions at the time they were formed.

Under highly reducing conditions such as prevailed when thucholite was formed uranium would appear as the lowest oxide (UO_2) which has a strong tendency to crystallize, so much so that artificial crystals can be easily prepared. Isomorphous oxides as ThO_2 (which is definitely known to be isomorphous with UO_2) and rare earth oxides (about which less is known at present) would be likely, if present, to crystallize with the UO_2 as uraninite. On the other hand, under less completely reducing conditions and probably at lower temperatures representing a later stage of the magmatic vapors, uranium would probably precipitate as U_3O_8 with which ThO_2 and rare earths are not isomorphous, and which has little if any tendency to crystallize. Thus we have a rather plausible explanation for the remarkable fact that uraninite always contains thorium and rare earths besides relatively large amounts of UO_2 , often greatly exceeding the ratio of UO_2 in U_3O_8 , and is always crystallized, whereas pitchblende contains only traces, if any, of thorium and rare earths and has a UO_2 content not greater in amount than is required for U_3O_8 and is never crystallized. The fact that carbon occurs in various forms in nickel-iron meteorites, as diamond in the blue ground pipes, etc., is sufficient evidence that it can be a primary constituent of deep seated rocks. In the case of the Parry Sound thucholite there seems to be little or no immediate ground for suspicion that the carbon may have come from limestones assimilated by the magma, the time-worn explanation usually advanced for all occurrences of calcite, graphite or other carbonaceous materials in igneous rocks, but unfortunately it cannot be said that the possibility of a limestone origin can be entirely ignored even here. There is comparatively little limestone in the Precambrian region bordering Georgian Bay and none in the immediate vicinity of the thucholite occurrence so far as the writer is aware, the nearest outcrop seen being a band perhaps a 100 ft. wide which crosses the junction of the concession road between McKellar and McDougall township and the road leading to McKellar and Dunchurch, about 16 miles from the thucholite dike. This band seems

to strike about N25° east magnetic and probably continues with increasing volume through Limestone Lake, Fairholme P. O. and past Dunchurch. The strike of this band apparently would carry it no closer than about 10 or 12 miles NW of the thucholite locality. The absence of contact pyroxenites (consisting largely of sahlite and calcite) formed by the action of granite on limestone, which are so characteristic of regions such as the Gatineau and Lièvre river, where limestone is abundant, is further evidence that such action has not occurred in the neighborhood of the thucholite dike.

During the present year the writer performed some further experiments on thucholite. It was thought for one thing, that possibly lead had been lost by volatilization during the combustion of the mineral, and that perhaps a greater percentage could be obtained by acid extraction. As material a single nodule was selected, which for apparent purity and homogeneity was the best of the whole collection. This was a solid nodule showing under the binocular only a few very small patches of decomposed cyrtolite intergrowth confined to the outer surface. The cyrtolite was cut away and the outer surface filed until no further impurities could be seen. The nodule was then broken into fragments and about 0.4 g. of apparently pure fragments were selected. Of this 0.1100 g. was burned in a platinum crucible yielding 0.0243 g. of ash equal to 22.09 per cent. The remainder, 0.3065 g. in weight, was leached with acids and the acid extract with the washings evaporated in a small platinum dish, ignited to oxide and weighed, the extracts of chlorides of course being first converted to nitrates before ignition.

		Grams
Extracted by H ₂ O only	3 hrs. on hot water bath	0.0005
“ “ 10% HCl	3 “ “ “ “ “	0.0289
“ “ 10 “ “	3 “ “ “ “ “	0.0087
“ “ 10 “ “	6 “ “ “ “ “	0.0072
“ “ 35 “ “	16 “ “ “ “ “	0.0068
“ “ *HF, H ₂ SO ₄ and final 35 per cent HCl		0.0088
		<hr/> 0.0609

* This extraction was first by HF only, followed by 3 hours with concentrated H₂SO₄ at the fuming temperature, and after washing by final treatment with 35 per cent HCl overnight on hot water bath, followed by a long treatment with hot water. All these separate extracts being small were combined and the total weighed 0.0088 as oxides. The long treatment with hot concentrated H₂SO₄ attacked the mineral somewhat, the acid becoming dark brown and on evaporation a small amount of a yellowish organic compound appeared in the dish.

It had been observed in the earlier work that there was a slight amount of water soluble substance. Microchemical and spectroscopic tests indicated that the water soluble material is mainly sulfate and chloride of sodium. A Nessler's test for ammonia gave positive indications also, but the ammonia may have come from the laboratory air or water.

The appearance of the thucholite fragments viewed under the binocular microscope was not noticeably changed after the severe acid treatment, except that some pieces developed ordinary fractures due to internal strain, and in some fragments very minute fissures, apparently resulting from solution, were visible. It has been mentioned previously that a polished surface examined by reflected light appeared to be entirely homogeneous except for the presence of a few very minute lines of harder material standing in relief. The solution fissures just mentioned have exactly the appearance that would result if these lines of harder material had been dissolved out. Otherwise the leached grains seemed to have retained their original brilliant lustre.

The specific gravity of the acid leached thucholite dried at 120° was determined after thorough boiling in a silica pycnometer, to be 1.75. This value which is nearly that (1.777) obtained on the unleached material of the earlier experiments is perhaps due to the leached mineral being more readily penetrated by water.

The oxides obtained by acid leaching amount to 90.09 per cent of the total obtained by combustion, and as probably much of the silica was retained by the carbon and volatilized by the HF treatment, it was concluded that the extraction was nearly complete. A 0.0969 gram portion of the original material dried at 120° was brought to a red heat in a covered platinum crucible, not more than one minute being required, and the material on weighing was found to have lost 0.0338 gram, so that there appears to be about 30 per cent of volatile matter present. About 15 minutes ignition with the crucible open in a large Meker flame was required to burn the remainder. The ash remaining weighed 0.0004 g. or about 4 per cent. Thus it appears probable that the ash could be entirely removed by acid leaching if the material were ground fine and given a sufficiently prolonged treatment.

The 0.0604 gram of extracted oxides were united and as a certain amount of silica had been extracted along with the other

substances, this was removed by HF and H₂SO₄. A few of the main constituents were then determined with minimum results for the carefully purified oxides as follows:

	Grams	Per Cent
PbO.....	0.0002	0.3
CaO.....	0.0018	3
ThO ₂	0.0224	37
R.E.....	0.0087	14
*ThO ₂ +R.E.....	0.0025	4

* Slightly impure recoveries not further treated.

There was a comparatively large amount of uranium present but owing to an accident it could not be determined. V, P₂O₅, Fe, etc., were present as usual. This analysis confirms the suspicion that the CaO of the earlier more complete analysis was low. No more lead was obtained than in the first analysis. Nickel and cobalt were sought in the 0.0243 of ash from the combustion of this lot, but no trace of either was found.

When the author in 1921 first identified the mineral here referred to as thucholite he was inclined to regard it with some justification as something entirely new and unheard of. As a matter of fact there appears to be no reference to any such mineral in the recognized works on mineralogy, though as it subsequently turned out this mineral had been found and superficially described long ago in both Sweden and Canada. A chance reference led to the following article in *Comptes Rendus*, Vol. **CXVI-1893**, pp. 677-678, entitled *Extrait d'une Lettre de M. Nordenskiöld à M. Daubrée*. After some remarks on the occurrence of diamond the writer continues:

Ne rencontrez-vous pas des bitumes ou de l'asphalte dans les roches cristallines? Comme vous savez, nous en trouvons des masses assez considérables en Suède, dans nos mines de fer, surtout dans celles des environs de Norberg et de Dannemora. Il existe deux types tout à fait différents de ces bitumes:

- (a) Bitumes donnant beaucoup de produits à la distillation et ne laissant après leur combustion presque point de cendres:
- (b) Bitumes ressemblant à l'anhracite, n'abandonnant à la distillation que des quantités insignifiantes et laissant un poids assez notable de cendres. J'ai analysé la cendre des bitumes anhracitiques et j'ai trouvé qu'elle contient toujours, outre silice, fer, chaux, magnésie, etc., plusieurs parties pour 100 des oxydes de nickel d'urane (3 pour 100), des terres de la célite et de la gadolinite. Ces mêmes oxydes, jusqu'à présent réputés si rares, se trouvent aussi dans la cendre d'une espèce de charbon formant de grands nodules dans nos plus anciennes couches sédimentaires (schiste à alun), ainsi que dans la grahamite de l'Amérique du Nord.

L'association dans les minéraux asphaltiques, de la matière charboneuse avec le nickel, l'uranium, le cérium, l'yttrium, etc., me semble fournir une indication: d'une part, sur l'origine des substances bitumineuses dans nos roches cristallines et nos plus anciens schistes sédimentaires, que seraient dues à des emanations de l'intérieur du globe; d'autre part, sur l'existence de combinaisons de l'oxyde de carbone avec l'uranium, l'yttrium, le cérium, etc., analogues au nickel carbonyl.

There can be little doubt that the material referred to by Nordenskiöld as "bitume (b)" was identical with thucholite. As a matter of fact, Nordenskiöld's statement as to the occurrence of the two kinds applies equally well to Canada. The writer has collected specimens which are almost like tar, rich in heavy hydrocarbons and which can be melted and ignited with a match but contain little or no rare element compounds. These also came from granite pegmatite, but the two varieties have not so far been found together in the same dike and it appears advisable to consider them entirely distinct minerals as they are quite different in composition and physical properties.

The mineral was apparently first discovered in Canada about 1894. In the *Journal of the Canadian Mining Institute*, Vol. VII, 1904, we find a paper entitled "On a Mineral Containing 'Radium' in the Province of Quebec" by J. Obalski, Inspector of Mines, Quebec. This short but interesting paper is reproduced in full below, the photographs and discussion only being omitted.

Since the new element, "radium" has been discovered, much attention has been called to the minerals containing it; so far it appears that the uranium ores are the only ones in which it has been found.

In our Laurentian formations, uraninite, composed of oxide of uranium and other rare metals, has been met with in some pegmatite dikes, which have been operated as producers of white mica (muscovite), and we have the records of the Villeneuve mine in Ottawa county, with monazite and uraninite, and of the Maisonneuve mine, in the county of Berthier, with samarskite; we have also several other white mica mines and prospects in the Saguenay district, but so far uranium ores from these have not been noted.

About ten years ago I found in a mine of the Saguenay district, then operated by the "Canadian Mica Co.," a remarkable specimen which I identified as "cleveite," and which is shown in the accompanying photograph.

This specimen has a specific gravity of 8.43, and weighs 375 grammes, or about 13 ounces. It is well crystallized in a dodecahedron form, deriving then from the isometric system. A complete analysis has not yet been made, but it contains 70.71 per cent of uranium oxide.

Radium having been discovered and found to exist mostly in connection with uranium ore, I experimented with the above mentioned crystal, and found it affected strongly the photographic plates, as shown by the accompanying radiographies.

The specimen has also a well marked action on the electroscope. I came then to the conclusion that it contained radium, and to have my opinion confirmed, I showed the specimen to Professor Rutherford, of McGill University, who, after testing it, stated that its radio-activity was equal to four or five times the activity the crystal would have if it was entirely composed of uranium and that it contained one-tenth of a milligram of radium, making it comparable with the best pitchblende so far operated for the production of radium. This crystal may be an accidental one, although I have found other small pieces of the same mineral in this vein, but I have also found in the same a carbonaceous material burning quite easily, and leaving ashes containing oxide of uranium. The specimens of the above coal have been tested by Mr. Milton L. Hersey, of Montreal, with the following results:

This "coal" has a fibrous, irregular structure, and contains a small amount of mica; it proved to be of a similar nature to a non-coking bituminous coal, yielding considerable gas, which burned with a bright yellow flame.

Volatile matter (including volatile combustible matter and a small quantity of moisture)	40.185
Fixed Carbon	52.590
Ash	7.225
	100.000

The ash itself was analyzed and found to contain 2.56 per cent of uranium, based on the coal, which is equal to 35.43 per cent of uranium in the ash itself.

It is important to note that the color of the ash was olive green, being due to the presence of oxide of uranium.

In another test, the coal was not burnt, but was merely finely pulverized and the powdered coal treated with boiling nitric acid, to dissolve the uranium compounds. It is then interesting to note that the uranium may be extracted by the direct treatment of the coal with nitric acid. A sample of this "coal" from the same place, tested in Montreal, was found to be radio-active as well as the ashes it gave; this radio-activity being due to radium.

I am not able to state what is the relation between the two specimens of cleveite and "coal" but I think this fact important, and I propose to make a further investigation next summer.

The white mica vein where these specimens come from is situated near the lake "Pied des Monts" about 18 miles back of Murray Bay, in the county of Charlevoix, on the north shore of the St. Lawrence.*

In spite of the meager chemical data for the Murray Bay mineral, it is quite evident that the material was very closely related to the Parry Sound mineral though apparently the former contained appreciable hydrocarbons which the latter does not.

* Note.—Since the above notes were read at the meeting, I had a letter from Mr. P. Curie, of Paris, in which he says that the small piece of cleveite from Murray Bay, I sent him, shows a radio-activity equal to six times the one of uranium, and he wants to be kept well posted about the new discoveries of radio-active minerals, in Canada.

One can be almost sure that the Murray Bay specimen contained vanadium, just as does the Parry Sound mineral, from the statement that the ash was colored olive green, this green color being due to vanadium rather than to uranium as stated, and there can be little doubt that the ash also carried thorium, rare earths, etc., like the Parry Sound and other examples. The similarity of the two occurrences is noteworthy. Both occur with uraninite in coarse granite pegmatites characterized by large muscovite crystals. The writer visited the Pied de Mont mine but it had been long abandoned and not the slightest trace of either uraninite or thucholite could be found. Nor was it possible to find any of Obalski's original material at Quebec. It is thought that he probably sent over most of his rare mineral finds to the Curies.

Thanks to the interest of friends among the mining men and by personal collecting the writer has secured specimens of carbon minerals from quite a number of Canadian localities. Of these some are tarry but three have been examined sufficiently to identify them as thucholite, so that including Obalski's Pied de Mont mineral we have at least four known occurrences, all in widely separated granite pegmatites of the Precambrian shield. Considering how seldom it is that pegmatite workings are visited by mineralogists or others likely to recognize this mineral and the brief periods of such visits when they do occur, it seems likely that substances such as thucholite and the tarry hydrocarbons are by no means rare in them, and that carbon as a constituent of igneous magmas is a much more important and frequently occurring element (if indeed in reality it does not always occur) than has been commonly supposed. The writer has in the past referred to this substance as a "mineral resembling anthraxolite," etc., which is true in regard to its appearance only, but as it is absolutely different from anthraxolite both in composition and origin, it seemed desirable to give it a distinctive name. Anthraxolite was the name first applied by E. J. Chapman⁸ to a substance from Lake Superior having the following composition:

Moisture	2.08
Volatiles	3.56
Fixed carbon	94.36
Ash	0.00
	<hr/>
	100.00

⁸ *Canadian Journal*, Vol. X, p. 410, as quoted by Ellis.

This was said to occur in a banded vein with quartz and pyrite.

The substance which has come to be accepted as the type anthraxolite, however, is the well known hard carbon of the Sudbury basin near Chelmsford, Ont., which is periodically rediscovered and put forward by promoters as anthracite coal. The writer has examined this mineral carefully both in the field and in the laboratory. Along with much quartz and pyrite it occurs as intrusive masses, pegmatite-like in form and sometimes of considerable size, cutting carbonaceous slates. The anthraxolite itself is almost pure carbon but it is much fractured and all the cracks, even the most minute, are filled with quartz along with, usually, a considerable amount of pyrite. It is scarcely possible to find a piece as large as an inch square, at most, which is not penetrated by quartz visible to the naked eye and the purest pieces when carefully burned in a crucible leave delicate films of silica where quartz had evidently been present as extremely thin veinlets. Apart from pyrite there is only the most minute amount of true ash actually belonging to the carbon itself, probably not more than a few hundredths of one per cent, and this appears to be chiefly iron. Tests for combustible volatile constituents and for heavy hydrocarbons gave negative results. Some analyses made long ago by Prof. W. H. Ellis⁹ of the University of Toronto, are produced below:

“Proximate Analysis of a Selected Sample of Sudbury Anthraxolite” by W.H. Ellis.

Moisture.....	4.0
Volatiles.....	1.8
Fixed Carbon.....	90.1
Ash.....	4.1
	<hr/>
	100.0

“Ultimate Analysis of a Carefully Picked Specimen (of Sudbury Anthraxolite) freed from Moisture” by W. H. Ellis.

Carbon.....	94.92
Hydrogen.....	0.52
Nitrogen.....	1.04
Sulphur.....	0.31
Oxygen.....	1.69
Ash.....	1.52
	<hr/>
	100.00

⁹ Analysis of some Pre-Carboniferous Coals, *Chemical News*, Oct. 15, 1897, pp. 186-188.

Ellis notes that the ash consists of silica and iron. Refined methods of gas analysis such as developed by Shepherd of course were not in use at the time of Ellis' work, but the analyses serve to show that there is not much volatile matter in anthraxolite.

The Sudbury mineral has always been considered to have originated from the distillation of the carbonaceous slates as a result of the intrusion of the Sudbury nickel eruptive. Thus it differs from thucholite in both composition and origin.

As there appears to be no suitable name in English for the carbon material described here the writer ventures to suggest the name thucholite as more or less appropriate, besides being easy to pronounce and to remember. The word consists simply of the chemical symbols of some of the main constituents with the letter "l" added for euphony, and the usual mineralogical ending "ite." The pronunciation suggested is thū'-chō-lite.

The writer wishes to thank Dr. A. L. Day, Director of the Geological Laboratory, Washington, and Dr. E. S. Shepherd of that institution, for the work of the latter on thucholite, also Mr. J. McLeish, Director of the Mines Branch, Department of Mines, Ottawa, for the use of apparatus and laboratory facilities of that branch.

Since the above article was written I have received (March 1928) from Prof. A. Fersman of Leningrad a communication informing me of the occurrence of thucholite in Russian pegmatites. The Russian mineral is described in a paper¹⁰ entitled "Mineralogy of Northern Karelii," very kindly sent me by Prof. Fersman.

A translation (from the Russian) of the portion of the paper describing the thucholite was very kindly made by A. Anrep, Geological Survey, Ottawa, and is as follows:

CARBON-URANIUM MINERAL

A carbonaceous uranium-bearing mineral of considerable scientific interest is found in certain veins both with uraninite and without it. The mineral is of black color, very friable and light (or soft) and is composed of grains ranging from 0.1 to 2 cm. in diameter embedded in quartz. Around these imbedded pieces as well as around the uranium are detected evidences of radium emanations; the periphery of the grains have short projections.

Experimentation by K. A. Nenadskevich showed that the mineral burnt easily and leaves an ash content of 9.5 per cent when heated in a closed tube. There is

¹⁰ A. H. Labunzof, *Bulletin de l'Academie des Sciences de l'Urss*, 1927, p. 617.

separation of water along with a slight amount of ammonia and a slight trace of sulphur.

The content of carbon is around 60 per cent. Moisture loss on drying at 100° 9.7 per cent and above 100° 19.3 per cent; 0.01 per cent NH_3 and H_2SO_4 . The ash (9.5 per cent) contains UO_3 54.20; PbO 17 per cent and Fe_2O_3 6 per cent.

This type of mineral with a decidedly magmatic origin is of great scientific interest as it shows that uranium does occur in certain kinds of carbonaceous particles.

Similar occurrences of carbonaceous uranium-bearing minerals are mentioned by Obalski of the Department of Mines, Province of Quebec, who states that in the pegmatite veins of Quebec as well as in the clayey wall-rock are to be found small carbonaceous grains, which after being burnt leave a residue of 7 per cent ash, containing 35 per cent UO_3 .

In an earlier part of the paper it appears that the author describes some 87 pegmatite dikes occurring in the district situated on both sides of the Murmanskoy railway from Kovda station on the north to Engozero station on the south. The district is about 100 miles long by 80 miles wide and is an eastern continuation of the Finnish mountain region which towards the east terminates near Kandalaschkii bay of the White sea.

Prof. Fersman states that further chemical investigations on the mineral are being carried on by Messrs. Nenadskevich and Wernadsky.

H.V.E.

II. CYRTOLITE INTERGROWTH ASSOCIATED WITH THE PARRY SOUND THUCHOLITE

As was noted in (I) the Parry Sound thucholite nodules almost invariably carry on their outer surfaces more or less embedded or intergrown cyrtolite. This is generally without definite crystal form but occasionally shows a development of the typical zircon-like square prisms with curved faces characteristic of cyrtolite. (Fig. 3). Usually also it is considerably altered. No detailed optical study of the mineral was made, but it was seen that there are at least three substances present, viz., the zircon-like prisms, a massive material which is more altered than the crystals, and minute inclusions of thucholite. Most of the grains had n greater than 1.75 but some were lower. All grains had indices greater than 1.65. From the analysis it appears that the constituent with the lower index is probably a phosphate which may be closely related to, or identical with, the material mentioned in (I) as being possibly a new silico-phosphate of calcium and the yttrium earths.



Fig. 3. Cyrtolite mass showing typical radiating structure at base and crude zircon-like crystal at top. Actual size about 1 cm. in longest direction.

Some typical examples of the cyrtolite intergrowth were selected and ground for analysis. A little thucholite was unavoidably present in the sample analyzed but not enough to appreciably affect the results, which are given below:

ANALYSIS OF CYRTOHITE INTERGROWTH ASSOCIATED
WITH THE PARRY SOUND THUCHOLITE.

${}^1\text{ZrO}_2$	43.03
U_3O_8	1.46
ThO_2	0.66
PbO	Not det. accurately
$(\text{Ce, La, Di})_2\text{O}_3$	0.34
$(\text{Yt, Er})_2\text{O}_3$	6.76
Fe_2O_3	4.16
Al_2O_3 , etc.	3.12
MnO	0.02
TiO_2	0.06
V_2O_5	Not detected
CaO	1.58
MgO	0.18
SiO_2	22.86
P_2O_5	3.53
Loss at 110°	1.64
2 Loss above 110°	10.90
	100.30

¹ Probably includes an appreciable amount of hafnium. The precipitation was by cupferron.

² Includes traces of carbon from thucholite.

Minerals such as cyrtolite and alvite (which apparently are much the same thing if not identical) have taken on new interest and importance as the richest source of element 72-hafnium. According to D. Coster and G. von Hevesy and associates the presence of considerable quantities of hafnium seems definitely to be directly related to an appreciable content of uranium, rare earths, etc. It is noteworthy also that zircons which are not altered contain very little hafnium and little or no uranium, rare earths, etc., while the cyrtolite group of minerals containing these elements are always considerably altered.

Cyrtolite is quite often found in Ontario pegmatites as nodules of crude crystal aggregates, sometimes as large as a fist or larger.