

URANINITE AND THORIANITE FROM ONTARIO AND QUEBEC¹

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ABSTRACT

The occurrence and mineral associations of 20 specimens of uraninite and thorianite are described. All the deposits occur in the Grenville Sub-province of the Canadian Shield.

The range in composition of these 20 specimens as determined by x -ray spectrography is U_3O_8 25–73%, ThO_2 7–55% and PbO 7–14%. Lattice constants (a_0) generally increase with increasing thorium content and range from 5.449 to 5.564 angstroms. After heating in air at 1000° C. for ten minutes the lattice constants are all reduced and more closely approach a lineal relation to the composition. The fact that none of the specimens oxidized to U_3O_8 on heating, indicates that they belong to the α group of Wasserstein.

The 20 specimens are readily subdivided into three distinct groups within each of which composition, lattice constants and type of deposits are distinctive.

An attempt to relate diffraction intensities of four different sets of planes to composition was inconclusive.

Introduction

Uraninite has been recognized from many pegmatite deposits in Canada and particularly from those in the Grenville rocks of Ontario and Quebec. Analyses of uraninite from many localities were published by Ellsworth (1932). In none of these analyses does the thorium dioxide content exceed 15%. However, during the summer of 1954 many new deposits were found in a region extending from Monmouth township in Ontario, 100 miles northeastward to Huddersfield township in Quebec, and from several of these deposits high thorian uraninite or uranian thorianite was identified.

Preliminary studies of these minerals have shown that high thorian uraninites and some of the thorianites have compositions differing from any previously reported. Moreover, their occurrence is restricted to deposits that are singularly alike in geology and mineralogy.

This investigation is part of a continuing study of uranium minerals in Canada. Examinations of most of the deposits were made by one of the authors (S.C.R.) and x -ray data and computations were provided by the other (A.P.S.). We are indebted to J. Satterly and D. F. Hewitt of the Ontario Department of Mines for some of the specimens and for descriptions of two of the deposits. The co-operation of E. A. Brown and S. Kaiman of the Mines Branch, Ottawa, in supplying additional material

¹ Published by permission of the Deputy Minister, Department of Mines and Technical Surveys, Ottawa, Canada.

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is much appreciated. Officers of the various companies that are prospecting and mining in the region have greatly facilitated our work. To them, and to colleagues of the Geological Survey of Canada who have assisted us in many ways, we express our thanks.

Mineralogy and Location of Deposits

The deposits are numbered and described in order of decreasing U:Th ratio of the uraninite or thorianite that they contain. The number of the deposit is the same as the number of the sample of uraninite or thorianite concentrated from it. Descriptions of deposits are taken from field notes except for that of Calumet Uranium Mines Ltd., whose description is based on specimens of the ore only.

Most of the accessory minerals of these deposits were identified by means of *x*-ray diffraction. Varietal names of feldspars, micas, pyroxenes and amphiboles are based on optical properties. Although uranothorite is reported from several of the deposits, its identification in all cases is tentative. It is metamict, and on heating in a vacuum to 1000° C. for five minutes, it yields a diffuse *x*-ray pattern equivalent to that of thorianite, as do other uranothorites. Continued heating of some specimens produced a product with a huttonite pattern. Many of the accessory minerals occur in small amounts. These were recognized in concentrates and middlings but were not seen in thin or polished sections.

In stating the location of the deposits, reference is given to province, township, concession or range, and lot in that order. Usually the concession (in Ontario) or range (in Quebec) is stated in roman numerals and the lot in arabic numerals but a letter is occasionally used for either concession or lot. Where work was known to have been in progress in the past year, the name of the company owning the deposit or holding it under option is given. Many of these deposits have been described more fully by Satterly & Hewitt (1955).

1. Ontario; Cardiff; XX; 7. Rounded masses of uraninite occur in a small concordant pegmatite, or migmatite band, in an area of hybrid granite gneiss. Accessory minerals include zircon, titanite and apatite.
2. Ontario; Faraday; XI; 16, 17. Faraday Uranium Mines Ltd., west drift in west adit. Small subhedral grains of uraninite occur in an ore shoot which consists of equal amounts of feldspar, magnetite and locally titanite. Quartz, zircon and fluorite are minor constituents. Uranothorite is also present in amounts slightly exceeding uraninite. The zircons exhibit sharply defined metamict and non-metamict multiple zoning. The ore shoot is a differentiate of granitic dyke rocks. These dykes are locally pegmatitic and are commonly radioactive. (See Satterly & Hewitt, 1955, p. 49.)
3. Quebec; Wakefield; III; 26. Uraninite crystals up to $\frac{1}{2}$ inch in diameter, occur at the contact between a zoned granite pegmatite and amphibolite. This border facies is a hornblende-rich syenite which contains accessory magnetite, pyrite, zircon and uranothorite.

4. Ontario; Cardiff; XI, XII; 27, 28. Centre Lake Uranium Mines Ltd., No. 4 dyke, 205' level. This dyke has a banded migmatitic texture suggestive of metasomatic replacement. In it, uraninite occurs disseminated in a lens-shaped band which is made up of oligoclase and microcline in equal amounts, with minor diopside, titanite, biotite and fluorite. Traces of uranothorite are also present. (See Satterly & Hewitt, 1955, p. 24.)
5. Ontario; Cardiff; XV; 6. Uraninite occurs in bands in a small syenite dyke composed of relatively fine grained oligoclase and microcline with minor titanite evenly distributed through it. Diopside is also present in patches. Uraninite grains attain diameters of $\frac{1}{4}$ inch and are subhedral to anhedral. (See Satterly & Hewitt, 1955, p. 33.)
6. Ontario; Cardiff; XI, XII; 27, 28. Centre Lake Uranium Mines Ltd., No. 1. dyke, adit level. Uraninite occurs as subhedral grains up to $\frac{1}{8}$ inch in diameter in a rather fine-grained footwall facies of the dyke. This rock is composed of equal amounts of microcline and albite (peristerite) with disseminated titanite and patches of diopside and hornblende. Some uranothorite and traces of pyrite and chalcopyrite are also present. (See Satterly & Hewitt, 1955, p. 28.)
7. Ontario; Cardiff; XXI; 9. This deposit is a very irregular pegmatite whose core is composed of a mixture of calcite and fluorite. Uraninite, in subhedral grains, occurs at the contact of the pegmatite with amphibolite, and in a tongue of fine grained syenite protruding from the main body of pegmatite. The syenite is composed of oligoclase and microcline with traces of diopside and titanite. Accessory minerals spatially associated with uraninite are hornblende, apatite, calcite, fluorite and traces of molybdenite and pyrite. Uraninite concentrates used in this investigation came wholly from the syenite differentiate. (See Satterly & Hewitt, 1955, p. 36.)
8. Ontario; Cardiff; XVII; A. Cardiff Uranium Mines Ltd., 125' level south. Deposits of this mine are veins or dykes of calcite and fluorite that change transitionally along strike to pegmatite. They are composed dominantly of calcite with subordinate purple streaky fluorite, minor apatite, and locally, feldspar and diopside. Partly rounded cubes of uraninite up to $1\frac{1}{2}$ inches in diameter are disseminated through the deposits. Accessory minerals include titanite, chondrodite, uranothorite, molybdenite and pyrite. (See Satterly & Hewitt, 1955, p. 17.)
9. Ontario; Cardiff; XIX; 2. Cardiff Uranium Mines Ltd., adit. This deposit was described by Wolfe & Hogg (1948); specimens collected by the writer were taken from the dump. Uraninite occurs in medium grained syenite and in calcite-diopside rock. Associated minerals are titanite, zircon, molybdenite and pyrrhotite. (See Satterly & Hewitt, 1955, p. 19.)
10. Ontario; Cardiff; XII; 29. Centre Lake Uranium Mines Ltd. Uraninite in cubes modified by octahedra, up to 1 inch in diameter, occurs in lenses of biotite-rich pyroxenite. These lenses together with bands rich in white calcite and leuco-granite occur in a series of amphibolites and crystalline limestone. Principal constituents of the radioactive lenses are biotite and diopside with minor hornblende, calcite and fluorite and accessory magnetite, titanite and apatite. No thorite or uranothorite were recognized in concentrates from this deposit. (See Satterly & Hewitt, 1955, p. 26.)
11. Ontario; Cardiff; XIX; 8. Anuwon Uranium Mines Ltd. Uraninite occurs in rounded to octahedral crystals in mica pyroxenite in which some pegmatite bands are emplaced. Associated minerals are apatite, salmon-coloured calcite, titanite, scapolite and fluorite. Crystals of uraninite occur in vugs in the rock. (See Satterly & Hewitt, 1955, p. 35.)
12. Ontario; Cardiff; XXI; 8. This well-defined lens-shaped pegmatite exhibits marked

differentiation from coarsely crystalline microcline and hornblende at the northeast end, to banded calcite-fluorite with an outer skin of hornblende and red apatite at the southwest end. Calcite-fluorite stringers and veins transect the coarse-grained facies in which feldspar and hornblende crystals attain lengths up to one foot and are partly oriented in comb texture normal to the walls. Uraninite in cubes modified by octahedra up to 1 inch in diameter occur in the 'skin' of hornblende and apatite with some accessory pyrite and molybdenite. (See Satterly & Hewitt, 1955, p.39.)

13. Ontario; Monmouth; VI; 5. Wadasa Gold Mines Ltd. The radioactive zones are composed dominantly of rounded diopside and phlogopite crystals in salmon-coloured calcite. Locally, tremolite is developed in crystals up to $\frac{1}{2}$ inch in cross section. Uraninite occurs in cubes, rarely modified by the octahedron, up to $\frac{3}{8}$ inch in diameter. Most of these cubes contain euhedral inclusions of diopside, Accessory minerals are pyrite and melanocerite. (See Satterly & Hewitt, 1955, p. 47.)
14. Ontario; Monmouth; XVII; 30. Homer Yellowknife Mines Ltd. Uraninite occurs as cubes up to $\frac{1}{4}$ inch in diameter in a skarn rock composed principally of calcite, diopside and phlogopite. Euhedral crystals of pyrite accompany the uraninite. This deposit was not visited by the authors. (See Satterly & Hewitt, 1955, p. 42.)
15. Ontario; Faraday; A; 29. Lockwood Farm. Rounded grains of uraninite and subhedral uranothorite occur in lenses of rock made up of coarse-grained tremolite and white calcite containing rounded grains of diopside. The presence of radioactive minerals is marked by a halo of hematite stain. Because the material tentatively identified as uranothorite occurs in and with uraninite, great care was taken in separation to use only the heaviest concentrate of a -200 mesh fraction. Examined under the microscope, the concentrate was found to be free from uranothorite. (See Satterly & Hewitt, 1955, p. 53.)
16. Quebec; Grand Calumet; VII; 29. Calumet Uranium Mines Ltd. This deposit was not visited. The crushed material which was studied, consists mainly of diopside, calcite, and mica with accessory uranian thorianite, monazite, rutile, and garnet.
17. Quebec; Huddersfield; V; 20. Yates Uranium Mines Ltd. Uranian thorianite in the form of small cubes, some of which are twinned, occurs in rock made up of salmon-coloured calcite, diopside and sparse phlogopite. Locally, actinolite and chondrodite are abundant. Veins of late white calcite with traces of fluorite transect all the above minerals. Thorite is present in about $\frac{1}{4}$ the amount of thorianite, and both are most abundant where phlogopite is plentiful. Lessingite, a silicate of thorium and rare earths is present in colourless grains.
18. Ontario; Dungannon; XVI; 14. Normingo Mines Ltd. Uranian thorianite occurs in zones marked by salmon-coloured calcite and phlogopite, in a country rock composed of impure micaceous marble and schist intruded by granite. The uranian thorianite is present as cubes up to $\frac{1}{4}$ inch in diameter, some of which are twinned. Accessory minerals in the thorianite zones are euhedral diopside, monazite, garnet, pyrite and pyrrhotite. (See Satterly & Hewitt, 1955, p. 48.)
19. Quebec; Huddersfield; V; 21, 22. Huddersfield Uranium and Minerals Ltd. Radioactive zones in impure crystalline limestone are composed of salmon-coloured calcite, euhedral diopside, and rounded subhedral phlogopite. Uranian thorianite occurs almost universally as interpenetration twins of the cube up to $\frac{1}{2}$ inch in diameter, and contains inclusions of calcite, diopside, and phlogopite. Traces of titanite and thorite were identified in the middling concentrates.

Acid pegmatites, worked for mica, occur within a few hundred feet of these deposits. Local weak areas of radioactivity were noted in the pegmatites but the minerals were not identified.

20. Quebec; Huddersfield; IV; 20. Yates Uranium Mines Ltd. This deposit is made up of salmon-coloured calcite, euhedral diopside and bands of phlogopite in an impure crystalline limestone host rock. Uranian thorianite is disseminated in the calcite but is slightly more concentrated in bands where diopside and phlogopite predominate. Accessory minerals are thorite and titanite with traces of chondrodite, allanite, and pyrite.

Laboratory Procedure

Concentrates of uraninite and thorianite were made by Mr. P. Lavergne from sized material using the Haultain superpanner and the Franz isodynamic separator. To avoid contamination due to composite grains, all material was crushed to pass 100 mesh. In two samples in which uranothorite was intimately admixed with uraninite, the material was ground to pass 150 mesh. All concentrates were examined under the binocular microscope and where necessary, were reconcentrated. It is thought that concentrates were at least 99% pure in all cases.

Concentrates were then ground to pass 325 mesh in a power mortar and the resulting powders were analysed for U, Th and Pb by *x*-ray spectrography. Standards were prepared to conform as closely as possible to the composition of each concentrate and analyses were made by the internal standard method, using strontium as the internal standard. It is intended to make chemical analyses of these concentrates at a later date. However, checks indicate that errors in these *x*-ray analyses do not exceed $\pm 5\%$ of the amount present.

X-ray powder diffraction patterns were made of all concentrates using a standard 114.7 mm. diameter camera and lattice constants were calculated from the same 14 lines for each pattern. Checks of accuracy, using two specimens of fluorite, indicate that errors in the lattice constants do not exceed $\pm 0.002 \text{ \AA}$.

A few milligrams of each sample were heated in air over a Meker burner at approximately 1000°C . for 10 minutes. *X*-ray patterns of these heated samples were made on standard 57.3 mm. diameter cameras, and lattice constants were calculated from 10 lines in each pattern. These lattice constants are thought to be correct within limits of error of $\pm 0.005 \text{ \AA}$.

For ten of the samples, intensities of lines indexed as (220), (311), (222) and (400) were measured on the *x*-ray diffractometer. These four sets of planes were chosen both because they represent major crystal directions and because they occur in a range of 2 theta in which peaks are well-defined and background level varies only slightly. For each sample two mounts were measured and background counts were made at four positions. It was found that reproducibility of ratios was not better than $\pm 5\%$ of the figure given. There was an even greater variation in absolute values.

Composition, Lattice Parameter and Diffraction Intensities

In the following table, twenty samples of uraninite and thorianite are listed in order of decreasing U:Th ratio. For each concentrate the percentage composition of U_3O_8 , ThO_2 and PbO is given, together with the U:Th ratio and the lattice constant (a_0) of unheated and heated material in angstroms. For ten of the samples the following ratios of diffraction intensities are also given: $I(311):I(220)$, $I(222):I(220)$, $I(222):I(311)$, and $I(222):I(400)$.

In Figure 1 the uranium and thorium composition as a ratio is plotted as the ordinate of a graph in which the lattice constants of both unheated and heated uraninite or thorianite are plotted as the abscissae. Ratios of U:Th are plotted down to unity but where that ratio becomes less than one, the Th:U ratios are plotted.

Results tabulated in Table 1 and plotted in Figure 1 show that there is a broad general increase in lattice constants of uraninites and thorianites with decrease in U:Th ratio, and that this relation is better defined when the composition is compared with lattice constants of heated material. In terms of Wasserstein's (1954) proposed classification, all of the uraninites and thorianites described in this paper are gamma type because on heating in air they did not decompose but their lattice constants did shrink. Unfortunately Wasserstein did not give the temperature and period of heating, nor did he mention whether he had attempted to determine conditions under which the lattice constant is stabilized. Our decision to heat at approximately 1000° C., for 10 minutes is roughly in line with conditions reported by Heystek & Schmidt (1953).

It is possible that when optimum conditions of heating are known and when more precise analyses of the samples are available, it will be found that the relation of U:Th to lattice constant will more nearly approach the linear function reported by Slowinski & Elliott (1952) for the artificial uranium and thorium dioxide isomorphous series. However, Brooker & Nuffield (1952) have shown that the lattice constant decreases with increasing $UO_3:UO_2$ ratio and this may explain the greater spread of points in the high uranium end of the graph in Figure 1. It will be observed that this spread is reduced for heated material as would be expected.

The effect of radiogenic lead in varying amount on the lattice constant has not been evaluated. In heating uraninite at 1000° C. in evacuated tubes, a pronounced sublimate of lead forms on the walls of the tube; it may be assumed, therefore, that some lead is lost in heating to 1000° C. in air.

All uraninites and thorianites of the group here reported, contain varying but small amounts of rare earths. The recent recognition by

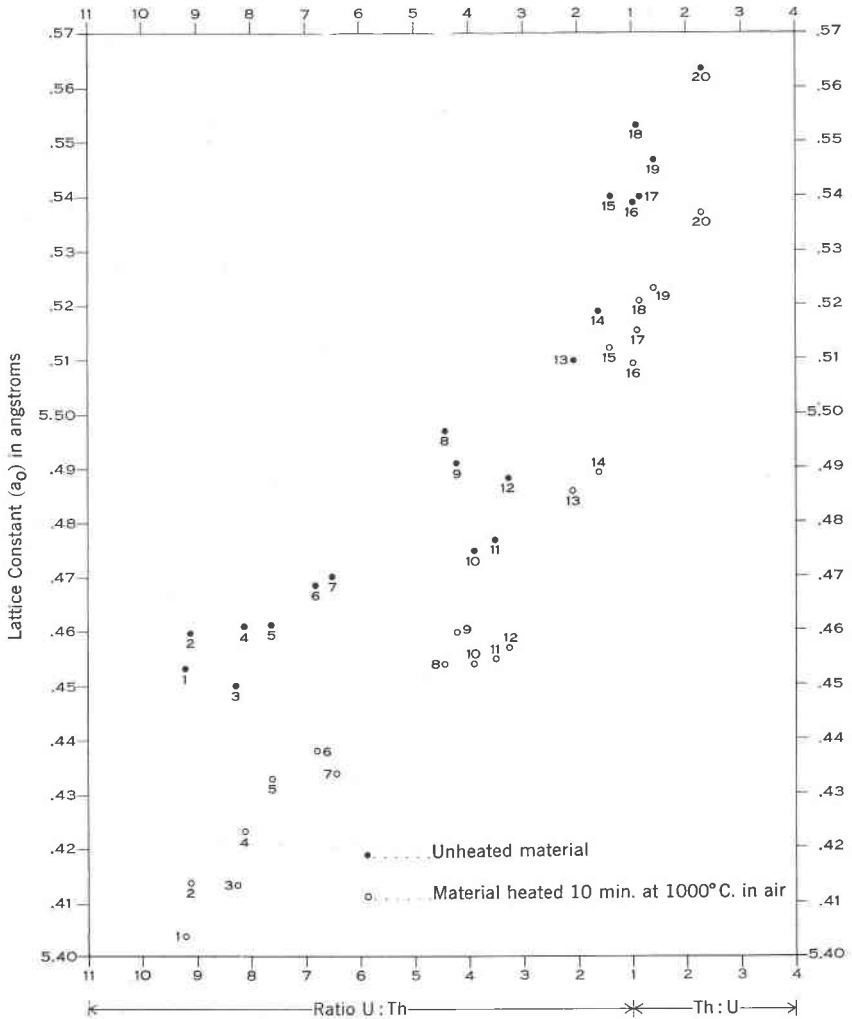


FIG. 1

Graham (1955) of a natural cerium dioxide, isostructural with uraninite and having a lattice constant within the range given for uraninite, suggests that cerianite must be considered as a third end member of the uraninite-thorianite series.

Attempts to use the lattice constant of uraninite or thorianite as an indicator of either composition or age must take account of all the above factors.

TABLE 1. COMPOSITION, LATTICE CONSTANT (a_0) AND RELATIVE DIFFRACTION INTENSITIES

Sample No.	Location	U:Th	Unheated $a_0(\text{\AA})$	Heated $a_0(\text{\AA})$	Composition (%)				Diffraction Intensity			
					U_3O_8	ThO ₂	PbO	$I(311)$	$I(222)$	$I(222)$	$I(222)$	
								$I(220)$	$I(220)$	$I(311)$	$I(400)$	
1	Cardiff; XX; 7	9.2	5.454	5.404	73.0	7.7	10.75	.82	.21	.26	1.8	
2	Faraday; XI; 16, 17	8.8	5.460	5.414	70.0	7.7	13.7	.81	.21	.25	1.7	
3	Wakefield; III; 26	8.25	5.449	5.413	69.0	8.1	13.9					
4	Cardiff; XI, XII; 27, 28	8.1	5.461	5.423	65.0	7.7	11.9	.84	.20	.24	1.6	
5	Cardiff; XV; 6	7.59	5.461	5.433	66.7	8.5	11.4					
6	Cardiff; XI, XII; 27, 28	6.79	5.468	5.438	64.5	9.2	11.9					
7	Cardiff; XXI; 9	6.43	5.469	5.434	66.7	10.0	8.4	.79	.20	.25	1.9	
8	Cardiff; XVII; A	4.44	5.497	5.454	62.8	13.6	12.8	.75	.22	.29	2.2	
9	Cardiff; XIX; 2	4.24	5.491	5.460	62.7	14.3	10.7					
10	Cardiff; XII; 29	3.86	5.475	5.454	68.0	17.0	10.75					
11	Cardiff; XIX; 8	3.53	5.477	5.455	59.6	16.3	10.3					
12	Cardiff; XXI; 8	3.24	5.488	5.457	59.0	17.6	10.5					
13	Monmouth; VI; 5	1.89	5.510	5.486	53.0	27.0	10.0					
14	Monmouth; XVII; 30	1.61	5.519	5.489	48.0	28.8	9.4	.78	.21	.27	1.9	
15	Faraday; A; 29	1.27	5.540	5.512	46.0	35.0	9.0	.77	.22	.30	2.5	
16	Grand Calumet; VII; 29	0.94	5.539	5.509	39.8	41.0	8.4					
17	Huddersfield; V; 20	0.88	5.540	5.515	36.8	40.1	7.7					
18	Dungannon; XVI; 14	0.83	5.553	5.521	40.0	46.4	8.0	.77	.22	.29	1.9	
19	Huddersfield; V; 21, 22	0.70	5.547	5.523	36.3	49.7	8.6	.78	.23	.29	2.1	
20	Huddersfield; IV; 20	0.43	5.564	5.537	25.0	55.5	6.8	.74	.22	.30	2.2	

It is apparent that the twenty samples can be divided into three distinct groups:

1. Uraninites containing 10% ThO₂ or less, and having a lattice constant less than 5.470 Å.

2. Uraninites containing between 10% and 18% ThO₂ and having lattice constants between 5.47 Å and 5.50 Å.

3. High thorian uraninites containing more than 20% ThO₂ and all thorianites. Lattice constants for this group are 5.51 to 5.65 Å.

Uranium, thorium and lead contents of groups 1 and 2 fall within, or close to, ranges of those published in Dana (1944) for uraninite. The three high thorian uraninites of group 3, (samples 13, 14 and 15), have compositions that are intermediate between those of published uraninites and thorianites. Composition of some of the thorianites, notably samples 18 and 19, also fall in a range in which other published analyses are lacking. Th:U ratios of thorianites for which analyses are published in Dana (1944) are: 19.6, 6.4, 2.2 and 1.07. For the six thorianites of samples 16 to 20, Th:U ratios are 1.08, 1.13, 1.2, 1.4 and 2.3. It is evident that the high thorian uraninites and thorianites of samples Nos. 13–20 extend the range in composition of known natural minerals in the uraninite-thorianite series and so strengthen the evidence that this series is fully isomorphous.

There is an interesting similarity in the types of deposits and mineralogy within each of the three groups. Deposits of *group 1* are pegmatite, granite, and syenite dykes which have well-defined walls but which in part, may be due to replacement. Dominant minerals are potash and soda feldspars with some quartz. Titanite is a consistent accessory mineral and zircon, apatite, and fluorite are also present in most deposits. In *group 2* the deposits include calcite-fluorite-apatite veins that are transitional to pegmatites, and mica pyroxenites containing abundant calcite, fluorite, and minor apatite.

Deposits of *group 3* are remarkably similar in essential characters and are quite unlike those of groups 1 and 2. They all occur in impure crystalline limestones as metasomatized zones which are apparently transitional to the country rock. In these zones, calcite is the dominant mineral and contains somewhat smaller proportions of subhedral diopside and phlogopite. In 7 of the 8 deposits the calcite is distinctively salmon-red in colour in contrast to the white and grey calcite in the impure limestone. Titanite, and less commonly, thorite, chondrodite and pyrite, are accessory minerals. It may be noted that Dana (1944) lists the one American deposit as occurring in serpentine at the contact of limestone and pegmatite.

Relative intensities of diffraction from the (220), (311), (222) and (400) planes in unheated material were measured to determine whether there

was any systematic variation of relative intensity with U:Th ratio, which might indicate some ordered, rather than random substitution of Th for U in the lattice. The atomic scattering factor of uranium is roughly 2% greater than that of thorium, so that differences in diffraction intensities due to possible ordered substitution of Th for U would be slight. Moreover, possible ordered effects resulting from auto-oxidation of UO_2 to UO_3 might well overshadow the Th for U substitution. The four sets of ratios shown do exhibit some indication of a trend with the U:Th ratio but reversals and lack of consistency combined with the stated limits of experimental error require that these results be treated with reserve. In general the intensity of the (222) powder diffraction relative to (311) and (400) appears to increase with thorium content. Random substitution of thorium for uranium would result in a relative decrease in the (222) intensities. Differences in the absorption of copper radiation by UO_2 due to oxidation of U, the presence of thorium, lead and rare earth elements such as cerium might also have a significant effect on the observed intensities. In this connection it may be significant that although the octahedron quite commonly modifies the cube in low thorian uraninites, it has not been observed in uraninite and thorianite crystals of group 3. It is also noteworthy that in 4 of the 5 deposits containing thorianite, twinned crystals were found with (111) as the twin axis. None of the uraninites exhibited this twinning.

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