

## THREE OCCURRENCES OF HIGH-THORIAN URANINITE NEAR EASTON, PENNSYLVANIA\*

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### ABSTRACT

The geology, mineralogy and paragenesis of three occurrences of high-thorian uraninite in serpentine near Easton, Pa., are described. Seven x-ray fluorescence analyses together with three corroborative chemical analyses show this mineral to be high-thorian uraninite rather than high-uranoan thorianite as supposed. Six samples contain from about 15% to 35% ThO<sub>2</sub>.

At the Williams quarry hydrothermal alteration of much uraninite to a sequence of secondary uranium- and thorium-rich minerals occurred. Frondel's Mineral C, thoringummite, then boltwoodite and uranophane were formed during serpentinization of fractured dolomite-diopside-tremolite contact-metamorphosed rock. Supergene coatings, chiefly hydrous uranium silicates and including boltwoodite and uranophane, formed much later.

At the reservoir site slight hydrothermal alteration of uraninite to thoringummite occurred. At the Royal Green Marble Co. quarry uraninite is found unaltered. At these localities fracturing or temperatures during later hydrothermal mineralization may have been of lower intensity.

It is concluded that thorium-rich Easton uraninite does not alter readily to secondary minerals, except when subjected to severe deformation and concomitant attack by serpentinizing hydrothermal solutions. The Th, U and Zr of this contact-metasomatic uraninite and the associated zircon originated in a granite-pegmatite magma rich in these elements, which intruded dolomitic rocks and through deformation while crystallizing yielded the hydrothermal solutions that deposited these minerals in those rocks, then serpentinized the rocks.

### INTRODUCTION

High-thorian uraninite, regarded until now as high-uranoan thorianite,<sup>1</sup> occurs in serpentine rock at three localities just north and northeast of Easton, Pennsylvania. The two earlier discoveries were made around 1930 by George W. Gehman (1936) at the C. K. Williams quarry and at the near-by site of the College Hill reservoir. In the past few years uraninite has been found also at the quarry of the Royal Green Marble Co. about a mile northeast of the first two occurrences and on the New Jersey side of the Delaware River.

This paper reports the results of a paragenetic study made of uraninite and associated minerals from all three occurrences. One result of the study reveals that the Easton mineral generally called thorianite is

\* Although not formally a contribution from the Mineralogy and Geology Departments of Harvard University, this may be considered one in view of the author's continuing work in the Berman Laboratory.

<sup>1</sup> The terms high-thorian uraninite and high-uranoan thorianite are arbitrarily taken for convenience at 15%-50% contained ThO<sub>2</sub> in uraninite and 15%-50% contained uranium oxides in thorianite.

actually high-thorian uraninite and that true Easton thorianite rarely exists. An early mineralogical study of this mineral from the Williams quarry was made by Wells, Fairchild and Ross (1933). The present investigation has depended on the large and unique collection of Easton uraninite specimens formerly belonging to George W. Gehman and now at Lafayette College. Mineralogy owes a debt to Mr. Gehman for making the original discovery of Easton uraninite at the Williams quarry, first identifying the mineral by chemical means, and then by unflagging effort throughout many ensuing years collecting nearly all of this material now available for study.

This study was much aided by Dr. Clifford Frondel of Harvard University and Dr. Brian H. Mason of the American Museum of Natural History, to both of whom sincere thanks are given. Grateful acknowledgment is expressed for  $\alpha$ -ray fluorescence analyses of uraninite specimens made in the Berman Laboratory of the Harvard Mineralogy Department by Dr. Robert M. Berman, for partial chemical analyses of three uraninite specimens made by Dr. Jun Ito at Harvard, and for photographs of two specimens taken by Dr. B. M. Shaub of Smith College. Dr. and Mrs. John S. Stevenson of McGill University kindly supplied samples of and information relative to thorianite from Pontiac County, Quebec. Dr. George Switzer kindly furnished a sample of thorianite from Betroka, Madagascar, from the collection of the U. S. National Museum. Clark C. McLean of Belvidere, N. J., generously made available for study all of his uraninite specimens from the Royal Green Marble Co. quarry.

#### GEOLOGIC SETTING

The mineralogy and paragenesis of these three uraninite occurrences cannot be fully understood without reference to the regional geology and the sequence of paragenetic events which controlled the formation of all minerals found in the serpentine-talc deposits near Easton. The geology of these deposits has been described by Peck (1905), Fraser (Miller, 1939), Bayley (1941), and Montgomery (1955).

A zone several hundred feet wide of serpentinized dolomitic limestone, of likely correlation with the Franklin metamorphic limestone of Precambrian age, follows the lower south slope of Chestnut Hill which borders Easton on the north (Fig. 1). This prominent northeasterly-extending ridge ends about a mile east of where the Delaware River cuts through it at Weygadt Gap. The uraninite occurrences and also a number of serpentine-talc quarries are located along this zone, mainly on the Pennsylvania side of the river.

In addition to its zone of serpentinized Franklin rocks, Chestnut Hill

is made up primarily of Precambrian para- and orthogneisses. The Precambrian crystalline rocks of the ridge occupy the core of an anticline formed during Appalachian folding, but they presumably owe their present position also to steep faults separating them from the flanking Cambrian limestones. Among the orthogneisses the Pochuck amphibolite and younger Byram granite gneiss predominate. The Pochuck is much

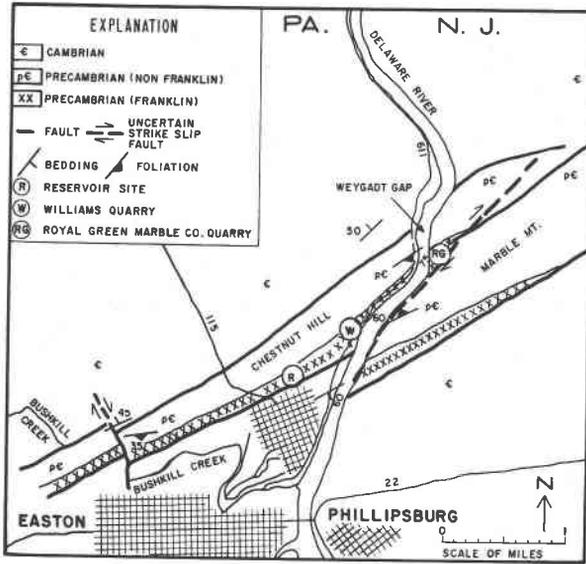


FIG. 1. Geologic sketch map of Easton area (adapted from maps of Peck (1905), Fraser (Miller, 1939) and Bayley (1941) and based on USGS Topographic Easton Sheet), showing location of three uraninite occurrences.

infiltrated in lit-par-lit fashion by Byram granite, and at Weygadt Gap these interlayered gneisses are crosscut by veins of coarse-grained tourmaline-bearing pegmatite probably related genetically to the Byram. Very coarse pegmatite outcrops on all sides of the Williams quarry a half-mile southwest of Weygadt Gap. Some of this pegmatite has clearly intruded Franklin rocks at the quarry and has been the unmistakable cause of contact-metasomatic effects in closely adjacent rocks. It seems beyond doubt that magma of such pegmatite, or magma related genetically to such intrusive pegmatite, gave rise to the hydrothermal solutions which serpentinized an originally dolomitic limestone and formed the numerous interesting and rare-element minerals now found in the serpentine deposits (Peck, 1905; Montgomery, 1955).

## MINERAL PARAGENESIS

Both field and laboratory study point to a long-continued series of related structural and metamorphic events in Precambrian time. These were the factors responsible for the complex paragenetic relationships found among the minerals of these serpentine deposits. That all of these events occurred during Precambrian time is attested by age determinations on two different contact-metasomatic minerals of the deposits. One gave 800 million years for thorianite from the Williams quarry (Wells, 1933), while the second of 850 million years is a recent determination by the Larsen method (E. S. Larsen, Jr., personal communication) on zircon from the quarry of the Royal Green Marble Co.

At the Williams quarry especially there is a bewildering variety of minerals—about forty distinct species have been found here—and metamorphic rock types. Nearly all of the latter, however, have been converted more or less in the direction of one end-product, serpentine. Most common of these rock types are those rich in gray-green serpentine, which may be massive, finely to coarsely micaceous, or else streaked or mottled with interstitial patchy areas of whitish calcite or pale brown to pink dolomite. The latter two minerals vary from coarsely crystalline character to finely fibrous material partly altered to tremolite. The micaceous serpentine is rich in flakes of pearly-gray to gray-green phlogopite from fine to very coarse size. A second common rock type consists chiefly of interlacing stubby to bladed prisms of gray-white tremolite. The tremolite prisms are in part fibrous and altered to greenish serpentine. This tremolite-rich rock is much in evidence at the north end of the Williams quarry. Another rock type is coarsely foliated talc, grayish or pale pink, and occurring in large-scale schistose masses above and west of the north end of the quarry. A less common rock type, found close to pegmatite above the southwest end of the quarry, consists of splintery gray-white diopside in crudely prismatic masses largely altered to fibrous tremolite. Relic parting surfaces belonging to the original diopside still show up prominently in this pseudomorphic tremolite.

The field and laboratory evidence in support of the paragenetic sequence of minerals given in Table 1 was described in detail by Montgomery (1955), chiefly for relations found at the Williams quarry. Some of the inferred paragenetic events and the main supporting points are summarized here.

*Thermal metamorphism:*

Following pegmatite intrusion certain silicious dolomitic Franklin beds were largely converted by thermal metamorphism to coarsely crys-

TABLE 1. PARAGENETIC SEQUENCE OF COMMON MINERALS OF EASTON SERPENTINE DEPOSITS

	folding and regional metamorphism	pegmatite intrusion and thermal metamorphism	shearing	hydrothermal metamorphism		supergene mineralization
				early	late	
calcite	=====	=====				
dolomite	=====	=====				
graphite	=====	=====				
diopside		=====				
phlogopite		=====				
tremolite		=====				
tourmaline (dravite)						
apatite						
zircon						
uraninite						
serpentine						
molybdenite						
sphalerite						
galena						
pyrite						
mineral C						
thorogummite						
boltwoodite						
uranophane						
taic						
quartz						
pyrolusite						
limonite						
cornellite (and several as yet unidentified U-mineral coatings)						

tallized diopside or tremolite. Relatively unaltered dolomite rock still occurs close to pegmatite, suggesting that the silica needed to form diopside and tremolite at this time did not come from a pegmatite source.

#### *Early hydrothermal metamorphism:*

The early diopside and tremolite formed as coarsely prismatic crystals, but even the freshest looking diopside now found is largely altered to fibrous tremolite. Beds of gray-white, graphitic and phlogopite-banded marble found above the southwest corner of the quarry also show partial alteration to fibrous tremolite. The formation of such fibrous tremolite, mainly pseudomorphic after diopside, reasonably marks an early stage of hydrothermal metamorphism. It is believed that crystals of uraninite and zircon were deposited at this time by early hydrothermal solutions. Grains of uraninite from the Williams quarry most commonly occur in massive serpentine enclosing patches of gray-white diopside partly altered to fibrous tremolite in finely gray-and-white-banded relic structures. These structures are due to selective filling or replacement by dark gray serpentine along parting planes originally developed in coarsely prismatic diopside. In a few specimens irregularly shaped grains of uraninite have unmistakably replaced diopside or fibrous tremolite pseudomorphic after diopside.

#### *Late hydrothermal metamorphism:*

Later periods of hydrothermal metamorphism were closely associated in time with powerful shearing forces. Extensive fracturing occurred and

the more permeable rocks were penetrated by hydrothermal solutions. While earlier mineral formation tended to be by replacement, here it tended to follow open spaces. The formation of the sulfides, molybdenite, sphalerite, galena and pyrite, occurred during this time. These sulfides largely formed in small fractures or along grain boundaries of earlier minerals. During this later hydrothermal mineralization serpentine formed on a large scale, in part directly from strongly sheared dolomite rock. Solutions from crystallizing pegmatitic magna now carried both water and silica. Coarse to fine flakes of colorless phlogopite formed in great abundance together with much of the serpentine, derived either from shaly impurities in the original rocks or from potash-bearing hydrothermal solutions. It must have been primarily due to these episodes of intense shearing, confined to particular rocks, and the resultant strongly-penetrative, but localized hydrothermal mineralization, that alteration of some of the uraninite in rocks now found at the Williams quarry to a sequence of secondary uranium-thorium minerals took place.

During latest hydrothermal metamorphism deformational forces continued to open up the rocks to widespread penetration by hydrothermal solutions. Open fractures became filled with whitish, silky, fibrous tremolite and calcite of finely acicular habit. Some of these tremolite-calcite fracture fillings are six inches wide and many feet long. One of the last minerals in the hydrothermal sequence was talc, which in part altered directly from serpentine along zones of very late shearing when lower temperatures prevailed. Final open spaces became partly filled with drusy quartz.

#### *Supergene mineralization:*

The effects of ground-water solutions during near-surface and surface weathering of rocks found at the Williams quarry are shown by the presence of thin fracture coatings of such minerals as limonite and pyrolusite and of such alteration products of uraninite as carnotite and uranophane.

Paragenetic relations seem much the same in rocks exposed at the quarry of the Royal Green Marble Co. and in specimens from the site of the College Hill reservoir. At the first locality massive apple-green serpentine is common, enclosing partly-engulfed patchy areas of pale-pink coarsely-rhombohedral calcite and crossed by fracture fillings of snow-white, fibrous calcite and tremolite. Molybdenite in thin flakes and coarse scaly masses and also pyrite in small cubes occur abundantly in some of this serpentine rock. Uraninite is found in association with these sulfides in this type of rock. Uraninite specimens preserved from the reservoir excavations on College Hill show either grayish-green, streaky and micaceous serpentine rock, or grayish rock rich in coarsely prismatic tremolite partly altered to pale green serpentine. Some of these specimens show

evidence of fracturing and shearing and marked alteration to gray-white, fine-scaly or powdery talc.

#### URANINITE AND ALTERATION PRODUCTS FROM THE WILLIAMS QUARRY

High-thorian uraninite from the C. K. Williams quarry (formerly called Sherrer quarry) occurs as black grains in several types of streaky or micaceous serpentine rock. Determinations of thorium-uranium content by the  $\alpha$ -ray fluorescence analyzer of the Berman Laboratory in the Harvard Mineralogy Department on four "thorianite" specimens from the Williams quarry, show this mineral to be much richer in uranium than thorium in all cases. A partial chemical analysis of one of these specimens ( $T_4$ ) by J. Ito at Harvard confirms the Th-U content as determined by  $\alpha$ -ray fluorescence analysis. The mineral is therefore high-thorian uraninite rather than high-uranian thorianite as formerly supposed. The original thorianite specimen from the Williams quarry analyzed by Wells (1933) and showing a  $\text{ThO}_2/\text{U}_3\text{O}_8$  ratio of about 51/49, cannot be regarded as typical for the locality.

The most common occurrence of uraninite from the Williams quarry is in a grayish-green serpentine rock showing grayish patchy areas of fibrous tremolite pseudomorphic after coarsely prismatic diopside. This tremolite has in turn been partly replaced by encroaching massive serpentine. The encroaching serpentine may also appear micaceous from an abundance of enclosed phlogopite flakes of pearly-gray to pale-green color. A second less common occurrence is in brownish or pinkish, fine-grained dolomite rock specked with small flakes of colorless phlogopite and streaked with grayish or brownish serpentine. Gehman (1936) reports that uraninite and also zircon were mostly confined to a narrow shoot-like zone soon mined out near the north end of the quarry. This would have been in association with the tremolite-serpentine rock still remaining in the walls there. Confirmation of this occurrence comes from traces of alteration products of uraninite still to be found near the floor of the quarry in this area. Well-formed zircon crystals, dark brown to creamy-gray in color and up to a centimeter and more in length, are associated with uraninite in several specimens of tremolite-serpentine rock from the Williams quarry.

The uraninite grains usually have a round shape, suggesting rounded cubic crystals, but some are highly irregular in shape. Some grains of the latter type have been fractured and broken apart by deformation. The grain size is from small specks up to a centimeter and more, with an average size of about 0.25 to 0.5 cm. The color is jet black in freshest appearing grains, but typically this grades into dark brown or reddish-

bronze shades because of slight alteration effects. The luster is resinous. The freshest material is opaque, compact, and breaks with a conchoidal fracture. In a few cases where diopside or tremolite have been partly replaced by patchy irregular grains of uraninite, the latter has a finely granular character.

The majority of uraninite grains from the Williams quarry have been altered to secondary uranium-thorium minerals to a greater or lesser degree. Although these alteration minerals have to some extent distinctive colors, they tend to grade into one another indistinguishably. The two earliest-formed secondary minerals, Frondel's Mineral *C* (Frondel, 1956a) and thorogummite have darker colors, ranging from chocolate-brown and brownish-orange in Mineral *C* to yellow-brown or pale orange-yellow in thorogummite, and have a marked resinous luster. The later-formed ones, boltwoodite (Frondel, 1956b), uranophane, carnotite, and several as yet unidentified species, are various lighter shades of yellow or yellow-green and do not possess a resinous luster.

Mineral *C* and thorogummite have largely formed within or along the edges of grains of altered uraninite, thus are pseudomorphic after those uraninite grains. The next-formed minerals, boltwoodite and some uranophane, have formed around the edges of altered uraninite grains or up to a few inches away, either by replacement of tremolite or diopside, or as fillings of small openings and along fracture surfaces. The latest-formed minerals, including boltwoodite and most of the uranophane, have commonly migrated some distance (whether in terms of a few inches or many feet) away from the grain sites of the original genetically-related uraninite, having been carried in solution to their new sites where they have been deposited as thin coatings on fracture walls or on exposed surfaces.

The alteration of uraninite from the Williams quarry to its secondary pseudomorphic products has proceeded in various ways and to a variable degree. Commonly patchy spots, irregular central cores, or narrow outer rims of dark brown, orange-brown, or brownish-yellow material appear in the grains and stand out in sharp contrast to the blacker color of relatively unaltered uraninite. Or else the original uraninite may have vanished, except for tiny scattered specks of black material, with its place wholly taken by minerals of brown or yellow colors. Where alteration of uraninite grains has progressed all the way to brownish-yellow or pale yellow material, made up chiefly of thorogummite or of boltwoodite and uranophane, the grain shapes are highly irregular due to obvious deformational effects. Deformation is especially evident where such grains are enveloped by massive serpentine. Most of the unaltered uraninite and its grains of more regular shape are associated with coarsely

prismatic diopside-tremolite material, which may have helped to protect these grains from the severest effects of deformation and the most penetrating attack by hydrothermal solutions.

The various alteration products of uraninite at the Williams quarry have been identified chiefly by optical properties and *x*-ray powder photographs. In refracted light the earlier-formed of these minerals appear to be minutely-grained aggregates. The mean or diagnostic index of refraction ranges from 1.78 to 1.80 for Mineral *C* and from 1.67 to 1.70 for thorogummite, and it is close to 1.670 for boltwoodite. The *x*-ray powder patterns for all three minerals are distinctive. Translucent reddish-bronze areas in several uraninite grains from the Williams quarry are isotropic and have an index of 1.84. This may be thorite, present as a very rare alteration product of high-thorian uraninite at this locality. In two specimens there are small irregular grains of a translucent red mineral with a high luster. It is isotropic and has an index of refraction of 1.94. After heating to 1000° C., a fragment of this red mineral gave the powder pattern of uraninite.

Color and luster aid in identification of these secondary minerals, especially when studied under the binocular microscope. Mineral *C* has a distinctive orange-brown or brownish-orange color when relatively pure, breaks with smooth conchoidal surfaces, and has a highly resinous luster. Thorogummite, which varies in color all the way from gray-white to yellow-brown but is most commonly a brownish-yellow at the Williams quarry, has a less-marked resinous luster and does not break to smooth conchoidal surfaces.

Where thorogummite grades into boltwoodite, the latter can be distinguished by its dull luster, rough-granular appearance, and a paler, more translucent yellow color. Boltwoodite also occurs as finely-fibrous material and as thin coatings on fracture surfaces. It is commonly closely associated with uranophane, and where the two minerals are intimately

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FIG. 2. High-thorian uraninite from Williams quarry showing differential alteration as controlled by serpentinization. The five black grains are rounded and deformed cubic or cubo-octahedral crystals of uraninite. They are rimmed and spotted with yellow-brown or orange-brown thorogummite and surrounded by gray fibrous tremolite with minor serpentine. Two yellow shapes near bottom center represent uraninite cubes engulfed by massive gray serpentine and almost entirely altered to brown thorogummite and yellow boltwoodite and uranophane.  $\times 1\frac{1}{2}$ . Photograph by B. M. Shaub.

FIG. 3. Secondary uranium minerals from Williams quarry associated with relic parting structures after diopside (light yellow areas with dark parallel lines). The structures were developed in diopside which first altered to fibrous tremolite. Parting planes were then infiltrated by dark-gray serpentine while intervening tremolite became replaced by yellow boltwoodite and uranophane derived from altered and destroyed uraninite close by. Gray-green serpentine surrounds the structures.  $\times 1\frac{1}{2}$ . Photograph by B. M. Shaub.

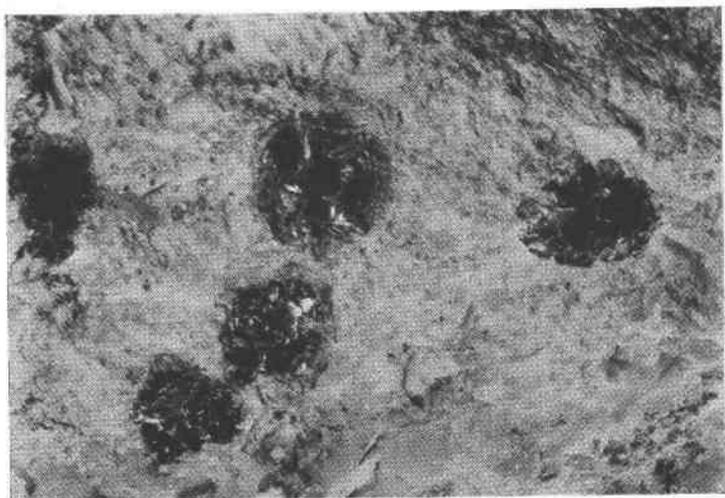


FIG. 2

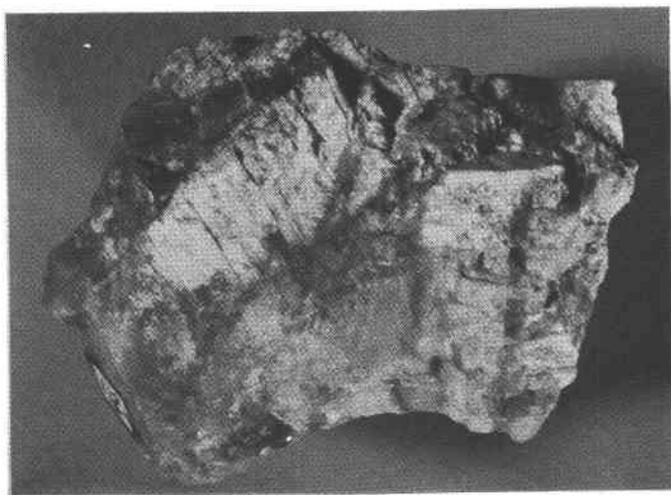


FIG. 3



mixed together as fibrous replacements of tremolite or diopside in the relic parting structures pseudomorphic after diopside, uranophane is more transparent and has a brighter lemon-yellow color than boltwoodite. Boltwoodite may be distinguished also by its rather weak yellowish fluorescence.

The later-formed secondary uranium minerals at the Williams quarry occur chiefly as thin fracture coatings or efflorescences on exposed surfaces. One of these minerals, found as a very rare coating of minute flakes of pale greenish-yellow color on phlogopite, fluoresces a bright yellow-green. Its high refractive index is 1.623 and it was first thought to be meta-autunite or uranocircite, but the *x*-ray powder photograph proves unidentifiable. A non-fluorescent mineral that occurs not uncommonly as yellowish fibrous or flaky coatings has been determined to be a new hydrous uranium silicate by mineralogists of the U. S. Geological Survey (S. Rubenstein and M. E. Staatz, personal communication). This mineral occurs with other secondary uranium minerals on the Colorado Plateau, as does boltwoodite. Carnotite occurs rarely as thin coatings of flaky aggregates with a distinctive yellow-green color. Uranophane occurs mostly as thin coatings of finely-fibrous character and pale-yellow to lemon-yellow color. An unusual occurrence of this mineral at the Williams quarry, found some years ago by John H. Bertrand, Jr., of Easton, shows tiny bright-yellow acicular crystals of uranophane intergrown with finely prismatic calcite and fibrous tremolite. The crystals are mostly aggregated in tiny prismatic bundles, but single crystals project into small cavities also. Among the specimens from the Williams quarry showing surface coatings of secondary yellowish or greenish-yellow uranium minerals, there may be additional species so far unidentified.

These alteration minerals of primary uraninite found at the Williams quarry are with one exception uranium-rich minerals with very little or no thorium present. The exception is thorogummite, which is high in thorium. It is likely that nearly all of the thorium once present in the primary high-thorian uraninite is preserved in thorogummite. Since in most cases studied Mineral *C* appears to have formed before thorogummite, it would suggest that once the primary uraninite had broken down to a secondary uranium-rich, thorium-poor mineral, the bulk of the thorium left over was used to make thorium-rich thorogummite.

#### URANINITE AT THE ROYAL GREEN QUARRY

In the past several years a small number of specimens of high-thorian uraninite have been found in material excavated from the serpentine quarry of the Royal Green Marble Co. (hereafter called the Royal Green quarry). This actively working quarry is on the New Jersey side of the

Delaware River, about a mile northeast of the Williams quarry and two miles north of Phillipsburg, N. J. The discovery was made in 1952 by J. Kent Perry of White House Station, N. J., but nearly all of the specimens since found were collected by Clark C. McLean of Belvidere, N. J.

This uraninite occurs as black grains in massive apple-green serpentine, in much of which coarse scales of molybdenite and small pyrite cubes are abundant. The mineral is jet-black, massive and compact, and shows a conchoidal fracture. It is opaque and shows a weakly resinous luster on freshly broken surfaces. The grains vary in size from mere specks up to more than a centimeter across. Some grain shapes are highly irregular, but most of the larger ones possess a crudely squarish or rounded outline suggestive of cubic or cubo-octahedral crystals. One rounded crystal 0.5 cm. in size is a modified cubo-octahedron.

Some uraninite grains are cut across by narrow veinlets of fibrous calcite and tremolite; others have been fractured and somewhat deformed and broken apart. In no case are there any signs of alteration effects. A Th-U determination by the Berman  $\alpha$ -ray fluorescence analyzer on a uraninite grain from this occurrence gave a contained Th/U ratio of 16%/32%. A partial chemical analysis of this specimen (T<sub>13</sub>) by J. Ito confirms the Th/U ratio but reports higher Th and U. This composition is very slightly higher in Th with respect to U than in analyzed samples from the Williams quarry. The lack of alteration to secondary minerals in uraninite from the Royal Green quarry seems hardly due, therefore, to higher-thorium, lower-uranium content, but more reasonably reflects less drastic deformational effects and weaker attack by hydrothermal solutions than in the case of the altered uraninite from the Williams quarry.

The close association of uraninite at this locality with molybdenite, pyrite, and also minor sphalerite and galena is of interest. These minerals occur at the Williams quarry, but much less commonly and not in close association with uraninite. Since scales of molybdenite and aggregates of small pyrite cubes are found following grain boundaries between uraninite and the surrounding serpentine in several specimens from the Royal Green quarry, the formation of sulfides at this locality came after uraninite. Molybdenite occurs commonly in certain zones of serpentine rock at the Royal Green quarry where no traces of uraninite have been observed. A few well-formed zircon crystals of reddish-brown color and up to a length of 2 cms. are also found associated with uraninite from the Royal Green quarry. Because they formed as isolated crystals and have been partly fractured and broken apart as have uraninite crystals from this locality, they are believed to have been deposited contemporaneously with the uraninite.

## URANINITE FROM THE SITE OF THE COLLEGE HILL RESERVOIR

Some years after his original discovery of uraninite at the Williams quarry, Gehman (1936) found a second occurrence of this mineral in material excavated from the site of the then-new College Hill reservoir and dumped along the banks of the Delaware River. It was already too late to examine the site of the occurrence, but Gehman managed to collect numerous excellent uraninite specimens from the excavated material. The rock proves to be similar in all respects to the typical serpentinized tremolite and dolomite rock found at the Williams quarry. The reservoir site is located on the south slope of Chestnut Hill about a half-mile west of the Williams quarry.

Nearly all of the specimens from this locality contain crude, but distinctly-formed uraninite crystals, ranging in size from about 0.25 cm. to 1 cm. Most crystals are simple cubes, but several are cubo-octahedrons. Two of the latter crystals show indistinct trapezohedral faces. The mineral is jet-black, opaque and compact, and exhibits a resinous luster on freshly broken surfaces. The occurrence is either in pale-green coarsely-prismatic tremolite, partly altered to greenish serpentine and grayish talc, or else in pale-pink to greenish-gray dolomite rock, specked with tiny white flakes of phlogopite and largely altered to olive-green serpentine and gray-white talc.

Many of the crystals have been fractured, deformed and broken apart. Powdery serpentine or talcose material thinly coats some of the broken crystal surfaces and surrounds crystal faces; in several cases this surrounding material has a reddish-brown color apparently due to staining by iron-oxide. In several specimens where the uraninite crystals are much fractured and broken apart and where the surrounding grayish matrix material is strongly talcose, the crystals show a partial alteration, especially on the outer surfaces, to a grayish or gray-brown, translucent and highly resinous substance. Some reddish-brown limonitic staining spreads outward also from these altered crystals. The resinous alteration material proves to be thorogummite of distinctly different character from the yellowish thorogummite found in intimate association with secondary uranium minerals at the Williams quarry. This material varies from a highly-lustrous, colorless, transparent substance to translucent grayish material, and it merges indistinguishably into opaque black thorianite. The colorless, transparent material seems isotropic, with a refractive index of 1.693, but gives a sharp  $x$ -ray powder pattern of thorogummite. The grayish material is anisotropic, gives a sharp  $x$ -ray powder pattern of thorogummite, and has a variable index ranging from 1.67 to 1.70. In one specimen, thin outer zones of several thorianite crystals, which have altered to grayish thorogummite, are covered with tiny, crude, parallel-

orientated crystals. These crystals, suggestive of somewhat rounded tetragonal prisms terminated by the dipyrmaid, were probably originally crystallized thorite present on the outer surfaces of pseudomorphs of thorite after thorian uraninite, which have since altered to thorogummite.

No signs of secondary uranium-rich minerals have been observed in uraninite specimens from this locality. Determinations of Th-U content by the fluorescence analyzer on two of the freshest appearing uraninite crystals from the reservoir site show slightly higher Th and slightly lower U than found at the other two Easton localities. A partial chemical analysis of one of these specimens (T<sub>11</sub>) by J. Ito confirms the Th/U ratio but reports higher Th and U. This is not enough of a difference in composition to explain the lack of alteration to secondary uranium-rich minerals at the reservoir site. The slight partial alteration of uraninite to grayish thorogummite found here in connection with strongly fractured uraninite crystals and a talcose matrix, indicates that shearing forces may have affected these crystals only during very late stages of hydrothermal mineralization when temperatures were low and talc was being formed from sheared serpentine rocks.

#### HIGH THORIUM CONTENT OF EASTON URANINITE

Table 2 lists the U-Th compositions and percentage ratios for a number of uraninite and thorianite specimens from Easton and elsewhere. The cell dimensions are also given for these samples, wherever possible, calculated from spacings between the 311 lines in the corresponding x-ray powder photographs. In general there is rough agreement between increasing cell size in these samples and increasing Th content; but when the samples are plotted on a graph using cell size and Th/U percentage ratios as the coordinates, the points do not fall on the virtually straight-line curve relationship which exists between ThO<sub>2</sub> and UO<sub>2</sub> in artificial material. The discrepancy is largely due to the variable state of oxidation of uranium in these samples. It is possible, nevertheless, to plot a steeper curve through these points, which could prove a useful guide in correlating between Th content and cell dimensions in samples of thorian uraninite or uranoan thorianite, subject to variability in the amounts and kinds of other elements present, in the radioactive decay, and in the oxidation of uranium.

Despite the variability in Th/U content found in Easton uraninite, six of the seven samples analyzed in this study contain from about 15% to 35% of ThO<sub>2</sub>. This is a high thorium content for uraninite, and the Easton mineral is noteworthy in occupying compositionally a portion of the wide central gap thought to exist between natural uraninite and thorianite.

TABLE 2. U-Th CONTENT AND CELL DIMENSIONS OF URANINITE AND THORIANITE

		X-ray fluores- cence analysis	X-ray fluores- cence analysis	Chemical analysis	Cell dimensions (in Å)
		% present (approx.) U:Th	% ratio (to 100%) U:Th	% ratio (to 100%) U:Th	calculated from x-ray powder photographs
T <sub>4</sub>	uraninite, Mi Vida, Utah			100: 0 <sup>1</sup>	5.42
T <sub>3</sub>	uraninite, Williams quarry	46: 7	86:14	86:14 <sup>2</sup>	5.44
T <sub>26</sub>	uraninite, Williams quarry	39:12.5	76:24		5.44
T <sub>5</sub>	uraninite, Williams quarry	42:16	72:28		5.51
T <sub>13</sub>	uraninite, Royal Green quarry	41:17	71:29	67:33 <sup>2</sup>	5.46
T <sub>2</sub>	uraninite, reservoir site	32:16	66:34		5.50
T <sub>11</sub>	uraninite, reservoir site	32:22	59:41		5.50
T <sub>12</sub>	thorianite, Betroka, Madagascar	29:22	57:43	54:46 <sup>2</sup> 5:95 <sup>2</sup>	5.53 5.60

<sup>1</sup> As generally reported for this locality.

<sup>2</sup> Jun Ito analyst, Mineralogy Department, Harvard University.

<sup>3</sup> As given for this locality in Dana's *System* (1944, 1, p. 621).

Even the freshest uraninite found at the three Easton localities occurs in serpentinized rock, thus must have been exposed to some attack by serpentinizing hydrothermal solutions. Thorianite, in contrast to the marked alteration behavior characteristic of uraninite, does not break down readily to secondary minerals. It is likely that high-thorian uraninite, of the type found at all three Easton occurrences, yields less readily to secondary alteration than low-thorian or non-thorian uraninite.

#### ORIGIN OF EASTON URANINITE

The origin of the high-thorian uraninite occurring in serpentine near Easton is bound up ultimately with a granitic magma which during Precambrian time intruded rocks now widely exposed over east-central Pennsylvania and west-central and northwestern New Jersey. The pegmatite end-products of this magma are characterized geochemically by the common presence of minerals carrying zirconium, rare earths, and thorium and uranium.

Coarsely crystallized allanite and zircon, usually in close association with granite pegmatite, have been reported from numerous localities in

the Precambrian crystalline rocks of east-central Pennsylvania and west-central and northwestern New Jersey. Recently rich deposits of uranoan thorite, monazite and zircon, of likely hydrothermal origin and closely associated with granite pegmatite, have been found near Chester, New Jersey (New Jersey State geologist, personal communication). Allanite occurs sparingly in coarse pegmatite collected from near the Williams quarry. Tiny zircon crystals are abundant in pegmatite stringers cutting Pochuck amphibolite a half-mile south of the Royal Green quarry.

The U and Th present in such granitic magma probably were used up in the normal case by dispersal in trace amounts throughout the crystal structures of accessory allanite and zircon formed in its intrusive bodies of granite. In the abnormal case where this magma was subjected to powerful deformational forces during crystallization following pegmatitic intrusion of dolomitic limestone, its U, Th, and also Zr, became free for transport into the intruded, strongly-sheared and highly permeable rocks by squeezed-off hydrothermal solutions which concentrated them in those rocks as contact-metasomatic crystals of Th-rich uraninite and of zircon.

#### CONCLUSIONS

Seven  $\alpha$ -ray fluorescence analyses (supported in three cases by chemical analyses, see Table 2) of typical specimens of the primary thorium-uranium mineral from three occurrences in serpentine near Easton, Pa., show this mineral to be high-thorian uraninite rather than high-uranoan thorianite as formerly supposed. In six of the analyzed specimens the ThO<sub>2</sub> content ranges from about 15% to 35%, a high thorium content for naturally occurring uraninite.

Although specimens of high-thorian uraninite and its secondary minerals from the Williams quarry present a complex problem in paragenetic relationships, field and laboratory study reveal a reasonable and orderly sequence of mineralogical changes. (1) Cubic or cubo-octahedral crystals of uraninite first formed by replacement of diopside or dolomite during early hydrothermal mineralization. (2) Where these crystals became exposed to long-continued attack by silica-bearing hydrothermal solutions, they were partly or wholly altered to the first-formed brownish-orange secondary product, Mineral C, and then to lighter-colored brownish-yellow thorogummite chiefly around the outer edges of the altered grains. (3) The formation of lighter-yellow boltwoodite and uranophane came only after deformation had affected the altered thorianite grains and, by fracturing of the surrounding minerals, had opened the way for thorough penetration by hydrothermal solutions which strongly serpentinized much of the matrix material. These penetrating solutions made

it possible for boltwoodite and uranophane to move outward from the edges of the altered and deformed uraninite grains, finding their way especially, through replacement of diopside or tremolite, into certain of the near-by relic parting structures pseudomorphic after diopside. (4) More recent near-surface attack by ground-water solutions dissolved uranium from the earlier-formed secondary uranium-thorium minerals, carried it some distance away, and redeposited it in the form of fracture coatings of pale yellow and yellow-green secondary uranium minerals that represent the final alteration products of the primary uraninite.

High-thorian uraninite from the reservoir site has altered in small part to gray thorogummite, or thorite later replaced by thorogummite. Strong fracturing promoted this alteration, but the absence of secondary uranium-rich minerals seems due to an attack chiefly by very late talc-forming hydrothermal solutions at lower temperatures than those prevailing during the hydrothermal alteration of uraninite from the Williams quarry.

High-thorian uraninite from the Royal Green quarry is found unaltered, despite exposure to serpentinizing solutions. Possibly pre-serpentine fracturing here was not sufficiently severe to expose this uraninite to a long-continued, penetrating hydrothermal attack.

It is concluded that because of its high thorium content none of the Easton uraninite alters readily to secondary minerals. Alteration to the earlier-formed secondary minerals has only occurred under exceptionally drastic, localized conditions of fracturing and concomitant attack by silica-bearing hydrothermal solutions. This alteration is known to have been hydrothermal rather than supergene, because it can be traced step by step through elucidation of the whole sequence of hydrothermally-imposed contact-metasomatic changes which affected the minerals of the associated serpentine deposits and to which the early-stage alteration of the high-thorian uraninite is inseparably linked.

The origin of the high-thorian uraninite found near Easton lies in a granite-pegmatite magma rich in thorium and uranium. This magma first intruded and thermally metamorphosed dolomitic limestones, then while subjected to powerful deformation during crystallization, yielded the rare-element- and silica-bearing hydrothermal solutions that penetrated the more permeable intruded rocks, deposited uraninite and zircon in them, and later serpentinized them on an extensive scale.

#### REFERENCES

- BAYLEY, W. S. (1941), Pre-Cambrian geology and mineral resources of the Delaware Water Gap and Easton Quadrangles, New Jersey and Pennsylvania: *U. S. Geol. Survey, Bull.* 920, 8-53.

- FRONDEL, C. (1953), Hydroxyl substitution in thorite and zircon: *Am. Mineral.*, **38**, 1007-1018.
- FRONDEL, C. (1956a), Mineral composition of gummite: *Am. Mineral.*, **41**, 539-568.
- FRONDEL, C. (1956b), Boltwoodite, a new uranium silicate: *Science*, **124**, 931.
- GEHMAN, G. W. (1936), Some minerals of the Serpentine Range near Easton, Pa.: *Rocks and Minerals*, **11**, 90-91.
- MILLER, B. L. (1939), Northampton County, Pennsylvania: *Pa. Geol. Survey, 4th Ser., Bull.* **C48**, 159-203, 435-466.
- MONTGOMERY, A. (1955), Paragenesis of the serpentine-talc deposits near Easton, Pa.: *Pa. Acad. Sci. Proc.*, **29**, 203-215.
- PECK, F. B. (1905), The talc deposits of Phillipsburg, N. J., and Easton, Pa.: *N. J. Geol. Survey, Ann. Report, 1904*, Pt. 3, 163-185.
- WELLS, R. C., FAIRCHILD, J. G. AND ROSS, C. S. (1933), Thorianite from Easton, Pa.: *Am. Jour. Sci.*, 5th Ser., **26**, 45-54.